National R-D Institute for Cryogenics and Isotopic Technologies - ICSI Rm. Valcea -

The 13th International Conference Tritium Science and Technology



Book of Abstracts

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Containment, safety, and environmental impact





ESTIMATION OF TRITIUM BEHAVIOR IN WATER COOLED SOLID BREEDER BLANKET FOR DEMO

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From the viewpoint of fuel control and tritium safety, it is necessary to correctly understand the tritium behavior in the subsystems constituting the tritium fuel cycle in a DT fusion reactor. The tritium breeding blanket is one of the major subsystems. In the solid breeder and water-cooled blanket, heat recovery and tritium recovery are performed at the same time. Previous studies have shown that the tritium generated by the nuclear reaction of lithium and neutron is released as both of HT and HTO from solid breeder materials. The HT/HTO ratio depends on the kind of breeding material and the purge gas. Since the permeation rate of HT through the metal wall is orders of magnitude faster than that of HTO, the permeation rate into the cooling water through the cooling tubes differs depending on the HT/HTO ratio, which affects tritium recovery efficiency. Some of the tritium implanted into the first wall of the blanket is also incorporated to the cooling water via the diffusion in the metal walls such as tungsten and F82H. Since mass transfer phenomena on the plasma facing wall are complicated, and it is difficult in the present to accurately evaluate the tritium transfer rate toward to the cooling water in consideration of this phenomenon correctly. Nevertheless, it is important to work on modeling with the aim of reflecting the results of fundamental experiments and improving the accuracy of tritium behavior prediction. The tritium permeated from the blanket module to the cooling water is transferred to the secondary cooling water via the heat exchanger. For safety management of primary and secondary cooling water and power generation equipment, it is required to evaluate the tritium transfer rate from the blanket modules to the cooling water. In this study, based on the research activities so far, the characteristics of tritium behavior from the blanket to the secondary cooling water in DEMO conditions were summarized.

 Li_2TiO_3 , which is the first candidate material of tritium breeder for JA-DEMO, has excellent chemical stability and causes only about 1% mass loss even in a water vapor of 500 ppm atmosphere at 900° [1]. In H₂ dry atmosphere, mass loss is less [2]. The effect of Li mass transfer on the surface reaction of tritium is not significant [3]. The chemical form of tritium released from the Li_2TiO_3 pebbles depends on the rate of isotope exchange reaction with H₂ and H₂O in the purge gas. In the numerical simulation using the isotope exchange reaction rate obtained in the fundamental experiment, when H₂O is 1 Pa and H₂ is 10 Pa,



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the ratio of T_2O in the released tritium is about 50% [1]. When H_2O is 1 Pa and H_2 is 100 Pa, the ratio of T_2O decreases to about 10%. Since the permeation rate of T_2O is about 3 orders of magnitude lower than that of T_2 , the T_2 partial pressure strongly contributes to the permeation rate of tritium to the cooling water. Under JA-DEMO reactor conditions, the coolant temperature is assumed to be 300°, 100 Pa of H_2 is expected to be added to the He purge gas, and then it was estimated that the produced tritium permeates to the cooling water at a rate of 2.3 g/day [4]. In addition, the tritium transfer rate to the cooling water due to the injection on the plasma facing wall was estimated to be 1.8 g/day at a uniform temperature of 400°. Considering the temperature distribution from 400° on the plasma facing surface to 300° on the cooling tube, the transfer rate of tritium was estimated to be 0.69 g / day [5]. When a tungsten deposition layer is formed on the plasma facing surface by the sputtering phenomenon, the tritium transfer rate is significantly reduced. On the other hand, due to the deposition layer, the tritium inventory on the wall surface increases [6]. In the presentation it will be also reported tritium behavior taking into account tritium transfer between high-temperature and high-pressure water.

Key words: Tritium, Blanket, Water cooled, Solid Breeder, DEMO

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OVERVIEW OF TRITIUM ACTIVITIES IN SOUTH KOREA

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The activities related to tritium technologies in South Korea have included developing the ITER Storage and Delivery System (SDS), ITER Test Blanket System (TBS) and investigating some systems for the fusion fuel. The ITER construction is one of the core activities for fusion energy. The developing technologies from the ITER can be combined with the experiences for building and operating the Wolsong Tritium Removal Facility (WTRF) handled the ppm-level of tritium. According to the Master Plan for Fusion Energy Development in South Korea, blanket technologies shall be developed for the DEMO including tritium handling technologies.

The SDS has been developed by observing the Depleted Uranium (DU) hydride behaviors as a tritium storage material, developing a DU hydride bed to store tritium safely and stably, and developing SDS unit processes to satisfy given the ITER functions and requirements. The performances of hydriding and dehydriding reactions of DU and DU hydride are observed using a full-size DU bed for the ITER by hydrogen and deuterium. The hydriding and dehydriding reaction rates are measured under various temperature and pressure conditions. The bed is one of the core components in the SDS and the lifetime of the bed has a key to determining the SDS availability. Heaters installed inside the bed can lead thermal cycles to perform the hydriding and dehydriding reactions. Heater manufacturing and bed assembly technology are the main factors that determine the life of the bed. The performance of the heaters with thermal cycles has been experimentally tested under practical operating conditions. The He-3 decayed by tritium can be re-used in the ITER and it can be recovered from the beds after storing tritium for a long time. The assessment of He-3 release from the DU bed is numerically performed and it can evaluate the impurity level of the tritium in the fueling gas. The He-3 with a bit remaining tritium can be purified by the Cryogenic Molecular Sieve Bed (CMSB). The regeneration procedure is developed and experimentally tested to compare the performances between purging and vacuuming methods.

The technical understanding of the ITER tritium plant has not only supported developing the SDS-related interfaces with the Isotope Separation System (ISS) and the



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Tokamak Exhaust Processing (TEP) but also initiated to analyze the tritium inventories in the high concentrated-tritium handling systems. The platform to simulate, analyze, and propose the ISS process design is developing and it can be applicable for various tritium handling systems later. Permeation membranes, catalytic reactors for CQ4 and Q2O, Ambient Molecular Sieve Bed (AMSB) for low concentrated-tritium recovery and their combinations are experimentally tested to confirm the availability as unit processes for the TEP and TBS. The WTRF has handled DTO and DT to remove and concentrate the tritium in the Wolsong Nuclear Power Plant (WNPP). The WTRF with the ppm-level of tritium in DTO and DT has targets to recover the tritium highly concentrated to reduce the tritium amount in the WNPP. Since the WTRF operation in 2007, the 80% of tritium concentration in the WNPP is lower and the tritium concentration of the exhaust in the site stack is similarly lower. The experience to build and operate the WTRF can be helpful to make the design of the low concentrated-tritium handling systems in the future.

The fusion power plant is the final goal for collaborating the ITER construction and developing the tritium handling technologies. In South Korea we have the Master Plan for Fusion Energy Development and the 4th period has started in 2022 during five years. Several blanket designs have been developed to verify the tritium breeding capabilities. And some of them are to be tested with neutrons to verify the tritium production ratio producing the tritium to be measured by sensors. The tritium shall be purified, separated, stored, and recovered in the blanket test site with a tritium handling facility to be designed. In order to develop the overall tritium plant of handing a large amount of tritium for a DEMO, a pilot using hydrogen and deuterium can be applicable to test the performance of the equipment and processes proposed as candidates. From the ITER to the DEMO the tritium handling technologies shall be developed with various aspects in South Korea: the amount of tritium, concentration of tritium, and material/equipment/process.

Key words: Tritium Technology, ITER, SDS, ISS, TEP, TBS, WTRF.



CLASSICAL THERMODYNAMIC ANALYSIS OF DEUTERIUM-BASED FUSION REACTIONS

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In classical thermodynamics, the spontaneity of a process is established through the assessment of the change in Gibbs free energy. So far, the feasibility of nuclear reactions has been characterized in terms of cross section and Q-value while the entropic term (T Δ S) has been neglected. Such an assumption is always justified for fission reactions where the term Δ S is positive. In the case of fusion reactions that operate at very high temperatures (10⁶ – 10⁷ K) and where Δ S is negative, the change in Gibbs free energy may result positive making the reaction non spontaneous.

This work describes a classical thermodynamic analysis of D-based reactions of interest for the magnetic-confinement fusion applications. The entropy contribution has been evaluated via the Sackur-Tetrode equation while the change in enthalpy has been considered constant and corresponding to the Q-value of the fusion reaction. The results of the thermodynamic analysis are compared with the nuclear reactions feasibility criteria relied on the reaction reactivity.

The reactions DT and D3He show a high degree of spontaneity although the second one presents a lower reactivity. The increase of the temperature could enhance the reactivity of the reaction D3He at the cost of decreasing its thermodynamic spontaneity. Both branches of the DD reaction are characterized by a much lower thermodynamic spontaneity than that of the DT and D3He reactions. Furthermore, at the temperature of their maximum cross section the DD reactions exhibit a largely positive change in Gibbs free energy and, therefore, are not spontaneous.

At the temperature of magnetic-confinement fusion machines (1.5×10^8 K), among the D-based reactions studied the DT one exhibits the highest degrees of spontaneity and reactivity.

Keywords: deuterium-based fusion reactions, magnetic confinement fusion, statistical thermodynamics.





> Containment, safety, and environmental impact





THE RADIOLOGICAL IMPACT ASSESSMENT OF ACUTE TRITIUM RELEASES IN ENVIRONMENT

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Tritium has a complex behaviour once released into environment. Tritium can be effectively incorporated into biological systems, including the human body, as organically bound tritium (OBT) with a larger residence time than tritiated water (HTO). There are few nuclear facilities with high tritium loads and the potential risk of short term atmospheric releases in case of techological incidents cannot be ignored. The International Atomic Energy Agency (IAEA) considered among its topics the accidental tritium releases [1, 2], in order to provide a harmonised model (research and/or operational grade) to be further used for emergency prepardness and for accident management in real time conditions. Regulatory requirements for an operational model are the following: to be relatively simple, transparent, easy to program, results should be conservative (but not too much), to provide deterministic calculations (the worst case assessment), probabilistic calculations (95% percentile for the worst case) [2,3]. The processes involved in tritium transfer in crops including soil are very complex and consequently, the previous requirements were not entirely achieved for tritium transfer by now.

Romania has two CANDU 6 reactors in operation and it is interersted to clarify the impact of a potential short term atmosperic tritium release for the local conditions. The present study is focused on the tritium transfer in foodchain. A previous study demostrated that the tritium dynamics in animal products can be predicted with a reasonable acuracy if the tritium intake in animal's foodstuff (in both forms HTO and OBT) is known [4]. It was also clarified that the food/feed contamination is predicted with an uncertainty lower than that of the atmopsheric contamination resulted from transport of tritium from source to receptor [3]. Some results of the Romanian model, CROPTRIT were reported [5]. In the present study further on going developments of CROPTRIT model are sumarised regarding the needs which must be accomplished by an operational model in order to be applied to Cernavoda Nuclear Power Plant (NPP) site. The nearest crop around Cernavoda NPP is vineyard (at 1.4 km from stack) and at about 3 km from stack, there are large agricultural lands cultivated with wheat, maize, sun flower (with root depth of 1-4 m). Vegetables are cultivated, also and pasture lands are spread at various distances. The winegrapes have a minor contribution to routine ingestion dose, but in case of accidental wet deposition of HTO, the situation can change due to very deep roots and long time until HTO in soil will be depleted in the rooting zone. For OBT production at day the requirements for



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a photosynthesis model were provided elsewhere [5] and the partition factors of dry matter to plant part are needed. The model of tritium dynamics in crops must be supplied with a crop growth model tested with site specific data in order to provide the input parameters for tritium dynamics. The OBT production in case of a tritium release during the night time depends on the transformation of starch to sucrose and then to amino acids. An important process is plant growth at night when the clean starch from reserves is mixed with HTO in leaves and then exported to other plant parts [6]. The OBT production in a case of night release is more complicated than for day release for crops which grow at night, also.

The challenging task is to find a simple solution for the atmosphere-soil interface and root uptake in order to have an operational model for HTO dynamics in soil-plantatmosphere complex, including the re-emission of HTO in air due to evapotranspiration processes. The CROPTRIT model is focused on the influence of various processes contributing to variability and uncertainty of tritium (OBT and HTO) at harvest and further studies are needed.

Keywords: tritium modelling, radiological impact assessment, operational model, uncertainty.

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TRITIUM PERMEATION AND INVENTORY IN DEMO EUROFER97 EXPOSED TO WATER

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Tritium containment and control are essential prerequisites for fusion to comply with the promise of a safe energy with low impact on the environment. In fusion reactors, the plasma-surface interactions will result in tritium contamination of the components. Safety concerns require a careful evaluation of tritium inventory and permeation in the walls of fusion reactors; this evaluation requires a better understanding of tritium permeation and trapping in plasma-facing components.

To that end, transport and trapping parameters in Eurofer97 were evaluated using hydrogen permeation and deuterium thermal desorption spectrometry experiments coupled to the reaction-diffusion code MHIMS. This analysis showed that three trapping sites are needed to model the behaviour of this material in its operating temperature range (room temperature to 873 K). The influence of these trapping sites, especially the third one (1.65 eV energy) has been studied through simulations of permeation cycles, showing that usual modelling parameters such as effective diffusivity and effective solubility oversimplify the dynamics of trapping, which results in underestimations of the inventory in PFCs [1].

Using the three-traps model MHIMS simulations as a basis, the Wapiti experiment (WAter-interface Permeation In Tritium-exposed materials) was designed. This experimental device consists in several permeation cells where thin metallic samples are exposed to gaseous tritium at room temperature. This experiment was performed on Eurofer97 with a downstream phase of air or water and air, allowing for a direct comparison of permeation in these media. The first results agree with MHIMS in terms of timelag prediction (around 25 days). Furthermore, the water permeation cell results indicate that tritium is more likely to form HTO through isotopic exchange than to recombine and form T2. The presentation will cover experimental and modelling results.

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APRIL: A NEW FACILITY FOR THE CHARACTERIZATION OF ANTI-PERMEATION COATINGS FOR FISSION AND FUSION APPLICATIONS

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Anti-permeation and anti-corrosion coatings are being developed in order to reduce tritium permeation from liquid metal (LiPb of WCLL BB and Pb for Lead Fast Reactors) to Primary Heat Transfer Systems. The facility APRIL (Alumina-coating for tritium Permeation Reduction for Innovative LFR) was designed and installed in ENEA Brasimone R.C to characterize the permeation reduction factor (PRF) of the candidate coatings in static conditions. APRIL is composed of three pipes, closed at one end, that simulate the heat exchangers of the fission and fusion reactors. Two of the pipes are coated with two different techniques: Pulsed Laser Deposition (PLD) and Atomic Layer Deposition (ALD). The third pipe is uncoated. During the tests, all the pipes will be filled with pressurized steam (100 bar) at relevant temperature for the fusion or fission application: 480°C, steam generator conditions of ALFRED LFR, or 328°C, water loop of the WCLL BB. The tests will be made in gas: the three pipes are installed in a chamber filled with helium having a known concentration of deuterium that simulates tritium. Deuterium will permeate inside the pipes, allowing to evaluate the PRF by means of the ratio between the measured permeated flux in the uncoated pipe and in the coated ones. Different deuterium partial pressures will be used in the tests to cover the entire range of interest.

Keywords: Tritium permeation, coating, fission reactor, fusion reactors



A PASSIVE SYSTEM TECHNICAL SPECIFICATION FOR CANDU 6 GENERATION // REACTOR

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Fukushima accident leads to enhancement of safety for the new nuclear systems technology and the existing fleet of nuclear power plants (NPPs) was required. An important objective is to increase the time window opportunity for plant operator to establish an alternate heat sink in case of Station Black Out (SBO) accident [6]. The efforts made by the RATEN in the frame of H2020 PIACE projects was to implement a passive safety system in CANDU 6 project [2,7]. In this project, RATEN was in charge of the engineering design and computational modelling aspects, required to integrate a passive safety system in the existing CANDU 6 project. In order to design a passive safety system, a three-days SBO accident was credited to occur at the Cernavoda NPP, a CANDU 6 type reactor. An Isolation Condenser (IC) system capable to transport the total energy produced in reactor core due to decay heat was designed and modelled. The engineering design solutions were made by RATEN CITON and the thermal-hydraulic analysis was performed by RATEN ICN using RELAP5 computer code, to confirm the natural circulation both in the secondary and primary circuit, during the SBO accident and heat transfer capability of IC with and without noncondensable gases. The passive safety system design consists in four (4x33%) closed loop independent circuits, one for each steam generator. Each loop has an IC design to transport 0.66% of nominal thermal power of the reactor [1]. In order to avoid a rapid transient during reactor cooldown, the system is endowed with four noncondensable gas tanks (one for each IC), connected to the outlet header of each IC, provided for reducing the IC heat flux simultaneously with reactor core residual heat decrease. The design concept was adapted to the CANDU 6 reactor power and the specific layout of Cernavoda site [3, 4], starting from the ALFRED patent [8] (the demonstrator of the lead fast reactors technology) passive system, to increase the plant operator window response frame time form 23 hours (current situation) to more than 72 hours.

Keywords: passive safety, SBO, CANDU 6, fission reactors, Isolation Condenser (IC), PIACE.

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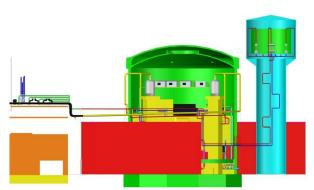


Figure 1. Passive Safety System for CANDU 6 Cernavoda Unit 2 NPP



PROCESS FOR THE DETERMINATION OF *EU-DEMO* HAZARDOUS INVENTORY NEEDED FOR SAFETY PURPOSES

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DEMO is an European project aiming at creating self-sustaining fusion reactions and to allow electrical power production. To achieve this goal, DEMO will create fusion reactions between deuterium and tritium (D-T) in a plasma within a vacuum vessel with specific temperature and particle density conditions. These reactions and the process to achieve them will handle or generate hazardous inventories for which safety provisions will be implemented in order to limit their effects on workers, members of the public and the environment.

The EU-DEMO inventory will contain radioactive nuclides, chemical materials as well as some other materials that may challenge safety functions. Tritium is one of the key major players in the impact of future fusion facilities but is not the only one.

Knowing the inventories, and controlling them, is important for DEMO Central Team for:

- Assessing the compliance with the safety objectives established for any nuclear facility

- Protecting the workers against the effects (e.g. external exposure, internal exposure) issued from the created or handled inventories in any situations, and in particular during maintenance,

- Protecting the members of the public against the hazards (e.g. doses from radioactive or chemical releases, dose rates) issued from the created or handled inventories in routine or accident situations,

- Protecting the environment against the hazards from radioactive, chemical or thermal releases (e.g., radioactive, thermal, environmental impact on soil, food, water, etc.) in routine or accident situations,

- Protecting the "legacy" via recycling/reducing/limiting/avoiding radioactive waste, and in particular high level waste with long term nuclides.

In order to cope with it, an inventory working group within EUROfusion has been setup with a roadmap based on the accessibility of available data, the R&D performed so far, and the lessons learnt from other fusion projects. Several sub-groups have been created for dealing with various inventories of interest for the most important design drivers:

- Tritium, as the nuclide that is underlying all the other nuclides, and that can be mixed or adsorbed in some particles from other nuclides,

- Activated Coolant Materials (activated corrosion products, activated fluids inside coolants),



- Activated dust (e.g. from erosion inside the vacuum vessel)
- Activated Breeding Blanket fluids, this will depend on the selected technology,

- Other inventories that would be considered as a design driver (e.g. cryogenic inventories, hydrogen/deuterium inventories...).

The goal of this working group, and sub-working groups, will be to deliver relevant inventories to be used for both the safety design of DEMO facility and the safety analyses that will be delivered for FP9 programme.

The presentation will provide the process implementation of this WG as well as the preliminary outcomes useful for the main DEMO stakeholders.

Keywords: Tritium, Nuclear safety, Environment, Hazardous Inventory

Acknowledgments:

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AN ASSESSMENT OF TRITIUM CONFINEMENT PHILOSOPHIES AND THEIR APPLICATION TO DEMO

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Tritium confinement systems are implemented to prevent and reduce the amount of tritium released from process equipment, in both accident and emergency scenarios. The purpose of this study is to provide recommendations on how to implement confinement strategies to the DEMO fuel cycle. This study firstly summarises and compares different confinement approaches employed at different tritium handling facilities. Also, considered are the best practice for tritium confinement from standards, recommendations and UKAEA practices. The application of tritium confinement is then considered for DEMO. Finally, recommendations are made on future work and considerations required to assist in the future designs of tritium confinement, including to minimise release in accident and normal operations and designs to achieve ALARA. The findings and recommendations are also applicable to other tritium handling facilities and equipment.

Keywords: Tritium Confinement, DEMO, Fuel Cycle



CONTAINMENT AND DETRITIATION IN BALANCE OF PLANT SYSTEMS

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All systems in the DEMO Balance of Plant will experience tritium contamination due to permeation through pipe and equipment walls. Tritium will be lost to the environment or may cause high tritium-in-air concentrations. This report investigates mitigations for these effects. The Balance of Plant design options are summarised and sources of tritium highlighted. Previous studies are analysed, and levels of tritium permeation compared to release limits and permissible tritium-in-air concentrations. Suggestions are made for minimising tritium loss to surrounding rooms and the use of vacuum or purged containments are investigated, commenting on possible issues. Estimates are made on parameters such as purge flow rates or vacuum required. A double layered containment consisting of a vacuum inner and purged outer layer is recommended to provide effective mitigation of permeation-to-rooms. The return of captured tritium to the fuel cycle is discussed, with amounts and compositions of gas streams estimated. Receiving systems are recommended, either Exhaust Processing or the Isotope Separation system. This work allows a new stream to the fuel cycle to be accounted for, and notes what might be required to avoid excessive loss of tritium to the environment. It also highlights additional considerations required by tokamak building designers.

Keywords: Tritium; Fuel cycle; Balance of Plant; Containment; Permeation

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ORGANICALLY BOUND TRITIUM LEVEL IN PLANTS DURING GROWING SEASON AND ENVIRONMENTAL FACTORS INFLUENCE

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In many countries, the fate of tritium (³H) in the environment has become a growing concern since the release rates of this radionuclide are expected to rise significantly over the upcoming decades. Changes in fuel management methods along with new tritium-emitting facilities, such as the International Thermonuclear Experimental Reactor (ITER) and Evolutionary Power Reactor (EPR), are thus expected to generate increasing discharges of tritium into the environment. Understanding the behavior of tritium in the environment is therefor an ongoing societal issue. Tritium can be found in several forms within the environment, e.g. gaseous, liquid or integrated into living organisms as Tissue Free Water Tritium (TFWT) or Organically Bound Tritium (OBT). Over the past several years, it has become increasingly acknowledged that OBT is the most pertinent tritium form for understanding its behavior and distribution within the biosphere. OBT plays a key role in radiation exposure to tritium because its ingestion through foods occupies a considerable fraction of tritium intake in man. [1]

Currently OBT is considered the sum of the exchangeable and non-exchangeable OBT and it can be expressed as Bq/kg fresh weight, Bq/kg dry weight or Bq/L combustion water [2], but in order to compare the tritium levels from different environmental compartments, it is important to remember that the OBT concentrations must be expressed in Bq /L of combustion water. [3]

The level of tritium around ICIT Rm. Valcea in all environmental compartments is not influenced by tritium releases from the Tritium Removal Facility (TRF), so far, TRF conducting only experiments under exclusion legal limits. Plant species chosen for this study were: fir tree, apple tree, vine, corn and wheat. In order to evaluate the OBT level during vegetation period, at least two samples per year from each studied plant species were collected and analysed. The environmental tritium level was monitored during vegetation periods in 2020 and 2021 (from April to October), in a specific location, near Tritium Removal Facility. A necessary step was considered the monitoring of tritium concentration with respect to the main environmental elements that can affect this evaluation: precipitation, air, and TFWT in soil and plants [4]. The combustion of the samples for OBT analysis was done using a Parr bomb 1121. All tritium measurements were performed by a low-background liquid scintillation spectrometer Quantulus 1220 (PerkinElmer), using a low background liquid scintillation cocktail (Ultima Gold uLLT). For



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this specific area the only variation of tritium level in the environment is the seasonal variation. The development stage of the studied plants correlated with the HTO (rain, soil and air), influence the OBT level in the samples. The measured level of OBT depends of the development stages of the plants, some differences being observed on OBT level from different plant organs (leaves, branches, fruits, buds, etc.), although the tritium concentrations as overall do not exceed 2.5 Bq/L.

Keywords: organically bound tritium, growing season, environmental factors, tritium level.

Acknowledgement: This work was carried out through the "Nucleu" Program, ctr. no. 9N/2019, developed with the support of the Ministry of Research, Innovation and Digitalization, project no. PN 19110302 "Research on variation trends specific to stable isotopes in different tree species: deepening of fractionation mechanisms and interconnected chemical processes on the soil-water-plant chain"

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ACCUMULATION OF ORGANICALLY BOUND TRITIUM IN KOMATSUNA CULTIVATED IN SOIL CONTAINING TRITIATED WATER

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A large amount of tritiated water will be generated in a Deuterium – Tritium fusion reactor because tritium can permeate through metal cooling tubes into the cooling water at high temperature conditions. Considering the safety of public and environment, the most important technical issue for the fusion reactor is confinement of the tritium within the fusion power generation site. On the other hand, from a viewpoint of social acceptance of the fusion reactor, it is also important to clarify tritium behavior in the environment assuming a severe accident including the release of tritiated water to the environment. When some amounts of tritiated water is released to the surrounding soil, there would be a possibility that tritium is absorbed into the plants and stay in the plants for a long time. Most of the tritium absorbed in the plants exists in a form referred to as tissue free water tritium (TFWT), which is not bounded to tissues. Some tritium exist as organically bound tritium (OBT), which is bounded to plant tissues through photosynthesis [1]. OBT is further classified into exchangeable OBT (eOBT) and non-exchangeable OBT (neOBT). neOBT is strongly bounded to carbon in plants until the organic substance is decomposed. On the other hand, eOBT is bound to sulfur, nitrogen, and oxygen atoms [2]. Since the behaviors of eOBT and neOBT are significantly different, it is important to evaluate the amount of tritium uptake into plants with discrimination between eOBT and neOBT. In this study, an airtight space was constructed in which plants were cultivated without tritium release to the surrounding, and an edible plant, Komatsuna was cultivated in the soil which had tritiated water. After that, leaves and stems were collected, and the amount of tritium uptake was measured using liquid scintillation.

Figure 1 shows the illustration of the cultivation equipment. Two airtight glove boxes were installed in the upper and the lower part of an incubator respectively. Six (6) samples of Komatsuna with all its leaves removed was placed in each box. 6 cc of tritiated water at a concentration of 150 kBq / cc was dropped to the culture soil of each sample in the upper box. Fresh air was continuously supplied by one pump and the air in the inside space containing tritium was evacuated by another pump. The tritium in the exhaust air was collected with water bubblers. The samples were always exposed to illumination. Leaves and stems were collected from Komatsuna cultivated for several days as samples for measuring the tritium uptake. In order to measure the amount of TFWT, water immersion and vacuum drying were performed. Next, in order to recover the eOBT by isotope



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exchange reaction with water vapor, the sample was exposed to moist air at room temperature. Finally, in order to measure the amount of neOBT, the sample was combusted at 800 °C in the gas flow including oxygen. Here, the amount of tritium retained in the stem is shown as follows. It was obtained that TFWT was 6.35 kBq/g, eOBT was 1.91 kBq/g, and neOBT was 0.77 kBq/g. The fraction of neOBT to total tritium uptake was found to be 8.55%. It was indicated that about 10 % of the tritium transferred from the soil into the stems of Komatsuna is stably accumulated as neOBT.

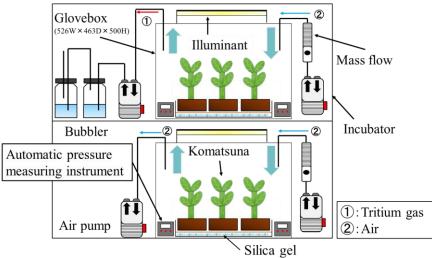


Figure 1. Experimental equipment for cultivation

Keywords: Environment, Komatsuna, Organically Bound Tritium

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STUDY ON METHODS FOR SUPPRESSING MIGRATION OF TRITIATED WATER IN SOIL

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It is assumed that the cooling water for the plasma-facing wall and the blanket of deuterium-tritium (DT) fusion reactor contains non-negligible amounts of tritium since tritium penetrates metal walls relatively easily under high-temperature conditions. In addition, tritium leaked into the workspace in each fuel processing system is recovered as water vapor and treated as tritiated water. Therefore, the fusion reactor facilities handle a large amount of tritiated water. Safety management of tritium is an important issue for the realization of the fusion reactor, and along with developing technology to prevent tritium from leaking into the environment, it is necessary to fully understand the behavior of tritiated water flowing in the soil is captured by soil particles during the percolation process. It is considered that tritium is captured mainly through the isotope exchange reaction because light hydrogen exists as inter-layer water and structural water in the clay minerals consisting of soil. In this study, to consider the method of suppressing the migration of tritiated water to the deep part of the soil, the percolation experiment of tritiated water in the soil mixed with adsorbents and swelling clay minerals was conducted.

5.1g of molecular sieve (MS3A, 1/16 pellet) was mixed with 15.0g of natural soil collected on the campus of Kyushu University. After the mixing process, the sample was packed in the soil column in the glove box and a certain amount of tritiated water was quickly poured from the top of the soil column. The mass of water flowing out from the soil column outlet was continuously measured using an electronic balance. A part of the effluent water was sampled non-periodically and the tritium concentration was measured with a liquid scintillation counter. Similarly, 15.0 g of natural soil was mixed with 15.8 g and 2.6 wt% of montmorillonite powder that a swelling clay mineral and 17.8 g of the mixture was packed in a column to conduct the percolation experiments of tritiated water.

In the MS-mixed soil, the outflow of tritiated water started earlier than the natural soil, and the decrease rate of the tritium concentration in the effluent water by tritium capture in the initial outflow was smaller than the natural soil. In other words, no effect on percolation suppression due to tritium capture into the MS was observed. Although the MS has a relatively large isotope exchange capacity, it is considered that tritium was not captured into MA sufficiently due to the slow migration of tritiated water into the pores of MS. The percolation rate of tritiated water into the soil mixed with 15.8 wt% of montmorillonite was very slow, and the height of tritiated water in the above column



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decreased only about 4 mm in 6 hours for a packing height of 44 mm. And it was observed that a swelling layer of about 15 mm was formed on the top of the soil column in 4 hours, and water penetration was strongly suppressed. In the soil mixed with 2.6 wt% montmorillonite, although the penetration rate of tritiated water was remarkably low, the outflow from the bottom began after 105 minutes. The tritium concentration in the effluent water was low, about 80-70% of the concentration in the poured water. The possibility of capturing tritium while allowing the penetration of water was indicated by using clay minerals that contain a large amount of light hydrogen.

Keywords: Tritiated water, Soil, Clay mineral, Adsorbent, Fusion reactor

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Decontamination and waste management





IN-SITU TRITIUM DECONTAMINATION OF THE *KATRIN* REAR WALL USING A *UV*/OZONE TREATMENT

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The KATRIN collaboration aims to determine the neutrino mass with a sensitivity of 0.2 eV/c² (90% CL). This will be achieved by probing the endpoint region of the β -electron spectrum of gaseous tritium with an electrostatic spectrometer. A gold-coated stainless-steel disk defines the reference potential for the high precision neutrino mass measurement and it terminates the β -electron flux as the physical boundary of the tritium source. This so-called rear wall is exposed to the gaseous tritium, which leads to ad- and absorption, which in turn leads to systematic uncertainties for the neutrino mass measurements that need to be understood and mitigated.

In maintenance phases during which the gaseous tritium source was emptied (< 10^{-5} of nominal gas density) the increasing activity of the rear wall was monitored using betainduced X-ray spectrometry (BIXS) and direct observation of emitted β -electrons with a silicon detector. The dependency of the observed activity increase to the integral tritium throughput was investigated and found to converge from a limited exponential growth to a continuous linear growth.

This presentation will give an overview of the results we obtained using several methods of in-situ decontamination of the rear wall while continuously monitoring the activity. The decontamination methods included bake-out during continuous evacuation, flushing the system with nitrogen, deuterium or air with residual humidity at different pressures and illumination of the rear wall with UV-light. These well-known methods lead only to a small (\approx 16%) decrease in the observed activity. However, a decrease of the surface activity by three orders of magnitude in less than a week was achieved by combination of different methods using UV light, a heated surface and a low (several mbar) pressure of air inside the chamber, leading to the production of highly reactive ozone. This proved to be the by far most efficient method, drastically reducing the contribution of the rear wall surface activity to the beta spectrum of the gaseous source.

Keywords: Tritium decontamination, surface activity, measurement and monitoring, ozone, UV light, KATRIN, gold, stainless steel.



HYDROGEN ENERGY ENGINEERING, PINK HYDROGEN, CHEMICAL ISOTOPE EXCHANGE WITH WATER, WASTE DETRITIATION

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Currently, an increasing number of countries, including Russia, are announcing large-scale programs for the development of hydrogen energy, including its production and use. In Russia, in 2021, the Concept for the Development of Hydrogen Energy was approved, according to which by 2024 Russia will have the ability to export about 200 thousand tons of hydrogen [1]. At the same time, special attention is paid to methods for producing hydrogen that do not have a carbon footprint. One of these methods is the electrolysis of water. At the same time, electrolytic hydrogen obtained using the off-peak electrical power of nuclear power plants according to the international classification is of the pink type.

In accordance with one of the projects within the framework of the Concept in Russia, it is planned to create a complex for the production of electrolytic hydrogen with a capacity of 150 tons/year on the basis of the Kola NPP with the prospect of increasing productivity up to 1500 tons/year. The appearance of a large-scale production of such hydrogen allows the authors of this report to propose a number of promising projects for its transit use for solving urgent problems for the nuclear industry, in particular, detritiation of waste water from nuclear power plants. This project assumes the application of the method of chemical isotopic exchange of hydrogen with water in the variant of CECE (Combined Electrolysis and Chemical Exchange) technology. A schematic diagram of a separation plant for solving this problem is given below.

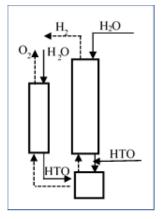


Figure 1 –Schematic diagram of separating installation for detritiation of NPP waste water

1 - electrolyzer, 2 - column of isotopic exchange of hydrogen with water,
 3 - column of purification of oxygen flow from water vapor



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The report presents the parameters of the separation installation for the detritiation of NPP waste water with a capacity of 1500 kg / day, obtained on the basis of an estimated calculation

Keywords: electrolysis, hydrogen production, transit use, isotopic exchange with water, detritiation of water waste of NPP

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DETRITIATION OF JET BERYLLIUM AND TUNGSTEN

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Investigations were undertaken into the thermal treatment of beryllium and tungsten to see if these materials can be detritiated in the Material Detritiation Facility (MDF) at UKAEA, allowing for the declassification of intermediate level waste (ILW) to low level waste (LLW). When heated in oxygen, both tungsten and beryllium readily oxidise, with a series of different oxides forming as temperature increases. These oxide layers act as a tritium barrier, reducing the amount that can be removed by thermal treatment and as such need to be avoided by heating at lower temperatures. Additionally, the formation of beryllium oxide presents health and safety concerns due to its toxicity and physical form. Experiments were undertaken using tungsten and beryllium samples from JET ILW campaigns. The samples were heated in a pyrolyser, and the tritium released was captured in a series of bubblers. The remaining tritium in the material was characterised by acid dissolution at external labs to allow for detritiation factors to be calculated. Tritium was successfully removed from the samples by heating in air, and detritiation factors of 16.95 for tungsten and 121 for beryllium were found. Future trials will use are being using JET ILW samples that have had their tritium content increased via soaking. This would allow for samples representative of ILW to be detritiated and could demonstrate the ability of the process to reduce the tritium inventory and allow declassification of ILW to LLW.

Keywords: Tritium separation, detritiation, decontamination and waste management, beryllium, tungsten



TRITIUM DECONTAMINATION OF COMPONENTS AND MATERIALS AT CNL AND UKAEA

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The minimisation of waste produced in fission and fusion applications is critical to the public acceptance of fission as a continuing and fusion as a future power source. UKAEA (United Kingdom Atomic Energy Agency) and CNL (Canadian Nuclear Laboratories) each handles significant quantities of tritiated components and materials prior to their storage and disposal. Tritium decontamination techniques have been developed by each organisation for various purposes including determining the tritium effects on materials, reducing the hazard associated with wastes, reusing the tritium, and reducing disposal costs.

Given the vast number of possible component materials that could be in contact with tritium in complex systems, ranging from metals, polymers, and other components, UKAEA and CNL are joining efforts to leverage each other's strengths and thus have a greater arsenal of decontamination recipes at their disposal.

In metals, studies were undertaken at UKAEA to determine the feasibility of thermal treatment to reduce the activity of tritiated materials (>12 kBq/g), considered Intermediate Level Waste (ILW) under UK regulations. UKAEA's Material Detritiation Facility (MDF) was designed and built in 2017/18 following the results of the studies reported here. The MDF includes a fume cupboard processing line, furnace, and gas handling system to enable waste to be added to the furnace, treated and tritium captured for re-use in another facility (JET Water Detritiation System). An important test of the JET MKIIa divertor was undertaken as validation of the facility. Detritiation percentages of 99.84% \pm 0.13% and 98.27% \pm 1.44% were demonstrated for Inconel and Stainless Steel, respectively. These levels of detritiation offer an additional option for UK permitted landfill disposal, further reducing cost and burden on UK nuclear infrastructure.

For polymers, long term investigations have been undertaken at CNL to determine the resistance of electrolyser ion-exchange membrane materials to the adverse conditions associated with the self-radiolysis of tritiated water. Detritiation results obtained on Proton Exchange Membrane (PEM) have been published previously [1]. Detritiation results obtained on the alkaline electrolysis ion-exchange membrane IMET[®] (Inorganic Material Electrolyser Technology) are discussed in this paper.

IMET membranes underwent accelerated testing by exposure to \sim 30 TBq/kg (\sim 830 Ci/kg) heavy water for periods of 16 and 32 months, resulting in theoretical doses of 390 and 690 kGy, respectively. CNL developed a decontamination method that would not impact the physical



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properties of the membranes so that the effect of tritium induced damage by the highly tritiated water could be studied. The materials were decontaminated to "free release" levels (< 1 GBq total quantity), and underwent tensile testing and electrolyser performance testing for comparison with fresh membranes. Details of this study will be discussed in this paper.

Keywords: Tritium, decontamination, detritiation, waste, H3AT

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THE RECOVERY OF TUNGSTEN FROM DETRITIATION PROCESSES

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Tungsten, or tungsten-rich alloys, thanks to their properties have been proposed as constructional materials for various plasma facing components in fusion reactors. During the fusion, tritium permeates into the walls of these lamellae, where hydrogen embrittlement occurs, forming blisters that rupture and form a tritiated waste. With new developments in fusion technology, it can be expected that the demand for metallic tungsten will skyrocket and it is therefore important to focus on the possibilities for tungsten recovery and recycling due to finite recourses and need for sustainability.

The aim of the experiments was to determine the possible reaction of metallic tungsten of the different grain sizes in the MSO (Molten Salt Oxidation). In the MSO process the waste is injected, together with excess air, into a molten alkali carbonate bath. During this process tungsten reacts with alkaline salt in an oxidative environment and is captured in the molten salt and the flue gases, including tritium, are lead through a series of water bubblers filled with demineralised water where the tritium is captured and then analysed. The captured tungsten then can be recovered from the molten salt.

A series of experiments were performed with metallic tungsten of a different grain size (20 μ m, 0.4-0.8 mm, 1-2 mm) and alkaline molten salts. The aim was to determine the reaction of metallic tungsten in molten salts (Li₂CO₃, Na₂CO₃ and K₂CO₃) at different temperatures varying from 800°C to 980°C. In the first series of experiments alkaline salt was melted in a metal crucible and cooled down. Tungsten was then poured onto the surface of the cooled salt and the crucible was inserted into the reactor vessel. After a set time of heating the molten salt was poured out, cooled, and analysed. In the second type of experiment the tungsten was gradually dosed through a dosing tube onto the surface of melted salt in the reactor vessel. The other experiments the cooled molten salts were dissolved, and unreacted metallic tungsten was separated. A XRF analysis was performed to determine if tungsten was present in the solutions. After this analysis various acids were tested to precipitate tungsten from the dissolved inorganic salts solutions in the form of WO₃. XRD analysis of solid samples also showed the presence of impurities, which entered the molten salt from the crucibles used for melting.

