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DEVELOPMENT OF THE JAERI FUEL CLEANUP SYSTEM FOR TESTS AT THE TRITIUM SYSTEMS TEST ASSEMBLY

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ABSTRACT

Tritium Process Laboratory (TPL) at the Japan Atomic Energy Research Institute (JAERI) has developed the Fuel Cleanup System (FCU) which accepts simulated fusion reactor exhaust and produces pure hydrogen isotopes and tritium-free waste. The major components are; a palladium diffuser, a catalytic reactor, cold traps, a ceramic electrolysis cell, and zirconium-cobalt beds. In 1988, an integrated loop of the FCU process was installed in the TPL and a number of "hot" runs were performed to study the system characteristics and improve system performance. Under the US-Japan collaboration program, the "JAERI Fuel Cleanup System" (JFCU) was designed and fabricated by JAERI/TPL for testing at the Tritium Systems Test Assembly (TSTA) in Los Alamos National Laboratory as a major subsystem of the simulated fusion fuel cycle. The JFCU was installed in the TSTA in early 1990.

I. INTRODUCTION

At the Tritium Process Laboratory (TPL) in the Japan Atomic Energy Research Institute (JAERI), the research and development of the fusion reactor fuel processing systems have been conducted. The fuel cleanup system(FCU) processes plasma exhaust to provide pure hydrogen isotopes to the Isotope Separation System and exhausts impurity elements as a tritium-free stream. Major components were developed in JAERI and tested without tritium[1,2,3]. Tritium testing of the components was performed under the US-Japan collaboration program at the Tritium Systems Test Assembly(TSTA) in the Los Alamos National Laboratory(LANL)[4,5]. Through these tests, fundamental characteristics of the components, compatibility with tritium, effects of impurities, and long-term operability and reliability were investigated.

An integrated loop of the FCU process was installed in the TPL for testing with a large amount of tritium[6]. The objective of the experiments was; to test the tritium processing components, to study the interaction between the components and the behavior of the loop; to obtain the experience of the design; fabrication and operation the tritium processing loop, and to establish a fundamental engineering data for the fusion fuel cycle. The experimental apparatus has been operational since 1988. A number of tests were performed with simulated plasma exhausts.

Based on these results, the complete subsystem of the FCU was developed and designed by JAERI for full scale demonstration of plasma exhaust reprocessing. The system, called JFCU, was installed in the TSTA in early 1990 and preliminary tests were conducted. The JFCU will be tested with simulated fusion fuels in the TSTA main loop beginning in early 1991. The integrity of the process, interaction with other subsystems, reliability and controllability , and process response under normal and off-normal conditions will be investigated.

The present paper describes the process and recent results of the tests of the FCU at the TPL, and reports the features of the JFCU apparatus developed and installed in the TSTA under US-Japan Collaboration.

II. SYSTEM DESIGN

A. Design Consideration

The FCU is required to process simulated plasma exhaust that contains major Q_2, (Q stands for any mixture of HDT) and up to 15% of impurities such as helium, water, methane and ammonia[7]. The pure DT product that will be sent to the isotope separation should not contain more than 1 ppm of impurities to avoid plugging in the cryogenic system. The process should recover all the tritium, either free or bound to other elements as impurities, and should discharge impurity elements, such as carbon, oxygen, nitrogen or helium as an essentially tritium free exhaust gas. Figure 1 shows the simplified schematic of the FCU in the TPL. Major components are; the Palladium Diffusers, two Catalytic Reactors, three Cold Traps, an Electrolysis Cell , three Zirconium-Cobalt Beds, and circulation/vacuum pumps. The process was selected to have following features; high purity product, low tritium concentration in the exhaust; small tritium inventory, minimal switch-over components, no solid waste generation, and continuous operation.

B. Process Description

The Palladium Diffuser separates simulated plasma exhaust into a pure Q_2 product stream and a bleed stream where impermeable impurities are concentrated. The permeated side of the palladium membrane is evacuated to drive permeation and reduce Q_2 concentration in the diffuser exhaust, "bleed". Selective diffusion of hydrogen through a palladium membrane provides superior purity of the product stream over other techniques. This feature provides less possibility of plugging in the ISS cryogenic distillation system. Compatibility of the component with impurities such as methane, ammonia and CO; separation characteristics; and long-term reliability with tritium and helium-3, produced by decay of tritium in the metal, have been verified in previous studies[4,5].

