

Tritium handling and processing experience at the Tritium Systems Test Assembly

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Abstract

In 1987 the Japan Atomic Energy Research Institute (JAERI) and the US Department of Energy (DOE) signed a collaborative agreement for the joint funding and operation of the Tritium Systems Test Assembly (TSTA) at the Los Alamos National Laboratory. This collaboration has continued through June 1994. During this seven year period the TSTA team has conducted many tritium experiments. These included a number of integrated process loop tests, as well as many off-line, stand-alone experiments to develop, test and validate new components and technologies for tritium service. The TSTA process loop has been operated in continuous mode for periods up to 25 consecutive days, and many loop experiments of shorter duration have been performed. In addition, a number of technical developments and component/system demonstrations have been accomplished. These tests have included development and evaluating of fuel cleanup systems and hydrogen isotope separation systems that are now available for consideration for ITER. This paper highlights some of the major accomplishments of this seven year collaboration.

1. Introduction

In 1987, the Japan Atomic Energy Research Institute (JAERI) and the US Department of Energy (DOE) signed a collaborative agreement (Annex IV) for the joint funding and operation of the Tritium Systems Test Assembly (TSTA) at the Los Alamos National Laboratory (LANL) for a 5 year period ending June 1992. After this initial 5 year collaboration, the Annex IV agreement was extended for another 2 year period ending June 1994.

During the first 5 years, a number of the integrated process loop tests of TSTA were conducted, as well as off-line testing of TSTA subsystems. During integrated loop testing the vacuum system, fuel cleanup systems, isotope separation systems, transfer pumping system

and gas analysis system were interconnected and tested using 100 g inventories of tritium to demonstrate steady-state operation of a tritium fuel processing cycle for a fusion reactor. These tests have resulted in a number of significant accomplishments and an experience database on research, development and operation of the fuel processing system. One of the most significant accomplishments during the initial 5 year period was the continuous operation of the fuel processing loop for 25 days [1]. During this 25 day extended operation, both the JAERI fuel cleanup system (JFCU) and the original TSTA fuel cleanup system (FCU) were operated under similar conditions of flow, pressure, and impurity content of DT gas. Both fuel cleanup systems were demonstrated to provide adequate impurity removal for plasma exhaust gas processing. The isotope

separation system was operated continuously, producing pure tritium while rejecting protium as an impurity.

During this collaboration, a new laser Raman spectroscopy system for in situ analysis of the gas in the TSTA loop was developed. Successful development of the laser Raman spectroscopy enabled us to study non-steady behavior of the isotope separation system and the integrated loop. During the extended 2 year period, preliminary testing for a blanket tritium recovery system was performed using the existing TSTA systems, including the JFCU. The experience on non-steady-state operation of TSTA, obtained during the 2 year extension, provides a valuable database for operation of tritium processing systems for practical, pulsed tokamaks such as ITER. Additional studies accomplished during the 7 year collaboration include studies on tritium interactions with gaseous materials such as CO, studies on tritium getter materials such as ZrCo and La–Ni–Mn alloys, and studies on tritium interaction with cryosorption pump materials. Through the collaborative experiments described above TSTA has also accumulated an impressive record of tritium safe handling.

2. Recent results

During the 2 year extension of the collaborative agreement at TSTA the programmatic emphasis has been focused on two areas: (1) operating the TSTA loop under conditions simulating the non-steady-state conditions expected for an operating tokamak; (2) operation of TSTA subsystems under conditions typical of those expected in processing product gas from a tritium breeding blanket.

Large tokamak fusion devices, such as ITER, will operate as pulsed machines, albeit with long pulse lengths. However, even for nominally steady-state machines, transient operations for ramp-up, shut-down, discharge cleaning or conditioning and normal operations must all be handled by the fuel processing system. TSTA has begun a series of tests, with the integrated fuel processing loop [2] to study the response of the fuel processing system to these changing scenarios. Operation of the fuel processing loop with tritium inventories of up to 100 g and nominal fuel processing rates of up to 390 g mol day⁻¹ (tritium processing rates of 1–1.2 kg day⁻¹) have been conducted with variations in flow rate and composition that mimic changes expected for practical pulsed DT torus operations.

