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# Design Of An Impurities Detritiation System For ITER Using A Palladium Membrane Reactor

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## 1 INTRODUCTION

The ITER exhaust will contain tritiated impurities such as water and methane. In order to recover the tritium from these impurities, ITER will need an Impurities Detritiation System (IDS). Potential processes for the IDS include catalytic-reactor/palladium-membrane systems, isotopic-exchange/palladium-membrane systems, and ceramic-electrolysis/palladium-membrane systems. To achieve the low tritium emissions established in the ITER design requirements, it was determined that a 2-stage system consisting of one or more of the above processes would be required. A simpler, 1-stage IDS is presented in this paper. The system uses a Palladium Membrane Reactor (PMR) to recover about 98% of the hydrogen isotopes. Although this is not an adequate recovery to fulfill the design requirements, the PMR is followed by the Process Waste Detritiation System (PWD). The PWD is a conventional oxidation-reactor/molecular-sieve system that converts hydrogen isotopes into water and then stores the water on molecular sieve. In this design, the PWD is incorporated into the IDS. The design allows for tritiated water stored on the molecular sieve to be periodically recycled through the PMR so that the required tritium recoveries are achieved.

### 1.1 PMR Background

A process to recover tritium from simulated fusion fuels and tritiated water has been successfully demonstrated at the Tritium Systems Test Assembly at Los Alamos National Laboratory. The ITER exhaust will contain tritiated impurities such as water and methane. Tritium will need to be recovered from these impurities for environmental and economic reasons. The PMR is a combined permeator and catalytic reactor. Catalysts are used to foster reactions such as water-gas shift,

$$Q_2O + CO \rightarrow Q_2 + CO_2, \tag{1}$$

and methane steam reforming,

$$CQ_4 + Q_2O \rightarrow 3Q_2 + CO \tag{2}$$

where Q represents the hydrogen isotopes H, D, and T. Due to thermodynamic limitations these reactions only proceed to partial completion. Thus, a Pd/Ag membrane, which is exclusively permeable to hydrogen isotopes, is incorporated into the reactor. By maintaining a vacuum on the permeate side of the membrane, product hydrogen isotopes are removed, enabling the reactions to proceed toward completion. For the water-processing application, only HTO and CO are injected into the PMR and it might be expected that only reaction (1) would be of importance. However, near the inlet of the PMR, some $CQ_4$ is formed by the reverse of reaction (2). Therefore,

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performance of the PMR system at water-processing conditions is similar to that of torus-exhaust conditions.

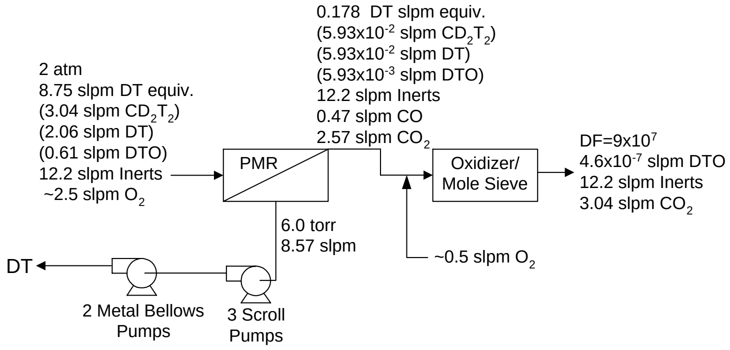

Figure 1. Schematic Representation of the Impurities Detritiation System

Results of palladium-membrane reactor experiments have been reported in previous papers. Simulated fusion fuels were processed with a PMR [1], but these early experiments contained no tritium. References [2 and 3] reported on tritium experiments with a single-stage PMR system and reference [4] reported on tritium experiments with a two-stage PMR system. The experiments were conducted at ITER relevant conditions and were found to have a 1st stage decontamination factor (DF=inlet hydrogen isotopes/outlet hydrogen isotopes) in the 150-400 range and up to $3x10^6$ for the 2nd stage alone. Reference [5] reported on a two-stage system for tritiated-water processing. Performance was similar to that of the fusion-fuel processing experiments. Reference [6] reported on methods of optimizing and controlling PMR performance.