The bleed gas from the diffuser is processed in the Catalytic Reactor - Cold Traps - Electrolysis Cell train. Hydrogen isotopes Q in the diatomic or molecular form (such as methane, and ammonia) is oxidized in the Catalytic Reactor. The Catalytic Reactor operates at 500 °C so that the conversion ratio of methane will be higher than 10^6. Oxygen required for oxidation of tritiated species is fed continuously to the Catalytic Reactor from the Electrolysis Cell and/or external sources.

The exhaust from the Catalytic Reactor passes through one of the Cold Traps to separate tritiated water and tritium-free species. The operation temperature of the trap is the lowest at which carbon dioxide in the stream is not frozen-out. The concentration of water vapor at the outlet is expected to be lower than 0.1 ppm. The exhaust of the trap, containing He, CO_2, N_2 O_2, and NOx is removed from the process. Traces of methane and water not removed in the loop will be the dominant source of tritium loss from the system. A part of the Cold Trap exhaust is recirculated by the Transfer Pump to the Catalytic Reactor inlet to dilute combustible species.

The Cold Traps are operated in a cycle of (1) trapping at 160K, (2) programmed regeneration by heat-up to 343K, and (3) precooling for the next trapping cycle. Three traps automatically switch-over every hour in accordance with the three steps in the cycle. Cold Traps are cooled by the freon refrigerator.

The catalytic oxidation-cold trap train utilizes the same process as the existing TSTA FCU. This process should have a high decontamination factor for tritium in the form of hydrogen, methane, ammonia, water and hydrocarbons in the exhaust stream . This process is expected to recover tritium bound in unknown chemical compounds, such as higher hydrocarbons.

The Cold Traps are periodically regenerated by externally supplied inert gas or by recycle of exhaust from the other trap. Tritiated water vapor from the heated Cold Trap is decomposed in the Ceramic Electrolysis Cell, where both free and bound oxygen are extracted from the feed stream containing tritiated water vapor. The Q_2 generated by the cracking of water is separated from the Cell exhaust by the Palladium Diffuser. Pure oxygen generated at the Electrolysis Cell is recirculated to the inlet of the Catalytic Reactor.

The Ceramic Electrolysis Cell (CEC) technique for decomposition of tritiated water

