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Commissioning and Operation of the JET Active Gas Handling System Flake Processing Facility

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ABSTRACT.

In 1998 following the DTE1 tritium experiment 150g of carbon material in the form of flakes and dust were remotely removed from the JET vacuum vessel. Subsequent measurement by calorimetry showed the flake material to contain 0.5g of tritium and a specific activity of 1.1TBq/g. Because of the specific tritium activity and out-gassing, the flakes were not considered to be a subject for disposal as waste but to be processed and the tritium recovered. A facility has been built within the Active Gas Handling System (AGHS) to recover the tritium and eliminate the flake material by its complete oxidation. The process yields HT which can then be transferred into the main AGHS processing loop for isotopic separation. Initially the process will be proven on batch sizes of up to 2g flake material and will eventually be scaled up to 50g batch sizes. This paper describes the inactive commissioning of the flake processing facility and the active processing runs on the flake material.

1. INTRODUCTION.

The facility for the processing of tritiated flake material was designed, manufactured and installed in the AGHS ready for functional testing and commissioning in Spring 2006. The basic plant and process has been previously described [1], the only change from the original design concept was the use of a commercial Thermal Gravimeter (STA 409 PC Luxx of NETZSCH) in place of a high temperature larger volume reactor (R2 in fig.1). The disadvantage of using the gravimeter was the limited volume of the gravimeter crucible which limited the flake batch size to 2g. However the gravimeter did enable the mass change within the crucible to be monitored during the combustion process. Figure 1 shows the process lay out of the facility. Figure 2 shows a diagram of the Thermal Gravimeter.

2. INACTIVE COMMISSIONING OF THE FACILITY.

Inactive commissioning commenced with functional testing of the individual components of the facility prior to being finally assembled on the plant. These tests confirmed that flakes/dust of CFC material can be completely oxidized in atmosphere of oxygen-argon mixture at temperature of 1120K. The SAES getter type ST909 is capable for quantitative decomposition of water vapour and production of molecular hydrogen at operation temperature of 970K [2,3]. Under some operational conditions a complete metal oxidation can be achieved. The palladium sorbent, 20wt% palladium coated on alumina, absorbs hydrogen at pressure above and below that needed for forming palladium hydride. Absorption occurs also in the presence of water vapour and after exposing the palladium sorbent to ambient air. Hydrogen can be quantitatively desorbed from the palladium sorbent by purging with argon at temperature of 570K. Similar effectiveness shall be expected for desorption under vacuum. In addition to the process tests, all safety interlocks and control operations of the facility were proven to function correctly prior to the inactive commissioning commencing.

The purpose of inactive commissioning was to perform an overall test of the fully assembled facility. The Cold Trap (CT) was filled with mini spirals of 2mm×2mm size made of stainless steel wires of 0.2mm diameter. The spirals were acid etched to increase their wetting properties for water.

The reactor R4 was filled with pellets of the SAES getter ST909 type. The getter was activated by purging with argon, which contained 4vol.% of hydrogen, at temperature of 770K for 4 hours. The palladium reactor R5 was filled with sorbent of 20wt% palladium coated on alumina. The sorbent was activated by purging with argon, which contained 4vol.% of hydrogen, at temperature of 570K for 4 hours. After the activation procedure the hydrogen absorbed by palladium was desorbed through the reactor purging with pure argon for 20 minutes at 570K.

A sample of CFC dust was prepared using non-contaminated material. The crucible was loaded with 633.4mg of dust. Water, 283.0mg, was added to the crucible. This was to simulate a presence of free water, which can be produced as a result of tritium self-oxidation when tritiated flakes/dust are in contact with air [2].

Initially the cold trap was cooled down to about 220K and thermal gravimeter was purged with argon initially at room temperature for 10 minutes and then heated to temperature of 370K. Reading of the humidity meter changed from Dew point of 250.0K at the beginning of the step to 272.9K at the end of the step. Total mass loss of 277.9mg recorded by the thermal gravimeter indicated that nearly all the water added to the dust sample was transferred to the cold trap.

