JAERI-Research
95-034

TSTA/FCU-JFCU TRITIUM EXPERIMENT ON BREEDING BLANKET INTERFACE UNDER THE COLLABORATION OF JAERI-US/DOE (EXTENDED ANNEX IV) —MARCH 1993—

May 1995

Mikio ENOEDA, Toshihiko YAMANISHI, Masayuki YAMADA
Satoshi KONISHI, Kenji OKUNO, Richard S. WILLMS*, David TAYLOR*
Wallace HARBIN*, John R. BARTLIT* and James L.ANDERSON*

日本原子力研究所
Japan Atomic Energy Research Institute

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編集兼発行　日本原子力研究所
印　　刷　㈱原子力資料サービス

TSTA/FCU-JFCU Tritium Experiment on Breeding Blanket Interface under the Collaboration of JAERI-US/DOE (Extended Annex Ⅳ)

— March 1993 —

Mikio ENOEDA, Toshihiko YAMANISHI, Masayuki YAMADA, Satoshi KONISHI
Kenji OKUNO, Richard S. WILLMS*, David TAYLOR *, Wallace HARBIN*
John R. BARTLIT* and James L. ANDERSON*

Department of Fusion Engineering Research
Naka Fusion Research Establishment
Japan Atomic Energy Research Institute
Naka-machi, Naka-gun, Ibaraki-ken

(Riceived March 4, 1995)

In the extended Annex IV (1992 ～ 1994), it was aimed to perform realistic non-steady condition experiments of the fuel loop and Breeding Blanket Interface (BBI) experiments. It was proposed to use the combination process of Cryo-Molecular Sieve Bed (CMSB) and Palladium Diffuser (PD) for the recovery and purification of tritium in the Breeding Blanket purge stream. The BBI campaign experiment by using the CMSB of TSTA-FCU and the PD of JFCU was performed for the recovery of tritium from the simulated Breeding Blanket purge stream in the flow rate of 12.61/min. It was demonstrated that the proposed combination process has the feasibility as the Blanket Tritium Recovery System in fusion plant. This report summarizes the experimental result and analysis of the simulated BBI experiment.

Keywords: The extended Annex IV, Breeding Blanket, Breedivd Blanket Interface, TSTA-FCU, Cryo-molecular Sieve Bed, JFCU, Palladium Diffuser Simulated Breeding Blanket Purge Stream, Flow Rate of 12.6 ℓ /min

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This Research report in the result of the joint study with Los Alamos National Laboratory.

日米協力Annex Ⅳに基づく増殖ブランケットトリチウム回収に関するトリチウム試験結果

日本原子力研究所那珂研究所核融合工学部
榎枝　幹男・山西　敏彦・山田　正行・小西　哲之・奥野　健二
Richard S. WILLMS*・David TAYLOR*・Wallace HARBIN*
John R. BARTLIT* and James L. ANDERSON*

（1995年３月４日受理）

原研は，日米核融合研究協力協定Annex　Ⅳ延長に基づき，米国ロスアラモス国立研究所のトリチウムシステム試験施設（ＴＳＴＡ）において，非定常運転対応核融合炉燃料循環系実証試験及び増殖ブランケットトリチウム回収系模擬実証試験を行っている。本試験は，ＴＳＴＡ燃料精製システムの低温モレキュラーシーブ塔と原研製燃料精製システムのパラジウム拡散器を組み合わせた世界初の増殖ブランケットトリチウム回収系模擬実証試験であった。

試験の結果，トリチウムを含む模擬ブランケットパージガス（流量12.6l/min)を低温吸着方式で３時間以上にわたって精製，回収されたトリチウムガスを３時間以内にパラジウム拡散器で純化する運転が可能であることが実証された。本報告は，この試験結果について詳細に解析，検討したものである。

本報告は，ロスアラモス国立研究所との共同研究の成果である。
那珂研究所：〒311-01　茨城県那珂郡那珂町向山 801-1
＊ロスアラモス国立研究所

Contents

目　　次

1. INTRODUCTION

Tritium Systems Test Assembly is a simulated fusion fuel loop that mainly consists of a torus mockup system with cryopumps (VAC), Fuel Cleanup System (FCU), JAERI Fuel Cleanup System (JFCU), Isotope Separation System (ISS), Transfer Pump Systems (TP1 and TP2), Uranium Tritide Bed (UTB) and Tritium Load-in/Load-out System (LIO). The TSTA has also safety systems such as Tritium Waste Treatment System (TWT), Secondary Containment System (SEC), Tritium Monitoring System (TM), Master Data Acquisition and Control System (MDAC) and Building Ventilation System (VEN). The FCU consists of cryogenic molecular sieve bed, catalytic reactor and hot metal bed, while the JFCU consists of palladium diffuser, catalytic reactor and ceramic electrolysis cell.

In June 1987, Japan Atomic Energy Research Institute (JAERI) and the United State Department of Energy (DOE) signed a collaborative agreement (originally 5 years) , Annex IV to the Japan/U.S. Agreement on Fusion Energy, regarding development of technology for fusion fuel processing. Under this agreement, JAERI and DOE have continued joint operation and experiments on Fuel Processing Technology with the Tritium Systems Test Assembly (TSTA) at Los Alamos National Laboratory (LANL). After a series of successful milestone runs for 5 years, the Annex IV was extended in June 1992, aimed to perform more realistic non-steady condition experiments of the fuel loop and Breeding Blanket Interface (BBI) experiments.

The major function of BBI is to recover tritium from the breeding blanket. In case of solid breeder blanket, tritium in the breeder will be removed and transferred to the helium purge gas containing about 0.1 % of H_2.[1] Hydrogen isotopes in the helium purge gas will be separated from helium by the helium separator. Then, pure helium will be circulated as the purge gas again, while recovered hydrogen isotopes will be purified and sent to the isotope separation process. The combination of cryogenic molecular sieve bed (CMSB) and palladium diffuser (PD) is one of the most major candidate system as the helium separator because of its simple mechanism and large capacity for dealing with more than 50000 l/min purge gas flow rate. Hydrogen isotopes will be roughly recovered by CMSB and purified by PD. By using the CMSB of FCU and the PD of JFCU, demonstrative experiments were possible up to 20 l/min of flow rate scale. This report presents H_2 + HT recovery experiments on the combination of CMSB and PD as the helium separator.

2. Test plan of this experiment

2.1 Purpose

One of the breeding blankets proposed for fusion reactors is composed of solid lithium oxide. tritium will be recovered from such a blanket by sweeping the blanket with a helium purge which contains 1 ~ 0.1 % H_2. This stream will extract T_2 from the blanket, resulting is a stream containing the concentration given in Table 1. It has been proposed that this stream be processed with cryogenic molecular sieve to adsorb the Q_2, leaving the He to be recycled back to the blanket. Once full, the cryogenic molecular sieve bed (CMSB) will be regenerated, sending the Q_2 to the isotope separation system (ISS). A schematic of such a process is given in Fig. 1. Initial tests of this concept have been performed at TSTA. Bench scale experiments were performed using about 50 g of MS5A at liquid nitrogen temperature. Various concentrations of H_2 in He were passed into the CMSB and the H_2 breakthrough curve was tracked using a gas chromatograph.[2] From this data an isotherm could be determined as shown in Fig. 2. It is the purpose of this experiment to demonstrate Q_2 can be separated from He using CMSB. The test will be performed at larger scale that for the bench-scale experiments and with tritium. Data collected will be compared with the bench-scale experiments and will be used for design for future breeding blanket interface equipment. Operating scenarios will be similar to those which are expected to be used for future fusion reactor plants.

2.2 Configuration

A schematic showing the overall configuration for this run is shown in Fig. 3. Two new lines will be installed between the FCU and JFCU as shown on the figure. Also, the 0 - 10 SLPM MKS mass flow controller in TP1 will be replaced with a 0 - 200 sccm MKS MFC and a new 0 - 20 SLPM Brooks MFC will be added to TP1.

The FCU total recycle path will be used to simulate the blanket purge. The loop is filled with about 800 toor of He and circulated at 15000 sccm. Added to the loop is 150 sccm of Q_2 (99% H_2 and 1 % T_2) so that 1 % Q_2 is fed to the CMSB. The estimated capacity of one CMSB is (35 scc Q_2 /g of MS5A) $\times$ 1600 g of MS5A = 56000 scc Q_2. Loaded at a rate of 150 sccm, bed breakthrough would be expected after 6.2 hours. This is a reasonable time for loading in an eight hour shift.

After bed breakthrough, the CMSB is regenerated. The Q_2 which evolved from the CMSB is sent to the inlet of the JFCU Palladium Diffuser (JFCU-PD). Q_2 that permeated through the PD is sent to the JFCU RT2 buffer tank and an LIO PC. The PD retentate is recycled back to the CMSB. This circulation continued until all Q_2 has been recovered from the CMSB. The remaining helium carrier can then be exhausted through a uranium bed to the TWT. The Q_2 recovered from the CMSB regeneration is stored in

RT2 and an LIO PC[3]. It is available for the next CMSB loading. Gas analysis to determine CMSB breakthrough, regeneration gas concentration, etc. will be by Raman spectroscopy. Samples will be sent to the Raman using the FCU gas analysis subsystem. Gas will return from the Raman to the FCU main feed.

2.3 Subsystems and Special Materials Required.

Subsystems required for this test include FCU, JFCU, LIO, TP1, UTB,FCU gas analysis, Raman, MDAC and UTIL. A mixture of 1 % T_2 in 99% H_2 must be prepared for this test.

2.4 Personnel Required

This test should be accomplished using two eight-hour shifts. Each shift must have a minimum of two operators and one staff. The shift assignments are given in Table 2.

2.5 Schedule

This experiment is scheduled to occur during the week of March 8 ~ 12, 1993. One to two days will be devoted to setup. Then, two to three days will be devoted to CMSB loading and regeneration. Normally, bed loading will occur during the A shift and bed regeneration will occur during the Bshift. Shift change meetings will be held at 7:30 and 15:30. All personnel involved with this test must attend these meetings. Other TSTA personnel are welcome to attend.

2.6 Hazards Associated with this Test

No hazards outside the normal working envelope of TSTA will be encountered during this test.

2.7 Data requirements

The primary data from this experiment will be the partial pressure of Q_2 fed to the CMSB, and the Q_2 concentration at the outlet of the CMSB. Concentrations will be determined via Raman (the FCU GC is a backup, the ISS GC). Flows and pressures will be monitored via MDAC and/or local readouts. Pertinent data will be logged in the TSTA Loop logbook.

2.8 Outline of the Test

The outline of the experiment is as follows.

(1) Regenerate FCU MSB 1 and 2.

(2) Prepare a 1200 torr mixture of 1% T_2 in He in an empty LIO PC. See Appendix A for details regarding how to make this mixture. Also, TTA-OP-125-04, R0 may be used as a reference.

(3) Backfill and evacuated all lines that will be used during this run with He. Use Figs. 3, 4 and 5 for reference.

(4) Cool FCU MSB1 with liquid nitrogen. CR1 should be heated to 175 °C (350 °F). The MSBF may be cooled if needed, but this should not be necessary.

(5) Line up the MSB1/TP1-MBP-B loop for circulation. Reference path "A" on Fig. 4.

(6) Load this loop (path "A") with 800 torr of helium.

(7) Turn on the TP1-MBP-B pump and adjust the flow to 15 SLPM using the 0 - 20 SLPM flow controller shown in Fig. 3.

(8) Set the 0 - 200 sccm flow controller shown in Fig. 3 to 150 sccm.

(9) Make sure the mass flow controller bypass in TP1 is closed. Begin flowing Q_2 from the LIO-PC into the 15 SLPM FCU circulation loop. Reference path "B" on Fig. 4. Mark the time.

(10) Use TP1 pumps as necessary to maintain the LIO to FCU flow at 150 sccm.

(11) Start heating JFCU-PD to 623 °C using heating program number 12.

(12) Start JFCU Scroll pump oil pump.

(13) Begin gas analysis of the FCU MSB (CMSB) exhaust using the FCU gas analysis subsystem. Samples are sent to the Raman subsystem for analysis and returned to the FCU main feed.

(14) Using gas analysis follow Q_2 breakthrough to completion (outlet concentration approximately equal to inlet concentration). Pressure in the 15 SLPM loop should begin to rise markedly after breakthrough.

(15) After breakthrough is completed, stop the Q_2 addition.

(16) Continue to circulate gas over the CMSB until the gas concentration stabilizes. Record the pressure and Q_2 concentration.

(17) Turn off the TP1-MBP-B pump.

(18) With the JFCU-PD hot (about 400 °C), establish the FCU/JFCU loop shown as path "C" on Fig.5.

(19) Turn on the JFCU-BP. Then, start JFCU-scroll pump, open HV512, close HV579 and exhaust gas into RT2 and the LIO PC. Reference path "D" on Fig.5.

(20) Open JFCU HCV575. Turn on JFCU-MBP. Begin circulation through the FCU MSB1(CMSB1) by slowly closing HCV575.

(21) Discontinue cooling to CMSB1.

(22) The CMSB1 heater may be used as long as Q_2 evolution does not overwhelm the

JFCU-PD.

(23) Helium may be added to the FCU/JFCU circulation loop as needed to maintain pressure for good circulation.

(24) Continue heating until CMSB1 is about room temperature, then secure the heating.

(25) Continue circulation until Q_2 in the FCU/JFCU loop is sufficiently low (as determined by the test director). this may be monitored by Raman analysis, PD permeate flowrate and by the ion chamber in JFCU.

(26) If the facility will be left with gas in process overnight, reduce the pressure in RT2 to below 590 torr by pumping the gas into the LIO PC. This may be done with the TP1 Scroll pump. Pressure in other tritium containing piping must also be reduced below 590 torr. Do not add impurities (e.g. He) to the LIO PC.

(27) Isolate the subsystems from one another (FCU, JFCU, LIO, TP1).