The experiments have shown considerable reactivity of metallic tungsten of the smallest grain size in the molten salt as the metallic tungsten recovery yield was less than 16%. The recovery yield of metallic tungsten increased with its grain size. There was also a significant difference between the Li_2CO_3 and the other two alkali salts, as the experiments with Li_2CO_3 showed higher recovery yield of metallic tungsten which was over 75% at the



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grain size of 0.4-0.8 mm and over 92% at the grain size 1-2 mm. The main reason was a lower melting temperature Li_2CO_3 . Using this salt has its' disadvantages as it has an inverse solubility in water, therefore the recovery of WO_3 from the cooled salt was more complicated compared to the other two salts and another disadvantage is its' price.

After the first two series of experiments Na_2CO_3 was chosen for the complete MSO process due to it having a lower melting point than K_2CO_3 and easier dissolution in water compared to the Li_2CO_3 . Although the highest grain size was more resistant in the molten salt, the grain size of 0.4-0.8 mm was chosen for the third series of experiments, mainly due to dosing conditions. In this experiment practically the entire tungsten sample oxidised with the yield of metallic tungsten being on average less than 2%. Compared to the first two experiments where the tungsten recovery yield was between 34 to 44% there was a significant loss of metallic tungsten. This wasn't just due to an aggressive alkaline molten salt environment but also to the presence of air which is standardly dosed into the apparatus together with the waste in the MSO process.

Keywords: detritiation, tungsten, molten salt oxidation, waste management, recycling



POSSIBILITIES OF REPROCESSING SOLID WASTE WITH TRITIUM FROM FUSION USING HIGH-TEMPERATURE HEATING

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The types of waste with tritium generated during nuclear fusion are evaluated.

Some methods of reprocessing and decontamination of solid waste using thermal processes are evaluated. The advantages and disadvantages of different methods are compared.

The selected high-temperature technology is intended for use in the EU DEMO project. It will be in the area where waste from nuclear fusion processes will be processed. The technology is also evaluated from the point of view of safety and the environment.

Solid wastes of various sizes are investigated with regard to their possibility of detriation. The focus is on wastes containing mainly tungsten grains of various sizes.

The possibilities and reasons for the use of high temperature technologies are investigated. The tests are mainly focused on waste with tungsten in powder form in various atmospheres. The problems related to induction heating and induction melting of metals and non-metals are monitored.

Keywords: Detritiation, dehydrogenation, EU DEMO fusion reactor, waste management, dust waste, tungsten, induction heating, induction melting in cold crucible, safety.



DEVELOPMENT OF HYBRID PROCESS FOR REMOVAL OF TRITIUM IN CONTAMINATED WATER

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At Wolsong NPP in Korea, a Tritium Removal Facility(TRF) was installed in 2007 to separate and remove tritium existing in the system. However, TRF is a process to remove HTO from D_2O from pure water, suitable only for heavy water with high tritium concentrations, and not suitable for surface water or groundwater contaminated with tritium and seawater caused by serious accidents such as the Fukushima nuclear accident. Therefore, in this study, four core technologies with potential for development were selected to select the elemental technology field of pilot facilities for treating ³H, and specialized research teams from four universities are conducting technology development.

The first technique is a zeolite membrane that is being studied at the K. University. The 3 H removal efficiency can be increased by substituting the Na⁺ ions in the pores with K ${}^{+}$ or Ca^{2+} ions to finely control the pore size of the zeolite membrane. The separation coefficient for D₂O or HTO was confirmed after ion exchange to confirm that there was a possibility of HTO separation. In HTO removal experiments of 40,000Bq/L by synthesizing a Linde Type A membrane, the removal efficiency was about 7% at one stage, and research is continuing to increase the processing efficiency by 60% with a large amount of stage design. The second technology is metal oxide and electrochemical treatment, which is being studied at KJ University. The researchers created a ³H removal device that included redox-based electrochemical multichannel membranes and filled the feed channel with ion exchange resins. During the decomposition of water and ³H, ³H is ionized and separated into two electrodes according to the electrochemical charge characteristics and removed (HTO \rightarrow T⁺ + OH⁻, HTO \rightarrow H⁺ OT⁻) [1]. As the number of carbon stacks increases, the removal efficiency gradually increases, and when acid is added, the removal efficiency may increase to about 30%. The third technique is tritium removal technology using hydrophilic inorganic adsorbents, which is being studied at the IH University. Boric acid and melamine precursors were used to synthesize an inorganic adsorbent BN consists of a layer of sp² hybridized boron and nitrogen atoms alternating in a honeycomb-shaped lattice, which are joined by the Vandervaals force[2]. The adsorption performance of BN at the removal target of 40000Bg/L was evaluated and it was confirmed that 43% of ³H adsorption was achieved. The final technique is the use of an aluminum-attached ion exchange resin in progress at GIST in Gwangju. This technology attaches an aluminum cation to the SO³-group of sulfonated PS-DVB resin, in this process, the ³H removal



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efficiency of 8% was shown in the single-column process, and it is expected that the ³H removal efficiency can be increased by increasing the length and quantity of the column.

After that, Elim Global intends to design a hybrid composite process by supplementing the problems in the operation of a single process, such as energy efficiency and processing capacity, and to fuse it with the pre-treatment process technology that has been carried out in prior research.

Keywords: tritium separation, adsorption of tritium, ion exchange resin, inorganic adsorbent, electrolysis

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DETRITIATION OF THE ORGANIC MATERIAL IN THE MSO PROCESS

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Tritium is a radioactive isotope of hydrogen, and due to its' high price, it is necessary to recover it from tritiated materials via detritiation technologies. The number of tritiated materials arises during fusion. The tritium from fusion reactors is deposited in the outer layer of plasma-facing materials, which must be dealt with during the maintenance or decommissioning processes. The secondary produced waste as laboratory equipment could also be contaminated with tritium. High-temperature treatment in an oxidation environment can achieve the release of tritium from metals or organic materials. The tritiated vapour is then captured in a series of water bubblers and reprocessed into a pure T_2 . One of the high-temperature methods is Molten Salt Oxidation (MSO), which uses high temperatures, alkaline salt, and oxidative environment for flameless oxidation of different types of waste.

This work aimed to simulate the detritiation processes of tritiated organic materials in MSO technology. Firstly, a series of experiments with D_2O absorb in ion resins was conducted. The organic waste was decomposed within the molten salt, and the flue gas was measured to determine the oxidation efficiency. The D_2O was captured in a series of water bubblers, and the water was then analysed with ATF-FTIR. The results showed that all deuterium in the form of D_2O was caught in the first water bubbler. The capture efficiency ranged from 22.32 % to 61.13 %. The lower efficiency capture can be explained as D_2O in water can form an HDO molecule, which wasn't possible to quantify correctly.

The second type of experiments was carried out with T_2O with an activity of 851 Bq and with tritiated oil with an activity of 2495 Bq. The T_2O was added to the set amount of ion resins and dosed into MSO. A peristaltic pump dosed tritiated oil. The flue gas was measured to determine oxidation efficiency, and the T_2O was captured within water bubblers. The water was analysed with Liquid Scintillation Spectroscopy. The results of the capture efficiency of the T_2O are shown in table 1. The higher yield in the third experiment could be explained that the tritium was captured in the soot on the walls of the reactor during the second experiment and was subsequently released during the third experiment. The capture efficiency is sufficient for the detritiation experiments. The results showed that this technology is suitable for the detritiation of tritiated organic materials.



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Experiment	Type of organic material	Activity (Bq)	Activity in first bubbler (Bq)	Activity in second bubbler (Bq)	Yield efficiency of T ₂ O (%)
1	ion resins	851	719.41	113.46	97.87
2	tritiated oil	2495	1478.2	355.8	76.42
3	tritiated oil	2495	3029.4	258.5	136.99

Keywords: Detritiation, Molten Salts, Waste Management, Tritium Analysis





Water and air detritiation





COMPARISON OF AWD TO CECE FOR ITER SCALE WATER DETRITIATION

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Tritium is used as a fuel in nuclear fusion and water detritiation is an important part of the overall fusion fuel cycle. This paper compares two competing technologies for an ITER scale water detritiation reactor, namely Advanced Water Detritiation (AWD) and the Combined Electrolysis and Catalytic Exchange (CECE) process. The processes are compared in terms of equipment size and footprint, energy demand, isotope separation characteristics, safety, and technology readiness level. An important technical concern discussed is management of deuterium accumulation since deuterium is enriched along with tritium and D-T separation is inherently more difficult than H-T separation. Interfacing with a downstream Isotopic Separation System (ISS) is also discussed.

Keywords: Tritium separation, LPCE, CECE, AWD, Deuterium, ISS.



MATHEMATICAL AND NUMERICAL MODEL FOR DYNAMIC SIMULATION OF LIQUID PHASE CATALYTIC EXCHANGE PROCESS WITH COMBINED ELECTROLYSIS FOR *ITER*

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For the Water Detritiation System (WDS) at ITER, a very specific unit operation is planned to be used, in which tritium is recovered from tritiated water using liquid-phase catalytic exchange (LPCE) process combined with electrolysis process. In the fusion reactors like JET or ITER, a considerable amount of tritiated waste-water is accumulated in various subsystems during operation and maintenance. To ensure the emission of tritium to the environment remains below very strict limits, the WDS is responsible for the recovery of tritium from moderately tritiated water and release of protium (and deuterium) to the environment.

The LPCE column, along with the electrolysis unit operation, is called the combined electrolysis and catalytic exchange (CECE) process. In CECE process the LPCE column can have two types of configurations of catalyst and packing arrangements in the column: trickle and segregated. For the system design as well as to make a choice between the two configurations for the column design, there is a requirement for parametric optimization to identify suitable process design and operating parameters.

The work presented here derives the analytical model which can captures the dynamic hydrogen isotopic distribution in the liquid, gas and vapor phases moving along the height of the column. The system considered, is based on counter-current phase flows, with liquid flowing from top to bottom while, gas and vapor flow in upward direction, counter-current to the liquid flow. Equations are valid for trace tritium and a full range of deuterium isotopic composition in the column, fulfilling one of the main requirement for ITER WDS design. The proposed model is stand-alone and flexible to be used in any environment as per the user's choice. The same set of equations can be used for the simulation of both the type of configuration of catalyst and packing in the LPCE column. The governing equations can be used to simulate the performance of the column with combined electrolysis and for optimization of the process parameter for the WDS system design. The simulation result of a case study is presented in this work to demonstrate compliance with given performance requirements of the WDS, thereby verifying the authentication of the governing equations used for the numerical simulation of the system.



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Developed model demonstrate the detritiation factor (DF) as 0.92e7, which is in very close conformance with WDS's DF 1.14e7.

Keywords: Tritium separation, detritiation, fusion reactors, ITER, LPCE, CECE, Electrolysis, MATLAB, Dynamic Model, Detritiation Factor.

"The views and opinions expressed herein do not necessarily reflect those of the ITER Organization."



COMPACTED MIXED CATALYTIC PACKING FOR WATER DETRITIATION

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In order to remove tritium from heavy water, used CANDU fission reactors or produced in ITER fusion reactors an effective and efficient technology for heavy water detritiation it is requested. One of the most studied and promising technology is liquid phase catalytic exchange (LPCE) coupled with cryogenic distillation of hydrogen. In LPCE process, the isotopic exchange take place in two steps:

$$H_{2}(gas) + HTO (vapor) \leftrightarrow HT (gas) + H_{2}O (vapor)$$
(1)

$$\frac{\text{H}}{2} \underbrace{O(\text{vapor}) + \text{HTO}(\text{liquid})}_{2} \leftrightarrow \text{HTO}(\text{vapor}) + \underbrace{H}_{2} \underbrace{O(\text{liquid})}_{2}$$
(2)

$$H_{2}(gas) + HTO (liquid) \leftrightarrow HT (gas) + H_{2}O (liquid)$$
(3)

Reaction (1) is a catalytic reaction and needs a *hydrophobic catalyst*, while the second process (2) is a conventional water distillation process and needs an efficient hydrophilic contact element. Often in practice is used a mixture in various ratio between catalyst and hydrophilic packing is so called *"mixed catalytic packing"*. In LPCE process, mixed catalytic pacing it's key element for the number and the sizes of LPCE columns and for the efficiency of the tritium separation process. Usually, the mixed catalytic packing can be arranged in random or ordered (alternated beds) structure and the inner geometry of LPCE column depend of the hydrophobicity of the catalyst.

The present paper it is focused on the last achievements and improvements of previously COMPACK C-P 001 packing, irrespective a more compact mixed catalytic packing and on a double activation procedure for increasing the wettability of packing. In the last developed mixed catalytic packing, called COMPACK C-P 002, the improved hydrophobic catalyst Pt/C/PTFE is manufactured with the compatible sizes with the sizes channels of hydrophilic packing and it is inserted in a certain ration on free channels inside of hydrophilic packing so that the height of LPCE columns is reduced to the necessary height hydrophilic packing. To increase the wettability of hydrophilic packing firstly a special chemical treatment and then a thermal treatment in controlled atmosphere have been applied. Chemical treatment increases the external surface (roughness) of metallic wire of metallic gauze and the thermal treatment has as effect formation of metal oxides more hydrophilic. Initial thickness and wettability of wire have been measured for untreated and treated materials as well as weight loss after the chemical treatment with three solutions of various composition at different duration and temperatures. A similar procedure has



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been applied for thermal treatment. Experimental results showed a significant increasing of wettability of improved packing.

Taking in consideration the significant advantages of the improved COMPACK C-P 002 packing, this was patented and selected to be applied in LPCE columns of the future Cernavoda Tritium Removal Facility.

Keywords: tritium separation, heavy water detritiation, mixed catalytic packing; hydrophobic catalyst

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THE STUDY OF THE CATALYTIC ACTIVITY USING A DIRECT-FLOW REACTOR

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The liquid phase catalytic exchange (LPCE) of hydrogen isotopes between hydrogen and water has proven itself well as an excellent process for detritiation and deprotiation of heavy water. For example, the LPCE process is used to maintain the required isotopic composition in reactors such as PIK or CANDU [1, 2]. To carry out the isotopic exchange process, it is necessary to use hydrophobic catalysts for the molecular activation of hydrogen. Currently, catalysts for this process are produced in Canada, Belgium, Romania, India, Korea and Japan. In Russia the hydrophobic catalyst RCTU-3SM is used – Figure 1, in which the platinum is supported on a styrene divinylbenzene copolymer (SDBC). This catalyst has been successfully operated for more than 25 years in the EVIO pilot plant located on the territory of the National Research Center "Kurchatov Institute" – PNPI [3]. This served as the basis for its use for isotopic water purification in the heavy water loop of the PIK reactor in the tritium removal facility (TRF) under construction. The process of heterogeneous isotope exchange between liquid water and hydrogen, which occurs on a hydrophobic catalyst, is rather complicated. The study of the catalyst is essential to optimize the process. An experimental setup was developed and assembled in the Hydrogen Isotope Separation Laboratory to study the process of co-current catalytic isotope exchange between hydrogen and water vapor on a layer of RCTU-3SM catalyst. The setup allows us to carry out experiments at different ratios of co-current flow values, temperatures, relative humidities, and of different flow isotope compositions. A description of the experimental setup, as well as the results of the experiments are presented. We have determined that internal diffusion limits the rate of reaction and proposed the measuring of catalyst activity by mass transfer coefficient *Kc*, that is the same for any isotope of interest (H, D or T) and acceptable in a wide range of deuterium concentration.

Keywords: Hydrogen isotope separation, hydrophobic catalyst, liquid phase catalytic exchange, direct-flow reactor, tritium, deuterium, activity constant.



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Figure 1. Hydrophobic catalyst RCTU-3SM

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PLANNED OPERATIONS OF THE SPARC'S TOKAMAK EXHAUST PURIFICATION SYSTEM

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The SPARC device is a deuterium-tritium fueled fusion tokamak, designed to operate with a 10g tritium inventory. As part of tritium emissions management, the SPARC facility has a water detritiation system that uses combined electrolysis and catalytic exchange (CECE) technology, a process with a successful track record in the production of heavy-water, and water detritiation for both the fusion and fission industries. The CECE process concentrates and separates hydrogen isotopes, enriching heavier isotopes in the working liquid [1], while discharging lighter isotopes as effluent. This system offers a high detritiation factor $(10^7 - 10^8)$, for processing tritiated water [2]. Using this approach, virtually all tritium is contained within the fuel cycle, while the system discharges gases, primarily protium, with activities on the order of 40 kBq/m³ (1 µCi/m³).

Minimizing tritium inventory in the CECE system is part of a broad strategy that allows SPARC to operate with low on-site inventory. Commercially available electrolyzers are not designed with the foresight of tritium operations, and typically operate with substantial liquid volumes, which lead to excessive tritium inventory. These large tritium inventories highlight the need to determine the minimum viable liquid volume in a electrolyzer that allows for unhindered operation but also minimizes tritium inventory. In this work, we design a bespoke alkaline electrolyzer package that minimizes liquid electrolyte volume and tritium inventory.

Electrolyzers require cooling to dissipate heat generated by inefficiencies in the electrolysis process. Typically, this cooling is provided by flowing feedstock in significant fractional excess (> 250), at high recycle ratios (> 0.99). Flow can be established actively, through means of a mechanical pump, or passively, through gas-lift; a two-phase flow phenomena that is crucial to the operation of passively cooled systems. The liquid flow induced by gas-lift is the primary means of cooling the electrolyzer. Gas-lift is commonly used on commercially available alkaline water electrolyzers and is of particular interest in water detritiation applications since the configuration eliminates a mechanical pump, a rotating shaft, and the associated leak and failure points.

In this study, we identify the key hydraulic parameters pertinent to gas-lift in the CECE process; firstly, the operating pressure, and secondly, several critical dimensions including submergence, lift, and tube diameter. We carry out sensitivity analysis to understand the effects of each parameter. Submergence and tube diameter are positively



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correlated with recirculating liquid flow, while operating pressure and lift are negatively correlated with recirculating liquid flow. Increased liquid flow results in increased cooling and thermal performance.

For SPARC operations, we determined that gas generation of 12.8 $\text{Nm}^3 \text{Q}_2/\text{hr}$. and 6.4 $\text{Nm}^3 \text{O}_2/\text{hr}$. (85% of name-plate gas production) induce a recirculating liquid electrolyte flow rate of approximately 40 liters per minute, which is used to cool the electrolyzer. This liquid flow keeps the temperature rise across the electrolyzer below 10 °C. Supporting equipment, in particular gas-liquid gravity separators, are designed to minimize liquid working volumes, and the associated tritium inventory. Compared to a reference commercial unit that produces 15 $\text{Nm}^3 \text{H}_2/\text{hr}$., our bespoke design reduces the electrolyte volume by 70%, from 350L to less than 100L, while maintaining the same gas production rates. With a target electrolyte concentration of 1850 GB/kg (50 Ci/kg), the commercially available unit would require as much as 2.5g of tritium working inventory, while our design uses up to 0.65g, freeing up the balance (1.85g) for use elsewhere in the fuel cycle.

Keywords: Water detritiation, combined electrolysis and catalytic exchange, fusion, heavy water, tritium, fuel cycle

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APPLICATION OF HYDROGEN FUEL CELL TECHNOLOGY TO HYDROGEN ISOTOPE SEPARATION PROCESSES

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Within tritium separation or heavy water upgrading processes, such as the combined electrolysis and catalytic exchange (CECE) process, a trickle-bed recombiner (TBR) is often employed to convert the gaseous hydrogen isotopes and oxygen to water in an overhead recombiner system. It has been proposed to replace the TBR with a hydrogen fuel cell to perform the same task of converting hydrogen isotopes to water. The purpose of the recombiner or fuel cell in the CECE process is to react hydrogen with oxygen to form water $(H_2 + \frac{1}{2}O_2 \rightarrow H_2O)$.

The main driver to replace the TBR with a fuel cell in the CECE process is economics. The key differences between the two arrangements are the energy generated from the hydrogen-oxygen reaction in the TBR unit is in the form of waste heat, whereas the fuel cell can produce electrical power. Additionally, the TBR requires pure oxygen as a feed while the fuel cell can operate with a feed of air. It is expected that the replacement of the recombiner with a fuel cell will improve the process economics and simplify the overhead recombiner system.

The following paper will discuss the assessment of the conceptually simple change in the CECE process to determine if any unexpected issues exist. The process details were reviewed to assess the integration of the fuel cell in the process in place of the recombiner and no significant issues were identified. A series of experiments to evaluate the performance of a 25 cm² polymer electrolyte membrane (PEM) fuel cell with deuterium demonstrated that a fuel cell will produce more power with deuterium than hydrogen. With the process review and fuel cell experiments completed, a simple assessment of the economics of replacing the TBR with a fuel cell in the CECE process was performed. It was found that there is no significant difference in the capital costs, but there is significant reduction (in the order of 10%) in operating costs by replacing the TBR with a fuel cell.

Keywords: Tritium separation, detritiation, recombiners, fuel cell, hydrogen isotopes.



EXPERIMENTAL INVESTIGATION OF DISTILLATION PROCESS WITH LOW TRITIATED/DEUTERATED WATER ON HYDROPHILIC PACKAGE

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National R&D Institute for Cryogenics and Isotopic Technologies (ICSI) was established in 1970 as a research focused Industrial Pilot Plant. ICSI was created with the purpose to develop the heavy water production technology. This technology has been successfully transferred to the heavy water production plant. At present, research within ICSI is focused on support for the National Nuclear Program, hydrogen and fuel cells, cryogenics, environment.

One of the main research directions of our institute is represented by the hydrogen isotopes separation process.

Within the institute, complex research was conducted on the separation of hydrogen isotopes starting with the Liquid Phase Catalytic Exchange (LPCE) process and continuing with cryogenic distillation.

This paper refers to the experiments performed with deuterated water and water with low concentrations of tritium on the hydrophilic packing in conditions of total reflux.

The experiments were performed on a column equipped with two types of hydrophilic packing.

Keywords: Hydrogen isotope separation, water distillation.

Acknowledgement: This work was carried out through the "Nucleu" Program, ctr. no. 9N/2019, developed with the support of the Ministry of Research, Innovation and Digitalization, project no. PN 19 11 01 04 – "Innovative CECE process solution for promoting a new decontamination technology of liquid waste poorly concentrated in tritium and for recovery of deuterium"



DEUTERIUM STREAM MANAGEMENT LEAVING THE *LPCE* COLUMN IN A CECE PROCESS FOR HEAVY WATER DETRITIATION

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In heavy water detritiation using the combined electrolysis and catalytic exchange (CECE) process, deuterium leaving the Electrolyzer is fed to the bottom of the Liquid Phase Catalytic Exchange Column (LPCE) in which tritium exchanges between the tritiated deuterium gas (moving upward in the LPCE column) and D₂O liquid (moving downward in the LPCE column). Once the deuterium gas leaves the LPCE column, typically a Trickle Bed Recombiner (TBR) is used to convert the incoming deuterium gas into the heavy water. This article studies a different approach in which instead of using a TBR, an additional LPCE column is used. In the additional LPCE column, deuterium gas is scrubbed with demineralized light water. This process alternative has many potential advantages over using a TBR. Firstly, oxidation of isotopic hydrogen is highly exothermic and thus requires a separate water-cooling circuit in order to maintain the temperature within TBR. Secondly, a TBR requires a relatively complex internal design to ensure proper distribution of the gas otherwise catalyst burn-up may occur. Using an LPCE column instead of a TBR eliminates these complications. This paper presents a high-level layout of the process plant in which an LPCE column is used instead of a TBR. Column modeling is also presented and effect of different parameters (such as G/L ratio, column temperature, etc.) on deuterium content leaving the LPCE is also discussed.

Keywords: Tritium separation, LPCE, CECE, Trickle Bed Recombiner, Deuterium, Detritiation, Heavy Water.



OXYGEN STREAM MANAGEMENT IN A CECE PROCESS

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In the Combined Electrolysis and Catalytic Exchange (CECE) process, the Electrolyzer produces both hydrogen and oxygen streams. Hydrogen is typically fed to bottom of the Liquid Phase Catalytic Exchange Column (LPCE). The oxygen stream however is processed and afterwards either fed back to the Trickle Bed Recombiner (TBR) in heavy water detritiation or released to the exhaust stack in light water detritiation. This article discusses in detail handling of the oxygen stream both in heavy and light water detritiation CECE processes. Oxygen leaving the Electrolyzer has a trace amount of tritium gas in it as well as well as water vapor (due to diffusion across the membrane). Trace tritium is converted to vapour using a catalytic converter and then either scrubbed using an Oxygen Vapour Scrubber (OVS) or captured in a dryer bed. This study analyses and compares the different options for handling the oxygen stream in a CECE process.

Keywords: Tritium separation, LPCE, CECE, Trickle Bed Recombiner, Deuterium, Detritiation, Heavy Water, OVS.



STUDY ON TRITIUM PERMEATION FROM PRIMARY WATER COOLANT INTO SECONDARY WATER COOLANT for *JA-DEMO*

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JA-DEMO plans to use two high-pressurized and high-temperature water coolants. Due to the high tritium mobility in high-temperature metal, tritium will inevitably transfer from the plasma side to the primary coolant and from the primary to the secondary water coolant. From the viewpoint of fuel control, tritium safety, and social acceptance of commercial fusion plants, it is compulsory to correctly understand the tritium permeation behavior through some experiments and modeling of permeation phenomena, which improves the accuracy of tritium behavior prediction. Therefore, in order to assess the tritium permeation amount and rate from primary to secondary and validate water detritiation system performance based on the recent JA-DEMO R&D activities [1], this study conducted an experiment that mocked protium permeation from the secondary water coolant to the first water coolant to consider the isotope effect and calculated tritium amount in the secondary water coolant and compare the obtained figures with the numbers of the existing water detritiation systems based on the previous studies [2-3].

The protium permeation from the distilled water confined in the Inconel tube to the gas phase was quantified with gas chromatography. As a result, the obtained protium permeation flux was higher than the tritium permeation by seven orders of magnitude. Based on the previous work [2], the concentration gap between protium and tritium was nine orders of magnitude. This result implies that when it comes to the two-way hydrogen isotope permeation between water coolants, tritium permeation was nearly proportional to tritium concentration in the primary coolant, even though gas-driven tritium permeation is theoretically proportional to the square of tritium concentration. Based on obtained experimental facts, the mass transfer coefficient for tritium permeation from the primary to the secondary coolant was defined by the assumption on the tritium permeation flux was proportional to the tritium concentration, 1.45×10^{-11} [m/s]. Based on the previous work [3], the tritium permeation rate from the plasma side to the primary water coolant through the first wall and divertor as well as cooling piping in the breeding zone was set to be 3.0 g/day. The time transition of tritium concentration in the coolants is shown in Figure 1, which explains that tritium concentration in the primary coolant will reach 1 TBq/kg, which is the safety guideline considering CANDU reactor after 100-day successive operation and that in the secondary coolant will reach 60 kBq/kg, which is the regulation value to release tritium contaminated water to the environment in Japan around 140-day continuous operation.

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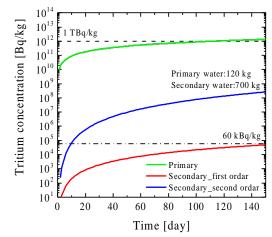


Figure 1. Ttime transient of tritium concentration in the water coolants

Keywords: Tritium, Blanket, Water coolant, Permeation, JA-DEMO

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WATER BALANCE AND WATER DETRITIATION REQUIREMENTS IN THE *EU-DEMO* FUEL CYCLE

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The European Fusion Programme for a demonstration fusion power plant (DEMO) has entered the concept design phase. Driven by the need to reduce as much as possible the tritium inventory, the fuel cycle of EU-DEMO follows a novel three-loop architecture.

Next to the processing of the torus exhaust, the fuel cycle of the EU-DEMO is also tasked with processing tritiated streams that arise from the operation of the plant. For this purpose several dedicated systems are located in the Outer Tritium Plant Loop. Especially detritiation of air flows via wet-scrubber columns may lead to the generation of significant amounts of tritiated water. Therefore a Water Detritiation System (WDS) is foreseen including a Combined Electrolysis and Catalytic Exchange (CECE) process for the final detritation and pre-enrichment of tritium for isotope separation. As the detritiation of water is laborious and energy intensive, minimization of tritiated water generation and efficient treatment thereof is an important design criterion for the Outer Tritium Plant Loop of the demo fuel cycle. This paper gives an overview of different water sources projected to be present in the EU-DEMO fuel cycle. Based on a water balance the load on the WDS system is estimated for different scenarios.

Keywords: DEMO, Fuel Cycle, Water Detritiation.



ASSESSMENTS RELATED TO ISOTOPIC EXCHANGE PROCESS DURING RMSB REGENERATION FROM TER HCPB

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One of the main challenges of the DEMO reactors is to prove the tritium selfsufficiency operation. Therefore, a tritium breeding rate (TBR) higher than 1.05 shall be achieved. Currently, in EU two types of breeding blanket types are investigated, water cooled lithium lead (WCLL) and helium cooled pebble bed (HCPB), and the associated tritium extraction and recovery systems (TER) are under development and proof of their performances.

In the case of HCPB option, the TER consists of a helium loop containing hydrogen and steam up to 100 Pa that removes the tritium from the pebble beds. Following the purging of the breeder blanket, the purge stream will be diverted through the TER to remove tritium from the helium stream and recycle helium back to the purged blanket. The process of the HCPB TER is based on the trapping/adsorption of Q2O on the RMSB (reactive molecular sieve bed) and the adsorption of Q2 on the CMSB (cryogenic molecular sieve bed) at 77K or alternatively tritium adsorption on getter beds. Tritium shall be recovered from the RMSB via catalytic isotope exchange (isotopic exchange between a purge gas H_2/D_2 and Q2O and from the CMSB/getter beds during regeneration of the beds. The RMSB regeneration consists of two stages. In the first stage the adsorbed Q2O shall be detritiated by isotopic exchange with a dedicated H_2/D_2 loop. The hydrogen/deuterium stream containing tritium will be processed in close loop in connection with Tritium Plant. The second phase is the removal of the trapped Q2O by circulating over the RMSB a purging helium stream while the desorption of the Q2O is provided by heating the RMSB.

In view of calculations of the amount of hydrogen, flow rate as well, that is required as swamping gas for promoting the isotopic exchange with the tritiated water adsorbed on the RMSB, a model for the isotopic exchange vapors/gas have been developed and will be presented.

The paper aims to present the results of the simulations in order to evaluate the mass transfer coefficients for the isotopic catalytic exchange process, based on experimental data from a mock-up system, and finally to provide a strategy for RMSB regeneration aiming to achieve tritium recovery efficiency.

Keywords: Tritium recovery, isotopic exchange, adsorption, regeneration, simulation.





Trítium processing (purification, isotopic separation, cryogenic distillation)





RECENT DEVELOPMENTS OF HYDROGEN ISOTOPE SEPARATION AND ENRICHMENT BY THE THERMAL CYCLING ABSORPTION PROCESS AT *CNL*

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Removal of tritium is extremely important in most CANDU and other types of nuclear reactors that generate substantial amounts of tritium by neutron capture. Furthermore, an effective hydrogen isotope separation is essential in fusion reactors, where deuterium and tritium are used as nuclear fuel. The thermal cycling absorption (TCA) process, which is a version of displacement chromatography separation technology, is viewed as a compact, scalable and, a viable economical alternative for tritium removal and enrichment in certain applications in fusion start-ups and small/medium size accelerators

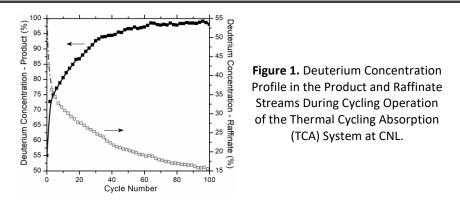
Canadian Nuclear Laboratories (CNL) has been actively advancing this technology with the aim of developing robust and promising high performance absorbents to facilitate the separation process, as well as optimizing the process to reduce cost and improve its operation. In addition to experimental studies, a computational fluid dynamics (CFD) based model has been developed to help understand the fundamental physics of the process and select absorbent materials to be used in the separation system.

In this paper, we present some recent experimental findings for deuterium/protium (D/H) separation, in a high-pressure (*ca.* 1000 kPa(g)) TCA system, which was used as a surrogate for tritium/deuterium (T/D) separation. CNL's system has demonstrated that high levels of deuterium enrichment (Figure 1) can be achieved by using a Pd-based absorbent produced by a CNL proprietary formulation. Various operating parameters were studied, both experimentally and numerically, and their impact on the separation efficiency was evaluated. Good agreement between the predicted and experimental deuterium concentration profiles was observed. The performance of the D/T TCA system is comparable to other TCA systems in operation. The experimental data collected to date, in conjunction with the modeling results, have been used to guide the preliminary design of an advanced TCA system for tritium separation, which is planned to be installed at CNL's licensed Tritium Facility by 2022. Preliminary modeling results of this system, for T/D separation, will be presented and discussed.

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Keywords: Tritium, deuterium, protium, isotope separation, enrichment, thermal cycling absorption process.



TRITIUM SEPARATION FOR MEDICAL ISOTOPE PLANT

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SHINE Technologies is building a medical isotope production facility to manufacture molybdenum-99 which decays into technetium-99m and other medical isotopes used in imaging procedures. The main production facility will be equipment with eight sub-critical irradiation units and a radioisotope production facility with three hot cells for moly-99 separation and purification. The technology relies on a sub-critical fission process in the irradiation units. The main components of the irradiation units include a neutron driver, a target solution, and a light-water pool surrounded by a biological shield. The light water pool for the target solutions and acts as a neutron reflector. The neutron driver comprises an ion source which accelerates deuterium atoms into a tritium gas target to generate 14 MeV neutrons generated by D-T fusion reactions.

The purity of the gas target is maintained by continuously siphoning a fraction of the tritium downgraded with deuterium, fusion reaction products, and protium arising from tritium interactions with the process lines. The impurities are segregated from the stream with a palladium-silver permeator. The protium and deuterium components in the hydrogenic stream are extracted using a Thermal Cycling Absorption Process (TCAP)¹ system. Preliminary, at scale, measurements demonstrate that high purity tritium and low activity raffinate are possible using protium and deuterium as the surrogate isotopic species. At scale tests using a tritium feedstock are envisioned for the summer of 2022.

This paper will discuss the performance of the TCAP system using protium and deuterium at the throughputs that are relevant to the production facility.

Keywords: Tritium separation, Thermal Cycling Absorption Process (TCAP)

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EXPERIMENTAL INVESTIGATION OF CECE PROCESS TO RECOVER TRITIUM AND DEUTERIUM FROM LOW TRITIATED/DEUTERATED LIQUID WASTE

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A current concern of researchers from the Experimental Pilot Plant for Tritium and Deuterium Separation (PESTD) within ICSI Rm. Valcea represents the promotion of an innovative solution of CECE isotopic separation process (Combined Electrolysis Catalytic Exchange), part of a new liquid waste decontamination technology.

Isotope separation by electrolysis is performed with a PEM (proton exchange membrane) electrolyzer, reconfigured for the nuclear field of tritiated water processing, in accordance with current regulatory body requirements (for nuclear field – CNCAN and for pressurized equipment – ISCIR). The new configuration includes software for electrolysis operation control and the catalytic isotopic exchange process takes place on a mixed catalytic packing of structured type, developed by ICSI.

In order to achieve the mentioned objective, an experimental installation was designed and built. This experimental installation, defined as module M1100, has interfaces with PESTD process and auxiliary systems and is placed within PESTD area. The installation has its own automation and control system that allows safe operation.

In a previous paper we presented modeling software for simulation of the nonsteady-state regime of the CECE separation process, specific to the deuterium/tritium isotopes concentration process in the liquid phase.

It has also been achieved an analysis for the concentration of various lowconcentrated tritium waste and shown the influence of the electrolyzer liquid holdup and the isotopic separation column holdup on concentrated water production.

The previous researches continued with those presented in this paper, the experimental investigations on the CECE installation in order to determine the separation performance of tritium, respectively deuterium.

There are presented two operating modes. The first mode is in "open circuit" to evaluate the individual separation performance on the two technological processes, water electrolysis, respectively water-hydrogen catalytic isotopic exchange. The second mode is



in "closed loop" which corresponds to the CECE isotopic separation process, the tritium/deuterium concentration in the water within the electrolyzer.

Experimental investigations were performed with low concentrated tritiated water (HTO) and low concentrated water in both deuterium and tritium.

The isotopic concentrations in the gas phase were measured at the exit of the electrolyzer and at several points on the catalytic isotopic exchange column. Measurements on deuterium were performed with a mass spectrometer, and on tritium in the liquid phase (by conversion to water on a bed of copper oxide), on a liquid scintillation analyzer.

The isotopic concentrations in the liquid phase were measured at the key points of the CECE installation, on deuterium they were measured on an infrared analyzer, and on tritium on a liquid scintillation analyzer.

The evaluation of the separation performances was made from the correlation of the measured data with the calculated ones provided by the software developed for the CECE isotopic separation process – the "open circuit" operating mode. For the "closed loop" operation mode, the data provided by the calculation program for the representation of the non-stationary CECE isotopic separation regime were verified. The results show a good correlation between the measured and the calculated data.

Keywords: Tritium separation, detritiation, combined electrolysis catalytic exchange, PEM electrolyzer

Acknowledgement: This work was carried out through the "Nucleu" Program, ctr. no. 9N/2019, developed with the support of the Ministry of Research, Innovation and Digitalization, project no. PN 19 11 01 04 – "Innovative CECE process solution for promoting a new decontamination technology of liquid waste poorly concentrated in tritium and for recovery of deuterium"



TRITIUM BEHAVIOR IN WATER AND GAS PRODUCED BY A FULLY TRITIUM COMPATIBLE HYDROGEN GENERATOR SYSTEM

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Enrolling in the research program of Cryogenic Plant for Tritium and Deuterium Separation, the water detritiation subsystem using Combined Electrolytic Catalytic Exchange (CECE) process has been developed. The CECE process uses tritiated hydrogen obtained by water electrolysis process to exchange tritium in a liquid phase catalytic exchange column. The electrolysis process is essential in the described process, which is why a special experimental program was designed to characterize a modified HOGEN H Series industrial hydrogen generator. The commercial equipment is a proton exchange membrane water electrolysis system furnished with one electrolysis cell able to deliver 2 Nm³/h of 99.999% pure hydrogen at a pressure of 15 bar gauge maximum. The major modifications were in the materials used in construction of the electrolizer, with special attention to obtain a low leak rate of the components and joints [1]. The tritium amount transferred to hydrogen gas, water enrichment factor, and number of hours necessary to attain stationary regime, were parameters of primary interest of the experiments.

Tritium activity concentrations of the electrolizer feeding water were around 180 Bg/l for the first experiment, around 1 kBg/l for the second experiment, and 1 kBg/l and 1.8% D_2O for the third experiment. The electrolyzed water was sampled every three hours for the first experiment, and tritiated hydrogen was sampled every 24 hours, after five days of the beginning of the experiment, when tritium activity concentration in electrolyzed water was relative constant (taking into account the measurement uncertainty). The other two experiments had different sampling approach. The electrolyzed water samples were sampled every three hours in the first five days, and every eight hours after the first five days. The Tritiated hydrogen was sampled every 24 hours from the beginning of the experiments. Hydrogen gas samples were collected using tubular reactor filled with copper oxide (CuO). The reactor was operated to 400°C, hydrogen gas reducing CuO to metallic copper, and the water vapour formed being then condensed in an alcohol-cooled trap to -40°C. The reactor operation to 400°C was established considering that the presence of water vapours can strongly inhibit the reduction reaction under 200°C [2] and the reduction of CuO to metallic copper is achieved after 350°C [3]. Tritium measurement was performed by liquid scintillation counting method [4] using TRICARB 2800TR liquid scintillation spectrometer. Counting time was 300 minutes (30min/repetition, 10 repetitions), the counting efficiency was around 30%, the background was below 11 Counts Per Minutes (CPM), conducting to an uncertainty around 6%.

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In order to minimize the necessary time for stationary regime, the hold-up of water electrolizer was chosen at the minimum value allowed for a safe and constant parameters operation in all experiments. The stationary regime was attained after 1200 hours, with an enrichment factor near 5, and an amount of 18% to 19% of tritium transferred from tritium enriched water to hydrogen gas. These parameters were obtained in all three experiments, the modelling software [5] of isotopes separation by electrolysis confirming the results.

Keywords: electrolyzer, Tritiated hydrogen, Tritium measurement, detritiation.

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D₂/H₂ SELECTIVITY IN LI-NA-*LTA* AND *CHA* ZEOLITES AT CRYOGENIC TEMPERATURE

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Separation of hydrogen isotopes can be of interest and achieved through selective adsorption in zeolites at temperature below 77 K. This separation process, known as quantum sieving, is based on the preferential adsorption of the heavier isotopes in micropores whose size is comparable with the kinetic diameter of hydrogen isotopes molecules (~0.29 nm). In our previous works we have showed that the aperture size of the zeolites and their composition play a key role in D_2/H_2 separation. [1-3]

In this talk we will report the novel results concerning the D_2/H_2 selectivity at 35 K – 77 K in two types of zeolites exchanged with lithium, sodium or calcium: LTA ($Li_{12}AI_{12}Si_{12}O_{48}$ and $Li_8Na_4AI_{12}Si_{12}O_{48}$) and CHA ($Na_{3.9}AI_{3.9}Si_{8.1}O_{24}$ and $Ca_{1.95}AI_{3.9}Si_{8.1}O_{24}$). D_2/H_2 equilibrium selectivity was measured by the co-adsorption technique combining manometry and mass spectrometry. Also, we recorded D_2 and H_2 single gas adsorption isotherms at different temperatures in order to calculate the isosteric enthalpy of adsorption.