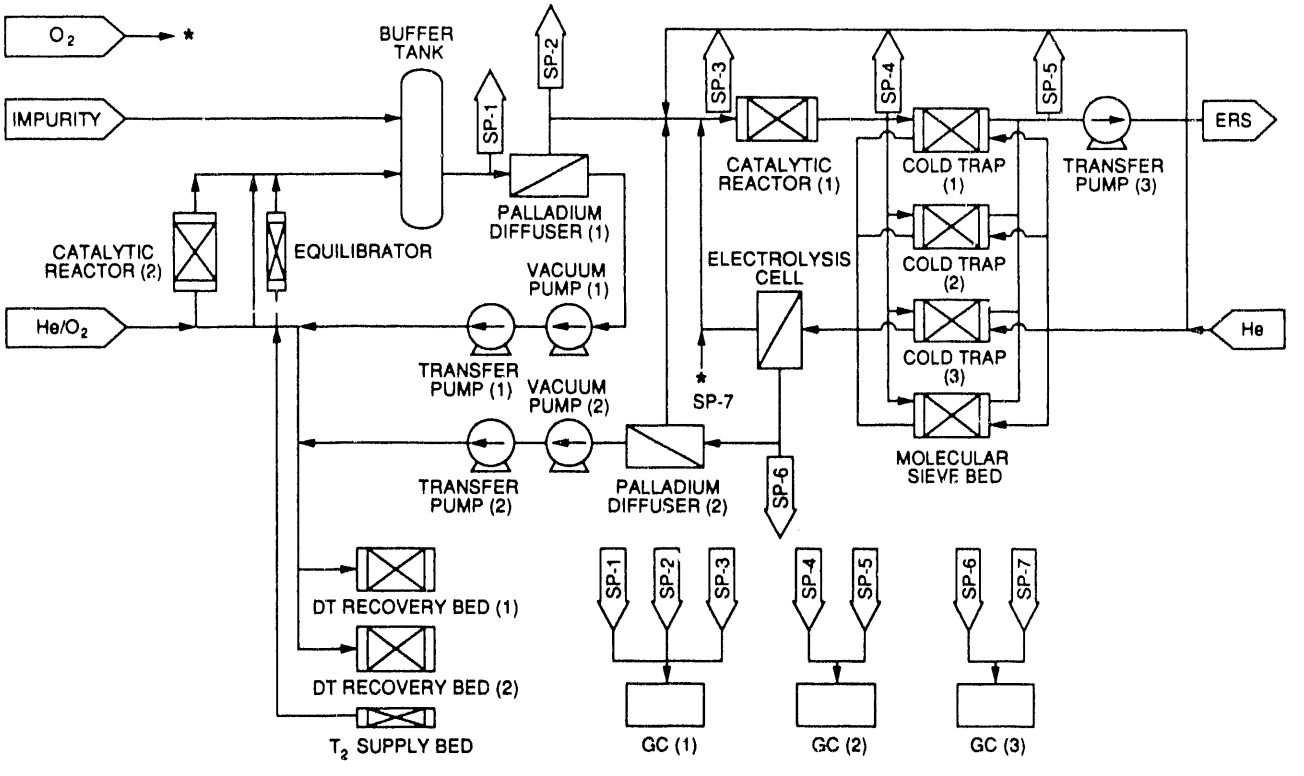

Fig.1 The simplified schematic of the FCU in the TPL.

has features superior to other options such as active hot metal beds, conventional wet cells, or catalytic shift reaction. The CEC generates no tritiated waste, has a minimum tritium inventory, and needs no further separation of oxygen exhaust from tritium.

A ZrCo bed supplies and recovers tritium in the experiments. Two ZrCo beds facilitate tritium recovery from gas chromatograph (GC) exhausts. The Zirconium-Cobalt intermetallic compound is an alternative to uranium. ZrCo has similar pressure-composition isotherms with hydrogen, it can be handled in air and is a non-nuclear material. These features were advantageous in fabrication and shipment of the apparatus.

Gas analysis is conducted by gas chromatographs(GCs) with Thermal Conductivity Detector (TCD) and ion chamber detectors. The GCs are installed in the glovebox, while controls are outside. Samples are taken from the process piping by vacuum pumping. Exhausts from sample pumping and carrier gas flow through small zirconium-cobalt beds for tritium recovery.

Detailed description of each component is written in a previous report[6].

III. RESULT OF THE TRITIUM TESTS OF TPL-FCU

Experiments with this apparatus have been performed since early 1988. One gram of tritium was introduced to the system in July, 1988. Full loop tritium runs for 3 to 6 days have been conducted several times to date. Performance of each component as well as the behavior of the integrated process were studied. Table 1 summarizes a typical operation condition of the FCU loop. Some modifications and improvements have been made based on the results.

Figure 2 shows the permeabilities of the pure hydrogen isotopes through the palladium diffuser 1 that has 48 of 600 mm long, respectively 1.6 mm and 1.44 mm in outer and inner diameters palladium alloy tubes. The diffuser is operated at 573 K. The permeation flow rates have linear relationship with the differential square root pressure across the palladium membrane. In the early tests in 1988, poor permeation was observed at the relatively low operation pressure. It was understood as an effect of the residual impurity gas in the diffuser and contamination on the membrane surface. After a few times of runs, permeation was restored. The ratio of permeabilities for H and T was approximately 2.3. This is in good agreement with our previous studies[4].

Control of the bleed flow from the diffuser operating with the Q_2-impurity mixture is shown in Figure 3. Pressure in the diffuser is controlled by the flow control valve at the outlet of the diffuser to obtain low concentration of tritium in the bleed. As seen

Table 1. Typical operating condition of the FCU at the TPL.

System	
Tritium Inventory	1~2 g, 20~70% T in Q_2
Impurity injection	N_2+CH_4, ~ 0.3 liter/min
Process exhaust	~0.3 liter/min
Palladium Diffuser	
Temperature	573K
Pressure	feed : 760 torr, permeated : 10 torr
Q_2 feed to Pd-1	0.5 ~ 2 liter/min
Bleed flow rate	~0.3 liter/min
Catalytic Reactor CR-1	
Temperature	773K
Flow rate	6.3 liter/min
O_2 addition	60 cc/min
Cold Traps CT-1~3	
Temperature	trapping : 160K regeneration : 343K switching interval : 1 hour
N_2 Carrier gas	regeneration : 2 liter/min
water throughput	~0.18 mol/h
Electrolysis Cell	
Temperature	870K
voltage	set potential : 1.3V cell voltage : 1.3 - 2.0V
N_2 Carrier gas	2 liter/min
water throughput	~0.18 mol/h

