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ABSTRACT

Nuclear waste management has to be taken into account for fusion machine in tritium experimentations. Soft housekeeping waste is produced during both operating and dismantling phases and is contaminated by tritium under reduced (HT) and oxidized (HTO) forms.

At CEA Cadarache, a lab-scaled facility has been built for soft housekeeping detritiation.

Moreover, the tritiated gas exhausted from the process described above is foreseen to be treated by a tubular Pd-Ag membrane reactor, for gaseous tritium recovery. As this membrane reactor uses hydrogen as swamping gas, one has to account for the compatibility towards explosive hazard.

Then, this work presents a double objective. A first study is presented in order to identify the best conditions for the declassification of soft housekeeping waste, without tritium recovery. Experiments carried out at 120°C are not efficient enough and do not allow one to choose the most efficient carrier gas. Some other tests are being currently performed at higher temperatures (150°C). Moreover, due to safety issues, the use of air has to be avoided during membrane reactor implementation phase. Preliminary results obtained with hydrogen hazard-free carrier gases are also presented.

1. INTRODUCTION

Soft housekeeping waste is produced during both operating and dismantling phases of fusion reactor and is contaminated by tritium under reduced (HT) and oxidized (HTO) forms. For example, concerning ITER project, the production of 1600 to 3800 tons of housekeeping and process waste is foreseen and can be split as follows [1]:

- 20% very low level waste
- 75% short lived intermediate level waste
- 5% long lived intermediate level waste

The higher the waste class, the more expensive and stringent the storage [2]. Moreover, tritiated waste induces the outgassing particularity compared to usual nuclear waste. Then, the implementation of a detritiation facility could be interesting to declassify this kind of waste.

In this work, the lab-scaled detritiation facility designed by CEA Cadarache is presented as well as its future coupling with a catalytic membrane reactor from ENEA Frascati [3].

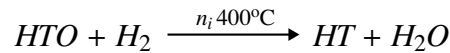
Consequently, this study consists in two main parts: first of all, the technical feasibility study is performed with different carrier gas at different temperatures and flows in order to optimize the experimental conditions for detritiation efficiency. Moreover, in order to avoid the explosive hazard due to the use of pure hydrogen as a swamping gas in the membrane reactor, the study of the influence of the type of carrier gas has also been carried out. Then, the most appropriate carrier gas, presenting the best compromise between safety and efficiency, will be determined.

2. PRESENTATION OF THE EXPERIMENTAL FACILITIES.

The housekeeping samples, produced during recent ITER Like Wall (ILW) shutdown, from work on divertor refurbishment/upgrade, are milled and placed in a glass reactor heated within a tubular oven up to a temperature lower than the housekeeping melting point. The carrier gas is then injected

in the detritiation reactor and, due to the cumulated actions of temperature and gas feed flow, the tritium is removed and finally trapped by a MARC 7000® bubbling system (see Fig.1) and the tritium activity is determined by liquid scintillation.

Moreover, the tritiated gas exhausted from the process, instead going into the liquid traps, is foreseen to be treated by a tubular Pd-Ag membrane reactor, as shown in Fig.2. In this way, the gaseous tritium (HT) recovery occurs, as HT is potentially reusable in fusion reactors fuel cycle. This catalytic membrane reactor consists of two parts [4-12]. The inner part of the membrane tube (so-called lumen side) receives the exhaust gas to be treated while hydrogen stream is injected in the inter-annular space (between the membrane and the reactor shell, so-called shell side) in a counter-current mode. The Pd-Ag membrane is known to be only permeable to hydrogen and its isotopes (deuterium and tritium), allowing hydrogen to permeate in the lumen side. The heating of the membrane up to 400°C and the presence of a Ni-based catalyst allow the isotopic exchange:



Tritiated gas HT formed by isotope swamping also permeates the membrane to be finally recovered in the shell side of the membrane reactor. Within the scope of the study, for analytic and waste management purposes, the gaseous tritium is oxidized in an oven containing a cartridge of copper oxide and the tritiated water formed is frozen in a liquid nitrogen cold trap.

