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Techniques for Tritium Recovery from Carbon Flakes and Dust at the JET Active Gas Handling System

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ABSTRACT

Detritiation of highly tritium contaminated carbon and metal material used as first wall armour is a key issue for fusion machines like JET and ITER. Re-deposited carbon and hydrogen in the form of flakes and dust can lead to a build-up of the tritium inventory and therefore this material must be removed and processed. The high tritium concentration of the flake and dust material collected from the JET vacuum vessel makes it unsuitable for direct waste disposal without detritiation. A dedicated facility to process the tritiated carbon flake material and recover the tritium has been designed and built. In several test runs active material was successfully processed and detritiated in the new facility. Samples containing only carbon and hydrogen isotopes have been completely oxidized without any residue. Samples containing metallic impurities, e.g. beryllium, required longer processing times and adjusted process parameter, resulted in an oxide residue. In order to simulate in-vessel and ex-vessel detritiation techniques, the detritiation of a carbon flake sample by isotopic exchange in a hydrogen atmosphere was investigated.

1. INTRODUCTION

Carbon-Fibre reinforced Carbon (CFC) tiles are extensively used in the JET tokamak as plasma facing first wall armour due to their excellent thermo-mechanical properties. Carbon is eroded from the areas of high power loading and is co-deposited together with hydrogen isotopes in areas of lower power density [1]. Eventually the deposition produces flakes that are only loosely attached to the tile surfaces. In tritium operations this can lead to a build-up of a tritium inventory in form of carbon flakes/dust, which subsequently have to be carefully removed and processed. In several shutdown campaigns flakes and dust material have been remotely removed from the JET vacuum vessel. The high tritium concentration of 1.2 TBq/g per gram of flakes and the very high out-gassing rates exhibited by the flakes/dust material make them unsuitable for direct waste disposal and require detritiation [2].

2. DESCRIPTION OF THE FACILITY

A facility was built within the tritium plant of the Active Gas Handling System (AGHS) at JET [3]. The main purpose of the facility is to reduce the activity of the flake material to allow its disposal as Low Level Waste (LLW) and within this step to recover the tritium and recycle it within the JET tritium plant. The LLW limit for tritium in the UK is 12 kBq/g of material. In the first test runs the process parameters were determined and optimised and provided important information for the planned larger scale detritiation facility. The process is based on the complete oxidation of the flakes/dust at high temperature under an atmosphere which contains oxygen. The basic plant and process has been described in detail in a previous paper [4].

In brief, the following steps are performed:

- 1) Remove water from the carbon sample by heating the flake sample under argon atmosphere to 570K and collect the water in a Cold Trap (CT)
- 2) Oxidation of the tritiated carbon sample under O₂/Ar (50%/50%) atmosphere at 1120K

- 3) Decomposition of water collected in the cold trap and in reaction with metal (R4, SAES St909 Getter $Zr(Fe_{0.5}Mn_{0.5})_2$) and finally hydrogen absorption on palladium sorbent (R5, 20wt% Pd/Al₂O₃)

Palladium is a good hydrogen absorbent which releases the tritium readily when heated up. The released tritium can be transferred to the AGHS sub-systems for processing and storage. A commercial thermal gravimeter (STA 409 PC LUXX, Netzsch) was used to heat the sample. The gravimeter includes a high temperature furnace, a very sensitive balance (resolution 2µg) and can be purged with gas mixtures. The maximum sample load is limited by the volume of the crucible to 2 g.

3. EXPERIMENTS

In a previous paper the inactive and first active run was described in detail [3] and only the key points are mentioned here.

