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A Systematic Approach to the Design of a Large Scale Detritiation System for Controlled Thermonuclear Fusion Experiments

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ABSTRACT.

The most common basic process of air detritiation employing oxidation of tritiated gases in a catalytic recombiner and subsequent collection of HTO on molecular sieve driers can also be used for a large scale detritiation system for the next step DT fusion device. Performance, economy and reliability can be improved by modifying the design of basic elements ie recombiners and molecular sieve driers, and by re-arranging them in a system permitting multiple process path choices for optimum performance depending on demand.

The proposed system is shown to be:

- free of secondary tritium releases by permeation,
- economical, with less than 1KW power required in a ready-to-operate "Hot Standby" condition,
- capable of reducing inlet humidity in the order of 10.000ppm (Volume) to 0.01ppm at the outlet by using two adsorber stages in series,
- capable of providing the best starting condition for water processing: little or no dilution by H₂O from isotopic swamping due to use of two adsorber stages.

The system detritiation factor DF is defined and discussed, demonstrating the overriding importance of high water retention efficiency.

Introduction

In the past, design and implementation of tritium removal systems was usually required as a remedial action to mitigate the hazards of tritium releases caused by defaults in process design and equipment (eg chronic leaks, permeation, lack of reprocessing facilities etc). Nowadays improvements in tritium handling technology permit to avoid many of these emissions at source.

The Next Step Device to demonstrate the viability of Controlled Thermonuclear Fusion - ITER - will make use of all know "best practicable means" to control potential emissions at source. It can, however, be expected that mandatory safety studies will demonstrate a finite probability of systems failure leading to tritium releases. Anticipation of such events requires implementation of a detritiation system. As a consequence, a relatively large plant will be conceived to cope with accidents that may or may not occur, with the requirement to be on continuous standby duty.

A similar situation was encountered at JET and led to the design of an Exhaust Detritiation System (JET-ED) [1] capable of dealing with up-to-air accidents of the JET Torus vacuum vessel and Neutral Beam Injector vacuum vessels at maximum tritium inventory (30 grams T₂). With a large plant on standby for this duty, it was considered economical to route all other (minor) potential release sources (mainly secondary containments of process equipment and several rooms containing equipment with substantial inventories) to this plant with the argument that the probability of serious release accidents arising simultaneously from more than one source would be acceptably low. An analysis of systems planned for ITER is likely to lead to similar conclusions. However, for a plant of substantially larger size, improvements in economy of operation , performance and reliability both in design of critical process elements and plant layout must be considered. This paper is intended to point out critical areas and to recommend solutions.

Main Features of Detritiation Systems

Tritiated gaseous effluents containing Q_2 (Q stands for any of the three hydrogen isotopes, ie $Q_2 = H_2$, HD, HT, D₂, DT, T₂), hydrocarbons CxQy, water Q_2O and other chemical compounds of Q, are reacted in a catalytic recombiner with Oxygen (air) to form primarily Q_2O and oxides of the other elements involved (eg CO, CO₂). A good summary of suitable catalysts and their main characteristics (reaction rate as function of temperature, aging effects etc) is given by Ellis [2]. All catalysts suitable for this purpose require operation at elevated temperatures. After the gas mixture has passed the recombiner, virtually all tritium is oxidised to Q_2O , which is removed from the air stream by chilled condensers followed by molecular sieve driers. For large capacity driers, disposal of saturated adsorber beds is not economical (or the adsorbed water may contain tritium in quantities suitable for reprocessing), hence saturated driers are regenerated in situ by heating and condensation of the evolving Q_2O on a chilled condenser. This general process is widely used and has proven to be simple, efficient and reliable.

Experience with systems such as the JET-ED designed for the same purpose and similar duty cycle (maximum design capacity rarely required) has shown that improvements in component design and plant layout are desirable and possible in order to cover the wide range of operating conditions economically while providing maximum safety for the worst case scenario. These improvements will be discussed in the following.

Component improvements

Catalytic Recombiner

1 Conventional Design

Conventional Recombiner systems are built from standard discrete elements as shown in Fig 1.