3. RESULTS AND DISCUSSION.

The experiments whose results are presented here have been carried out during 3 days and 6 hours a day effective working time. In order to clarify the plots, the X-axis represents the effective experimental time. The night-gaps between two experimental days have not been considered in the graphs.

The results presented here only concern the detritiation part of the study (i.e. without the membrane reactor coupling). The experiments have been carried out under several atmospheres:

- Argon + 5% H₂
- Argon + 2% O₂
- Argon + 25% and 40% H₂O
- Air

3.1 SAFETY CONSIDERATIONS.

Considering that the catalytic membrane reactor uses hydrogen as a swamping gas and that the efficiency of the isotopic exchange depends on the temperature (the optimal temperature has been determined at 400°C), it appears essential to assess the explosive hazard between hydrogen and the carrier gases listed above.

As an explosion can occur only in presence of a fuel, a combustive gas and an ignition source, the gases containing only argon and water (neither fuel nor combustive gases) are not concerned here.

Figure 3 shows the flammability limits of a ternary nitrogen/oxygen/hydrogen mixture. Point 0 on Fig.3 is a virtual mixture aiming at the diagram understanding. It would represent a mixture composed by 6% O₂, 2% H₂ and 92% N₂.

Point I represents an original 5% concentration of H₂ in nitrogen (simulating here argon). It is remarkable to notice that if an oxygen entrance occurs (represented by the dotted line), the flammable region can be crossed from a temperature approx. 150°C. On the other hand, the full line represents the behaviour of a ternary mixture resulting from hydrogen entrance in an original 2% O₂ in nitrogen (point II). In this case, up to 400°C (corresponding to the working temperature of the membrane), the flammable region is never crossed.

Nevertheless, it is important to keep in mind that the interaction between the Ar-O₂^{2%} and H₂ occurs under the normal mode of the experiment. On the other hand, a mixture Ar-H₂^{5%} with air would only occur under an incidental mode, if the system is not air-tight.

Thus, air excluded, all other gases are hazard-free towards hydrogen under normal mode.

3.2 DESCRIPTION OF OPERATING CONDITIONS.

The samples used in this study have been provided by JET fusion machine. The initial mass activity of the samples has been assessed to 20kBq/g, i.e. very close to the British ILW/LLW threshold. However, due to the expected heterogeneity of the initial activity of the samples, the residual activity is always measured by liquid scintillation at the end of the experiment under the operating mode schematized on Fig.4.

First of all, a well-known housekeeping quantity (between 12 and 15 g) is placed in the glass reactor. As the samples may contain some beryllium traces, the sample preparation is made in a dedicated room for Be handling. Then, the air-tight reactor is placed in the oven and a leak test is performed by reading of the pressure variation.

The temperature of the oven is regulated and the carrier gas is injected in the system. The demineralised water in MARC 7000® bubblers is changed every one or two hours and a given quantity of this recovered water is analysed by liquid scintillation.

At the end of the experiment, the activity of the residue is also measured by liquid scintillation after calcination in a Hastelloy reactor placed in an oven heated up to 500°C under air for 3 hours and the tritium is trapped in a MARC 7000® device (Fig.4).

Finally, the detritiation yield is calculated using the following equation:

$$Y(t) = 100 \times \frac{\sum_{j=1}^n a_i}{\sum_{j=1}^N a_i + a_{res}} \quad (2)$$

With $Y(t)$: the detritiation yield (%).

a_i : ³H removed activity as a function of time (Bq)

a_{res} : ³H activity of the residue (Bq)

n_i : sampling number at t-time (dimensionless)

N : sampling number at the end of the experiment (dimensionless)

As the samples come from JET, it has been decided to fix the threshold between Intermediate Level Waste (ILW) and Low Level Waste (LLW) according to the British regulation, i.e. a residual activity of 12 kBq per gram of waste [13].