3.1. INACTIVE RUN

The detritiation facility was first commissioned using inactive material and all individual components were tested before assembly on the plant. A tritium free experiment was carried out to confirm and establish the system parameters. A sample of 633.4 mg in-active CFC dust was prepared and 283.0 mg water was added. Water is expected to be released from the active samples as a result of (i) water adsorption during storage in an air atmosphere [2] and (ii) when the H-isotopes associated with the active dust are oxidized under 50% oxygen in argon atmosphere. During the inactive run the three reactors, Cold Trap (CT), intermetallic compound (R4) and palladium sorbent (R5), were in operation. The sample was completely oxidized by heating to 1120K in a 50% oxygen in argon atmosphere. During this step more than 90% of the water was collected in the cold trap and the oxidation rate was estimated to be 3.89 mg/min. No residue was left

3.2. FIRST ACTIVE RUN

Flake and dust material from the JET Machine was collected with a vacuum cleaner during the 2001 shutdown and stored in a cyclone pot. This material was formed in the JET vacuum vessel during tritium and subsequent deuterium operations. Approximately 1.0g of this material was prepared for full oxidation. After the detritiation experiment it was found that the sample contained a high amount of beryllium (33%). Beryllium evaporation is regularly used to condition the Torus first wall for plasma operation. After heating the sample at 570K under an argon atmosphere a mass loss of 2.7 wt% water was observed. After cooling down the sample was heated under a 50% oxygen in argon atmosphere where further mass loss was observed. This was due to the oxidation of carbon material into carbon oxides. The mass loss continued until the temperature of 920K was reached. In the temperature range from 920K to 1120K a mass gain was observed. This was caused by oxidation of metal contained in the sample. A mass gain of 35.4 % of the initial sample mass was measured. Gas chromatography analyses of the palladium sorbent reactor gas were carried out as the last step of the

process. The composition of the gas was 97.52% H₂, 2.47% D₂, and 0.01% T₂. The total tritium activity recovered from the flakes/dust sample was calculated via PVT-c measurements to be 16.1 GBq/g. Table I shows the elemental composition found after chemical analysis of the solid residue and a sample retrieve from the cyclone pot. The solid residue had a tritium concentration of 790 kBq/g and contained 20.3% Be and 36.0% BeO. Taking into account the tritium activity of the flakes/dust sample (16.1 GBq/g), this gives a Decontamination Factor of about 20400. The first active test showed that a low active sample residue is above the limit of 12 kBq/g and therefore still requires disposal as Intermediate Level Waste.

3.3. SECOND ACTIVE RUN

During a second active run a sample of 819 mg of flake/dust material was prepared from a poloidal limiter tile from the first Deuterium Tritium Experiment (DTE1) and the Trace Tritium Experiment (TTE). The initial tritium activity was 1.16 MBq/g and the sample had no beryllium content. Due to the small amount of water adsorption found in the first run and the small tritium activity in this sample less water was expected and therefore the 3 reactors of the facility were not used (Fig.1, CT, R4, R5). All the combustion gases were collected in the reservoir and the water vapour content was continually monitored with a humidity sensor (HM). The aim was to achieve complete oxidation similar to the inactive test to confirm the process parameters with a sample without metallic content. Purging with 35ml/min of the 50% oxygen/argon mixture through the gravimeter and humidity sensor, the highest humidity signal of -25.78°C (DP) was measured at a temperature of 1000K. But overall during the oxidation of the flake/dust sample at 1123K the humidity signal was stable at around -29°C DP, this is also an indication that the sample had a very low tritium activity. As expected, with increasing temperature a mass loss occurred due to the oxidation of the carbon flakes (Fig.2). After 4.5 hours the whole sample was fully oxidized and this is illustrated in Fig.2 by a flat signal from the thermogramme. The total mass loss was 817.6mg. This gives an oxidation rate of 3.0mg/min or 182mg/h. The difference from the initial mass of 1.4 mg is due to measurements under different gases.

It was not possible to detect any significant amount of water. 15.9 L of exhaust gas were collected in the reservoir (RES). The ionisation chamber in the reservoir detected no increase in the activity (background level was 3.23GBq/m³). The gas chromatographic and mass spectrometer (RGA) analyses yielded 54.3% Ar, 24.9% O₂, 0.22% CO, and 17.9% CO₂ with a ratio of 1/0.5/0.004/0.3 for Ar/O₂/CO/CO₂ respectively. However it was not possible to measure any H, D or T. The second active test demonstrated that it is possible to fully oxidize CFC flakes/dust material with no metal content using 50% oxygen atmosphere at 1123K and the gas chromatographic analyses confirmed that the produced gases are mostly carbon oxides.

