Decontamination Studies of Japan Atomic Energy Research Institute Fuel Cleanup System and Glovebox Installed at the Tritium Systems Test Assembly

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ABSTRACT

The JAERI Fuel Cleanup System, a major fusion fuel processing system that has been used for four years with DT and impurity mixtures, has been decontaminated and decommissioned at the Tritium Systems Test Assembly. The system is composed of several processing components that were exposed to either DT gas or tritiated water vapor during operations. The entire system was contained in a large glovebox. Decontamination was first performed by purging and circulating various gases such as He, H_2 and room air through the system. The effectiveness of the decontamination methods are compared. Residual contamination of the system materials such as metals, catalysts and molecular sieves were measured and will be presented here. After gas purging, the plumbing and components were disassembled and packaged for ultimate disposal according to the procedures and regulations for the TSTA, LANL, and DOE. The waste packages will be buried at the LANL waste disposal site.

INTRODUCTION

The JAERI Fuel Cleanup System (JFCU) was a tritium fuel processing system capable which handled ITER quantities of DT fuel and fuel impurities. The JFCU was designed to process 15 mol per hour of a Q_2 mixture (Q represents any mixture of H, D and T) containing up to 15 percent impurities. The glovebox was installed in the Tritium Systems Test Assembly (TSTA) in 1989, and operated from 1990 to 1994 under various ITER relevant fuel processing conditions. The process utilized a palladium diffuser for DT gas stream purification, a catalyst bed for conversion of impurities to water, and a ceramic electrolysis cell for cracking water and to recover hydrogen isotopes. The process utilized zirconium cobalt beds for Q_2 storage and scavenging. Cold traps combined with small molecular sieve beds were used to collect water. A combination of metal bellows pumps, a turbomolecular pump, and a scroll pump were utilized for process pumping. The process components of the JFCU were exposed to primarily Q_2 gas or Q_2O water vapor in experimental operations which typically lasted a week. Tritium isotopic concentrations were approximately 30-50%. After each run, tritium and tritiated water were removed by the ZrCo bed and the electrolysis cell, respectively. The system was back filled with inert gas until the next experiment. After completion of the test program in 1994, the JFCU was decontaminated and disassembled for disposal.

This paper presents the results of the decontamination and disposal (D&D) of the JFCU process systems, tritium holdup studies performed on materials in the process, and lessons learned. Internal decontamination of components was conducted utilizing various purge gases and residual contamination levels were evaluated by water soaking and high heat decontamination at 923 K.

Tritium holdup on materials used in a fuel processing system is an important issue for safety as well as design. The data on tritium holdup in stainless steel piping, copper pellets from a cold trap, oxidation catalyst, and molecular sieve, taken from the JFCU are reported.

COMPONENT DECONTAMINATION

A. Procedure

Prior to the start of the JFCU decontamination, all process components were shutdown in the normal operational fashion. This removed all readily recoverable quantities of tritium by evacuation.

Internal decontamination of the JFCU process components and associated tubing, transducers and valves was performed by a series of three-stage steps—1) fill the subject components with a purge gas; 2) circulate the purge gas through the components for 8 hr/day, leaving the gas to soak the components for 16 hr/day; and 3) evacuate the purge gas, measuring its tritium content. These three stages were repeated until the amount of tritium removed was negligible. Purge gases used were helium, hydrogen, carbon monoxide, ozone, and air. Components were decontaminated at both room temperature and at their heated operating temperature.

The evacuated purge gases were sent to the TSTA Tritium Waste Treatment system (TWT). The TWT utilizes an ion chamber and integrating program which allowed the total quantity of tritium from each D&D evacuation to be determined. The ion chamber did not distinguish the chemical form of the tritium sent to the TWT system. However, samples of some of the evacuations were collected and analyzed using a mass spectrometer.

B. *JCR1 and PD*

For this decontamination the circulation loop consisted of the Catalytic Reactor (JCR1) and the Palladium Diffuser (PD). When heated these two components were held at temperatures of 773 K for JCR1 and 673 K for the PD. This decontamination occurred over a period of 22 days and a total of 6310 Ci of tritium were removed.