For Na-CHA and Ca-CHA zeolites high equilibrium D_2/H_2 selectivity were obtained: 25 and 18 respectively (measurement conditions: 35 K, 600 hPa, 25% D_2 -75% H_2 mixture). These values are among the highest reported for zeolites under similar conditions. Moreover, they are obtained at high loadings (~10 mmol/g). The selectivity in CHA zeolites increases exponentially when temperature decreases as it was previously observed for other zeolites in our works. [2, 3] In contrast, for LiA and LiNaA zeolites the D_2/H_2 selectivity changes in a rather surprising manner. It increases down to ~55 K, but below this temperature it diminishes. Such a "bell-shaped" temperature dependence of D_2/H_2 selectivity in zeolites is observed for the first time. This effect can be correlated with a homogeneous energetic character of H_2 and D_2 adsorption in LiA and LiNaA zeolites which is suggested by a constant value of the isosteric enthalpy of adsorption for different loadings.

Keywords: Isotopic separation, cryogenic adsorption, quantum sieving, zeolites, cation exchange, D_2/H_2 selectivity.

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PROCESS MODELING OF LIGHT AND HEAVY WATER DETRITIATION FACILITIES

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For over 50 years, the Canadian Nuclear Laboratories (CNL, formerly Atomic Energy of Canada Ltd.) has been actively involved in developing technologies and facilities for detritiation of light and heavy water as well as for heavy water production and purification. During this time, a process model (HWPsim) has been developed for steady state conditions by CNL to help design such facilities. This document will review CNL's process model, and highlight some of the recent facilities that have been designed with the help of this model.

This model works by linking gas/vapour and liquid streams between equipment submodels to accurately predict the change in composition, flow, temperature and/or pressure of the relevant streams. Some of these sub-models include, liquid phase catalytic exchange (LPCE) columns, oxygen vapour scrubbers, condensers and dryers, gas phase and trickle bed H₂-O₂ recombiners, cryogenic distillation columns, and electrolyser stacks. Figure 1 provides an example of the model's ability to accurately reproduce measured tritium concentrations (within 2 % accuracy) in LPCE columns for detritiation factors (DFs) of 100 and 46,000. These data were measured at CNL's Combined Electrolysis and Catalytic Exchange for Upgrading and Detritiation facility (CECE-UD; Chalk River, Canada).

Other notable examples of facilities that CNL has been involved in designing with the use of the model include: the Combined Industrial Reforming and Catalytic Exchange plant (CIRCE; Hamilton, Canada), Wolsong Tritium Removal Facility (WTRF; Gyeongju, South Korea), NSSI CECE Detritiation Facility (Houston, USA), and the planned Heavy Water Detritiation Facility (HWDF; Chalk River, Canada). Sample performance metrics for these facilities include: DFs reaching 10^7 (HWDF), tritium concentrations reaching 99 % (WTRF), and throughputs reaching 100 kg-D₂O/h (WTRF).

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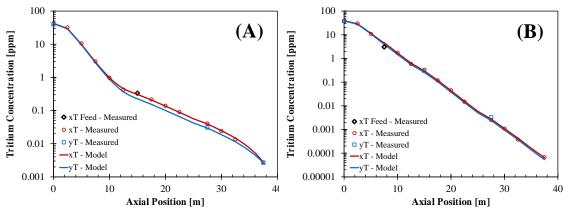


Figure 1 - Comparison Between the Measured and Modeled LPCE Profiles at a DF of (A) 100, and (B) 46000. The Axial Position Refers to the Position From the Base of the LPCE Column, and the Variables xT and yT in the Legends Refer to the Tritium Atom Fractions in the Liquid (xT) and Gas Phases (yT).

Keywords: Tritium separation, detritiation, CECE, exchange, simulation.

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STEP FUEL CYCLE DEVELOPMENT ACTIVITIES FOR A COMPOUND CRYOPUMP TO SEPARATE TRITIUM FROM SEEDING IMPURITIES FOR RECYCLE TO THE TOKAMAK

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The fuel cycle architecture for the Spherical Tokamak for Energy Production (STEP) considers a percentage recycle of the tokamak exhaust gas which bypasses tritium plant processing as proposed by the EU DEMO design team. Hydrogenic species are removed from the exhaust gas and routed to the fuel matter injection to be reintroduced into the tokamak. This separation provides two key benefits; firstly, there is a reduced inventory of tritium in the fuel cycle, and secondly, the tritium processing plant's size is reduced [1]. To achieve this, a modified version of a compound cryogenic pump [2, 3] which separates hydrogen and its isotopologues from impurities is considered. The modifications enable the pump to exhaust helium continuously, operate at temperatures no lower than the range between 10K and 20K, lower power consumption and shorter regeneration cycles.

To assess exhaust gas behaviour and their interactions with the cryopanels, a Monte Carlo simulation using PI-DSMC code offering multicore processing was performed. A 2D axisymmetric geometry was generated with the cryo-panel surfaces defined as full diffuse reflectors with a set tritium sticking coefficient. Only tritium, deuterium and helium were considered in the simulations to reduce computational resources. Initial results indicate that most deuterium and tritium is cryosorbed, whilst a fraction of helium remains stagnated in the pump. Figure 1 shows a cross-section of the pump and inlet reservoir with the tritium profile within the pump obtained in one of the simulations. These initial simulations suggest the feasibility of compound cryogenic pump in separating hydrogenic species from impurities.

A further study was carried out to identify the pumping requirements, spatial arrangements and thermal loads for the STEP concept design. The Monte Carlo simulations provided data to assess material balances and determine an optimum cryopanel area for species adsorption. Another numerical model was developed to calculate the optimum tritium recycling fraction during cryopump operation.

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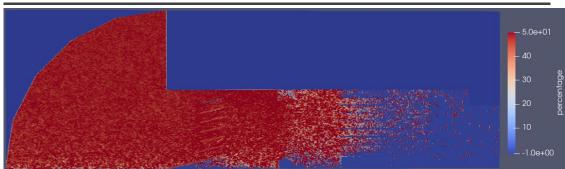


Figure 1. Monte Carlo results showing percentage of tritium left within the compound cryopump.

Keywords: Compound cryopump, recycle, selective pumping, STEP concept design, Monte Carlo simulation, PI-DSMC and tritium.

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CRYOGENIC IMPURITY REMOVAL FOR HELIUM AND HYDROGEN ISOTOPE PURIFICATION

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In tritium processing, impurities accumulate in the hydrogen isotope stream due to slow atmosphere in-leakage, off-gassing of components, line purging, and degradation of materials from contact with tritium over time. The bulk of the hydrogen isotopes can be removed via hydrogen permeators, but the hydrogen isotope concentration is too high in the permeator bleed to be discharged through the atmospheric stack. Also, in processing 3He the concentration of impurities dictates if the 3He will require additional purification prior to consumer use. Both streams can be purified through the use of consumable Zralloy getters, but this also generates a stream of radioactive waste. The purification could also be performed using a series of adsorption beds at varying temperatures to selectively capture impurities on each bed. The focus of this work is adsorption of nitrogen and hydrogen on a cryogenic adsorption bed. Several adsorbents have been screened for desorption profiles as a function of temperature for several gas mixtures of nitrogen, hydrogen, and helium. The bakeout conditions of the adsorbents was also tested to determine the effect of the presence of adsorbent-bound moisture had on the desorption profiles. Results will be presented on the adsorption capacity, adsorption profile, desorption profile, effect of bakeout conditions, and hydrogen retention for each adsorbent.



COMPARISON OF EXPERIMENTAL AND SIMULATION FOR SEPARATION OF HYDROGEN FROM INERT GAS USING DENSE METALLIC Pd₇₇Ag₂₃ MEMBRANES

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Hydrogen purification with dense metallic membranes such as palladium alloy membranes is a well-known technology. The high permeability and selectivity of such membranes for hydrogen lead to ultrapure H_2 [1]. Such production is necessary for several applications such as the production of ammonia and alcohols, development of clean energy sources and nuclear fusion reactor [2].

This study aims to investigate the best operating conditions of a $Pd_{77}Ag_{23}$ membrane to reach a low hydrogen content in the retentate. Two $Pd_{77}Ag_{23}$ diffusers units are used to separate mixture of hydrogen and inert gas such as He or N₂. The diffuser unit is composed of a dense metallic $Pd_{77}Ag_{23}$ membrane under hollow fiber configuration, placed into a thermal oven. A pilot scale apparatus has been set up in order to purify up to 200 L_{TPN} of gas mixture per experiment.

Permeability of the membrane has been measured with pure hydrogen and compared with the literature. Different operating parameters have been studied (gas flow in the retentate, transmembrane pressure, gas composition, feed flow rate...) and the consequences on hydrogen permeation flux, permeate composition and retentate composition have been systematically analyzed as well as the purification factor (representing the quantity of hydrogen in the permeate flow divided by the quantity of hydrogen in the feed flow). Additionally, a predictive model has been developed with Matlab to simulate the behavior of these Pd₇₇Ag₂₃ membranes including the concentration polarization effect. Finally, the modelling approach and experimental study have been compared.

Depending on the parameters, the purity of hydrogen is above 99,9 % and the purification factor above 97%. The experiment conducted evidences the impact of the concentration polarization on both diffusers units. This effect induced a limitation of the mass transfer, with a decrease of the permeate flux up to 70% depending on the operating conditions. The polarization concentration has been characterized with the mathematical model developed: the k-factor representing the mass transfer coefficient in the retentate side reached 10^{-2} to 10^{-3} m.s⁻¹ according to the diffuser unit.

Keywords: hydrogen purification, PdAg membranes, simulation



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EVALUATION OF BRAZE JOINTS FOR HYDROGEN PURIFICATION DIFFUSERS

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Hydrogen isotope purification technology uses a thin wall palladium – silver tube that is typically brazed into a manifold. The typical commercial hydrogen purifier uses relative short tubes and significant number of them to produce high purity hydrogen with little regard for the residual hydrogen in the raffinate. Our diffuser design requires both the product and raffinate streams to be as pure as possible, to extract as much hydrogen as possible with very low residual hydrogen in the raffinate. In the past, the diffuser vendor was responsible for both alloy selection and process development, that has left a knowledge gap. In order to fill this gap, this project was initiated to develop alloy process knowledge to compare alloys, to understand the processes, to better facilitate production of diffusers, and to own the technical requirements to produce diffusers that meet design goals and requirements. The diffuser operates between 350 and 430°C. The goal is to have a braze that 1) has with a melting temperature where the operating temperature is between 0.4 and 0.6 Th(abs), 2) is compatible with hydrogen, 3) can be vacuum furnace brazed, and 4) does not require flux. Results will be presented on the braze evaluation, parameter determination, and functional testing of the brazes.



CONTINUOUS CRYOGENIC PUMPING SYSTEM FOR FUSION NEUTRON GENERATORS

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Torion Plasma Corporation and SHINE Technologies have developed a continuous cryogenic pumping and purification system that will be used as the front-end of a fueling system for a gaseous deuterium-tritium fusion neutron generator.

The neutron generator has shown to continuously produce 14 MeV neutrons at a rate of 4.6×10^{13} neutrons per second through fusion of linearly accelerated deuterons driven into a low-pressure tritium gas target. The stream of deuterons dilutes the tritium gas target which reduces the overall neutron flux. Additionally, tritium induced isotopic exchange in the process loop is expected to contaminate the fuel with protons. To maintain the high neutron flux, the tritium gas target must be isotopically purified, and the fuel ratio re-adjusted for optimal performance. A neutron generator fueling system has been designed to continuously draw the low pressure (< 100 Torr) tritium from the remotely (> 25m) located target chamber, remove all gaseous impurities, isotopically purify the gas stream, and deliver 98% pure tritium back into the target chamber at a low flow rate (< 150 sccm.)

This presentation will focus on the frontend system that draws the low-pressure tritium, removes impurities, and delivers high pressure gas to the isotopic separation system. Cryogenic pumps coupled to a palladium permeator were selected as the core technology for reliability, to reduce moving parts and to limit permeation losses to the environment by avoiding mechanical pumps. A quad cryopump system was demonstrated to continuously draw low pressure hydrogen from a simulated chamber, permeate the gas across the palladium filter and pressurize the gas to 2,000 torr with a 30-minute duty cycle between each pair of cryopumps.

Keywords: Cryogenic pumping, tritium recovery and delivery, palladium permeation, DT fusion fueling.



HYDROGEN ORTHO PARA SEPARATION BY CRYOGENIC DISTILLATION

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Cryogenic distillation is commonly used for isotopologue separation [1]. Additionally, the nuclear spin isomers ortho and para of H₂, D₂ and T₂ can be separated. The latter can be used for the measurement of the separation performance or the height equivalent of theoretical plates (HETP). Due to the very high achievable separation performance, binary mixtures with components A and B have almost 100% of component A at the top and almost 100% of component B at the bottom of rather short columns. This is a backdraft for accurate measurements of the separation performance. To overcome this there are several strategies. One is to use many sampling points along the column and to adjust the total amount of both components A and B in such a way that a suitable gradient can be measured between at least two of the sampling points. However, this means that already in the design phase the column and the distribution of the sampling points have to be designed accordingly. The distillation of ortho and para on the other hand relaxes a lot of this. The gradient is much smaller all along the column and by this the separation performance and the HTP can be measured accurately.

The separation of ortho and para hydrogen also enables the production of samples above the thermal equilibrium. While it is straightforward to produce high purity para H_2 and T_2 (ortho D_2) by cooling it down and catalysing the conversion one cannot exceed the room temperature equilibrium of 75% ortho H_2 and T_2 (66% para for D_2). Samples above this equilibrium are of interest when studying fundamental interactions in the dense gaseous, liquid or solid phase [2]. Fiannly the combination with HD above the equilibrium [3] and highly accurate hydrogen and tritium samples [4] enables us at TLK to produce unique gas samples for a very broad range of applications.

In this contribution, we report about the experimental verification of ortho para distillation at the TLK including high precision in-situ Raman and IR spectroscopy for the measurement of the ortho para ratio of gaseous and/or liquid hydrogen isotopologues.

Keywords: ortho-para, isotope separation, cryogenic distillation, Raman spectroscopy, IR spectroscopy, tritium measurement and monitoring, TLK

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RECENT PROGRESS OF TRITIUM RELATED R&D ACTIVITIES FOR CFETR

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The Chinese Fusion Engineering Test Reactor (CFETR) is the next-step device for the development of fusion energy in China. The engineering design of CFETR was started in the middle of 2017, and has been successfully finished by the end of 2020. During the engineering design of CFETR, the tritium plant, as a main auxiliary system, has accomplished its detailed conceptual design. However, the feasibilities of the technology options in the detailed conceptual design still remain to be demonstrated from the engineering point of view. In the meantime of engineering design of CFETR, three dedicated tritium related projects aiming at engineering feasibility demonstration for CFETR tritium plant have been launched one after another since the middle of 2017. One of the projects is responsible for the establishment of a laboratory scale tritium plant demonstration system with multiple confinement barriers and for conducting tritium cycle experiments with gram level tritium. The other two projects are responsible for the establishment of a full scale inner fuel cycle demonstration system and a full scale outer fuel cycle demonstration system, respectively, and conducting experiments with protium/deuterium instead of tritium. In his paper, the most recent progress of these tritium projects, as well as some other tritium related R&D activities, and the next step tritium related R&D plans will be presented.

Keywords: CFETR, tritium plant, demonstration system, R&D activities.



DESIGN CONSIDERATIONS AND LATEST ARCHITECTURE OF THE EU-DEMO FUEL CYCLE INNER LOOPS

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The development of the fuel cycle for the European DEMO reactor has moved in 2020 from the pre-concept to the concept design phase. In this new phase, significantly more details on the design of the fuel cycle have to be included, including tritium control and tritium storage for all operational scenarios of the fuel cycle like start-up, steady state plasma operation, dwell time operation, plasma termination and fuel cycle shut-down.

This paper presents the design considerations behind the development of the Direct Internal Recycling Loop (DIRL) and the Inner Tritium Plant Loop (INTL) and explains the latest system architecture. Special focus is given on operational aspects during the different scenarios mentioned above and explains how it is intended to monitor and control the tritium composition. The discussion is done in a qualitative and quantitative way, wherever possible, based on the latest throughput numbers available for DEMO.

Keywords: DEMO, fuel cycle.



DETRITIATION AND RECOVERY OF VACUUM PUMP FLUIDS FOR COMMERCIAL FUSION POWER

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Current fusion test facilities, such as ITER, use cryopumps to produce the high vacuum required for commercial fusion power. However, cryopumps are not practical for commercial-scale fusion power due to the high energy requirements, tritium inventory, and capital costs [1]. In this work, we present a series of potential oils and a novel detritiation process which can potentially enable traditional oil-based vacuum pumps for commercial fusion power. Traditional oil-based vacuum pumps (roughing and diffusion) are ideal for maintaining the necessary vacuum for fusion power, but organic molecules, such as vacuum fluids, are damaged by exposure to tritium [2]. In particular, mineral oils, the most common vacuum fluids, are rapidly degraded by tritium, due to the highly energetic beta decay, isotope exchange, chemical reduction, and recoil triton effects [3]. However, some modern vacuum oils may be more resistant to tritium due to their highly aromatic chemical structure, which impart radiation and radical stability to the molecule [3]. By experimentally simulating tritium damage through the 4 primary modes of tritium degradation, an array of potential vacuum fluids was explored, and some of the tested vacuum fluids displayed significant resistance to all modes of damage. However, these resistant vacuum fluids still experience minor damage and represent a potential tritium inventory, so a novel EM-thermocatalytic process is being developed to simultaneously remove tritium through isotope exchange and targeted catalytic dehydrogenation.

Keywords: Vacuum, oil, detritiation, commercial fusion, decontamination, power cycle

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R&D ACTIVITIES ON TRITIUM TECHNOLOGY FOR *ITER* AND *JA DEMO* AT THE TRITIUM PROCESS LABORATORY

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The DEMO reactor is the next step after ITER, which aims to realize power generation from fusion energy by the middle of this century and to achieve a sustainable energy source. The challenges are to demonstrate the potential advantages of fusion reactors from the safety and environmental viewpoints and to develop self-sufficiency technology for tritium fuel. In a fusion reactor that aims to continuously burn deuterium-tritium as fuel, the fuel burn rate continuously supplied to the reactor is only a few percent at most. To recycle the unburned fuel gas, it is necessary to establish a closed fuel circulation loop (fuel cycle) that the process of separating hydrogen isotopes from impurity gases, separating hydrogen by isotope, and supplying it to the fuel system. This fuel cycle consists of a large chemical plant with many subsystems connected. Most of the knowledge on the operation of the ITER fuel cycle will be acquired in the late 2030s when the DT operation of ITER will be started. It is important to note the difference of the fuel cycle between ITER and DEMO as a chemical plant (Figure 1). Therefore, it is necessary to develop tritium-related technologies for DEMO in parallel with the construction and operation of ITER for the realization of DEMO in the middle of this century. The tritium technology group is developing the detritiation system (DS) for DEMO through the DS procurement activities of ITER, and developing the main subsystems of the fuel cycle for DEMO as follows. 1) Qualification test of the integrated performance of the ITER DS, which simulates the accident and abnormal events in the ITER facility intended to be reflected in the design of the DEMO DS; 2) Verification of the rapid hydrogen isotope analysis technique using the palladium membrane diffuser and the laser Raman method as a part of the technical study on the Tokamak Exhaust Processing System; 3) Technical study on the DEMO blanket tritium recovery; 4) Accumulation of characteristics data on tritium inventory in the plasma-wall interaction under the condition closer to ITER and DEMO to understand the behavior of tritium in the reactor analyzing the JET ILW-3 test sample, and numerical modeling of the DEMO fuel cycle for evaluation of tritium behavior in the fuel cycle, tritium inventory and initial loading requirement. In this presentation, the status of R&D toward the realization of the DEMO will be introduced.







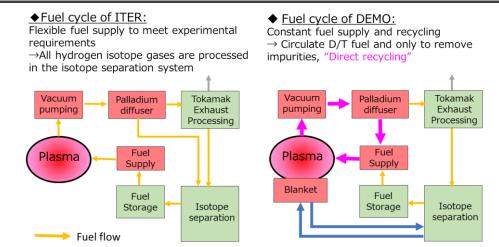


Figure 1. Schematic diagram of the fuel cycle of ITER and DEMO

Keywords: Tritium, detritiation system, fuel cycle, hydrogen isotope analysis, ITER, DEMO.



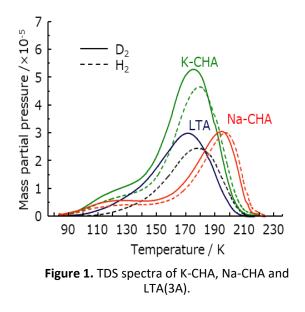
THERMAL DESORPTION OF HYDROGEN ISOTOPES FROM CHA-TYPE ZEOLITE

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There are only a limited number of techniques for the separation of the hydrogen isotopes hydrogen (H, or protium), deuterium (D) and tritium, including cryogenic distillation and the Girdler-Sulfide process. However, selective sorption or molecular sieving by microporous materials such as zeolites, metal organic frameworks (MOF) and covalent organic frameworks (COF) may provide simple and versatile approaches to hydrogen isotope separation [1]. Our group has previously reported that a CHA-type zeolite having an 8-membered ring structure exhibits D_2/H_2 separation at an adsorption temperature of 201 K [2]. In the present study, the H₂ and D₂ desorption abilities of CHA zeolites were investigated using thermal desorption spectroscopy (TDS).

A K^+ -type CHA zeolite (K-CHA) was prepared via a hydrothermal conversion method, using a mixture of an FAU-type zeolite (H-Y; Tosoh Corporation, Japan) and an aqueous solution of KOH. This K-CHA was subsequently ion-exchanged with Na⁺ using an aqueous solution of NaOH to produce Na-CHA. X-ray diffraction analyses confirmed that both the K-CHA and Na-CHA were successfully synthesized. A commercial LTA-type zeolite (3A, Tosoh Corporation, Japan) was also used in this study for comparison purposes.



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TDS was performed using a gas sorption apparatus designed in our own laboratory. In each trial, a sample (about 1.0 g) was loaded into a quartz cell and then held under vacuum (below 5.0×10^{-5} Pa) at 673 K for 2 h. After cooling to room temperature, the sample was transferred into a cooling bath at 77, 201 or 250 K and held at that temperature. At this point, gaseous H₂, D₂ or a mixture of H₂ and D₂ (50.7/49.3, volume/volume) was introduced into the cell to a pressure of 10 or 50 kPa and the specimen was allowed to adsorb the gas for 30 min. The specimen was subsequently immersed in liquid N₂ and any residual unadsorbed gas was removed from the cell. The pressure in the sample cell was then reduced to below 5.0×10^{-5} Pa (typically within 30 min). Following, the liquid nitrogen cooling bath was removed and the sample desorbed the isotopes while the extent of desorption was monitored using mass spectroscopy, to obtain TDS spectra.

Figure 1 shows the TDS spectra obtained from the K-CHA, Na-CHA and LTA(3A) following the adsorption of hydrogen isotopes at 250 K and 50 kPa. It is evident that, in each case, D_2 was desorbed beginning at a slightly lower temperature than H_2 . In addition, the peak temperature for D_2 desorption was somewhat lower than that for H_2 . As an example, the desorption peak temperatures for D_2 and H_2 when using the K-CHA were 172 and 177 K, respectively. This shift in the desorption peak can be explained by the faster diffusion kinetics of D_2 compared with H_2 [3]. These results suggest a kinetics quantum sieving effect that is dependent on both the structure and counter cations of the zeolites.

Keywords: Hydrogen isotope separation, zeolite, porous materials, kinetic quantum sieving

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INTEGRATION OF A MICRO-CHANNEL REACTOR IN A CECE PROCESS FOR H₂ REMOVAL FROM THE O₂ STREAM PRODUCED BY THE H₂ GENERATOR

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A Tritium Laboratory (TRI-Valcea) is under development on ICSI site from Rm. Valcea. The lab will serve as infrastructure for fusion associated research activities related to tritium and will enable a wide variety of experimental work like tritium processing and development of materials for detritiation processes. The capabilities of the lab will ensure an advanced tritium infrastructure to feed, recover, store and recycle tritium for safely managing the radioactive isotope.

One of the processes used to recover tritium from the lab infrastructure or experiments for re-storage is the thermal cycling adsorption process (TCAP), which is a semi-continuous chromatographic process for hydrogen isotope separation that uses metal hydrides as packing materials.

Based on the lab requirements for tritium recycling (e.g. "poor" tritium containing mixtures), throughputs and storage concentration for tritium, a TCAP process is in view for implementation. This paper will present the mathematical models and computer simulation programs in view for the design, calculation and optimization of the future TCAP operation.

Keywords: hydrogen isotopes separation, tritium, TCAP process, gas chromatography, mathematical models, computer simulation programs, heat and mass transfer.

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INVESTIGATIONS OF A MEMBRANE-COUPLED TEMPERATURE SWING ABSORPTION PROCESS FOR HYDROGEN ISOTOPE SEPARATION IN THE *EU-DEMO* FUEL CYCLE

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One processing step of the torus exhaust gas within the EU-DEMO fuel cycle is to reestablish the required DT ratio of the recycled unburnt fuel. For this purpose, an Isotope Rebalancing (IR) and Protium Removal (PR) unit is provided which has the advantage of avoiding a complete isotopic separation of the exhaust gas and furthermore keeping the total tritium inventory low. To achieve this, a membrane-coupled temperature swing absorption (MC-TSA) process was developed which combines isotopic separation via gaseous diffusion and metal-hydrogen-absorption.

In the TSA process stage two columns are used. Due to anticyclical heating, a hydrogen isotope mixture is transferred multiple times. Since the columns are filled with absorbing materials which have opposite affinities to the isotopes a separation is reached. The process performance depends on the operation parameters, i.e. temperature, pressure, gas composition and number of cycles as well as on the selection of the material couple. Objective of this study is to identify efficient operation limits and suitable material pairs using the test facility HESTIA (Hydrogen Experiments for Separation with Temperature Initiated Absorption). Additionally, preliminary material characterization studies support the material selection which is restricted to various requirements. These include reasonable absorption and desorption temperatures, pressures and fast kinetics as well as safety conditions. Furthermore, a distinct isotope effect during absorption is essential for the separation efficiency.

This poster will give an overview of the developed MC-TSA technology and the current status of process investigation. First results of analyses of possible material candidates and the resulting conclusions for the separation task will be presented.

Keywords: Hydrogen Isotope Separation, Metal-Hydrogen-Interactions, Hydrogen Absorption, EU-DEMO



ENHANCEMENT OF THERMAL CONDUCTION FOR THE WELDED CONDENSER OF A CRYOGENIC DISTILLATION COLUMN FOR HYDROGEN ISOTOPES SEPARATION

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In the cryogenic distillation columns used for the separation of hydrogen isotopes an important task is to enhance the thermal transfer from the cooling power throughout the column condenser to the vapours, from the column boiler, to cool and condense it's. It is obvious that the best candidate for a higher thermal transfer is OFHC copper which has an increase in thermal conductivity coefficient with decreasing of the temperature before to 4.2 Kelvin. Any new model of a condenser must be in accordance with safety regulations and among all, to fulfil the allowable leaks limits. The cryogenic distillation columns are made of 304L or 316L stainless steel grades, one of the most suitable metallic materials to withstand the safety requirements. Therefore, a dissimilar bond technique has to be developed for those two materials. In this sense can be used brazing or electronic welding, but, of great interest would be to find a more economical and faster solution to make such a dissimilar bound. In [1] are mentioned that good results have been obtained for the leak tightness of dissimilar welding, between 304L stainless steel and OFHC copper, using a GTAW (TIG) welding, with pre-heat of the copper between 200 and 540 Celsius degrees. This paper analysed how the welding of these different materials influences the thermal conductivity in the contact area, respectively the efficiency of heat transfer in the condenser. For this purpose, has been developed a cryostat to measure the thermal conductivity for different samples welded in similar conditions at the condenser. The results showed the evolution of the thermal conductivity of copper OFHC inclusive for the heat-affected zone (HAZ).

Keywords: Cryogenic distillation, thermal conductivity.

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PROTON EXCHANGE MEMBRANES CONTAINING MONOLAYER 2D MATERIALS FOR WATER ELECTROLYSIS AND ENHANCED HYDROGEN ISOTOPE SEPARATION

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The separation of hydrogen isotopes is important for enrichment of deuterium and tritium, which are useful resources for fusion technology. Furthermore, the tritium separation and management technologies have gained importance in recent years because a large amount of tritiated water has been generated after the nuclear accident in Fukushima.

Recently, it was reported that two-dimensional (2D) single-layer materials such as graphene or hexagonal boron nitride (hBN) have a remarkable proton conductivity, and the permeability of deuteron is lower than that of protons. [1] On that basis, monolayer 2D materials was considered to have potential applications for tritium separation. Although the deuterium removal from hydrogen gas was demonstrated by using a monolayer graphene [2], water electrolysis approaches that utilizing a proton exchange membrane based on monolayer 2D materials has received little attention. [3]

We envision an water electrolyzer containing proton conductive and tritium blocking 2D layer within the proton exchange membrane (PEM) for enhanced tritium separation and enrichment. We hypothesized that a monolayer 2D material coated on the PEM can reduce permeation of triton through the PEM, and it would improve enrichment of tritium in water and depletion of tritium in hydrogen gas. Furthermore, the superior gas barrier property of the monolayer 2D materials can potentially minimize degradation of perfluorosulfic acid (PFSA) membranes and a safety-related explosion issue which can be originated from oxygen and hydrogen gas crossover through the membrane. [3]

We synthesized composite membranes of PFSA coated with monolayer 2D materials, such as hexagonal boron nitride and graphene monolayers. Proton/deuteron conductivity and hydrogen gas permeability of the composite membranes were analyzed. Furthermore, effects of the monolayer 2D materials on water electrolysis performances and hydrogen isotope (deuterium or tritium) enrichment were investigated.

Keywords: Tritium separation, enrichment, water electrolysis, 2D materials, proton exchange membrane



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exchanger [1]



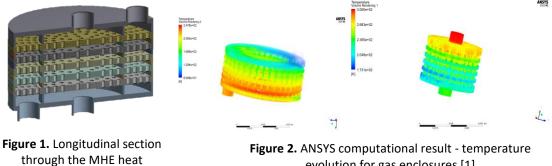
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DESIGN, FABRICATION AND TEST OF A PROTOTYPE OF MATRIX HEAT EXCHANGER FOR CRYOGENIC DISTILLATION OF HYDROGEN ISOTOPES

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To increase the efficiency of refrigeration, liquefaction, and cryogenic separation cycles, the heat exchangers must have high effectiveness doubled by high compactness, small temperature differences between incoming and outgoing flows must be ensured in order to increase efficiency; large heat transfer surface, relative to the volume of the heat exchanger, to minimize heat loss; there must be a high heat transfer rate to reduce the transfer area; small pressure drop to reduce compression costs; high reliability with minimal maintenance. All these properties are entirely fulfilled by the Matrix Heat Exchangers (MHE). This paper presents the results of the research program developed by the team of the Cryogenic Laboratory from ICSI Rm.Valcea, in order to achieve a proper solution for a matrix heat exchanger to be used for cooling the gas mixture at the entrance of a column for cryogenic distillation of hydrogen isotopes, running at low pressure and flows. The Matrix Heat Exchanger (MHE) prototype has been modelled using Ansys Fluent computational fluid dynamics. The heat exchanger mainly consists of a high-conductivity part made from perforated copper plates and a low-conductivity part represented by stainless steel inner and outer rings spacer, to separate the counter-current gas flows (figure 1). The temperature profile for gas flow zones (figure 2) shows its dependence on the way in which the holes are arranged on the plate, respectively their distribution on the active plate's surfaces.



evolution for gas enclosures [1]



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Within several experimental campaigns, different assembly and testing techniques of the MHE prototype had been performed, in order to achieve numerical data for the temperature and pressure drops along the heat exchanger and to verify the ANSYS Fluent numerical simulations results. The results revealed that the matrix heat exchanger prototype designed and tested has a high quality of heat transfer, low-pressure loss (within a few millibars), affordable dimensions and weight.

Keywords: cryogenic distillation, hydrogen isotopes, heat transfer, Matrix Heat Exchanger

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TRITIUM COMPATIBLE HYDROGEN GENERATOR CONSTRUCTION

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The electrolysers are used as hydrogen generators and represent one of the two key components of the CECE process for water detritiation. CECE process is foreseen to be implemented in both ITER and DEMO plant for water detritiation. The availability on the market for tritium compatible hydrogen generators is very scares. This paper reports on the construction of a tritium compatible hydrogen generator, concerning materials, joints and quality documentation.

Keywords: tritium electrolyser

Acknowledgement: This work was carried out through the "Nucleu" Program, ctr. no. 9N/2019, developed with the support of the Ministry of Research, Innovation and Digitalization, project no. PN 19 11 01 04 – "Innovative CECE process solution for promoting a new decontamination technology of liquid waste poorly concentrated in tritium and for recovery of deuterium"



INVESTIGATIONS ON He-3 – HYDROGEN ISOTOPES SEPARATION EMPLOYING Pd/Ag MEMBRANES

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Tritium resulted from separation processes is being stored on metal hydrides. In time, due to radioactive decay, tritium converts into helium-3, which accumulates in the storage vessel. The recovery of helium-3 is a topic of high interest due to its wide range of applications in healthcare, security and advanced research. Currently at ICSI is under development a method based on gas-chromatography, Pd/Ag membrane permeation and cryogenic distillation for helium-3 separation and enrichment having as sources both the cover gas of nuclear reactors and tritium storage containers.

This paper reports the investigation of using Pd/Ag membranes for helium separation from hydrogen isotopes by experimentally determination of the operating performances of the membrane in view of process integration.

Keywords: helium 3, tritium, gas chromatography, helium isotope separation

Acknowledgement: This work was carried out through the "Nucleu" Program, ctr. no. 9N/2019, developed with the support of the Ministry of Research, Innovation and Digitalization, project no. PN 19 11 01 04 – "Innovative CECE process solution for promoting a new decontamination technology of liquid waste poorly concentrated in tritium and for recovery of deuterium"



ENGINEERING DESIGN OF THE TRITIUM ADVANCED TECHNOLOGY FACILITY

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The UKAEA is building an advanced tritium processing facility, H3AT, at their site in Culham, UK. The facility comprises a tritium circulation loop which will provide a technical resource to test tritium fuel cycle process configurations for ITER and facilitate additional experiments in support of the UK fusion program.

Atkins has delivered the concept and preliminary design of all process systems. This poster describes the design process, including the development of process flow diagrams, process and instrumentation diagrams, mechanical systems, a dynamic process simulation, hazard and operability studies and control system deliverables.

A 3D model was developed in each stage of the design in increasing levels of detail. This was used to optimize layout and support operability and maintenance studies. Figure 1 shows a 3D visualization of a H3AT glove box used to support the design process.

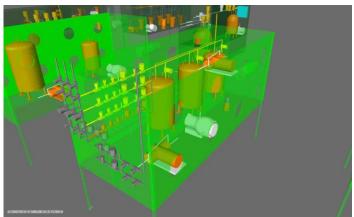


Figure 1 – Preliminary layout for H3AT glovebox

Keywords: Tritium separation, detritiation, plant layout, mechanical engineering, process engineering, containment, simulation



EXPLOITING THE SPECIFIC ISOTOPE-SELECTIVE ADSORPTION OF METAL-ORGANIC FRAMEWORK FOR HYDROGEN ISOTOPE SEPARATION

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Adsorptive separation using narrow micropore adsorbents has demonstrated the potential to separate hydrogen iso-topes. Herein, we employ an isotope-responsive separation using cobalt formate. A D2 responsive third sorption step was revealed, and consequently, a noticeable difference was observed in the uptakes of D2 and H2. This may have resulted from the additional space created for D2 due to its dense packing, as DFT calculations revealed that cobalt formate possesses 2.26 kJ/mol higher binding strength for D2 than that for H2. The exploitation of this D2-responsive third sorption step renders a promising separation performance, with a D2/H2 selectivity of up to 44 at 25 K/1 bar. Lastly, cobalt formate was synthesized on a gram scale here, which makes it a prospect for commercialization

Keywords: Tritium separation, Metal-Organic Framework, Isotope separation



NEW TYPE OF MEMBRANE CONTACT DEVICES FOR WATER DETRITIATION

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One of the most promising methods for separating hydrogen isotopes is isotopic exchange between hydrogen and water. Isotopic exchange in this system occurs in two stages, the first of which - isotopic exchange between hydrogen and water vapor occurs in the catalyst system, and the second is a phase isotopic exchange of water. Traditionally, the process is carried out in countercurrent columns filled with beds or a uniform mixture of hydrophobic platinized catalyst and hydrophilic packing [1- 3]. The hydrophobic properties of the catalyst lead to a decrease in the throughput of the separation column and, therefore, limits the field of application of the method to relatively small-scale problems. In Mendeleyev University of Chemical Technology of Russia, an alternative membrane-type contact device (MCD) is being developed, in which the catalyst located in the vapor-gas space is separated from the water flow by a Nafion-type membrane, on the surface of which phase isotopic exchange of water takes place [4, 5].

This report presents the results of a comparison of the mass transfer characteristics of contact devices of various configurations: MCD - with a flat membrane, MCD-TM - with a bundle of tubular membranes, and MCD-X - with a single tubular membrane wound in the form of a spiral. The work used a hydrophobic platinized catalyst RCTU-3SM [3] and domestic membranes: flat MF-4SK 250 μ m thick and tubular TF-4SK 150 μ m thick. The ratio of the catalyst volume to the membrane surface was 0.24 cm³/cm² in MCD, 0.095 cm³/cm² in MCD-TM, and 0.071 cm³/cm² in MCD-X. **Table 1** shows the dependences of the mass transfer coefficient (K_{oy}) in calculate on the hydrogen flow on the different speed of vapor-gaseous flow (w_{v-g}) at a pressure of 0.01 MPa and a temperature of 333 K.

Table 1. Dependences of the mass transfer coefficient in calculate on the hydrogen now								
Hydrogen flow, Ndm ³ /h	MCD		MCD-X		MCD-TM			
	w _{v-g} , m∕s	K _{oy} ·10 ³ , m ³ /(m ² s)	w _{v-g} , m/s	K _{oy} ·10 ³ , m ³ /(m ² s)	w _{v-g} , m∕s	K _{oy} ·10 ³ , m ³ /(m ² s)		
20	0.06	2.66	0.38	1.51	0.77	2.31		
40	0.12	3.08	0.76	1.99	1.54	2.88		
80	0.24	4.27	1.51	3.30	3.07	3.62		
100	0.30	4.74	1.89	4.70	3.84	5.17		

Table 1. Dependences of the mass transfer coefficient in calculate on the hydrogen fl	he mass transfer coefficient in calculate on the hy	drogen flow
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From the presented data, it can be seen that with an increase in the hydrogen flow, an increase in the coefficient of mass transfer is observed, and for all the studied samples, the K_{oy} values are close to each other. At the same time, due to the design of MCD-X and MCD-TM devices, the flow rates of the vapor-hydrogen mixture were much higher than in MCD, which allows us to conclude that the contact devices with tubular membranes are more efficient. In conclusion, it should be noted that on the basis of such contact devices, compact multistage counterflow modules can be created for separating hydrogen isotopes, including for the purpose of obtaining heavy water, which, unlike traditional counterflow columns, do not require vertical configuration.

Keywords: Membrane contact device, water detritiation, Nafion membrane, chemical isotope exchange, water-hydrogen system

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INVESTIGATING THE REACTION OF NITROGEN WITH URANIUM DEUTERIDE USING QUADRUPOLE MASS SPECTROMETRY

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Uranium has been and continues to be a useful media for the storage of tritium [1-3]. Uranium powder readily hydrides at room temperature and dehydrides when heated to ca. 400°C under vacuum [1-3]. Tritium processing lines are typically sub-atmospheric and installed in inert nitrogen gloveboxes and therefore a potential exists for nitrogen inleakage over time. Nitrogen is reported to react with uranium and uranium hydride at elevated temperatures [4-5], however the reaction of nitrogen with uranium hydride at room temperature has been less extensively researched.

To investigate this reaction further this work utilised 100 g of uranium that was repeatedly hydrided and dehydrided with deuterium to a stoichiometry of UD₃. D₂ was used to understand the chemistry as a precursor to conducting experiments using T_2 . Six successive cycles were performed to produce a powder with a high surface area to promote observation of surface reactions between UD₃ and N₂.