A major issue for operating tokamaks will be the recovery of tritium from a breeding blanket. During the

7 years of this collaboration the project has investigated how the stream containing tritium recovered from the blanket will interface with the plasma exhaust gas processing system. Within the frame of the breeding blanket interface (BBI) study [3], ways of using the existing equipment and subsystems at TSTA to simulate and evaluate this interface were examined. Several experiments at TSTA were performed using combinations of the components of the fuel processing loop to demonstrate the recovery of tritium from a purge gas stream simulating tritium recovery from a solid ceramic breeding blanket.

2.1. Isotope separation system

In order to meet the above objectives, major modifications had to be made to the isotope separation system (ISS) at TSTA. This new ISS configuration more closely resembles the proposed isotope separation system for ITER.

The modifications to the ISS encompassed removing all of the original intercolumn connecting piping, flow controllers, valves, etc. and replacing this highly tritium contaminated piping with new and upgraded components. New flow controllers, valves, piping and instrumentation were installed. Also, the first column of the TSTA four-column cascade has 84 theoretical stages, with three alternate feed-injection points, and seven sample withdrawal locations. This column piping was modified to permit a stream of hydrogen isotopes to be withdrawn from any of the sample taps through a flow control valve, equilibrated over a precious metal catalyst bed at ambient temperature, and pumped back into any other sample tap (sidestream recycle). Fig. 1 shows the column layout schematically. These major modifications to a tritium system that had processed more than 10⁹ Ci tritium required developing new procedures and technologies. This included the use of a Cajon™ orbital welder system for welding all of the new piping and components to the existing, tritium contaminated system. The use of the orbital welder permitted these modifications to occur in very crowded spaces, without any worker uptake of tritium, and without any significant release of tritium to the room or environment. Extensive pumping and purging of the system before performing any line breaks contributed significantly to the safe accomplishment of these modifications.

Experimental results obtained with the modified ISS, including the sidestream recycle, demonstrate a dramatic effect on the product streams. The sidestream recycle results in higher purity products with fewer columns in the cascade and, therefore, with lower tri-

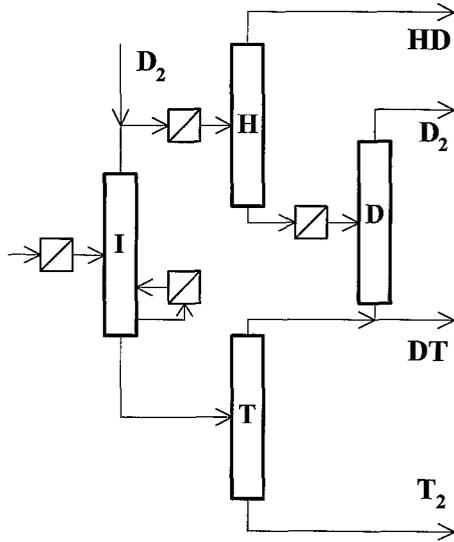


Fig. 1. Flow schematic diagram for ISS showing sidestream recycling on column 1.

tium inventories in the system. These results are significant confirmation of this technique as a means of reducing tritium inventory in fusion fuel processing loops.

Other changes that have been applied to the ISS during this period include the addition of more sophisticated control loops. Previously the ISS had been operated in “open loop”, i.e. all manual control. This required semi-continuous, focused attention to the ISS by an expert or extremely well trained and experienced operators. The new control loops were added to maintain material balanced in the column by controlling (1) liquid levels in all four columns, (2) using reboiler heat input to control column ΔP on columns I and H, and (3) controlling the ratio of feed flow to top flow for columns I, D and T. The effectiveness of the new control system was demonstrated in a recent operation of the TSTA fuel processing loop. Using a nominal 50:50 mixture of DT with impurities of nitrogen, oxygen, helium and hydrogen in the mixture, and a nominal flow rate of 6 l min^{-1} in the loop, it was demonstrated that the ISS could be started up, within a matter of hours, with minimal operator intercession. This demonstration test used all liquid level and ratio controls on the four columns. Once stable operation was achieved loop flow was upset, both up and down by 33%. That is the 6 l min^{-1} flow was lowered to about 4 l min^{-1} and equilibrium re-established, and later increased to about 8 l min^{-1} and equilibrium again re-established. The liquid levels in all columns remained very