The amount of tritium removed during each of the fill/circulate/evacuate operations is shown in Fig. 1. Shown on the x-axis for each of these steps are 1) the purge gas used, 2) whether or not the components were heated (H) or Cold (C, i.e. ambient temperature), and 3) the circulation/soak time (days). Each of these were found to be important factors affecting the degree of tritium release.

As shown on Fig. 1, decontamination began by using He as the purge gas in the first two tests. Heating the components was found to dramatically increase the quantity of tritium removed from 13 to 520 Ci. The next six tests used H_2 for the purge gas. This further increased the amount of tritium recovered to 1770 Ci in test 3 using a one day-soak and even further to 3271 Ci by using a three-day soak. After this it appears that the amount of tritium in the system has been substantially reduced as the amount of tritium removed per test dropped off dramatically with the last H_2 purge removing only 2 Ci. Thereafter four more tests were conducted using either CO or air as the purge gas. Only small amounts of tritium were removed in these last tests.

As can be seen by comparing test 2 (He) and test 3 (H_2), purging with inert gases such as helium was found to be less effective at decontamination compared to hydrogen purging. Hydrogen is believed to be a more effective tritium removal purge gas because it chemically exchanges with tritium-containing species on surfaces. However, it must be remembered that cleanup operations using hydrogen generally create more waste water than inert gas purging.
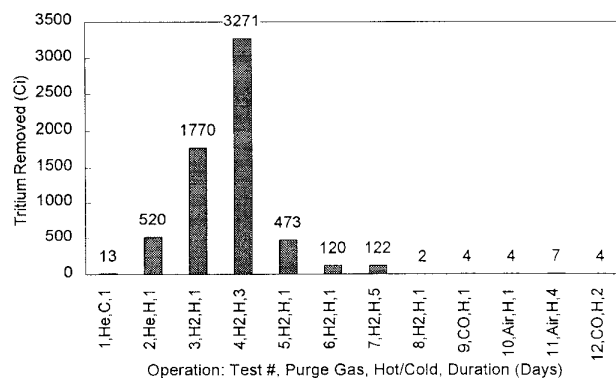

Fig. 1 Decontamination of JCR1 and PD

It can also be observed on Fig. 1 by comparing, for instance, test 3 (one day soak) and test 4 (three day soak) that gases recover more tritium per cycle with longer soak times. If H_2 is used as the purge gas, using less H_2 will reduce the amount of tritiated water waste collected in the TWT (all hydrogen and hydrogen-containing species are oxidized to water and collected on molecular sieve).

Mass spectrometer analysis was performed on samples which were collected at the conclusion of some of the decontamination steps. These results are summarized in Table 1. Shown are all of the tritiated species which were detected by the magnetic sector mass spectrometer. From these data it appears that carbon monoxide may have had beneficial decontamination effects especially after the introduction of room air which contains water vapor. This is observed in test #12 which showed an increase in T_2O released compared with levels in test #10. This may indicate that a water gas shift reaction took place in the catalyst bed. However, the amount of tritium removed by this specific operation was trivial compared to the amount of tritium still retained in the catalyst as determined in subsequent analysis.

Table 1
Mass Spectrometer Analysis of JCR1 and PD Purge
Gases (mole %)

Sample	DT	T_2	T_2O
post test #3 H_2	1.531	0.734	0
post test #8 H_2	0.086	0.094	0
post test #9 CO	0.032	0.018	0.025
post test #10 AIR	0.038	0.011	0.029
post test #12 CO	0	0	0.049

C. *Cold Traps*

The decontamination data for the circulation loop containing the three cold traps and their associated molecular sieve beds are shown in Fig. 2. This loop was decontaminated over a 25 day period during which a total of 2257 Ci of tritium were recovered. Overall, as observed in the JCR1/PD decontamination, relatively large amounts of tritium were removed initially followed by very little tritium recovered in the last tests. In this case, the largest single tritium removal test was the first one which removed 1600 Ci using a He purge gas. This was followed by a He purge which removed only 192 Ci. Then a H_2 purge was performed and the tritium removal was increased to 289 Ci. Two more H_2 purges removed reduced, but substantial amounts of tritium. The campaign was completed with four air purges with ever decreasing amounts of tritium removed per cycle. The last air test used a soak time of 2 days with the components heated followed by 10 days of soaking with the components cold (ambient temperature).