3.3 INFLUENCE OF THE NATURE OF THE CARRIER GAS.

In order to identify the respective efficiencies of the carrier gases, several atmospheres have been tested:

- Low oxidative (Ar + 2% O₂)
- Low reductive (Ar + 5% H₂)
- Intermediate oxidative (air)
- Humid gases (Ar + x% H₂O, x = 25 and 40)

The gas feed flow has been fixed at 300 NmL/min (“N” represents a gas flow normalised at 273K and 1 atm.) which corresponds to a normal workflow for the membrane reactor. In order to avoid the housekeeping burning during the experiment, the temperature has been fixed at 120°C.

Figure 5 shows the behaviour of the detritiation yield of soft housekeeping as a function of the effective experimental time. First of all, one can observe that none of the 5 plots shown on Fig. 5 reaches a plateau and all seems to obey a quasi linear regime which tends to demonstrate that the experimental time is not optimized under these experimental conditions.

Moreover, on the one hand, the dark dots (diamonds, circles, triangles and squares) of the graph establishes that the detritiation yield does not exceed 8 % whatever the carrier gases employed at 120°C during 18 h effective experimental time. The amount of water in argon does not seem to have an influence on the detritiation yield (see dark circles and triangles on Fig.5).

Table 1 summarises the experiments carried out at 120°C and 300NmL/min carrier gas feed flow. First of all, it is visible that the expected heterogeneity of initial activities of the samples is accurate. Indeed, the initial activities vary from 10 to 30 kBq/g. It is also to notice that none of the Ar-based carrier gases allows the declassification to LLW after 18h-experiment. Furthermore, the initial activity of the sample detritiated under air (10 kBq/g) being slightly lower than LLW/ILW threshold, it is not possible to conclude that these experimental conditions are sufficient to declassify the waste to LLW.

3.4 INFLUENCE OF THE TEMPERATURE.

The previous section has shown that the initial experimental conditions (120°C and 300NmL/min) are too soft to declassify the housekeeping waste. Thus, in order to optimize the operating conditions and to avoid the combustion of the housekeeping sample, experiments have been carried out at 150°C. At this moment of the study, we are only able to compare the influence of temperature with Ar+ 2% O₂ and Ar + 25% H₂O, the experiments using the other carrier gases are currently being carried out.

Figure 6 establishes the comparison of the detritiation yields as a function of time for two different carrier gases at 120°C (dark dots) and 150°C (light dots). It is visible that a reasonable temperature increase of 30°C leads to a considerable increase of the detritiation yield. Indeed, for both Ar-^{2%}O₂

(diamonds) and Ar-^{25%}H₂O (triangles), one can notice that the detritiation yield increases nearly fivefold (5.0 to 23.6 % for Ar-^{25%}H₂O, 7.6 to 56.1% for Ar-^{2%}O₂).

It is also interesting to notice that a slope break occurs after 15h (0.6 day) for 150°C Ar+ 25%H₂O experiment and seems to enlighten the beginning of equilibrium between tritium removal and residual activity of the sample.

As Table 1 does, the residual activities listed in Table 2 show that 15h are enough to declassify to LLW the soft housekeeping treated by Ar-^{2%}O₂ at 150°C. Indeed, its residual activity is much lower than the ILW-LLW threshold. On the contrary, using Ar-^{25%}H₂O at 150°C does not allow one to declassify the waste yet. Nevertheless, if the behaviour of this experiment remains the same as its end (yield increase of approx 0.75% per hour), the declassification to LLW may be possible after extra 13h experimental time.

3.5 INFLUENCE OF THE CARRIER GAS FLOW.

This study is useful for two different reasons. First of all, determining the influence of the residence time on the detritiation yield will allow one to assess the flow within an industrial detritiation facility and then the cost of the gas consumption, especially coupled with a membrane reactor, where air can not be used for safety reasons. Then, this study may also allow one to determine the contamination mode of housekeeping by tritium. Indeed, a detritiation yield increase with carrier gas feed flow would indicate that the sample is contaminated by tritium on its surface while a constant detritiation yield with gas flow increase would show that the most part of tritium has penetrated the waste.

At this time of the study, only experiments with air have been carried out. More experiments performed with other carrier gases are foreseen to confirm the following conclusions which can be only considered, up to now, as tendencies.