Recombiner 1 oxidises Q_2 to Q_2O at 150°C, Recombiner 2 oxidises C_xQ_y , in particular Methane CQ_4 to CO/CO_2 and Q_2O at 500°C. This conventional approach has several disadvantages:

- the in-line heater being separated from the Recombiner 2 and Recombiner 1 being isolated from the heat source by counterflow heat exchanger HX2 requires continuous air flow (even in absence of Q_2/C_xQ_y) to maintain both recombiners ready for operation in "hot standby".
- The air flow necessary for maintaining "hot standby" is usually running through a drier bed in adsorption, effectively shifting any residual moisture (including HTO) toward the drier bed outlet. This can be expected to result in premature HTO break through.
- The recombiner, heater and heat exchanger process boundary walls are at high temperature with the consequence of potential tritium release by permeation: a small fraction of the HTO formed will re-dissociate on vessel surfaces in areas where the oxide layer develops cracks (especially in the 500°C region) and permeate as H and T.

2 Improved design

An improved recombiner design avoiding all disadvantages of the conventional approach is shown in Fig 2

A counterflow heat exchanger based on standard tube bundle or plate design is partially filled with recombiner catalyst. The upper end is heated by low power density heaters embedded in the catalyst itself. High efficient thermal insulation is fitted on the inside of the process boundary in a metal cladding vented to the process. This design has the following advantages:

- in "hot standby" continuous air flow is not required: thermal conduction will establish a temperature gradient in the bed, similar to full flow conditions.
- Using, eg, 10 cm of Microtherm thermal insulation [3], typical standby power to maintain recombiner catalyst at 500°C is in the order of 0.5 KW for a catalyst volume of 1m³.
- With no air flow, shifting of any moisture inventories
 (including HTO) in subsequent air driers does not take place.
- The temperature gradient in the catalyst below the heated upper zone of the counterflow heat exchanger effects oxidation of more reactive species closer to the recombiner inlet (eg Q₂ at ≥ 80°C), spreading release of reaction energy over a large volume.

<u>Air Driers (AD)</u>

1 Conventional design

As already seen on recombiners, conventionally designed AD systems are assembled from discrete elements. A typical example is the drier assembly of the JET ED system (fig 2) which consists of three identical AD systems in parallel (only AD1 shown in detail) connected to common inlet and outlet manifolds.

In adsorption, Valves 1 and 2 are open, Valve 3 is closed, the heater is off and the blower running. Moist air passes from the drier inlet manifold through the drier bed, the cooler/condenser, and the blower to the outlet manifold and is then discharged to the stack. This particular arrangement has been selected for reasons of keeping all valves near room temperature (also during regeneration). For regeneration, valves V1 and V2 are closed, V3 open, blower running and heater energised. The heater is controlled to heat the recirculating air to 300°C. As the drier bed is heated up, adsorbed moisture is released, recovered in the cooler/condenser and collected in a tank for disposal or reprocessing. The disadvantages of this system are:

- During adsorption, the drier bed is not actively cooled. Experience at JET [4] has shown, that in case of high inlet humidity and high flow rate (8°C dewpoint, 500 m³/h) the sorption heat released in the mass transfer zone raises the bed temperature by 10 to 30°C This is most likely to lead to premature tritium breakthrough by displacement of residual HTO contamination - HTO roll-up [5] toward the bed outlet
- During regeneration, the total energy required for heating and desorption is supplied by an external heater. Heat transport by

means of air requires a correspondingly high mass flow rate (≥ 300 m³/h) for an acceptable duration of the regeneration cycle (approximately 8 hours). This is in conflict with efficient moisture collection: at the end of the regeneration cycle, the air leaves the bed and enters the cooler/condenser with a temperature of 300°C, leaving it with a temperature of 40°C, hence, the final regeneration is done using air with 40°C dewpoint recirculating through the bed. This in turn leads (after cooldown) to a residual moisture content of $\geq 3\%$ by weight on the sorbent, more or less equally distributed through the bed. Even if the cooler/condenser would have met its design performance of 15°C outlet temperature, the residual moisture content would still be $\approx 2.5\%$. It is again clear, that in case of HTO contamination immediate tritium release proportional to HTO contamination in a subsequent adsorption run is bound to occur.

The only way to remove residual contamination would be the introduction of large amounts of tritium-free H₂O for isotopic swamping during the final regeneration phase. The resulting dilution of collected HTO by H₂O would however, severely increase difficulties for waste disposal or potential reprocessing. For both reasons, it is desirable to avoid the necessity for isotopic swamping.

