OVERVIEW OF CURRENT TRITIUM RELATED CHALLENGES

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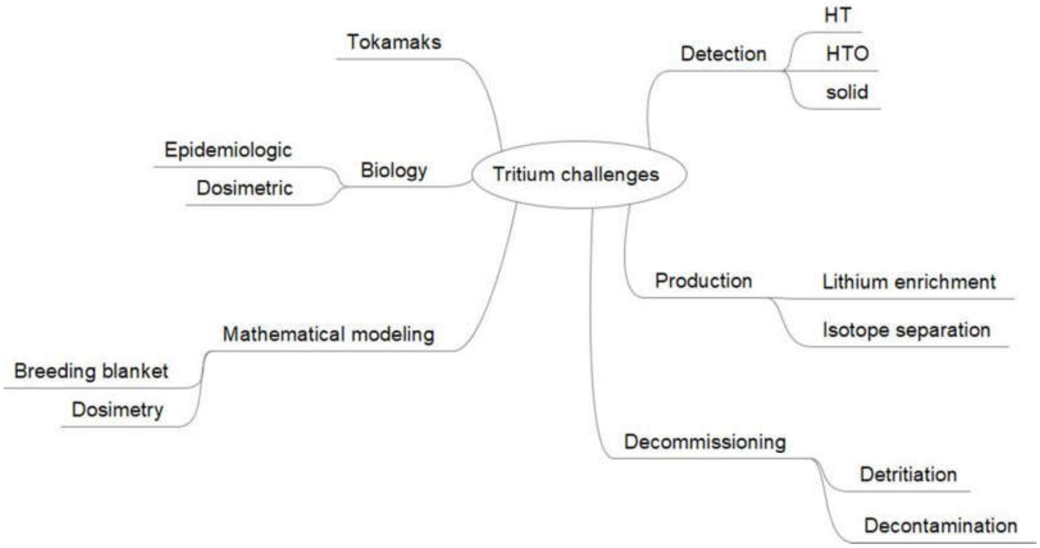
Abstract: The aim of the contribution is to provide an overview of current challenges related to the tritium. Tritium as a radioactive isotope is subjected to the regulation. It is produced artificially at some types of nuclear power plants as a by-product of the fission reaction. Knowledge of tritium behaviour and properties become important because it is used as fuel in tokamaks. Although tokamaks exist for sixty years, not all related challenges have been successfully solved yet and new questions have arisen along the way. The challenges related to tritium are divided into categories based on corresponding scientific disciplines: tritium detection and analysis (technological tokamak parts), tritium production connected with lithium enrichment, hydrogen isotope separation for tritium purification (fuel for tokamaks) or decontamination (decommissioning of experimental facilities and future plants). The decontamination of tritiated parts needs to be optimized for future decommissioning of tokamak parts. Biological studies, mathematical modelling, and radiology focused on tritium is being developed to increase public safety and to describe the transfer of tritium in materials, biological systems and the environment. Identified and listed challenges are known problems for experts in the field. Authors are focusing on main issues that are currently under investigation in scientific community as were presented on the first Tritium School organised by the TRANSAT project in Ljubljana 2019.



Keywords: *tritium use, tritium analysis, tritium production, tritium challenges.*

1. INTRODUCTION

The First Tritium school [1] held in Ljubljana 2019 was chosen for summarizing current challenges related to tritium because it provided well-balanced information about well-established and newly developed technologies. The First Tritium School was organized within the TRANSAT project [2] (TRANSversal Actions for Tritium) with main objectives to provide technical solution and new technologies that mitigate the tritium release, improve tritium management and among others also deepen the knowledge on tritium radiology.

The information provided on the conference enabled newcomers in the tritium field to obtain basic knowledge as well as advanced information required to comprehend the problematic. Authors of this contribution are summarizing basic information and highlight some challenges that are under investigation in the tritium field. The work should help anyone new in the field to easily obtain a basic overview on the problematic.

2. 1. Tritium properties

Tritium [1], [3], [4] is a radioactive isotope of hydrogen with symbol T or ^3H. It consists of one proton and two neutrons. Its half-life is approximately 12.3 years, it decays by beta decay into ^3He. Beta particles from tritium decay can penetrate about 6 mm of air and about 6 μm of water. It has a very low mean energy of 5.7 keV, and the maximal energy of 18.6 keV. It is naturally produced in the atmosphere by cosmic ray via nitrogen spallation. The most common form of tritium is gas (HT or T_2), tritium oxide (HTO) also called tritiated water, and organically bound tritium (OBT) in methane, metal hydrides, solvents, etc. Chemically tritium behaves as hydrogen and it is naturally present in surface waters (0.4 – 1.2 Bq/l). Tritium can enter the human body by inhalation (HT), ingestion (HTO, OBT) and skin transfer (HT, HTO). Blood absorption of tritium is rapid and complete, that leads to a uniform distribution in body tissues. The passage of tritium into the blood as OBT (i.e. T bound to carbon in the non-exchangeable form) is about 20 times higher than in form of HTO. The retention of tritium in the form of HTO in the human body is within the period of 10 days, in the form of OBT 40 days for adults.