(28) After isolating the LIO PC, the system may be left unattended for the night in this configuration.

(29) Repeat steps 4 to 25, except use FCU MSB2 (CMSB2) instead of 1. The CMSB1 and CMSB2 operations will not be conducted concurrently.

(30) Final cleanup of the loops used in this test is done by moving as much gas as possible from Raman, FCU GAN, and TP1 (all piping except that between RT2 and the LIO PC) into the FCU/JFCU circulation loop. Recover as much Q_2 from the gas as possible using the PD.

(31) Evacuate the remaining "dregs" gas (mostly He) through a UTB bed and out to the TWT. Use UTB bed 2 for this step.

(32) The LIO PC gas may be stored indefinitely in the PC until a future ISS run when the tritium can be recovered.

(33) Perform an orderly shutdown of all subsystems used in this experiment.

Note: A few newly installed lines and fittings will be used during this test. These include the 0 - 20 SLPM and 0 - 200 sccm flow controllers in TP1 and the new lines between FCU and JFCU that pass through TP1. A pressure leak-up test should be performed on these before they are exposed to tritium. then low level tritium gas should be staged into these new lines, first at sub-atmospheric pressure followed by over atmospheric pressure.

3.Experimental results and Discussions

3.1 High temperature regeneration of CMSB1 and 2

Cryogenic molecular Sieve Beds in FCU were heated in about 220 °C for 14 hours before experiments for the purpose of the complete regeneration. Figure 6 shows the temperature of the CMSB1 and CMSB2 in high temperature regeneration. Adsorption capacity is largely influenced by the regeneration condition such as temperature, purge gas condition, regeneration time, etc. Insufficient regeneration or impurity purge out causes less adsorption capacity of Q_2. Soon after regeneration was completed, the CMSB1 is cooled by liquid nitrogen for Q_2 loading experiment. The CMSB2 was set in stand-by status.

3.2 CMSB1+PD Experiment

Simulated BBI experiment in this study consists of adsorption of Q_2 in He purge flow on the CMSB and purification of Q_2 regenerated from the CMSB by PD in JFCU. The operation of the CMSB was in butch wise operation of CMSB1 and CMSB2. Figure7 shows the temperature of CMSB1 and CMSB2 in the course of the CMSB1-PD experiment. In this figure, important sequences in the experiment are shown as A, B, ···, E and F. Each sequence is described as follows.

A: Initiation of He purge gas flow and Q_2 injection.

B: Stop Q_2 injection.

C: Stop He purge gas flow.

D: Evacuate residual gas in the CMSB1 and start regeneration of the CMSB1.

E: Start collecting purified Q_2 by PD.

F: Complete recovery of Q_2.

Figure 8 shows the flow rate of He purge and Q_2 injection. In the figure, dashed line represents Q_2 injection rate, which was supposed to be 150 sccm, while the He purge gas flow rate was about 9.5 SLPM. The dimension of the CMSB1 is 60 cm height × 7.5 cm f and the particle size of MS5A was 80 mesh. the superficial; gas velocity was calculated as 3.6 cm/s and the Reynolds's number was about 0.5. Because of small size of MS5A particle, the pressure drop in the CMSB1 was as high as 750 torr as shown in Fig. 9. In Fig. 9, the solid line represents the entrance pressure of the He purge gas and the dashed line represents the exit pressure of the CMSB1. As soon as the He purge gas flow was terminated, both of pressure were equalized to the uniform pressure in the line. After termination of the He purgegas flow, residual He gas in the CMSB1 was evacuated to the TWT through regeneration loop (path C in Fig. 5). the regenerated gas was continuously transferred to the regeneration loop. The solid line and dashed line show the pressure and the HT concentration in the regeneration loop in

Fig. 10. The pressure peak between Sequence D and E represents the residual gas transferred from the CMSB to the regeneration loop. As HT concentration did not show any peak, the major composition of the residual gas was considered to be helium. The regeneration of the CMSB1 in early stage was carried out at liquid nitrogen temperature. Consequently, the CMSB1 was heated up to about 100 °C. As soon as the regeneration started, the purification of regenerated Q_2 began in PD. Figures 11 and 12 show the pressure of purified Q_2 by PD and the flow rate of the purified Q_2 transferred to LIO PC as the final product. Recovered Q_2 in LIO PC was used as the injection Q_2 for the next CMSB2 + PD experiment.

3.3 CMSB2 + PD Experiment

Figure 13 shows the temperature of the CMSB2 in the CMSB2 + PD experiment. Operation sequences are almost the same as the CMSB1 + PD experiment. The sequences are summarized as follows.

K: Initiation of He purge gas flow and Q_2 injection.

L: Stop Q_2 injection.

M: Stop He purge gas flow.

N: Start regeneration of the CMSB2 and collection of purified Q_2.

O: Stop regeneration of the CMSB2. Final evacuation of the simulated BBI loop and regeneration loop.

P: End of the CMSB2 + PD experiment.

The flow condition of the He purge gas and Q_2 injection was the same as in the CMSB1 + PD experiment. Figure 14 shows the flow rate of the He purge gas and the pressure of Q_2 injection. Figure 15 shows the inlet and outlet pressure of the CMSB2 in the experiment. As can be seen from this figure, the pressure drop in the CMSB2 was about 300 torr. The regeneration of the CMSB2 and purification of Q_2 proceeded in the same sequences as the CMSB1 +PD experiment. The pressure and HT concentration of the regeneration loop are shown as the dashed line and the solid line in Fig. 16, respectively. In CMSB2+PD experiment, He gas was filled and circulated in the regeneration loop. Thus, the pressure in the regeneration loop was kept as high as 720 torr. On the other hand, regenerated Q_2 gas was purified by PD, which could be seen in Fig. 16 as the decrease of HT concentration in the regeneration operation. (sequence N and O).

Purified Q_2 gas was collected into the PC in LIO. Figures 17 and 18 shows the pressure and the flow rate of purified Q_2 gas. As can be seen from these figures, the major amount of Q_2 gas was purified by PD in 1 hour after the initiation of the regeneration operation.

3.4 Breakthrough Curves of Q_2 Adsorption on CMSB's

3.4.1 Measurement Procedure

In this Breeding Blanket Interface Experiment, a gas stream of helium with nominally 1% H_2 and 0.02% HT was passed through a cryogenic (77K) molecular sieve bed (CMSB's) to simulate processing of the tritium extraction purge gas stream from the lithium breeding blanket in a fusion reactor. The CMSB adsorbs the hydrogen, leaving helium to recycle back to the blanket, until the CMSB becomes fully loaded with hydrogen, at which time the process stream would be diverted to a second CMSB while the first CMSB would be regenerated to send its hydrogen to the isotope separation system (ISS). The hydrogen loading of the CMSB was monitored in these experiments by real-time Raman analysis of a small fraction of the the gas stream exiting the CMSB. Raman analysis provides characterization of CMSB breakthrough and regeneration, as demonstrated in these experiments, even in case of varying tritium concentration, for which an ion chamber diagnostic would be unreliable.

The principal components of TSTA used in this run were the CMSB's in the FCU, a loop to simulate the blanket purge using TP1 and regeneration of the CMSB's using the JFCU-PD during the CMSB loading premixed hydrogen gas (T_2:H_2=1:100) in a LIO-PC was injected through a mass flow controller into the flow controlled helium stream ahead of the CMSB. Details of the flow paths can be found in the section 2. Raman analyses were performed on either the CMSB entrance stream or the CMSB exit stream by sending a small fraction through computer-controlled valves in the FCU gas analysis subsystem to the Raman cell in EXP2 glovebox and back to the FCU main feed. raman analyses were usually made of the H_2 vibration lines (Q-branch) with analysis times of 1 ~ 4 minutes, but occasionally Raman analyses were also made for HT and T_2. Raw Raman spectra were displayed in real-time by the Raman controller adjacent to EXP2 and were interpreted manually for guiding the course of the experiment[4]. The raman spectra were also archived in the Raman controller for later quantitative analyses, which are presented here.

3.4.2 Breakthrough Curves in CMSB1+PD experiment

a)Preliminary CMSB Loading

Hydrogen injection into helium stream flowing through CMSB1 at 77K was conducted in a series of bursts, which concluded at 16:40 on 3/9/93. During most of this time the Raman system was not on-line, as it was being optically aligned and calibrated using nominally pure H_2. Raman analyses of the CMSB1 exit stream, conducted beginning at 16:33, verified that hydrogen breakthrough had already occurred. Initial estimates of the H_2 concentration were too high due to two factors: 1)The calibration gas had been significantly diluted by helium, to which Raman spectroscopy is insensitive; 2)Non equilibrium nuclear-spin distributions in the H_2 evolving from the CMSB required calibrations and analyses for H_2 in even-numbered rotational states ("para-hydrogen") and for H_2 in odd-numbered rotational states ("ortho-hydrogen"). Recalibrations conducted on 3/10/93 yielded proper Raman analysis of the CMSB1exit stream following cessation of H_2 injection on 3/9/93 of 1.82%(±0.02%) H_2, HT:H_2 of 0.03(± 0.1%), and T_2 undetectable (T_2:H_2<0.02%). The high uncertainty in HT reflects the fact that these low HT levels were close to the Raman detectability limit. A single Raman analysis of the CMSB1 exit gas at 21:06 on 3/9/93 during CMSB1 regeneration, when the temperature had risen to about 200K, yielded H_2 concentration of 2.80% with an equilibrium nuclear-spin distribution.

b) CMSB1 Loading

Hydrogen was injected at constant 0.15 SLPM into the 650 torr helium stream flowing at 12.6 SLPM through CMSB1 at 77K from 11:32 to 17:43. Raman analyses were conducted of the helium stream exiting CMSB1 both before and after beginning injection to verify that no H_2 was present (<0.02%). Occasional Raman analyses during injection of the gas stream entering CMSB1 showed H_2 concentrations of 0.51%(± 0.02%) around 12:00 and 0.78% around 15:30; these values are lower than expected from the flow rates. Most Raman analyses were conducted for H_2 in the CMSB1exit stream during and following the H_2 injection; these Raman results are plotted in Fig. 19., which shows separately the partial pressures of even-H_2 and odd-H_2 as well as their sum. These are plotted as partial pressures rather than as concentrations because during injection the total pressure in CMSB1 steadily increased, as shown in Fig. 20. Breakthrough is indicated by the sharp jump in H_2 density in the CMSB1 exit gas at 280 min after initiation of H_2 injection. Interestingly, eve-H_2 was observed significantly earlier than odd-H_2, as has also been observed in gas chromatography of H_2 on alumina columns at 77K. However, after about 20 minute the relative densities of even- and odd-H_2 became consistent with equilibrium at 77K (theoretical even fraction 50.6%, vs. 25% at 300K). As injection continued, a second inflection point occurred in total-H_2 density. After injection was stopped, the H_2 density at CMSB1 exit continued to increase for about 20 minutes to a final equilibrium density, corresponding to a final concentration of 1.68% resulting from injection for 90 minutes following breakthrough. Careful inspection of the CMSB1 pressure trend in Fig. 20 shows the expected increase in slope at the time of breakthrough. After being loaded and equilibrated, CMSB1 was regenerated.

3.4.3 Breakthrough Curves in CMSB2+PD Experiment

a) CMSB2 Loading

Hydrogen was injected at constant 0.15 SLPM into the 450 torr helium stream flowing at 12.6 SLPM through CMSB2 at 77K from 9:38 to 14:38 on 3/11/93. Raman analyses right after beginning H_2 injection verified the absence of H_2 in the CMSB1 exit stream (<0.02%) and showed a concentration in the entrance stream of 0.91 %. Raman analyses of CMSB1 exit stream during and following H_2 injection are plotted in Fig. 21, along with the CMSB2 total pressure. These results are qualitatively similar to those of 3/10/93 (Fig. 19), discussed above, with breakthrough time of about 176 min and final equilibrium H_2 concentration of 2.56 % resulting from injection for 124 minutes following breakthrough. Higher noise and jumps in the CMSB1 pressure trend of Fig. 22 make the slope increase at breakthrough more difficult to discern than in the data of 3/10/93 (Fig.20).

b) CMSB2 Regeneration

After being loaded and equilibrated above, CMSB2 was regenerated with Raman analyses of exit and entrance streams. To regenerate CMSB2 the liquid nitrogen coolant to CMSB2 was stopped, the CMSB2 heater was turned on, and the flow was established through the JFCU PD. The time origin for regeneration is considered to be the time at which the flow was established, signified pressure decrease at CMSB2. The results of Raman analyses of H_2 partial pressures in the CMSB2 exit stream during regeneration are shown in Fig. 23. Figure 24 shows the pressure and temperature change during the regeneration of CMSB2. As the CMSB2 temperature rises, the H_2 partial pressure should rise, but circulation past the PD should then remove H_2 from the gas stream. Interestingly, the H_2 density shows an initial peak at 15 minutes, then a period of lower densities before rising to an even higher peak at 52 minutes with concentration of almost 18 %. The Raman data show that regeneration was essentially completed within 120 minutes, as the CMSB2 temperature was reaching 290 K. The raman analyses indicated that the H_2 nuclear-spin distribution was always consistent with equilibrium at the temperature of CMSB2 throughout the regeneration cycle. The temporal gaps in Fig.23 signify periods of Raman analysis of gas entering CMSB2. Coinciding with the H_2 peak in exit gas at 60 min, the H_2 concentration in the entrance gas corresponded a 98 % removal efficiency of H_2 by the Palladium Diffuser.

3.4.4 Analysis of Breakthrough Curves

Figure 1 shows the modeling concept for adsorption of hydrogen isotopes on CMSB (fixed bed). Cryogenic Molecular Sieve Bed is treated as a fixed bed packed with Molecular Sieve pellets. Helium purge gas is assumed to flow as the plug flow in the bed. It is assumed that the concentration profile of hydrogen isotopes in the radial direction of CMSB is negligible. In general, mass transfer phenomena in a adsorption bed consists of complicated combination among the gas film diffusion, adsorbent surface diffusion, diffusion in micro-pore of adsorbent, and so on. So, it is practical to introduce the overall mass transfer coefficient which represents all the mass transfer steps. The followings is the assumption on which the experimental data was analyzed in this work.