3.4. THIRD ACTIVE RUN

In order to compare possible in-vessel and ex-vessel detritiation techniques, the detritiation of a carbon flake sample using isotopic exchange was investigated. A sample of 578mg of flakes/dust material

from the same pot as the first active run was introduced into the thermal gravimeter. The tritium activity was found from the first active test i.e. 16.1GBq/g calculated via PVT-c measurement. It is worth mentioning that the sample contained a high percentage of beryllium as determined by the chemical analysis (Table I). As shown from the experience from the first active run, only a small amount of water was expected, therefore none of the three reactors (Fig 1, CT, R4, R5) were in operation. The aim of this test was to investigate the detritiation effect through isotopic exchange and not by oxidation. Therefore the gravimeter was filled with 4% hydrogen in argon mixture and a purge route with a flow rate of 35 ml/min was established through the gravimeter passing over the sample, the humidity sensor and finally collected in the reservoir. In this first step any water was transferred from

the sample at 590K for 6 hours into the reservoir. During the heating of the gravimeter to 590K the humidity sensor showed a signal of -34.2°C DP . The outlet pressure of the gravimeter was 998 mbar. After ten minutes at 590K the humidity sensor signal increased to $-18.34^{\circ}\text{C DP}$. This was a clear indication that water was released. After a further 160 min the humidity signal recovered to the value of -34.2°C DP . Purging continued for a further 180 min to ensure that all water was released. This resulted in a total time of 6 hours at a temperature of 590K. 16 L of gas was collected in the reservoir having an activity of 148 MBq. Results from the gravimeter signal revealed a water mass loss of 9.8 mg or $5.45\text{E-}4$ mol. Related to the initial mass this amounts to $9.4\text{ E-}4$ mol/g or 1.7%.

The second step was to re-circulate 4% hydrogen in argon mixture at 590K for 8 hours over the sample to investigate whether there is a continuing isotopic exchange effect. For this the reservoir was filled with 540mbar of gas and a gas flow rate of 100 ml/min was established. The humidity sensor showed no release of water during the 8 hours re-circulation. 19.8 L of gas with an activity of 0.1GBq was collected in this step. No free water was left from the first step, which meant the removal of activity came through off-gassing of the flakes or isotopic exchange. On completion of recirculation, the system was evacuated and the gravimeter filled with argon. The flakes/dust residue was then sent for chemical analysis to Waste Management Technology (WMT) Winfrith, the results of which are shown in Table I. After each step the gas in the reservoir was directly discharged to the Exhaust Detritiation System (EDS) of the AGHS. An ionisation chamber reading was taken before and after the discharge and gave a recovery rate of 2.80% of Tritium. The purged sample after the third run has a very similar tritium concentration to measurements of the activity of material remaining in the cyclone pot by the chemical laboratory (2.66 GBq/g and 2.74 GBq/g respectively). The outcome of this test was that no efficient detritiation factor could be determined after recirculation using 4 % hydrogen in argon.

SUMMARY AND CONCLUSIONS

The new detritiation facility at JET AGHS is functional and demonstrates the ability to detritiate flake and dust material generated during tokamak operations using tritium gas and a carbon first wall. It was also discovered that some batches of carbon flakes/dust can contain substantial amounts of