2. TRITIUM CHALLENGES

2. 1. Tokamaks

The tokamak is one of several types of magnetic confinement devices being developed to produce controlled thermonuclear fusion power. The most preferred reaction in thermonuclear fusion [1], [5] is

$$D + T \rightarrow \alpha + n + 17.6.\ \text{MeV} \qquad (1)$$

based on its largest probability cross-section and lowest energy with comparison to other reaction as D-D, D-3He, and T-3He that are also considered for the fusion.

Former Tore Supra (operated between 1988 and 2010) nowadays WEST [6] (upgraded between 2013 and 2016) is located in France. Joint European Torus [7] (JET) is located in the UK. The ITER (International Thermonuclear Experimental Reactor) under construction in France is designed to demonstrate the scientific and technological feasibility of fusion energy. One of the challenges connected with tokamaks reactors is the increasing need for fuel that rises with the increase of plasma volume needed. Tritium consumption in fusion system is very high and is considered to be about 56 kg per 1 GW per year, therefore DEMOnstration Power Station[8] (DEMO) will consume 112 – 224 kg of tritium per year.

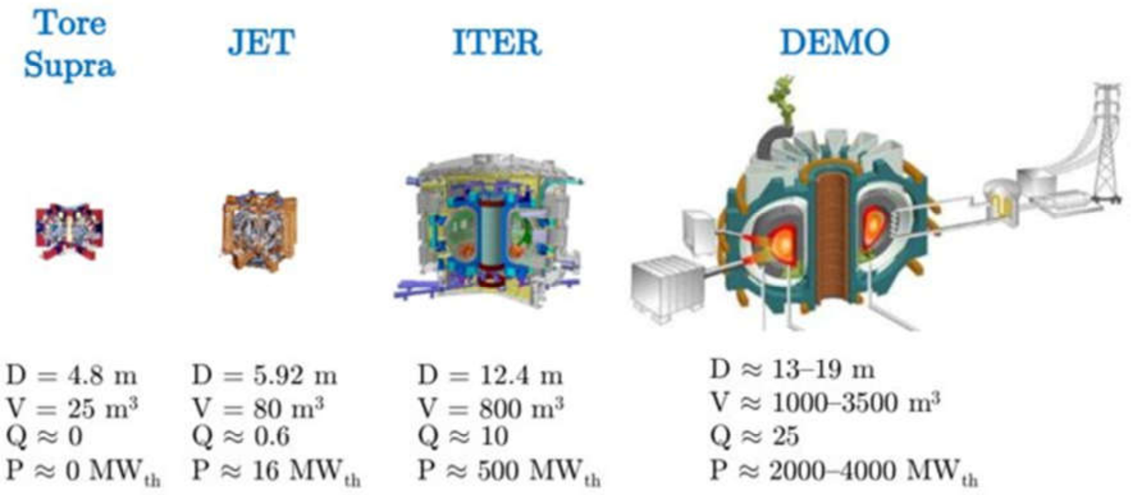


Figure 1 Comparison of Tore Supra, JET, ITER and DEMO [9]. *D - diameter, V - plasma volume, Q - the ratio of energy output to the energy needed for heating the plasma.*

2. 2. Detection

Tritium is a radionuclide that is difficult to detect mainly because of its energy, type of emission (β-) and its chemical and physical behaviour [4], [10]. The most used technique for liquid samples is LSC – liquid scintillation counting. However, with this technique is problematic to measure tritium in coloured or opacity liquid because the technique is based on light detection (the transfer of electron energy to photons via phosphor). Any other liquid than clear transparent one would absorb released photons. The detection limit (DL) for LSC is in the scale of tens Bq/l.

To analyse tritium in a gaseous form it can be used an ionization chamber (not specific to tritium, DL around 2.5 kBq/m^3), proportional counters, mass spectrometry, Raman spectroscopy [11] or gas chromatography.