(1) Plug flow of He purge gas in the CMSB
(2) Uniform concentration distribution in radial direction of CMSB
(3) Isothermal operation of adsorption and desorption
(4) Negligible axial diffusion
(5) Overall mass transfer coefficient and linear driving force for microscopic mass balance
(6) Adsorption isotherm is represented by (modified) Langmuir's equation

On the assumption summarized above, mass balance equations of H_2 and HT in CMSB could be written as follows.

$$u\frac{\partial C_H}{\partial x}+\rho\frac{\partial Q_H}{\partial t}+\varepsilon\frac{\partial C_H}{\partial t}=0 \qquad \text{------------------(1)}$$

$$u\frac{\partial C_T}{\partial x}+\rho\frac{\partial Q_T}{\partial t}+\varepsilon\frac{\partial C_T}{\partial t}=0 \qquad \text{------------------(2)}$$

$$\frac{\partial Q_H}{\partial t}=K_F^H a_v\left(C_H-C_H^*\right) \qquad \text{------------------(3)}$$

$$\frac{\partial Q_T}{\partial t}=K_F^T a_v\left(C_T-C_T^*\right) \qquad \text{------------------(4)}$$

$$Q_H=\frac{a_1^H C_H^*}{1+b_1^H C_H^*+b_1^T C_T^*}+\frac{a_2^H C_H^*}{1+b_1^H C_H^*+b_1^T C_T^*} \qquad \text{------- (5)}$$

$$Q_T=\frac{a_1^T C_T^*}{1+b_1^H C_H^*+b_1^T C_T^*}+\frac{a_2^T C_T^*}{1+b_1^H C_H^*+b_1^T C_T^*} \qquad \text{--------(6)}$$

,where, C_H and Q_H represent H_2 concentration in bulk He purge flow and loading on MS5A. C_T and Q_T represent HT concentration in bulk helium purge flow and loading on MS5A. Equations (1) and (2) are macroscopic mass balance of H_2 and HT in CMSB. Equations (3) and (4) are microscopic mass balance of H_2 and HT on MS5A. These equations gives the boundary conditions between bulk gas flow and MS5A pellets.

Equations (5) and (6) are Markham-Benton's isotherm equations for H_2 and HT. Nishikawa etal[5] recommended the Markham-Benton's isotherm by observing H_2, HD and D_2 experiments. The coefficients in eqs. (5) and (6) in terms of H_2 are reported as 4.05×10^{-7} [mol/g ppm], 2.74×10^{-4} [mol/g ppm], 2.03×10^{-7} [1/ppm]and 1.32×10^{-5} [1/ppm] for a_1^H, a_2^H, b_1^H and b_1^H, respectively. Enoeda etal[6] has performed the bench-scale experiments with H_2 and HT and has obtained isotherm coefficients of HT as 7.90×10^{-7} [mol/g ppm], 5.67×10^{-4} [mol/g ppm], 3.04×10^{-7} [1/ppm] and 1.22×10^{-5} [1/ppm] for a_1^T, a_2^T, b_1^T and b_1^T, respectively.

With respect to the boundary condition, the exit gas is directly circulated to the entrance of CMSB's with injected Q_2, which injection rate is constant. Thus the boundary condition for the experiments in this work is described as,

$$C_{H,in} = C_{H,out} + C_{H,inj} \quad \text{-------------------} (7)$$

Equations (1) ~(6) are numerically solved with the equation (7) by using improved Eurer method[7] for various values of the isotope effect on isotherm (K_E) and overall mass transfer coefficients (K_F^H and K_F^T). The best fit between the experimental observation and calculated solution gives the values of K_E, K_F^H and K_F^T.

In Figures 19 and 21, the solid linesshows the comparison of the Raman observation and simulation results of CMSB1-PD run and CMSB2-PD run, respectively. The values of mass transfer coefficient K_F^H, K_F^T are already observed by the bench-scale experiments and known to be about 1.6 cm/min in this work (u=4.6 cm/s, a_v =333.3cm²/cm³ for 80 mesh). The injection concentrations of Q_2 are measured by Raman- analyses as 5577 ppm and 5388 ppm in CMSB1+PD experiment and CMSB2+PD experiment, respectively. The simulation results agreed with the experimental observation relatively well. The bench-scale experiments were performed with 1/16 inch pellet MS5A. The mesh size in this work was different from the bench-scale experiments which were performed with 1/16 inch pellet MS5A, which can be a major reason why the correspondent mass transfer coefficient deviated.

4. Conclusion

The combination of the Cryo-Molecular Sieve Bed (CMSB) and Palladium Diffuser (PD) was proposed for the tritium recovery system in Breeding Blanket Tritium Recovery System of the fusion plant. It was demonstrated that the CMSB + PD process could deal with the simulated blanket purge Gas Stream (H_2 5400 ~ 6200 ppm, H/T ratio 33) in the flow rate of 12.6 l/min. In the demonstration campaign, H_2 + HT gas was successfully recovered by the CMSB for more than 3 hours. Recovered H_2 + HT gas was successfully purified by the PD in about 3 hours. This result implies that the continuous recovery of hydrogen isotopes in the blanket purge gas will be achieved by the combination of the Cryo-Molecular Sieve Bed (CMSB) and Palladium Diffuser (PD). This demonstration campaign was one of the major activities expected in the extended Annex IV.

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2) Willms, R. S., the Proceedings of 15th SOFE at Hyannis, Mass. USA, 61(1993).
3) Konishi, S., M. Inoue, T. Hayashi, K. Okuno, Y. Naruse, J. W. Barnes and J. L. Anderson, "Development of the JAERI fuel cleanup system for tests at the tritium systems test assembly", Fusion Technol., 19 1607~1611(1991).
4) Inoue, M., S. Konishi, T. Yamanishi, S. O'hira, T. watanabe, K. Okuno, Y. Naruse and J. L. Anderson, "Isotope separation system experiments at the TSTA", Fusion Technol., March 1992 Vol.21, No.2 Part 2, the Proceedings of 4th Topical Meeting on Tritium Technology in Fission, Fusion and Isotopic Application, Albuquerque, NM. USA. Sept. 29 ~ Oct. 4 1991.
5) Nishikawa, M., K. Tanaka and M. Uetake, Journ. Fusion Technol., 17-26(1994)26.
6) Enoeda, M., T. Kawamura etal, the Proc. of 5th Topical Meeting on Tritium Technol. in Fission, Fusion and Isotopic Applications, Belgirate Italy May 1995. to be printed.
7) Weber Jr., W. J., J. C. Crittenden, Jour. WPCE, 47, 924(1975)5.

Nomenclature

u : Superficial gas velocity [cm/min]
C_H, C_T : H_2, HT concentration in bulk He purge gas flow [ppm]
Q_H, Q_T : H_2, HT loading on MS5A [mol/g(MS5A)]
C^*_H, C^*_T : Equilibrium gas concentration with Q_H, Q_T [ppm]
x : Bed height location [cm]
t : Operation time [min]
ρ : Bed density [g/cm³]
ε : Bed porocity [-]
K^H_F, K^T_F: Mass transfer coefficient of H_2 and HT [cm/min]
a_v : Specific surface area [cm^2/cm^3]
K_E : Isotope effext on isotherm HT/H_2
a^H_1, a^H_2, b^H_1 and b^H_1 : Coefficients for Markham-Benton's isotherm of H_2
a^T_1, a^T_2, b^T_1 and b^T_1 : Coefficients for Markham-Benton's isotherm of HT

References
1) ITER Documentation Series. #31, IAEA Vienna 1989.
2) Willms, R. S., the Proceedings of 15th SOFE at Hyannis, Mass. USA, 61(1993).
3) Konishi, S., M. Inoue, T. Hayashi, K. Okuno, Y. Naruse, J. W. Barnes and J. L. Anderson, "Development of the JAERI fuel cleanup system for tests at the tritium systems test assembly", Fusion Technol., 19 1607~1611(1991).
4) Inoue, M., S. Konishi, T. Yamanishi, S. O'hira, T. watanabe, K. Okuno, Y. Naruse and J. L. Anderson, "Isotope separation system experiments at the TSTA", Fusion Technol., March 1992 Vol.21, No.2 Part 2, the Proceedings of 4th Topical Meeting on Tritium Technology in Fission, Fusion and Isotopic Application, Albuquerque, NM. USA. Sept. 29 ~ Oct. 4 1991.
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6) Enoeda, M., T. Kawamura etal, the Proc. of 5th Topical Meeting on Tritium Technol. in Fission, Fusion and Isotopic Applications, Belgirate Italy May 1995. to be printed.
7) Weber Jr., W. J., J. C. Crittenden, Jour. WPCE, 47, 924(1975)5.

Nomenclature

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C_H^*, C_T^* : Equilibrium gas concentration with Q_H, Q_T [ppm]
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t : Operation time [min]
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a_v : Specific surface area [cm^2/cm^3]
K_E : Isotope effext on isotherm HT/H_2
a_1^H, a_2^H, b_1^H and b_1^H : Coefficients for Markham-Benton's isotherm of H_2
a_1^T, a_2^T, b_1^T and b_1^T : Coefficients for Markham-Benton's isotherm of HT

Table 1 Concentrations Expected from Breeding Blanket Purge Stream.

Component	Concentration (%)
He	99.0
H_2	0.99
T_2	0.01

Table 2 Shift Assignments.

Shift	Time	Operators	Staff
A	7:30～15:30	Dahlin, Wilhelm	Willms, Enoeda, Yamada, Barnes
B	15:30～23:30	Hamerdinger, Harbin	(Willms), Yamanishi, Bartlit(M-W), Carlson(Th-F)

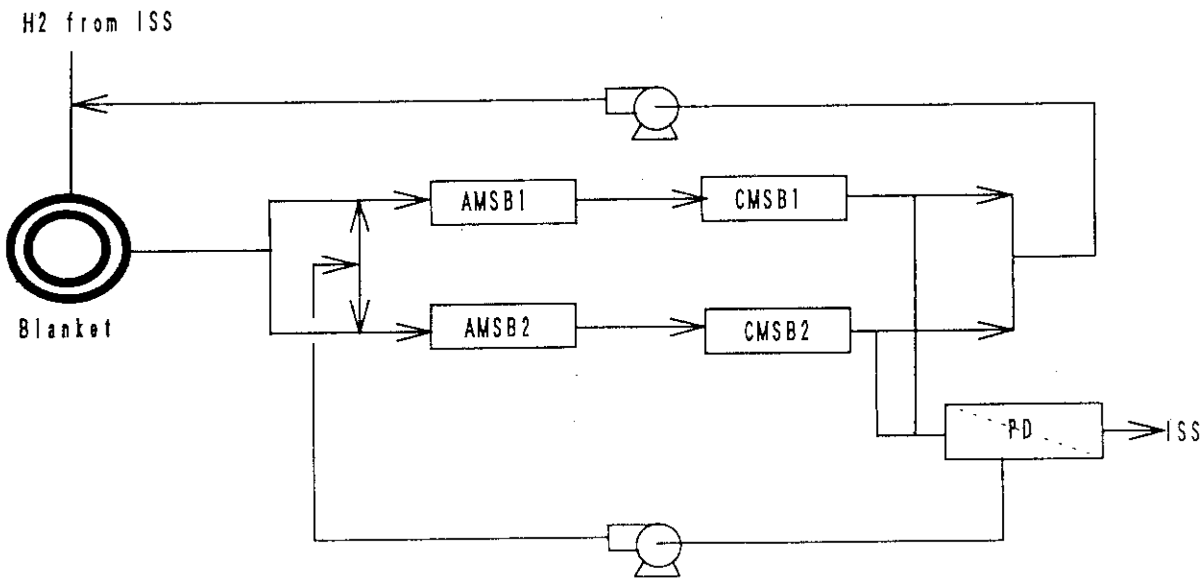

Fig.1 Breeding Blanket Interface Proposed for Solid Lithium Blanket Processing.

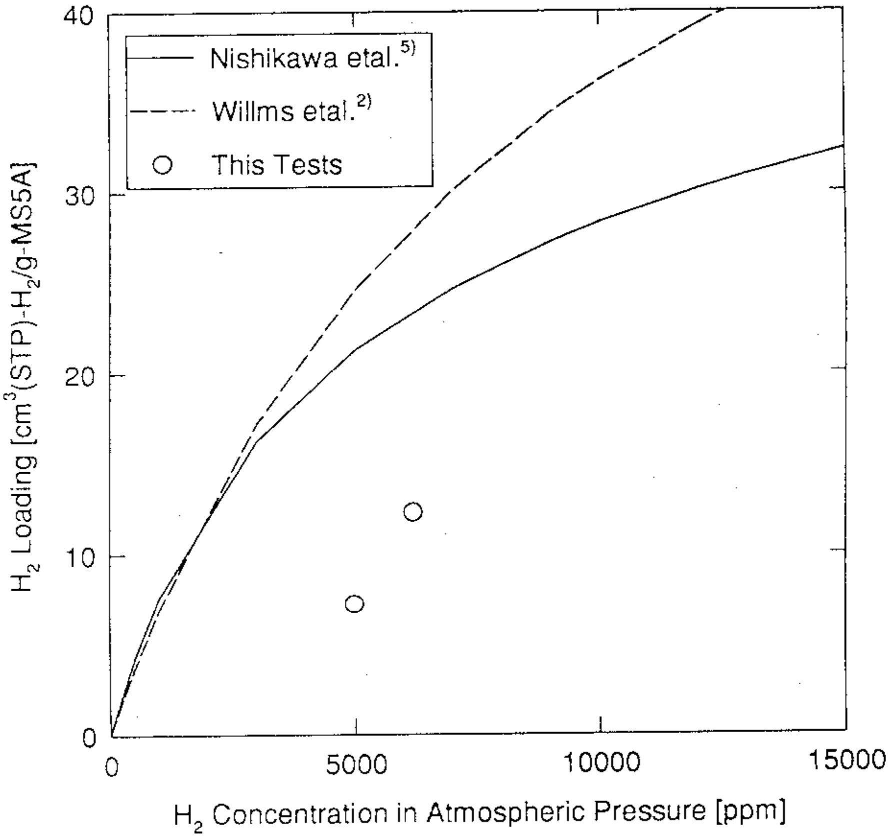

Fig.2　H_2 Isotherm on MS5A in LN_2 Temperature.