For this campaign it was quite apparent from test 13 that He was quite effective for at least the initial removal of tritium from the system. By comparing this series of tests to the

JCR1/PD tests, it is observed that H_2 is not so dramatically better than He for removing tritium. This may have been due to residual, tritiated humidity being present on cold trap surfaces which the He removed by a simple drying process. However, while not so dramatic as in the previous tests, H_2 was again shown to be more effective than He at recovering tritium. This was concluded by observing the increase in the amount of tritium in test 15 (H_2 purge) over that in test 14 (He purge). Again this is believed to be due to H_2's ability to chemically exchange with tritiated species.
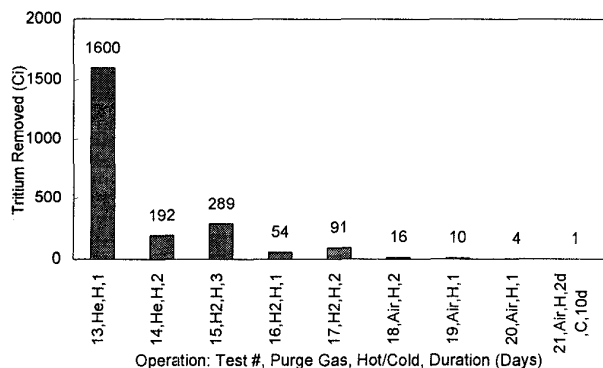

Fig. 2 Decontamination of Cold Traps and Molecular Sieve Beds

D. *CEC*

The ceramic electrolysis cell decontamination began with a quick flush using dried nitrogen with the CEC cold. This removed 122 Ci of tritium. Thereafter the CEC was heated and purged with room air over a 9 day period. The first air flush removed 79 Ci of tritium which was less than the initial N_2 flush. The subsequent air flushes removed ever decreasing amounts of tritium. A total of 285 Ci of tritium was removed in the CEC decontamination. The results of this series of tests are shown on Fig. 3.

The cell was expected to be contaminated mostly by tritiated water. The ceramic cell, which did not have a large surface area, cleaned up quickly, with a relatively small amount of tritium needing to be removed.
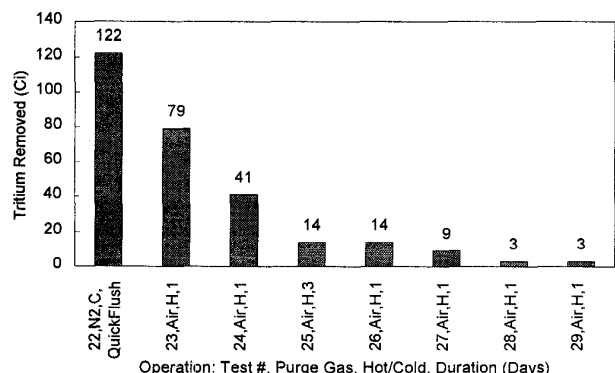

Fig. 3 Decontamination of Ceramic Electrolysis Cell

In total, these three series of tests removed 8852 Ci of internal contamination from the JFCU components.

E. *Ozone treatment*

Subsequently, ozone was used in an attempt to remove further internal contamination. After exposure to air, an ozone generator was used to produce air containing about 3.5% ozone which was pumped through the catalyst, cold trap and associated piping. Each test was metered to insure that 50 liters of air, or air with 3.5% ozone, was placed into the components. The tests were done with components in the heated and unheated condition. Test runs were each of the same duration.

Results of the decontamination of the cold traps at room temperature indicates that nearly twice as much tritium was liberated with the ozone present, 0.513 Ci vs. 0.284 Ci. While these levels are low, the fact that the ozone treatment followed immediately after the air run does indicate some effect. A run with components heated (773 K for the catalyst bed and 343 K for the cold traps) yielded 1.310 Ci with air and only 0.702 Ci with air/ozone. This lack of ozone effectiveness could be a result of the rapid ozone breakdown over heated components. From these tests the overall benefit of ozone decontamination is not conclusive.

At the conclusion of the component internal decontamination the glovebox atmosphere which contained them was in the 10 to 30 mCi/m^3 range.

