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Can tritium monitoring and control requirements be met by existing technologies?

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Can tritium monitoring and control requirements for DEMO 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 practices 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.

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1. Introduction

Fusion power plants such as DEMO will use the fusion reaction between the hydrogen isotopes deuterium (D) and tritium (T) for energy production. Using tritium involves the same difficulties as working with the other hydrogen isotopes, hydrogen and deuterium, such as metal embrittlement. Additional handling challenges are also introduced due to its radioactivity and potential non-proliferation issues caused by tritium's capability to increase the sophistication of nuclear weapons programmes.

Due to these challenges, the DEMO fuel cycle will need a Tritium Management and Control system. This system will account for tritium inventories and flows at all locations in the fuel cycle and at all times, to ensure that the following four requirements are met:

1. Safety; reduce the amount of tritium that is potentially released in accident scenarios to specified limits, maintain concentrations and amounts of substances at safe values to avoid dangerous situations such as creating explosive mixtures, monitor and minimise worker radiation exposure

2. Environmental protection; limit the amount of tritium (gas, liquid, solid) released both during normal operations and accidents
3. Non-proliferation; reduce the likelihood of material diversion/theft and identify its occurrence by specifically monitoring for any diversion of tritiated material, which might be used for purposes related to nuclear weapons
4. Plasma and process control; ensure stable compositions of plasma and gas mixtures in fusion reactors and monitor processes to detect leaks promptly, enable adequate control of processing systems within the fuel cycle

These requirements each impose the need for tritium detection at different locations in the fuel cycle [1] and at different time intervals. Current experience with a tritium management and control system only exists from experimental nuclear fusion reactors like JET. However, simply adopting this system for a DEMO fuel cycle is not practical, due to various reasons: JET operates in pulse mode and uses small quantities of tritium, allowing for most measurements to take place in batch mode. Moreover, most of the accountancy operations that accompany any operation in the JET Active Gas Handling Facility (AGHS) are done by hand. In contrast, DEMO will operate in (near) continuous mode, requiring suitable detection methods, while the implementation of automated systems is necessary to reduce the staffing requirements and likelihood of human errors. Further, a DEMO plant will also both consume and produce significant amounts of tritium, and some of that which is produced will be exported to other fusion power plants. The large amounts of tritium present will provide a larger risk of diversion of relevant quantities for proliferation activities and the additional complexity of future fusion power plants may make material diversion easier to conceal. Although there is no regulation from the IAEA regarding tritium accountancy right now, and control is imposed on a national basis, it can be expected that this may change. It should however be noted that the proliferation threat posed by tritium is that of improvement of existing nuclear weapons programmes, rather than the spread of nuclear weapons to new countries. Without weapons-grade nuclear material the possession of tritium is not a proliferation concern, although it may reduce the breakout time of nation states.

The outline of this paper is as follows. First the different requirements will be discussed in more detail, explaining the purposes they serve and the locations in the fuel cycle where these measurements need to take place. The locations all come with varying sets of conditions, that need to be categorized, and the applicability of available detection technologies to these condition categories will be discussed. This paper concludes with recommendations for improvements in the applicability of detection technologies, how these can be tested and for which conditions new technologies will need to be developed.

2. Tritium detection locations

There are four different purposes for which tritium accounting and monitoring is needed in the DEMO fuel cycle, each with its own requirements regarding accuracy, frequency, and locations where tritium detection needs to take place.

