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ABSTRACT

The JET Fusion Technology (FT) work programme was launched in 2000, in the frame of the European Fusion Development Agreement, to address issues related to JET and ITER. In particular, there are four topics related to tritium being investigated .

Based on the experience gained on the existing tokamaks, first calculations indicate that in-vessel tritium retention could represent a burden for ITER operation. Therefore erosion/deposition studies are being performed in order to better understand the layer co-deposition and tritium retention processes in tokamaks. Moreover, testing of in-situ detritiation processes, in particular laser and flash lamp treatments, should assess detritiation techniques for in-vessel components in the ITER-relevant JET configuration.

To reduce the constraints on waste disposal, dedicated procedures are being for detritiation of metals, graphite, carbon-fibre composites, process and housekeeping wastes.

During the operational and decommissioning phases of a fusion reactor, many processes will produce tritiated water. Key components for an ITER relevant water detritiation facility are being studied experimentally with the aim of producing a complete design that could be implemented and tested at JET.

This paper describes these topics of the FT-programme, the strategy developed and the results obtained so far.

1. INTRODUCTION

Because of its plasma size, divertor configuration, heating systems, diagnostics, tritium and beryllium capabilities, remote handling and other auxiliary facilities, the JET device can access a wide range of operating regimes that allow to address issues likely to affect ITER design and operation. It can also be used as a test bed for ITER-relevant equipment and technology and provide data and samples whose analysis will help ITER licensing.

The JET Fusion Technology (FT) work programme was launched in 2000, in the frame of the European Fusion Development Agreement, to cover R&D areas that can provide relevant contributions to the research programmes of both ITER and JET. Until now, laboratories in Belgium, Finland, France, Germany, Italy, Romania and United Kingdom have taken part in these studies. The activities cover tritium issues, engineering, neutronics and safety. Results and strategies are discussed on erosion and deposition of plasma facing component materials (PFCs), in-situ detritiation, waste management and water detritiation.

2. EROSION AND DEPOSITION

During the 1997 Deuterium–Tritium Experiment (DTE1), tritium accountancy performed within the Active Gas Handling System (AGHS) revealed that up to 40% of the tritium injected into the tokamak was, at least temporarily, trapped in the machine [1]. After a three month clean-up campaign, about 17% of the site inventory remained in-vessel. Carbon layers co-deposited with hydrogen up to H/C ratio of 0.8 had been observed before DTE1. Such layers were believed to retain the majority of the injected tritium in DTE1. Various FT tasks were started to locate and characterise the areas of erosion and deposition on the plasma facing components.

2.1. EXPERIMENTAL

Different divertors have been implemented in JET leading to different distributions of hydrogen isotopes, carbon and beryllium on in-vessel surfaces [2]. Within JET FT, the work has been focused on the MkIIIGB configuration.

The MkIIIGB divertor was first mounted on JET (see Figure I) in 1998. To characterise better the deposition and erosion of the PFCs, tiles were removed in 1999 and replaced by other tiles with special marker coatings [3].

These tiles were introduced in JET at different locations inside the vacuum vessel (Divertor, Inner Wall Guard Limiters (IWGL) and Outer Wall Limiters (OPL)).

2.2. RESULTS

Coated marker tiles from the Divertor, OPL and IWGL have been analysed. The major part of the analyses performed within the FT work-programme concerned the tiles installed in 1999 and removed in 2001. Some tiles present in the torus from 1998 to 1999 and from 1998 to 2001 were also analysed. On the inner divertor wall (tiles 1 and 3), the surface analyses have shown a double structure layer in the tiles exposed from 1999 to 2001:

- The surface layer (approx. 1-5 μm on tile 1 and 10-15 μm on tile 3) contains mostly carbon and beryllium with high deuterium content and some nickel (< 1% at.). Nickel is one of the constituents of the inconel, which is used for the JET vessel wall material, internal metal fitting, bolts etc.
- The film underneath is very rich in beryllium and nickel traces are also present in this region.