Figure 7 shows the behaviour of the detritiation yield as a function of time for two different air flows. It is visible that the beginning of the curves are quasi superposed for the 7 first hours (0.3 day), which corresponds to the first day of experimentation. It is also remarkable to observe that the following black diamond shows a jump, probably due to a tritium degassing during the night. This phenomenon occurs again during the second night stop (see the next-to-last black diamond on Fig. 7). This is only visible on the 450 NmL/min experiment because that one has been carried out before the 300 NmL/min one and the natural tritium outgassing occurs much more easily with fresh samples. Nevertheless, these two points excluded, the slope is higher for higher feed flows (0.32%/h for 300 NmL/min, 0.46%/h for 450 NmL/min).

Despite that, Fig. 7 establishes that the higher the feed flow, the higher the detritiation yield. Furthermore, the gap between the detritiation yields obtained using these two feed flows tend to demonstrate that the tritium contamination is mainly on the waste surface.

Table 3 sums up the results obtained for the detritiation of soft housekeepings under air at different gas feed flows. As already mentioned in section 3.3, due to the weak initial activities, it is impossible to conclude about the possibility to declassify the waste treated at 300 NmL/min. This conclusion is also valid for a 450 NmL/min-gas feed flow. Indeed, the initial activity of the sample is clearly too close to the ILW/LLW threshold.

CONCLUSION AND PROSPECTS

In this study, the detritiation efficiencies of low activity-soft housekeeping by different carrier gases at different temperatures and flows have been assessed. In the future, it appears essential to perform the same study on much higher sample amounts and much higher tritium contaminated housekeeping waste as expected during the ITER project.

The initial goal of this study was to demonstrate the technical feasibility of the detritiation of soft housekeepings under soft conditions, i.e. without housekeeping combustion. After 18h-experiments, the 120°C-initial reference temperature is too weak to declassify weakly contaminated soft housekeeping waste but also to establish a hierarchy concerning the most efficient carrier gases, even if the oxidative gases give higher detritiation yields. Thus, the reference temperature was to be slightly increased up to 150°C.

The first results of experiments carried out at this new reference temperature reveal that this temperature increase leads to a fivefold detritiation yield increase, leading to the possibility to declassify waste treated with Ar-2%O₂.

Further experiments currently carried out may confirm the surface contamination of the waste by tritium as well as the influence of the feed flow on the detritiation yield.

The most appropriate carrier gas to be employed for housekeeping detritiation and gaseous tritium recovery by the membrane reactor designed by ENEA is Ar-2%O₂, which will be used at 150°C. This gas will allow to obtain reasonable detritiation efficiency and to have the advantage to be hazard-free towards hydrogen. The feed flow rate needs to be optimised by a compromise between waste detritiation and membrane permeability efficiencies.

ACKNOWLEDGMENTS

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Carrier gas	a _{init} (kBq/g)	a _{res} (kBq/g)	Y (%)
Ar- ^{2%} O ₂	23.0	21.3	7.6
Ar- ^{5%} H ₂	23.0	22.2	3.5
Ar- ^{25%} H ₂ O	29.7	28.2	5.0
Ar- ^{40%} H ₂ O	29.4	27.9	5.4
Air	10.2	9.4	8.1

Table 1. Housekeeping initial, after 18h-experiment tritium activities and detritiation yield at 120°C and 300NmL/min.

Carrier gas	T (°C)	a _{init} (kBq/g)	a _{res} (kBq/g)	Y (%)
Ar- ^{2%} O ₂	120	23.0	21.3	7.6
Ar- ^{2%} O ₂	150	17.3	7.6	56.1
Ar- ^{25%} H ₂ O	120	29.7	28.2	5.0
Ar- ^{25%} H ₂ O	150	18.8	14.3	23.6

Table 2. Housekeeping initial, after 18h-experiment tritium activities and detritiation yields at 120 and 150°C (15 h for Ar-^{2%}O₂ at 150°C).

Carrier gas	Feed flow (NmL/min)	a_{init} (kBq/g)	a_{res} (kBq/g)	Y (%)
Air	300	10.2	9.4	8.1
Air	450	13.1	11.4	13.5

Table 3. Housekeeping initial, after 18h-experiment tritium activities and detritiation yield at 120°C for different gas flows.