Reactions were performed by flowing a mixture of N_2 and ³He (95:5) over the UD₃ sample. ³He was used as an internal reference species against which the consumption of N_2 could be monitored via real-time quadrupole mass spectrometry. These measurements provided evidence of a reaction between N_2 and UD₃ at room temperature with a partial consumption of N_2 and production of D₂. Partial consumption of N_2 indicated the reaction may be limited to the hydride surface at room temperature. Increasing the sample furnace temperature to 230°C resulted in further reaction until all N_2 had reacted.

These results highlight that reaction between N_2 and uranium-hydrogen storage media can occur at room temperature. The impact of this reaction on tritium processing is currently undetermined.

Keywords: Tritium, Processing and Storage, Uranium Hydride, Nitrogen

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BENCHMARKING HYDROGEN ISOTOPE SEPARATION EFFICIENCY OF Pd/k PACKED TCAP COLUMNS

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The Thermal Cycling Absorption Process (TCAP), developed at the Savannah River Site, has successfully facilitated the separation of hydrogen isotopes on a broad scale. A fundamental piece to the success of this process involves the ability for the isotopes to be efficiently separated by a column packed with palladium deposited onto an inert kieselguhr support (Pd/k). As this technology continues to experience wider implementation in the field of fusion science, there comes a need to design adaptable columns for varied throughputs and performance requirements. Through the continued operation of this system and with changing requirements to its usage, a major problem to address lies in the separation efficiency of the Pd/k material. Many factors can influence this column efficiency, such as the particle size and homogeneity of the packing material, the packing density, and the absorption kinetics of hydrogen onto the Pd component. This work will outline the current performance of the TCAP column by utilizing previously tested pulse experiments and investigate the effect of particle size and distribution on the separation efficiency.

Keywords: Hydrogen isotope separation, TCAP, column efficiency



INSTRUMENTATION & CONTROL FOR TRITIUM COMPATIBLE HYDROGEN GENERATOR

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Hydrogen generators that use PEM cell stack for water electrolysis have various applications, one of them being CECE (Combined Electrolysis Catalytic Exchange) installations. This process separates a stream of tritiated water into detritiated water and a stream of tritium-enriched hydrogen. The stream of tritium-enriched hydrogen then can be used in tritium recovery processes. The development of hydrogen generator capable of processing tritiated water still arouses increased interest due to their low commercial availability. This paper presents the construction of a hydrogen generator compatible with tritium, in a two PEM cell stack configuration, in terms of instrumentation and control as well as the active safety measures implemented.

Keywords: tritium, electrolysis, instrumentation, control

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VERIFICATION OF THE APPLICABILITY OF VACUUM REGENERATION FOR CRYOGENIC MOLECULAR SIEVE BED

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Cryogenic Molecular Sieve Bed (CMSB) is one of the technologies capable of purifying and recovering helium-3 produced in a tritium storage bed. After the completion of purification step, the regeneration step to remove impurities adsorbed on the CMSB is performed typically by heating and purging with helium-4. However, the remaining helium-4 used as purge gas through the adsorption bed is mixed with and inevitably contaminates helium-3, which will be the product of the subsequent purification step. In order to avoid this contamination, in the regeneration step, impurities adsorbed on the bed are capable to be removed through vacuuming instead of purging. This study attempts to confirm the applicability of vacuum regeneration for CMSB in the helium-3 recovery system. Two types of regeneration methods, heating/purging and heating/vacuum, were compared in the aspect of adsorption performance which is the adsorption capacity and the moment of breakthrough start. The experimental results show there is no significant difference between two methods. From the viewpoint of process operation optimization, the adsorption performance according to heating temperature, heating time, and degree of vacuum was also investigated. These results are expected to be useful for establishing the design of the helium-3 purification process.

Keywords: Cryogenic Molecular Sieve Bed, helium-3 purification, vacuum regeneration, adsorption capacity, breakthrough.



EXPERIMENTAL STUDY ON VAPOR ADSORPTION AND DESORPTION USING AMBIENT MOLECULAR SIEVE

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An ambient molecular sieve bed (AMSB) in tritium extraction system (TES) is considered to be adopted to separate tritiated vapor in purge gas of pebble bed breeding blanket [1]. Recently, research apparatus for vapor adsorption and desorption (RAVAD) as presented in Figure 1 has been installed in the Korea Institute of Fusion Energy (KFE) to test adsorbent performance of AMSB in low vapor concentration as in the purge gas and various experiments are in progress now [2].

In this study, experimental results using compact AMSB are introduced to investigate absorbent performance. Dry air which has vapor concentration less than 40 ppb(v), is used instead of helium for the purge gas. The air flow to the AMSB with the target vapor concentration is supplied by the vapor generator which can consistently produce vapor up to hundreds ppm(v). In the full adsorption cases, the flow rate affects breakthrough time, but the total adsorption capacity is determined only by the vapor partial pressure. In the cyclic cases where AMSB was not completely desorbed, the remaining vapor in adsorbent affects subsequent adsorption. Therefore, operational scenario needs to be carefully prepared in order to maintain the adsorption performance for AMSBs in TES. Further works are planned to establish model for TES design and to validate it with experimental data.

Keywords: Tritium Extraction System (TES), Ambient Molecular Sieve Bed (AMSB), Vapor, Adsorption, Desorption, Dry Air.

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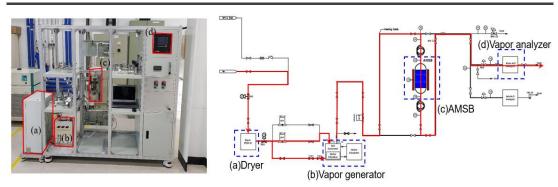


Figure 1. Layout of RAVAD; (a)Dryer, (b) Vapor generator, (c) AMSB, (d) Vapor analyzer and flow diagram for adsorption (red line)



ESTIMATION OF THE EFFECTIVE PUMPING SPEED FOR DEUTERIUM AND TRITIUM OF THE LINEAR DIFFUSION PUMPS IN THE *EU-DEMO* TORUS EXHAUST VACUUM PUMPING SYSTEM

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As the minimization of the tritium inventory of the EU-DEMO has been identified as one of the main design drivers, a continuous pumping process termed KALPUREX [1] has been suggested. In this process, Linear Diffusion Pumps (LDPs) are employed as continuously operating substitutes for cryopumps. Diffusion pumps are kinetic vacuum pumps and feature species dependent pumping speeds. In view of torus exhaust pumping, this is most prominently reflected in different pumping speeds for deuterium and tritium. In this work, we investigate the expected impact of the aforementioned effect on the effective pumping speed at the divertor outlet. We estimate the effective pumping speed from the transmission probabilities of the individual systems. In total, we employed three models: A Test Particle Monte Carlo (TPMC) model of the duct including the MFPs and a second TPMC model for the adapter including the two-stage baffle for the LDPs. The LDPs themselves are simulated using a model based on the Direct Simulation Monte Carlo (DSMC) method, as recently proposed [2]. The results confirm that the effective pumping speed is dominated by the conductance in the duct, metal foil pumps and baffle between divertor and LDPs.

Keywords: Fusion reactors, torus exhaust pumping, vacuum pumping, isotope effect, Linear Diffusion Pump

References:

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Tritium facilities and operation





QUALIFYING COILS FOR CRYOGENIC SERVICE

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Cryogenic coils comprising a combination of an LN2 line, a process line and a sheathed heater brazed together are a critical component in the arsenal of tritium handling tools. These coils appear in cryosorption pumps, tritium compressors and form the heart of the Thermal Cycle Absorption Process (TCAP). By their very nature, these coils handle large quantities of high-activity hydrogen gas. Any compromise in their integrity can lead to an untenable series of events that could eventually result in the release of large quantities of tritium into the environment. Design to the ASME code provides the first step in ensuring that these coils operate within the required pressure regime with a defined safety factor. However, long-term metal fatigue due to thermally induced stresses should be evaluated when designing these coils, to ensure the pressure boundary integrity remains intact for the service lifetime. For example, coils in the TCAP system are expected to undergo on the order of 600,000 thermal cycles in their operational lifetime. Coils used in cryosorption pumps are project to achieve upwards of 1,000,000 thermal cycles.

Two prototypic assemblies have been constructed to form a cohesive assembly with high thermal conductivity between the LN2 line, the process line and the sheathed heater: a paper-clip format proposed for isotope separation systems and a helix layout planned in cryosorption pumps. Both configurations were outfitted with an array of up to 31 thermocouples along the length of the coils. The coils were thermally cycled between 77 K and 420 K. The evolution of the thermal profiles was recorded. Additionally, a thermal imaging camera was used to visualize the propagation of the heat and cooling fronts as the coils were cycled between the temperature extremes. The time dependence of these profiles was used as the input data into the modelling of the thermally induced stresses that work harden the 316 stainless-steel walls of the coils and the braze.

A short length of two stainless steel tubes was brazed together and subjected to a 'pull' test to determine if the tube integrity along the line of attachment would be compromised should the braze joint failed. A cross-section of the separated tubes indicated that the tubes separated along the brazing bond without tearing the walls of the stainless-steel tubing.

This paper will present the thermal maps, discuss the stress analysis of both configurations, and present the pull test results.

Keywords: Cryogenic coils, Thermal Cycling Absorption Process (TCAP), cryosorption pumps.



CURRENT AND FUTURE LLNL TRITIUM FACILITY CAPABILITIES

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This overview is to highlight LLNL's current and projected operations in Tritium Facility that include tritium and tritiated materials and handling systems R&D, ICF target filling and R&D, tritium recovery, actinide and tritium decontamination, and actinide waste assay. Tritium operations include tritium-related research, tritium recycling and recovery, decontamination and renovation activities, legacy waste processing, and tritium target filling, isotopic analysis, and operational support. The Tritium Facility has a state-of-the-art light isotope gas analysis capability, calorimetric assay capability (being installed), and a full featured, dedicated National Ignition Facility (NIF) ignition target processing station (Figure Along with the existing hydrogen(H)-deutrium(D)-tritium(T) fill station, the full 1). spectrum of tritium R&D operations can be accommodated at B331. For example, the tritium gloveboxes can provide storage and purification of D2 and T2, custom isotopic mixing of D_xT_y , H_xT_y , $H_xD_yT_z$, mixing with noble gases and trace gaseous compounds, and recovery/recycling of lightly contaminated isotopic mixes. The Tritium Facility has a dedicated mass spectrometer for analysis of gas samples containing tritium for isotopes and contaminants. Tritium fill stations are equipped with easy-to-use manifold connections to interface with the user experiments and various fill assemblies. These fill stations enable testing the experimental assemblies in-line with the on-going H/D/T fill operations without the need to install a new experimental system.

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Keywords: Tritium isotopic mixes, ignition target fill stations, tritium recovery, mass spectrometry, calorimetry.



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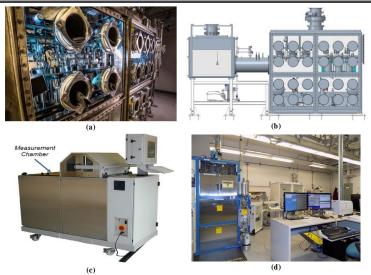


Figure 1. LLNL tritium operations. (a) Tritium glovebox for isotopic mixing with H/D/T and with other gases. (b) Design of a new tritium processing system to support ignition research. (c) Adding calorimetric assay capability to non-destructively obtain the mass of the tritium in storage containers and material in process. (d) Mass spectrometer for light isotope gas analysis.



FOUR YEARS OF TRITIUM OPERATION OF THE *KATRIN* EXPERIMENT AT *TLK*

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The Tritium Laboratory Karlsruhe (TLK) has a long history of developing technical tritium handling techniques and processes, reaching back almost three decades. Consequently, when the astroparticle community was reaching out to possible hosts for the "next generation" neutrino mass experiment, TLK with its proven and reliable tritium infrastructure was the obvious choice [1].

Since the start of the tritium operation of the KATRIN experiment back in May 2018 [2], more than 500 days of 24/7 measuring campaigns were conducted, that necessitated a reliable supply with tritium. This validated the standard procedures and technical rules applied by TLK. Nevertheless, several defects and off normal events had to be managed since the commissioning of the KATRIN tritium loop. Again, the decade-long experience and expertise at the TLK helped to solve any issue with the involved tritium processing systems on a short timescale.

This contribution will give an overview of the current operational conditions of the TLK tritium facilities involved as well as the relevant technical, analytical and administrative procedures implemented. Furthermore, an analysis will be given for system and component malfunctions in the tritium loop as well as the associated actions for problem-solving and repair. In addition, an end of live investigations for the component failure will be presented.

Keywords: Tritium Laboratory Karlsruhe, TLK, Tritium processing, KATRIN, closed fuel cycle

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RESULTS FROM THE CHARACTERIZATION OF IRRADIATED JET FIRST WALL CARBON MATERIALS AND THE IMPLICATIONS FOR TRITIUM RETENTION PROPERTIES

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The Joint European Torus (JET) reactor vessel has operated with carbon Plasma Facing Components (PFCs) since the mid-1980s. JET is the only fusion reactor operating with tritium. The graphite and Carbon Fibre Composite (CFC) plasma facing materials retained significant quantities of tritium [1]. For this reason, JET now operates with all-metal PFCs and ITER has abandoned a plan to initially operate with carbon PFCs, to avoid regulatory issues with radioactive tritium inventory 'lost' to retention in the reactor vessel.

Graphite moderators are widely used in fission reactors, especially gas-cooled types such as Magnox, Advanced Gas Reactors and RBMK. Activation of 2H, 6Li and 10B present as contaminants in the graphite leads to a significant buildup of tritium. The global inventory of irradiated graphite is estimated to be 250kt [2], with significant quantities in the UK, Russian Federation, USA and France, along with large liabilities in Ukraine, Lithuania, Spain, North Korea, Japan, Italy, Belgium and Germany.

Tritium contaminated graphite represents a significant liability, one which could be substantially reduced by the removal and recovery of this critical fusion fuel.

We present new results from a characterisation study of JET CFCs and JET and Magnox graphite with various tritium contamination levels. X-ray Diffraction, X-ray Tomography, Scanning Electron Microscopy, Raman spectroscopy and Secondary Ion Mass Spectroscopy analysis were used to characterise the carbon material and analyse the physical properties affecting tritium retention levels.

The tritium retention levels and depth profiles in JET graphite have been the subject of much study [1,3]. Our new results complement previous studies and help improve the understanding of tritium binding to, and release from, nuclear reactor carbon materials.

Keywords: irradiated graphite, tritiated carbon, waste repurposing.

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OVERVIEW OF SPARC TRITIUM HANDLING AND EMISSIONS MANAGEMENT FOR DT FUSION

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SPARC is a deuterium-tritium burning tokamak designed to produce net fusion power and develop a pathway for commercially viable fusion. SPARC is on track to start plasma operations in 2025 with a fully self-sufficient tritium handling and processing facility equipped to support it. The tritium processing systems are designed to collect, purify, store, and deliver tritium to the torus.

Leveraging the experience of the tritium community, SPARC will deploy a full tritium handling facility with qualified operators within 5 years of concept design. The tritium handling systems completed concept design review in 2020 and will complete detailed engineering and procurement of detritiation systems by early 2023. Following the acceptance of systems on-site, 12-15 months are planned for installation, testing, and qualification with inactive gases. The tritium plant will then ramp up to tritium operations and tokamak operations will transition to deuterium-tritium plasmas over 12 months. SPARC will leverage high-maturity technologies for processing and initiate design, procurement, system installation and, commissioning in parallel to reduce the timeline between conceptual design and operations as much as possible.

The tritium inventory required for fueling SPARC is 10 g. The tokamak's use of hightemperature, high-field superconducting magnets enables a compact design ($R_o = 1.85$ m, a = 0.57 m, $B_0 = 12.2$ tesla), roughly 2% the plasma volume of ITER [1]. Fusion gains (Q) greater than 2 are expected on SPARC with less than 0.5 g of tritium fueling and 0.3 mg burn up. Burning plasmas are expected to require less than 1g of tritium fueling and 2.5 mg burn up. The tritium fueling system is designed to receive gas from the tritium facility (40 m distance) stored at the tokamak port in a reservoir and deliver up to 1g of tritium fuel per 10 second pulse using commercial piezo valves compatible for tritium handling.

The in-vessel tritium retention is anticipated to remain low. SPARC will use ICRF heating [2], with no neutral beam injection which has been a source of tritium retention on previous tokamaks [3]. In addition, tungsten has been selected for plasma-facing materials in the divertor and limiter regions over a carbon wall which is anticipated to retain significantly less tritium than carbon plasma facing components. Regular glow-discharge



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cleaning and vessel bake-outs up to 350° C are planned to recover tritium from the first wall.

Tritium permeation into auxiliary systems such as the Vessel Heating and Cooling (VHC) System will be monitored. The VHC system is a gas-based heating and cooling system and is expected to collect more than 1 Ci of tritium that has permeated through the torus wall during vessel bake-outs. Humidification of the purge gas encourages permeant tritium to desorb from the wall as HTO which can be trapped on adsorbent beds to maintain a low activity gas stream. The tokamak ports are doubly contained and are maintained at 150°C during vessel bake-outs. The inert gas in the interspace will be purged to the tritium facility for recovery.

Commonwealth Fusion Systems has started construction of the facility on a 47-acre site in Devens, MA, USA. The site is designed to restrict annual gaseous emissions below 112 Ci and to maintain concentrations in the effluent below $0.1 \,\mu\text{Ci/m}^3$. Rooms that contain tritium will be ventilated between 4-8 room air exchanges per hour to minimize surface contamination. Tritium monitoring on-site will include the use of real time monitors in rooms, diffusion cells with weekly liquid scintillation counts, and surface swabbing to maintain activities below 1000 dpm/cm². In this presentation, we will discuss the status of SPARC, design, and operations for tritium handling, monitoring, and emissions management.

Keywords: Tokamak fusion reactors, deuterium-tritium fusion, measurement and monitoring, decontamination and waste management, tritium emissions

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R&D PROGRESS OF *D-T* FUEL CYCLING FOR *CFETR*

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CFETR (China Fusion Engineering Test Reactor) is a tokomak reactor which has been designed by China national integration designing group, and one of its objectives is to demonstrate the tritium self-sufficiency. A D-T fuel cycling system has also been designed that included D-T fuel recovering system which functioned as tritium recovery from the exhaust gases in tokomak plasma and feeding back to the plasma vessel, tritium breeding system which functioned as tritium containment and tritium recovery from gases and liquid effusions. A series of D-T fuel cycling related R&D have been carried out, and a D-T fuel cycling evaluation system has also been developed which fuel cycling properties were demonstrated recently.

Keywords: Tritium, fuel cycling, design, evaluation, progress, CFETR.



TRITIUM INFRASTRUCTURE OF THE AKULINA-2 FRAGMENT SEPARATOR

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RFNC-VNIIEF has developed a tritium infrastructure for the AKULINA-2 [1] fragment separator. It includes a complex that allows you to work with tritium in the amount of 2,7 kCi, and a family of tritium targets for conducting relevant experimental studies.

Components of the tritium infrastructure are located in a special room designed to work with tritium on II class of work with open sources of radiation.

The report provides a description of this infrastructure, the main principles of organization of works on this infrastructure, as well as structural features of gas and liquid-tritium targets.

The availability of this infrastructure makes it possible to conduct a wide class of studies of light exotic nuclei, such as ${}^{6.7}$ H, 10 He, 13 Li, 16 Be, ${}^{19-21}$ C, 24,26 O et al., formed in direct nuclear reactions (t,p) and (t,d).

Keywords: Tritium target, tritium infrastructure, light exotic nuclei, fragment separator.

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CONSTRUCTION OF A TEST FACILITY AND THE TEST OF AN ACTIVE TRITIUM PERMEATION BARRIER

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In future fusion or fission reactors, tritium permeation may present a serious challenge. In order to separate the water steam cycle from gas streams containing significant amounts of tritium, a permeation barrier is necessary. Tritium permeation into the environment through steam generators and heat exchangers can be a significant hazard regarding radiation and environmental safety.

In the scope of the project Transversal Actions for Tritium (TRANSAT) a facility has been set up to perform tests on various scaled and functioning permeation barrier mockups at the Tritium Laboratory Karlsruhe (TLK). The facility was built in a standard glove box unit in accordance to the technical terms and requirements of tritium handling at TLK. The behavior of an active permeation barrier was investigated.

Within the first series of TRANSAT experiments, four different mock-ups have been tested for tritium permeation. Migrated tritium is oxidized to tritiated water (HTO) using carulite reactors and molecular sieves for HTO trapping. This paper will present the construction, set up and commissioning of the facility as well as the first series of TRANSAT experiments including their evaluation.

Keywords: Tritium migration, Tritium permeation, permeation barrier, fusion reactor, reactor safety, tritium release mitigation, test facility



LUMPED-PARAMETER MODELLING FOR THE DESIGN OF MODULAR PAV TRITIUM EXTRACTION UNITS FOR THE EUROPEAN DEMO REACTOR

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In the framework of the technologies proposed for the Tritium Extraction and Removal System (TERS) of the Water-Cooled Lithium-Lead Breeding Blanket (WCLL BB), one of the promising solutions is the Permeator Against Vacuum (PAV) adopting a niobium membrane. While PAV mock-up, based on a shell-and-tube design with Nb U-shaped pipes, is currently under installation at ENEA C. R. Brasimone, Italy [1], the preliminary design of a scaled-up configuration for the TERS of the European DEMO is ongoing. A modular approach has allowed identifying the optimal size of a PAV Tritium Extraction Unit that could fit both the Inboard and the Outboard Blanket loops, adjusting the number of PAV Units in each loop, simply based on a suitable permeation model for the hydrogen isotopes through the Nb pipe wall. However, if a U-shaped pipes design is adopted, the large number of pipes (~400) in the PAV Unit will lead to some deviation from the ideal condition and will impact the PAV performance, in view for instance of the non-uniform flow distribution among the pipes and of the effects of radiative heat losses to the surrounding. Both effects could be in principle investigated thorough 3D CFD, but such a large number of pipes prevents the use of any CFD tool to perform a detailed thermal-hydraulic analysis. Recently, a lumped-parameter model of a PAV system has been developed using the Modelica language. In the model, the U-shaped pipes are simplified in 1D thermal-hydraulic lines, the hydraulic and thermal characteristics of which have been derived and calibrated from a detailed CFD model. The permeation model for the hydrogen isotopes through the Nb pipe wall has been also implemented, so that the new RadiaTube component can account for the PbLi flow, radiative heat transfer and Tritium permeation through the pipe wall. The RadiaTube Modelica model has been calibrated against results from the PAV mock-up [2].

The aim of the work here is to use the new *RadiaTube* Modelica component to develop a lumped parameter model of the entire PAV unit, and compute the extraction efficiency during operation, accounting also for the thermal losses to the environment and for the flow non-uniformity among the pipes in the Unit. Th will allow the validation of the preliminary design of the PAV Unit.



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Keywords: Tritium Extraction and Removal System, Permeator Against Vacuum, lumped model, DEMO reactor.

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LONG TERM OPERATION OF THE TRITIUM SOURCE OF THE KATRIN EXPERIMENT AND RECENT NEUTRINO MASS RESULTS

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The Karlsruhe Tritium Neutrino (KATRIN) Experiment aims to measure the neutrino mass in a model-independent manner with a sensitivity of 0.2 eV/c^2 (90% C.L.). This is achieved by spectroscopy of the beta-decay electrons of tritium close to the kinematic endpoint using a high-resolution (<1 eV @ 18.6 keV) integrating spectrometer. For this purpose, a windowless gaseous tritium source (WGTS) with a high stability in activity and hence tritium throughput (0.1 %/h) is necessary to provide 10^{11} β-electrons per second [1]. The WGTS and the connected tritium processing infrastructure have been operated successfully with high purity (>98%) tritium at the Tritium Laboratory Karlsruhe since 2019 [2]. The first campaign in 2019 set a new upper limit on the neutrino mass of 1.1 eV/c² (90% C.L.) [3]. A second campaign in the same year but with higher source strength allowed a further reduction of the upper limit to 0.8 eV/c² (90% C.L.) [4].

The beta-electrons generated in the strong tritium source partly ionize the molecular gas and form a 30 K-cold plasma inside a source magnetic field of 2.5 T [1]. Spatial inhomogeneities in this plasma and thus in the starting potential of the β -electrons lead to a distortion of the β spectrum. This can lead to a systematic bias in the neutrino mass analysis. By measuring the lineprofile of mono-energetic conversion electrons from traces of ^{83m}Kr [5], used as an atomic reference standard, co-circulating with the tritium inside of the WGTS, these distortions can be precisely quantified. Due to the low vapor pressure of Kr at the design source temperature of 30 K, the plasma calibration has to performed at an elevated source temperature (>80 K).

During the second KATRIN campaign at nominal column density it has been shown that the start potential of the beta-electrons defined by the plasma was drifting due to drifting surface potentials at the boundaries of the source. In 2020, the source setpoint for neutrino mass measurements was changed from T = 30 K to T = 80 K. We will show how this choice reduced the maximum achievable tritium activity by 10%, but allowed performing the ^{83m}Kr calibration under identical operational conditions as the during the neutrino measurements. Furthermore, after changing to the higher temperature setpoint, the observed drift has decreased in magnitude by about a factor of 10.

After this change, between early 2020 and late 2021, four measurement phases were successfully conducted. With the reduced systematic impact of the source potential drift after the source temperature change, the limit on the neutrino mass will be able to be improved upon significantly in the near future.

Keywords: KATRIN, cold tritium plasma, 83m-krypton, windowless gaseous tritium source



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DESIGN OF THE SPHERICAL TOKAMAK FOR ENERGY PRODUCTION (STEP) TRITIUM FUEL CYCLE

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The Spherical Tokamak for Energy Production (STEP) mission is to "deliver a UK prototype fusion energy plant, targeting 2040, and a path to commercial viability of fusion". Measures of performance underpin this mission. These include net electrical power generation, safety, tritium sustainability, cost, and maintainability. A concept maturity level system engineering approach has been adopted during the initial concept design.

The STEP fuel cycle has numerous primary objectives to support the fusion reactor operation. These include:

- To provide the fusion reactor vacuum environment
- To maximize tritium recovery
- To separate tritium and deuterium to achieve the fueling ratio
- To remove impurities
- To provide matter to maintain the fusion plasma

Gathering interface requirements between fuel cycle and the reactor has been critical to support definition of the fuel cycle's functional specifications. Detailed technology assessments have been conducted focused on achieving the fuel cycle objectives above. The technology evaluation covers:

- Separation of tritium from impurities
- Separation of hydrogen isotopologues
- Injection of tritiated and non-tritiated matter
- Detritiation of trace tritium streams
- Provision of reactor vacuum environment
- Extraction of bred tritium
- Mitigation of tritium permeation
- Storage of tritium
- Accounting of plant-wide tritium

Modelling capabilities are being developed using Aspen Plus[®] and Aspen Custom Modeler[®]. This capability supports analysing different configurations and technology options. In addition, it enables an early steer on matters such as tritium breeding, tritium inventory and STEP's fuel sustainability objective.



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The STEP fuel cycle team will demonstrate the challenges of a continuously operated fusion fuel cycle at the STEP commercial scale. The impact of these challenges on the fuel cycle configuration and technology selection will be explored.

Keywords: Tritium separation, detritiation, fusion reactor, tritium extraction, modelling, fuel cycle.

	Content Description	Presenter
1	Introduction and background on STEP programme	Mohamad Abdallah
2	Concept maturity process and integrative nature of the tritium fuel cycle	Mohamad Abdallah
3	Overview of the STEP fuel cycle	Mohamad Abdallah
4	Separation systems and plasma exhaust processing	Elaine Loving
5	Recycle and vacuum pumping	Antulio Tarazona
6	Modelling tool development	Iryna Bennett

Table 1. Design of the Spherical Tokamak for Energy Production (STEP) Tritium Fuel Cycle Presentation Contents.



Figure 1. STEP Prototype Reactor Illustration – UKAEA



TRITIUM INFRASTRUCTURE FOR THE "TARGET FACILITY" OF THE LASER FUSION PLANT

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The RFNC-VNIIEF has developed a tritium infrastructure for the "Target Facility" of the laser fusion plant. It includes a research complex for testing the technology of filling microtargets with deuterium-tritium fuel and a system of tritium purification, collection and disposal (TPCD). The TPCD system, which is the part of the "Target Facility" complex, is designed for safe operation, maintenance and constant supply of a gaseous mixture of hydrogen isotopes, including tritium, specialized stands for filling of thermonuclear targets, as well as for filling target assemblies, execution dosimetric control and disposal of radioactive waste.

The "Target Facility" ensures the functioning of two stands (filling systems): system for filling glass micro-shells with a hydrogen isotopes mixture by diffusion with operating pressures up to 10 MPa and temperatures up to 300 °C and system for filling polymer shells by low-temperature distillation of a hydrogen mixture through a capillary. The TPCDsystem includes gas purification units operating on the principle of conversion of hydrogen isotopes on catalysts into water with further retention of water vapor on molecular sieves (zeolite-based adsorbers).

The components of the tritium infrastructure are located in special rooms designed for work with tritium according to II-class work with open sources of radiation.

Keywords: Tritium infrastructure, laser fusion plant, gas purification, tritium retention, target filling system.



SELECTING FUEL ISOTOPE COMPOSITION FOR HEATING INJECTORS OF COMPACT FUSION NEUTRON SOURCE FNS-ST

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For FNS-ST [1], a compact neutron source, the dependence of the neutron yield on the tritium content in the core plasma is studied to optimize the operation of heating injectors with different isotope compositions of the neutral beams. Self-consistent modeling of the FNS-ST operating modes is performed using the SOLPS4.3 and ASTRA codes [2, 3] for different bulk plasma densities and diffusion coefficients. The FC-FNS code [4-7] is used to calculate the required flows of the fuel components into the plasma provided by various injection systems: pellet injectors and neutral beams. During simulation, the plasma density varies in the range $n_e = (7-10)\cdot10^{19}$ m⁻³ and the ratio of the particle to the heat diffusivities in the range $D/\chi_e = 0.2-0.6$. For scenarios with different beam composition, the neutron yield was evaluated for the parameter range – **Figure 1**. For operation with the maximum neutron yield of $(4.5-5.5)\cdot10^{17}$ s⁻¹, the tritium accumulation on the site up to 370 g for different isotope compositions of the heating beams is estimated.

This work was supported in part by the Russian Science Foundation (project no. 18-72-10162).

Keywords: fusion neutron source, FNS-ST, spherical tokamak, neutral beams, hydrogen isotopes, fuel cycle, tritium inventories, neutron flux, integrated plasma simulation.



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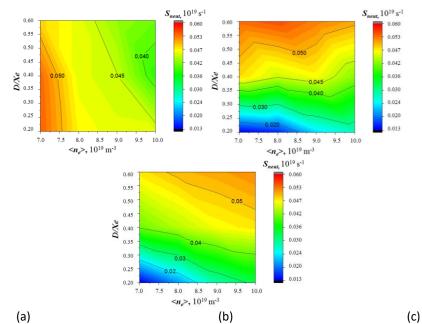


Figure 1 - Neutron yield S_{neut} as a function of n_e and D/χ_e parameters for (a) D+T-, (b) D- and (c) Tbeams. Scale on the right shows the correspondence of the color and S_{neut} values.

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SPARC TOKAMAK TRITIUM PROCESSING SYSTEMS

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SPARC is a compact (R_o = 1.85 m, a = 0.57 m), high-magnetic-field (B_0 =12.2 T), superconducting, tritium-burning tokamak designed to produce a fusion gain (Q) exceeding 2 [1]. The working inventory of SPARC will be 10 g of tritium.

The SPARC tritium-handling infrastructure comprises five major systems: tokamak exhaust purification, isotope separation, trace tritium recovery, water treatment, and tritium storage and delivery. This presentation will provide an overview of each of these systems and discuss how those systems are interlinked, with particular attention given to the management of tritium inventories within each system.

The Tokamak Exhaust Purification System collects gas during regeneration of the Divertor Neutral Cryopumps and effluent from the Torus Vacuum Pumping System on cryogenic molecular sieve columns. Gases released from the cryosorption pumps are passed through a palladium/silver permeator to separate the hydrogenic species from inert gases and impurities. The hydrogenic species are directed to the Isotope Separator. The remainder of the gas is treated over a nickel catalyst to recover tritium bound to impurity gases. The treated impurities are sent to the Trace Tritium Recovery (TTR) System to convert any residual tritium to tritiated water.

The Isotope Separation system is based on the Thermal Cycling Absorption Process (TCAP) [2,3], where hydrogen atoms are separated on a palladium on kieselguhr matrix and hydrogen isotopologues are separated on cryogenic molecular sieve. Pure tritium is directed to the Tritium Storage and Delivery System (TSDS). Raffinate is sent directly to stack or, if the tritium content is unacceptably high, to the TTR. This system is housed in a glovebox using a helium cover gas.

All air-bearing effluents containing trace quantities of tritium and tritiated hydrogenic effluents from the various process loops are discharged into the TTR System. TTR is a classic burnand-dry system in which the incoming hydrogen is oxidized and collected as tritiated water on room-temperature molecular sieve driers [4,5]. The primary function of TTR is to reduce emissions from the facility. Tritiated water is periodically recovered from the driers and sent to the Water Treatment System.

The Water Treatment System is based on the Combined Electrolysis Catalytic Exchange (CECE) Process by coupling an alkaline electrolyzer to a liquid-phase catalytic-exchange column to concentrate tritium in the electrolyte [6,7]. Column detritiation factors of the order of 10⁷ to 10⁸ are projected. The electrolysis cell will reside in an air box. High-activity tritiated gas is sent to the isotope separator.



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The Tritium Storage and Delivery System (TSDS) is based on uranium beds [8]. TSDS will utilize several U-beds to collect, store, and deliver gas to the Fuel Injection System. Pure tritium will be delivered from one U-bed while a second, cold U-bed will be on standby should the need arise to recover tritium from the Fuel Injection System rapidly. TSDS will be fitted with a rf quadrupole and a calibrated volume to assay gas on a regular basis. The TSDS will be housed in a glovebox using a helium cover gas.

Keywords: Tokamak fueling system, tritium recovery, purification, emission reduction

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RUSSIAN TRITIUM CYCLE TECHNOLOGY READINESS LEVEL ANALYSIS FOR THE DEMO-FNS HYBRID (FISSION-FUSION) REACTOR

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DEMO-FNS is a hybrid (fusion-fission) reactor with a DT fusion capacity of 40 MW [1]. It is now being designed in Russia [1]. The tokamak-based reactor specific features are stationary operation, $Q \sim 1$ and a blanket with fissile materials - these factors largely determine the configuration and basic fusion cycle (FC) technologies [1,2].

FC technologies are based on technologies for handling tritium and deuterium, which are now being developed and used in various fields of science and technology. The need to improve tritium technologies in Russia is due to the transition to larger gas flows (orders of magnitude) and reserves of tritium in the fuel cycle of hybrid and fusion systems in the context of restrictions on the import of dual-use technologies. This paper compares the matureness of the previously selected candidate technologies for tritium-deuterium FC [2] in the Russian Federation and in the world.

We have carried out an assessment of the readiness of existing in Russia technologies for handling tritium and deuterium for use in the DEMO-FNS fuel cycle. For the analysis, the Technology Readiness Level (TRL) methodology was used, in accordance with which each technology was assigned a readiness level from TRL 1 (the basic principles of the technology were demonstrated) to TRL 9 (the technology was tested by successful operation). The TRL methodology has become widespread and has recently been actively used in the scientific field - in particular, it was used to assess the readiness of technologies in the fusion: for a fusion power plant [4]; for the fuel cycle of the DEMO reactor [5]; for plasma diagnostics [6]. The following technologies were analyzed by authors in [7]: membrane separation of hydrogen-containing gas mixtures, cryogenic hydrogen rectification, chromatographic separation of hydrogen isotopes, cryoadsorption separation, gas detritiation in a scrubber, the CECE process. Tritium storage technology, breeding materials and tritium extraction process and tritium safety systems were considered specifically in this report and generalized conclusions including data from [7] were made. This includes all major systems and technologies for the tritium fuel cycle of the DEMO-FNS facility.

The listed technologies have been developed in Russia and are used in various areas of industry and science. However, the operating conditions for the use of technologies differ from the planned parameters of the DEMO-FNS FC, for which most technologies are under development phase (TRL 4-6), certain technologies, such as cryoadsorption separation and chromatographic processes correspond to the research stage (TRL 1-3). The



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state of these technologies is "below" or "corresponds to the world level". For the further development of the considered technologies, specialized installations and stands are needed, which will allow testing their joint use under conditions that simulate the operation of a fusion installation. Based on the analysis results a Roadmap for the development of technologies for the tritium fuel cycle in Russian Federation was formed

The work was supported by the NRC "Kurchatov Institute".

Keywords: Fuel cycle, tritium, deuterium, hydrogen isotopes, technology readiness level, TRL, technology maturity, DEMO-FNS, fusion reactor, tritium technologies in Russia, palladium membranes, hydrogen cryogenic rectification, CECE-process, chromatographic separation hydrogen isotopes, removal of tritium-containing impurities by adsorption at cryogenic temperatures, removal of tritiated water vapors in a scrubber, tritium storage, breeding materials and tritium extraction, tritium safety systems

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OVERVIEW OF THE CURRENT STATUS IN TRITIUM BATTERY DEVELOPMENT, MANUFACTURING AND APPLICATIONS

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Nowadays, more and more unattended electronic devices that require low power consumption are used in various fields such as industry (sensors for remote systems, analog digital microchips), telecommunications (wireless communication devices), medicine (e.g. ., pacemakers, defibrillators, brain neurostimulators), etc. The most suitable power supplies for these types of devices are batteries. However, in order to be used, batteries must meet certain requirements, such as small size and long life, so that they do not require replacement too often or never. These requirements are met by radioactive isotope batteries. The energy density of radioactive isotopes is a few orders of magnitude higher than in the case of stored chemical energy, making isotopic batteries an important option, despite the limitations in terms of output power, required by radiation regulation and safety. In this paper, various aspects related to tritium batteries are reviewed and discussed, including state-of-the-art manufacturing technologies, as well as their applications.

Keywords: tritium battery

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TRITIUM CATEGORIZATION METHOD OF COMPONENTS AND SYSTEMS TO DRIVE REQUIREMENTS AND GUIDANCE FOR THE DESIGN AND OPERATION OF TRITIUM WETTED SYSTEMS

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Tritium categorization is a convenient way to design and manage tritium processing plants as it allows consistent and balanced application of requirements and guidance for the safe handling of tritium. The goal of the tritium categorization is to provide a basis and support for design decisions, operation concepts and safety strategies concerning the specific radiological properties and hazards of tritium. General hydrogen specific hazards like flammability are not considered by this tritium categorization process and should be covered elsewhere.

The assessment of tritium hazards and the implementation of specific design concepts and solutions for tritium process systems needs to be performed in an organized and methodological manner. The paper gives an introduction to the decision process for categorization of tritium process systems into defined tritium groups, and the selection of appropriate requirements for the these tritium groups. It provides an overview of the various fields and topics concerning tritium characteristics, material interactions and hazards, and presents methods and concepts to be considered before and during the design development phase of tritium process systems in view of process performance, occupational and nuclear safety.

The first step of the tritium categorization method is the definition of tritium groups based on handled activity concentrations (gases, liquids) and pressure/temperature systems of the respective process systems. This is followed by the identification of requirements to manage the tritium hazards within the given operation boundaries for each tritium group. The third step in this method is the selection or development of design solutions for the tritium system linked to the identified requirements. In addition, major concepts and features like confinement systems and safety schemes need to be defined in parallel depending on the tritium activities at risk.

Keywords: tritium hazards, tritium process systems, material compatibility, tritium requirements, tritium safety, tritium accountability.

"The views and opinions expressed herein do not necessarily reflect those of the ITER Organization."





METHODOLOGY FOR EXPERIMENTAL STUDIES OF TRITIUM GENERATION AND RELEASE FROM TWO-PHASE LITHIUM CERAMICS UNDER IRRADIATION CONDITIONS IN THE *WWR-K* REACTOR

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The development and use of new structural and functional materials in fission and fusion energy require experimental data on their properties under conditions of influence of operating factors. However, it should be noted that there are few works on the study of material samples during reactor irradiation, which are due to both technical and methodological complexity of such experiments.

This paper presents the results of the development of a methodology for studying lithium ceramics, the blanket material of fusion reactors, during irradiation in the WWR-K reactor at the Institute of Nuclear Physics (Almaty, Kazakhstan).

The stages of preparation of in-reactor studies of two-phase lithium ceramics of $LiSiO_4+Li_2TiO_3$ type in the reactor core are defined, the description of the experimental installation CIRRA which provides measurements of tritium and helium yield by vacuum extraction, description of the reactor ampoule design and results of thermal physical calculations of irradiation ampoule parameters with samples are given. The results of methodical calibration experiments with hydrogen, deuterium, and helium influx are presented, and calibration coefficients are determined that allow the fluxes of gases released from the samples to be calculated by the magnitude of the measured partial pressures.