RESIDUAL TRITIUM MEASUREMENTS

A. *Procedures*

At the end of the internal decontamination work an effort was made to characterize the amount of residual tritium remain on the various materials. Representative samples of the following material were collected: copper pellets from the cold traps, 1% Pt-doped alumina (Engelhard) catalyst, molecular sieve from the beds associated with the cold traps, and stainless steel tubing. Each of these were soaked in water with several dilutions of the water necessary to assess tritium holdup using a scintillation counter. The copper pellets, catalyst, and piping were then heated to 923 K and the liberated tritium was measured.

In addition, smears of the inner surfaces of process piping and components were made at various locations in the system to ascertain removable tritium holdup after decontamination.

B. *Cold trap copper pellets*

Five copper pellets from one of the cold traps were removed and observed to be heavily oxidized as indicated by a grayish

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black appearance. These pellets were individually soaked in water, and the water was replaced with fresh water after 24, 48 and 72 hours. Each contaminated water sample was analyzed and the results are recorded in Table II. Thereafter, all five pellets were heated together to 923 K and the liberated tritium was measured. This result is also given in Table II. A grand total of 4619 μCi of additional tritium was recovered in these steps. This translates into an average of 920 μCi removed from each pellet or 570 μCi removed per gram of copper. How much tritium remained in the copper even after this procedure is not know, but it is believed that the value is small.

Table II
Tritium Holdup in Cold Trap Copper Pellets

Pellet	24 Hours Water Soak (μCi)	48 Hours Water Soak (μCi)	72 Hours Water Soak (μCi)	Heated to 923 K (μCi)
1	50	55	120	
2	42	48	58	
3	53	57	83	
4	37	44	64	
5	34	37	48	
Total	216	241	373	3789

C. Molecular sieve

After its decontamination (heating to 343 K), a sample of the molecular sieve was removed from one of the beds associated with one of the cold traps. The sieve was noted to still be white and solid with no appearance of deterioration. Two of the cylindrical sieve pellets were separately soaked in water for 24 hours with the water subsequently analyzed. The pellets yielded 1.21 and 0.78 mCi. The difference between the two numbers may be due to the location of the sieve pellet within the trap. Using these measurements it can be estimated that the tritium holdup was nearly 100 mCi per gram of sieve material.

D. Catalyst

A single pellet of catalyst was recovered from catalytic reactor JCR1 which had been decontaminated as summarized on Fig. 1 (heating was to 773 K). The Engelhard 1% Pt-alumina was silver gray and appeared unchanged from its unused form. Upon soaking in water the pellet yielded 67 μCi after 24 hours with identical results at 48 hours. This translates to a holdup of approximately 6.7 mCi per gram of catalyst. Tritium holdup in the pellet after water soaking is unknown at this time.

E. Smear tests of selected tubing

At the conclusion of the in-place decontaminations described above, swipes were taken inside tubing at the entrance and exit of components. Swipes were done with a dry cotton swab, 0.6 cm in length, passing around the inside of the

tubing, making two complete revolutions on each sample. The primary form of tritium to which each tubing section had been exposed was either Q_2O or Q_2. All the tubing samples were 1.25 cm OD, type 316 stainless steel. The swipe area was estimated to be 1.7 cm^2.

The amount of tritium on each cotton swab was analyzed and the results are summarized in Table III. The tubing that carried Q_2 swiped, on average, 64% higher than the Q_2O tube samples. The swipeable tritium holdup per square centimeter of tube is used to categorize material for waste disposal.

Table III
Results of Swipes Inside Tubing

Sample Location	Sample Exposure Type	Surface Contamination μCi	Specific Surface Contamination μCi per cm^2
outlet of cold trap CT3	Q_2O	8.8	5.2
inlet of cold trap CT3	Q_2O	25.4	14.9
inlet to catalyst bed	Q_2	31.5	18.5
outlet to catalyst bed	Q_2O	11.7	6.9
inlet to PD diffuser	Q_2	30.2	17.8
outlet to PD diffuser	Q_2	44.4	26.1
inlet to electrolysis cell CEC	Q_2O	14.2	8.4
outlet to electrolysis cell CEC	Q_2	17.1	10.0
inlet to zirconium cobalt bed ZCB1	Q_2	16.3	9.6
outlet to zirconium cobalt bed ZCB1	Q_2	8.6	5.1