## 2 SYSTEM DESIGN

A schematic representation of the IDS is shown in Figure 1. The inlet to the system comes from the outlet of the Front-End Permeator System. The inlet rates are specified in the ITER design requirements as 3.04 slpm $CD_2T_2$, 2.06 slpm DT, 0.61 slpm DTO, and 12.2 slpm inerts (mostly He). Approximately 2.5 slpm $O_2$ is added before injection to ensure high performance.[1] 8.57 slpm DT is produced in the permeate and sent to either the storage or isotope separation systems. 98% of the hydrogen isotopes are recovered in the permeate. The remaining 2% are mixed with approximately 0.5 slpm $O_2$ before being injected into an oxidation-reactor/molecular-sieve system for storage of DTO on molecular sieve. The IDS decontamination factor is $9x10^7$, or a recovery of 99.9999989% of the hydrogen isotopes. DTO is periodically driven off the molecular sieve by heating and recycled back through the PMR to recover the DT.

Details of the PMR and oxidation-reactor/molecular-sieve components are discussed in the following sections. Also, a process and instrumentation diagram will be available in the ITER Detailed Design Document. The IDS is estimated to have an installed cost of $1.4M (without glove box). Details of the cost estimation will be available in the ITER Cost Report.

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[1] Addition of approximately 2.5 slpm $O_2$ will result in optimal performance at the inlet conditions shown. At other inlet conditions, different $O_2$ injection rates or CO may be required. Good performance of the PMR is not dependent on precise control of these additives. Extensive testing has shown that good performance occurs over a wide range of carbon-to-oxygen ratios, and the control of this ratio is not difficult [6]. Control can probably be monitored with an outlet moisture measurement, but the additional measurement of CO or $CO_2$ could also be required at times.

## 2.1 PMR Design

The PMR component shown in Figure 1 actually consists of 2 PMRs, 2 metal bellows pumps, and 3 scroll pumps. During torus exhaust processing, flow enters the system from the FEP. If the ratio of $CD_2T_2$ to DTO is not within the range required for good performance, then either $O_2$ or CO is added to this stream. The flow then enters the 2 PMRs (in parallel), where 98% of the DT is separated and sent to either the isotope separation or the fuel storage system. Each PMR is loaded with $Pt/\alpha\text{-}Al_2O_3$ catalyst (Engelhard A-16825) and has about the same Pd/Ag membrane area as permeators commonly used at the USDOE facilities Savannah River Site and Los Alamos National Laboratory (~0.25 $m^2$). These permeators are small (~0.3 m in diameter and 0.6 m in length), however, the PMRs will be somewhat larger due to the use of 0.635 cm diameter Pd/Ag tubes rather than the 0.318 cm tubes used in the permeators. With this permeator area, the PMRs are over-designed so that in the event of a PMR failure, the IDS would still achieve the ITER design requirements.

A vacuum is applied to the PMR permeate with 3 Normetex PV-12 scroll pumps backed by 2 Metal Bellows MB-601 pumps. If a scroll pump failure occurs, PMR performance will decrease to about 97% recovery of the DT (0.29 slpm DT equivalent in the retentate stream). This would put an increased load on the molecular sieve beds, but the ITER design requirements would still be achieved. If a metal bellows pump failure occurs, a similar small decrease in performance would occur.

## 2.2 Oxidation-Reactor/Molecular-Sieve Design

Organic and molecular hydrogen are oxidized to water in the reactor. The reactor was designed using $Pt/\alpha\text{-}Al_2O_3$ catalyst (Engelhard A-16825), which has been used with good performance and lifetime in tritium facilities in the US. Reactor dimensions of 1 m in length and 0.02 $m^2$ in area at 650 °C can, conservatively, process the PMR effluent with conversions (inlet/outlet) in excess of $10^8$. This will convert essentially all of the organic and molecular hydrogen in the PMR effluent into DTO. During off-normal performance of the FEP or the PMR, much higher flow rates at much higher $CD_2T_2$ and DT concentrations than the design values (Figure 1) can be processed.

