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A.N. Perevezentsev, A.C. Bell, P.D. Brennan, J.L. Hemmerich

EURATOM/UKAEA Fusion Association , Culham Science Centre, Abingdon, Oxfordshire OX14 3DB, U.K.

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ABSTRACT

A water detritiation facility, based on a world-wide adopted Combined Electrolysis Catalytic Exchange (CECE) process, for the JET Active Gas Handling plant is described. A research and development programme is presented. The programme includes the testing of structured inert packing with an incorporated hydrophobic catalyst for increased throughput of a Liquid Phase Catalytic Exchange (LPCE) column, a Vapour Phase Catalytic Exchange (VPCE) process for reduction of tritium inventory in the alkali electrolyser and a column of high effectiveness for alkali retention.

1. INTRODUCTION

Ever since the JET experiment with deuterium-tritium plasmas in 1997, known as DTE1 [1], a stream of tritium-contaminated water waste has been continuously generated [2] in the Exhaust Detritiation System (EDS) of the JET Active Gas Handling (AGH) plant, which provides detritiating ventilation to chronically off-gassing volumes of the machine [3]. An average rate of the water generation is approximately 10 tonne a year. Tritium concentration in this water varied from 60GBq/kg observed just after the DTE1 campaign to the current, ie more than 4 years later, value of 2GBq/kg. A larger stream of tritiated water is expected from the waste detritiation facilities being developed [2]. An increase of tritiated water flow in the future is the reason for carrying out a conceptual design study for a water detritiation facility for JET.

The Combined Electrolysis and Catalytic Exchange (CECE) process widely adopted for water detritiation [4-9] will be used for the JET water detritiation facility. To reduce the cost and time required to construct the facility, it has to be incorporated into the JET AGH plant. A simple schematic of the facility is shown in Fig.1. The key elements of the facility are an electrolyser (EL) for conversion of tritiated water to gaseous hydrogen, a LPCE column where this hydrogen is detritiated via isotopic exchange with liquid water, a Cryogenic Distillation (CD) column for tritium pre-enrichment and a Palladium Permeator (PD) for chemical purification of hydrogen to be fed to the CD column. Tritiated feed-water is supplied to the electrolyser. The hydrogen generated in the electrolyser passes a PD where part of the hydrogen stream is purified by selective diffusion through membranes of palladium alloy and then fed to the CD column. The rest of the hydrogen stream is injected into the LPCE column. A tritium-depleted product of the CD column is also injected to the LPCE column. The LPCE column is fed with natural (tritium-free) water at the top. Hydrogen detritiated in the LPCE column is discharged to the environment. A tritium-enriched product of the CD column is transferred to the Gas Chromatographic (GC) isotope separation system of the AGH plant for final enrichment to pure tritium.

The existing equipment and operating procedures of the AGH plant dictate the limits for the water detritiation facility:

a) Tritium discharge from the water detritiation facility should contribute a fraction of <10% to the daily limit of 40GBq adopted for the AGH plant,

b) A cooling capacity of the AGH plant helium refrigerator available for the CD column is 500W (at 20K),

c) The CD column should provide tritium enrichment to ≥0.5at.% and tritium inventory below 37TBq,

d) Maintaining tritium concentration in the water of the electrolyser, which will be the only not high vacuum standard equipment operating at elevated pressure, as low as reasonably achievable is desirable.

This paper describes the features of the JET water detritiation facility.

2. DESCRIPTION OF THE FACILITY

The flow diagram of a water detritiation facility is given in Fig. 2. It is proposed that an electrolyser of hydrogen throughput of 10Nm^3/h and a CD column of the height equivalent to 100 theoretical plates [1] will be used in the facility. The numerical evaluation shows that to enrich tritium from $5 \cdot 10^{-7}$at.%, which is equivalent to 60GBq/kg of tritium concentration in tritiated feed-water, to ≥0.5at.% recovering >99% of tritium from the feed and maintaining a tritium inventory <37TBq, the CD column should operate with a re-flux ratio of 6 or larger. A cascade of two CD columns would be needed if tritium concentration in the feed-water were below 6GBq/kg. The re-flux ratio and cooling capacity available for the CD column limit the feed rate to 200mol/h.