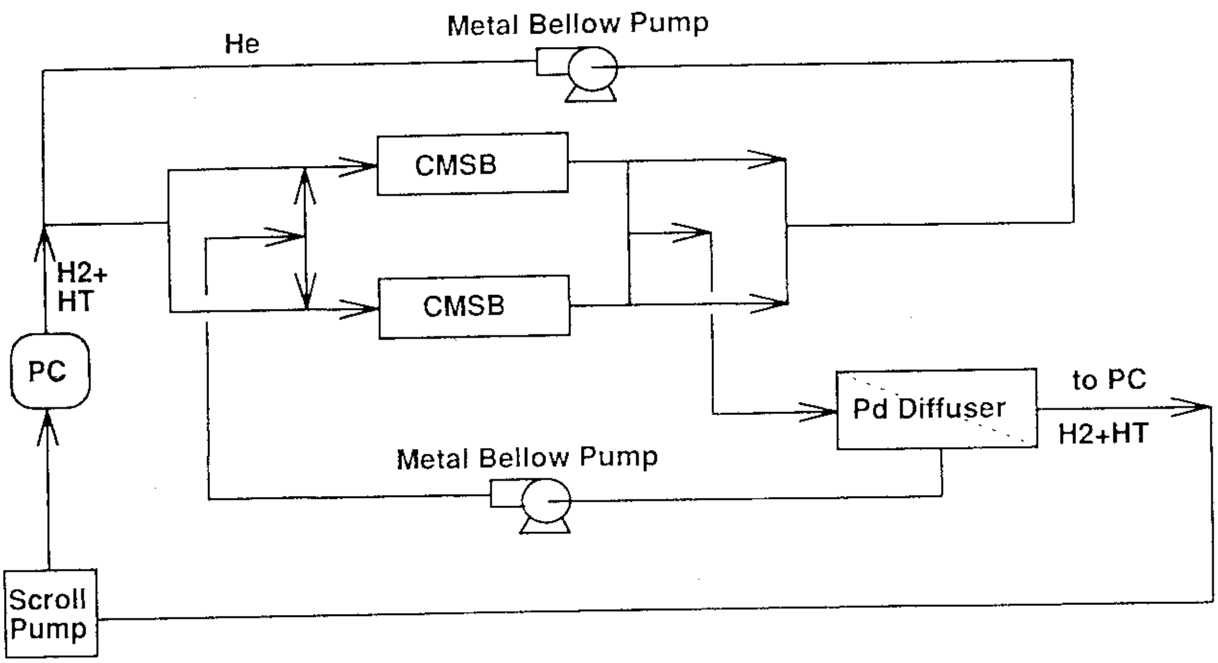

Fig.3　Schematic Flow Sheet used in FCU-JFCU BBI Experiments.

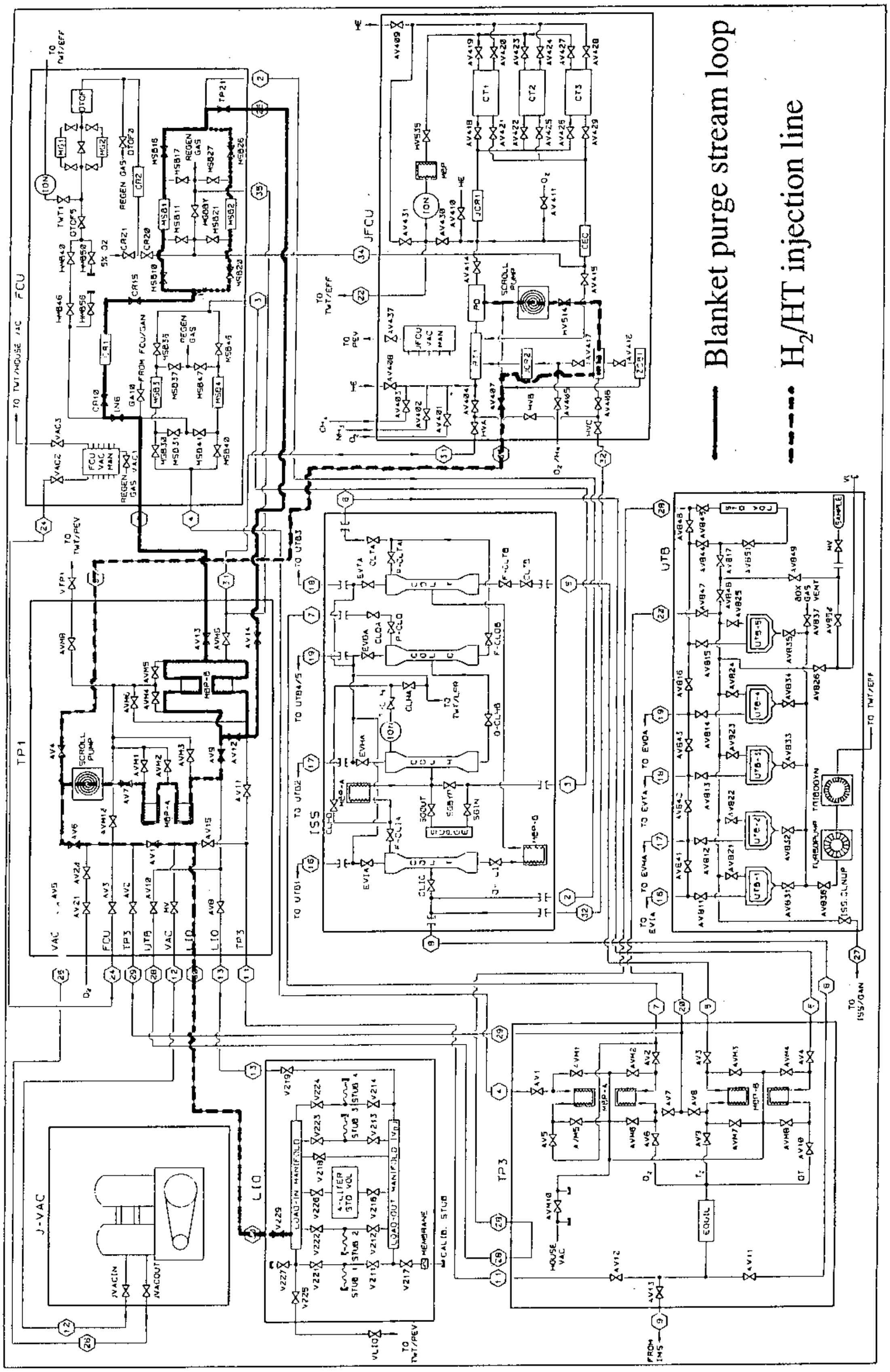

Fig.4 Flowpath of Simulated Blanket Purge Stream Loop and H_2/HT Injection Line.

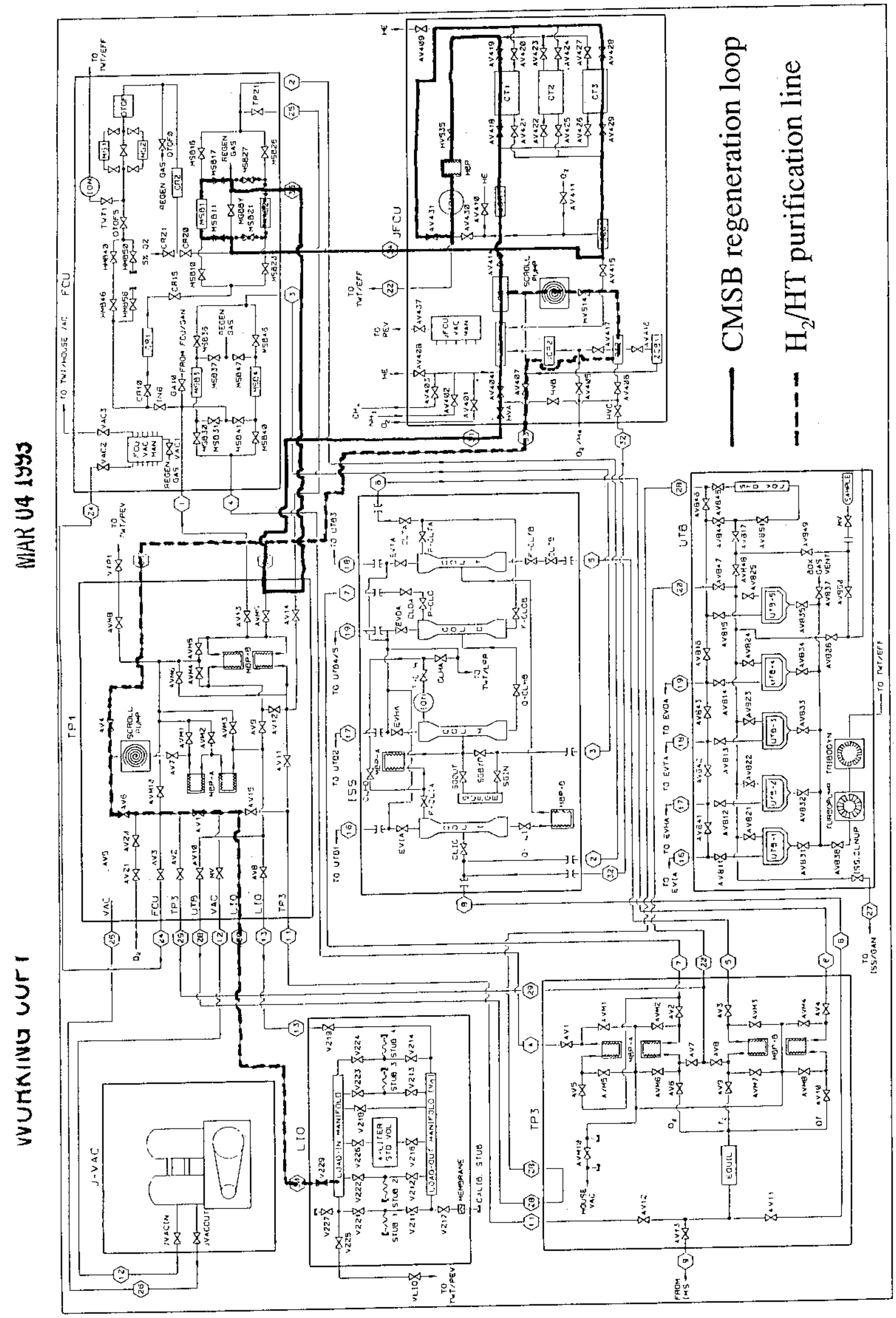

Fig.5 Flowpath of CMSB Regeneration Loop and H_2/HT Purification Line.

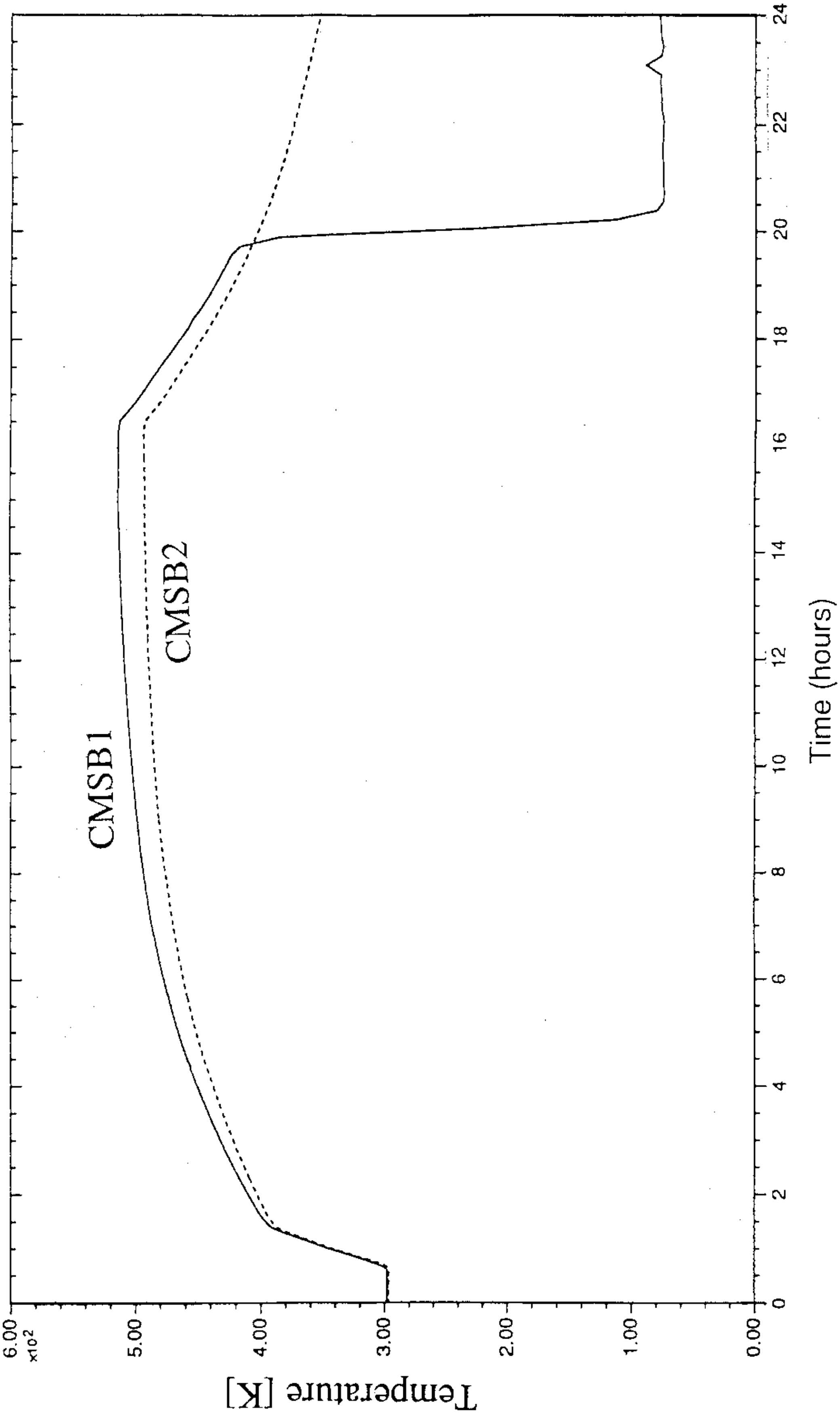

Fig.6 Temperature of CMSB1 and CMSB2 in High Temperature Regeneration.