beryllium. Carbon flakes/dust with no metallic content can be fully oxidized without any residue under 50% oxygen/argon atmosphere at 1120K and therefore no further disposal is required. Oxidation of samples with high metal contents have shown a mass increase at high temperature (above 920 K) due to the formation of metal oxides. These metal oxides have a residual tritium inventory and therefore required disposal as Intermediate Level waste, but nevertheless the detritiation factor is 2×10^4 . It was found that detritiation of the material from those batches required a much longer time than that foreseen for pure carbon materials. The experience gained from the test runs indicates a need to optimise the procedure for detritiation of the carbon flakes/dust in order to process efficiently the large quantity of material which is expected after the eventual JET decommissioning. This is an important step for future decommissioning activities and directly relevant for ITER. Isotopic exchange of flakes and dust material at 590K under 4 % hydrogen in argon atmosphere is not efficient. The analyses have shown that the flakes/dust sample collected from the sub-divertor region contains some beryllium (~9 %) which after oxidation represents ~33% the residue mass. The determined tritium activity from the first active run was 16.1 GBq/g whereas the chemical analysis yielded 2.7 GBq/g. The discrepancy results mainly from the use of different analytical techniques. A further explanation could also be the nonhomogeneity of the sample.

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	1 st active run, Residue	Cyclone pot	3 rd active run 4% H2 in Ar
Analysis date	23/11/06	21/06/07	21/06/07
Units (unless otherwise stated)	mg/g	mg/g	mg
Tritium Activity	790 ± 40 kBq/g	2.74 ± 0.13 GBq/g	2.66 ± 0.11 GBq/g
Carbides as C		0.2 ± 0.1	0.6 ± 0.1
Carbon	103 ± 12	603 ± 41	812 ± 67
Ag	< 0.20	< 0.1	< 0.1
Al	180	54.2	16.9
Ba	< 0.71	< 0.1	< 0.1
Be	203	89.8	92.0
BeO	360	6.0	1.1
Ca	4.9	2.8	2.2
Cd	< 0.12	< 0.1	< 0.1
Co	0.05	< 0.1	< 0.1
Cr	5.8	27.1	15.6
Cu	1.3	3.4	1.9
Fe	19.2	25	7.6
K	< 1.1	< 1.6	< 1.1
Li	< 0.05	< 0.1	< 0.1
Mg	1.8	2.5	0.5
Mn	0.65	1.3	0.6
Mo	0.51	< 0.3	< 0.3
Na	0.98	< 0.3	< 0.2
Ni	44.9	108	66.8
Sr	< 0.03	< 0.1	< 0.1
Ti	0.56	0.2	0.2
V	1.6	0.4	0.5
Zn	0.93	0.7	0.3