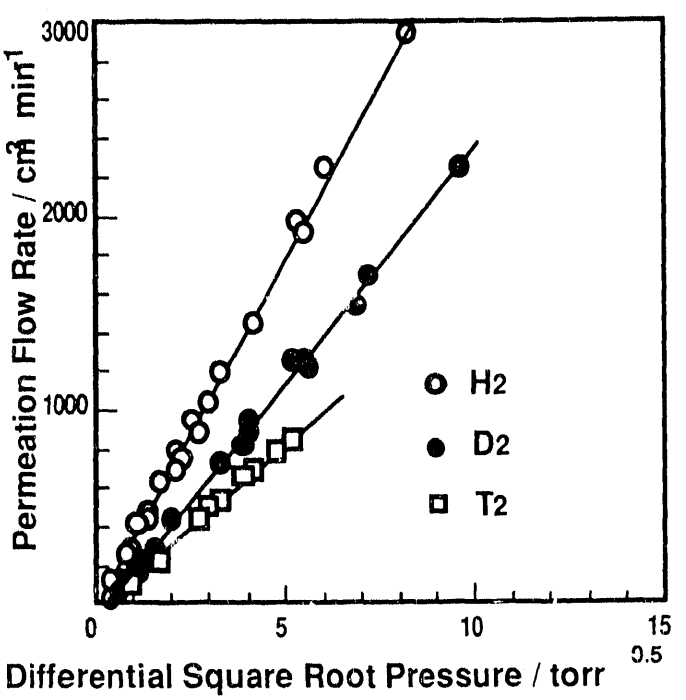

Fig.2 Permeabilities of the pure hydrogen isotopes through the palladium diffuser.

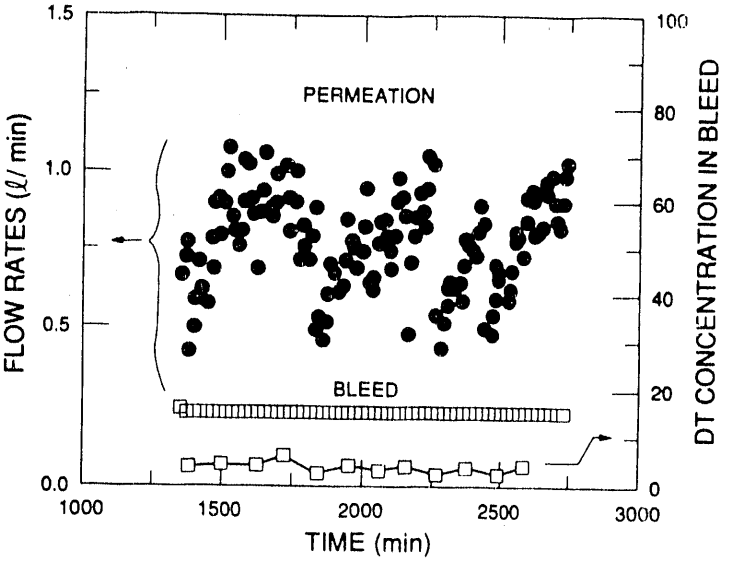

Fig.3 Control of the bleed flow from the diffuser in the operation with the Q_2-impurity mixture.

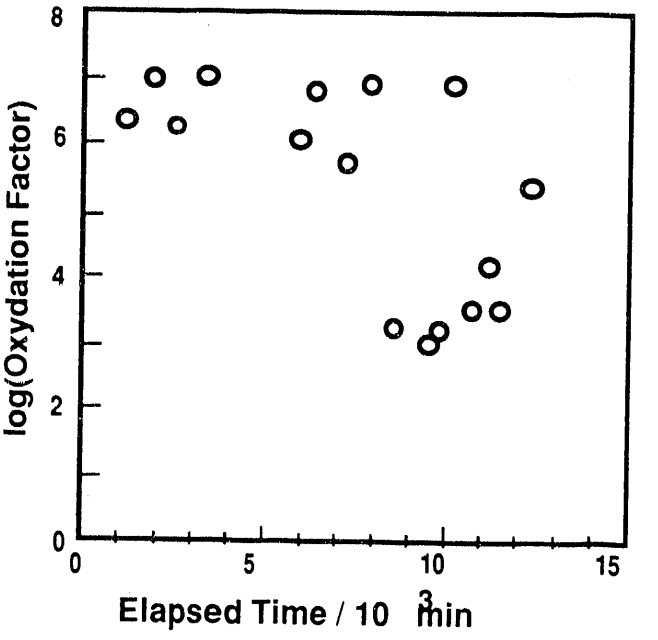

Fig.4 Change of the oxidation factor of hydrogen at the catalytic reactor.