2 Improved design

An air drier of improved design is proposed in Fig 4.

As in the proposed advanced recombiner (Fig 2), the process boundary is at room temperature, the thermal insulation being contained in a metal cladding inside the vessel and vented internally at the "dry" side (=top) of the vessel.

A heating/cooling device is embedded in the sorbent. Heating or cooling will be supplied from external sources via a suitable heat transfer medium. A cooler/condenser is arranged at the lower end of the vessel. Flow direction in adsorption is upwards, in regeneration downwards. Since the energy for heating the sorbent and desorption of Q_2O is directly transferred to the sorbent, a much smaller air flow is required to transport released moisture to the cooler/condenser. For regeneration, dry air (DP \leq 60 °C) from the drier outlet manifold can be "borrowed" (since only approximately 10% of the air flow as compared to conventional systems is required) to achieve full regeneration with negligible residual humidity on the sorbent. This will make isotopic swamping, except in cases of very heavy HTO contamination, during regeneration unnecessary. At the end of the regeneration run cooldown is performed without air flow, which might otherwise lead to undesirable HTO deposition in the top layer of the bed. The proposed design is more complex than the conventional. However - as will be shown later - this is justified by the necessity for high water retention efficiency.

System layout

A system making full use of the improved characteristics of the proposed recombiner and drier designs is shown in fig 5.

This system is presented using a method which has proven very flexible for developing complex flow diagrams containing multiple elements with multiple process path choices for the JET Active Gas Handling System - the matrix presentation: two rows of pipes running at right angles to each other in two parallel planes can be interconnected by valves at crossover points; in pipe runs between valves, process elements are inserted as required. Depending on which valves are open or closed, multiple process paths within the matrix can be

selected. Elements can be simply named (and subsequently very easily located) by their function and coordinates in the matrix, eg:

VA 1-2 can connect the lines X=1 and Y=2

AD 5-2/4 is an Adsorber on line X=5, connected between lines Y=2 and Y=4.

The sequence of X or Y coordinates can also be used to indicate flow direction: AD 6-4/0 is an Adsorber on line X=6 with flow from line Y=4 to line Y-0.

A further advantage is the possibility to directly use this type of flow diagram as the operator interface mimic between plant and control system. As shown in fig 5, closed values are represented by an open circle (\equiv no connection made) and open values by filled-in circles (\equiv connection made). Global status of process elements may be presented by colour (eg green = running OK, red = defective/not available, blue = ready for use etc) and more information may be called up via cursor on auxiliary pages.

The clear presentation makes it easy for the operator to select a suitable process path, while control system hardwired or software interlocks would prevent him from committing errors such as bypassing recombiners via VA 0-1, VA 3-1, VA 3-2 etc.

Normal Operation

Air to be detritiated enters the system at point 0-0. It passes through a diagnostic block D0-0/1 where parameters of interest are measured (Tritium concentration, humidity, flow rate, pressure etc). After valve VA 0-1, a cooler/condenser CC 1/2-1 removes excess humidity to approximately 2-3°C dewpoint. Recombiner RE 1-1/2 (or RE 2-1/2 as redundant spare) oxidizes tritiated compounds to HTO. A cooler/condenser CC 3/5-2 reduces humidity to

2-3°C dewpoint. A (optional) refrigerated condenser RC 3/5-2 may be energised in series to reduce the dewpoint to -20°C. This would increase the adsorption time of the subsequent adsorber(s) by a factor of 7 in case of maximum production flow, in particular during an emergency requiring maximum design flow rate. A further diagnostic block D 3/5-2 measures dewpoint, flow rate etc. The air then passes through the first stage adsorber AD 5-2/4, reducing the air dewpoint to \leq -60°C. A diagnostic block D7/8-4 detects humidity and tritium breakthrough after the first stage adsorber bank. The second stage adsorber AD8-4/6 buffers any accidental humidity excursion at the outlet of the first stage adsorber bank. Normally loaded with inlet air at dewpoint \leq -60°C and regenerated alternatingly with their own output in counterflow, the second stage driers should achieve an outlet dewpoint in the order of -100°C. A bank of blowers B 5,6,7 - 6/7 provides main system flow, ranging from zero to maximum design flow. Flow rate/blower speed is controlled to maintain the system inlet pressure measured at D0-0/1 subatmospheric. The blowers can be selected for specific duties: a rather small fan being sufficient to maintain system at underpressure in standby with zero flow, while the other two cover the full range of design flow conditions.