The same double structure layer has been observed on tiles exposed from 1998 to 2001 but not on tiles exposed from 1998 to 1999. Experimental data obtained by SIMS and RBS were used to determine the composition of the different layers. RBS spectra were modelled using SIMNRA program [4] with the different layers suggested by the SIMS results and adding a ^{13}C layer. The results of the simulation are given in Table I.

The model for erosion/deposition predicted a 2-stage process. The impurities in the scrape-off layer (mostly carbon and beryllium) arrive at tile 1 and 3. Then the carbon is chemically re-eroded and transported to the shadowed regions of tile 4 and the inner louvre region, leaving a beryllium- rich layer on tiles 1 and 3.

The presence of carbon-rich layer on the surface of tiles 1 and 3 (installed in 1999 and removed in 2001) may indicate that the re-erosion of carbon was reduced by decreasing the wall temperature of JET from 320°C to 200°C during the last 12 weeks before 2001 shutdown. The helium plasmas during the last 3 weeks before shutdown might also have contributed to the change of composition of the layers.

At the outer divertor (tiles 7 and 8), erosion (up to several microns) is observed in almost all areas. The ^{13}C from the puffing experiments was not detected. SIMS analyses show implanted hydrogen isotopes at the surface of an eroded carbon-fiber composite (CFC) tile.

On the IWGL tiles there are regions both of erosion and deposition, but no double layer structure is observed in the deposits. Deposited layers of up to 80µm thickness have been found (on the upper part of the limiter). The surface is covered by a deuterated layer of carbon plus some beryllium. At areas of the limiter closest to the plasma, erosion of the coating is noticed with some hydrogen isotope implantation

On the OPL tiles, the central regions are areas of net erosion (up to several microns) with implanted hydrogen isotopes at the surface of the CFC. Some deposition is found at the ends of the OPL tiles (up to about 60µm) with relatively low hydrogen isotope, beryllium and nickel contents.

2.3. NEXT STEPS

A fresh set of tiles and other components will be removed from JET during the 2004 shutdown. All the 2002-2004 campaigns have been at the lower vessel temperature, so it might become clear if that was the reason for the duplex film in the inner divertor, and if so, what are the consequences for transport to and deposition in the shadowed areas.

3. IN SITU DETRITIATION

Tritium retention inside the machine is an issue for ITER, since tritium inventory limits are set for safety reasons. To have a minimal impact on operation, in-situ detritiation has to be studied with the aim of decreasing the retained tritium inventory within the chamber without de-conditioning.

The reduction of the tritium inventory can be done by ablation or heating of the co-deposited layer, which contains the highest concentrations of the tritium.

3.1. RESULTS

Within JET FT work-programme, detritiation techniques using two different devices are being studied: laser and flash lamp.

Tests using lasers have been performed, at laboratory scale, on samples from Tore Supra and TEXTOR [5]. Ablation thresholds for graphite ($>1\text{J}/\text{cm}^2$) and co-deposited layers ($<0.5\text{J}/\text{cm}^2$) have been determined experimentally by measuring the crater depth obtained with different energy densities (Fig.2).

The ablation efficiency obtained was estimated to allow a 20µm co-deposited layer removal, without damaging the graphite substrate, at about $1\text{m}^2/\text{h}$, with a Nd-YAG laser beam of 250W mean power, repetition rate of 10kHz, and 100ns pulse duration,. Laser energy densities were also adjusted to be lower than the co-deposited layer ablation threshold in order to perform laser heating experiments. After laser treatment with different time durations, Glow Discharge Optical Spectroscopy (GD-OS) was applied for in-depth analysis of graphite samples with co-deposited layers. The (H+D) profiles obtained by GD-OS are presented in Fig.3.

Heating by nanosecond laser is thus efficient for the surface detritiation of thin co-deposited layers.

