FUSION FUEL PURIFICATION DURING THE TRITIUM SYSTEMS TEST ASSEMBLY 3-WEEK LOOP EXPERIMENT

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INTRODUCTION

During the time period from 4/19 - 5/5/89, the Tritium Systems Test Assembly (TSTA) at Los Alamos National Laboratory (LANL) conducted its longest continuous integrated loop operation to date. This provided an opportunity to test some hitherto unproven capabilities of the TSTA Fuel Cleanup System (FCU). Previous FCU tests were reported in [1]. The purpose of the FCU is to remove impurities from a stream of hydrogen isotopes (Q_2) representative of torus exhaust gas. During this run impurities loadings ranging from 60 to 179 sccm of 90% N_2 and 10% CH_4 were fed to the FCU. Each of the two FCU main flow molecular sieve beds (MSB's) were filled to breakthrough three times. The MSB's were regenerated during loop operations.

DESCRIPTION OF THE TSTA FCU

Purification

The FCU, shown schematically in Figure 1, employs 77 K MSB's (Linde 5A) to remove impurities from Q_2. MSB1 and MSB2 are used to purify the main flow path (nominally 50/50 D/T), while MSB3 and MSB4 purify the neutral beam gas (nominally 100% D_2). During this run impurities were only added to the main flow path. Before the impurities break through on one bed, its exhaust is routed to the inlet of the companion bed (path not shown). In this way an MSB can be taken to 100% loading, leaving an insignificant amount of co-adsorbed Q_2 when an MSB is regenerated.

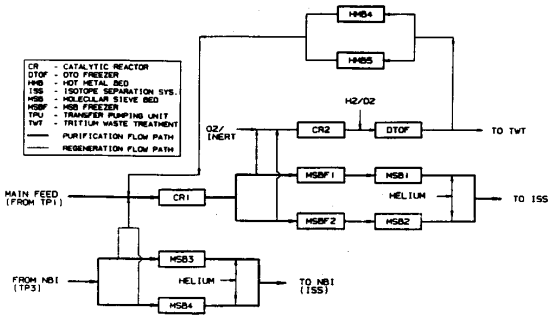

Figure 1. FCU Flow Schematic

The purpose of catalytic reactor 1 (CR1) is to cause any free oxygen impurity to combine with Q_2 so that an explosive mixture will not develop on the MSB's. The molecular sieve bed freezer (MSBF's) serve to keep water out of the MSB's. If water reachs the MSB's, higher regeneration temperatures are required.

Regeneration

When an MSB breaks through (detected by gas chromatography), flow is diverted exclusively to the companion bed and the bed needing regeneration is taken off-line. Since the adsorbed impurities contain tritium (in this case as tritiated methane), the impurities cannot simply be exhausted to the tritium waste treatment system (TWT). Rather, as the bed is warmed and the impurities desorb, they are mixed with O_2/He before being admitted to catalytic reactor 2 (CR2). Therein, tritium-containing impurities are converted to Q_2O and other oxides (in this case, CO_2). The Q_2O is captured as ice in the deuterium tritium oxide freezer (DTOF), and the remaining impurities containing negligible tritium are exhausted to the TWT.

A mixture of 5% O_2 and 95% He is used to mix with the impurities. This supplies the oxygen necessary for combustion, but will not form an explosive mixture. The amount of O_2/He added is controlled by an oxygen monitor in the CR2 exhaust. Generally the control system is set to maintain a 2% O_2 concentration at this point.

Periodically the DTOF must be regenerated. This involves heating the DTOF and driving the water over a hot metal bed (HMB) which contains uranium. Therein, the water is reduced to uranium oxide (solid) and Q_2 (gas) which is readmitted to the FCU main flow. Table 1 summarizes the properties of each FCU component.