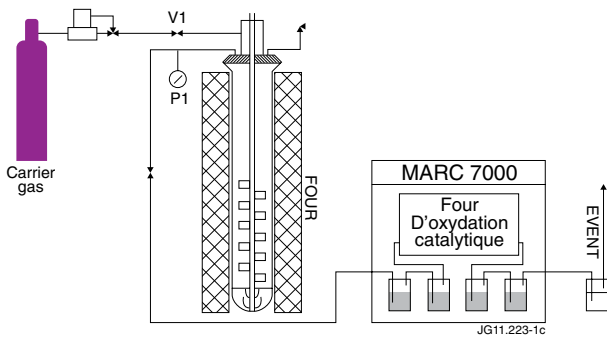


Figure 1: Schematic representation of the original housekeeping detritiation facility.

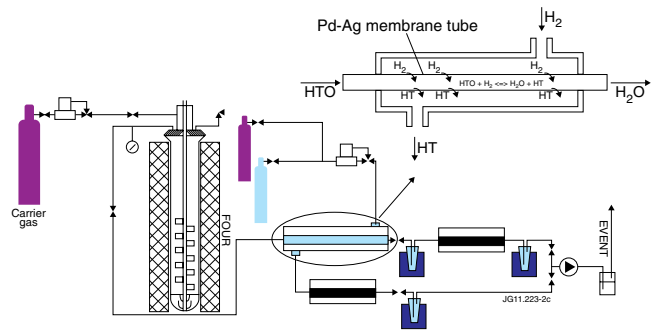


Figure 2: Schematic representation of the housekeeping detritiation facility coupled to the catalytic membrane reactor.

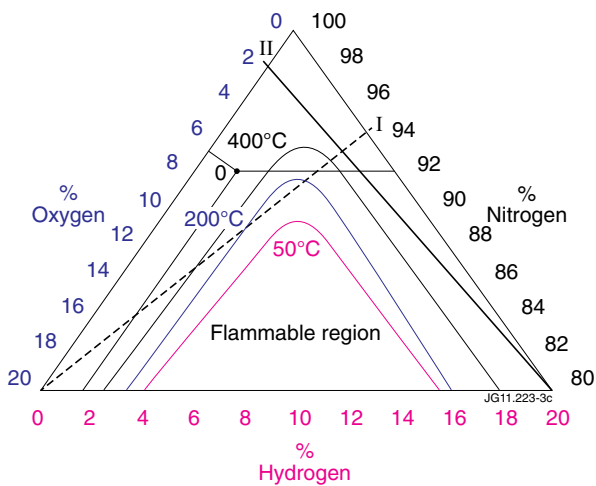


Figure 3: Flammable region limits of a ternary $O_2/H_2/N_2$ mixture as a function of the temperature.

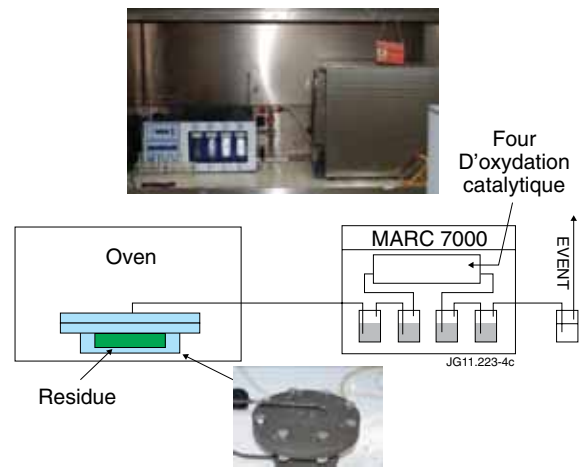


Figure 4: Schematic representation of the residue calcination facility.