After effective tests at laboratory scale on simulated deposited layers [6], it was decided to test photon cleaning by flash lamp in the ITER-relevant JET configuration (tritium and beryllium environment, use of remote handling tools). To perform the tests (ablation of inner divertor tile deposits and heating of OPL) during the JET 2004 shutdown, the flash lamp system was adapted for remotely manipulation. A head unit containing the 5Hz flash lamp and optics was held by the MASCOT robotic arm, with power and cooling water being supplied via an umbilical. Ablation tests have been performed in situ at 300J in a region where the deposit thickness is assumed to be between 60 and 80 μm . The OPL tiles, where the tritium content in the film is supposed to be high, will be removed later during the shutdown in order to perform the heating treatments. They will be performed at 100J, energy, insufficient to remove co-deposit but sufficient to allow tritium out-gassing, inside the Beryllium Handling Facility (BeHF). Further test will be also performed in BeHF in order to characterise the particulates and gases produced during ablation.

3.2. NEXT STEPS

Following the flash lamp test, the tritium content for treated and adjacent untreated tiles will be measured by calorimetry (tile content) and full combustion (surface and bulk contents). Ion Beam Analyses will be also performed to assess the changes in the surface film composition. The next step in terms of laser detritiation, focussing on ablation, is its implementation in a real tokamak configuration with the use of remote handling tools. This was foreseen for the 2004 shutdown in collaboration with the USA but had to be cancelled. It is now proposed to develop all the technology necessary for the adaptation of the laser to the JET remote handling arms, including the use of optical fibres and galvanometer for the tile surface scanning.

4. WASTE MANAGEMENT

In fusion devices operating with tritium, different tritiated wastes are produced. As an example, at JET tritiated stainless steel, graphite or CFC items, organic liquids together with process and housekeeping wastes are common. The constraints on the storage and final disposal can be limited by reducing the tritium content inside the materials. Therefore, dedicated procedures are being developed for each type of materials, with the major goal of obtaining good detritiation factors with easily manageable secondary waste.

4.1. RESULTS

In organic oils, tritium seems to be bound in organic molecules, which leads to unsuccessful thermal desorption. The use of additional chemical reactions, such as oxidation, to transfer the tritium to a more stable and easier treatable inorganic matrix (water) is necessary. One of the treatments studied consists of mixing equal volumes of oil (specific activity 8.5kBq/L) and water at room temperature for 72 hours. In these conditions, 10% of the tritium remained in the oil, which would then need further processing. In another set of experiments, oil and water were injected separately, via nozzles, into a furnace to allow a complete oxidation. In these tests, 7ml/min of

oil (specific activity 301 kBq/L) and 9ml/min of water were injected during about 350min into a furnace at about 1000°C. The total organic carbon was below the detection limits (2mg/L). Less than 0.6% of the tritium was discharged through the ventilation system, the remainder of the tritium being in the condensate. The first results show that the tritium can be transferred, more or less efficiently, to the water and that the volume of waste is multiplied by a factor 2-3. But it is important to notice that the processes developed are based on simple technologies and lead to secondary wastes which are easier to manage (HTO).

Concerning carbon materials, heating treatment in air cannot be used for tiles since it leads to the destruction of the material by oxidation. Nevertheless, the highly tritiated dust and flakes produced at JET could be processed by such treatment.

Thermo-desorptions have been performed on samples from JET CFC tiles from 20 to 1100°C under a stream of He containing 0.1% H₂. The tritium is significantly released only at temperatures above 400°C. The highest tritium release rate is observed at about 850°C [7]. The first type of released tritium (between 400 and 800°C) is attributed to the break of T-C bond when the carbon has a *sp*³ hybridization and the second (above 800°C) to the break of T-C bond with *sp*² or *sp* hybridization.

Some desorptions have been performed under a stream of Ar containing 5% H₂ showing that the optimal detritiation temperatures are between 300 and 800°C [8]. For two CFC samples, the species removed, as a function of the temperature is shown in Fig.4. Decontamination factors (DF = initial activity / final activity) between 20 and 90 are obtained.

After tests on small samples (thickness about 1.8mm), the detritiation of full graphite tiles is ongoing using radio frequency heating. The amount of tritium before and after detritiation will be measured by calorimetry.