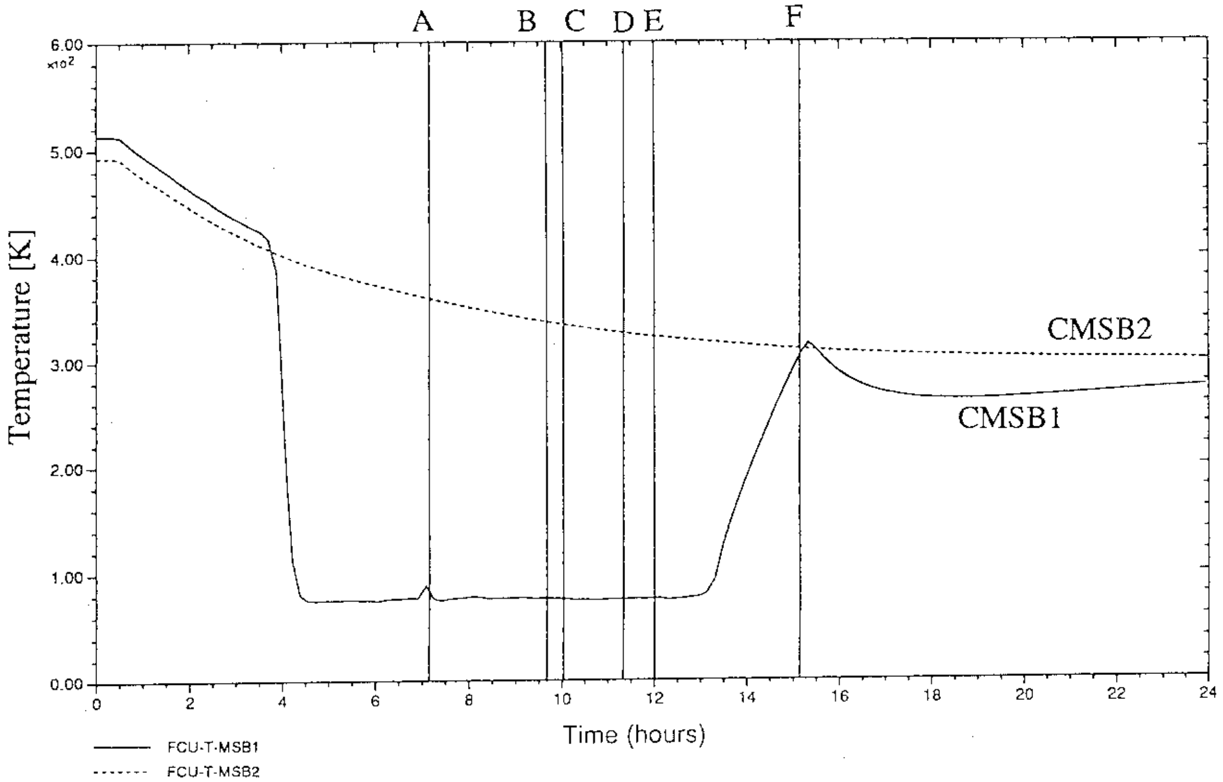

Fig.7 Temperature of CMSB1 in Adsorption and low temperature regeneration.

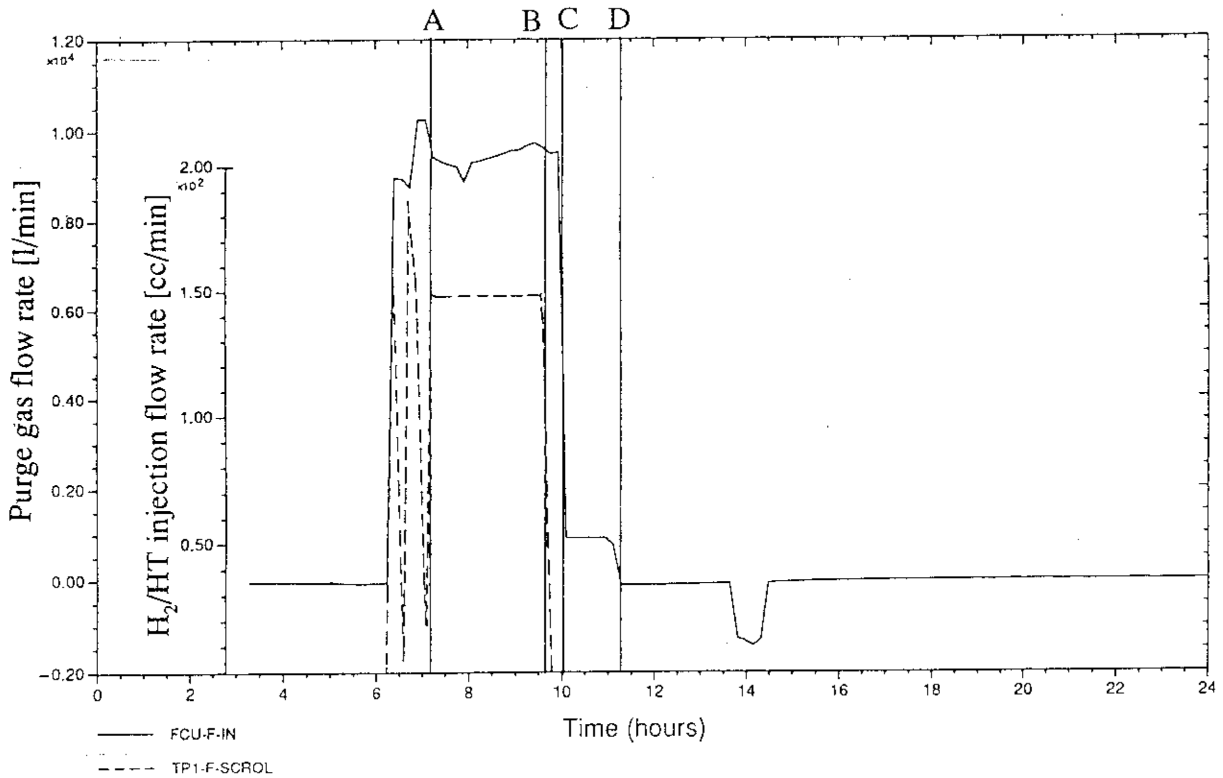

Fig.8 Flow Rate of Simulated BBI Purge Gas and H_2/HT injection in CMSB1 Run.

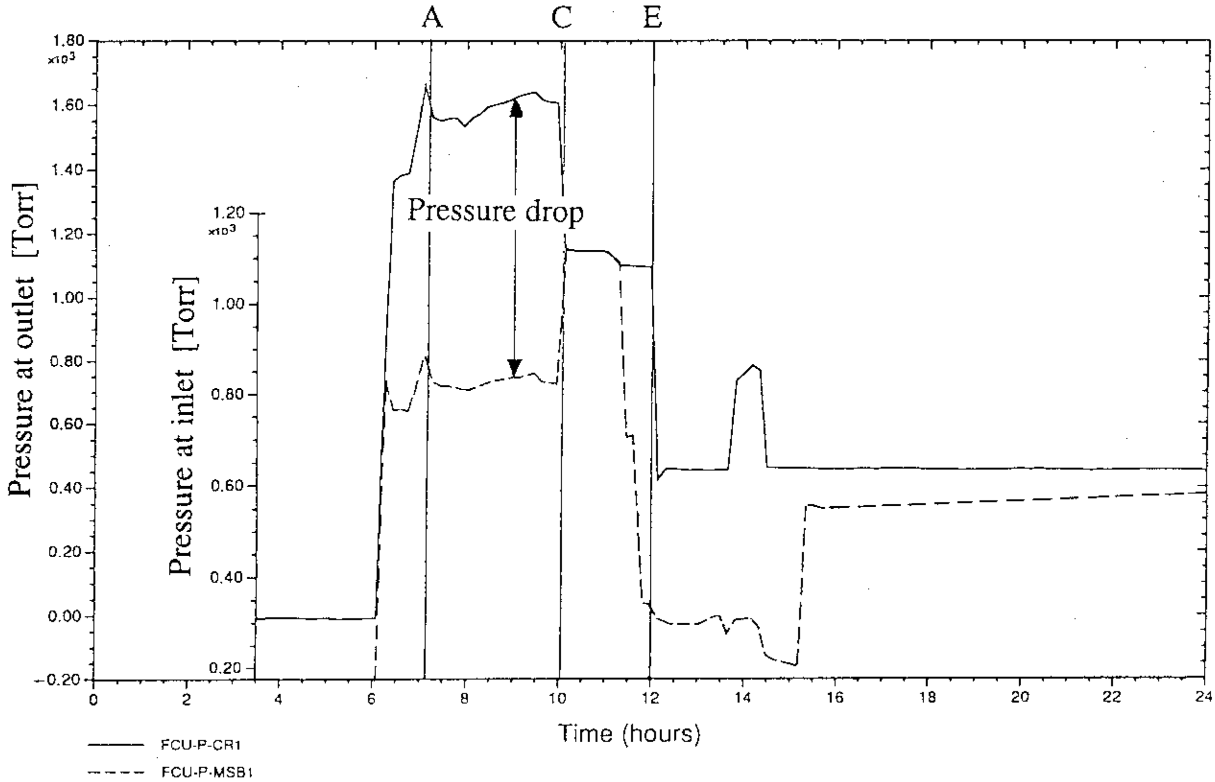

Fig.9 Pressure Drop in MSB1.

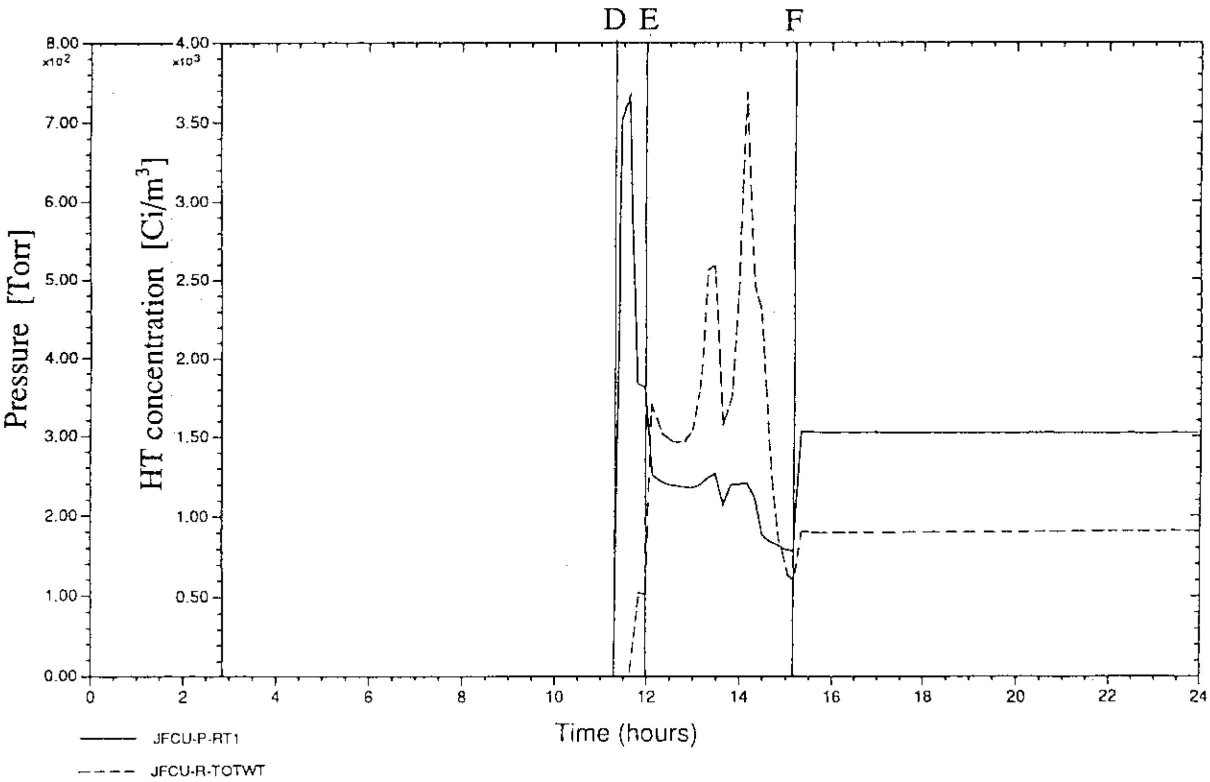

Fig.10 Regenerated Gas Pressure and HT Concentration Recieved in JFCU-PD Breed Loop. (CMSB1 Run)