To meet the tritium discharge limit, the water detritiation facility should provide an overall decontamination factor (DF) ≥10 000. The numerical evaluation of performance of a water detritiation facility of a layout given in Fig. 1 shows (see Table 1) that ≥25 theoretical plates are required to provide DF ≥ 10 000. It was considered desirable to employ a single LPCE column, which should be characterised by a HETP (Height Equivalent to Theoretical Plate) of ≤0.35, to fit inside the AGH building. A ratio, $R = L_{HTO}/L_{H2O}$, of flow-rate of tritiated feed-water to tritium-free feed-water is a parameter strongly influencing the decontamination factor and giving a large extent of flexibility in terms of DF provided. The smaller this ratio, the larger DF can be achieved, however at the expense of the amount of tritiated water processed.

Table 1: Evaluation of the water detritiation facility performance (operation temperature of LPCE column of 333K, no equilibrators are foreseen for the CD column).

R	N*	DF	Product*	
			CD (at.%)	LPCE (kBq/mol)
1	25	1.6E9	0.5	4.2E-4
1.7	25	3.5E6	0.5	0.2
2	25	1.8E5	0.55	4
2.5	25	6.1E3	0.5	1.1E2
3	25	380	0.45	1.8E3
2	35	3.7E7	0.55	2E-2

* Number of theoretical plates in LPCE column (N), tritium concentration in the product of the CD column and in hydrogen detritiated in the LPCE column are given.

The electrolyser consisting of a cascade of electrolytic cells has the potential to be a major source of tritium leakage from the facility. Tritium concentration in the water of the electrolyser is much larger than that in the feed-water. This is inevitable due to a large isotopic effect of the water electrolysis. The amount of water required for the electrolyser's operation depends on its type. Employment of electrolysers of two basic types has been considered: a conventional electrolyser with alkali electrolyte and an electrolyser with a solid polymer electrolyte. The latter type, which does not need a liquid electrolyte and auxiliary devices to remove alkali from the gas streams, is more attractive. However, it still requires an extensive and long-term testing programme to reach the level of confidence necessary for safe operation with tritium-contaminated water. Therefore, a conventional, commercial electrolyser of elevated operation pressure with a potassium hydroxide (KOH) electrolyte, which has demonstrated a reliable performance for many years, has been chosen. To provide an additional protection against the electrolyte leakage, it is foreseen to modify a commercial electrolyser by adding a water-filled channel surrounding the gas flow ducts and electrolyte flow ducts. This channel will operate as a secondary containment. Electrolyte leakage will be detected by monitoring tritium activity and electrical conductivity of the distilled water circulated through the channel.

It is foreseen to operate the facility in a steady state mode, which supposes that tritium flow with the feed and with the tritium product of the CD column is equal to that in the tritiated feed-water. Because only a fraction of the hydrogen generated in the electrolyser will be fed to the CD column, the tritium concentration in the electrolyte will rise until a balance of tritium flows has been achieved. An additional increase of the tritium concentration in the electrolyte may be necessary if the tritium concentration in the hydrogen fed to the CD column is too small, to secure enrichment to 0.5at.%. The LPCE column should have sufficient detritiation capacity to compensate for the increase in tritium concentration in the feed-hydrogen and sustain tritium discharge within the adopted limit. To reduce the increase of tritium concentration in the electrolyte, as described above, it is foreseen to employ a cascade of two CD columns and pre-enrich tritium in hydrogen fed to the CD cascade. The latter option is based on the use of a Vapour Phase Catalytic Exchange (VPCE) reactor. The electrolyte will circulate through the electrolyser and a boiler, which is purged with hydrogen generated in the electrolyser. Water vapour generated in the boiler and then purified of potassium hydroxide aerosol is passed through the VPCE reactor together with gaseous hydrogen. In the VPCE reactor, tritium is transferred from the water vapour to the hydrogen. The increase of tritium concentration in the hydrogen is related to a molar ratio of gaseous hydrogen to water vapour flows (λ) by a simple correlation

$$K = (\lambda + \alpha_e)/(\lambda + \alpha_i) \tag{1}$$

where K is the tritium enrichment factor, α_e is a tritium separation factor for water electrolysis in the employed electrolyser, α_i is the equilibrium separation factor for a reaction of tritium isotopic exchange between hydrogen and water vapour.

Tritium enrichment in the hydrogen by a factor of K>4 can be achieved for a VPCE reactor operated at a temperature ≥570K and a ratio of $\lambda \leq 0.5$. The ratio λ can be maintained by controlling the temperatures of the boiler and the condenser.