In a solid form, tritium can be analysed using calorimeters, pyrolizers, phosphor screens, Beta Induced X-ray spectrometry, SiPMs with solid scintillators [12] and other methods. Usually, the solid material is pyrolyzed, tritiated gas is oxidized to tritiated water that is trapped in aqueous solution which is analysed using LSC.

2. 3. Biology

To determine the effect of tritium on living organisms and to estimate the associated risk, epidemiological (the study of the distribution of disease in populations and the factors that determine the risk of disease in these populations) and dosimetric methods that deals with relative biological effectiveness [13] (RBE) are combined. RBE is a ration of reference absorbed dose of radiation of a standard type, and the absorbed dose of radiation of evaluated type that causes the same amount of biological damage. Both doses are quantified by the amount of energy absorbed in the cells. The RBE for tritium varies between 1 and 5 depending on the reference radiation used in a study. Biokinetic models consider a homogenous distribution of tritium in tissues, but that is not relevant for DNA/RNA precursors. Tasks needed to be done are an investigation of tritiated organic molecules and their biokinetic. There is still a lack of technical standards in areas such as plants sampling methodology. As a result of the tokamak operation, there is a need to investigate tritiated dust and its biological activity. There is a lack of more precise data in the epidemiological studies [14]–[16] that quantify tritium-specific effects and distinguish it from another radiation modes.

2.4. Mathematical modelling

Mathematical modelling is useful for the preparation and validation of tritium mass transfer models that are used for calculating concentrations and flows in different reactor components. Models can be stratified based on their complexity on systematic models forming process flow diagrams and detail models focusing on tritium transport in specific geometries**.** Mathematical modelling is also utilized in dosimetry to interpret biological effects of internal emitters causing energy depositions in a small volume related to cellular size. The modelling focused on nano or micro scale dosimetry estimates potential DNA damage. There are still areas that needs to be explored such as determination of HTO role in DNA hydration shell or damage caused by T incorporation in DNA.

2. 4. Production

To discover low cost procedure for tritium generation is of great importance because the price of tritium, that can be used as a fuel for the tokamaks, is about 10 000 $ per 1 g [1]. That is because of natural occurrence of tritium is low and still needs to be purified prior usage as a fuel. To ensure fuel self-sufficiency there is a need to generate tritium during the fusion. Tritium is primarily produced in a breeding blanket by following reactions:

$$ {}^{1}_{0}n + {}^{6}Li \rightarrow {}^{3}_{1}T + He + 4.8\ MeV \tag{2} $$

$$ {}^{1}_{0}n + {}^{7}Li - 2.466\ MeV \rightarrow He + n' \tag{3} $$

A breeding blanket covers the interior of the fusion reactor vessel and enables to use neutrons in the reaction with Li (reactions 2 and 3). Tritium generated in the breeding blanket will be used in the reactor as a fuel. The ^{6}Li reaction is the preferable because it has positive energy output and thus fulfils the aim of the breeding blanket to ensure energetically self-sufficiency. Therefore, in a breeding blanket typically is used high enriched lithium $^{6}Li \approx 90$ %. There are two major concepts of breeding blankets: Helium Cooled Pebble Bed (HCPB) and Helium Cooled Liquid Lead (HCLL). First concepts use Li in the form of Li_4SiO_4 or Li_2TiO_3 as a ceramic breeding blanket material, where the enrichment of ^{6}Li is between 40 to 60 %. The second concept uses eutectic LiPb with 15,8 atom% Li with ^{6}Li enrichment around 90%. If the HCLL concept would be used in DEMO breeding blanket, the total weight of pure ^{6}Li is estimated to 53 t.

Lithium enrichment

The ^{6}Li isotope [17] is sparse in the natural lithium (only 7,5% of the ^{6}Li in natural lithium) and therefore lithium must be enriched before it can be used in the breeding blanket. Well documented and effective enrichment system is the COLEX which is based on chemical exchange of Li-amalgam vs LiOH-aqueous solution. COLEX has the separation factor $\alpha = 1.059$ at 0 °C. The enrichment process is acceptable when $\alpha > 1.03$ [1]. Although there are several newer and promising lithium enrichment systems (Chemical exchange with ion exchangers: organic - $\alpha = 1.014$, inorganic - $\alpha = 1.022$ [1], Chemical exchange with intercalation systems $\alpha = 1.023$ [1]), these methods have been tested only in a laboratory or technical scale. The challenge is to improve the COLEX process to obtain a better separation factor and to improve environmental safety.