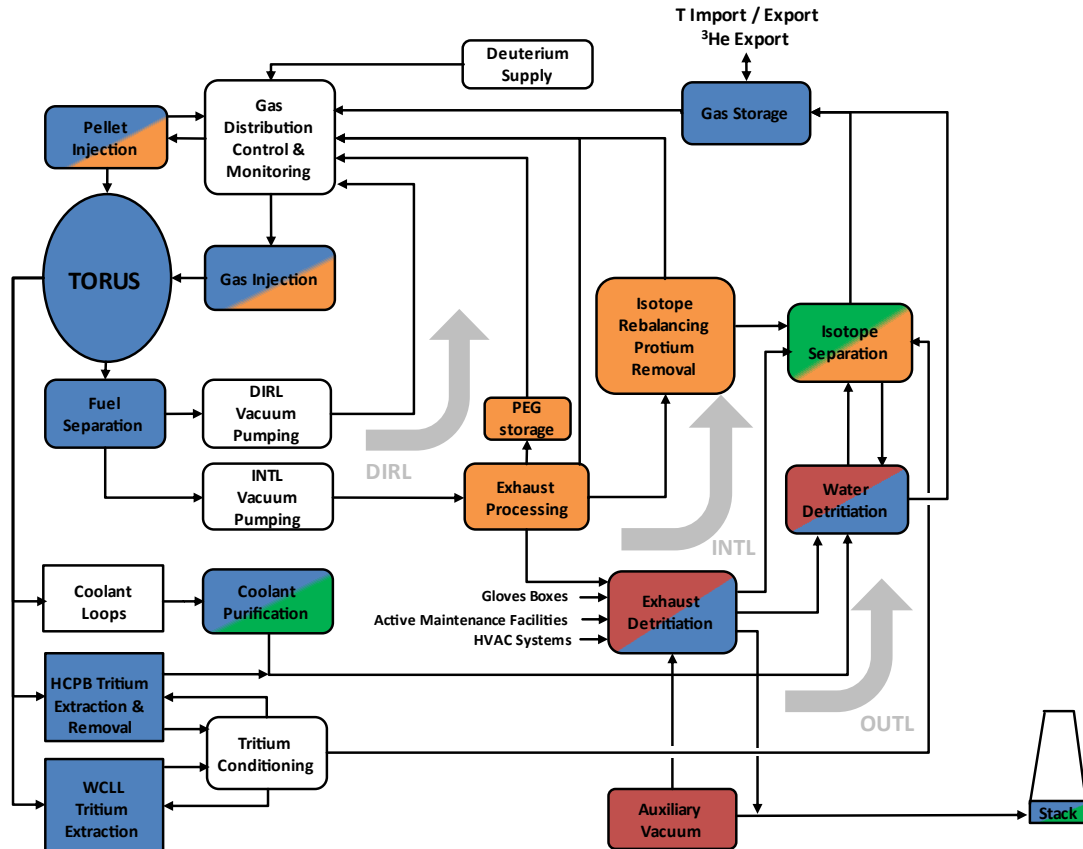


Figure 1: Diagram of subsystems in the fuel cycle [2], with colours indicating for which purpose tritium measurements need to take place. Blue: non-proliferation, green: environment, red: safety, orange: plasma composition control. General process control measurements can be expected to take place in almost every location and are hence not displayed here.

Non-proliferation

Tritium itself is not a fissile material, but it can be used to enhance the performance of nuclear weapons. As such, Ontario Power Generation has made an agreement with the UK that all tritium that is supplied to JET is being used for peaceful non-explosive uses of nuclear energy only [3]. As such, inventory audits need to take place annually, even though tritium does not fall under the Non-Proliferation Treaty as defined by the IAEA.

For now, since there is no specific guidance regarding tritium accountancy in fusion reactors, only assumptions on future regulations can be made, drawing from experience from the fission industry and other tritium handling facilities. It can be assumed that for non-proliferation purposes, all tritium quantities both entering and leaving the plant will need to be monitored. These include all tritium quantities that are imported from elsewhere, produced and extracted from the breeder blankets, burnt in the fusion reaction, exported to elsewhere or discharged as waste in either gaseous, liquid, or solid form. These quantities need to be measured, at least annually, and will be reported to the regulators. It can be expected that most of these measurements, such as the amount of tritium imported or exported, will take place in batch-mode. Moreover, a (planned) shutdown of the plant for maintenance will provide an opportunity to pull back tritium from some subsystems and perform accountancy on these. However, other processes such as the burn-up and production rate of tritium in the fusion reaction can only be determined indirectly, for example using neutron diagnostics [4], [5]. Another challenge will be to determine the “losses” of tritium by being

trapped in areas such as the first wall and breeder blanket, although these are expected to remain relatively constant once equilibrium is reached after the start-up of the plant. Finally, the radioactive decay of tritium needs to be considered for the total inventory of the plant.

To make sense of these measurements, analogous to practise with fissile materials in fission power plants, it needs to be defined what constitutes a significant amount of tritium to be diverted, and during what timeframe [6]. These accountancy activities need to occur with enough precision and high enough frequency to detect any tritium losses that exceed these limits. A useful and widely used tool for this purpose is the introduction of Mass Balance Areas (MBAs); areas where the quantity of tritium transported into or out of is determined, and of which the total tritium inventory can be measured when required.

Safety

As there is no location yet for DEMO, local safety agencies cannot be consulted regarding their requirements and regulatory processes. However, the two key goals will be, firstly, the prevention and mitigation of accidents, and secondly the monitoring of radioactive material releases and doses to workers and the public.

The former can be achieved through appropriate plant design, including application of safety case methodologies, and limiting the tritium inventories in subsystems. These measures will reduce the source terms of tritium in an accident scenario. Radioactive releases and worker doses can be achieved using in-plant radiation monitoring such as ionisation chambers for tritium and gamma and alpha detectors for other radiation releases, personal dosimeters and bioassay sampling of workers. Other measures, such as limiting the airborne tritium inventories in each room and a zoning system for ventilation aid the achievement of both goals [6].