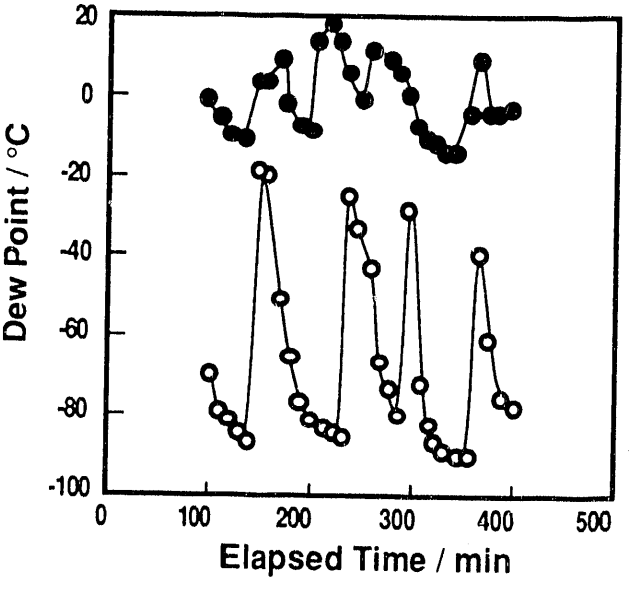

Fig.5 Change of the humidities at the inlet and the outlet of the cold traps.

in the figure, bleed flow rate was kept stable while flow rates of DT permeated through the diffuser changed. Change of permeation flow rate is due to the change of impurity concentration in the feed side of the palladium membrane. Hydrogen concentration in the bleed measured by the GC is also shown. Bleed flow rate and hydrogen concentration were stably controlled to be lower than 4% during the operation. More than 98% of the Q_2 fed to the diffuser permeated into the pure product stream.

Figure 4 shows the reduction in hydrogen concentration at the catalytic reactor. During stable operation, the ion chamber in the GC at the outlet of the reactor did not detect the tritium in Q_2 form and thus indicated that the hydrogen concentration was reduced by a factor of 10^7. Under some system upset conditions, oxidation factor dropped to 10^2, due to the lack of oxygen at the reactor.

Figure 5 shows the change of the humidities at the inlet and the outlet of the cold traps. Poor moisture trapping was observed when the traps were switched. After stabilizing pressure drop and flow rate across the traps the trapping efficiency improved to as high as 10^6 to 10^7, although moisture at the trap outlet is still suspected.

The CEC worked well in both decomposition of water and removal of oxygen in the feed stream. Oxygen flow across the cell and the decomposition rate of the water are shown in the Fig.6. Decomposition efficiency was higher than 90% .

As an integrated loop, the FCU successfully processed Q_2 with impurities such as N_2-CH_4 and N_2-Q_2O in the above tests. Both purification and recovery of Q_2 were performed continuously. After most of the mechanical or electrical problems found in early operations were corrected, the system has been operated with improved stability and performance. Approximately 3.3×10^6Ci of tritium, 250 mol Q_2, and 88 mol of impurities have been processed to date, and average loss of the tritium to the exhaust stream was 10^{-4} of the total throughput including system upset condition. Thus it is concluded that a technique to process plasma exhaust stably and safely was established.