Isotope separation

The higher price of tritium opens a potential for recovering and concentrating tritium from tritiated materials or waters contaminated by tritium. Tritium gained during the detritiation of solid materials is mainly captured into water in the HTO form. Nowadays, well documented cryogenic distillation is used [18]–[20]. A promising separation (HT from HTO) is a membrane separation using Combined Electrolysis Catalytic Exchange (CECE). Membrane separations have the potential to replace a cryogenic distillation. A pilot plant based on membrane separation were reported [21], but the challenge is to construct an efficient membrane in large scale; as can be a graphene membrane. The size of separators based on graphene membrane was reported only in a laboratory scale [22]. Consequent challenge will be in developing a scaleup technology.

2. 5. Decommissioning

Tritium as well as hydrogen has the ability to penetrate into metal materials [23]. This ability is undesirable as the inner wall of reactor vacuum vessel is covered by tungsten. Tungsten is used for its physical properties such as high melting point, high thermal conductivity etc. Plasma facing tungsten components are irradiated and absorb high portions of tritium which can lead to the formation of blisters [1.] These blisters can erupt after receiving a high amount of tritium and tritiated tungsten dust is distributed in the reactor. Due to the high price of tungsten and tritium, there is a need to recycle tungsten dust collected from the reactor and to remove the tritium from tritiated tungsten parts in the reactor wall.

One of the methods for detritiation of the metal parts is a high temperature metal treatment. The high-temperature method [24] uses high temperatures for the detritiation of metal materials. Tritiated metal parts are moved into the furnace with the temperature around 1000 °C with flowing air. After the heating time around 6 hours, the captured tritium is released from metal and due to the process air, the flue gas is oxidized:

$$2HT + O_2 \rightarrow 2HTO \tag{4}$$

$$2CO + O_2 \rightarrow 2CO_2 \tag{5}$$

The tritiated water vapor is captured within the series of scrubbers (bubblers in laboratory scale) filled with water. In the next step, tritium in the water is purified for reuse in the fusion reactors. The actual process of refining tritium is the series of electrolysers for gaining the HT gas and the cryogenic distillation process for purifying the HT from H_2 gas. The separation factor is high, but high costs limit this process. There are other processes with lower cost and higher separation factor (such as membrane separation), but they are still being tested in the laboratory scale. The challenge is to convert these lab-scale technologies into pilot plant scale and compare it with the existing methods.

3. CONCLUSION

This contribution provides short summary of current challenges in the area connected to tritium. World-wide interest in tritium and related technologies increases because of its use as a fuel in tokamaks and its presence as a radioactive contaminant. More intense work with tritium leads to exploring the effects of both real and hypothetical contamination on nature and humans. More technologies have been developed in the area of decontamination and decommissioning.

There exist over 22 thousands publications connected to a tritium subject listed at the Web of Science [25] and over 140 thousand on the Taylor & Francis Online [26] database, therefore only small selection of possible challenges was highlighted in the text. The increasing interest within tritium can demonstrated by graph based on data obtained from the Web of Science. This text should be beneficial for anyone who seeks for a short overview.

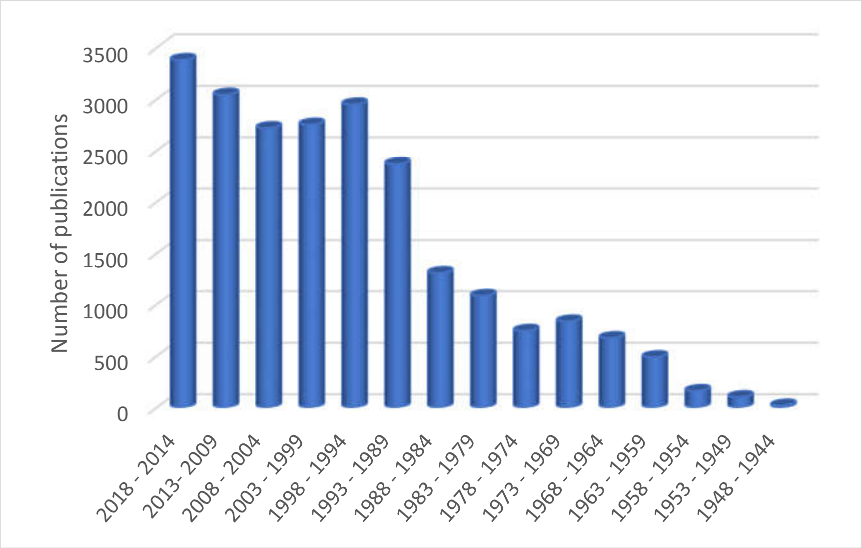


Figure 2 Number of publications in 5 year span based on data from Web of Science [25]

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