The work is supported by the Ministry of Education and Science of the Republic of Kazakhstan with Grant No. AP09259535.

Keywords: fusion blankents, lithium ceramics, reactor experiments, irradiation, tritium release.



Biological effects

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EFFECT OF TRITIATED WATER ON MITOCHONDRIAL MUTANT CELLS AND ITS RADIATION SENSITIVITY

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There have been a small number of reports of radiation-induced mitochondrial DNA(mtDNA) damage [1,2], but mtDNA damage which decide cell state induced by tritiated water(HTO) has not been investigated before. MtDNA is an important contributor to the ATPgenerating oxidative phosphorylation complex and energy deficiency from mitochondrial mutations in these cells [3]. Therefore, in the case of mitochondrial mutant cells, we will investigate the radiation sensitivity of mutant cells through the effects of HTO radiation on ATP synthesis, cell apoptosis assay and growth inhibition. Using human lymphoblastoid cells with Leber optic atrophy(GM 10744) and Leigh's syndrome(GM 13740), which are two such diseases arising from point mutations in the mitochondrial genome and comparing whit normal human lymphoblastoid cell line (GM 15036). The result of 0 to 72h postirradiation, the different increase phenomenon of apoptosis signal and proliferation toxicity in mutation cells indicated the response of mitochondrial mutant cells to HTO radiation (0-1Gy) was very different from normal cells. Leigh's syndrome cells exhibit radiation hypersensitivity after HTO irradiation. In contrast, the mutation of the Leber optic atrophy cells conferred radioresistance. These observations were explained preliminarily by the change of ATP generation in different mutant cells with time after irradiation.

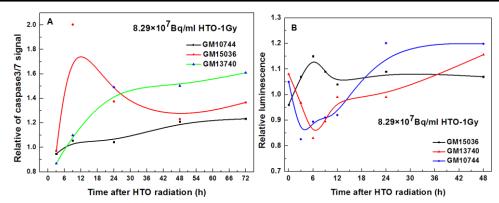
Keywords: Tritiated Water, Mitochondrial Mutant Cells, Radiation Sensitivity

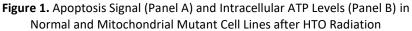
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Interactions with materials





SYNTHESIS OF ERBIUM DEUTERIDE AND ERBIUM TRITIDE BY ION BEAM IMPLANTATION

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Erbium hydrides are suitable materials for the long-term storage of hydrogen and this work details the first reported instance of erbium tritide synthesis via ion beam implantation. This method offers an alternative loading methodology to conventional gas loading with several advantages and these are discussed within. The work also demonstrates the capabilities of the newly upgraded DELPHI implantation facility which now operates with tritium.

The findings of an experimental study are presented in which the DELPHI facility was used to implant ions of deuterium as well as a protium/tritium mixture to form erbium deuteride and tritide compounds. Following implantation, samples were analysed by thermal desorption spectroscopy to quantify the hydrogen species retained and identify the formation of hydride phases. The effect of ion beam parameters on the film composition and total hydrogen uptake are investigated and discussed.

Keywords: Tritium implantation, materials science, tritium facility, measurement and monitoring.



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Figure 1. A sample during ion beam exposure in DELPHI



EQUILIBRIUM PRESSURES OF HYDROGEN ISOTOPES OVER HYDRIDE, DEUTERIDE AND TRITIDES OF TITANIUM

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Measurement of equilibrium pressures of hydrogen desorption in the system Ti-X, where X - H or D, in the temperature range from 820 to 978 K and the range of change in the content of hydrogen isotopes from a.r. X/Ti=1.65 to a.r. X/Ti=2.01. The range of equilibrium pressure change in the experiments, for different hydrogen isotopes, ranged from 2 to 277 MPa. The synthesis of hydrides was carried out at a pressure of 100 MPa and a temperature of 773 K.

According to the obtained gas desorption isotherms, the heats of formation of hydride and deuteride of titanium in the indicated range of atomic ratios (a.r.) X/Ti was found.

Data on P-T-C diagrams for Ti-D system, obtained on titanium of various grades, in different temperature ranges at different pressures, were found in some literature. Based on the data available in the literature on P-T-C ratios according to the Vant-Goff equation, the authors found differential heats of hydrides formation depending on a.r. X/Ti. Based on these calculation data and data obtained in the present work, the dependence of the differential heat of titanium hydride and deuteride formation on its hydrogen content in the range of atomic ratios X/Ti from 0.01 to 2.01 is built.

Based on literature-known isotopic effects for Ti-H, Ti-D and Ti-T systems and the experimental data obtained in this work, the heat of titanium tritides formation was calculated.

Keywords: Hydrogen isotope, titanium hydride, equilibrium pressure, desorption isotherms, P-T-C diagrams, differential heats of hydrides formation.

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TRITIUM PERMEATION FROM/TO HIGH TEMPERATURE, HIGH PRESSURE WATER THROUGH INCONEL 600

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A nuclear fusion power plant will use a steam turbine to generate electricity. Tritium (T) permeation through steam generator piping results in the risk of uncontrolled T leakage to the environment. Therefore, T permeation must be precisely evaluated and minimized. Nickel alloys are widely used as pipe materials. In this study, the permeation of T from/to high temperature, high pressure water through Inconel 600 film was examined.

Thin disks of Inconel 600 were used as samples. The thickness of the samples was 0.1 mm and the surface area exposed to water was 200 mm² for each side. The permeation device used was made of type 304 stainless steel and separated into two chambers by a sample disk. The upstream chamber was filled with tritiated water (0.9 MBq/cm³) and the downstream side was filled with non-radioactive water. The volume of each chamber was 3 cm³. The device was placed in a forced convection oven and heated to 280°C for 14–60 h. The vapor pressure of water at this temperature is 6.4 MPa. After heating, the downstream chamber was opened and the concentration of T in water was measured using a liquid scintillation counter.

Correlation between heating time and the amount of T permeated to the downstream side is shown in Figure 1. The initial permeation rate up to 14 h was 1 Bq/h. After 14 h, the data points spread in a relatively wide range but the permeation rate in average was 3 Bq/h after 14 h. Namely, the permeation rate increased after 14 h. Here, T cannot permeate through the sample in the form of a HTO molecule. T atoms are liberated by the oxidation of metals and a part of the liberated T atoms permeate to the downstream together with H atoms. The other part of liberated T is released into the upstream chamber as HT. At the downstream side surface, a T atom is released in the form of HTO via isotope exchange with H₂O or as HT via recombination with a H atom. Namely, the partial pressure of HT increases with heating time in both chambers. This increase in HT partial pressure is one of the possible reasons for the increase in permeation rate. A more detailed discussion on the permeation mechanisms will be given at the presentation.

Keywords: tritium, tritium permeation





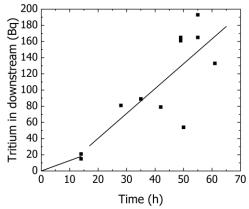


Figure 1 - Change in T permeation with elapse of time



THE EFFECT OF HEAVY ION IRRADIATION ON THE MICROSTRUCTURE EVOLUTION AND DEUTERIUM RETENTION IN PURE W AND W-ZrO2 ALLOYS

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In this work, microstructure evolution, deuterium retention and mechanical behavior on pure W and W-1.5ZrO₂ alloys were investigated through heavy ion C^{\dagger} irradiation for up to 4 dpa under different temperature of 400-700°C. TEM observation results showed that dislocation loops density decreased but voids number increased and the diameter of dislocation loops and voids obviously increased with the increasing of irradiation temperature in two materials. By comparison, W-1.5ZrO₂ alloys had less and smaller dislocation loops and voids owing to the more grain boundaries and interfaces between the matrix and ZrO2 phase. Meanwhile, the percentage of <111> and <100> type loops at the irradiation conditions was determined. High resolution TDS and EELS analysis demonstrated total deuterium retention amount and depth after different temperature irradiation and deuterium permeation. Simultaneously, internal correlation relationship between radiation induced defect types and deuterium retention state were revealed. The surface hardness measurement showed that the irradiation hardening of pure W and W-1.5 ZrO₂ alloys were saturated at 600°C for 4 dpa. But W-1.5 ZrO₂ alloys had less irradiation hardening effect, indicating ZrO2 phase could absorb more irradiation defects, to be the promising candidate as plasma facing materials in the future fusion devices.

Keywords: plasma facing materials; W alloys; microstructure evolution; deuterium retention; mechanical behavior



SMALL ANGLE NEUTRON SCATTERING TO CHARACTERIZE DECAY HELIUM BUBBLES IN TRITIUM PRECHARGED STAINLESS STEELS

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Long term exposure of stainless steels to tritium gas results in a decay helium bubble microstructure that greatly affects their deformation and fracture properties. Therefore, developing an understanding of the long-term embrittlement effects of tritium and decay helium on the structural properties of stainless steels is one of the main goals of the Savannah River Tritium Effects on Materials programs. In this study, small angel neutron scattering (SANS) was used to probe tritium exposed steels to learn as much as possible about decay helium bubble size, spacing and distribution. The information complements transmission electron microscopy observations, particularly in forged microstructures where nanometer-sized bubbles are not easily resolved due to high dislocation densities. Samples were sliced from tritium-exposed-and-aged fracture mechanics specimens tested at Savannah River. The samples included 3 and 17-year aged specimens, the latter representing the longest aged specimens tested in the program. Companion specimens in the as-forged or hydrogen-precharged condition were also examined. Measurements were performed on beamline CG-2 at the High Flux Isotope Reactor (HFIR) at Oak Ridge National Lab (ORNL). Small angle incoherent neutron scattering (SAINS) measurements were also performed on the hydrogen-precharged samples to measure the hydrogen content in the samples, and across weldments. The data show that helium bubbles in tritium charged welds fit well to a hard sphere model, while the as-forged and hydrogen-precharged welds fit well to a Gunier-Porod model with and increased incoherent background for the hydrogen-precharged condition. The results and analysis suggest the technique has good potential for providing better statistical information on the size, spacing, and distribution of the decay helium bubbles in tritium exposed and aged stainless steels.

Keywords: Tritium-material interactions, Mechanical Properties, Helium Bubbles, Embrittlement.



TRITIUM TRANSPORT IN MATERIALS FOR TRITIUM PRODUCTION

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Tritium-producing burnable absorber rods (TPBARs) are irradiated in a commercial power plant to produce tritium for US defense needs. A comprehensive experimental and computational research program exists to reduce technical and programmatic risk by improving fundamental scientific understanding of the irradiation performance of tritium-producing materials irradiation performance. The presentation will provide examples of these activities with focus on integration of results from studies that are providing insight into tritium transport in g-LiAlO₂ pellets that produce tritium in TPBARs, and the aluminide-coated Type 316 stainless steel that provides both a tritium barrier and structural cladding.

Before irradiation, the LiAlO₂ pellets are essentially single-phase with a small quantity of pores and LiAl₅O₈ precipitates located mostly on grain boundaries. After irradiation, grain boundaries are clean, although open in some cases due to partial amorphization near the boundary, the periphery of each grain is dense LiAlO₂, and large LiAl₅O₈ precipitates with associated pores are located in the middle of the grains. Understanding microstructural evolution during irradiation is important for understanding the mechanisms and kinetics of tritium transport. Keys to building this understanding include postirradiation microstructural characterization of TPBAR pellets to quantify the extent of LiAl₅O₈ precipitate formation, porosity formation and amorphization; neutron irradiation experiments with in-situ measurement of tritium release; complementary ion irradiation studies to observe irradiation damage mechanisms and defect mobility as well as to quantify tritium diffusivity and release; atomic-scale density functional theory and molecular dynamics computational studies to elucidate fundamental material properties and irradiation effects; and phase field meso-scale models to bring all of this information together to interpret the mechanisms driving microstructural evolution. Recent experimental and computational results related to tritium release from the LiAlO₂ surface and gas-phase transport within the TPBAR will also be discussed.

The modelling efforts for the cladding are less mature than for the pellets, but a combination of experimental data and computational results are providing significant insight into the irradiation performance of this material also. Key findings to date include in-reactor measurement of tritium permeation enhancement in 316 stainless steel that may be related to the interaction of tritium interstitials with irradiation-induced defects as suggested by recent atomistic modelling studies. The aluminide coating process causes some sensitization with associated chromium carbide precipitation at the grain boundaries, and irradiation enhances the Cr depletion as well as causing radiation-induced segregation of Ni and Si to dislocation loops. Void nucleation in Cr_3Si precipitates in the aluminide



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coating have also been observed in irradiated cladding. All of these radiation-induced features are possible tritium trapping sites that could affect in-reactor performance of the TPBARs. Finally, studies focusing on the near-surface region of the aluminide coating offer clues about tritium and lithium transport within the TPBAR that could have implications for tritium permeation through the cladding during irradiation.

Keywords: tritium production, tritium transport, tritium trapping, microstructural evolution



TRITIUM TRAPPING AND RELEASE IN W MATERIALS: IMPACT OF TUNGSTEN OXIDE

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The Plasma-facing materials (PFM) for next generation fusion devices like ITER will be submitted to intense fluxes of light elements, notably He and H isotopes (HI). Interaction of the first wall materials with particles escaping the magnetic confinement is particularly significant for the divertor components; and tungsten (W) was chosen for ITER due to its low sputtering yield, low HI retention and high melting point. However, the extreme plasma conditions at the divertor can jeopardize those favourable properties, triggering concerns for the reactor efficiency and safety: notably, incident He particles can drastically affect the surface, with the formation of dislocation loops, bubbles or W-fuzz [1]. Retention/permeation of HI that are crucial processes to monitor since fusion reactors operate with tritium (T) [2]. The W surface properties plays a major role on HI retention/permeation being the physical interface between the vacuum vessel, the plasma and the material. Yet, W PFM present a native oxide layer at the surface that can survive during conditioning and operational conditions. Little works exist on the consequences of this oxide layer on tritium trapping: does it introduce new traps for hydrogen, does it act as a diffusion barrier? These are the questions we are trying to answer in this work. With one objective: to improve our modelling effort and thus achieve a better evaluation of the long term inventory in W PFC of ITER with the help of our predictive rate equation codes. In that frame, we coupled a multi-scale material characterization of W samples with tritium inventory studies led at the Saclay Tritium Lab. Thus, we developed an oxidation protocol to create stable enhanced W oxide layers (200-300 nm thick) which is compared with unoxidised polycrystalline W sample and a W sample with native oxide, These materials are fully characterized by different techniques as Raman spectroscopy, Electron BackScatter Diffraction, Scanning Electron Microscope, Focused Ion beam and Transmission Electron



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Microscope. Then we exposed the W samples to tritium gas-loading, which does not introduce any damages in the material and helps to characterize the trapping sites originally present. Tritium desorption is measured by Liquid Scintillation counting (LSC) at ambient and high temperatures (800°C), the final step being full material dissolution to reach the total T inventory. It appears that T trapping is strongly dependent on surface condition, with a major change in the desorption dynamic at room temperature: only 3 days are needed to desorb 75% of all T trapped in un-oxidised W, whereas it extends to 30 days in the enhanced W oxide case. Based on data analysis, we will show that this room temperature desorption indicates original trapping sites with low energy e.g 1.13 eV. In this paper, we will also present experimental results of deuterium thermo-desorption spectrometry confirming the above-observed results. Finally, we will assess the consequences of such outcomes on the time dependent tritium inventory in W PFM.

Keywords: Tritium inventory, fusion reactors, first-wall materials, tungsten, oxide

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TRITIUM AGING OF REGENERATED LANA.75

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The Savannah River Tritium Enterprise (SRTE) has used LaNi4.25Al0.75 (LANA.75 or LANA) as a hydride material to store tritium for over two decades. LANA.75 beds that store significant quantities of tritium have a limited service life due to radiolytic decay of tritium to He-3 within the metal matrix. He-3 has a very low solubility in the metal; it becomes trapped and alters the crystal structure of the hydride. The altered structure causes the formation of a heel of trapped hydrogen isotopes and reduces the reversible capacity of the hydride under normal processing conditions. Depending on tritium exposure history, LANA.75 tritium storage beds are replaced after 8-12 years of service.

It has been shown that heating tritium-aged LANA.75 under vacuum can reverse tritium aging effects, eliminating the heel of trapped hydrogen, and restoring the reversible capacity. An additional benefit is the release of He-3 trapped in the metal. Though promising, further study is needed to ensure there are no unexpected changes to the hydride before this restoration technique is employed in full scale beds.

Isotherms were collected on the previously regenerated sample after approximately two years of quiescent tritium aging. Isotherms were collected at 80, 100, and 120°C. As expected, there was a decrease in the plateau pressure, an increase in plateau slope, and a portion of the "heel" of tritium trapped in the metal had been reestablished. Unexpectedly, it appeared that the plateau had shortened at the higher tritium to metal ratios as well. This is typically seen in older samples. After testing, the sample was reloaded with tritium for continued aging.

Keywords: Tritium Storage, Tritium Aging, Regeneration.



ON THE STUDY OF HELIUM BUBBLE EVOLUTION IN METAL TRITIDE BY ADVANCED TRANSMISSION ELECTRON MICROSCOPY

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Tritium is the key nuclear fuel for nuclear fusion reaction and metal tritides are important functional materials in the field of nuclear fusion energy. The helium atoms generated by the decay of tritium atoms and their evolution seriously affect the macro performance of metal tritides. The formation and residence of helium bubbles will affect the tritium storage properties of metal tritides. The research on the growth mechanism of helium bubbles in metal tritides is an important scientific problem and attracts more attentions. Limited by the radioactivity of tritium and the lack of microanalysis technology for light element atoms such as tritium and helium, the study of helium bubble growth model has been in the stage of phenomenological theory for a long time. With the progress of electron microscopic analysis technology, a new transmission electron microscopic technology for more accurate analysis of helium bubble information is proposed, and a new breakthrough has been made in the research on the microscopic mechanism of helium bubble growth.

Understanding the helium bubble evolution is the key basis to solve the scientific problem in the practical application of metal tritides, which is of great significance to promote the development of science and technology of nuclear energy engineering. In recent years, the micro analysis technology has made remarkable progress. The focused ion beam microscope has solved the difficulty of preparing electron microscope samples of radioactive tritides [1]. The resolution of double spherical aberration transmission electron microscope is better than 60 pm, which helps to reveal the mechanism of helium atom migration and helium bubble growth at the atomic scale [2, 3]. The three-dimensional reconstruction technology of transmission electron microscope provides the three-dimensional spatial number density and interval size distribution information of helium bubbles [4, 5], which avoids the possibility that the two-dimensional plane information may mask the three-dimensional information of helium bubble growth. The resolution of electron energy loss spectrum has reached 0.1 eV, which solves the difficulty of analyzing the pressure in helium bubble and the number density of helium atoms in helium bubble [6-7]. In addition, the preparation technology of ultra-thin electron microscope specimens is necessary to obtain microscopic information at the atomic level. The progresses of low-energy ion thinning technology and electrochemical flash polishing [8] all increase the feasibility of studying the microscopic mechanism of helium bubble evolution. This report will focus on introducing the developed technology for the microscopic analysis of helium bubbles in metal tritide. Some of the major results on the helium bubble evolution in the metal tritide will also be summarized.



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Keywords: titanium tritide, helium bubble, growth mechanism, microscopic study, electron-energy loss spectrum

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THE MEASUREMENT OF HYDROGEN FLUXES WHILE FORMING TUNGSTEN DEPOSITION LAYER AND THE OBSERVATION OF MICROSTRUCTURE

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For the demonstration of D-T fusion reactors using deuterium (D) and tritium (T) as fuels, understanding the behavior of hydrogen isotopes in the plasma facing wall, such as dissolution, diffusion, and re-emission, is an important issue from the perspectives of fuel density control and tritium safety management. Tungsten (W), which has excellent thermal properties, has been studied as a candidate for the plasma-facing material. When highenergy particles are injected into the wall material, the constituent atoms are ejected, called sputtering. W is an element with a high atomic number and the sputtering rate is relatively low, but the effect of sputtering cannot be ignored in consideration of the longterm operation of a fusion reactor. Since the ejected W atoms are deposited on the plasma-facing wall, it is necessary to understand the hydrogen isotope behavior in the deposition layer in addition to the original W wall. It has been reported that the structure of the W deposition layer differs greatly from the original W, and it retains hydrogen isotopes at high concentration [1]. However, there is a lack of quantitative understanding of the re-emission rate to the plasma side and the permeation rate when the formed deposition layer is continuously irradiated with hydrogen isotopes. Since the fuel tritium which permeated through the plasma facing wall is mixed into the coolant, the understanding of the permeation characteristics is important for the safety of fusion power generation systems.

In this study, we observed the hydrogen permeation phenomenon in the process of forming W deposition layer, and quantitatively evaluated the change of permeation flux with time. Hydrogen permeation flux through W deposition layer was measured by using the radio frequency (RF) plasma sputtering device. Hydrogen plasma was generated by applying RF power of 200 W and supplying hydrogen gas (pressure of 60 Pa) in the vacuum vessel. W target was attached to RF electrode and sputtered by hydrogen plasma. Then, sputtered W atoms were deposited on the nickel (Ni) substrate (ϕ 12.8 mm, 0.020 mm). At the same time, hydrogen particles also injected into Ni substrate or W deposition layer. The back side of Ni substrate was connected to quadrupole mass spectrometer (QMS), and the amount of hydrogen gas which permeated W deposition layer on Ni substrate was measured. The hydrogen permeation flux and substrate temperature were measured when the distance between the Ni substrate and the RF electrode was set to be 40 mm and 60



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mm, respectively. After the experiment, the cross section of W deposition layer was observed by transmission electron microscopy (TEM).

Regardless of the distance between the Ni substrate and the RF electrode, there was a tendency for the hydrogen permeation flux to increase rapidly and show a peak, then, it continued to decrease until the end of the experiment. This result can be attributed to the decrease in the injection of hydrogen particles into the Ni substrate due to the increase in the thickness of the W deposition layer and the temperature increase of Ni substrate. As the distance between the Ni substrate and the RF electrode becomes closer, the distance between the tungsten deposition layer and the plasma becomes smaller, and the heat load and the incident flux from the plasma are expected to increase. Therefore, when that distance was 40 mm, the substrate temperature was higher and the hydrogen permeation flux was larger than when it was 60 mm. From the TEM observation of the cross section of the deposition layer, it was found that about 260 nm thick deposition layer was formed and there were differences in the microstructure in the depth direction of the deposition layer. In addition, defects and voids such as crack and cavity were observed. Previous studies have reported that the hydrogen diffusion rate in the W deposition layer is slower and the hydrogen solubility is higher than that in W bulk [2,3]. These differences between W deposition layer and W bulk are caused by the presence of numerous micropores and grain boundaries which were observed in this study.

Keywords: hydrogen isotopes, permeation, tungsten deposition layer

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EVALUATION OF COMMERCIAL AMMONIA DECOMPOSITION CATALYSTS FOR TRITIUM PROCESSING

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In tritium processing, ammonia is formed at room conditions through the radioactive decay of tritium in the presence of nitrogen. Nitrogen is a typical glovebox containment atmosphere for tritium processing, and by way of in-leakage, line breaks, and the use of nitrogen in the process piping, ammonia isotopologues may form. The ammonia may be retained in adsorbent drier beds, as it readily be adsorbed by standard adsorbents. To prevent inadvertent build-up of tritium in the form of tritiated ammonia, it must be decomposed back to hydrogen or hydrogen isotopes and nitrogen. This is traditionally done through the use of packed catalyst beds at high temperatures. At plant-scale tritium processing and fusion facilities, the ammonia decomposition catalyst should be commercially available. Additionally, the catalysts used for ammonia decomposition are typically used in high-ammonia concentration streams for the purpose of generating hydrogen. Results will be presented on the evaluation of four commercially available catalysts for their ammonia decomposition effectiveness, activation temperatures, poisoning resistance, structural stability, and overall suitability for tritium service in high-hydrogen isotopologue concentration processes.



ISSUES ON DUST PARTICLES CONTAINING TRITIUM IN ITER AND DEMO

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The important tritium issues relating to dust particles is that they are floating materials and have higher residual tritium concentrations than fixed bulk tiles in fusion reactors. These dust particles have a wider diameter, including invisible sizes to human eyes. Because it is floating material, it reached the reattached areas through interactive processes with plasmas from produced source areas of plasma-facing walls. In the case of dust particles adsorbed by electrostatic force, they remain stable on the walls.

Toward the next-step fusion devices such as ITER and DEMO, it is required that quantitative analyses of reduced tritium and retained tritium in the plasma vacuum vessel, tritium decontamination methods for plasma-facing material, protections of remote handling systems for in-vessel maintenance, so on.

It has been investigated those characterizations of remained dust particles and their tritium residues [1], and many reports were researched including carbon particles. But data on metallic dust particles is required in the case of ITER and DEMO. The produced amount of dust particles in JET-ITER Like Wall (ILW) was two orders of magnitude lower than that in the carbon wall experiments of JET. Also, the total residual tritium of ILW dust particles in the plasma vacuum vessel was two orders of magnitude lower than that in the carbon wall [2]. It was also shown that the tritium accumulation in the ILW materials, W and Be, is lower than carbon [3]. The result is also consistent with the data in the literature by J. Roth [4]. For the protection of the machines, such as remote maintenance systems, from contamination of dust particles, it is necessary to consider the schedules of inserts into the in-vessel. Dust particles of micron or less adhere to surfaces of walls by static electricity force. It is known that one of the factors that cause floating dust particles is when the pressure changes. Therefore, it is preferable to insert the manipulators after it reaches the operating pressure of machines. The details will be discussed in this presentation.

Keywords: Tritium decontamination, quantitative tritium analyses, DEMO, measurement and monitoring

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TRITIUM ION IMPLANTATION AND GAS SOAKING FACILITY FOR MEASURING RETENTION AND OUTGASSING FROM FIRST WALL MATERIALS

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Presented are results from the inactive commissioning of a new facility within the Hydrogen-3 Advanced Technologies (H3AT) Department at the Culham Centre for Fusion Energy. The facility can be used to measure retention and outgassing from various plasma facing materials, such as tungsten or beryllium tiles, after in situ tritium ion implantation or neutral tritium gas soaking at temperatures up to 1000°C. Such plasma facing materials become damaged during fusion pulses, which affects the amount of tritium retention. The knowledge of tritium retention and outgassing in plasma facing components is of great importance for JET and ITER operation, particularly because ITER will use a larger tritium inventory than JET which will enhance the negative effects of tritium retention. Plasma damage leads to increased outgassing timescales which has significance for maintenance safety.

To study tritium retention in ion and molecular form, two laboratory-scaled facilities have been designed by CCFE and ENEA: the Tritium Loading Facility (TLF) and the Tritium Soaking Facility (TSF). Tritium retention and outgassing after implantation of energetic tritium ions into components will be studied in the TLF, while tritium retention in divertor materials will be investigated with molecular tritium in the TSF. Both facilities can handle small material samples of mm thickness or larger component size samples of cm thickness. The capability of in situ outgassing measurements is a first for the H3AT department.

This work presents the design of both the TLF and TSF with all their ancillary equipment and components, as well as the first commissioning results obtained for tungsten using deuterium gas.

Keywords: Tritium retention, Tritium outgassing, First wall materials, JET3 project, Tungsten



EVALUATION OF HYDROGEN ISOTOPE RETENTION BEHAVIOR AND IMPURITIES DEPOSITION FOR PLASMA EXPOSED TUNGSTEN SAMPLES IN QUEST

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W (tungsten) is one of candidate PFMs (plasma facing materials) for future fusion reactors due to its higher melting point, lower sputtering yield and lower hydrogen solubility. In the actual fusion conditions, 14 MeV neutron will be produced by D (Deuterium) - T (tritium) fusion reaction and will be irradiated to W at temperature above 623 K. In addition, irradiation damages will be introduced into W accompanied with impurity deposition. The introduction of irradiation damages by the neutrons and charge-exchanged particles will change the hydrogen isotope retention property. QUEST (Q-shu University Experiment with Steady-State-Spherical Tokamak) at Kyushu University has several unique features, in especially only H plasma experiment (no helium discharge was done), all metal plasma facing walls and higher wall temperature of 473 K. Therefore, the evaluation of hydrogen isotope behavior for plasma facing wall in QUEST will be quite useful to understand the dynamics of fuel retention in future fusion reactor. Polycrystalline W samples (10 mm[®], 0.5 mm^t, 99.99% purity from A.L.M.T. Corp. Ltd) and were preheated at 1173 K for 30 minutes under the vacuum less than 10^{-6} Pa to remove impurities and damages. These samples were exposed to QUEST H plasma during 2019A/W(Autumn/Winter) campaign. The samples were installed on the plasma facing wall located at Top, Equator and Bottom walls. The unique future of 2019A/W was long term discharge (~ 21 mins), and wall temperature was 473 K. After the H plasma exposure in QUEST, the chemical state and the depth profile of atomic concentration were evaluated by XPS (X-ray photoelectron spectroscopy) using the combination of Ar⁺ sputtering technique at Shizuoka University. In addition, TEM (transmission electron microscopy) observation was also performed at Kyushu University. To evaluate the enhancement of D retention after exposure of QUEST H plasma, additional 1 keV deuterium ion (D_2^+) implantation was performed with the ion flux of 1.0×10^{18} D⁺ m⁻² s⁻¹ up to the ion fluence of 1.0×10^{22} D⁺ m⁻² at room temperature. The H and D desorption behaviors were evaluated by TDS (thermal desorption spectroscopy) from room temperature to 1173 K with a heating rate of 0.5 K s⁻¹. The H TDS results showed the samples installed on Top and Equator walls had the higher H retention. For D retention, major D desorption was found at 800 – 1100 K for the sample on Equator wall, indicating that most of D was trapped by deposition layer due to thick deposition. However, most of D was desorbed at the temperature less than 700 K for Top and Bottom samples, where thin deposition region.



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This fact indicates that most of D was trapped by W. In the presentation, correlation of hydrogen isotope retention with chemical state on the surface will be discussed in detail, based on TDS and XPS results.

Keywords: Tungsten, Hydrogen isotope retention, Plasma exposion



EVALUATION OF TRITIUM DISTRIBUTION IN BE LIMITER TILES USED IN JET ITER-LIKE WALL CAMPAIGNS

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In order to test plasma-wall interactions in ITER tokamak, Joint European Torus (JET) in U.K. has been operated with ITER-like wall (ILW) where beryllium was used in the main chamber as limiters whereas components made of bulk tungsten and tungsten coated carbon-fiber composite (CFC) were installed in the divertor region. The JET-ILW experimental campaigns were carried out with deuterium plasma three times since 2011: ILW-1 in 2011-12, ILW-2 in 2013-14 and ILW-3 in 2015-16.

After each campaign, selected tiles were retrieved from the tokamak and analyzed for plasma-wall interaction studies [1]. Tritium retention and distribution in JET-ILW beryllium limiter tiles were evaluated by applying beta-ray induced X-ray spectrometry (BIXS) and imaging plate (IP) technique after ILW-1 and ILW-3 [2,3]. Tritium retention was higher in tiles used in third campaign and tritium distribution was different to that of deuterium which indicated different retention mechanism between tritium and deuterium.

In succession to previous study, tritium inventory in selected castellation pieces cut from beryllium limiter tiles were examined in this study under the framework of Broader Approach Activities. The castellation pieces were parts of limiter tiles, i.e., inner wall guard limiter (IWGL), outer poloidal limiter (OPL) and upper dump plate (DP), exposed to plasma during ILW-1 or ILW-3 or all three campaigns (ILW-1,2,3). In the plasma-facing surface, OPL showed the highest tritium concentration and the concentration increased as the tokamak operation time extended. On the other hand, tritium retention in IWGL and DP tile pieces did not show simple increase with plasma exposure time. Analysis of X-ray spectra with consideration of generation and attenuation of X-rays in the sample indicated implantation of high energy tritium ions to plasma-facing surfaces up to depth of several micrometers. The castellations pieces with high tritium concentration in the plasma-facing surfaces



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presented high tritium concentration also in castellation grooves. However, unlike plasmafacing surfaces, tritium was dominantly retained by deposition in the castellation grooves.

Keywords: tritium, JET, ITER-like wall, beryllium limiter, tritium retention

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TRITIUM EMBRITTLEMENT OF NICKEL-BASED ALLOYS

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Tritium exacerbates hydrogen's embrittlement of structural materials with the additional effect of its helium decay product. Helium is not soluble in the atomic lattice, and when bred in through tritium decay leads to the formation of high-pressure nanoscale helium bubbles which impede dislocation motion and harm mechanical performance. The tritium-decay-derived-helium bubble distribution evolves with time and further decreases fracture toughness. Primary containment material mechanical performance is critical for tritium handling, processing, and storage, which are enabling capabilities for fusion energy. Savannah River National Laboratory has an ongoing program studying tritium embrittlement. Traditionally, austenitic stainless steels are the gold standard for resisting the embrittling effects of hydrogen and tritium. However, these are not suitable for all applications. For high temperature strength and creep resistance, high carbon variants and nickel-based alloys are frequently preferred in pressure vessel design. However, little to no data is available in the literature for their performance in tritium. Thus, a testing program systematically combining some of these environmental conditions has been initiated. Specifically, 347H stainless steel, as well as Inconel 625, Hanes 230, and Hastelloy X were tested in the as received condition, heat treated, and precharged at 623K with 34.5 MPa hydrogen and tritium. Here, we present initial results on tensile testing of the baseline, high temperature aged, hydrogen precharged and the first age of tritium precharged specimens. Deuterium permeation data in these same materials is also presented.

Keywords: Tritium-material interactions, Mechanical Properties, Embrittlement.



COMPARATIVE ANALYSIS OF THE RESULTS OF REACTOR EXPERIMENTS WITH TWO-PHASE LITHIUM CERAMICS (25%Li2TiO3-75%Li4SiO4)

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In the presented work the comparative analysis of two different experiments on irradiation of lithium ceramics (25%Li2TiO3-75%Li4SiO4) is presented. Particularly, the dependences of helium and tritium-containing molecules release from the investigated samples, in the process of irradiation, under different conditions are analyzed. (Table 1 shows experiments parameters.)

(The results of the first reactor experiment were given by the authors earlier in [1]).

Table 1. Parameters of reactor experiments with two-phase itilian ceranics					
Experiment	Pebble sizes, μm	Irradiation	Thermal	Fluence of	Range of
		duration,	neutron flux,	neutrons during	investigated
		days	n/(cm2*s)	irradiation, n/cm2	temperatures, C
Nº1	250 -1250	4.3	0.5-5*10 ¹³	1.9*10 ¹⁹	250-750
Nº2	500 -710	12	0.5-5*10 ¹³	5.2*10 ¹⁹	250-685

Table 1. Parameters of reactor experiments with two-phase lithium ceramics

Qualitative differences observed in the release of helium and tritium from different (in diameter) pebbles of two-phase lithium ceramics were found and described.

In work the formal model describing process of release of tritium and helium from samples of lithium ceramics during reactor irradiation is presented, and also parameters of release of these gases are determined.

Comparison of the obtained dependences of tritium and helium release from samples of lithium ceramics with the calculated data is carried out.

Keywords: Tritium, helium, lithium ceramic, neutron irradiation, fusion reactor, blanket.

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STUDIES CONCERNING TRITIUM RETENTION IN CONTROLLED/ UNCONTROLLED W DUST MORPHOLOGIES, AND WITH/ WITHOUT MICROWAVE (2.45 GHz) PLASMA TREATMENTS

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Tritium retention inside fusion related materials represents a crucial topic, due to the development of fusion facilities (e.g., ITER, SPARC, and so on). Here in, a thermonuclear plasma will interact with the inner walls, producing local damages, especially dust. In our paper, we have studied the Tritium retention in Tungsten (W) dust, with or without microwave (2.45 GHz) plasma treatments. For these experiments we have used two sets of W dust: dust with uncontrolled shapes and with a mean diameter of 500 nm, and dust with controlled shapes (spherical shapes) and with a mean diameter of 5 µm [1]. W dust was exposed at different Tritium gas pressures (40 mbar, 100 mbar and 400 mbar) with the protocol described in reference [2]. From these experiments we have established the Tritium release activity trend at room temperature. The second part was to remove the Tritium from W dust via Hydrogen gas exposure, for decontamination process as theoretically predicted [3]. In this line, the tritiated dust was exposed in 1 bar of Hydrogen gas during 18h, for isotopic exchange purpose. After 18h, the dust was dissolved and measured the remaining Tritium activity. Afterwards, other experiments were dedicated to Hydrogen plasma treatments applied to both dust sets (with uncontrolled and controlled shapes) before tritiation process, in order to enhance the Tritium retention inside W dust. Here on, the plasma treatment parameters include an applied power = 100 W, Hydrogen gas pressure = 7 mbar, W dust quantity = ~40 mg, and a total plasma exposure time = 44 min. After plasma treatments, the dust was exposed to 100 mbar Tritium gas, for loading process. W initial and plasma treated dust, was investigated via Scanning Electron Microscopy and X-ray Photoelectron Spectroscopy. Tritiated W dust was analysed via room temperature desorption and dissolutions methods [1]. The plasma discharge was analysed via optical emission spectroscopy measurements.

Results have highlighted the fact that, by exposing W dust with uncontrolled shapes, to 400 mbar Tritium gas, the release of the tritiated dust will be in two steps during 12 days. Moreover, by



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treating the same type of W dust to a Hydrogen plasma before tritiation process, it can be enhancing the capacity of dust to retain Tritium inside them. By exposing W dust to a plasma, the specific surface area (SSA) will be increased, resulting a high retention of Tritium inside them.

Keywords: Tritium retention, tungsten dust, microwave plasma treatment, detritiation, tritiated dust

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ACHIEVEMENTS ON THE INTERACTION OF HYDROGEN AND HELIUM WITH MATERIALS

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The evolution of hydrogen and its isotope and helium bubble in structural and functional materials have always been bottlenecks restricting the development of nuclear engineering, including the properties of hydrogen and tritium storage, the tritium isotope separation, tritium diffusion and transport, tritium retention and prevention, and the helium bubble evolution in the tritium-related materials [1-3]. With the advancement of the ITER [1] and CFETR [2] major research and development plans, the improvement of the performance of tritium-related materials and the development of novel tritium-related materials have attracted more attentions. As an important collaborator of ITER and CFETR plans, we have made some achievements in the field of interaction of hydrogen and helium with materials in recent years [4-8].

A variety of advanced electron microscopy techniques have been developed and applied to study the helium bubble evolution in metal tritides. The helium bubble pressure [9], bubble density [10] and local residual stress have been obtained, which helps to disclose the microscopic state of helium and establish the the evolution mechanism of helium bubbles in metal tritides [7-8]. A series of Ti-Zr-Hf-Mo-Nb high-entropy alloys (HEAs) have been developed for tritium storage, which has good tritium storage capacity, kinetics, anti-disproportionation as well as the anti-poisoning performance [4-6]. The Ti-Zr-Hf-Mo-Nb HEAs is more resistant to He bubble formation, which may have resulted from the suppressed He clustering, weaker coarsening effect, and severe lattice distortion in HEA [5]. An adsorption analyzer was employed to perform the absorption experiment of palladium hydrides [11], the relationship between pressure and the content absorbed strictly followed the Langmuir equation in the hydride β -phase, which confirmed that the hydrogen atoms in the octahedral interstices follow a monolayer coverage model. Cooperated with the analyzer, an on-line micro gas chromatography (on-line μ GC) technique has been developed and applied to study hydrogen isotope effect in metal hydrides, zero effect of hydrogen isotope was investigated in LaNi₅-H(D) [12]. The adsorption and diffusion of hydrogen at M-doped (M=Pd, Cu, Ir, Mo, Zr, Y, Al, Mg, Si) Ti(0001) surfaces are investigated by first-principles calculations. All the involved M dopants, especially Si, hinder the hydrogen adsorption at Ti surface. The optimal hydrogen diffusion path at M-doped Ti(0001) surfaces are predicted. Despite of the passive effect on hydrogen adsorption, these M dopants generally decrease the hydrogen penetration barrier diffusion from the surface site to the sublayer interstice and Pd performs most



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significantly. The above achievements play an important role in the development of nuclear fusion energy.

Keywords: Hydrogen behavior, Helium bubble evolution, Isotope separation, High-entropy alloy, Microscopy analysis.

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HYDROGEN ISOTOPE EFFECT ON PLASMA DRIVEN PERMEATION FOR NEUTRON DAMAGED W

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Evaluation of the physical parameters related to hydrogen isotope transport through plasma-facing materials is required to design future fusion reactors. To accurately predict hydrogen isotope behavior, damage by neutrons and energetic charged particles, combined with He ash effects, should be extensively studied at fusionrelevant higher temperatures. In our previous studies, D retention in tungsten (W) was enhanced by neutron irradiation due to the uniform damage distribution. The D permeation rate for damaged W was reduced due to reduced D permeation path. For the detailed design activity for DEMO, the isotope effect on retention and permeation by concurrent hydrogen isotope (H/D/T) exposure at fusion relevant temperatures must be evaluated. Consideration of these factors, along with the effects of displacement damage and He irradiation conditions is being pursued under the Japan-US collaborative research, FRONTIER, and Japanese domestic research. In this study, 50:50 H and D mixed plasma exposure was performed for neutron damaged W and plasma driven permeation (PDP) behavior was evaluated. After PDP experiment, H and D retentions were also observed by thermal desorption spectroscopy (TDS). These permeation and retention behavior for neutron damaged W was compared with that for undamaged W and the role of irradiation damages on hydrogen isotope transport was discussed.