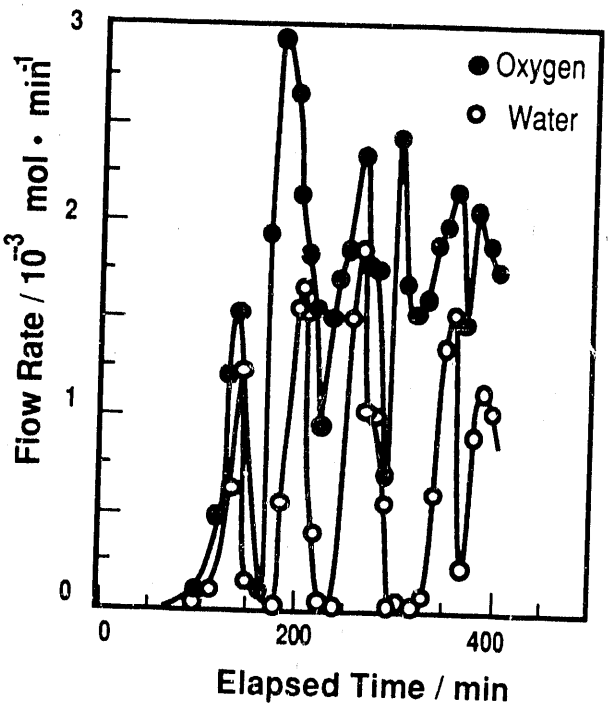

Fig.6 Oxygen flow across the cell and the decomposition rate of the water.

IV. DEVELOPMENT OF THE JFCU

A. Design of the JFCU

The JFCU is designed to be compatible with the existing TSTA loop and serve as one of the major TSTA systems. The function of the JFCU is; to process simulated plasma exhaust from the TSTA Transfer Pumping Unit (TPU), to supply pure hydrogen isotopes to the ISS, and to exhaust elements other than hydrogen to the Tritium Waste Treatment (TWT) system. It must meet all performance requirements of the existing TSTA FCU that has successfully operated in integrated loop runs[8]. Figure 7 shows the diagram of the JFCU process. Flow rates of the chemical species in the process is also shown. Inlet Q_2 flow is 15 mol/h, the same as the original design feed flow rate for the existing FCU, and 1/5 of the throughput for ITER. Major difference of this process from the FCU in TPL is that while the JFCU has only one Palladium diffuser, the TPL-FCU has a second diffuser for separation of Q_2 from the CEC product.

The JFCU system is operated from outside of the glovebox aided by a computer. All the physical variables are transmitted through CAMAC interfaces to the JFCU computer, that is subordinate to the Master Data Acquisition and Control computer (MDAC) of TSTA. The data is stored in the JFCU computer and sent to MDAC for monitoring and archiving. The JFCU computer locally operates the JFCU as a Front End Controller for the TSTA MDAC. It accepts a few macro commands from the MDAC.

B. Installation and Testing

The JFCU apparatus was fabricated by the Mitsubishi Heavy Industries and shipped to the TSTA in late 1989. Installation in the main cell of the TSTA followed by the initial checkout was completed in March, 1990. Technical requirements for operation at TSTA, such as leak rate, electrical insulation and other qualities were met. .Figure 8 is a photo of the JFCU in the TSTA. The apparatus is installed in a glovebox of ca. 5m x 1.2m x 2m. Component tests were begun in May 1990. Integrated process testing was conducted in June.Tests were performed with D_2, CH_4, NH_3 and He. All the components and their process functions such as, purification of D_2, oxidation at the catalytic reactor, trapping of water, decomposition of water, and analysis