SUMMARY OF OPERATIONS

Narrative

On day 0 of the run (4/19/89), as seen on Figure 2, loop flow was started through MSB1. The following day impurities addition was begun. Late on day 2 flow was routed from the exhaust of MSB1 to the inlet of MSB2 so that MSB1 could be taken to complete breakthrough which occurred on day 3. Flow was then routed through MSB2 alone and impurities were turned off while MSB1 was regenerated (on some of the subsequent regenerations impurities addition remained on).

Table 1. FCU Components

Component	Contents	Operating Temperature (K)
CR1	387 gm Precious Metal Catalyst	450
CR2	387 gm Precious Metal Catalyst	800
DTOF	3 L Baffled Vessel	160
HMB4,5	4 Kg Uranium	750
MSB1,2,3,4	1.6 Kg 5A Molecular Sieve	77
MSBF1,2	3 L Baffled Vessel	160

Loop flow was interrupted on day 4 due to an N_2 plug in the isotope separation system (ISS). After loop flow resumed on day 5, an excessive pressure buildup occurred upstream of MSBF2 due to a water plug. Flow was diverted through MSB1 while MSBF2 was regenerated. After resuming flow through MSB2, it broke through on day 7.

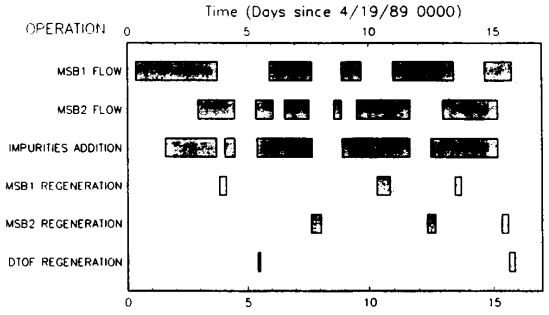

Figure 2. Summary of FCU Operations

Soon thereafter a number of problems were encountered. It was discovered that the loop flow was inadvertently opened to the ISS surge tank, resulting in a low loop flow inventory. After this was corrected, an ISS plug, possibly caused by O_2 ice, occurred. Loop flow resumed on day 8. At this point a water ice plug in MSBF1 necessitated routing loop flow through MSB2 until MSBF1 could be regenerated. Resuming MSB1 flow resulted in its breakthrough on day 9. Though these problems were somewhat disheartening, the quick resolution of them, resulting in the continuation of operations, attest to the versatility of the TSTA piping layout.

From this point on, operations became somewhat routine. MSB2 broke through on day 11, and impurities were off at the end of this day to arrange for the convenient timing of the MSB1 breakthrough on day 13. The last loading of MSB2 was completed on day 15 after which impurities addition was turned off.

Each MSB breakthrough was followed by its regeneration. The DTOF was regenerated after the first MSB regeneration, and then not again until the last day of the experiment.

Impurities Addition

Impurities (90% N_2 and 10% CH_4) were mixed with the main TSTA loop flow in the transfer pumping unit (TPU) upstream of FCU. Usually the impurities addition flowrate was 60 sccm, but other flowrates--97, 120, 179 sccm--were also used. Impurities addition was on for 10.2 days (discontinuously) resulting in a total of 967 liters (870.3 liters N_2 and 96.7 liters CH_4) added to the loop. Table 2 shows the quantity of gas adsorbed on each bed at breakthrough. The average quantity of impurities adsorbed per bed was 161.2 liters. This is 84% of the theoretical capacity of these 1.6 Kg beds. The remaining 16% of the bed is believed to be occupied by water which cannot be driven off of the beds because of their 500 K temperature limit (a temperature of about 600 K is required for completely drying molecular sieve).

Table 2. Molecular Sieve Bed Performance

Loading #	1	2	3
Bed		Liters Loaded	
MSB1	162.1*	158.1	161.1
MSB2	162.1*	158.0	165.2

* Precise loading information was not available for these loading since the exact MSB1 breakthrough time is not known.