For stainless steel, no tritiated sample from JET was available, therefore work has been performed on samples from an SCK-CEN oxidation installation and a French fast breeder reactor. Large samples (250 to 700g) were used in order to determine the impact of the treatments on tritium trapped both at the surface and in the bulk. Heating treatments have been performed under different atmospheres: superheated steam, air and Ar + 5% H₂

Initial tests with superheated steam have shown that the optimal conditions are a working temperature around 370°C, without carrier gas and with a low steam flow rate (~ 220g/h) over approximately 6 hours. A DF of about 210 was obtained (initial activity 41kBq/dm²). In that case, the tritium contents were determined by smear tests so only the surface contamination is considered. Measurements of the tritium content in the bulk are necessary in order to fully determine the efficiency of this process in reducing the tritium in the waste.

Other samples with initial activity around 40Bq/g were treated in a furnace up to 1100°C in air or Ar + 5% H₂ [8]. The total tritium content was measured by melting of the stainless steel and counting of the outgassing species. Heating the samples during 3 hours at 400°C leads to DF of about 5 in air and 8 in Ar + 5% H₂. The DF increase respectively to about 130 and 110 when heating at 1000°C during 0.5 hour.

These studies on relevant samples (no artificial loading and large dimensions) provide data on stainless steel detritiation by heating. Even if the DF obtained at present are not very high, these processes could allow significant reduction of the tritium content in the waste.

All these treatments, of course, lead to the production of tritiated gases or liquids that will have to be treated. The right balance between the production of secondary waste and the reduction of waste classification has to be reached.

4.2. NEXT STEPS

An inventory of the waste produced by JET Active Gas Handling System, basement equipment and torus diagnostics is ongoing. It will provide information on the masses, materials and activities (contamination and/or activation) of the JET waste in order to identify the ones requiring more R&D for processing and packaging. At a laboratory scale, work continues on stainless steels and housekeeping waste in order to optimise the decontamination factor while reducing the production of tritiated water.

5. WATER DETRITIATION

Tritiated water is produced during fusion reactor lifetime and decommissioning. The Water Detritiation System (WDS) is based on an enrichment of the tritiated water inside a Liquid Phase Catalytic Exchange (LPCE) column, then a dissociation of oxygen and hydrogen isotopes in an electrolyser and the separation of the hydrogen isotopes inside a Cryo-Distillation (CD) column and Gas Chromatography (GC) (Fig.5).

With the aim of proposing a complete design for an ITER relevant water detritiation facility that could be implemented and tested at JET, the key components for such a system are being experimentally studied, particularly the LPCE column and two types of electrolyser.

5.1. RESULTS

The main requirements for the design of a WDS for JET are to process 10 tons tritiated water of $\sim 1\text{Ci.kg}^{-1}$ during one operation campaign of 2-3 months, to achieve a decontamination factor of 10^4 along the stripping section of the LPCE column and to deliver a tritium content in the bottom product of CD column in the range of 0.5-1% atomic ratio to allow further separation by the GC systems.

The LPCE column is based on a mixture of catalyst and packing. Laboratory experiments were carried out to investigate the separation performances of different catalysts and catalyst/packing mixtures [9][10]. The separation performances have been measured against several parameters: the operating temperatures (40 to 80°C), the composition of the catalysts (platinum concentration and ratio between the support materials), catalyst particle size, type of packing (etched stainless steel or phosphoro-bronze), catalyst /packing filling ratio and the mixing mode between catalyst and packing (homogenous or layers). Two catalyst/packing mixtures, which have both good technological behaviour and separation performances, were selected for further investigations and improvements (see Table II). The separation performances, assessed with the height equivalent of theoretical

plates (HETP), are highly dependent on the ratio G/L between the specific molar flow rates of hydrogen and water (typically between 3 and 4).

Tests are ongoing on the two selected catalyst/packing mixtures in order to check the achievable decontamination factors on longer columns (more than 2†m).