The molecular sieve beds are designed to each have 22.7 kg of 4A molecular sieve. The 1st stage has 2 beds plumbed in parallel, only 1 of which is on-line at any given time. After 2.9 days of torus exhaust processing (continuous burn), the inventory of the on-line bed will reach 99 g of tritium, which is the inventory limit for any given component in ITER. At this point, the other (unloaded) 1st stage bed is put on-line. The tritiated water stored on the loaded bed is then processed through the IDS system to recover the tritium. Processing is accomplished by heating the bed and adding the appropriate amount of CO. Two operating scenarios are possible for bed regeneration: during torus exhaust processing or during times of no torus exhaust processing.

During torus exhaust processing, 0.875 slpm of DTO (10% of IDS design capacity in equivalent DT flow) can be processed without impacting the performance of the IDS. 14 hrs. are required to regenerate a bed at this rate. During times of no torus exhaust processing, the bed can be regenerated at 8.75 slpm DTO (100% of IDS design capacity in equivalent DT flow). 1.4 hrs. are required to regenerate a bed at this rate. Tritiated water processing with a PMR system has been demonstrated in extensive testing at Los Alamos.

The performance of the 2nd stage bed is dependent on the operating cycle of the 1st stage beds because more water breaks through the 1st stage beds as the water inventory increases. If each 1st stage bed is allowed to be fully loaded to 99 g of tritium before regeneration, then the 2nd stage bed will have a lifetime of 883 days of continuous burn. At the end of 883 days, the 2nd stage will have a breakthrough of 1 Ci/day at continuous burn. Initially, the breakthrough will be close to 0 Ci/day since residual water on the molecular sieve will isotopically exchange with the tritiated water. After 200 days of continuous burn, the breakthrough will be 0.23 Ci/day. However, these breakthroughs can be significantly reduced by not allowing the 1st stage beds to be fully loaded to 99 g of tritium before regeneration. The 2nd stage bed will contain 5.0 g of tritium when it fully loaded. This bed can also be regenerated through the PMR if necessary.

In the event of a failure of a 1st stage bed, the other 1st stage bed would be put on-line. In the event of a failure of the 2nd stage bed, 11.2 Ci/day (continuous burn) would breakthrough to the VDS when the bed was dry and 112 Ci/day would breakthrough if the bed was fully loaded.

## 3 PMR OPERATING EXPERIENCE

PMR technology has been extensively demonstrated at Los Alamos. A glove box system has been used to process simulated ITER torus-exhaust gases in a 32-hr. test. Also, 2250 std. L of tritiated water were regenerated from a molecular sieve bed and processed through the PMR system in 172 hrs. of testing. This demonstration resulted in the recovery of approximately 4 g of tritium and, essentially, complete regeneration of the bed. About 30 start-up/shut-down cycles were performed in the course of these demonstrations with high performance and reliability. In addition, a non-tritium PMR system has been used for experimental purposes. This system has accumulated 215 days of round-the-clock operation over an 18-month period, while also recording high performance and reliability.

## 4 CONCLUSIONS

A relatively simple, 1-stage Impurities Detritiation System has been designed for ITER. The system also simplifies the Tritium Plant design by incorporating the Process Waste Detritiation System into the IDS while achieving the design requirements of both systems. The IDS consists of a palladium membrane reactor followed by an oxidation-reactor/molecular sieve bed. 98% of the hydrogen isotopes are recovered in the PMR and sent to the isotope separation or storage systems. The remaining 2% of hydrogen isotopes are temporarily stored on molecular sieve. The molecular sieve beds are periodically regenerated back through the PMR to recover the hydrogen isotopes.

The IDS has a robust design that allows for individual PMR, pump, or molecular sieve failure while still achieving the design requirements. PMR technology has been demonstrated with 204 hrs. of tritium operation and 215 days of non-tritium operation at high performance and reliability. The system is estimated to have an installed cost of $1.4M (without glove box).

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