Table I: Chemical analyses of flakes/dust samples

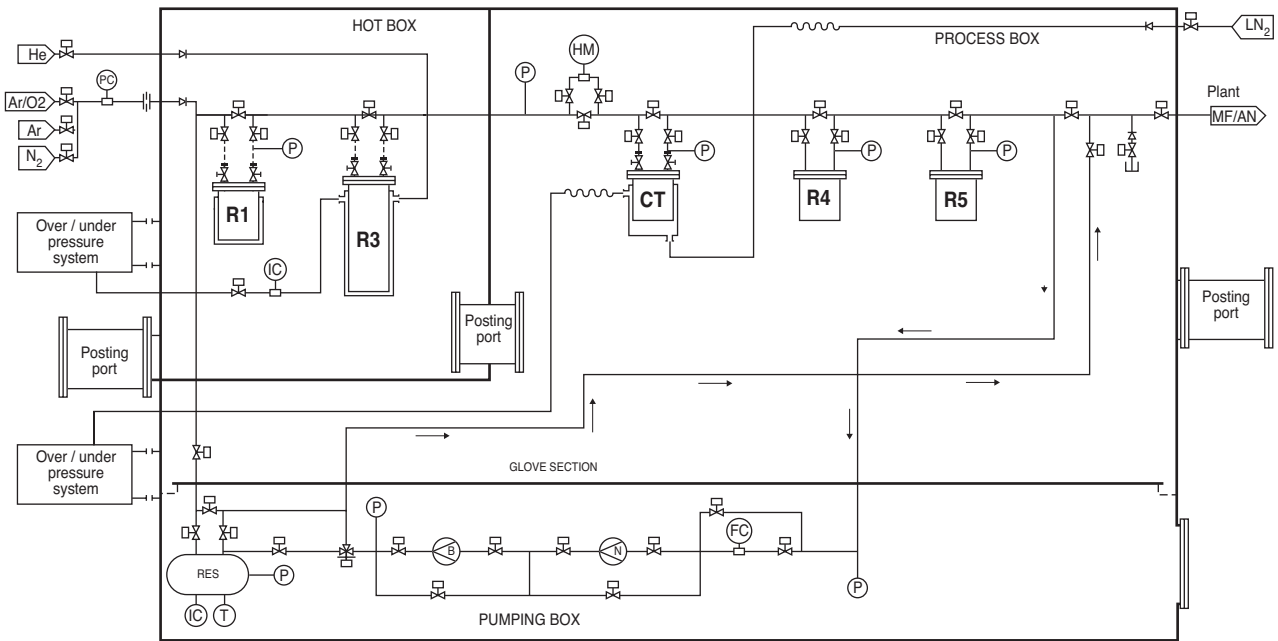


Figure 1: Simplified low diagram of the detritiation facility:
R1: Furnace, HM: Humidity sensor, CT: Cold trap, R4: Intermetallic compound
R5: Palladium sorbent, RES: Reservoir, IC: Ionisation chamber

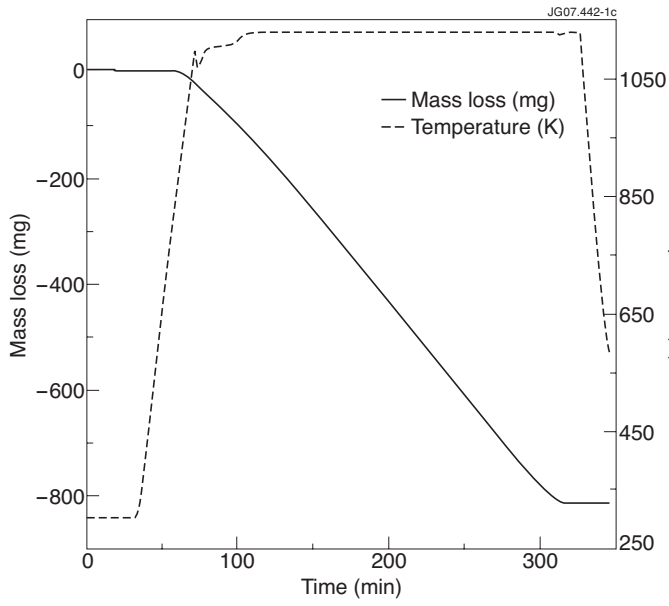


Figure 2: Record of thermal gravimeter signal during oxidizing process for the second active test.

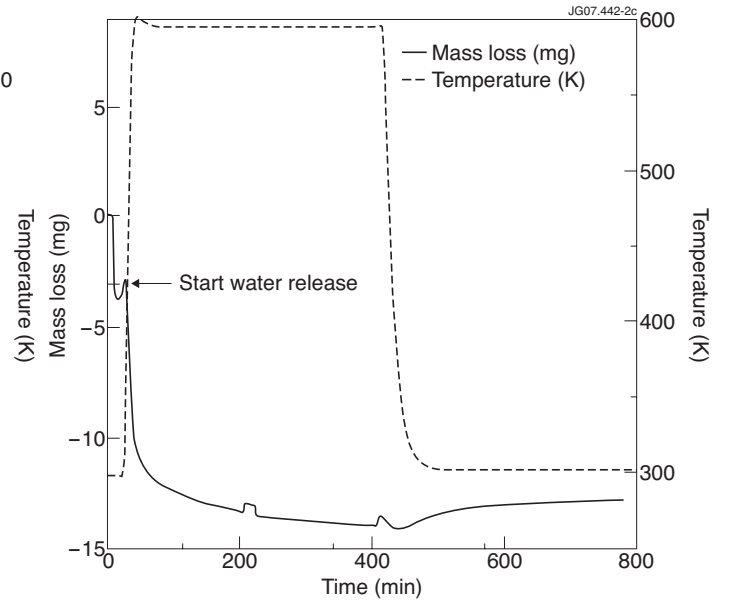


Figure 3: Record of thermal gravimeter signal during water release for the third active test.