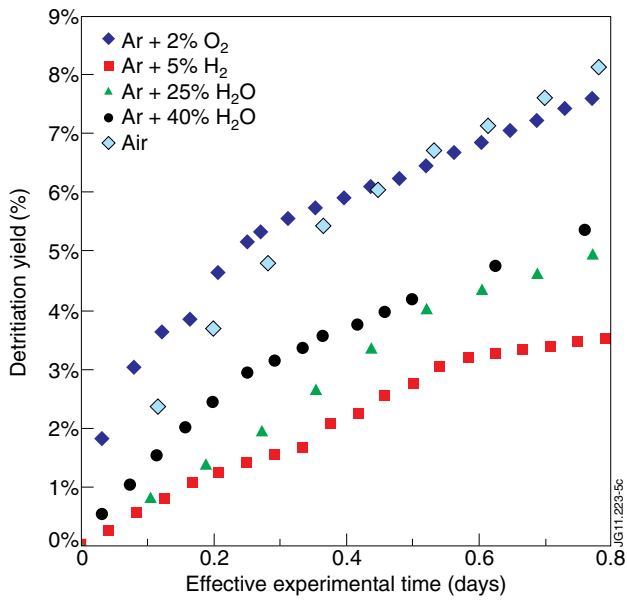


Figure 5: Soft housekeeping detritiation yield as a function of effective experimental time for different carrier gases. $T = 120^{\circ}\text{C}$ and $Q = 300\text{NmL/min}$.

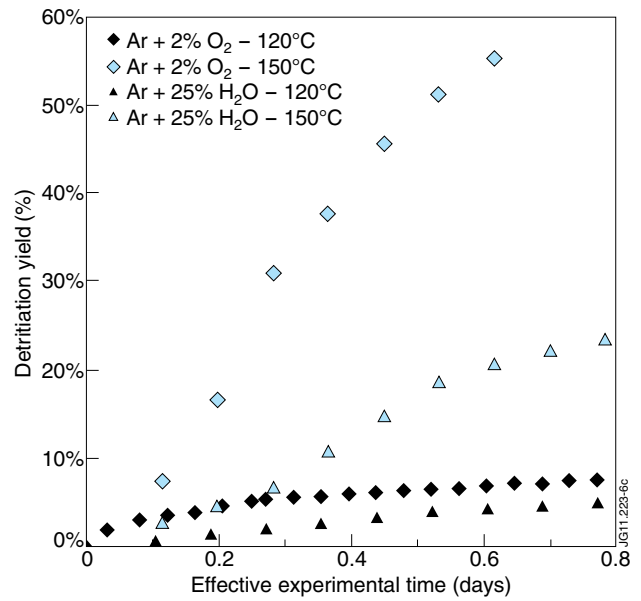


Figure 6: Detritiation yield as a function of effective experimental time with Ar + 2% O₂ and Ar + 25% H₂O. $Q = 300\text{NmL/min}$.

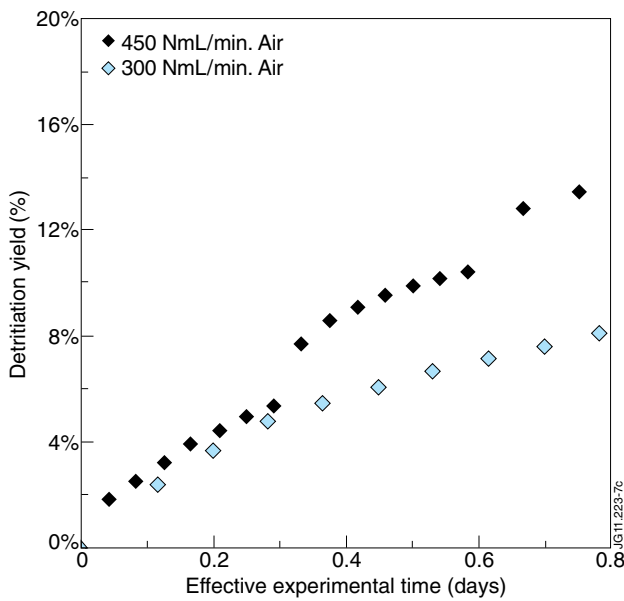


Figure 7: Detritiation yield as a function of effective experimental time with air. $T = 120^{\circ}\text{C}$.