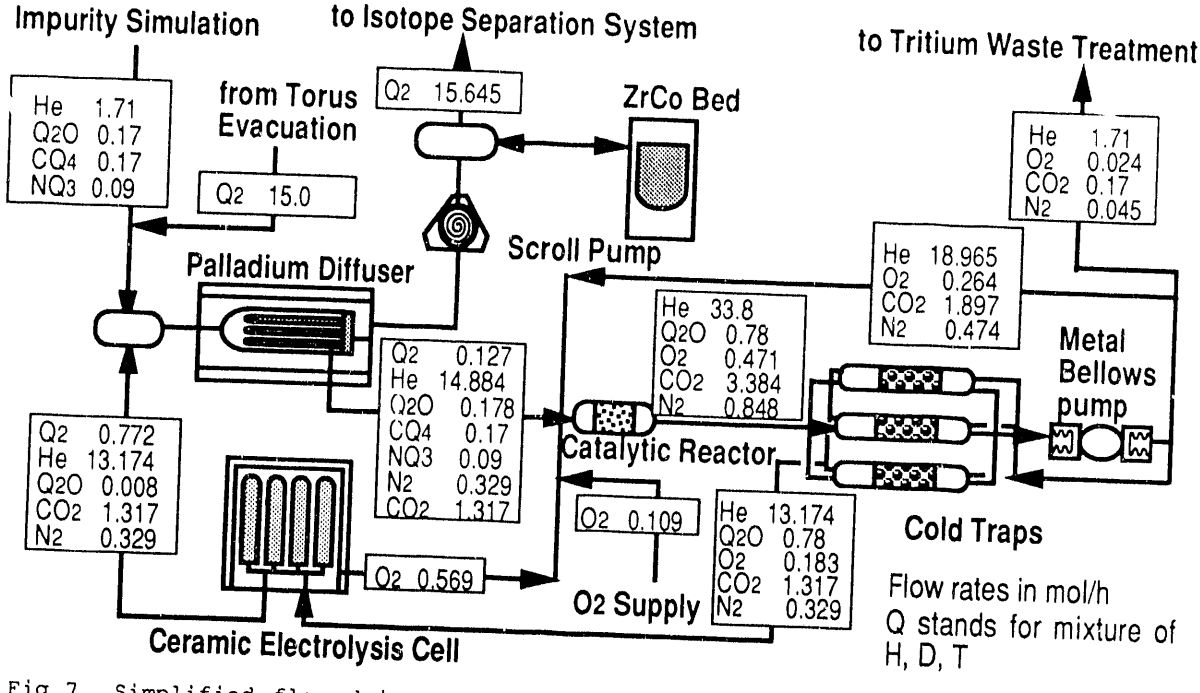

Fig.7 Simplified flow daigram of the JAERI Fuel Cleanup System. Flow rates of chemical species in the process is shown in mol/h.

with GCs were tested. The cold testing of the JFCU for optimization and modification of the system, better understanding of the characteristics, and the training of the personnel will continue through the end of 1990. Tritium testing is expected to begin in early 1991 after the approval by the DOE is obtained.

V. CONCLUSION

The JAERI Fuel Cleanup System(JFCU) was developed as a full-scale subsystem of the fusion fuel cycle to be tested in the TSTA under US-Japan collaboration program. This process utilizes a palladium diffuser and a ceramic electrolysis cell. It has some attractive features for fusion fuel processing. Studies under collaborative programs at TSTA have proved the feasibility and long-term reliability of the main components of this process with tritium. An integrated system has been tested successfully in the TPL in JAERI. The process will be studied with simulated fusion fuel in an integrated loop. The result is expected to demonstrate tritium technology needed for the next step fusion devices.

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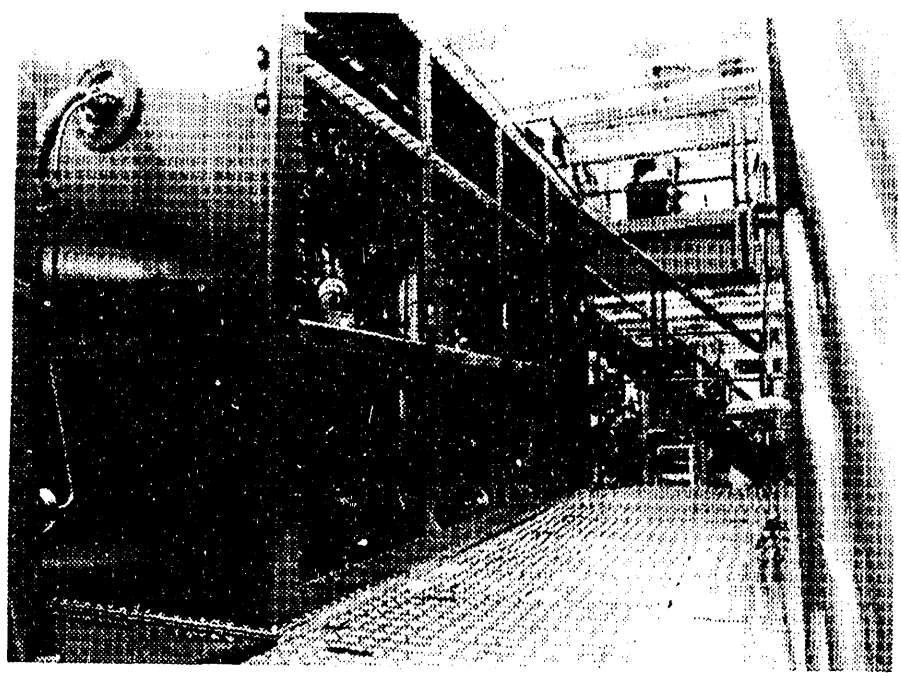

Fig.8. Photo of the JFCU in the TSTA.

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