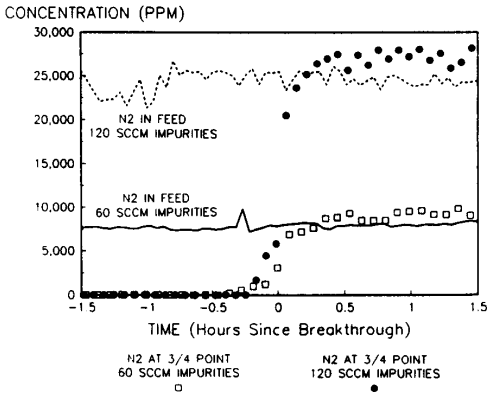

Figure 3. MSB Breakthrough at Two Impurities Addition Rates

Figure 3 shows two typical MSB breakthrough curves. The markers represent the N_2 concentration at a point 3/4 down the length of the MSB. The methane is not observed at breakthrough because it is more strongly adsorbed and is trapped at the bed inlet. The lines represent the N_2 concentrations in the feed to the MSB based on the ratio of N_2 addition flowrate to the total loop flowrate. In the case of the solid circles and dashed line, the impurities were being added at 120 sccm during the breakthrough, while the impurities flowrate for the squares and solid line was 60 sccm. In both cases the breakthrough N_2 concentration is higher than the N_2 feed concentration. This phenomenon is called "roll-up" or "roll-over" behavior [2]. It is caused by the CH_4 displacing previously adsorbed N_2 from the front of the bed. Thus, the measured N_2 concentration is the sum of the N_2 in the feed and the CH_4 displaced N_2.

The tritium inventory on an MSB varies as it progresses from preloading to impurities loading to regeneration. Before impurities are loaded on a bed, it is cooled to 77 K and preloaded with O_2. This requires 148 liters of DT which represents 20 grams of tritium. As impurities are loaded on the bed, the O_2 is displaced and returned to the loop. When a bed is completely loaded with impurities the tritium inventory on the bed depends on the amount of tritium in the impurities. In this experiment, if the CH_4 exchanged to $CH_2D_1T_1$ (a reasonable estimate of what actually occurred), the tritium inventory on a bed would be 2.2 grams. This inventory is returned as O_2 to the loop when the DTOF is regenerated. Neither the operation of FCU nor the operation of other TSTA systems (most notably ISS) were adversely affected by these changes in MSB inventories.

MSB Regeneration

Regeneration of an MSB was accomplished by first starting the O_2/He flow through CR2 and the DTOF, and out to the TWT. Then a 1.4 SLPM He purge was started to sweep desorbing gases out of the MSB to CR2 while the MSB was heated. The gas evolution from the bed could be monitored using gas chromatography (GC) to sample the helium purge as it exited the MSB. These analyses could be performed at a maximum frequency of one every four minutes.

The results of such analysis are shown in Figure 4. The markers represent the CH_4 (squares) and N_2 (triangles) concentrations. The solid line is the

temperature measured at the center of the bed. The reason for the broad nature of these desorption curves is related to the MSB configuration. The beds are constructed of two hemi-cylinders oriented with the cylinder axis vertical (process flow is down one hemi-cylinder, then up the other side, and out). This assembly is submerged in a liquid nitrogen bath. During regeneration the liquid nitrogen level drops as it boils away. This creates significant temperature gradients. Gas desorption begins at the top of the bed and eventually works its way to the bottom. This broad desorption pattern is advantageous since it prevents overloading of the O_2/He addition system.

The O_2/He addition rate was automatically adjusted to maintain a 2% O_2 concentration at the CR2 exhaust. The resulting O_2/He flowrate is shown in Figure 5. As expected the maximum flowrate coincided with the maximum CH_4 concentration. Each mole of CH_4 requires 2 moles of O_2, while the only effect of N_2 on the O_2/He is that of dilution.

Also monitored during regeneration was the radiation of the gas leaving FCU for the TWT. This is shown in Figure 6. The radiation values recorded where only the background readings of the ion chamber. Thus, no measurable tritium was lost in this step of an MSB regeneration.

Though anticipated as a possible problem, no plugging (due to ice formation in the inlet) of the DTOF occurred.