The CFC dust was oxidized at temperature of 1120K under Ar/O_2 (in 1:1 ratio) mixture of $50cm^3$/ min flow rate. Sample of dust was completely oxidized in 160 minutes with the recorded mass loss of 633.4mg. No noticeable change in the humidity meter reading was recorded. The cold trap was then warmed up stepwise for controlled release of carbon dioxide, which was collected in the reservoir and then discharged.

For water decomposition in reaction with the getter ST909 the reservoir was filled with argon, which then re-circulated through the cold trap, reactor R4 maintained at operation temperature of 770K and through palladium sorbent in the reactor R5 maintained at room temperature. Argon flow rate was $100cm^3$/min. The cold trap was heated stepwise up to 470K. At the end of the water decomposition the reading of the humidity meter decreased to Dew point of 241.75K. At the final stage of water decomposition the palladium reactor R5 was cooled down to 250K while continuing argon re-circulation. Finally the argon was evacuated from all the reactors, compressed in the reservoir and discharged. The palladium sorbent was then heated to 570K and the released hydrogen was compressed in the reservoir. Volume of molecular hydrogen desorbed from the palladium sorbent was 0.330NL, which is equivalent to decomposition of 266mg of water. More than 90% of water added to the crucible was collected in the cold trap, converted to gaseous hydrogen and absorbed by palladium sorbent. This showed a good efficiency of the facility operation for processing even a very small amount of water. Inactive commissioning confirmed that the integrated facility is capable of processing carbon flakes/dust contaminated with tritium.

3. ACTIVE COMMISSIONING OF THE FACILITY.

A 1g sample of flake material removed from the JET vessel sub divertor region was dispensed into a ceramic crucible. The flakes/dust loaded into the crucible was covered with the fine mesh made of

stainless steel. This plug was exposed to temperature of 1120K under atmosphere of Ar/O_2 prior to being used for the active commissioning. The open end of the crucible was then sealed with paraffin film before the crucible was loaded into the plastic containers for transferring from the Beryllium glovebox to the Flakes/Dust glovebox. The crucible with the flakes and mesh were then loaded into the Thermal Gravimeter.

The first step of the process was to transfer water, which might be adsorbed on the surface of the CFC flakes/dust, to the cold trap. This was carried out by connecting the thermal gravimeter to the cold trap and heating the flakes/dust up to 570K. The thermal gravimeter was purged with argon at flow rate of 50cm3/min through the cold trap to the reservoir. The total mass loss recorded by the thermal gravimeter was 26.5mg, which is equivalent to 2.7wt% of water content in the sample of CFC flakes/dust.

The highest reading (263.64K) of the humidity meter was observed after first 9 minutes at flakes temperature of 570K. To collect the residual water remaining in the manifold after this stage of the water transfer from the flakes/dust to the cold trap, the gas in the reservoir was re-circulated through the cold trap. This resulted in reduction of the humidity meter reading down to the Dew point of 236.15K.

The thermal gravimeter was then purged with Ar/O2 mixture (in ratio 1:1) at flow rate of around 35cm3/min through the cold trap to the reservoir. The cold trap was maintained at temperature around 180K. This step took 3 days. For the first two day oxidation process was carried out at temperature of 1120K. For the last day the operation temperature was increased to 1270K. The humidity meter showed increase of the Dew point from 237.75K before start of oxidation to 272.55K once operation temperature of 1120K was achieved. The Dew point then quickly fell to below 246K. The averaged rates of mass change of 1.0mg/min, 0.20mg/min and 0.03mg/min were observed for days 1, 2 and 3, respectively. The average rate of mass change for the whole duration of step 3 was 0.47mg/min. This value is considerably smaller than that observed for CFC dust oxidation in the functional tests. In addition a mass gain, not mass loss as expected from the oxidation of carbon, was observed. The total mass gain was 338.1mg, which is equivalent to 35.4wt% of mass of the original sample after removal of adsorbed water. After completion of the active commissioning the thermal gravimeter was opened for investigation of the solid residue left in the crucible. The picture showing that residue is given in Fig.3. This solid residue will be sent for analysis of its chemical composition together with a sample of the original flakes/dust.