stable at the selected set-point. This test was important in establishing the ability of a fuel processing loop to respond, unobtrusively, to rather significant changes in input conditions. An operating tokamak, such as ITER, surely will be subject to this type of large swing in fuel processing inputs. More detailed discussions of these recent results are presented by Sherman et al. [4,5].

2.2. Fuel cleanup system

Recent activities at TSTA have emphasized specific studies on both the original TSTA fuel cleanup system (FCU) and the JAERI fuel cleanup system (JFCU), but with no major modifications to either system. Fig. 2 shows a flow schematic for the two fuel cleanup systems. The FCU has been used for tests on the practical-scale testing of cryogenic molecular sieve beds for separating low-concentration hydrogen isotopes from helium [6] as part of the ongoing BBI studies [3]. Here, the emphasis was on tritium recovery from a solid ceramic breeding blanket using a helium purge gas, containing about 1% hydrogen, to recover the tritium. This gas mixture (about 99% He and 1% protium) can be purged through the solid blanket where the hydro-

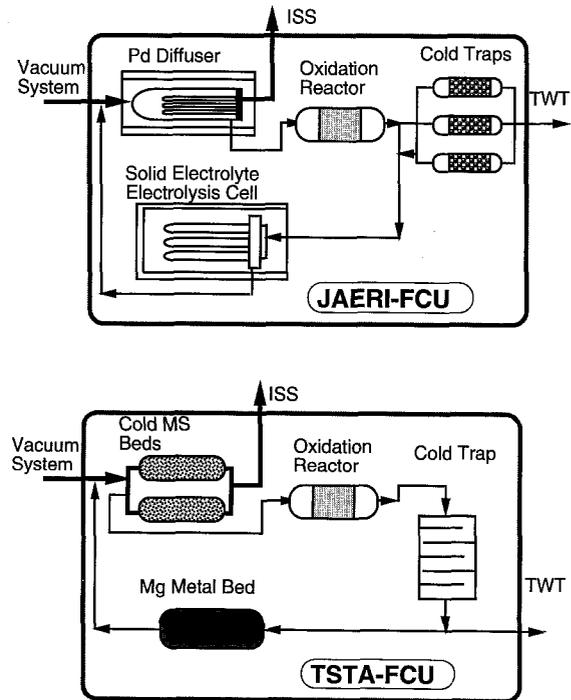


Fig. 2. Flow schematic diagrams for the JAERI fuel cleanup system and the original TSTA fuel cleanup system.

gen exchanges with the tritium. The resulting gas (about 99% He, about % hydrogen and about 0.01% tritium) flows out of the blanket and, for further processing, requires separation of the hydrogen isotopes and the helium.

For these tests the existing cryogenic molecular sieve beds in the FCU were used. These beds each contain about 1.6 kg of Linde 5A molecular sieve. Gas was fed to the cryogenic molecular sieve bed at about 13 standard $l\ min^{-1}$ with a nominal composition of 99% helium, 0.98% hydrogen and 0.02% HT. These tests showed that the cryogenic molecular sieve beds are effective in removing low concentration hydrogen (tritium) from a helium stream. When the beds became saturated with hydrogen, the analytical system (Raman spectroscopy) showed a very sharp breakthrough curve. These measurements were performed during a TSTA process loop experiment designed to demonstrate the use of the cryogenic molecular sieve as a means of separating and recovering tritium from a solid ceramic breeding blanket. The cryogenic molecular sieve studies are described in more detail by Willms et al. [6] in a companion paper.