DTOF Regeneration

The last step in the FCU operations is the regeneration of the DTOF. The system is designed for DTO steam to pass over hot uranium where it is converted to uranium oxide and Q_2 gas. However, when

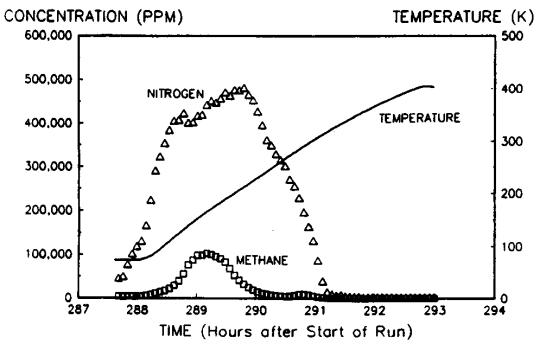

Figure 4. MSB Regeneration: Gas Evolution and Temperature

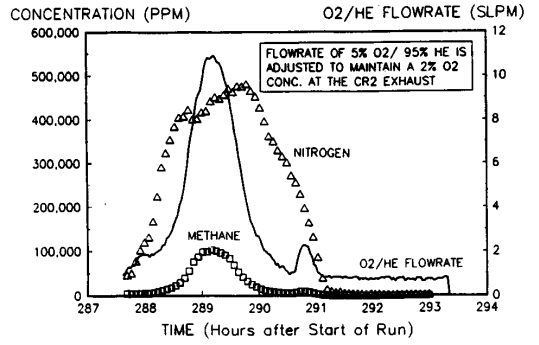

Figure 5. MSB Regeneration: Gas Evolution and O_2/He Flowrate

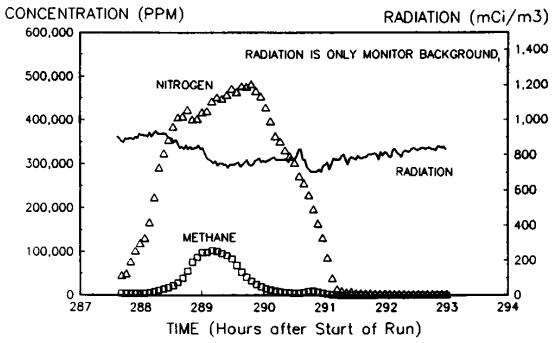

Figure 6. MSB Regeneration: Gas Evolution and Radiation

this step was attempted, the MSBF's plugged with ice in their inlet tubing (heater tape should solve this plugging problem in the future). The fact that so much water passed unconverted through the HMB indicates that conversion was quite poor. The bed was well activated and had plenty of capacity, 60%, before being used. However, after completion of this run the HMB capacity was measured and found to be reduced to 15%, much more than could be accounted for by the 8.6 moles of water produced during the experiment. Though it is unclear what caused this loss of capacity, it is feared that the formation of oxides on outer surfaces is preventing access to active material beneath the surface.

CONCLUSIONS

The two principle findings of this experiment were:

- The use of cryogenic molecular sieve is a very robust method for removing impurities from fusion fuels. While loop flowrates, pressures and impurities concentrations varied, the MSB's continued unaffected, completely removing impurities. No ISS plugging occurred during this run due to impurities passing through the MSB's.

- The first step in the MSB regeneration process also proved to be very effective. An automatic system, requiring minimal operator action, added the appropriate amount of O_2/He for complete impurities oxidation. No measurable tritium was lost to the TWT.

The principle problem uncovered during this experiment was the poor conversion of water to UO_2 and Q_2. Work is continuing to determine why this process did not work as designed.

REFERENCES

[1] R. Scott Willms, "Recent Operating Results at the Tritium Systems Test Assembly Using Molecular Sieve at 77K for Purifying a Fusion Fuel Process Stream", Presented at the First International Symposium on Fusion Nuclear Technology, Tokyo, April 10-15, 1988.

[2] Ralph T. Yang, Gas Separation by Adsorption Processes. Boston: Butterworth, 1987, p. 173.