Meanwhile, the two catalyst/packing mixtures are currently investigated in endurance experiments over 3, 6 and 9 months. The aim is to determine the influence of tritium decay and possible erosion phenomena due to the water flow. If necessary, an appropriate procedure for regeneration of the catalyst will be developed. Two different exposure modes are used: immersion in tritiated water and exposition to water-vapour as in a total reflux distillation process [11].

There are two types of electrolyser: conventional electrolyte based on aqueous solution of potassium hydroxide and solid polymer electrolyte (SPE). On one hand, currently used with tritiated water, the conventional electrolyte needs auxiliary systems for KOH purification since KOH acts as a poison on the catalysts of the LPCE column and the palladium membranes of the permeator and could lead to waste not easily manageable. On the other hand, SPE, which has low tritiated water hold-up, does not need a liquid electrolyte and auxiliary systems for KOH purification but the lifetime in an environment of highly tritiated water is not known. Tasks have been launched within the Fusion Technology work programme to deal with these issues.

Removal of caustic materials from hydrogen and oxygen streams generated by an alkali electrolyser has been studied. It has been demonstrated that hydrogen and oxygen with a very small residual caustic content, below 0.1mg/m^3 , can be supplied by an electrolyser equipped with a simple rinsing column and a final fine filter. If required, tritiated water can be removed from the electrolyte of the electrolyser employed within a water detritiation facility. A simple distillation column would provide water with chemical purity sufficient for its further processing. These results show that employing an alkali electrolyser should not present a problem in respect of safety and operation. In the same time, the behaviour of an electrolyser cell based SPE exposed during three months to tritiated water ($1\text{ to }10\text{ Ci.kg}^{-1}$) has been investigated. The cell current during the test, the chemical composition of the tritiated water and the comparative tensile strength, before and after the test, have been monitored.

After enhancements, the two types of electrolyser can be considered for JET-WDS process evaluation.

5.2. NEXT STEPS

The configuration and the evaluation of the length of the LPCE column for the JET WDS have been done on the assumption that the deuterium concentration in the tritiated water is at natural level. However, it has been shown that the real deuterium content in the water to be processed ($\sim 0.2\text{-}0.5\text{†}\%$ at $\text{D}/(\text{H}+\text{D}+\text{T})$) has to be taken into account. In order to precisely determine the length of the LPCE column and complete the design, the impact of H, D and T species present in the LPCE column, at relevant concentrations, will be investigated in 2005.

CONCLUSIONS

Fusion Technology activities at JET are providing relevant contributions to JET operation and ITER design in the areas of erosion/deposition, in-situ detritiation, waste management and water detritiation. There are still some pending questions, in particular the understanding of all the phenomena involved in impurity transport. But on the other topics, processes and technology are being developed in the different laboratories to make available solutions for JET and ITER.

Other topics covered by FT at JET and not addressed in this paper are being developed in order to provide data for ITER design and licensing. These are tritium processes (cryo panels, gas purification), engineering studies (optical fibres test under JET irradiation, laser in-vessel viewing system R&D, etc.), neutronics and safety tasks (tritium spreading modelling, development of shutdown dose rate codes, collection of data on component failures, remote handling experience feedback, occupation radiation exposure, etc.).