The presence of potassium hydroxide requires special attention to eliminate its poisoning effect [4] on the catalysts in the LPCE column, the recombiner of the hydrogen and oxygen purification devices and on the palladium membranes in the palladium permeator. Deterioration of the catalysts in gas purification devices has been observed for commercial electrolysers. Destruction of the palladium membranes in the palladium permeator as a result of interaction with potassium hydroxide could lead to the more severe problem of accidental oxygen and water vapour ingress to the CD column. Accumulation of oxygen and water in a form of solid clusters in liquid hydrogen in the CD column could form a potentially explosive mixture. Considerable improvement of effectiveness of KOH retention columns in the gas purification devices of the commercial electrolysers is necessary, to eliminate potassium hydroxide breaking through. Molecular sieve dryers may be installed at the inlet and pure hydrogen outlet of the palladium permeator to provide the additional protection for the permeator and the CD column.

3. EXPERIMENTAL PROGRAMME

A research and development programme has been set up for a JET water detritiation facility. Reproducibility in performance reported in the earlier studies for the LPCE columns filled with random layers of hydrophobic catalysts (all CECE pilot-scale water detritiation facilities [4-9] are filled with random packing) has been checked. Two basic types of hydrophobic catalysts, ie catalysts being hydrophobic by nature and catalysts being hydrophilic by nature but made hydrophobic, have been tested in this work. As an example of a catalyst of the first type, a catalyst consisting of platinum (0.8w.%) coated on styrene-divinylbenzene copolymer was prepared. A catalyst of the second type was prepared by platinum (1w.%) coating on activated charcoal (19w.%) and then mixing with PTFE (80w.%). The performance of LPCE columns filled with catalysts of both types, together with inert packing made of stainless steel mini-spirals, was tested at different ratios R and showed HETP ≈0.25m [10]. The hydrogen throughput was limited to ≈20mol. $(H_2)/m^2 \cdot s$ by the column flooding. This result, being similar to earlier reported [7, 11, 12] values for pilot-scale facilities, confirms the reproducibility of the LPCE column performance. A noticeable increase in throughput at high effectiveness of isotopic exchange is expected for columns filled with the structured inert packing, which incorporates the same catalysts. Such packing has been developed and are being tested.

Testing of sieve-tray columns for KOH retention additionally equipped with layers of packing of high effectiveness for water distillation and gas scrubbing, and evaluation of a VPCE reactor performance under the real gas flow-rates and pressure are foreseen in the programme. The above-mentioned equipment to be developed and tested is shown in Fig.2 within the boundary of 1MPa pressure. No research and development were planned for cryogenic distillation columns. Their design is well developed with the operation experience already available at the AGH plant [1]. A final stage of the experimental programme will be the construction and testing of the pilot-scale facility for processing of 10 tonne a year of the JET tritiated water waste. This feed flow rate covers the current rate of tritiated water generation. When upgraded to full-scale, the detritiation facility should process all the water to be generated by EDS and by the various waste detritiation facilities, providing tritium discharge much below the limit adopted for the AGH plant.

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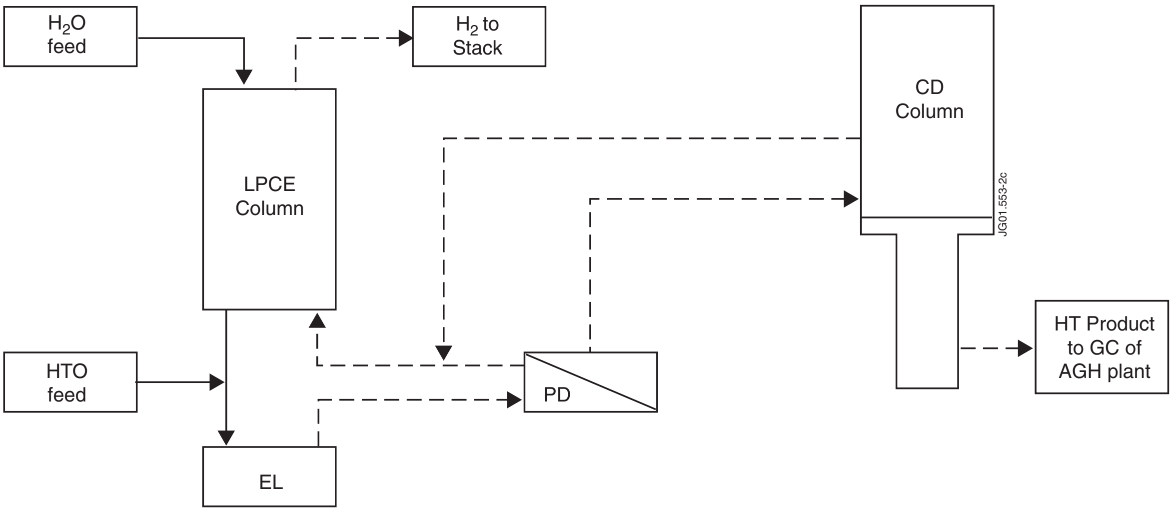