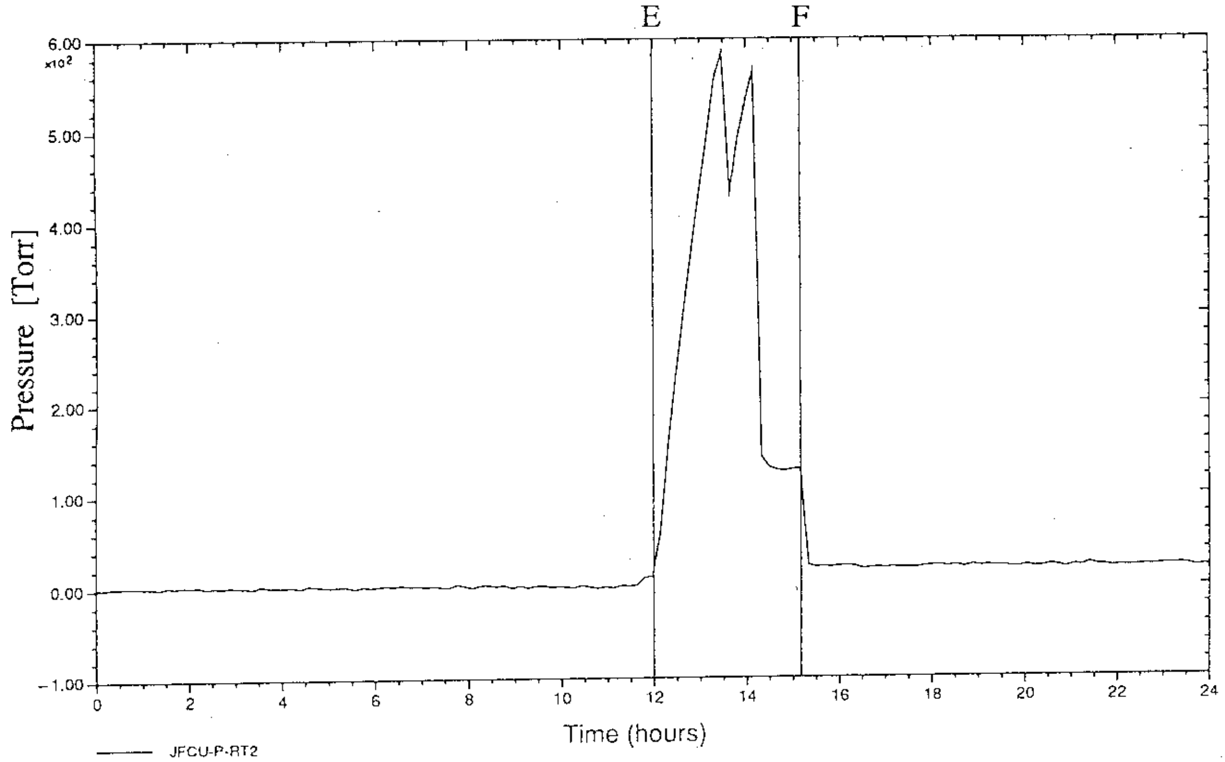

Fig.11 Purified H_2/HT Gas Pressure sent from JFCU-PD to PC in CMSB1 Run.

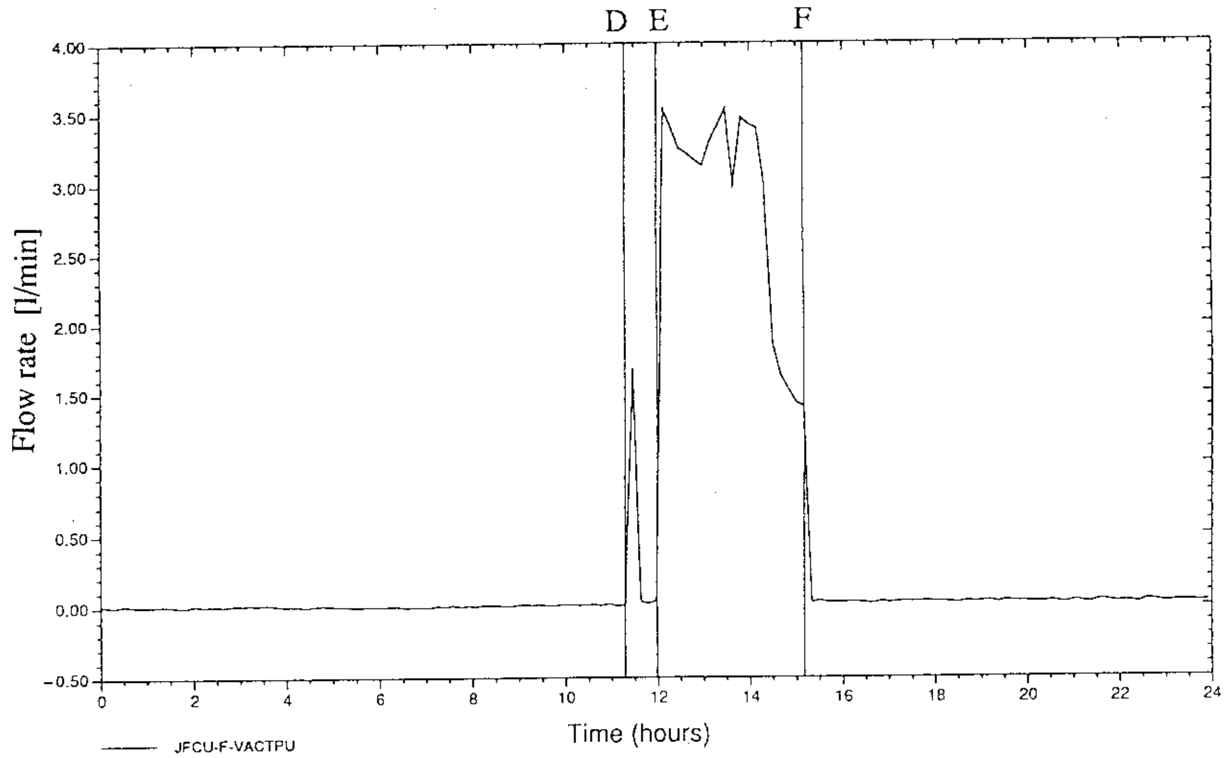

Fig.12 Flow Rate of Purified H_2/HT Gas Recovered to PC in CMSB1 Run.

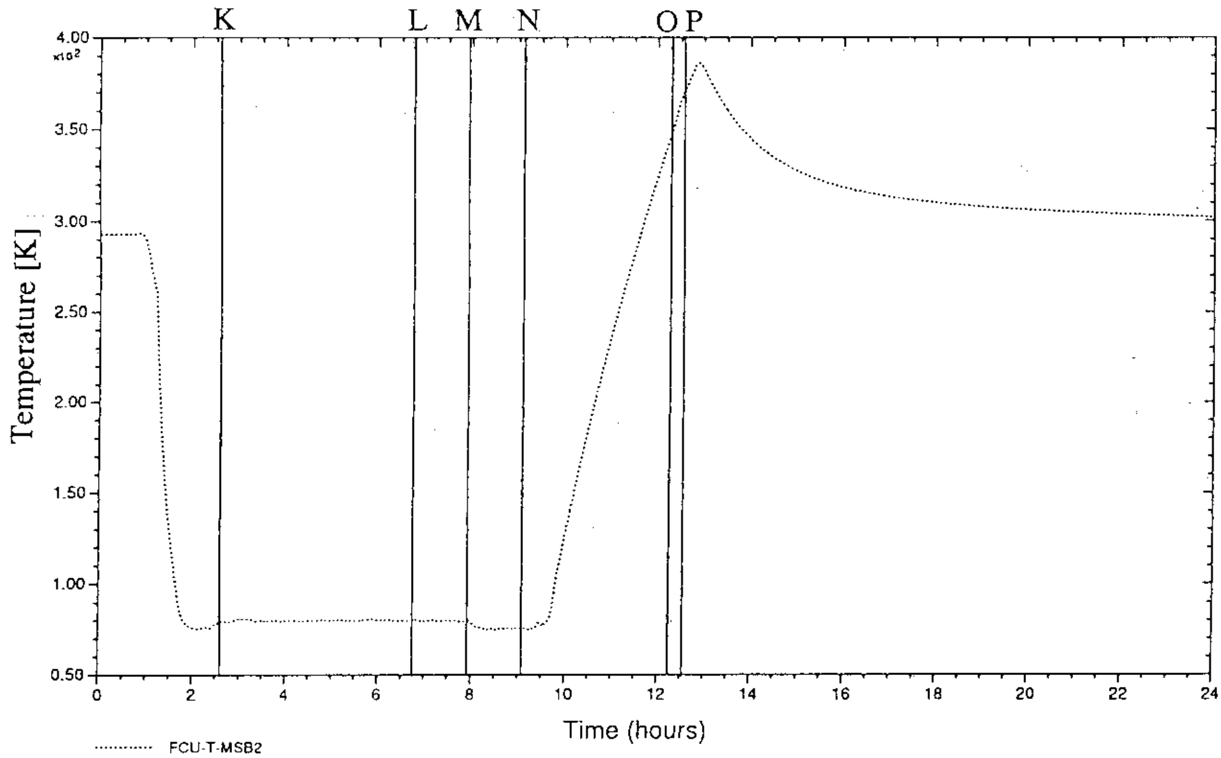

Fig.13 Temperature of CMSB2 in Adsorption and low temperature regeneration.

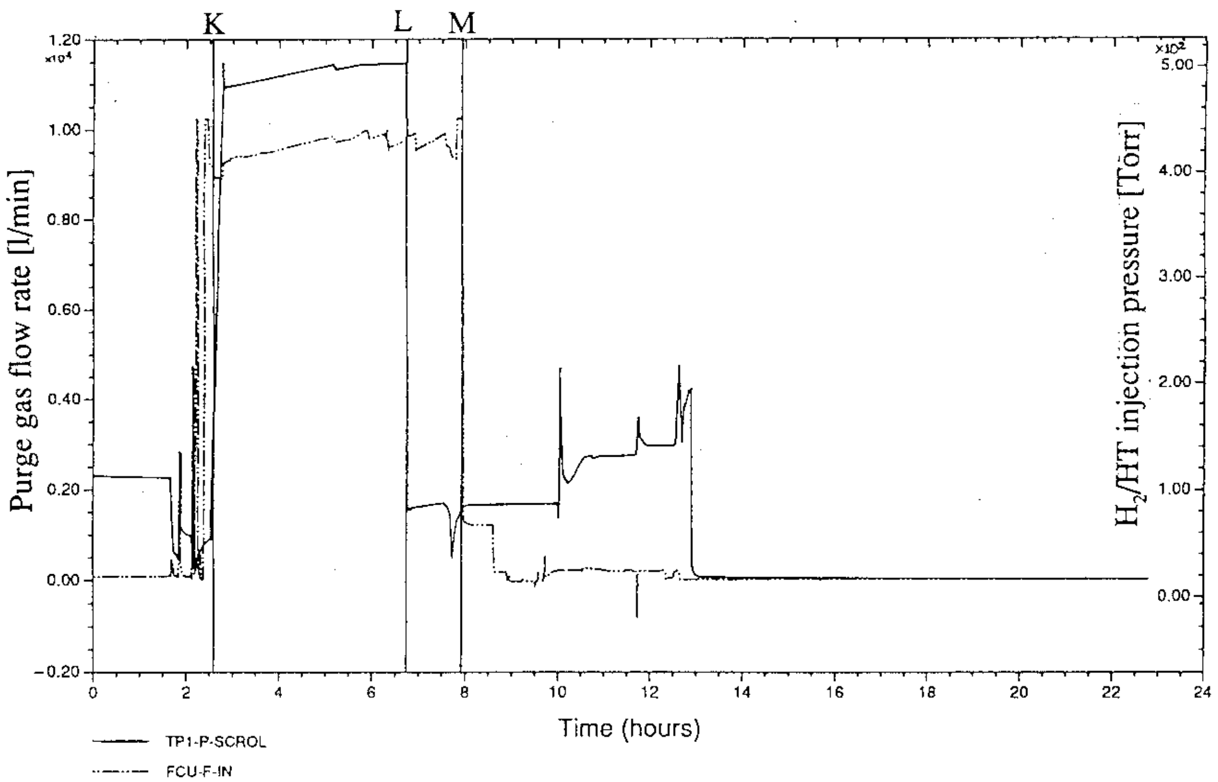

Fig.14 Flow Rate of Simulated BBI Purge Gas and H_2/HT Injection Pressure in CMSB2 Run.