F. Water soaking and heat treating of selected tubing samples

To better assess the tritium holdup in stainless steel tubing, a sample of tubing from the outlet of a cold trap (CT3) and a sample of tubing from the outlet of the zirconium cobalt bed (ZCB1) were soaked in water for 24 hours and then heated to 923 K until there was no evidence of further tritium release. The tubing samples were 1.25 cm long. The amount of tritium liberated in each of these two steps is recorded in Table IV. These steps liberated 20% more tritium from the Q_2-exposed tubing (ZCB1) compared to the Q_2O-exposed tube sample (CT3). This technique removed much more tritium than did the swipes because the soaking and heating removed tritium from more than just the surface and with greater efficiency.

Table IV
Tritium Recovered from Water Soaking and Heat Treatment of Selected Tubing

Tubing Sample Location	24 h water soak (μCi)	Heating at 923 K (μCi)	Total specific T_2 recovered (μCi/cm^2)
Outlet CT3	114	101	126
Outlet ZCB1	181	76	151

WASTE PACKAGING

The waste acceptance criteria at Los Alamos National Laboratory requires that medium level waste packages not exceed 10 Ci/m^3 total activity. This criteria dictated that the catalyst bed (JCR1) and the zirconium cobalt bed (ZCB1) be packaged separately from the glovebox. With these removed along with certain other components which were potentially reusable, the entire glovebox was placed in a strong tight metal container for burial. The container met DOT Type A shipping requirements and was required to be leak tight at 10 psi above atmospheric. The total activity of the glovebox and its contents packaged within this container was estimated to be under 150 Ci.

The zirconium cobalt bed and the catalyst bed were required to be overpacked in polyethylene drums with heat welded plastic covers. No attempt was made to assay the tritium hold up in the zirconium cobalt bed as this would have required compromising a double walled stainless steel container to draw samples. The holdup on the zirconium cobalt bed was roughly estimated to be 1000 Ci +/-500 Ci, based on experience with depleted uranium storage beds.

PERSONNEL EXPOSURES

During these D&D activities exposures to personnel and releases to the environment were tracked to ascertain the overall hazard associated with this type of work which included the study of components, the removal and handling of samples, and the packaging of decontaminated materials. This resulted in small increases in personnel exposures above the normal exposures at TSTA. On average, personnel associated with these tasks received a total additional exposure of 1 to 3 mrem. Most of this additional exposure is related to absorption or inhalation of glovebox atmosphere during open glovebox operations to remove samples or salvage items. During window off operations care was taken to avoid moving the trunk and face of the body past the glovebox plane and into the box itself. Supplied air was not required for these operations. Portable ventilation ducts were utilized to limit exposures, and a radiation technician was present to monitor all potential exposure situations. All tritium operations were reviewed on separate work instructions or covered under a radiation work permit.

STACK RELEASES

These D&D activities resulted in four Ci total released to the TSTA stack above normal operations. This number was kept low by utilizing the Portable Water Removal (PWR) unit [1] each time the glovebox was opened. The PWR is a blower with a molecular sieve cartridge on the outlet which is exhausted to the stack. Two of the units were used at the same time due to the relatively large volume of the JFCU. A total of 12 Ci was captured on molecular sieve in the PWR's during three separate open glovebox operations.

LESSONS LEARNED

For D&D of process systems the original design of the glovebox systems should include multiple pathways to a waste treatment or vacuum system and recirculation paths, even if these paths are not necessary for the experiment itself. The ability to evacuate components before breaking process connections and to maintain partial vacuum within while working on them reduces exposures and releases.

Decontamination efforts should include multiple system flushes and, if possible, extended soak times when using hydrogen or moist gas. A highly contaminated system may require 20 to 30 system flushes over several months before it can safely be handled outside of the glovebox. This should be balanced against the amount of waste generated by the decontamination effort itself.

Check valves in any piping system should not be relied upon to prevent the spread of contamination. Every check valve (eight total) in the JFCU failed to prevent contamination spread over the years of operation.

Welded systems should have adequate mechanical joints to insure that components can be removed without having to cut a welded pipe. Cutting welds and piping inside of the glovebox is difficult.

Do put magnetic tool holding strips or a designated tool storage area inside every glovebox. Incorporate a staging area or workplace into the glovebox design whenever space permits.

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