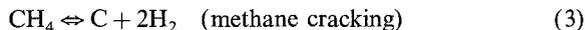
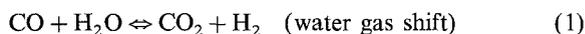
In the BBI tests, the JFCU was used for recovery of tritium when regenerating the cryogenic molecular sieve beds (CMSB) in the FCU. The regenerated gas from the CMSB, containing helium and hydrogen isotopes, was recycled through the palladium diffuser in the JFCU. High purity HT gas was recovered here and sent to the isotope separation system to separate tritium from the hydrogen. In a separate test, moisture, which is anticipated to be the dominant tritiated impurity in the helium purge, was decomposed in the ceramic electrolysis cell and the released hydrogen was recovered by the diffuser. This series of tests demonstrated that the components of the TSTA processing loop, that is the FCU, JFCU and the ISS, provide an excellent test bed for demonstrating the interfacing or a breeding blanket tritium recovery stream with the plasma exhaust gas processing loop.

The JFCU has been used during all TSTA loop tests during the past 2 years. A new “batch” mode operation of JFCU was tested for impurities processing [2]. Previously, within the JFCU all impurities containing hydrogen isotopes were oxidized and the resulting water was collected on cold traps for later processing via electrolysis, while tritium-free impurities were discharged continuously for steady operation. For the most recent tests the water resulting from oxidation was continuously fed, in the gas phase, to the electrolysis cell. The resulting hydrogen isotopes are fed to the ISS input and the tritium-free impurities are sent intermittently to the

tritium waste treatment (TWT) system for final processing and release to the plant vent. In the most recent test, ca. 0.3% of tritiated methane in the processing loop was decomposed without the introduction of additional oxygen for oxidation. These tests demonstrated that impurities can be processed without condensing water, thus resulting in operation with lower tritium inventory and reduced risk of tritiated water release. This batch mode of operation is acceptable for gas streams containing small quantities of impurities.

2.3. Non-loop experiments

Palladium membrane reactor studies. A major study at TSTA during the past two years has been the development of the palladium membrane reactor (PMR) concept [7] for fuel cleanup. This investigation, led by R.S. Willms, uses a combined catalytic reactor and palladium membrane permeator. In this combination a catalyst is used to promote various reactions leading to the release of hydrogen isotopes from the impurity molecules such as water, methane and other hydrocarbons. Reactions such as water gas shift, steam reforming and methane cracking,



can be carried out over the reactor catalyst, and the product hydrogen can be removed simultaneously from the mixture by permeating through the palladium membrane. Because the reaction product is removed continuously, conversions greater than calculated from thermodynamic equilibrium can be obtained. In addition, ultrapure hydrogen is produced, eliminating the need for an additional processing step between this component and the isotope separation system. PMR has been built and tested at TSTA. The results show that a nickel catalyst, operating at about 600 °C is very effective at promoting all three reactions listed above. The fact that the PMR operates better at this temperature than at lower temperatures indicates that the permeation of hydrogen isotopes through the palladium membrane is the controlling step. The water gas shift reaction is more efficient at lower temperatures, but the permeation of hydrogen through palladium increases with temperature. These studies have shown that the PMR is a potentially important tool for fusion fuel processing.

Laser Raman spectroscopy. Laser Raman spectroscopy has been developed as an on-line analytical tool at

TSTA [8]. This device is used primarily to perform rapid, real-time analysis of hydrogen isotope gas streams in the TSTA loop. For instance, the composition profiles in the four columns of the ISS are measured by pulling a small flow of gas from each analytical tap in turn along the length of the column. This gas flows through a scattering cell where the Raman scattering of the laser light provides unequivocal analysis of the hydrogen isotope ratios. Each analysis is the average of 100, 1 s scans, while the interval to switch to the next sample is about 3 min. Alternatively, consecutive samples can be taken at the same location, thus providing rapid information on composition changes at that location with a temporal resolution limited by flow to the Raman cell (less than 1 min). These analyses provide dynamic data that can be compared with the predicted response of the system. These data thus provide valuable validation of the dynamic code used to design new isotope separation systems.