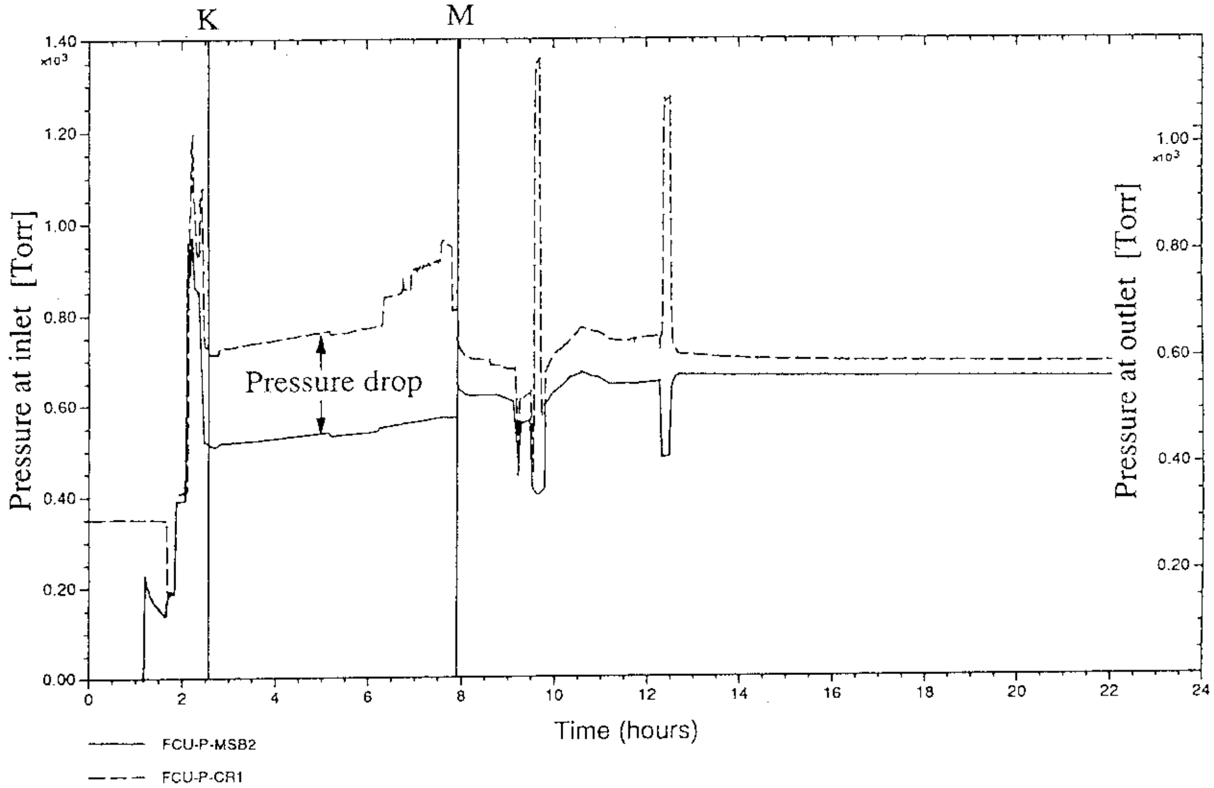

Fig.15 Pressure Drop in MSB2.

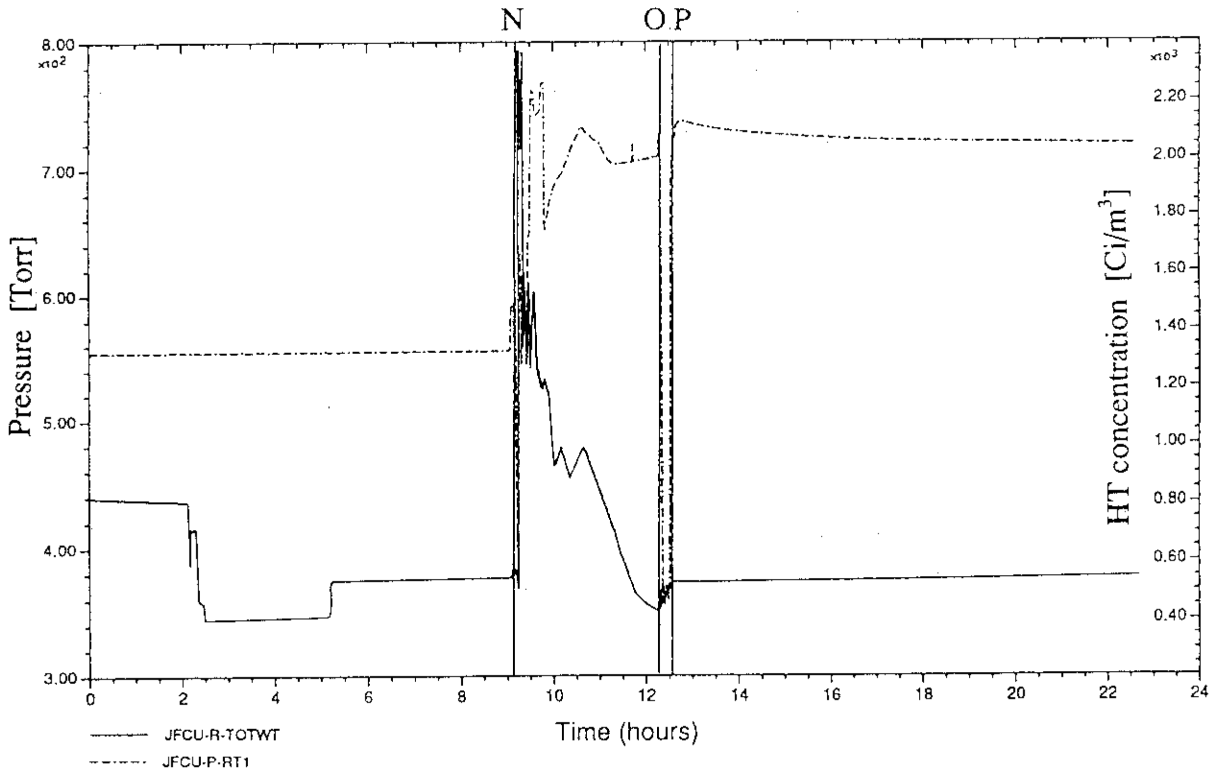

Fig.16 Regenerated Gas Pressure and HT Concentration Recieved in JFCU-PD Breed Loop. (CMSB2 Run)

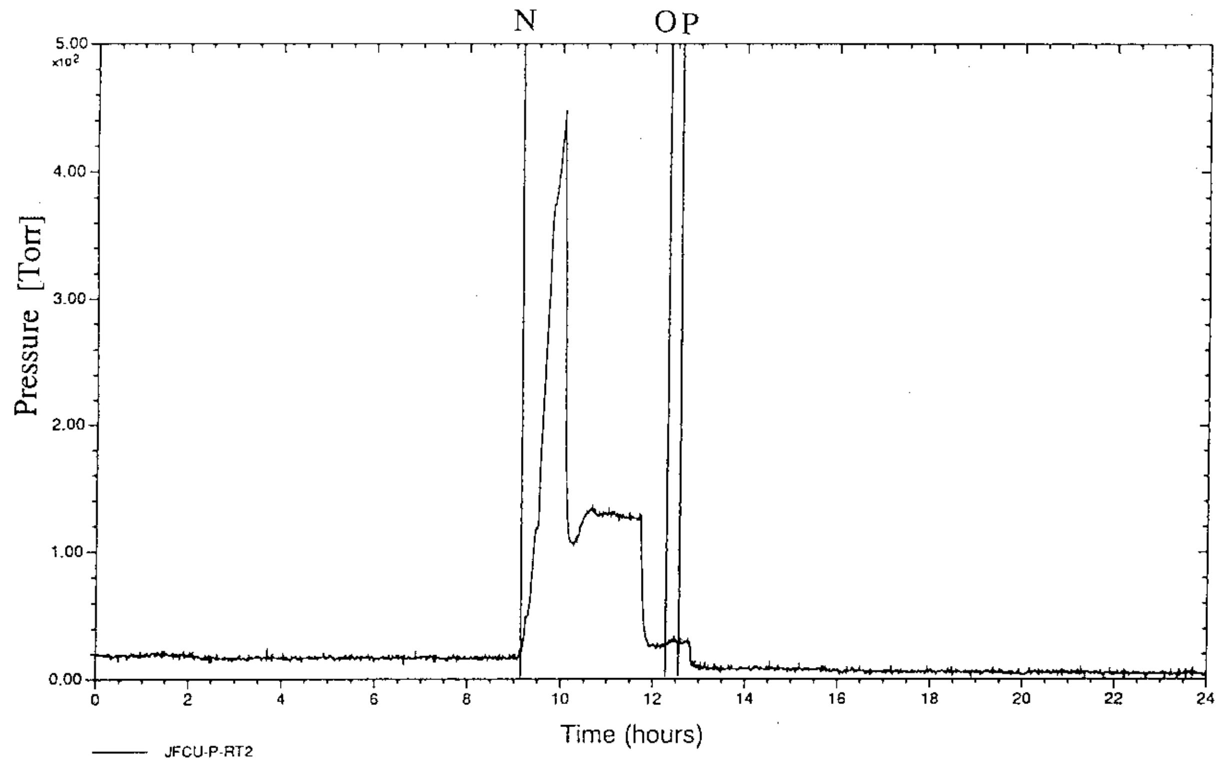

Fig.17 Purified H_2/HT Gas Pressure sent from JFCU-PD to PC in CMSB2 Run.

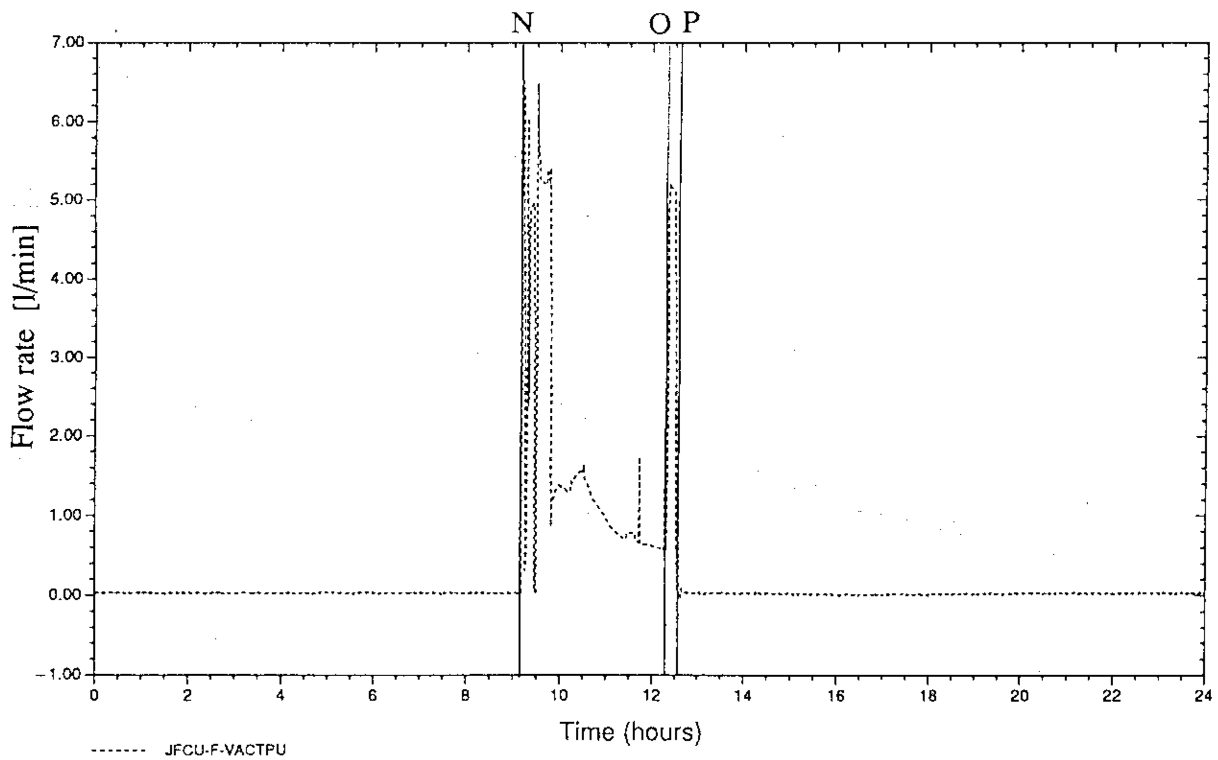

Fig.18 Flow Rate of Purified H_2/HT Gas Recovered to PC in CMSB2 Run.

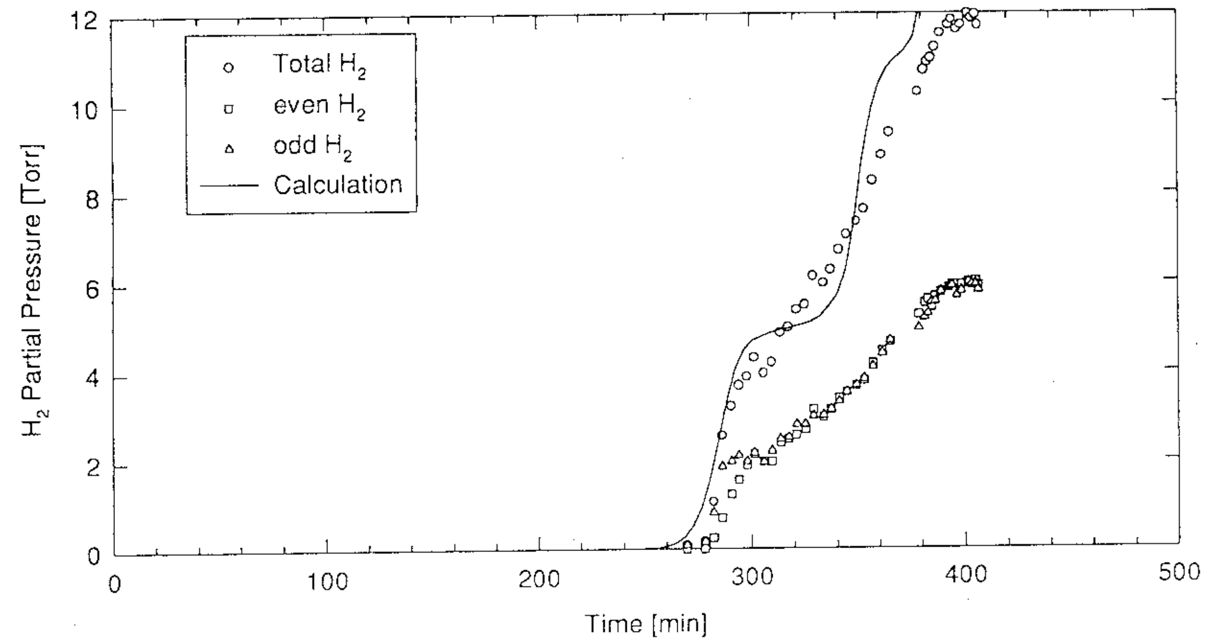

Fig.19 Raman Measurement Results and Calculation of Breakthrough Curve in CMSB1-PD Run.