The kinetic curve of the sample mass change as a result of contact with oxygen is depicted in Fig.4. The mass change started with mass loss at temperature just above 670K. This process continued till temperature reached 920K. At temperature above 920K a mass gain, not mass loss, was evident.

The mass loss observed at temperature below 920K can be attributed to oxidation of carbon dust. Mass gain observed for temperature above 920K can be attributed to oxidation of metal present in the crucible. Firstly, the stainless steel of the plug inserted to the crucible could react with oxygen. However, the plug was used in other runs at the same temperature and under atmosphere of Ar/O2 mixture.

Therefore it is reasonable to assume that stainless steel was already, at least partially, oxidized. Total mass gain for complete oxidation of stainless steel in plug of initial mass of 120mg should be about 50mg, which is much smaller than that observed during oxidation of the sample of tritiated flakes/dust. Source of the mass gain might be beryllium, which could be present in the sample of flakes/dust. Beryllium is evaporated, on a regular basis, on CFC tiles inside the JET vacuum vessel and is then sputtered from the tiles together with the carbon. Presence of beryllium was observed in the flakes/dust removed from the vacuum vessel after DTE1 [4,5]. The operation temperature of the flakes/dust oxidation was sufficient for beryllium to react with oxygen [6]. To provide the mass gain of 338.1mg, the sample of flakes/dust used for active commissioning would need to contain 190mg of beryllium. However it shall be noted that reaction of carbon with oxygen should occur at the same time as the oxidation of metal. The mass gain shown in Fig.4 therefore presents the apparent effect of two possible processes that work against each other in respect to mass change to be detected by the thermal gravimeter. Therefore mass gain designated to beryllium oxidation is larger than the 338.1mg mass gain recorded . Provisional evaluation based on the assumption that the sample of flakes/dust contained beryllium, which was oxidized and produced BeO, shows that content of beryllium in the flakes/dust used for the active commissioning tests might be in excess of 50%wt.

Gas chromatographic analysis of the gas collected in the reservoir at this step showed the presence of carbon dioxide and carbon monoxide in ratio 2.5:1. This indicates that oxidation of carbon occurred preferably with formation of CO_2. Gaseous hydrocarbons were also detected but with concentrations of 2 to 3 orders of magnitude smaller than that of carbon oxides.

The water collected in the cold trap was then processed. Firstly carbon dioxide was released from the cold trap to the reservoir and the reservoir evacuated. Argon was then circulated through the cold trap, reactor R4, with SAES getter ST909 at 770K, and reactor R5 with palladium sorbent at room temperature. At the last stage the cold trap was gradually heated to 470K and the palladium sorbent cooled to 140K and gas circulation continued until the dew point fell below 230K

To collect residual hydrogen from the gas at the end of the Step 4, the cold trap was isolated and argon was re-circulated through the reservoir, reactor R4 with SAES getter ST909, at 770K, and the reactor R5 with the palladium sorbent cooled to 140K. Reading of the humidity meter showed a reduction of Dew point below 230K.

After collecting the hydrogen, from tritiated water decomposition, the reactor R5 and reservoir were pumped down. The temperature of the reactor R5 was maintained 140K during the pumping. Hydrogen absorbed by the palladium sorbent was then desorbed by heating reactor R5 to 570K and pumping the gas to the reservoir. The total amount of molecular hydrogen in the reservoir was 1.65Nl. Analysis of isotopic composition of this hydrogen showed 97.517% of protium, 2.473% of deuterium and 0.01% of tritium. Tritium activity removed from the sample of flakes/dust was therefore 15.8GBq. This gives a tritium concentration in the flakes/dust of 16.09GBq/g. Total yield of hydrogen isotopes from processing the flakes/dust is 0.075mol(Q2)/g. The yield related to the water removed from the sample of flakes/dust by thermal desorption is 1.5•10-3mol/g, which presents only 2.2% of total yield

of hydrogen isotopes. The specific tritium activity of 16.09GBq/g in the flakes/dust removed from the JET vacuum vessel in 2002 is approximately 70 times less than the 1.1TBq/g measured for the flakes/dust removed from the vessel after DTE1.