Environment

Similar for safety agencies, it is not known yet which local environment agency needs to be consulted regarding tritium emissions from a DEMO plant. These emissions will constitute gaseous discharges of T₂, HT and DT, liquid discharges of HTO and organically bound tritium compounds, and solid waste containing tritium. Other discharges that need to be monitored for radiological reasons are discharges of activated plasma-enhancement gases and activated products from sputtering and corrosion, especially tungsten dust from plasma-facing components.

Gaseous discharges can be detected using real time ionisation chambers located in the stack (stack monitors), although the accuracy of these instruments is typically not considered sufficient for quantitative monitoring. Instead, ionisation chambers provide an indicative measurement and are linked to recording, plant interlock and alarm systems. For quantitative monitoring, emissions can be further monitored using tritium absorbing gel packs which are routinely replaced and measured using scintillation methods to give accurate emissions data. Vegetation patches can also be used to monitor local absorption of tritium by crops. Sampling and scintillation analysis of all liquid outlets of the plant can be undertaken, with samples being taken to an on-site or off-site laboratory for scintillation analysis. Solid waste can similarly be sampled and surveyed before being taken away for controlled disposal in appropriate facilities.

Process monitoring and control

Process control is required for operating the plant safely and efficiently, and to ensure that gas mixtures are of the right compositions. It can be assumed that the process control data is the information provided to the control room, to serve as indicators as to whether processes are occurring as normal, and all equipment is functioning correctly, or whether there is a fault somewhere in the plant. Further, the plasma composition needs to be controlled to ensure the DT fusion occurs at its optimum for power output. This will depend on the correct functioning of isotope separation systems and fuelling systems, such as pellet and gas injection.

3. Measurement conditions

Considering all these requirements leads to a large collection of measurement locations in the DEMO fuel cycle, that are each characterised by a set of measurement conditions. To find a suitable tritium detection technology for every location, these measurement conditions need to be categorised first.

This is a challenging task as there are many different flow rates, temperatures, matter phases, tritium concentrations and external factors such as magnetic fields and radiation in the DEMO fuel cycle. Further, some of these flow rates are still unknown. Another important factor is whether the measurement is on a subsystem that works in batch mode, or on a continuous flow. In the case of continuous flows between subsystems, the tritium detection must be continuous, either on-line or in-line, in real time. Although most measurements will fall in this category, there will be some subsystems where static measurements, for example calorimetry, can take place, such as storage systems.

To overcome these difficulties, all system blocks in the fuel cycle have been examined, considering the in- and outflow of all gases and liquids, while also eliminating duplicate mass flows. This results in 46 different connections between system blocks. The conditions occurring at each connection have been categorised using Table 1 [7].

Table 1: Measurement conditions categorisation

	Categories				
Flow rate (gas, Pa m³ s⁻¹)*	0 – 10	10 – 100		100 – 1000	> 1000
Flow rate (liquid, kg s⁻¹)	0 – 10	10 – 100		100 – 1000	> 1000
Phase	Gas		Liquid		Solid
Temperature	Cryogenic (20 K)	Cold (20-298 K)	Room (298 K)	Warm (298-373 K)	Hot (>373 K)
Tritium concentration	High (90-100%)		Medium (10-90%)		Low (<10%)
Presence of ionising radiation or magnetic fields	Yes			No	
Operational mode	Static			Continuous	

The conditions occurring at some connections may fall in the same category, while there are also combinations of conditions that do not occur at all. As a result, there are 17 different sets of

* These are calculated at 273.15 K

conditions identified. For each of these conditions, the most suitable tritium detection technology needs to be found.

4. Detection technologies

Previous work [8], [9] studying the suitability of tritium detection techniques at DEMO fuel cycle relevant measurement conditions has produced a short list of 7 detection technologies that will be discussed briefly.

BIXS

Beta-Induced X-ray Spectroscopy (BIXS) only depends upon the beta particles produced from a source. Hence the method can be used to analyse tritium in a wide range of chemical states. BIXS can be used for water samples, although with a lower sensitivity than for gas, and on solids. One advantage of using BIXS is that it can be used under dynamic conditions in liquids [10], as opposed to e.g. liquid scintillators. However, BIXS is not commercially available and suffers from memory effects [11].

Scintillation counter

Liquid scintillation counting (LSC), which uses a scintillation cocktail that is mixed with the tritiated liquid, is commonly used due to its low limit of detection but can only be used in batch mode. Solid scintillators, such as fibres, can be used for real time monitoring of tritiated liquids [12]. Gaseous scintillation detection systems are not commercially available but have been tested for on-line static measurements [13]. However, this was more than 25 years ago and has not been followed up since.