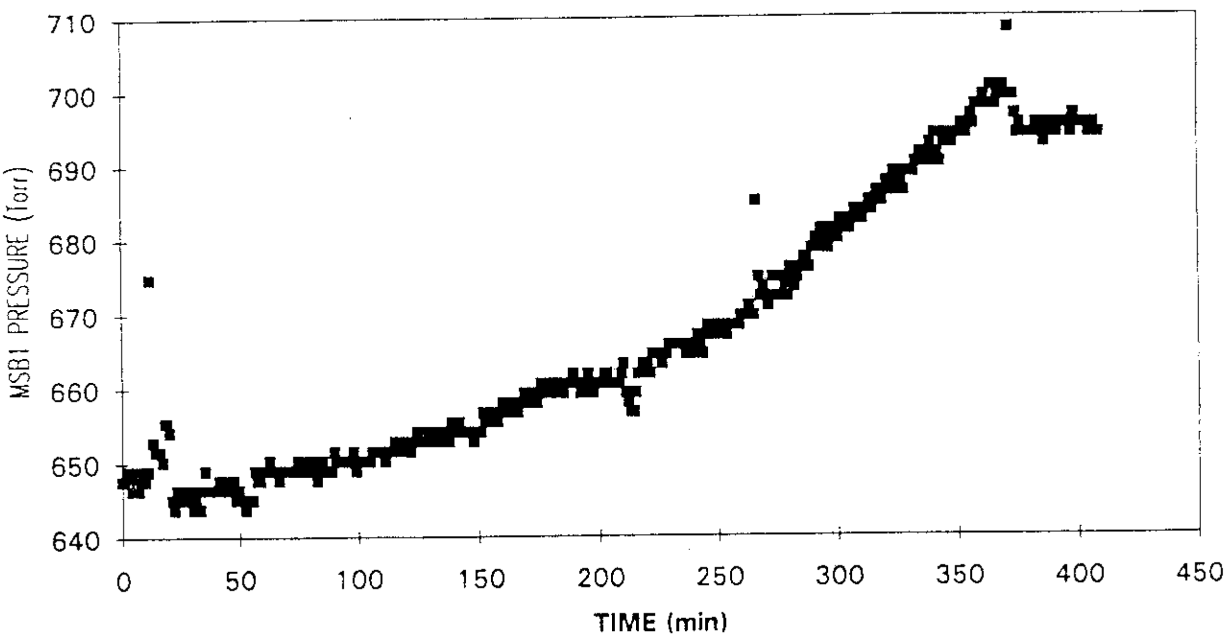

Fig.20 Pressure Change in CMSB1-PD Run.

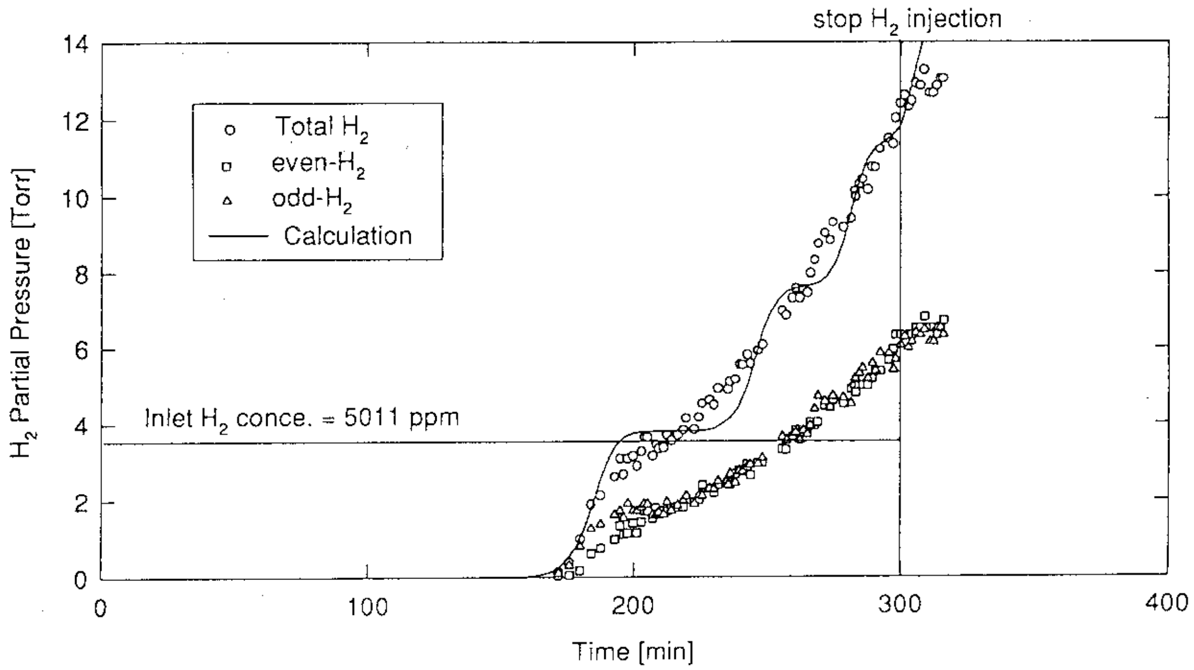

Fig.21　Raman Measurement Results and Calculation of Breakthrough Curve in CMSB2-PD Run

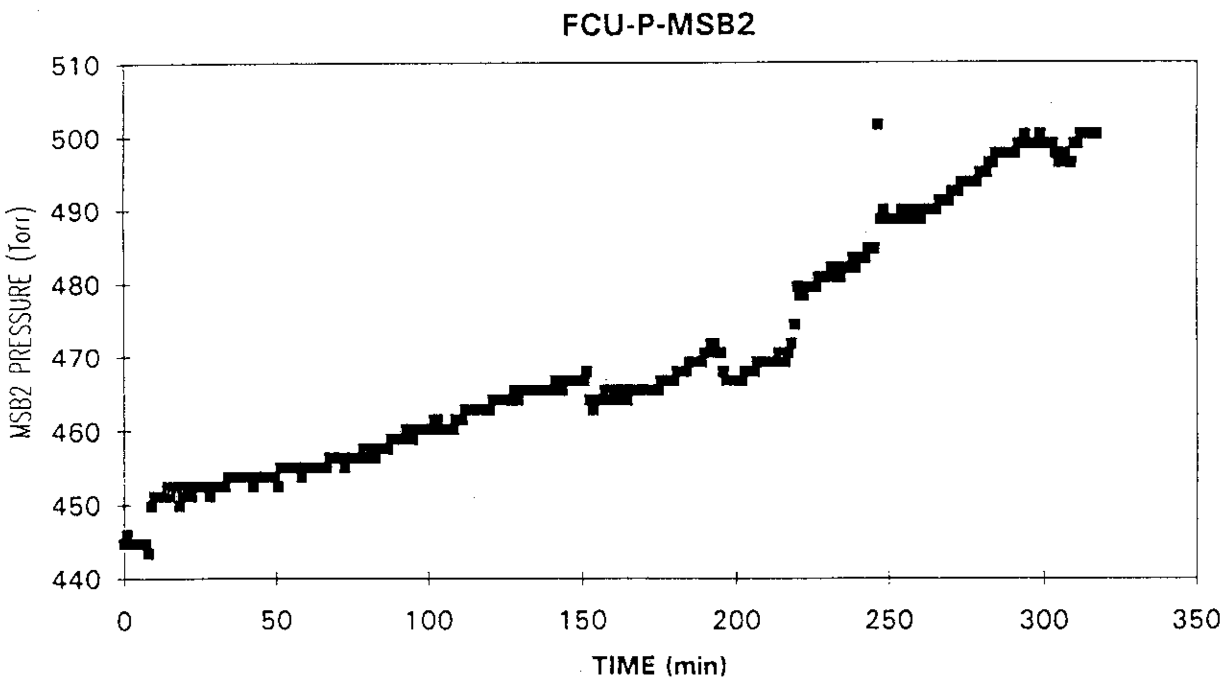

Fig.22　Pressure Change in CMSB2-PD Run.

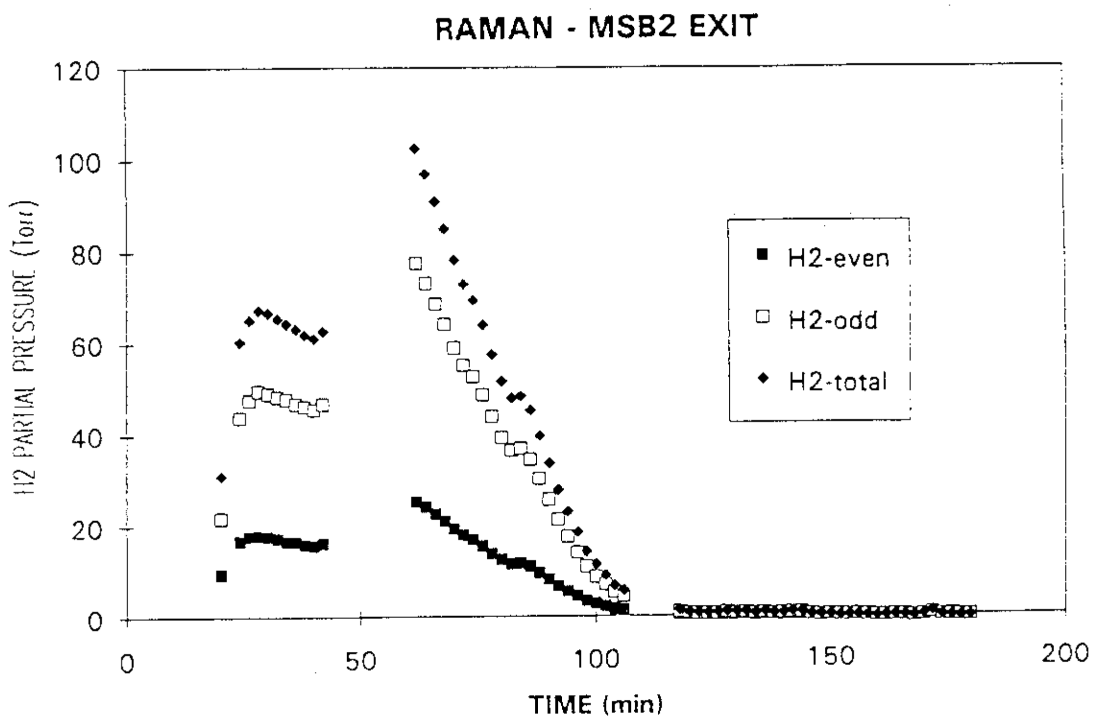

Fig.23 Raman Measurement Results in CMSB2 Regeneration.

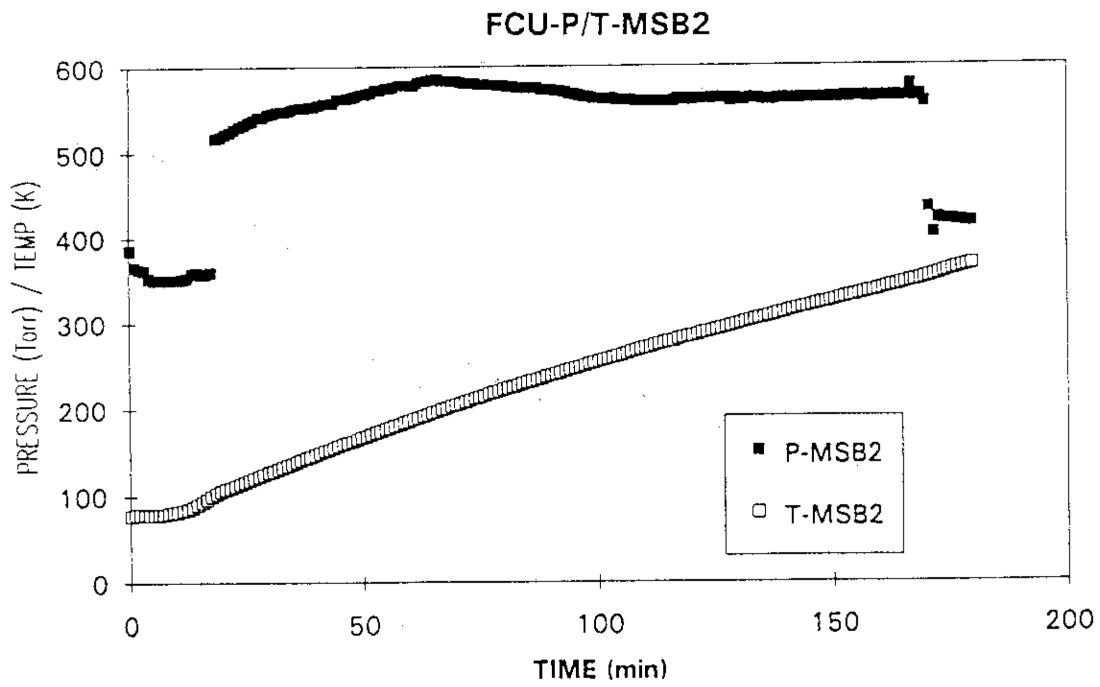

Fig.24 Pressure and Temperature Change in CMSB1 Regeneration.

Appendix　Preparation of Q2 Mixture

A 1 % mixture of T_2 in H_2 is required. The total quantity of gas to be prepared is
(1200 torr/760 torr)50 liters = 78.9 liters
Thus, (0.01)x(78.9liters)=0.789 liters tritium is required.
PC831 contains 91.52 % T_2. The LIO standard volume is 3.728 liters. Thus the LIO standard volume should be filled with
(1/0.9152 fraction tritium)x(0.789 liters/3.728 liters)x760 torr=176 torr of PC831 gas
After filling the LIO standard volume with 176 torr of PC831 gas, the standard volume should be isolated. All LIO lines should be emptied and an empty PC attached to LIO. All of the LIO standard volume gas can then be transferred to the empty PC. H_2 is used to raise the final pressure of the PC to 1200 torr.