For the PDP experiment, large temperature dependence on H and D permeation was observed for undamaged W and large H permeation was found at lower temperature, 450 K, H and D permeation fluxes were almost 2:1. However, as the temperature was increased up to 650 K, they became almost unity. The TDS results showed the H and D retention was almost the same. For neutron damaged W, H and D permeation fluxes were almost 3:2. Even if the temperature was increased, no large change was observed. But higher D retention compared to H was found by TDS. These



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results showed that the irradiation damage may contribute on hydrogen isotope transport and the details will be discussed in this presentation.

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Keywords: Hydrogen isotope, isotope effect, plasma driven permeation, neutron damaged W.



IMPACT OF IRRADIATION ON FUEL RETENTION IN FUSION REACTOR MATERIALS

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Accurate evaluation of tritium inventory in a vacuum vessel is one of the most important issues for a safety assessment of a future fusion reactor. Current design of a fusion demonstration reactor (DEMO) uses tungsten as the plasma-facing material and reduced activation ferritic/martensitic (RAFM) steel as the structural material. These materials will be exposed to 14 MeV neutrons produced by DT fusion reactions. One of the possible mechanisms underlying accumulation of tritium in a vacuum vessel is penetration of tritium into these materials and trapping at defects induced by neutron irradiation.

To understand hydrogen isotope trapping in neutron-irradiated tungsten and RAFM steel, the author and colleagues have examined the hydrogen isotope retention in neutron-irradiated tungsten [1-3] and self-ion irradiated RAFM steel [4,5]. The objective of this presentation is to discuss possible tritium inventory in a vacuum vessel of DEMO reactor based on the data reported in [1-5] by considering the amounts of tungsten and RAFM steel expected to be used in a vacuum vessel and temperature distributions.

The hydrogen isotope retention in tungsten and RAFM steel significantly increased due to trapping at defects induced by irradiation. However, the trapping in RAFM steel was far weaker than that in tungsten, and the accumulation of tritium in RAFM steel was evaluated to be moderate at the expected service temperature (~300°C or higher). The tritium content in tungsten was also evaluated to be small near the plasma-facing surface due to high temperature (>1000°C). However, in the case of water-cooled tungsten monoblock divertor, the temperature of tungsten near cooling pipes should be ~300°C, and significant tritium accumulation was expected in this region by the combination of relatively low temperature and strong trapping of hydrogen isotopes at radiation-induced defects in tungsten. Namely, the "cold parts" of tungsten can play dominant roles in tritium inventory if tritium diffuses to this cold part under long-term exposure to steady state DT plasma. Effectiveness of bake out process for tritium removal will be also discussed.

Keywords: tritium inventory, vacuum vessel, radiation-induced defects, trapping, plasmafacing material, structural material

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THEORETICAL INVESTIGATION OF TRITIUM DISTRIBUTION IN IRRADIATED GRAPHITE SPHERES OF HIGH TEMPERATURE GAS-COOLED REACTORS

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The very high temperature gas-cooled reactor (VHTR) as well as its prototype high temperature gas-cooled reactor (HTGR) has been considered as one of the six promising types of Generation IV nuclear systems which has broad prospects in hydrogen production, cogeneration, and process heat application [1]. The tritium has been identified as one of the dominant radioactive nuclides in the primary loop and secondary loop in HTGRs. The tritium distribution in irradiated graphite spheres from the reactor core of the 10 MW high temperature gas-cooled test reactor (HTR-10) and Arbeitsgemeinschaft Versuchsreaktor (AVR) have been determined experimentally [2-5]. Both results show that the tritium activity concentration is higher at the surface of the irradiate graphite spheres, while in the interior, the tritium activity concentration is nearly even distributed. A typical tritium activity distribution in the irradiated graphite sphere is shown in Fig. 1 [3]. Previous studies attributed this phenomenon as a strong interaction between tritium in the helium and the graphite matrix of irradiated spheres. In this study, a theoretical model will be established to describe the boundary between tritium in the helium and surface of an irradiated graphite sphere. We will simulate the tritium accumulation process at the surface of the sphere and illustrate the interaction between tritium and graphite based on the molecular dynamics theory and first-principles calculations. This study can provide a preliminary investigation on the mechanism of interaction between tritium and graphite surface from a microscopic view and supply a reference for the research on tritium sink in the primary loop of HTGRs.



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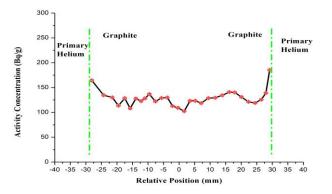


Figure 1 – Experimental results of ³H activity distribution in an irradiated sphere of HTR-10 [3]

Keywords: High temperature gas-cooled reactor, irradiated graphite spheres, tritium, molecular dynamics

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ACTIVATION OF GROUP III AND IV METALS FOR TRITIUM ABSORPTION

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Thin film samples of several Group III and IV metals, after being exposed to ambient atmosphere, were probed using XPS as a function of temperature. Passive oxide and hydroxide layers were found to form during exposure, with varying composition and relative stability. Upon thermal annealing, the layers were seen to decompose and give way to sub-oxide and suspected carbonaceous species that persisted until higher temperatures. A temperature is identified where the surface has changed character from insulating to primarily metallic when the stable oxide peak had completely disappeared and the metallic peak had emerged. This transition temperature is determined to be 400°C for Zr, 500°C for Y, 400°C for Sc, and 300°C for Ti. After the transition temperature the metal is now prepared to readily absorb all hydrogen isotopes and form stoichiometric metal hydrides.

Keywords: Metal Tritides, Zirconium, Titanium, Yttrium, Scandium

This paper describes objective technical results and analysis. Any subjective views or opinions that might be expressed in the paper do not necessarily represent the views of the U.S. Department of Energy or the United States Government.

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THE GENERATION AND ANALYSIS OF TRITIUM-SUBSTITUTED METHANE

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Tritium, the radioactive isotope of hydrogen, can form a plethora of molecular species. While reaction paths of stable molecules can often be controlled externally, radioactive species tend to undergo radiolysis. An important category of species are tritium-substituted hydrocarbons, which form by radiochemical reactions in the presence of tritium and carbon. In large-scale tritium applications, such as nuclear fusion, these species are unwanted and need to be removed from process gases. In order (i) to test removal procedures; (ii) to determine molecular structure by high-resolution spectroscopy; and (iii) to study fundamental exchange reactions, it is in general necessary to generate various gas species in significant concentrations. Tritium-substituted methane species, CQ_4 (with Q=H,D,T), are often the precursor for higher-order reaction chains, and thus are of particular interest.

 CQ_4 production has been carried out in the CAPER facility [1] of the TLK. The reagents were methane and high-purity tritium. A range of methods, reactors and reaction conditions were tested, and a few additional components were integrated into the system, which allowed us to concentrate synthesis products of interest. For particular reaction-paths the catalyst had to be prepared, activated and reduced with pure T₂ at 400°C for the reaction to take place (see e.g. ref. [2]). Using heterogeneous catalysis (with nickel catalysts) and circulating the gas through the permeator, we synthesized the complete CQ_4 isotopologue family with Q=H,T. By controlling the overall reaction conditions in successive steps during production, we were able to shift the reaction equilibrium towards species with high tritium content, specifically enriching the product molecule CT_4 . In addition, we observed that under certain reaction conditions longer-chain tritiated hydrocarbons [3] were generated.

The CQ₄ samples were transferred to the TRIHYDE facility at TLK, using various allmetal sample cylinders (up to 1000 cm³ at ~850 mbar) and dedicated tritium-compatible Raman cells (volume ~8 cm³ at ~850 mbar; CQ₄-content up to ~25 %), in order to avoid potential (uncontrolled) changes in the gas composition when sending the gas via the TLK tritium transfer lines. Sample analysis was performed using the TRIHYDE analytical instrumentation [4]; besides convenient sample handling and gas processing capabilities, it incorporates the species-sensitive methods of mass spectrometry and Raman spectroscopy



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[5], to determine gas composition. Here, we emphasize the discussion of results from the mass analysis, for species identification and semi-quantification.

Keywords: tritium-substituted methane, mass spectrometry, Raman spectroscopy, measurement and monitoring

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TRITIUM PERMEATION DEPTH STUDY IN FUSION RELEVANT MATERIALS

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Tritium accumulation in fusion relevant materials is a challenge for many areas including waste management and tritium inventory control. The University of Rochester has published tritium permeation depth profiles of tritium soaked 316L Stainless Steel. In collaboration with UKAEA, there are imminent plans to expand this programme to many more fusion relevant materials. There are over 100 samples from 8 different metals including Eurofer 97, ODS, XM-19 as well as erbia coated 304 stainless steel waiting to be analysed. A slew of measurement techniques will be used at both UKAEA and the University of Rochester to assess the surface and depth profile of tritium inside these metals. Analysis methods include acid etching, Secondary Ion Mass Spectroscopy (SIMS), Focussed Ion Beam (FIB) milling and Thermal Desorption Spectroscopy (TDS) amongst others. The University of Rochester also has an Atomic Layer Deposition (ALD) system which can be used to coat samples in anti-permeation coatings to investigate the permeation seen in these. The UKAEA can also employ the DELPHI system to ion damage samples which will exhibit properties more representative of materials exposed to fusion reactor conditions. An opportunity is also offered for the attendees to suggest their own materials to be analysed.

Keywords: Tritium, Permeation, Coatings.



HIGH-ENERGY TRITIUM IONS AND A-PARTICLES RELEASE FROM THE NEAR-SURFACE LAYER OF LITHIUM DURING NEUTRON IRRADIATION IN THE NUCLEAR REACTOR CORE

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In recent years, one of the promising directions in the development of fusion facilities is research, related to the use of lithium [1] and its various compounds [2] as a plasma-facing material. The use of lithium as a plasma-facing material in most cases is realized by using a capillary-porous structure (CPS) as a stabilizing matrix [3]. To ensure effective tritium generation, it is necessary to study its interaction with the elements of fusion reactor systems and foremost with the material itself, in which the tritium will be generated.

In [4] describes experimental studies that show the possibility of using the method of optical diagnostics of high-temperature plasma in fusion reactors. In experiments to study the optical radiation of nuclear-excited plasma conducted at nuclear reactors, gas excitation is usually carried out by using products of exothermic nuclear reactions occurring the interaction of thermal neutrons with ³He, ²³⁵U, ¹⁰B, ⁶Li nuclei [5]. Less studied before our works [6,7] was the use of a nuclear reaction with lithium-6 with thermal neutrons. The large mean free path of tritium nuclei in lithium (130 μ m) and gaseous media (35 cm in atmospheric pressure helium) makes it possible to excite large volumes of gases and provide a larger amount of power nested in the gas in comparison with reaction products with ¹⁰B.

The present paper examines *in-situ* spectroscopic measurements of nuclear-excited plasma of noble gases, in particular, the luminescence of noble gases in a wide temperature and spectral range, excited by the ${}^{6}Li(n,\alpha){}^{3}H$ nuclear reaction products in the core of a nuclear reactor. A thin layer of lithium applied on the walls of the experimental device [8], stabilized in the matrix of the capillary-porous structure, serves as a source of gas excitation. During in-pile tests, conducted at the IVG.1M research reactor, thermal neutrons interact via the ${}^{6}Li(n,\alpha){}^{3}H$ reaction, and the emergent alpha particles with a kinetic energy of 2.05 MeV and tritium ions with a kinetic energy of 2.73 MeV excite gaseous medium. The intensity of tritium release from the lithium layer in noble gases was



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estimated by the intensity of the alpha line of the Balmer series of the tritium atom T_{α} (656.2 nm). The temperature dependence of intensity of the emission of atoms of noble gases and tritium release were studied. A significant tritium release was observed at 673 K due to the beginning of desorption of thermalized tritium atoms dissolved in the liquid phase of lithium.

The results are of interest in terms of clarifying the mechanisms and developing models that allow describing the processes of generation, diffusion, and release of tritium from lithium during neutron irradiation.

This work was supported by the Ministry of Energy of the Republic of Kazakhstan (Program "Development of nuclear energy in the Republic of Kazakhstan for the 2021–2023 years").

Keywords: Emission spectra, lithium capillary-porous system, tritium ions, nuclear reactor.

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EVALUATION OF ALL-METAL ALUMINUM SCROLL PUMP WITH SURFACE PASSIVATION COATING

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In tritium processing, vacuum pumps are used to evacuate tanks, vessels, and lines, as well as transferring gas between parts of the facility. Historically, this was done using a Normetex 15 all-metal scroll pump backed by a Senior Aerospace MB-601 metal bellows pump. However, Normetex was liquidated in 2012 and the pumps were not available. Eumeca, founded by former Normetex engineers, currently fabricates Normetex-style scroll pumps and have been successful. However, after the loss of the only vendor when Normetex was liquidated, an alternative vendor was needed to strengthen the supply chain.

Air Squared has been working with SRNL to develop an all-metal air cooled stainless steel version to replace the Normetex. The increased weight and heat transfer of the stainless steel was a concern. To mitigate this, it was proposed to develop an all-metal air cooled scroll pump made out of aluminum, as well as lower the cost due to ease of machining aluminum. Since aluminum readily forms an oxide layer that can interact with tritium to form an appreciable surface tritium contamination, coatings were investigated to prevent the tritium from interacting with the aluminum oxide surface. Four coatings were compared to bare aluminum to determine surface adhesion during thermal cycling and an optimal coating was chosen for pump fabrication. The fabricated pump then underwent a series of tests for various gases: ultimate vacuum as a function of back pressure, and flow rate as a function of inlet pressure while backed by a MB-601 pump. The gases tested were pure hydrogen, deuterium, helium, nitrogen, argon, and krypton. Results will be presented for the ultimate vacuum and flow tests, as well as evaluation of the stability of the surface coating over time.



CATALYST DEVELOPMENT FOR AMMONIA DECOMPOSITION PALLADIUM MEMBRANE REACTOR IN TRITIUM PROCESSING

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In tritium processing, the energy associated with tritium decaying into 3He is high enough to self-catalyze the formation of tritiated ammonia in the presence of nitrogen. In the operation of fusion reactors, tritiated ammonia will be formed through the contact of the nitrogen divertor shield gas with the hot D/T plasma. Ammonia decomposition is typically performed using fixed catalyst beds at high temperatures. However, the reaction is limited by the reaction equilibrium. In order to decompose ammonia to levels lower than standard equilibrium, one of the decomposition products must be removed. By using a palladium membrane reactor (PMR) the ammonia can be decomposed in close contact to the palladium-alloy tubes, allowing for the quick removal of hydrogen from the process stream and further shifting of the equilibrium to lower decomposition concentrations. The industry-leading catalyst for ammonia decomposition is a Ru-based catalyst, but it requires temperatures greater than 550°C to be active, which is higher than standard Pd-Ag tubes can withstand. By developing a catalyst with lower activation temperatures, it is possible to develop a PMR that can efficiently decompose ammonia at standard hydrogen permeator temperatures. Results will be presented on the synthesis and evaluation of low-temperature ammonia decomposition catalysts, as well as evaluation of the catalyst in the presence of Pd-Ag membranes.



EXPERIMENTAL TESTS ON THE PERMEATION OF DEUTERIUM THROUGH NICKEL MEMBRANE

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Tritium permeation through structural materials of the fusion reactors is an important issue both from radiological safety and tritium self-sufficiency points of view. [1] Because the fuel of a fusion reactor will consist of a mixture of deuterium and tritium, but also because the Helium Cooled Pebble Bed (HCPB) concept of Breeding Blanket (BB) uses helium gas with a small addition of hydrogen as purge gas in order to promote the release and extraction of the tritium bred, [2], mutual influences among isotopes have to be taken into account in the assessments of permeation fluxes. Various theoretical and experimental works regarding the permeation of hydrogen isotopes through metals have been carried out so far, some of them being focused on this kind of synergistic isotope effect. However, there still are some controversies regarding the mutual influence among isotope species in the multi-isotope permeation. [3] The processes of dissociation and recombination at the surface of the metal have a decisive role in the occurrence of this isotopic effect, therefore multi-isotope permeation studies should be performed in the limited surface regime of permeation (SLR). As pointed out in other previous works, [4, 5], carefully designed experiments and accurate measurement methods are required to be able to (experimentally) highlight the small differences between multi-isotope and mono-isotope permeations (especially in SLR where driving pressures should be very low, and hence the measured permeation flows are very small). That is why in a previous work a versatile experimental installation has been design and realized, its multi-purpose features being described elsewhere. [5]

This paper presents the experimental testing of the gas-driving permeation of deuterium through thin membrane of nickel in the temperature range of 200° C- 500° C, and for driving pressures within the range of 10^{-1} Pa-10 Pa, so that the permeation can be very well described by the surface limited regime (SLR). In order to improve the measurement accuracy, the calibration of the measuring instruments where performed by injecting of known (and very low) flows of deuterium (in the range of 10^{-13} mol/s -10^{-10} mol/s) directly into the permeation cell where the testing membrane of nickel were replaced by a thick copper disk. The results obtained in this first campaign of experimental testing are extremely useful for the continuation of the proposed work as they are the reference basis for comparison with the results given by multi-isotope permeation experiments (the future planned campaigns of experimental testing), so that influence of an isotope species on the permeation of another could be emphasized experimentally. Also, the (mono-isotope) transport coefficients for the surface (dissociation and recombination) processes, experimentally determined in the here presented test conditions, will be used as input data to the



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theoretical models of multi-isotope permeation, so that they can be benchmarked against the experimental results given by the future experimental tests on multi-isotope permeation..

Keywords: Deuterium/Tritium permeation, isotope effects, fusion structural materials, fusion safety.

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HYDRIDE BED PERFORMANCE: JUXTAPOSITION OF *LANA85* AND *LANA75* HYDRIDE BED ABSORPTION RATES, CAPACITY, AND OPERATIONAL CAPABILITIES

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Savannah River Tritium Enterprise (SRTE) has used LaNi4.25Alo.75 (LANA75) hydride beds to store hydrogen isotopes for over 2 decades.

One benefit of using LANA75 in hydride storage beds is that helium-3, produced from the decay of tritium, is retained in the hydride material. Bed overpressure is pulled prior to transfers allowing the hydride beds to deliver high purity product gas. One disadvantage is that over time, the helium-3 accumulates in the material, resulting in the formation of a heel that cannot be removed under normal operating conditions. This heel traps hydrogen in the bed which reduces its operational capacity. In addition, the helium-3 eventually begins to evolve from the material, preventing the delivery of high-purity product.

SRTE plans to replace four hydride beds. Two beds contain LaNi4.15Alo.85 (LANA85), and two contain LANA75. Overall, LANA85 performs similarly to LANA75 as it readily forms a hydride when introduced to hydrogen isotopes, and retains helium-3 produced from tritium decay. However, LANA85 and LANA75 differ in a few ways; these slight differences can be utilized at ambient temperatures to aid operational efficiency.

Based on the isotherm at ambient temperature generated by the Savannah River National Laboratory, LANA85 has a lower plateau pressure in the α +ß phase than LANA75. This occurrence is expected to improve operational capabilities by holding onto more product gas during overpressure evacuations. The LANA75 and LANA85 bed performances will be tracked over several months and key parameters such as hydrogen absorption rates and capacity will be monitored.



EVALUATION OF TRITIUM DISTRIBUTION IN BE LIMITER TILES USED IN JET ITER-LIKE WALL CAMPAIGNS

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In order to test plasma-wall interactions in ITER tokamak, Joint European Torus (JET) in U.K. has been operated with ITER-like wall (ILW) where beryllium was used in the main chamber as limiters whereas components made of bulk tungsten and tungsten coated carbon-fiber composite (CFC) were installed in the divertor region. The JET-ILW experimental campaigns were carried out with deuterium plasma three times since 2011: ILW-1 in 2011-12, ILW-2 in 2013-14 and ILW-3 in 2015-16.

After each campaign, selected tiles were retrieved from the tokamak and analyzed for plasma-wall interaction studies [1]. Tritium retention and distribution in JET-ILW beryllium limiter tiles were evaluated by applying beta-ray induced X-ray spectrometry (BIXS) and imaging plate (IP) technique after ILW-1 and ILW-3 [2,3]. Tritium retention was higher in tiles used in third campaign and tritium distribution was different to that of deuterium which indicated different retention mechanism between tritium and deuterium.

In succession to previous study, tritium inventory in selected castellation pieces cut from beryllium limiter tiles were examined in this study under the framework of Broader Approach Activities. The castellation pieces were parts of limiter tiles, i.e., inner wall guard limiter (IWGL), outer poloidal limiter (OPL) and upper dump plate (DP), exposed to plasma during ILW-1 or ILW-3 or all three campaigns (ILW-1,2,3). In the plasma-facing surface, OPL showed the highest tritium concentration and the concentration increased as the tokamak operation time extended. On the other hand, tritium retention in IWGL and DP tile pieces did not show simple increase with plasma exposure time. Analysis of X-ray spectra with consideration of generation and attenuation of X-rays in the sample indicated implantation of high energy tritium ions to plasma-facing surfaces up to depth of several micrometers. The castellations pieces with high tritium concentration in the plasma-facing surfaces



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presented high tritium concentration also in castellation grooves. However, unlike plasmafacing surfaces, tritium was dominantly retained by deposition in the castellation grooves.

Keywords: tritium, JET, ITER-like wall, beryllium limiter, tritium retention

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PLASMA-DRIVEN PERMEATION WITH NOBLE GAS ADMIXTURES

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One major design driver for the fuel cycle of a DEMO reactor is the reduction of the tritium inventory. To this end, the Karlsruhe Institute of technology (*KIT*) has developed a smart fuel cycle architecture under the *Direct Internal Recycling* (*DIR*) concept. It foresees the separation of a large fraction of hydrogen from the tokamak exhaust with direct recycling back to fueling, bypassing the tritium plant. Prime candidate to fulfil the separation task is a metal foil pump (*MFP*), which employs plasma-driven permeation.

This technology relies on the interaction of two main components: A metal foil of a group 5 metal and a source of suprathermal hydrogen. In theory, plasma-driven permeation is capable of producing fluxes similar in magnitude to those of the impingement rate of particles on a given metal foil. The specified metal materials can naturally grow non-metallic impurity monolayers, which impair molecular hydrogen absorption but are, to some degree, transparent to suprathermal, i.e. atomic hydrogen. In this way, the plasma-facing side of a metal foil can feature large absorption fluxes, which lead to high desorption fluxes on the rear side, where only negligible amounts of molecular hydrogen are re-absorbed. We investigate plasma-driven permeation using a scalable cylindrical microwave plasma source from industry in our experimental facility HERMES*plus* at *KIT*.

In this work, we give an introduction to the working principle of a *MFP* and its application in *DEMO*. We present parametric studies of the permeation flux and its dependence on operation parameters like foil temperature, pressure in the plasma chamber and microwave plasma power. We demonstrate the plasma-driven permeation fluxes independence on hydrogen isotope using protium and deuterium as feed-gas. Furthermore, the results of admixing noble gases according to the tokamak exhaust gas composition will be showcased as they will be processed by the *MFP*.

Keywords: Direct Internal Recycling, DEMO, Metal foil pump, Plasma-driven permeation, Tritium separation



DETRITIATION OF LOW-LEVEL TRITIATED WATER USING HYDROGEN BONDED POLYMER: AN EXPERIMENTAL AND THEORETICAL STUDY

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We have experimentally and theoretically investigated the detritiation performance of polymers for low-level tritiated water (5 x 10^5 Bq/L). Twenty-one types of polymers (Fig. 1) containing hydrogen-bonding functional group were evaluated in terms of tritium uptake capacity and separation factor through batch experiments. Among the polymers, p-toluenesulfonyl hydrazide (CH₃C₆H₄SO₂NHNH₂), sulfonyl amide (S(=O)₂-NH₂), and 2-chlorotrityl chloride (C₁₉H₁₄Cl₂) showed the higher tritium uptake capacity than other types of polymers as 39.8±0.14 Bq/mmol, 68.0±7.57 Bq/mmol, and 45.4±4.17 Bq/mmol, respectively. Separation factor (α) of the polymers were calculated according to the equation:

$\alpha = (\text{Tritium}_{\text{polymer}} / \text{Hydrogen}_{\text{polymer}}) / (\text{Tritium}_{\text{solution}} / \text{Hydrogen}_{\text{solution}})$ (1)

The values were 3.8 ± 0.02 for p-toluenesulfonyl hydrazide; 9.9 ± 1.27 for sulfonyl amide; 5.5 ± 0.50 for 2-chlorotrityl chloride. The bonding properties of tritium on the hydrogen position of the functional groups were studied using density functional theory at the B3LYP/6-311G** level. Two parameters influencing tritium uptake performance by polymers were calculated, 1) zero point energy differences between hydrogen and tritium and 2) hydrogen binding energy on hydrogen position of functional groups. The results revealed that polymers with higher tritium uptake capacity has lower hydrogen binding energy implying lower binding energy of hydrogen to the bonding site of functional group might facilitate isotope exchange between H+_{polymer} and T+_{solution}.

Keywords: Tritium separation, Hydrogen bonding polymer, detritiation of tritiated water, hydrogen binding energy, zero-point energy



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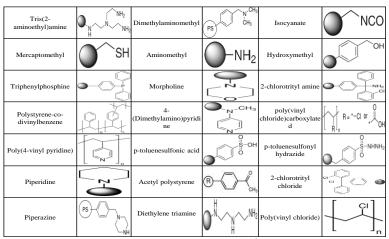


Figure 1. Various types of polymers containing H^{+} bonding functional group

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REACTOR STUDIES OF TWO-PHASE LITHIUM CERAMICS 35%Li₂TiO₃-65%Li₄SiO₄

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This work presents data on the study of gas release from two-phase lithium ceramics ($35\% \text{ Li}_2\text{TiO}_3-65\% \text{ Li}_4\text{SiO}_4$) under neutron irradiation conditions.

Two types of ceramic pebbles were investigated: one type with pebble sizes 250-1250 microns, the other one with pebble sizes 500-710 microns.

Irradiation was carried out at the WWR-K reactor (Almaty, Kazakhstan) for ~21 days (for each campaign) at a ceramic temperature of ~665°C. The total fluence during irradiation was ~9.10¹⁹ n/cm² for each campaign. In the course of irradiation, experiments were carried out on heating ceramics during irradiation, as well as experiments with hydrogen isotopes supply into the chamber with samples. During the entire irradiation, the gas composition in a continuously evacuated ampoule with samples was recorded.

The dependences of the release of tritium-containing molecules and helium during irradiation are obtained, and a qualitative analysis of the obtained dependencies was carried out

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Keywords: lithium ceramics, fusion reactors, tritium, neutron irradiation





Tracer technics and isotopic effects





REFRACTIVE INDEX MEASUREMENTS OF SOLID HYDROGEN ISOTOPES

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Solid deuterium (D) - tritium (T) mixture has been attracted as a fuel pellet in both inertial confinement and magnetic confinement fusion reactors. However, because of the isotope effect, the triple point temperatures of hydrogen isotopologues are different. The fractionation of hydrogen isotopologues has been noticed during the solidification process [1]. Since the isotopologue fractionation phenomenon occurs, the preparation of homogeneous DT solid at a given composition is one of the challenging issues.

High-quality DT solid fuel pellets are indispensable for the effective ignition of fusion reaction in an inertial confinement fusion reactor. Inhomogeneity of the pellet should result in the deterioration of the fusion reaction. Thus, the characterization method for fuel pellets should be developed to know the isotope distribution and the uniformity of the pellet. The refractive index of DT solid gives the composition of isotopologues, and the mapping of the refractive index in the DT solid shows the uniformity of the pellet.

We aim to develop a dedicated refractive index detection system at cryogenic temperature. The line laser (543 nm) is refracted after passing through a prism cell containing solid DT - Figure 1. By measuring the angle of refraction, the refractive index is evaluated with Snell's law. The temperature dependence of the refractive index and the refractive index as a function of the D/T ratio were observed. Then, by mapping the refractive index in the solid DT, the distribution of DT was cleared. The difference of triple point of hydrogen isotopologue plays an important role in the solidification process.

To confirm the influence of tritium beta decay, the refractive index distribution measurement of the solid protium (H) - deuterium (D) mixture was completed at first. The inhomogeneity in the solid HD mixture was observed and results from the difference of the triple point of the HD mixture. Therefore, the inhomogeneity in a solid DT should appear. The refractive index distribution of the solid DT mixture will be measured to confirm the influence of the decay heat and the decay product (helium-3) of tritium. The study of DT uniformity will encourage the study of the higher-quality DT fuel pellets.

Keywords: Hydrogen isotopologues, isotopic effects, refractive index, deuterium, tritium, fusion fuel, isotopologue fractionation, etc.

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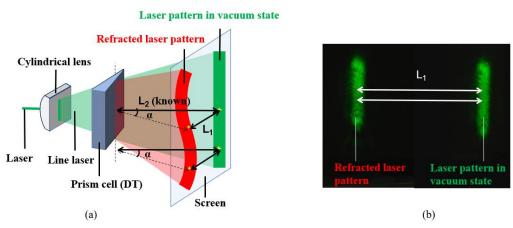


Fig 1 - (a) Schematic of optical measurements (b) Laser pattern on the screen

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STUDY OF HYDROGEN TRAPPING IN BEARING STEEL BY TRITIUM TRACER TECHNIQUE

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Understanding of hydrogen behavior in bearing steels is important to clarify the contributions of hydrogen embrittlement to failure of bearing. Influence of trapping at lattice defects (vacancies, dislocations, etc.) and nonmetallic inclusions (Al₂O₃, MnS, etc.) on hydrogen behavior can be significant, but the details of trapping has not been fully clarified.

A b-particle emitted from tritium atom (T) generates x-rays via interactions with a matter surrounding the T atom. Non-destructive depth profiling of T in a solid is possible by analyzing x-ray spectrum, and this technique has been called as b-ray induced x-ray spectrometry (BIXS) [1]. The range of b-rays from T (\geq 18.6 keV) in a steel is > 1 mm. Hence, the intensities of characteristic x-rays provide information on local chemical compositions around T atoms in regions within the range of b-rays. Hence, in addition to the non-destructive depth profiling, the BIXS technique can be used for investigation of hydrogen trapping at nonmetallic inclusions. This paper reports the results of proof-of-principle study by using a bearing steel.

The samples used in this study were plates of high-carbon chromium steel type SUJ2. The plates were fabricated from rods of the steel water-quenched from 1073 K or 1143 K and tempered at 453 K for 2 hours. After polishing the surfaces to mirror-like finish, the samples were exposed to deuterium (D)-3% tritium (T) mixture gas at 453 K and 2 kPa for 5 hours. The spectra of x-rays generated by b-rays from T were measured using a silicon drift detector under Ar gas atmosphere. Other set of samples were exposed to D₂ gas under the same conditions and subjected to thermal desorption spectrometry (TDS) at the temperature ramp rate of 0.5 K/s.

The x-ray intensity from the samples quenched from 1143 K was far higher than that from the samples quenched from 1073 K. The TDS analyses gave consistent results: the D retention in the former sample was larger than that of the latter. Crystallographic analysis indicated that the larger T/D retention after quenching from 1143 K was due to presence of retained austenite after quenching. In both type of samples, small but clear peaks of characteristic x-rays of Mg, Al and Si were observed in addition to the large peaks of Fe and Cr characteristic x-rays. Monte Carlo simulation of x-ray spectra by assuming uniform distribution of T atoms provided no noticeable peaks of Mg, Al and Si because the concentration of these elements were at impurity level (~0.5 at.% of Si, and >0.02 at.% for Mg and Al). This clear difference between observed x-ray spectra



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and simulated ones showed that trapping of T in nonmetallic inclusions containing Mg, Al and Si both in ferrite and austenite phases.

Keywords: hydrogen embrittlement, b-ray induced x-ray spectrometry, thermal desorption spectrometry, nonmetallic inclusions.

Reference:

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Measurement, monitoring and accountancy





THEORETICAL INVESTIGATION OF TRITIUM CONCENTRATION QUANTIFICATION METHOD FOR *DT* FUEL SYSTEM USING B-RAY INDUCED *X*-RAY

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Tritium measurement is one of the most important issues in DT fuel system of a fusion reactor. Beta-ray Induced X-ray Spectrum (BIXS) method is a promising way to obtain tritium concentration in-line, especially for tritium of high concentration. However, till now there is no theory to establish the relationship between tritium concentration and gas information including compositions and pressure. A novel method was proposed to quantify tritium concentration in BIXS measurements for tritium in gaseous form. Benchmark experiments and Non-linear Response Coefficients (NRCs) are introduced in the method. Effects of both gas compositions (hydrogen/deuterium/tritium) and gas pressure (1 Pa to 100 kPa) have been investigated quantitatively by Monte-Carlo simulations to determine the key parameters in the proposed method. Results indicate that for gas composed of hydrogen, deuterium, and tritium, NRCs are independent of gas composition within gas pressure from 1 Pa to 100 kPa. Benchmark experiment used to calibrate BIXS method is found to be universal for all the hydrogen isotopes. Therefore, with the calculated NRCs, tritium concentration can be obtained using only one benchmark experiment performed with any gas compositions at any gas pressure in BIXS measurements for gas composed of hydrogen isotopes. For gas composition including hydrogen, deuterium and tritium with different concentration, the error of NRCs in the proposed method is less than 1.0%.

Keywords: Tritium, Fusion, Tritium measurement, BIXS



μ*RA* - A NEW COMPACT EASY-TO-USE RAMAN SYSTEM FOR ALL HYDROGEN ISOTOPOLOGUES

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Based on the extensive experience with Laser-Raman systems at the Tritium Laboratory Karlsruhe (TLK) [1] we developed a new and compact Raman system tailored to the needs of everyday tritium process control. The focus was set to an easy to install and operate system with a small footprint and comparatively low cost of acquisition and negligible operating cost.

Based on this, we developed a fully tritium compatible system able to detect all six hydrogen isotopologues (H₂, D₂, T₂, HD, HT, DT) simultaneously with a lower level of detection of approx. 1 ... 5 mbar (<300 sec acquisition time) in < 1000 mbar (absolute) gas mixtures. An accuracy of < 10 % with a good reproducibility should be achieved while using mainly commercially available components to ensure an easy set-up process. This led to the use of a fully fibre-coupled system with an optimized configuration providing easy and quick setup (10 min from unboxing to measurement) while being small and flexible enough to be added to virtually any existing tritium primary system. Only minor modifications to an existing primary system (free $\frac{1}{2}$ " VCR port) and the glove box/secondary containment (KF25 port or equivalent) are necessary. Complex alignment procedures are rendered obsolete due to the fibre-coupled design.

After evaluation of different optical components, we came up with the system presented in this contribution. Followed by extensive tests with inactive mixtures of H_2 and D_2 , the first system was deployed in a crucial part of the tritium infrastructure at the Tritium Laboratory Karlsruhe, the "tritium transfer system" (TTS [2]). Performance studies with mixtures of all hydrogen isotopologues utilising the versatile TriHyDe [3] system and a cross-check to the high-performance LARA system [1] were conducted to explore the limits of the micro-Raman (μ RA) system.

The first installation was followed by two more systems providing for the first time in-situ compositional monitoring of the tritium separation process in another crucial TLK infrastructure system, the isotope separation system (ISS) [2].

Based on the experience gained with the first μ RA operation, we developed a dedicated data-taking and online/offline analysis software. With this, we now have a complete and easy-to-use, lightweight yet powerful inline spectroscopy system near to atmospheric pressure down to approx. 1 mbar. This makes the μ RA system perfect for nearly all practical operations in a typical tritium process environment.



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Further μ RA systems will be deployed in the TLK, not only in infrastructure systems but also in various experimental setups handling tritiated gases and liquids. Due to its flexibility and modularity, fine tuning the system to special needs can be facilitated with comparatively low effort.

Keywords: raman spectroscopy, fibre coupling, tritium measurement and monitoring, micro-Raman, TLK, μ RA

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THE TRITIUM MIGRATION ANALYSIS PROGRAM, VERSION 8 (TMAP8)

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The original Tritium Migration Analysis Program (TMAP) was developed at Idaho National Laboratory in the 1980s-2000s, culminating in the release of TMAP4 in 1992 [1] and TMAP7 in 2004 [2]. Its primary application was safety analysis of fusion energy systems, though it has also been frequently used in the modeling and interpretation of tritium and hydrogen transport experiments. Written in Fortran 77, the original code solves for the transport of hydrogen isotopes (and other molecules they react with) in a network of zero-dimensional enclosures via advection by fixed, user-specified flow rates, and to/from one-dimensional solid segments through which they can diffuse and become trapped at defect sites. Interfaces between enclosures and segments may have a fixed flux or concentration, follow a solution (e.g. Henry's or Sieverts') law, or have competing kinetic (e.g. dissociation and recombination) effects. TMAP7 addressed issues with multiple-isotope transport in TMAP4, increased allowable model size and complexity, and added the ability to model species that can adsorb on surfaces but not diffuse through them.

Despite the relatively comprehensive treatment of the relevant macroscopic tritium transport physics, in light of the greatly increased abilities of present-day computers, TMAP's relatively coarse nodalization, 0D/1D representations, and limited problem size are now unnecessarily restrictive. An increasing desire to model tritium transport in complex, 3D components and couple to other physical models (e.g. that solve for carrier fluid velocities) has prompted the development of TMAP8. TMAP8 is a complete re-implementation of TMAP's physics in the C++-based Multiphysics Object-Oriented Simulation Environment (MOOSE) [3]. TMAP8 preserves the ability to model large reactor systems as networks of 0D/1D components in the style of legacy TMAP, but with a completely revised, more user-friendly input file format typical of other MOOSE-based applications; it can also solve the same equations on arbitrary 2D or 3D meshes. Coupling to other MOOSE-based or non-native applications is facilitated by MOOSE's "MultiApp" and "Transfer" systems. Some examples of both 1D and 3D modeling are presented, and the status of TMAP8 verification and validation is discussed.

Keywords: Tritium transport, fusion reactors, Multiphysics modeling.

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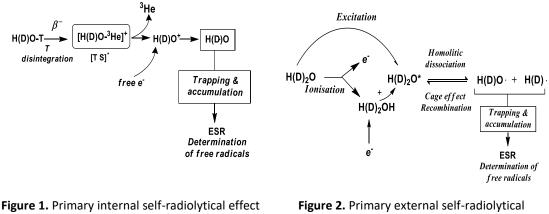


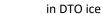
CHARACTERIZATION OF TRITIATED HEAVY WATER USING MAGNETIC RESONANCE SPECTROSCOPY

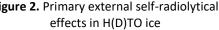
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In this paper, a novel method for determination of radioactive concentration and hydrogen isotopes distribution in tritiated heavy water is proposed. The method relies on the determination of free radicals radioinduced by self-radiolysis of frozen tritiated heavy water [1, 2] using electron spin resonance (ESR) spectroscopy (see Figure 1 and 2).







The preliminary ESR analysis of the HTO and DTO samples stored in liquid nitrogen confirmed the accumulation of HO• (asymmetric doublet with $g_1 = 2.048$, $g_2 = 2.026$ and $g_3 = 1.996$) [3] and respectively DO• (triplet with g = 2.013 and a = 0.69 mT) radicals. The secondary radicals, D•/H• generated in Primary external self-radiolytical effects of the HTO/DTO, were not identified by ESR spectroscopy.

The linear dependence between ESR signal of the $H(D)O\bullet$ radicals and storage time at -196° C suggests a constant accumulation of $H(D)O\bullet$ radicals. At high accumulation time and radioactive concentration a recombination tendency of the formed radicals has been identified.

Our preliminary results sustain the possibility of the use of ESR spectroscopy in determination of radioactive concentration for high activity samples (0.1 ... 4 TBq/I).

Also the T-Nuclear Magnetic Resonance spectroscopy (T-NMR) has been employed for radioactive concentration determination of DTO. The T-NMR amplitude signals have a



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linear dependence with radioactive concentration, but the accuracy of the T-NMR method is lower compared to ESR. The advantage of the T-NMR method consists of determination of nuclides distribution in DTO, respectively the presence of the protium traces in the system.

Keywords: Tritiated water, measurement and monitoring, waste management, ESR spectrometry, NMR spectrometry

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OPTIMISATION OF PYROLYSIS OPERATING CONDITIONS FOR H-3 EXTRACTION FROM RADIOACTIVE WASTES MATRICES BY EXPERIMENTAL DESIGNS

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Tritium is one of the most important radionuclide to measure for waste management. Due to its labile and chemical behavior (organic or inorganic forms), tritium extraction efficiency is the key for its evaluation content in waste before its evacuation.