REFERENCES

- [1]. P. Andrew et al, "Tritium recycling and retention in JET", *Journal of Nuclear Materials* **266-269** (1999), pp 153-159
- [2]. J.P. Coad et al, "Distribution of hydrogen isotopes, carbon and beryllium on in-vessel surfaces in the various JET divertors", This Conference.
- [3]. J.P. Coad et al, "Erosion/deposition in JET during the period 1999-2001", *Journal of Nuclear Materials* **313-316** (2003), pp 419-423
- [4]. M. Mayer, SIMNRA User's Guide, Report IPP 9/113, Max-Planck-Institut für Plasmaphysik, Garching, Germany, 1997.
- [5]. F. Le Guern et al., "laser ablation and heating tests performed on Tore Supra and TEXTOR graphite samples", *Proceedings SOFE Conference*, Sept. 2003, San Diego (USA), to be published.
- [6]. K. Gibson et al., "The removal of co-deposited hydrocarbon films from plasma facing components using high-power pulsed flashlamp irradiation", *Proceedings PSI Conference*, May 2004, Portland (USA), to be published.
- [7]. N. Bekris, "Surface and depth profiling of tritium in selected JET tiles", Final report for the Fusion Technology task Jw0-FT-1.6, Dec. 2003.
- [8]. C. Poletiko et al., "Dedicated procedures for the detritiation of steel and graphite", This Conference.
- [9]. I. Cristescu et al., "Investigation of separation performances of various isotope exchange catalysts for the deuterium-hydrogen system", *Fusion Science and Technology*, vol. 41, May 2002, pp 1087-1091.
- [10]. J. Braet & A. Bruggeman, "Development of an improved hydrophobic catalyst for liquid phase catalytic exchange", *SOFE conference*, San Diego, USA, October 2003.
- [11]. G. Ionita et al., "Endurance Test for the SCK-CEN Catalytic-Mixed Packing Proposed for Water Detritiation System at JET", This Conference.

Layer	Composition in at. %					Thickness (nm)**
	D	Be	¹² C	¹³ C*	O	
1	12	3	27	55	3	276
2	22	5	66	0	7	1784
3	21	24	35	0	20	2067

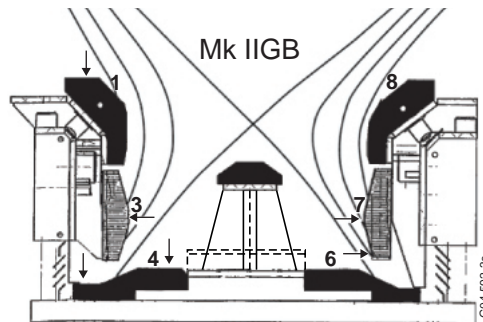
JG04.592-1c

Table I: Results of the simulation on middle of tile 1 exposed from 1998 to 2001.

	Mixture A	Mixture B
Catalyst	Pt: 1%w C: 19%w PTFE: 80%w	Pt: 1.5%w C: 15%w PTFE: 83.5%w
	Pellets	Pellets
	fl = 1.9mm×2mm	fl = 2mm×2mm
Packing	Minispirals	wiremesh
	Stainless Steel	Stainless Steel
	fl = 2mm×2mm	
Filling ratio (catalyst/packing)	1:2 homogeneous	1:2.5 sandwich method
Temperature	40°C	60-80°C
Column diameter	20mm	38mm
[T] in H₂ inlet stream	100kBq/mol	0
[D] in H₂ inlet stream	0	1-2%
G/L	3.51	3.35
HETP (cm)	11.38	26

JG04.592-2c

Table II: Characteristics of the 2 selected mixtures.



G04.592-3c

Figure 1: MkIIGB Divertor installed in 1998

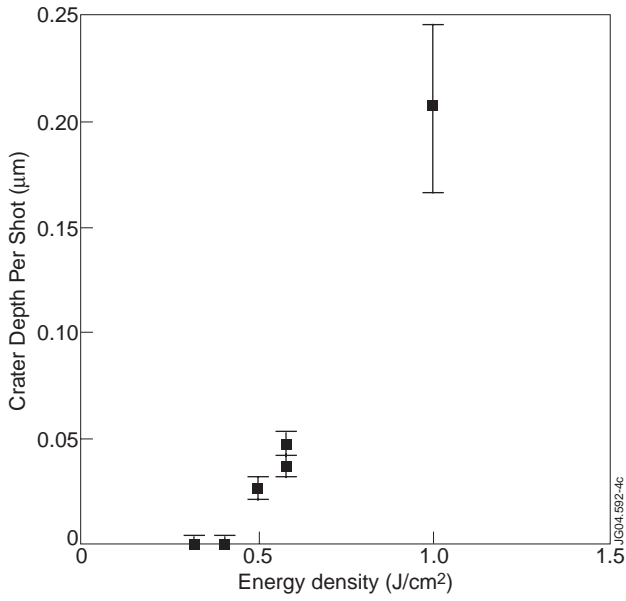


Figure 2: Crater depth vs. laser energy density on co-deposited layer (4-10ns pulse duration, 532nm, homogenized beam, 20Hz repetition rate)