CONCLUSIONS

The new facility, designed and assembled at the JET AGHS, was successfully commissioned for tritium recovery from carbon flakes/dust removed from the JET vessel. The facility is functional and allows tritium recovery from tritiated carbon materials. Full scale processing requires employment of reactor to accommodate a large batch of the flakes/dust.

During flakes/dust oxidation tests in active commissioning it was discovered that the flakes/dust contain some material that reacts with oxygen forming solid oxides. This solid residue, and a sample of original flakes/dust, will be analysed to determine their chemical composition.

The successful commissioning of this facility demonstrates the ability to detritiate flake and dust material generated from a tokomak operating with tritium and a carbon first wall. In future material will be collected from within the JET vacuum vessel following operations with a Beryllium first wall. The processing of both types of material is directly relevant to the operation of ITER and future decommissioning activities.

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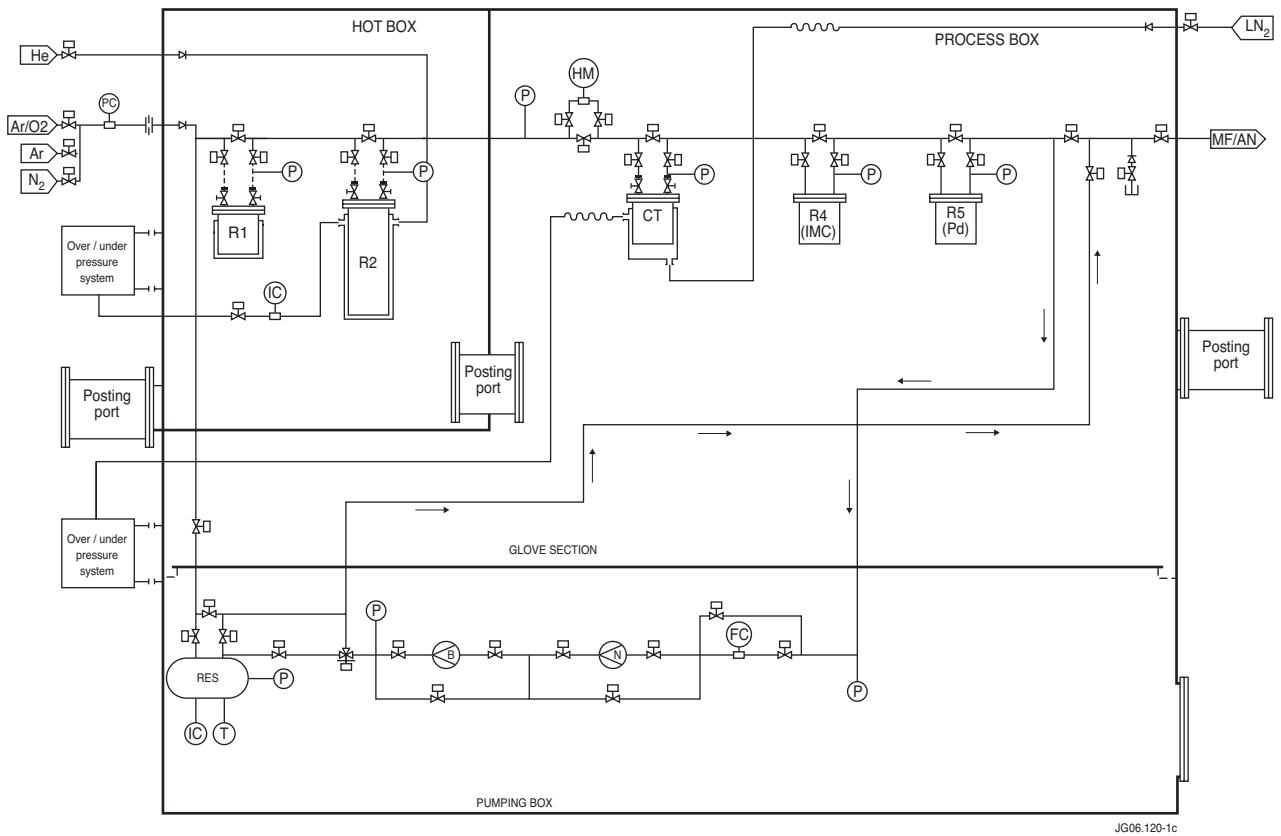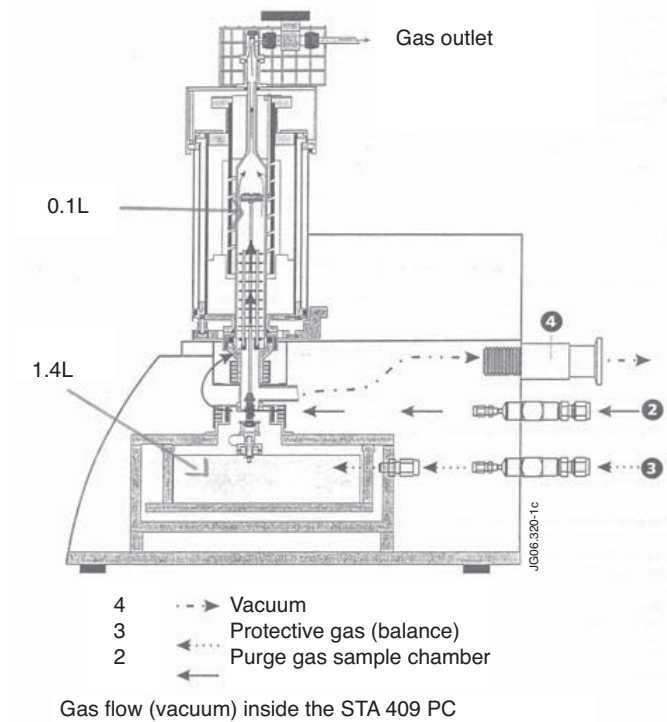
Figure 1: The facility flow diagram [1].