Liquid Scintillation Counting (LSC) is the method of reference for tritium measurement. To extract tritium from solid sample generally generated during decommissioning projects, the pyrolysis method has been used and described in previous studies [1]. Recent evolutions in pyrolysis furnaces design such as separate bubbler temperature control, regulation of gases flow rates in the pyrolysis tubes gases have led us to perform an optimization study of the operating conditions for the pyrolysis step before tritium measurement for different matrices. For each matrix studied: aqueous effluent, concrete, oil and graphite, experimental designs were used to study pyrolysis parameters influence on tritium extraction efficiency.

For each matrix, a first screening design was used to determine influent parameters on tritium extraction efficiency and in secondly, optimization design (DOEHLERT, pentagon, Hocke, ...) was performed to determine optimum conditions for tritium extraction. We will present results concerning optimization of the pyrolysis conditions for four different matrices: liquid wastes (oil and aqueous effluent) and solid waste: concrete and graphite. For optimization, the available CRMs proficiency tests for aqueous samples and intercomparison between laboratories for oil were used. For concrete, a specific study has been performed on the influence of concrete storage conditions and quantity of sample analyzed on the recovery of H-3 after pyrolysis.

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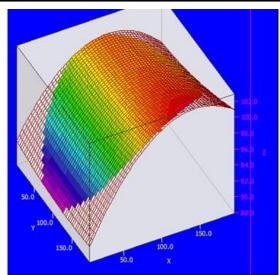


Figure 1. Extraction yield of tritium in function of pyrolysis gases flow rates

Keywords: Tritium extraction, measurement, pyrolysis, aqueous effluent, graphite, concrete, oil.

Reference:

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MINIATURIZING TRITIUM MONITORS FOR PROCESS GASES

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Tritium monitors used to measure activity within a process loop are generally large instruments that require a rack mounted controller outside of a glovebox, multi-wire cabling per unit that use expensive custom glovebox feed throughs and preamplifiers mounted to ionization chambers which take up valuable glovebox space. A development campaign was initiated to miniaturize all the components of a process gas tritium monitor into one small package. The goal was to reduce the entire footprint of the system both inside and outside of the glovebox and to reduce the complexity of the glovebox penetrations for multiple units.

A process monitor comprises an ionization chamber and the electronics. The ionization chamber must operate at a high DC voltage to provide a potential gradient between the anode and walls of the chamber for the separation and measurement of ions generated when energetic electrons due to tritium beta decay ionize the process gas. The electronics must provide this bias voltage, amplify the femtoampere $(1 \times 10^{-15} \text{ A})$ currents with transimpedance amplifiers to generate millivolt signals, process the signal in a microprocessor, allow relay activation for a greater dynamic measurement range, and communicate with the user through a visual display, analogue and digital outputs.

The first challenge in miniaturizing all the electronics into a single, small footprint mounted directly onto the ionization chamber requires the use of surface mount technology. Using the power over ethernet (POE) protocol, DC to DC inverters and microprocessors, the electronics have been compressed into a 4" wide x 2" deep x 5" height enclosure. All power and communications are connected by an industry standard RJ-45 ethernet connection from a small POE ethernet switch that can be installed inside a glovebox. This allows all process monitors within a glovebox to be networked such that only one commercially available glovebox ethernet jack penetration is required.

The second challenge centred on miniaturizing the ionization chamber. The goal was to design a high activity, low weight, small footprint device that could be integrated into a process loop with the same ease as a pressure transducer or mass flow meter. The ionization chamber was designed to meet the best practices for applications in tritium process-loops: high leak tightness (< 1×10^{-9} atm-He cc/s), non-organic seals and compatible wetted materials, high pressure boundary (> 100 psig), low surface retention, and metal face seal connections. This was achieved with a 10cc ionization volume integrated into a single machined stainless-steel block with $\frac{1}{2}$ " male VCR connections at either end. A ceramic insulated feedthrough was welded into the block to apply the accelerating voltage and collect the beta induce current.



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This paper will present the design and performance of a 10 cc tritium process monitor with the characteristics described above. The device was vetted for leak tightness, its response to tritium activity into the PBq/m^3 (100 kCi/m³) range and gammas from a Cs 137 source, pressure testing to destruction and, stability to environmental conditions.

Additional ionization chamber designs which can be mated with the electronics are also discussed. A 1 Liter detection volume for more sensitive applications in the 10s of kBq/m³ (1 μ Ci/m³) range, a 20cc virtual wire cage and a glovebox atmosphere monitor on a KF40 fitting are highlighted.

Keywords: Tritium monitoring, process monitor, measurement and monitoring, high activity monitoring



ENVIRONMENTAL BEHAVIOR OF TRITIUM AND ITS IMPACT ON THE FUSION DEMO AND NEAR TERM POWER PLANTS

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This paper will summarize the study of the entire vision of tritium control issue for fusion energy. Design of the processing of tritium in fusion reactors, its emission to the environment, its behavior, and impacts from social aspects of the near term fusion power plants are discussed, and the result to be reflected in the design is suggested. Not only previous DEMO designs, but also recent smaller and earlier commercial fusion attempts and social perception aspects are also considered and presented.

Fusion power plants will be major tritium sources to be released to the environment. The fusion power plants, regardless of the types of their energy conversion processes, will have to discharge large amount of waste heat from the generation thermal cycles to the environment. The fusion power obtained as heat is first sent from the breeding blanket with high concentration of tritium by heat transfer media, that will then be transferred to a fluid to drive turbines, and is finally discarded in either water or air to be released to the environment.

In each steps, heat exchanger, steam generator, or similar devices are used for heat transfer, but they have some tritium permeability or leak rate for tritium, and thus can never be free from contamination. Large amount of air or water with tritium contamination will of course controlled to contain considerably lower concentration of tritium required by the regulation. Since the amount of this final coolant is extremely large, for instance, several degree warmer waste heat emission at the level of 1GW requires 20 tons of water per second, to meet the concentration limit of 60000 Bq/kg can be achieved by reasonable technical efforts.

However, it should be noted that this is a emission by the normal operation of fusion plant, and such a large amount of tritium above the natural background of ~1Bq/kg is detectable with the modern instruments by local public without any special technical expertise. Concentration limit of tritium is far higher than the natural background, and thus the slow and steady increase of tritium level due to the operation of fusion plant will be visible, and should be agreed before the construction. The authors have reported that typically its consequence is 10 micro Sieverts per year level of public dose, but the detected tritium will be 100 Bq/kg, that is easily detected. Affected area will be the order of 100km, and the tritium concentration will continue to increase 60 years until equilibrated with the decay and natural dilution. Such environmental effect is predictable, and must be explained to the public prior to the start of fusion plant projects.



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Particularly by the process of the public explanation and agreement on the release of tritiated water from the Fukushima-Daiichi power plant in Japan suggests this kind of public understanding is inevitable. Tritium behavior in the environment is important to understand because the tritium will finally be contained in agricultural or fishery products that will be evaluated in the consumer market. Reputational damage is sometimes far more important than actual dose or health effects. When the tritium concentration in the food products is detectable, it may not be acceptable even if all the regulation would be met and no radiological effects are anticipated.

Demo and next generation DT fusion reactors will have to be designed with the consideration of such kind of tritium related issues in the fairly early stage of conceptual study. Tritium emission can be controlled technically to follow the regulation limit, but at the same time, its consequence in environmental and social aspects must be well considered and explained to the public. In the past nuclear programs, the responsible body for the nuclear programs are public entities or electricity/utility sectors. However in the recent fusion development, private startup companies are playing major roles to develop and build DT burning machines to demonstrate fusion energy considerably earlier than previous public programs. They plan relatively smaller amount of tritium to be handled, and the regulation and tritium related discussions for such a small fusion plants needs more social involvements.

Keywords: Environmental emission, contamination, fusion power plants, monitoring, demo design.



SPEED OF SOUND MEASUREMENTS FOR VERIFICATION OF REFERENCE GAS SAMPLES CONTAINING TRITIATED MOLECULES

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The development and characterization of tritium monitoring devices is of great importance in the diverse field of tritium processing, e.g. process control and accountancy [1]. For this purpose, highly accurate reference gases containing well-defined admixtures of tritium are essential. At the Tritium Laboratory Karlsruhe, the TRIHYDE facility has been commissioned, capable of fabricating gas samples that may contain any of the six hydrogen isotopologues with sub-percent accuracy of the relative isotopologues concentration [2].

TRIHYDE produces these samples by precise manometric gas mixing of the homonuclear isotopologues (T_2 , D_2 and H_2) and consequently the sample accuracy is limited by the available purity of the initial feed gases. The stable isotopologues are commercially available as reference gases; however, no certified isotopically pure tritium samples are available on a technical scale.

Therefore, in order to verify the accuracy of the TRIHYDE produced samples, the relative isotopic ratio of individual samples is evaluated using in-line composition analyses based on speed of sound measurements. This method is especially sensitive to common impurities like helium, nitrogen and argon. Using literature values for the individual speed of sound of the respective isotopologues, an agreement between measured and manometrically derived composition on a <0.1% level was achieved. This demonstrates the high accuracy of the prepared samples and their possible utilization in the characterization of in-line analytical devices. Furthermore, this setup allowed us to perform first measurements of the speed of sound of tritium, an up to now missing experimental value for the tritiated isotopologues [3]. In general, the presented methodology can be expanded to include other gas species of interest, e.g. helium and argon, as well as the ortho-para ratio[4], therefore covering a wide range of tritium-containing mixtures present in typical tritium processing applications [5,6,7].

Keywords: Tritium analytics, reference gas samples, measurement and monitoring, process control, accountancy, speed of sound

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EVALUATION OF ENVIRONMENTAL TRITIUM LEVELS AROUND CERNAVODA NUCLEAR POWER PLANT AFTER 25 YEARS OF OPERATION

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Cernavoda Nuclear Power Plant (NPP), the only Canadian Deuterium Uranium (CANDU) in Europe, is located in the South-East of Romania, at the confluence of the Danube River and the Danube – Black Sea Channel, about 60 km from the Black Sea (Constanta County) and about 160 km from Bucharest. Cernavoda NPP has 2 operating Units of 700 MWe and produces about 20% of the electric power of Romania (Unit 1 - since December 1996; Unit 2 – since November 2007) [1].

The Environmental Control Laboratory (ECL) of the Cernavoda Nuclear Power Plant, located in Cernavoda town, 2 km from the Cernavoda NPP, is part of the Radiation Protection Department and is equipped with performant analyzing systems to determine the natural and artificial radionuclide levels in the environmental samples within 30 km area around the Cernavoda NPP.

The Environmental Radioactivity Monitoring Program for Cernavoda NPP started in 1996 and all necessary activities are performed in the Environmental Control Laboratory, which has a Quality Assurance Program according to the appropriate international standard and is participating in many intercomparison exercises and proficiency tests to validate analyzing methods and to demonstrate the effectiveness of the Environmental Program, as mandatory condition in the certification of the ECL and the reauthorization process of the Cernavoda NPP by the Romanian regulatory body (CNCAN) [2].

The annually report contains all the results for the environmental radioactivity monitoring and effluent monitoring; the results of the monitoring program are annually compared with the results of The Preoperational Environmental Monitoring Program performed between 1984 and 1994 [3].

This paperwork presents the evaluation of tritium levels in environmental samples around Cernavoda NPP and the impact on the public health after 25 years of operation (1996-2021) [4].

Keywords: tritium, measurement and monitoring, environmental levels, CANDU.



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CAN TRITIUM MONITORING AND CONTROL REQUIREMENTS BE MET BY EXISTING TECHNOLOGIES?

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The tritium inventory of future fusion power plants needs to be monitored in the fuel cycle for several reasons; to comply with limits imposed by environment and safety regulators, adhere to practises required by nuclear regulators and for process control purposes. Fulfilling all these requirements leads to a comprehensive list of locations in the fuel cycle where tritium monitoring needs to take place, each characterized by different measurement conditions and required accuracies.

Meanwhile, existing tritium detection technologies all come with specific applicabilities such as accuracy, material phase, and ability to detect tritium in a continuous manner. These do not necessarily correspond to the required measurement conditions. As an example, one tritium detection technology will be matched up with the previously defined measurement conditions, which allows for the identification of gaps in the existing detection capabilities of this technology.

This work leads to several recommendations: for developments to expand the applicability of tritium detection technologies, for experimental proposals to test detection techniques at more extreme conditions, and to expand the regulatory framework regarding tritium handling and breeding. These developments are critical for a functioning tritium management and control system and this talk outlines the first step in that process.

Keywords: Tritium detection, accountancy, measurement and monitoring, fuel cycle.

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HIGH-RESOLUTION SPECTROSCOPY ON TRITIATED MOLECULAR HYDROGEN

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Small molecules represent a benchmark for testing quantum chemical theories. Ultra-high-resolution spectroscopy allows to measure the energy levels of molecular hydrogen with a relative accuracy of the order of 10^{-10} . Extending these studies to different isotopologues of molecular hydrogen has emerged as promising method for testing mass-depending contributions to quantum electrodynamics (QED). The technological challenges of handling tritium in a delicate high-resolution spectroscopy environment has restricted these studies to be extended to the radioactive isotopologues. Recently, we have performed Coherent Anti-Stokes Raman Spectroscopy (CARS) in a closed tritium cell (A < 1GBq) which allowed to probe fundamental vibrational transitions in HT, DT and T₂ with an accuracy up to $5x10^{-4}$ cm⁻¹ [1,2] which reflects a hundred-fold improvement over prior measurements. This technique is limited by the Doppler-broadening (~370 MHz for T₂ at 300 K) from the thermal motion of the molecules and by systematic effects, e.g. related to the intensity of the interacting laser pulses.

In a next step, we aim to make use of a very sensitive, Doppler-free technique such as Noise-Immune Cavity-Enhanced Optical Heterodyne Molecular Spectroscopy (NICE-OHMS). For the heteronuclear HD molecule, a linewidth of only 400 kHz and an accuracy of about 10⁻⁶ cm⁻¹ was achieved [3]. The experimental transition energies are currently in tension with the theoretical calculations by 2-sigma significant shift [4]. High-resolution spectroscopy of the isotopologue HT will therefore shed light onto this effect by making use of the different reduced mass. At this level of accuracy, the systematic uncertainties from experimental parameters have to be carefully controlled. NICE-OHMS measurements will be performed in a closed cavity system without connection to an actively pumped vacuum system. For this reason, a new method for tritium storage and pressure regulation within 0.1 to 5 Pa based on a temperature-controlled mini-getter system is employed.

In the presentation we will show our recent progress of high-resolution spectroscopy of tritiated molecular hydrogen isotopologues by CARS spectroscopy. In the main part, we focus on the new mini-getter system (SAES ST171) for compact tritium systems such as the NICE-OHMS cavity which allows a pressure regulation of molecular hydrogen in a range of 0.1 - 10 Pa while other gases species are absorbed. We show the measurements results of



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the equilibrium (hydrogen) pressure for all six hydrogen isotopologues as functions of temperature and total amount of gas.

Keywords: high resolution spectroscopy; CARS; NICE-OHMS; getter.

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APPLICATION OF THE RAMAN SPECTROSCOPY FOR DETERMINING THE COMPRESSIBILITY FACTOR OF REAL GASES AND GAS MIXTURES

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Currently, the task of experimental determining the compressibility factor of hydrogen isotopes remains relevant in connection with the prospects of hydrogen energy, information on the thermodynamic properties of hydrogen in wide range of pressures and temperatures is needed for the production, use and storage of hydrogen.

The paper considers the possibility of using Raman spectroscopy as a method for determining the compressibility factor of real gases and gas mixtures. The consideration uses the fact that the intensity of the radiation of the Raman scattering is directly proportional to the concentration of the molecules of the irradiated substance.

Thus, by recording the Raman scattering spectra at an increase in pressure, it is possible to determine the dependence of the concentration (or density) of the gas on its pressure. From the obtained dependence, you can derive the compressibility factor of the gas at a given temperature. Optical probes developed in RFNC-VNIIEF allow you to record the spectra of Raman scattering of gases at pressures up to 400 MPa.

The method under consideration may be promising for determining the equations of the state of gas mixtures, since peaks of its components are present in the Raman scattering spectrum of the mixture, and the concentration of these components can be determined independently of each other.

The method under consideration may be more accurate than conventional methods, since it does not require measuring the volume or mass of the gas under investigation. In addition, processes such as sorption of molecules of the analyzed gas on the walls of the vessel, and their desorption do not affect the results obtained by this method, which can also positively affect the accuracy of measurements.

The results of the experimental determination of the compressibility factor of pure H_2 , a as well as the mixture of H_2 with He at pressures up to 400 MPa and a temperature 293 K are provided in the paper. The comparison of the experimental and calculation data was also carried out.

Keywords: Hydrogen isotopes, Raman spectroscopy, compressibility factor, gas mixture.



THE DEVELOPMENT OF AN INSTRUMENT TEST FACILITY FOR THE EVALUATION OF GASEOUS MEASUREMENT TECHNIQUES FOR CONTINUOUS TRITIUM ACCOUNTANCY

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The continuous nature of future fusion power plants such as DEMO is in stark contrast to the batch-wise operation of current fusion facilities. The requirement to operate in a continuous manner poses many challenges in accurate tritium measurement, as well as in tritium accountancy. This study describes work underway within the UKAEA to assess the suitability of analytical techniques and supporting technologies to conduct continuous tritium accountancy.

An instrument test facility is currently under development which will provide quantifiable data to inform the development of tritium accountancy systems and provide characteristic data on the application of analytical technologies to fusion-relevant process conditions. A bespoke Raman spectroscopy system utilizing an innovative hollow-fibre sampling technique has been procured for testing, as has a high resolution, continuously sampling mass spectrometer. The research aims, objectives and intended outputs of this facility are presented, as are its planned capabilities and the current progress of the test facility. Also presented is the outline experimental programme which will be conducted using this facility, as well as inactive instrument characterisation activities conducted so far.

Keywords: Fusion reactors, measurement and monitoring, experimental facility, accountancy, mass spectrometry, Raman spectroscopy



A NOVEL APPROACH FOR TRITIUM PROCESS MONITORING WITH HYBRID MODEL-BASED AND DATA-DRIVEN APPROACHES

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Nuclear fusion depends on the concept of tritium breeding and self-sufficiency to become a feasible technology [1]. However, the use of tritium represents a hazard due to its radioactivity. Also, it is extremely hard to monitor: it appears in small quantities at ranges where sensors perform poorly [2] and easily contaminates environments due to its permeation properties. Because of these difficulties, ITER, the largest fusion experiment so far, relies on a conservative static procedure to monitor the tritium inventory [3]. This strategy fixes a limit in its flexibility of operation and, in future fusion power plants, may jeopardize fusion performance due to the periodic shutdowns that become mandatory to perform the tritium accountability.

We can avoid plant halts if a dynamic monitoring strategy proves itself valid. Tritium Plant models have been developed for this kind of monitoring and analysis tasks [4] but the issue of the underperformance of sensors is still to be addressed and the path to dynamic monitoring remains unclear. The present work shows a novel approach for tritium monitoring based on the principles of fault detection and diagnosis, first-principle process simulation and historical data usage through machine learning. A demonstration using test bench models (model-based perspective) will show the performance of the proposed method. These models will generate data (data-driven perspective), which in turn will feed a machine learning model trained to perform the monitoring task.

The output of this work is the verification of the use of hybrid methods as a continuation of a previous work [5] in the road of enhancing tritium monitoring and the usefulness of new tools to track the tritium life-cycle.

Keywords: measurement and monitoring, tritium inventory, machine learning

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AUTOMATED, HIGH-THROUGHPUT MASS SPECTROMETRY OF TRITIUM, DEUTERIUM, HYDROGEN, AND OTHER GAS MIXTURES IN SUPPORT OF THE LAWRENCE LIVERMORE NATIONAL IGNITION FACILITY

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Fusion experiments performed at the National Ignition Facility (NIF) require ultrahigh purity mixtures of tritium and other species [1]. These experiments include inertial confinement fusion and other high energy density and national security investigations. Fuel mixtures for the fusion reactions generally consist of tritium and deuterium sometimes in combination with hydrogen or noble gases. To validate computational models of neutron yield, the fuel composition must be known precisely and must conform to narrow elemental and isotopic ratios [2]. Additionally, the tritium mixture must be delivered to the containment capsule through a microscopic tube, often as narrow as 2 μ m in diameter, to minimize perturbations to implosion symmetry [3]. When experiments are performed at cryogenic temperatures, contaminants such as water and hydrocarbons can solidify and plug the fill tube during formation of the tritium ice layer. For these reasons, the fuel composition must adhere to stringent purity requirements. NIF executes 400 laser-driven shots per year requiring multiple fuel and process gas analyses per week or day.

We developed analytical methods to characterize the NIF tritium fuel mixtures with high resolution, speed, precision, and accuracy by gas phase mass spectrometry. These methods allow us to analyze several dissimilar samples per day to a detection limit of five parts per million with resolution of identical nominal masses, such as HT and D₂. We built a custom inlet system to connect multiple samples of differing pressure and composition to a Finnigan MAT 271 magnetic sector mass spectrometer. We developed software that operates inlet system valves, configures hardware parameters, and processes raw data. These workflow improvements allowed us to automate calibration and sample analysis to drastically increase sample throughput.

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Figure 1. Finnigan MAT 271 mass spectrometer customized for high-throughput analysis of tritium and other gas species



Figure 2. False-color illustration of laser beams irradiating a NIF target holding a spherical deuterium-tritium fuel capsule

Keywords: Tritium analysis, mass spectrometry, inertial confinement fusion, measurement and monitoring

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EXTENDED HYPOTHETICAL CALCULATION OF TRITIUM CALORIMETRY USING MULTI-HYDRIDE COMPONENT SYSTEM

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In-site static or in-bed calorimetry seems not to be applied to the depleted uranium hydride system for the hydrogen isotope, especially tritium, accountancy. The main reason is that the depleted uranium hydride bed has absolutely the restriction in thermal decay energy measurement due to the heat generation comparing with the high thermal mass of the hydride bed itself. It is understandable that the surroundings ought to have great affection to this small value of rate-controlled tritium calorimetry [1].

On the other hand pressure sensitive measurement during hydride bed heating, accompanying by several step-by-step system temperature changes, seems to be approachable even to the small amount of tritium residence. The experimental validation is not easy, however, to be realized by handling of the depleted uranium hydride alone, because of rather flat PCI(Pressure-Composition-Isotherm) plateau. Even in dual complex system of hydride there may be a weak point to estimate the expected hypothetical phase equilibria without any experimental evidence. Nonetheless, the estimation of tritium inventory is very useful during tritium (hydrogen isotope mixture) operation at any arbitrary measurement of hydrogen isotope composition, within a meaningful time, in the depleted uranium hydride bed system.

The authors have introduced this theoretical approach of tritium calorimetry accountancy measurement before, assuming of two complex mixture of hydride materials together [2]. But unlikely the theoretical premise there seemed to be important unknowns to calculate the complex hydride material system equilibrium. In this study, therefore, the authors are to modify the former approach and to extend case studies of tritium calorimetry using pressure sensitive methodology for three component complex hydride material system.

Keywords: Tritium, calorimetry method, ternary complex, multiple hydride material, etc.

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PROCESS MONITOR EVALUATION FACILITY

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Tritium process monitoring equipment is not characterised across most of the conditions relevant to their users. Manufacturers lack the capability to work with the required tritium concentrations and gas mixtures due to the high cost and unique skills required to produce and operate safety infrastructure. Instead, extrapolation is used to predict performance. The University of Rochester has a new facility. The Tritium Process Monitor Evaluation Station will enable the assessment of existing and newly developed tritium monitors under a broad range of relevant conditions, including the loss of dynamic range due to surface contamination. In collaboration with the UKAEA, the test stand aims to improve accuracy and confidence in readings provided by tritium process monitors and establish new production standards, no longer relying on extrapolation and estimates. Flow conditions, humidity, temperature, and carrier gases can be varied to cover a range of operating parameters and the commissioning of these systems has progressed. This system is primarily aimed at qualifying tritium monitors and calibrating these monitors in process relevant conditions. However, the facility can also determine the relationship between permeation through organic materials and temperature, by using a qualified process monitor. The facility aims to become a user facility following commissioning and characterisation by the collaboration.

Keywords: Tritium; Measurement; Process Monitor.



AUTOMATED HEAVY WATER SAMPLING AND ANALYSIS SYSTEM AT PIK RESEARCH REACTOR

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This work is devoted to the creation of heavy water sampling and analysis system (ASSA HW) at the PIK reactor [1]. A necessary condition for obtaining a high neutron flux (up to $5 \cdot 10^{15}$ neutrons/cm²·s) at the PIK reactor is the use heavy water reflector (HWR). Handling of heavy water with tritium, including chemical control, is radiation hazardous. Unlike the existing automatic chemical control light water systems at nuclear plants, ASSA HW should not only control the chemical and isotopic coolant composition, but also return of heavy water back to the HWR circuit. This is due to heavy water is an expensive coolant, which belongs to special non-nuclear materials. Now there isn't an analogue of such a system in Russia.

In the ASSA HW project, four sampling points were defined: HWR circuit, HWR tanks and after ion-exchange filters. The system controls eight parameters of heavy water: deuterium concentration (aD); volumetric activity of tritium (CT); electrical conductivity; factor pD; the concentration of dissolved oxygen; the concentration of dissolved hydrogen; turbidity; the concentration of chloride ions.

To control the chemical parameters of heavy water, it was decided to use several types online analyzers manufactured by Russian companies.

To measure the deuterium concentration, it is used an industrial NIR Fourier spectrometer type Matrix-F-(Bruker). Adaptation this device to ASSA HW has been carried out since 2016 [2]. Four calibration models were constructed for different ranges of D_2O concentrations. It was shown that, the Matrix-F sensitivity is sufficient for operational monitoring of HWR heavy water. At present, an on-line heavy water analyzer model has been created at the PNPI laboratory, and a calibration technique has been developed before its installation in the PIK reactor circuit.

Flow-through radiometer Wilma (LabLogic) was included in to ASSA HW for tritium activity monitoring. The use of a liquid scintillator had obvious drawback – irreversible waste of heavy water is generated. Therefore, the Wilma is equipped with solid scintillator flow cells on base CaF_2 powder for use in the PIK reactor. Since 2017, work has been carried out on this analyzer to measure the tritium activity [2]. However, the results obtained do not yet allow the use of Wilma in the ASSA HW.

All on-line flow analyzers were calibrated at laboratory prior to install in the HWR loop. Currently the ASSA HW system is mounted in the HWR circuit.

The presented system organizes automated remote monitoring the isotopic and chemical composition of heavy water, return the heavy water after measurements back to the circuit and reduce the dose load on the reactor personnel.



Keywords: ASSA HW, the PIK reactor, on-line monitoring, deuterium, NIR spectrometer, tritium, flow-through radiometer.

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Tritium supply, transport and storage





BALL MILLING – A LOW COST, HIGH THROUGHPUT ANALOG FOR TRITIUM-AGING OF HYDRIDES?

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Materials exposed to tritium accumulate helium as tritium diffuses into the material matrix and decays into He-3. These He-3 atoms can coalesce and form He nano bubbles that can grow to larger sizes and can cause deleterious effects on materials properties. He incorporation also notably affect the properties of metal hydrides used for tritium storage. Investigations of He bubble formation, properties, and damage are significantly limited by the methods currently available to generate them – tritium aging or He ion implantation. These techniques are costly, time consuming, pose radiation hazards, create collateral ion damage, and are limited to the near surface region in the case of implantation. Recently, it has been reported that mechanical alloying via ball milling can incorporate inert gases, such as Ar and He, into material matrices and subsequently produce numerous gas bubbles trapped in the metals. This presentation describes the potential application of ball milling to simulate the effects of tritium aging in hydride materials. The advantages of using ball milling to simulate aging are reduced costs and increased throughput compared to traditional tritium aging. This will allow faster development of hydride regeneration processes, as well as enable investigations into new materials that may be more resistant to tritium aging effects.

Keywords: Hydrides, tritium aging, helium bubble



CHARACTERISTICS OF URANIUM HYDRIDE BED FOR FUSION FUEL CYCLE: ABSORPTION/DESORPTION PERFORMANCE AND PRACTICAL OPERATION TEST

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The authors developed an improved uranium hydride bed based on the design, fabrication, experiment and troubleshooting of an original design DU bed [1]. We focused on robust and simple design, reliability and longevity considering a service in the tritium plant of nuclear fusion machine, fabricated an experimental bed named DU-FB2 (2nd Depleted Uranium Full scale Bed) with a 1.86 kg of depleted uranium which is storage capacity equivalent to 70 g of tritium.

We performed absorption and desorption performance verification with both of hydrogen and deuterium, and investigated the effect of practical operation condition such as the initial temperature for recovery and the heating temperature for delivery. The rate and time for absorption and for desorption were compared respectively. The equilibrium pressure of uranium hydride and uranium deuteride with this large-scale bed were also studied. Additionally, as a

preliminary test for practical operation in tritium plant, we conducted a cyclic operation test composed of five sequential absorption and desorption of deuterium. Summary of these experimental results and analysis will be presented.

These experimental results are expected to be the basis for the operating scenario of a uranium bed in the tritium plant of nuclear fusion facility.

Keywords: Tritium storage, metal hydride, uranium, deuterium.



Figure 1, 2nd 70g tritium scale depleted uranium bed for experimental validation (DU-FB2) – KFE

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MEASUREMENT OF ABSORPTION AND DESORPTION ISOTHERMS OF PALLADIUM HYDRIDE USING VARIOUS H₂/D₂ MIXTURES BETWEEN 20°C AND 120°C

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Among the pure metals, palladium ranks second best among the hydride-forming elements. As such, it is widely used as a storage medium for hydrogen isotopes including tritium. However, the absorption and desorption isotherms of mixed isotopes has not been reported in open literature. In the present study, these isotherms have been measured for various H_2/D_2 mixtures between 20°C and 120°C. Equilibrium pressures in the $\alpha + \beta$ plateau region are in between those of the pure isotopes at the same temperatures. Calculated van't Hoff constants are shown in Table 1. Values for the pure isotopes were measured in a previous publication [1] and agree well with van't Hoff constants reported by other authors [2, 3]. An empirical formula has been derived to predict the equilibrium pressure given the composition of the absorbing gas and the palladium temperature.

% H_2 in absorbing gas	A (kJ/mol)	В (J/K)
0 (pure D ₂)	-16.2	-44.6
25	-16.6	-44.4
50	-15.9	-41.1
75	-16.5	-41.0
100 (pure H ₂)	-18.1	-44.2

Table 1. Measured van't Hoff constants for pure and mixed isotopes

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Keywords: Palladium, hydride, isotherm, storage

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THE TRITIUM WINDOW: AN OPPORTUNITY FOR ACCELERATED FUSION DEVELOPMENT

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The first demonstration D-T fusion reactors are expected to require in the order of up to several kilograms of tritium for start-up. Before that, experimental facilities may require several tens or hundreds of grams. As no natural source of tritium exists, it must be obtained from CANDU-type heavy water fission reactors, which produce tritium at a rate of around 2 kg per year contributing to a saleable tritium inventory that is currently estimated to be approximately 30kg. Parts of the CANDU fleet are coming to end-of-life, and the existing stockpile of tritium is decaying. The availability of tritium for fusion is considered as a risk to programs globally. ITER, anticipated to require a more than 10 kg tritium for its experimental program, will not operate with D-T until beyond 2035. Recent research has suggested that current stocks of tritium are more than sufficient until this time [1, 2], and thus there is an opportunity to obtain tritium without competition: a tritium window has opened. For the next 15 years or so there will be an abundant supply of tritium directly from CANDU heavy water, but also from the stockpile of tritium that has already been collected which, if unsold, will otherwise decay. This window presents a unique opportunity for organizations pursuing accelerated development, specifically for private fusion developers who have ambitions to operate demonstrators within the next decade, operating on a different development model [3]. This study presents updated modelling of global tritium production and inventory projections of CANDU supply against near-term demand from prospective accelerated programs. Tritium has not been procured in quantities in the order of kg for fusion before now. Steps for developers to procure, transport, receive, store and use tritium in their fusion facilities are outlined, and the challenges associated are discussed and a route forward charted. Finally, the need to prove tritium breeding in a parallel, to ensure that external tritium is not again needed for the first commercial plants and beyond, is analyzed with respect to the potentially accelerated tritium supply and use timeline. Altogether, this study presents a strategy for ensuring that this tritium window is not missed, facilitating developers to push forward with their accelerated programs.



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Keywords: Tritium supply, tritium breeding, tritium procurement, strategy, accelerated development, commercialization.

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HIGH PURITY TITANIUM SPONGE PRODUCT DEVELOPMENT AND PRODUCTION SYSTEM

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Westinghouse has developed a high-purity titanium sponge product to act as a getter bed for long-term tritium storage in lethal service immobilized tritium containers. Westinghouse manufactures several types of tritium storage and transport containers for the Nuclear Industry however they are the sole producer of this high-purity specification which is required to meet exacting tritium removal facility and customer requirements. The high-purity of the product is critical for safe and efficient tritium immobilization and the product's impurity levels are closely monitored through independent chemical analysis to ensure all chemical interface requirements are achieved. In order to meet the exacting customer requirements for this product, Westinghouse designed and manufactured a firstof-its-kind laboratory production system to effectively produce the titanium sponge product while ensuring the highest safety standards were met, especially when dealing with the associated production hazards. For example, production of the titanium sponge involves a multi-step process where raw titanium is alternatively reacted with extremely high temperature hydrogen at 1000 deg C and then exposed to high vacuum at 0.5 millitorr. Additionally, material reduction processes produce highly flammable explosive dust which must also be managed accordingly. Overcoming these safety hazards required extensive research of hydrogen and explosive dust hazards as well as implementation of an extensive safety control system with associated safety shutdown systems. The chemical reaction process involves many key parameters that had to be researched and experimented with during both the research and development phase and also during system commissioning and start-up.

Once manufacturing is complete the high-purity titanium sponge product is loaded into a specialized pressure vessel which has been optimized for tritium immobilization as tritium tritide on the titanium sponge getter bed [1]. Additionally, these vessels must accommodate the eventual radioactive decay of the tritium into the helium-3 isotope. The product is used by numerous nuclear generating stations across Canada for decontamination to remove tritium from the heavy-water moderator and heat transport fluids [2]. The high-purity titanium sponge product can be used in a wide range of storage or transportation containers to support both the fission and fusion industry.



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Keywords: Tritium immobilization, water detritiation, getter bed, storage container, transportation container, decontamination, helium-3, tritium tritide, immobilized tritium container (ITC)

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Figure 1. High Purity Titanium Sponge and Immobilized Tritium Container – Lethal Service



Figure 2. Titanium Sponge Production System – Laboratory Setup



THE EFFECT OF CO-AND COUNTER-PERMEATION ON TRITIUM TRANSPORT IN *HCPB* AND *WCLL* BREEDING BLANKET ENVIRONMENTS

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Ensuring a safe operation of future fusion power plants requires tritium transport into the coolant of the breeding blanket to be kept as low as possible. Previous experimental studies have found that a simultaneous co- or counter-permeation of protium and deuterium through a metal membrane can for certain partial pressure compositions influence the net flux of permeating deuterium. This finding gave rise to the assumption that a controlled addition of H₂ to the breeder fluid and/or coolant of a helium-cooled pebble bed (HCPB) and a water-cooled lithium lead (WCLL) breeding blanket could significantly affect the tritium release rate to the coolant. With the purpose of deeper understanding this phenomenon and its implication for future blanket designs this work focuses on the numerical study of co- and counter-permeation processes occurring in two representative HCPB and WCLL permeation environments (fig. 1). Therefore, numerical simulation tools of the considered permeation systems have been developed using EcosimPro which are capable of calculating the time evolving particle concentrations and fluxes in the considered sub-systems. In addition to multi-isotopic recombination and dissociation processes at the membrane surfaces the models take into account relevant chemical reactions between water molecules and hydrogen isotope gas molecules occurring in the breeder/coolant fluids (see figure 1). The models of both systems are used to simulate steady state tritium permeation fluxes for different H₂ partial pressures in both the coolant and the breeder fluids. Studying the considered WCLL system reveals that adding H₂ gas to the lithium lead or the water causes an increase of tritium permeation into the coolant which excludes this method as a tritium mitigation technology for WCLL breeding blankets. However, in case of the considered HCPB permeation environment the tritium permeation flux to the helium coolant significantly decreases with increasing H₂ partial pressure in the purge gas. When adding H_2 gas to the coolant the permeation flux rises with growing H_2 partial pressure until reaching a maximum. For higher H₂ pressures the net flux of permeating tritium decreases until dropping significantly below its initial value. These findings suggest that for the HCPB system a controlled co- and counter-permeation of protium and tritium could serve as a tritium mitigation method as long as the required partial pressure combinations are technically feasible.

Keywords: HCPB, WCLL, tritium modeling, multi-isotopic transport, co-permeation, counter-permeation

National Research and Development Institute for Cryogenics and Isotopic Technologies



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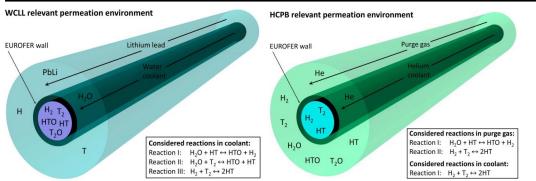


Figure 1. Numerically observed tritium permeation systems. [left] Considered water cooled lithium lead environment. [right] Considered helium cooled pebble bed environment.



CALCULATION OF SOLID HYDROGEN PROPERTIES BY ATOMISTIC SIMULATIONS

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Accurate data on the thermal and physical properties of hydrogen isotopes are needed for various calculations required for the design and optimization of devices and systems needed for the fusion fuel cycle, including tritium storage and delivery system. For solid hydrogen, currently available data are limited, especially for species containing tritium. Since the experimental facilities available for basic tritium experiments are limited worldwide, it is of great significance to develop a computational simulation method and to obtain the missing data on tritium, which is the purpose of this study.

It is known that nuclear quantum effects are large in solid hydrogen crystals. Therefore, it is necessary to use a calculation method that can cope with them. We combine two computational methods in the present study: vibrational analysis using quantum partition function, and path integral molecular dynamics (PIMD). First, PIMD calculations are performed using the PIMD code [1]. As in many previous studies, for simplicity, we treat each hydrogen molecule in the hcp crystal as a particle and use an isotropic two-body potential model constructed by fitting to first-principles calculations [2]. The numerical setup for the PIMD calculations is as follows: 200 molecules ($5\times4\times4$ hcp supercell); 96 beads; 0.1 fs time step; massive Nose-Hoover thermostat with chain length 4; 50000 steps for equilibration and 100000 steps for production. Next, a vibrational analysis of the hydrogen dimer is performed to account for rotational and vibrational contributions within the molecule. Finally, we calculate the equilibrium volume, bulk modulus, enthalpy, and heat capacity at 1 bar for all six isotopes (H₂, HD, HT, D₂, DT, and T₂).

As an example of the calculation results, Figure 1 shows the temperature dependence of the equilibrium volume of hydrogen solids at 1 bar. When compared to the experimental data of H_2 and D_2 solids, it can be seen that the volume is systematically overestimated by up to 3% in the calculations, indicating that the T_2 calculations contain similar errors. Likewise, in comparison with the available experimental data for the H_2 and D_2 solids, the errors in the present calculations are estimated to be up to 20% for the bulk modulus and 5% for the enthalpy, but much larger for the heat capacity due to statistical errors in the PIMD calculations. The details of the calculation results will be explained in the presentation.



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Keywords: Solid hydrogen, thermodynamic properties, bulk modulus, tritium, isotope effects, quantum effects.

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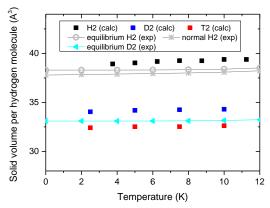


Figure 1. Solid volumes of hydrogen isotopes. The experimental data are taken from Ref. [3].



INVESTIGATION OF ZERO EFFECT OF HYDROGEN ISOTOPES IN LaNi₅ APPLIED FOR *ITER SDS*

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During ITER plasma operation, deuterium-tritium gases with well-defined ratios of D/T are required to be supplied by the different hydrogen storage beds. An inverse isotope effect was found during the desorption of the 1:1 H-D mixture from zirconium cobalt (ZrCo) hydrogen storage beds [1]. Zero effect of hydrogen isotopes was investigated in various metal-hydrogen systems [2-4]. It is worthwhile to find a candidate with zero isotope effect to meet the required D/T ratio during supplying of D-T gases. As a promising tritium storage material, LaNi₅ obtains its rapid and reversible kinetics performance, and its zero effect was investigated near room temperature [2].