Raman spectroscopy

Raman spectroscopy is a method based on the inelastic Raman scattering of laser light off a sample, which can be in gaseous, liquid or solid form. Raman spectroscopy can distinguish between all six hydrogen isotopologues due to the relatively large mass differences, making it suitable for qualitative measurements, and can be calibrated for taking quantitative measurements. However, it is unsuitable for monatomic species such as helium. Measurement times are typically of the order of seconds [14] making it suitable for continuous measurements. However, to increase the accuracy usually the average of several spectra is taken, increasing the measurement time. Raman spectroscopy is commercially available, although developments are needed for gas phase detectors and to ensure tritium compatibility.

Mass spectrometry

Mass spectrometers are generally used for quasi-static gaseous measurements, where a small amount of gas is sampled by breaking the particles up into ions and analysing their mass/charge ratio. Mass spectrometry has the potential to measure gas compositions up to pure tritium with no memory effect and there is a high confidence that this technique can be used to measure continuously in fusion relevant conditions with good reliability. Recent developments have made it possible to sample gas streams up to 30 bar and 200 °C. Mass spectrometry measurements can be combined with other detectors such as flow meters to provide information in terms of g/m^3 or mol/m^3 .

Further development is needed towards the calibration of fraction patterns containing tritium of, for example, CQ₄, and to address the issue of line overlap, which is a primary concern for hydrogen isotopologues as their fragments occupy a very narrow mass range with very small mass differences between some species.

Gas Chromatography

Gas chromatography is a separation technology, and needs to be combined with another technology to act as detector. For example, using knowledge of characteristic retention times and monitoring physical properties of the elution allows for identification of the eluent.

Micro-gas chromatographs work faster, with a retention time of several minutes, which is not quick enough yet for continuous measurements [15]. Moreover, due to the use of a carrier gas this method cannot be used in a continuous manner as this gas cannot re-enter the gas stream.

Calorimetry

Calorimetry is a tritium detection method that is commonly used for solid storage systems, as measurements can typically take hours. By measuring the temperature increase in the sample due to the radioactive decay the total amount of tritium can be determined. As a result, it needs to be combined with pressure-volume-temperature calculations to determine the concentration of tritium in a mixture. Calorimetry measurements generally need to be performed in tandem with a reference volume.

Ionisation Chambers

Ionisation chambers produce a small electric current with its magnitude a measure of the amount of radiation present. Ionisation chambers are very sensitive to memory effects, moisture, and gas pressure. Their use in tritium laboratories is common and widespread, for example for gas discharges and air monitoring, however, due to these limitations are usually only used as indicators and not for quantitative measurements.

Other technologies

A possible candidate technology for solid measurements such as on the first wall materials are Laser-Induced Breakdown Spectroscopy (LIBS) and Laser-Induced Ablation Spectroscopy [16], [17]. Here a high-energetic laser produces a plasma of particles released from the solid material, which can be analysed using spectroscopy. These technologies can be used for determining the thickness and composition of deposition layers on the first wall.

Gap analysis and recommendations

The properties of the measurement technologies discussed are summarised in Table 2, focussing on whether the technology can be applied in a continuous manner, and which state of matter can be detected.

Table 2: Overview of main conclusions regarding the detection technologies

Technology	Phase	Continuous	Measure for tritium concentration?
BIXS	Gas / liquid / solid	Yes	Yes
Scintillation counter	Liquid	Yes – development of scintillation fibres	Yes
Raman	Gas / liquid / solid	Yes	Yes
Mass spectrometry	Gas	Yes – in development	Yes – in development
Calorimetry	Solid	No	Yes
Gas Chromatography	Gas	No – microGC shorter retention times, but not enough for continuous	No
Ionisation chambers	Gas	Yes	Yes
LIBS / LIAS	Solid	No	Yes

Comparing the measurement conditions at which tritium detection is required with the available technologies, shows that, in general, the main difficulty lies in finding technologies that are suitable for in-line measurements, with real-time results. Raman spectroscopy is a very promising candidate for these conditions, having the additional advantage that it is suitable for all three phases of matter. BIXS, although having the same advantages, is probably of a technological readiness level that is too low for application in DEMO right now. However, DEMO is still many years away and it may be worth developing BIXS further as it could offer many applicabilities. Further, with developments towards higher resolutions, mass spectrometry could be exploited for composition analysis. Other technologies, such as calorimetry, gas chromatography, and ionisation chambers, are of a more mature readiness level and can be applied in those conditions that they are already being used for. However, modifications might be desired to widen their applicability, especially to continuous modes or higher flow rates.

Future experimental activities will be necessary to test the applicability of these technologies to tritium detection at various conditions. Currently at UKAEA, an experiment [18] is being designed with this aim. As gaseous conditions make up most conditions present in the DEMO fuel cycle, these conditions are prioritised in this rig. However, it is important to keep in mind that the detection of tritium in especially solids, and detection in the presence of ionising radiation or magnetic fields is at a lower technology readiness level and will need to develop further before being applicable to the DEMO fuel cycle.

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