The Raman system has also been used to perform radiochemical studies of tritium with various molecules [9]. A recent study has been the tritium plus sulfur hexafluoride system as SF_6 is used as an insulating gas in neutral beam systems that provide auxiliary heating to tokamaks. If an accident were to result in mixing SF_6 with tritium in the tokamak, what reaction products would be produced, at what rates, and would they poison the precious metal catalyst in the tritium cleanup systems? In a Raman experiment with a nearly equipolar mixture of these two gases, at a total pressure of 825 Torr, TF remained undetectable in the gas mixture and was thus less than 2% of the initial tritium after 6 days. However, new Raman lines believed to be from SF_4 gradually appeared during these 6 days; pending calibration of the SF_4 Raman spectrum, the final SF_4 concentration was estimated to be several per cent. Although the complete reaction mechanism has not been determined from this experiment, the relatively slow reaction rate is encouraging for fusion technology, as any accident would result in lower partial pressures of reactants and would be contained in a time much shorter than several days. In a companion study Talcott et al. [10] showed that the presence of SF_6 , at concentrations of 35–100 parts per million, does not result in deterioration of the catalytic ability of the precious metal catalyst of the type used in normal cleanup and air detritiation systems in tritium facilities.

In the latest work, the study of the interaction of tritium with carbon monoxide has been initiated with experiments conducted jointly both at TSTA and the Tritium Processing Laboratory (TPL) in Japan. In the initial studies of the CO-T_2 system a visible, solid

product developed. This particulate matter deposited onto the surfaces of the Raman cell, and showed an unidentified broad Raman peak. It is believed that some type of polymerization has occurred, but the product has not yet been identified. Further investigation of this system is planned.

Oil-free vacuum pump. A large, oil-free, reciprocating pump has been developed by JAERI and has been installed at TSTA for tritium testing. The pump has been installed in such a manner that it can be used as a circulating pump in loop experiments, or can be used in stand-alone experiments. The initial experiments measured pumping speed and evacuation performance of the pump with hydrogen, deuterium, helium, nitrogen, tritium and mixtures of these gases at discharge pressures of 400, 500, 760 and 875 Torr. The pumping speed for pure tritium was measured at more than $120 \text{ m}^3 \text{ h}^{-1}$ at 4 Torr suction pressure. The ultimate pressure obtained was 0.8 Torr.

This pump subsequently was used to circulate tritium semicontinuously for some 3 months to determine pump compatibility and reliability in tritium service. The pump performance did not degrade significantly through this long-term testing. The pump was demonstrated to be an acceptable circulation pump by incorporating it into the TSTA fuel processing loop, and demonstrating that the pump can be used for tritium service in fusion tritium applications.

Tritium storage beds. The use of metal or intermetallic materials for tritium storage has long been an accepted practice in tritium facilities. At TSTA uranium beds have been used to store the entire tritium inventory (up to 150 g tritium) during shut-down periods. As part of the US–Japan collaboration at TSTA some other storage materials have been investigated. The use of zirconium cobalt (ZrCo) and lanthanum–nickel–manganese (La–Ni–Mn) alloys for tritium storage has been studied. Isotherms for these systems with tritium have been measured. These materials have been shown to be acceptable storage media, with properties that are fairly similar to those of uranium. Fig. 3 shows the measured dissociation pressure of tritium over ZrCo as a function of temperature. A ZrCo bed with a storage capacity up to ca. 700 l tritium (about 180 g) has been installed and used at TSTA. It has not, however, been used to store these very large quantities of tritium. The performance of this bed has been entirely satisfactory with no unexpected behavior observed.