***Fig. 1:** Schematic of the JET water detritiation facility*

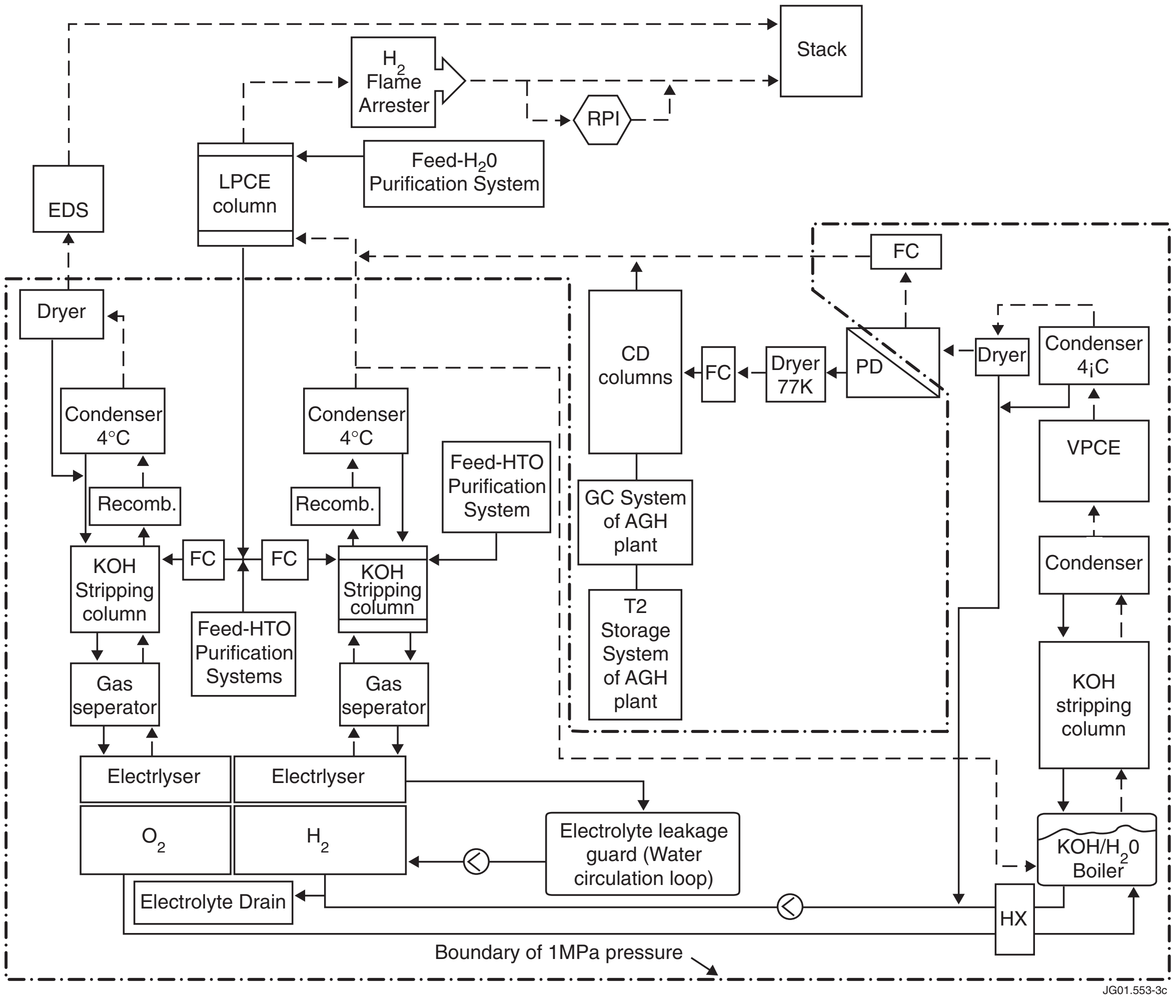

***Fig. 2:** Flow diagram of the water detritiation facility.*