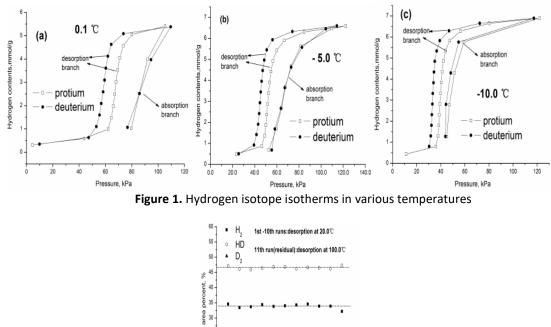
In this work, hydrogen isotope isotherms were performed in a Micromeritics ASAP 2460 volumemetric adsorption analyzer, which was described in our previous study [5]. LaNi₅ sample (4.0g) was immersed in a thermal stat powered by a Refrigerated Heating Circulator (huber Unistat 625w). An online micro gas chromatograph [6] was installed and coupled with the adsorption analyzer, in order to characterize the possible isotope effects during 10 desorption runs of the hydrided sample at 20.0°C and the final run at 100.0°C, which was pretreated at -5.0°C under 100kPa 1:1 H-D mixture.

The results show that zero effect of the absorption branch was found at -5.0°C, as shown in Fig.1b. Inverse isotope effect was shown during absorption when the temperature was higher than -5.0°C (Fig. 1a). Obvious normal isotope effect was investigated in the desorption branch (Fig. 1), which will be applied in hydrogen isotope separation [6]. Zero effect was observed during the desorption process, as shown in Fig. 2. LaNi₅ will be a good candidate for D-T supply near room temperature.

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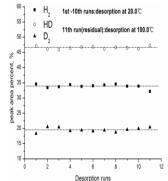


Figure 2. Zero isotope effect of LaNi₅ hydride during desorption

Keywords: Tritium supply, LaNi₅, hydrogen isotope effect, P-C isotherm, online GC

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ANALYSIS OF PRODUCTION OF FUEL TRITIUM FOR NUCLEAR FUSION USING *HTGR*

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Tritium is thought to be used as fuel for fusion reactors, which are expected to be the next generation of energy sources. One of the challenges is to secure tritium needed when starting a fusion reactor, which is estimated to be several kilograms [1, 2]. However, since the amount of tritium naturally present is very small, a method for producing tritium using a high-temperature gas reactor is being considered. Currently, in Japan, tritium production tests are planned in testing furnaces (HTTR) for high-temperature gas reactors. Although the tritium production volume is 1 GBq, the retention behavior of tritium in the material is not known in detail. Based on the experimental results of hydrogen isotope behavior in Zr, Al_2O_3 and $LiAlO_2$ conducted in this laboratory, this presentation introduces the tritium analysis method after conducting tritium production tests in HTTR. The page length must be one page including figures and tables.

A test specimen used in HTTR is shown in Figure 1. In addition to an Al_2O_3 container having a hydrogen transmission inhibiting effect, a tritium production test is performed by further covering the outside of the test body with a quartz tube about 1 mm thick. From the previous evaluation of hydrogen absorption characteristics [3] in Zr, Ni coating is applied to Zr to suppress the formation of an oxide film and provide corrosion resistance. HTTR is scheduled to produce tritium for one year, and the amount of tritium produced will be analyzed in this laboratory. An analyzer (Figure 2) has already been manufactured. Tritium is heated and discharged from both sealed samples in a dry Ar atmosphere. The amount of tritium released is measured through the ionization chamber.

Keywords: Tritium production, Fusion reactors, HTGR, Tritium confinement.



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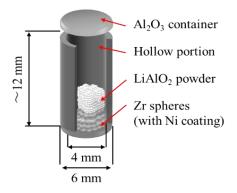


Figure 1. Schematic representation of the test specimen

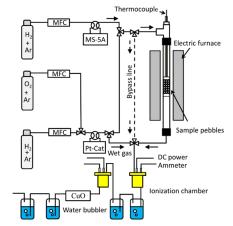


Figure 2. Schematic diagram of the analyzer

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ASSESSMENT OF He-3 RELEASE FROM DEPLETED URANIUM TRITIDE BED CONSIDERING CYCLIC OPERATION USING EMPIRICAL MODELS

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In this study, the amount of He-3 release from tritium-loaded depleted uranium bed considering long-term operation in the fusion fuel cycle is assessed using empirical models. Several works are surveyed to investigate the trend of He-3 released from uranium tritide. The model for He-3 release is developed consisting of two parts, the natural release model and the heating release model, based on the experimental data of Northall and Knowles [1] and Limacher et al. [2]. The natural release model is validated by applying it to other experimental research. The model is applied to cyclic and long-term operation scenarios to assess the amount of He-3 in the headspace of the metal tritide bed. The results show that the He-3 release after long maintenance can be significant and must be separated from tritium before supply to the fuel cycle. During plasma operation and short maintenance, it is shown that the He-3 release is not significant and there is no need for separation.

Keywords: Helium-3, release, tritium, depleted uranium, empirical model, metal tritide bed.

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Tritium breeding and extraction





IN-PILE TRITIUM RELEASE EXPERIMENTS FOR SOLID BREEDERS IN CHINA MIANYANG RESEARCH REACTOR (*CMRR*)

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Tritium release and inventory in lithium containing ceramic pebbles are key properties to be tested. Due to the lack of fusion neutron sources, using fission reactors to build on-line irradiation-induced tritium production facilities is an effective way to study the production/release behavior of tritium breeders under irradiation, as well as to develop and validate the online tritium extraction technology. Recently, a new in-pile experimental loop (Fig.1) has been established in CMRR for ceramic tritium breeder irradiation. The most typical of this loop is the capability of online reloading of irradiated samples. The irradiation temperature in the pellets bed can be controlled within the range of $300 \sim 750$ °C, with a spatial inhomogeneity of ± 10 °C. Up to now, a series of irradiation experiments have been carried out on different breeder materials, i.e. Li₄SiO₄ (7.5 % ⁶Li), Li₄SiO₄ (90 % ⁶Li) and Li₂TiO₃ (7.5 % ⁶Li) pebbles, indicating the stability of the facility. In this paper, the in-pile tritium behavior will be reported during normal operation and during transients in temperature, purge gas chemistry and gas flow. The analysis of the tritium residence time is done based on the in-pile tritium release measurements and the post irradiation examination.

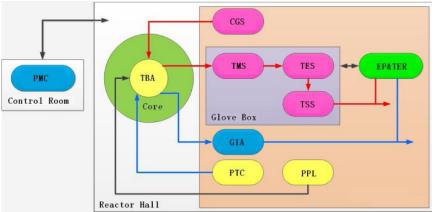


Figure 1. Overall layout of the in-pile experimental loop at CMRR

Keywords: in-pile tritium behavior, tritium residence time, fusion reactors, measurement and monitoring, decontamination and waste management, etc.



TRITIUM PRODUCTION AND NEUTRON ACTIVATION OF 2LiF-BeF₂ (FLiBe) IN FLUORIDE SALT-COOLED HIGH-TEMPERATURE REACTORS AND FUSION REACTOR BLANKETS

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FLiBe (2LiF-BeF₂) salt is proposed as a tritium breeding blanket. It is also used in as a coolant or fuel solvent in fission reactors. In both cases tritium is produced because of neutron irradiation, alongside several other activation products, including other hydrogen isotopes and oxygen. Quantifying production rates of tritium and other hydrogen isotopes is important for the design of tritium management systems. Tritium production and all the other neutron activation reactions on Li, Be and F contained in the salt also have chemical effects on the molten salt coolant, changing its redox potential and thus providing a continuous corrosion driver for metal components. In this talk, the production rates of tritium in the ARC fusion blanket and the Mark-I Fluoride-Salt-Cooled High-Temperature Reactor (FHR) and are estimated and compared to tritium production in other reactors. Production rates to predict the rate of oxidation of structural metal in contact with the molten salt. The effects of isotopic composition of lithium, operational time, reactor power, and primary salt inventory are quantified and discussed.

Keywords: Tritium production, neutron activation, breeding blanket, fluoride-salt-cooled high-temperature reactor, FLiBe, Corrosion



TRITIUM RELEASE BEHAVIORS IN Li₂TiO₃ PEBBLES WITH DIFFERENT GRAIN SIZES AND DEUTERIUM PERMEATION IN CORROSIVE *RAFM* STEELS

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In future D-T fusion reactor, self-sufficiency of tritium is one of the critical issues to maintain steady-state operation of the fusion reactor. Li₂TiO₃ have been proposed as prominent tritium breeder candidates for water cooled ceramics blanket (WCCB). The tritium release behaviors are affected by many effects including grain size, porosity, adsorption water and so on. In present work, the effects of different grain sizes on tritium release from Li₂TiO₃ were investigated. Tritium breeding pebbles have been irradiated by thermal neutrons in Kyoto University. Tritium release experiment was performed in Shizuoka University. Tritium release spectra were obtained by tritium thermal desorption spectroscopy (Tritium-TDS). Liquid scintillation counter (LSC-5100) was used to calibrate the tritium amount. The kinetic parameters of tritium breeders and structure materials (RAFM steels) will be contact during operation of the fusion reactor under high temperature for a long time. The RAFM steels will be corroded by tritium breeders. It will affect tritium recovery. In present work, the permeation and retention of deuterium from corrosive RAFM steels were also investigated.

Tritium retention reduces as the heating temperature goes up. There is large tritium retention in Li_2TiO_3 with grain size of 40µm than that of 2µm at lower temperature. However, almost all the tritium has been released when the heating temperature is higher than 450°C. The normalized amount of tritium releases from breeders with grain size of 2µm and 40µm is similar. The temperature of tritium release peak from Li_2TiO_3 pebbles with 40µm is higher than that with 2µm. The activation energy of tritium release from pebbles with large grain size is higher than that with small grain size. Grain size has a significant effect on tritium release. The tritium release performances are compared between the breeders with micro-grains and nano-grains. The temperature of tritium release peak from Li_2TiO_3 pebbles with nano-grains is a little lower than that with micro-grains. There is smaller tritium retention compared with that of 2µm and 40µm. The activation energy of tritium release from nano- Li_2TiO_3 is the smallest. It is attributed to the effect of nano-grain which has larger specific surface. The comprehensive analysis and comparison of tritium release in breeding materials have been performed.



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In the solid breeding blanket, the blanket structural materials will be in contact with ceramic breeder pebbles under high temperature for years. A corrosion layer will be formed which will affect tritium recovery. Recent research focuses on the corrosion characteristics and deuterium permeability of CLF-1 RAFM steel by Li_4SiO_4 and Li_2TiO_3 pebbles under sweep gas (He + 0.1% H₂) flow. Preliminary study found the formation of double corrosion layers, which is the rich chrome in inner layer and the iron in outer layer. The parameters of deuterium permeability and retention were obtained. The comprehensive analysis for deuterium permeability from original RAFM steel and corrosive RAFM steel was performed.

Keywords: Tritium release, tritium breeder, fusion reactors, corrosion, permeation.



CHEMISTRY CONTROL IN IRRADIATED 2LIF-BeF₂ (FLIBe) MOLTEN SALT BLANKETS AND FLIBE IRRADIATION STUDIES

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Tritium management in 2LiF-BeF_2 (FLiBe) molten salt at temperatures above 500 °C is of interest for fusion and fission reactor. Tritium management is also directly related to chemistry control and management of corrosion of metal components exposed to molten salt because neutron activation reactions impact corrosion and salt chemistry, and salt chemistry impacts tritium management. In this talk a back-ground on chemistry control in FLiBe blankets will be provided, and the relationship between activation reaction rates and chemistry control strategy will be discussed. The design of irradiation-corrosion studies relevant to irradiated FLiBe will also be discussed. The impact of tritium production and other activation reactions of FLiBe on fluid chemistry is quantified. The results are then used to estimate the required supplies of salt and reducing agents for redox and stoichiometry control in the fission and fusion reactors that utilize FLiBe as a tritium breeding blanket or a coolant, respectively.

Keywords: Tritium management, chemistry control, neutron activation, breeding blanket, fluoride-salt-cooled high-temperature reactor, FLiBe, irradiation-corrosion studies



MODELING THE PbLi FLOW INCLUDING TRITIUM TRANSPORT AND PERMEATION WITH GETTHEM

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Tritium self-sufficiency is one of the challenges on the path towards a reliable electricity production from fusion energy by means of the EU DEMO reactor [1]. Its Breeding Blanket (BB) and Tritium Extraction and Removal System (TERS) must be designed to minimize the tritium inventory. The Test Blanket Module (TBM) programme on ITER will provide a validation of the different designs. Both TBM programme and EU DEMO design require substantial R&D, in turn asking for support from reliable modeling to prepare the experiments and compare different solutions.

One of the BB designs that is being considered for the EU DEMO and is going to be tested in a TBM, is the Water Cooled Lithium Lead (WCLL) BB; it adopts water for the cooling and flowing PbLi as a breeder material. The tritium exiting the BB in the PbLi mass flow rate should then be extracted in the TERS to close the fuel cycle. One of the possible technologies identified for the EU DEMO TERS is the Permeator Against Vacuum (PAV): the tritium dissolved in the liquid PbLi flowing within several parallel channels will permeate towards the vacuum pumped on the other side of the channel wall (the membrane). In the geometry considered here, based on the shell-and-tube configuration, the PbLi channels are "U-pipes" surrounded by a vessel, where vacuum is pumped.

The new, accurate model of the tritium permeation across the membrane in the PAV, involving both transport phenomena in the wall and surface processes, was already used to size the EU DEMO PAV [2]. However, besides the component itself, it is important to properly define the interfaces of the PAV in the TERS, and of the TERS in the entire PbLi and tritium loops. The model of such a complex system will be implemented in system-level tools as the GETTHEM code [3], that already includes a model of the WCLL PbLi loop.

As a first step, in the work presented here the permeation model is implemented in the Modelica, object-oriented language used by GETTHEM. The resulting, lumpedparameter component will be able to capture the thermal-hydraulic behavior of the PbLi, to model the tritium transport in the fluid and to estimate the tritium permeated flux supplied to the tritium processing. Such a model is tested here in a loop mimicking the PbLi one in the WCLL and is coupled to the vacuum pumping system on the secondary side. This



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will allow to simulate the normal operation and, in the future, all the possible (also accidental) operating conditions of the EU DEMO TERS using the GETTHEM.

A future validation campaign is also foreseen, possibly exploiting the PAV mock-up tests that will be performed at ENEA Brasimone in EU DEMO-relevant operating conditions. The qualified tool will thus be used to provide feedbacks to the EU DEMO PbLi and tritium loop design.

Keywords: EU DEMO, fusion reactors, modeling, PbLi, permeator against vacuum, tritium extraction.

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LIBRA (LIQUID IMMERSION BLANKET RAPID ASSESSMENT) EXPERIMENT

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Fusion power plants (FPPs) must breed tritium (T) fuel for self-sufficiency and to provide fuel to start additional FPPs. However, conventional T breeding technology, based on solid lithium ceramic breeder material, results in too-low T breeding ratios for fusion market penetration. T supply will become a bottleneck, with limited T available to start new plants, and fusion power will be limited to ~1 GWe on the grid by 2050, assuming first FPP operation in ~2030. The liquid immersion blanket (LIB) concept [1], which utilizes a molten FLiBe salt breeder, enables much higher T breeding ratios (TBR) and a simplified, more economic FPP design. With the LIB, 1 TWe of fusion power on the grid by 2050 becomes possible. While T breeding in FLiBe has been experimentally demonstrated in small salt volumes [2-3], the technology is still at a very early stage of development, and no T breeding tests in a representative fusion environment have been carried out in FLiBe.

The MIT PSFC is pursuing an innovative FLiBe T breeding platform called LIBRA that will demonstrate the validity of the LIB concept and enable the development of robust T accountancy and recovery methods. A D-T neutron generator is surrounded by a cylindrical tank containing >1300 kg of molten FLiBe (**Fig. 1A**), providing breeding in a representative fusion neutron energy spectrum. Sparge and sweep gases carry bred T to detector systems (**Fig. 1B**). LIBRA allows for integrated development and testing of the LIB concept at a lab scale. It is a flexible test platform that will provide a deep understanding of T behavior in FLiBe (**Fig. 1C**), which will in turn inform full-scale LIB designs. The LIBRA tank is divided into quadrants, each of which will have independent salt chemistry and T monitoring systems, facilitating 4 simultaneous T breeding experiments under independent salt conditions. In this talk, we will discuss the history of FLiBe T breeding, the design of LIBRA, its implications for the LIB and fusion power more broadly, near-term testing plans, and eventual plans to demonstrate TBR > 1. We will also provide an update on the progress of the LIBRA facility construction at MIT and supporting FLiBe science research activities.



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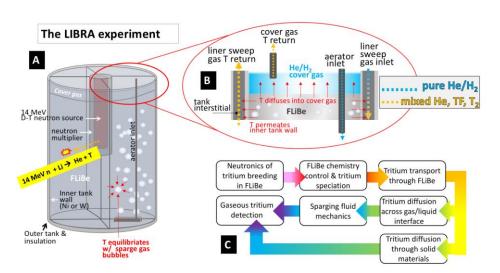


Figure 1. (A) The LIBRA experiment consists of a 14 MeV D-T neutron generator surrounded by a cylindrical volume of FLiBe salt, divided into four sectors, each with an independent T recovery/detection system (B). LIBRA will be the most comprehensive, fusion-relevant test of T breeding and behavior in FLiBe to date (C).

Keywords: Tritium breeding, detritiation, fusion reactors, measurement and monitoring, FLiBe

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SIMULATIONS OF TRITIUM TRANSPORT IN A VACUUM SIEVE TRAY AND RESULTS FOR EXTRACTION EFFICIENCY AT SCALES RELEVANT FOR COMMERCIAL FUSION REACTORS

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Vacuum Sieve Tray (VST) technology that recovers tritium from falling droplets of liquid media containing tritium under vacuum shows strong potential to be the de facto solution for extracting tritium fuel from liquid breeders/coolants used in future fusion reactors. Existing experimental devices have demonstrated that the concept works for liquid metal. However, these devices can only process small flow rates and understanding of the commercial viability relies on extrapolation to relevant scales. We present a threedimensional Monte Carlo simulation of the VST specifically for modelling tritium reabsorption by neighbouring droplets during the fall. It has already been confirmed that the transport of tritium within the droplet is fast, the recombination is not a ratecontrolling process, and the performance of VST is predictable by numerical modelling based on geometry. The distribution of nozzles in various spatial arrangements, the nozzle density, and multi-layered designs to extend fall time given a certain chamber height can be analysed within this framework. Simulations have been performed to demonstrate device sizes large enough to accommodate liquid metal flow rates up to 1 m³/s, which is expected from breeder circulation for the current blanket designs. The impact on plant cost from tritium extraction and the feasibility of manufacturing are discussed in order to design the VST device to be integrated in Kyoto Fusioneering's test facility (UNITY) to accelerate the development of fusion engineering technology.

Keywords: Tritium extraction, vacuum sieve tray, commercial fusion reactor, lithium-lead



STUDY OF CHEMICAL REACTION RATES FOR TRITIUM RECOVERY IN BREEDING BLANKET PURGE GAS

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One of the important issues for the development of fusion reactors with high tritium self-sufficiency is the tritium recovery rate in the outer fuel cycle. For efficient extraction processing, tritium in the form of HT is more desirable to exist in the purge gas from the breeding blanket. It was reported that the tritium released from the breeder pebbles was mostly in the form of HTO. Then, the HTO is expected to react with H₂, which is added in a small amount into the helium-based purge gas, and to produce HT through the isotope exchange reaction HTO+H₂ \rightarrow H₂O + HT.

In our previous works, the tritium recovery rates for the HCCR-TBM operation conditions were estimated by minimizing the Gibbs free energy in the HTO-H₂ mixture assuming the final chemical equilibrium state was reached in the purge gas flow. Moreover, the possible chemical reaction channels and their finite reaction rate coefficients for the isotope exchange reactions have been being studied by the transition state theory (TST) by the authors.

The present study will present a set of chemical reaction rates for the analysis of chemically reacting $HTO-H_2$ mixture flow, and results of the rate equations integrations for the chemical species in the purge gas.

Keywords: HT production, Chemical Kinetics, Purge Gas Flow, Tritium Breeding Blanket



ISOTOPE ENRICHMENT EFFECTS OF LITHIUM BETWEEN LIQUID METAL AND CHLORIDE MOLTEN SALT

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Tritium is necessary as fuel for fusion reactors. It's produced by the nuclear reaction between ⁶Li and neutron in fusion blanket. However, natural abundance of lithium is only about 7.8% for ⁶Li, and the enrichment to 30-90% is required in fusion reactors [1,2]. The mercury amalgam method [3] is the only method of lithium isotope enrichment in practical use. Mercury is difficult to use in the world because of its environmental impact. Although alternative methods of lithium isotope enrichment have been proposed, the best methods have never been established.

Therefore, this research focuses on the lithium isotope exchange equilibrium between liquid metal and chloride molten salt and aims to experimentally clarify the lithium isotope enrichment effects. The experiment is performed with a double liquid layer of lithium-based liquid metal and chloride molten salt (LiCl 58.5 mol.%-KCl 41.5 mol.%) in a capsule made of SS316. The capsules are placed in electric furnace and heated to temperatures above the melting points of them. The capsules of liquid metal and chloride molten salt are stirred inside the furnace by turning them upside down (**Figure.1**). After standing for 12 hours, liquid metal and chloride salt are removed from the capsules. The metal is treated with aqueous solution to become nitric acid solution and the chloride salt is dissolved in water. Each liquid phase sample is measured with Inductively Coupled Plasma Mass Spectrometry (ICP-MS). The ratio of ⁶Li to ⁷Li can be obtained.

Trial runs were completed at 653K with Li-Pb eutectic alloy (Li 17 at.%, LiPb) as liquid metal and ICP-MS measurements were performed. Based on the trial results, Main experiments will be performed at 633K, 673K and 713K with Li and LiPb. The goal is expected to clarify how useful the lithium isotope enrichment effects are in the system with liquid metal and chloride molten salt, and to investigate its temperature dependence. The main results will be shown at the conference.

Keywords: isotope effect, isotope enrichment, liquid metal, chloride molten salt, lithium



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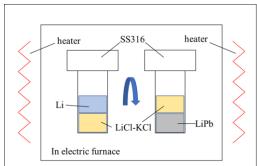


Figure 1. The capsules of liquid metal and chloride molten salt in electric furnace

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DEVELOPMENT OF THE CONTINUOUS COOLING SYSTEM FOR THE CATHODE IN THE GLOW DISCHARGE-TYPE FUSION NEUTRON SOURCE

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The glow discharge-type fusion neutron source generates neutron by inducing the nuclear fusion reaction of deuterium and/or tritium [1]. This type of neutron source has been used for the evaluation of the tritium breeding ratio inside the blanket mock-up in the nuclear fusion field [2]. In this device, a fusion reaction occurs by applying a high voltage between the electrodes and inducing a glow discharge. The fusion reaction is mainly caused by collisions between charged or neutral particles and the hydrogen isotopes trapped on the surface of the electrode [3]. However, the relation between the neutron production rate (NPR) and the abundance of the hydrogen isotopes on the surface is not revealed. The purpose of this study is to clarify this relation.

In our study, three types of cathodes that have different hydrogen affinity from each other were prepared; the titanium-coated, the palladium-coated and the uncoated cathode. The base material was stainless steel (SUS304). The gas inside the chamber was deuterium. As shown in **Figure.1-a**, the case of the titanium cathode showed the enhancement of the NPR. Hence, it confirmed the tendency that the NPR depends on the hydrogen retention of the electrode material. In addition, the result also indicated that deuterium tends to desorb from the surface and the NPR gain becomes lower as the temperature of the electrode becomes higher at high electric input operation. Therefore, it is difficult to control the NPR at the range of high electric power input. If this problem is solved, the stability will be improved and the kind of applications will be expanded.

To solve this problem, the continuous cooling system for the cathode is proposed (Figure.1-b). The feature of this system is that the tube of the water chiller itself has the function of the cathode. Flowing water cools the cathode surface from the inside throughout the discharge operation time. During the experiment, the cathode temperature is controlled by changing the water temperature. In addition, the abundance of deuterium on the cathode is calculated based on the cathode temperature and gas pressure. The transition of the NPR under the same voltage and current condition with changing the abundance of deuterium will be measured. The result is expected to reveal the relation between the NPR and the abundance of hydrogen isotopes in more detail. Additionally, the effectiveness of this cooling system will be discussed at the conference.





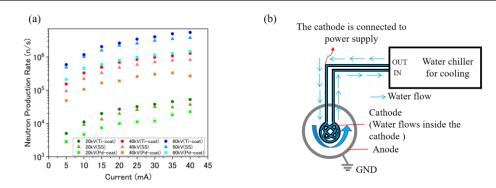


Figure 1. (a) The material dependency of the NPR. Ti, SS, and Pd denote titanium, stainless steel and palladium respectively, (b) the conceptual diagram of the continuous cooling system

Keywords: Fusion reaction, Glow discharge, High voltage feedthrough, Neutron source

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Others

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INTENSIVE TRITIUM SOURCE OF ANTINEUTRINOS FOR EXPERIMENTS ON COHERENT ELASTIC NEUTRINO - ATOM SCATTERING AND SEARCHING OF ELECTROMAGNETIC PROPERTIES OF NEUTRINOS

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We present an analysis of using the intense tritium antineutrino source to measure the coherent elastic neutrino-atom scattering and possibilities to study the neutrino electromagnetic properties (the magnetic moment and electric millicharge). It is predicted that in a such scheme of an experiment the sensitivity to the neutrino magnetic moment will be two orders of magnitude better than the present world best limits on neutrino magnetic moments.

The advantages of tritium antineutrino source:

- more intense antineutrino fluxes at the detector axis with respect to the reactor and accelerator sources;
- a strongly suppressed correlated background;
- small sizes allowing the use of low-background underground laboratories and flow modulation to subtract uncorrelated background;
- the knowledge of the antineutrino spectrum with high accuracy;
- the low energy realize in the tritium beta-decay ($E_0 = 18,6$ keV) guarantees that the bremsstrahlung does no penetrate beyond the source region and therefore there is no need for passive protection between the source and the detector.

The analysis of various types of detectors for realization of the experimental goals is also presented.

Keywords: Tritium antineutrino source, coherent elastic neutrino-atom scattering, neutrino magnetic moment, detector.



TRITIUM OPPORTUNITIES AND TECHNICAL ROADMAP FOR FUSION DEVELOPMENTS WORLDWIDE – CNL AND UKAEA VIEW

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Commercial generation of electricity via fusion technology remains one of the promising alternatives to help meet the challenging targets to decarbonize the global energy system. Fusion technology can play a significant role as part of the long-term switch from carbon-based fuels to electricity, due to superior energy output per energy generation area, usage of abundant fuel elements that can be made available without environmental degradation, and relatively benign activation products that avoid transuranic waste.

Many countries have their own fusion R&D programs and large research efforts are being undertaken in multi-country collaborative efforts such as ITER. Recently, various small company start-ups have been successful in initiating their development programs, funded by government grants and private investments.

While fusion reactors have many different designs, many share common challenges, especially in the area of the deuterium-tritium fuel cycle and associated auxiliary systems. The development of many of the new fusion designs remains dependent on the availability of comprehensive deuterium-tritium expertise. However, the fusion technology developers, in most cases, are having to focus on the physics and mechanical aspects of their technologies due to their complexities. As a result, many of these developers are keen to acquire their tritium management needs through external contracts instead of establishing their own in-house capabilities. As world-leading experts in deuterium-tritium technology, Canadian Nuclear Laboratories (CNL) and the United Kingdom Atomic Energy Agency (UKAEA) are best positioned to support such needs of the fusion industry. In addition, the technology developers would also benefit from the understanding of regulatory requirements related to tritium management that CNL and UKAEA can offer.

This paper broadly explores the fusion market worldwide, identifies opportunities where tritium expertise is key to the development of technology, and presents a view of how CNL and UKAEA are addressing those opportunities for the various fusion developers, ranging from small company start-ups to large multi-country efforts. The paper presents a preliminary holistic view of a technology roadmap and necessary capabilities to address the upcoming challenges.

Keywords: Tritium separation, roadmap, fusion reactors.

National Research and Development Institute for Cryogenics and Isotopic Technologies



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Figure 1. The newly renovated Tritium Facility at CNL – Chalk River Campus

Figure 2. The new H3AT facility currently under construction at UKAEA



PROFICIENCY TESTS AS A TOOL FOR LABORATORY IMPROVEMENT – A VIEW FROM THE OTHER SIDE

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Humans have always observed natural phenomena around them, but their quantitative descriptions are rather new in human history. In relatively closed communities, it was important that the relative results were comparable and traceable to local arrangements. But in the time of raising globalization, we need and expect more than that. In addition to absolute values, traceable to uniformly and accurately known agreed measurement units, we also want additional information and answers to the questions: how accurate, precise, repeatable, sensitive, trustable is the result? The answers to these challenges are detailed descriptions of measurement methods and records of their validation, identification and regular control of key parameters that may affect the measurement result. A quality system QA/QC is born.

Interlaboratory comparisons and proficiency tests (PT) are an increasingly popular necessity used by laboratories as an external, independent confirmation of the quality of their results. The process is known: the laboratory introduces the measurement method, validates and test it with the participation at PT. How to find and choose the most appropriate PT, how to respond to results that are not just a confirmation of the laboratory's capabilities but also an opportunity for the laboratory to identify its weaknesses, to improve its measurement practices, to plan the progress? How can the laboratory influence the provider of PTs?

But, there is also the story of the other side, other perspective. How does the proficiency test look like from the side of PT organizer? Is providing the samples with a known true value and issuing the evaluation report to the laboratory the only task of the organizer? Can the provider of PT do more for the labs and how? What are the challenges, opportunities, problems, weaknesses?

In the lecture, I will try to answer the above questions on the example of IARMA, a relatively new and small provider of proficiency tests and reference materials on the world stage on the example of tritium.



VIMA – THE SPINNING ROTOR GAUGE TO MEASURE THE VISCOSITY OF TRITIUM BETWEEN 77 AND 300 K

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Experimental values for the viscosity of the radioactive hydrogen isotope tritium (T_2) are unavailable. Values for the viscosity of tritium found in literature are either ab initio calculations using classical kinetic theory, which do not reflect the importance of quantum mechanical effects at low temperatures [1], or based on extrapolation by mass ratios as well as an empirical factor derived from deviations of the H₂ and D₂ viscosity measurements from this simple scaling [2]. One reason for the empirical factor might be differences in the interaction potentials of the isotopologues [3]. Accurate data of the tritium viscosity will allow for further testing of the models for the viscosity of diatomic molecules and can be used as a test of their interaction potentials.

As a first step towards measuring the viscosity of tritium, an experiment, based on a spinning rotor gauge (SRG) [4], to measure the viscosity of the non-radioactive isotopologues of hydrogen and their binary and tertiary mixtures, has been set up at the Tritium Laboratory Karlsruhe. We were able to show, that for measurements at room temperature and at liquid nitrogen temperature, the uncertainty on the measurements is between 1 and 2 % and our measurements are in good agreement with literature values [5][6].

The highest impact on the uncertainty of the measurements is given by the thermal stability of the system. This has more than one effect: First, as the SRG can only measure the viscosity relative to a reference gas, helium in our case, it has to be assured, that the measurements with helium and the sample gas are done at exactly the same temperature. Secondly, there are diverse effects which change the gas temperature slightly, depending on the measurement procedure. Therefor thermal stabilization is most important for such an experiment, to guarantee high accuracy in measurement data. We developed a system, with which we will be able to have a temperature stabilization of 0.2 K.

In this contribution we present our cryogenic setup which will be able to measure the viscosity of gases in a continuous range between 77 and 300 K with an uncertainty of 1%.

Keywords: tritium, viscosity, TLK, spinning rotor gauge, cryogenic setup



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DEVELOPMENT OF Ti-³H TARGET FOR RADIOACTIVE ISOTOPE PHYSICS EXPERIMENTS

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The exotic nature of radioactive isotopes (RIs) has been attractive research subjects in modern nuclear physics. The unbalance of neutron and proton numbers in RIs introduces novel shape and cluster structure of nuclei which are different from the natural stable nuclei. RIKEN RI beam factory (RIBF) can provide intense fast RI beam, and allows us to study fundamental properties of rare RIs.

In our project, we newly introduce a tritium target for experiments at RIBF. The tritium has a neutron-rich nucleus consisting of two neutrons and one proton. By transferring this neutron-richness to the RI beams provided by RIBF, we can reach unexplored region of neutron-rich nuclear states.

Among various implementations of tritium target, we chose the physical form of tritiated titanium (Ti-³H). A Ti-³H target had already been used in low-energy experiment (a few MeV per nucleon) [1], but the target thickness was thin and just about 1 \square m^t. We needed a thicker target with far larger amount of ³H for high-energy experiment at RIBF (up to 350 MeV per nucleon).

In this work, we have produced a self-supporting Ti-³H target with a thickness about 80 times larger than the previous study [1]. We first optimized the fabrication procedure through the production of the Ti-²H targets, where we searched for the conditions of making flat and crack-free Ti-²H foil. Second, we constructed a compact Sieverts device accommodated in the tritium glove box environment at HRC, Univ. Toyama, and produced Ti-³H target based on the optimized procedure. We successfully obtained a Ti-³H target with a dimension of 9.2 mm × 12.0 mm × 80 \square m and a total tritium amount of 1.58 TBq (Figure 1).

The target was sent to RIBF for the RI beam experiment. The first experiment was performed in June 2021. We performed a triton-triton scattering experiment and searched for tri-neutron states, a quantum state consisting of only three neutrons. A high-intensity triton beam (170 MeV per nucleon) was produced by a fragmentation process of ⁴He beam



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[2], and was irradiated onto the Ti-³H target. The experiment was successfully done and the data analysis is now ongoing.

In the presentation, we will explain scientific background, production procedure of our thick Ti-³H target, and experimental conditions in RIBF.

Keywords: Tritium target, metal hydrides, nuclear physics, radioactive isotope beam experiments

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Figure 1. Produced Ti-³H target. The amount of ³H loaded in the Ti foil with 80 $\mathbb{D}m^t$ was 1.58 TBq. The foil is supported by a frame made of type 304 stainless steel.



USE OF MAGNESIA CONCRETE FOR SOLIDIFICATION OF AQUEOUS LIQUID RADIOACTIVE WASTES DONTAINING TRITIUM

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The article discusses the possibility of using magnesia concrete for solidification of liquid radioactive wastes containing tritium. The authors proposed using partially or completely dehydrated magnesium chloride to convert tritium water contained in liquid radioactive wastes into magnesia concrete – the final product of solid radioactive wastes. At the same time, the filling capacity of the final solid compound with water containing tritium reaches 48 percent of compound's mass.

Keywords: Tritium liquid radioactive wastes, magnesia concrete, solidification wastes.



NEUTRONIC ANALYSES OF TRITIUM PRODUCTION BY IRRADIATION OF THE LITHIUM CERAMICS PEBBLES IN THE WWR-K REACTOR

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One of the six tritium breeder module concepts for fusion facilities is the helium cooled pebble bed. In this concept, the breeder material is lithium ceramic pebbles [1, 2]. In Kazakhstan, on the basis of the WWR-K reactor [3], research is being carried out to support the development of test breeder modules of tritium. For this purpose, special irradiation capsule is being developed in which conditions are created close to nuclear operational ones in fusion facilities. Predict the production of tritium in order to select the optimal technique for in-reactor gas release measurement and further utilization of tritium, as well as the effect of the irradiation capsule on the neutron-physical characteristics of the reactor is necessary up to performing reactor experiments. Neutronic analysis was performed using the MNCP6 transport code and the both ENDF/B-VII.1 and JEFF-3.1 nuclear data libraries. In this work, both single-phase lithium ceramics (Li₂TiO₃) and twophase ($Li_2TiO_3 + Li_4SiO_4$) were considered. The production of tritium while irradiation of two-phase lithium ceramics during one regular irradiation cycle was 1.8 Ci per gram of sample, and while irradiation of highly enriched single-phase lithium ceramics - 11.3 Ci per gram of sample. The study of the self-shielding effect of pebbles along the diameter of the irradiation capsule demonstrated that the thermal neutron flux in the center of the irradiation capsule is almost two times less than at the periphery.

Keywords: neutronic analysis, lithium ceramics, pebbles, WWR-K, tritium breeder module

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TRITIUM INGRESS DEPTH STUDY IN FUSION RELEVANT MATERIALS

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Tritium accumulation in fusion relevant materials is a challenge for many areas including waste management and tritium inventory control. The University of Rochester has published tritium permeation depth profiles of tritium soaked 316L Stainless Steel. In collaboration with UKAEA, there are imminent plans to expand this programme to many more fusion relevant materials. There are over 100 samples from 8 different metals including Eurofer 97, ODS, XM-19 as well as erbia coated 304 stainless steel waiting to be analysed. A slew of measurement techniques will be used at both UKAEA and the University of Rochester to assess the surface and depth profile of tritium inside these metals. Analysis methods include acid etching, Secondary Ion Mass Spectroscopy (SIMS), Focussed Ion Beam (FIB) milling and Thermal Desorption Spectroscopy (TDS) amongst others. The University of Rochester also has its own Atomic Layer Deposition (ALD) system which can be used to coat samples in anti-permeation coatings to investigate the permeation seen in these. The UKAEA can also employ the DELPHI system to ion damage samples which will exhibit properties more representative of materials exposed to fusion reactor conditions. An opportunity is also offered for the attendees to suggest their own materials to be analysed.

Keywords: Tritium; Permeation; Coatings.



NEW ACHIEVEMENTS OF H₂-HD-D₂ ISOTOPIC SEPARATION WITH THE CRYOGENIC DISTILLATION EXPERIMENTAL STAND FROM ICSI CRYOGENIC LABORATORY

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At ICSI since 1996 a hydrogen and deuterium distillation module based on a liquid nitrogen cycle combined with a J-T valve has been developed. In this older version the CD system has only one distillation column, with low cryogenic power and without any possibility of separation of the 6 isotopologues. Therefore, was decided to modified the total cryogenic power of the CD system and to have four columns for H2, HD, HT, D2, DT and T2 separation. The "CRYO-HY" structural funds project which was finished in 2015 and which, besides the construction of a low-temperatures laboratory for cryogenic fluids, also, increases the processing power of the actual ICSI-PESTD (Experimental Pilot for Separation of Tritium and Deuterium) for all the future demands. The new refrigerating system has a cryogenic helium cycle with a power of 1 kW at 20 K, and is designed to provide various operating modes of cryogenic distillation at different flows. The cooled helium gas (16 K -24 K) supplies the condensers of the 4 distillation columns (CD 301, CD-302, CD-303 and CD-304). In order to lower the tritium inventory, it is necessary to lower the diameter of the last columns from a cryogenic distillation cascade. The proposed diameters of the last two columns were 15 mm and 7 mm, with lengths of 3.5 m and filled with Heli-pack B. ICSI-PESTD is a detritiation experimental plant which is built to make the separation of a quantity of tritiated heavy water with an activity which must be in accordance with the operating license, and has several modules, like: Liquid Phase Catalytic Exchange module, Purification module, Tritium Gas Handling System, HVAC system, Monitoring and storage system. There have been campaigns to verify the functionality of this installation [1] but it has proved that is quite difficult to achieve the parameters resulting from simulation and design, especially for the last two columns with small diameters [2]. It was, therefore, important to carry out further research into the behavior of small diameter columns and to find appropriate solutions so that when the whole plant is commissioned, the parameters can be adjusted for a higher separation factor and for safe and correct operation. Prior to 1996, three mathematical models for the design of cryogenic distillation columns for multicomponent mixtures were developed at ICSI using the Lewis-Matheson, Underwood and Wang-Henke methods. Based on the last-mentioned model, Wang-Henke, simulations



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were performed to find a possibility to link the existing cryogenic distillation process within ICSI-PESTD, to a new CECE module [3].

In the ICSI Cryogenic Laboratory, with an old experimental setup for cryogenic distillation of H2-HD-D2, were measured the ICSI B5 and B7 ordered package at diameters up to 50 mm [4].

Starting from these experiments have been decided to develop a cryogenic distillation technology, step by step, starting from the design and intensive testing of the key elements, i.e. cryogenic distillation column condenser and boiler, ordered and disordered packing, as well as an inter-column heat exchanger model and those were tested using the new cryogenic distillation stand developed in the Cryogenics Laboratory ([5] and [6]). In the framework of Romania's National Research Plan, one of the research projects carried out by ICSI Rm.Vâlcea in the last 4 years (2019-2022) was "Development of test methods for materials used in cryogenics and cryogenic technological systems for isotope purification and separation". The project had broader areas of research related to materials testing at cryogenic temperatures and helium isotope separation. The following research activities have been developed in relation to hydrogen isotope separation: experimental stand for measuring the thermal conductivity of materials used at cryogenic temperatures, cryo-adsorption/cryo-condensation tests and experiments, tests of cryogenic distillation equipment for the development of cryogenic hydrogen isotope separation solutions, development of a conceptual model of a matrix heat exchanger (MHE). All the experiments have been concentrated to characterize new types of packing at total reflux (without product extraction), in order to determinate the HETP, and to verify the operating properties of a new condenser and boiler and a new heat exchanger, based on the work described in performed in [7].

Keywords: cryogenic distillation, hydrogen isotopes, heat transfer, Matrix Heat Exchanger

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