Recently a self-assaying, portable uranium storage bed has been constructed and tested at TSTA [11]. This

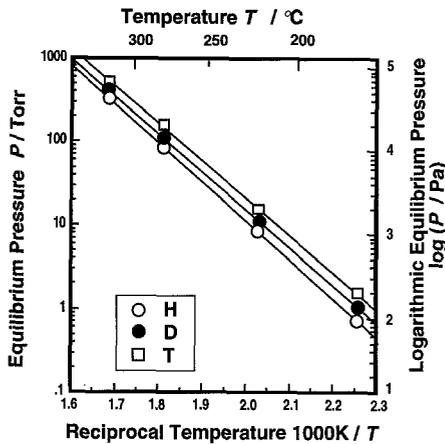


Fig. 3. Dissociation pressure as function of temperature for hydrogen isotopes over zirconium cobalt.

bed contains 400 g uranium and has a maximum tritium capacity of about 15 g. The bed has been designed to store tritium, but also to measure the heat of decay of the stored tritium, thus giving a measure of tritium inventory by measuring a rise in temperature. The bed is insulated within a highly evacuated secondary enclosure which provides containment in the event of a tritium leak from the uranium bed. The high thermal resistance between the bed and the secondary enclosure surfaces, which are at room temperature, cause any heat generation within the uranium bed to result in a large, easily measurable and repeatable temperature rise. The design of this bed is such that a 1 g tritium inventory will result in a temperature rise of about 15 °C. The temperature rise of the bed (compared with the temperature of the vacuum jacket) can be sensed to less than 0.1 degree celsius, which results in an ability to detect tritium down to 100 Ci. Several tritium loadings of this bed have been performed, demonstrating the ability of this bed to assay the stored tritium. Design is now underway for a large self-assaying storage bed, capable of storing up to 200 g of tritium.

Molecular sieve regeneration system. Molecular sieve regeneration system (MSRS) is being added to the existing TWT system within TSTA [12]. This MSRS will provide more accurate tritium measurement of the liquid waste generated within the TWT. Within the TWT, hydrogen isotopes in all waste gas streams are removed from the effluent gas stream by the catalytic conversion to water and subsequent removal of the water by adsorption on fixed molecular sieve beds. The quantity of tritium in the water is calculated from the measured

concentration of tritium in the gas stream going into the catalytic reactor. The remaining gas stream, now free of tritium, is then released to the plant vent.

Periodically it is necessary to regenerate these molecular sieve beds, transferring the water to disposable molecular sieve beds for long-term storage of this tritiated water. The MSRS will capture this water as it is driven off the fixed bed. The tritium content of this trapped water is then measured by both calorimetry (measuring the heat of decay of the contained tritium) and by collecting a small sample of this water for liquid scintillation counting. After these measurements are complete, the water is transferred to a disposable molecular sieve bed. This direct measurement of the tritium will improve the tritium accounting and inventory control within the TWT.

3. Summary

During the 7 years of the TSTA–JAERI collaboration this project has made a significant contribution to the development and understanding of fusion related tritium technology. The TSTA loop has been operated in continuous mode for long periods (up to 25 consecutive days), and many loop experiments of shorter duration (1–2 weeks continuous operation) have been performed.

More recent loop tests have been devoted to studying the operation of the fuel processing loop under non-steady conditions, thus more closely simulating the operation of a fuel processing system interfaced to an operating tokamak. Operating tokamaks are subject to many different operating scenarios such as ramp-up, long-pulse operation, shut-down and discharge cleaning and conditioning. The exhaust gas coming to the fuel processing loop under these conditions varies widely in flow rate and composition. The TSTA loop has been demonstrated to respond quickly and smoothly to rapid changes in input flow rate and to gas composition.

Another operating scenario that has been studied is the processing of a fluid stream that simulates the recovery of tritium from a breeding blanket. This work, identified as the breeding blanket interface studies at TSTA, has demonstrated the technology required to recover low concentrations of tritium from a helium stream. This stream simulates the purge gas stream postulated for recovery of tritium from a solid ceramic blanket.

In addition to these loop operations, a number of technical developments and components or system demonstrations have been accomplished. These non-

loop experiments and testing have led to development of a number of components and systems that will be included in the design of future, fusion related tritium facilities. A brief summary of some of these key developments has been presented in this paper.

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