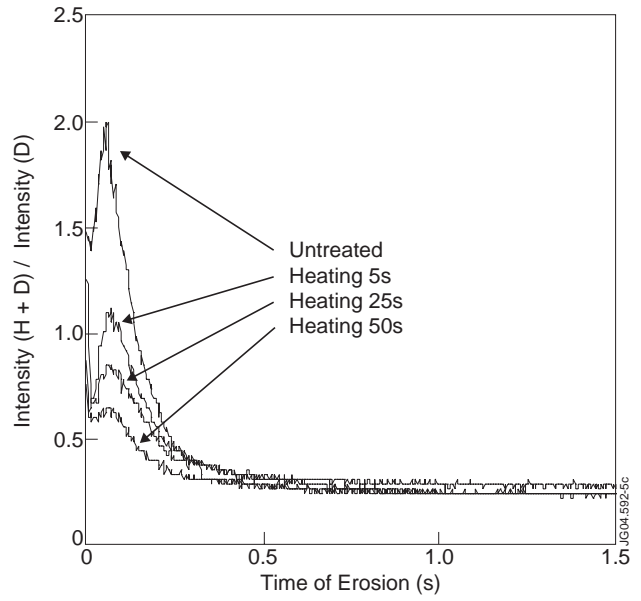


Figure 3: H+D profile obtained by GD-OS.

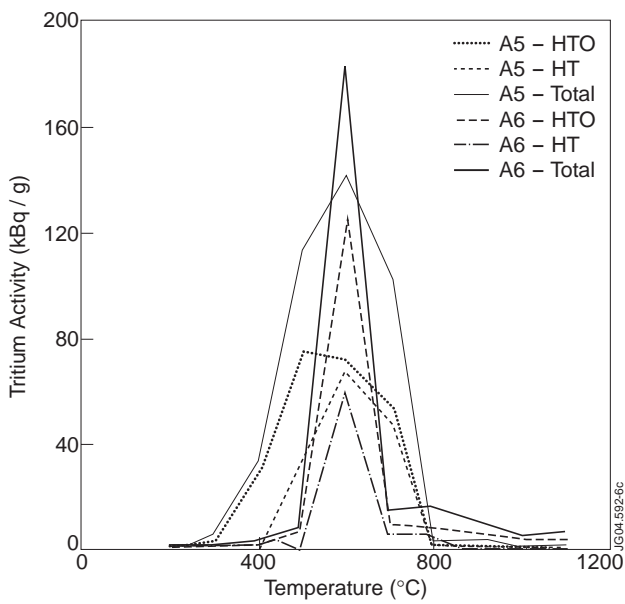


Figure 4: HT and HTO desorption as function of temperature.

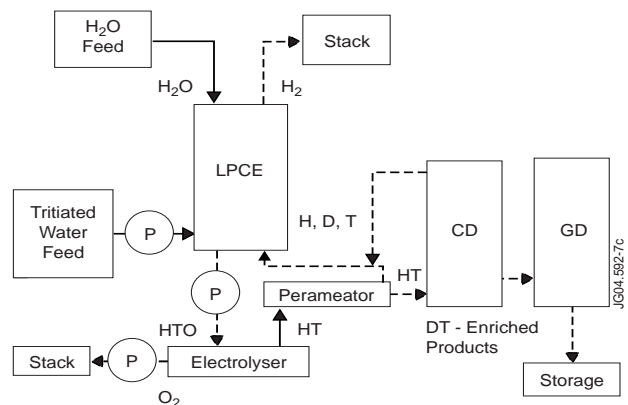


Figure 5: JET WDS design (P: purification, only main fluxes are indicated).