4	· - ▸	Vacuum
3	◄────	Protective gas (balance)
2	◄····	Purge gas sample chamber

Gas flow (vacuum) inside the STA 409 PC

Figure 2: The schematic of thermal gravimeter.

Figure 3: Picture of solid residue left after oxidation of the CFC flakes/dust
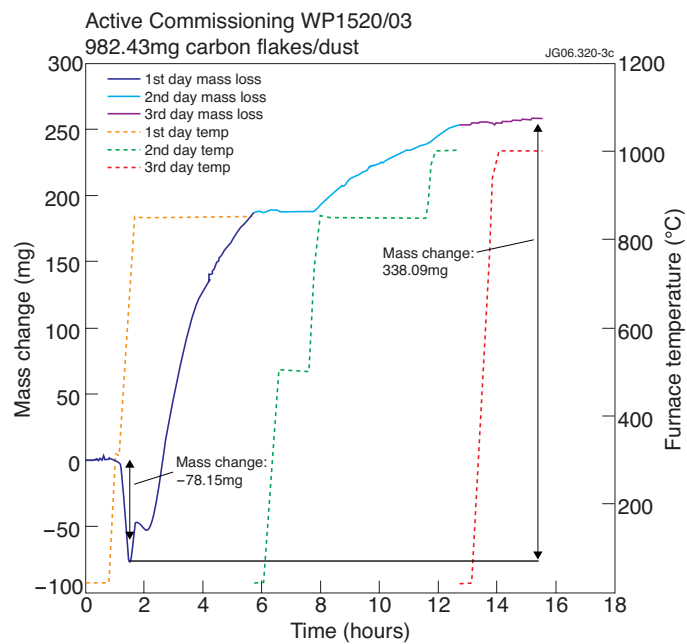

Figure 4: Kinetic curve of the sample mass change during Stage 3 of the active commissioning.