Drier regeneration

The regeneration path for AD 6 is shown (dotted line) in fig 5: dry air enters the heated AD 6-4/0 via VA 6-4. The main fraction of recovered water is collected in the integral cooler/condenser and routed to water collection. The exiting air of dewpoint approximately 3°C is driven by the regeneration blower BL3-0/2 (the second blower shown is a redundant backup) through CC and RC 3/5-2 and AD 5-2/4 in a closed loop. Only a small fraction of the recovered water is thus shifted to the adsorbing drier AD 5-2/4 (less than 5% if system is correctly tuned) to be recovered during regeneration of AD 5-4/0. Regeneration of second

stage driers AD8 and AD9 is done accordingly. In case of heavy HTO contamination, the last phase of regeneration could make use of isotopic swamping by admitting tritium-free moist air through VA 6-3, followed by final dry-out.

Recombiner decontamination/recirculation

During operation of the JET ED System it was found that the recombiner catalyst carrier has a non-negligible water sorption capacity: if dried with recirculating dry air from system outlet when hot and subsequently cooled, the air at the recombiner outlet reaches a dewpoint \leq -80 °C. In case of work on recombiners, contamination must be removed prior to work (eg catalyst replacement). With the recombiner to be decontaminated in hot standby, dry air from VA8-4 can be recirculated through the recombiner and the first stage adsorbers to remove residual humidity (HTO) from the recombiner. A short recirculation loop through recombiner(s) only could also be selected to achieve efficient processing of small gas batches by efficient mixing and multiple passage through the recombiner(s).

Instrumentation

Standard instrumentation is available for all parameters (pressure, flow, temperature, humidity) with one exception: for very low humidities (dewpoint \leq -60°C to -70°C) no reliable and accurate instrument exists, which means that the full performance especially of the second stage adsorbers cannot be demonstrated by hygrometry; final performance tests can only be done with HTO using tritium monitors of adequate sensitivity.

An additional set of instrumentation was found useful at JET: temperature profile monitors (15 thermocouples spaced by 10 cm each)

embedded in the adsorbers give very valuable information on location, width and movement of the mass transfer zone during adsorption and regeneration. At high adsorption rates, this information can be used to predict humidity breakthrough before the outlet humidity rises.

Reliability and maintenance

The recombiner design with very low power density heater and the drier design with external source for heating and cooling ensures virtually unlimited life of critical process components. The detailed mechanical design of these units should foresee easy and safe replacement of catalyst and sorbent. Further redundant units (easy to add in the presented flow scheme as required) can be foreseen if systems analysis shows risk of deterioration (eg by poisoning of catalyst) in case of low probability accidents.

The most vulnerable components appear to be the blowers; therefore redundant blowers were suggested and they can even by replaced with the system in full operation provided the mechanical design permits the necessary access and foresees suitable, reliable and safe couplings.

Detritiation Factor - definition and discussion

Detritiation systems are usually characterised by their Detritiation Factor DF which is defined by the ratio of tritium activity at the system inlet to tritium activity at the system outlet.

For systems dealing with HTO collection only, the definition is quite simple: assuming a water collection efficiency η (fraction of water arriving at inlet, retained in the adsorber bed) gives the DF for HTO:

$$DF_{HTO} = \frac{1}{1 - \eta} \tag{1}$$

If a recombiner is used in series and a mixture of various tritiated gases (including HTO) passes the system, the situation is a bit more complex: we assume an input mixture with a tritiated water fraction W_0 and a convertible fraction C_0 of other tritiated gases, both expressed in relative tritium activity such that

$$W_0 + C_0 = 1 \tag{2}$$

We now attribute a conversion efficiency ε to the recombiner for catalytic oxidation of convertible gases to HTO. Assuming further, that the original water fraction W₀ passes the recombiner without being altered, we obtain at the recombiner outlet a water fraction W_R of

$$W_{\rm R} = W_{\rm o} + C_{\rm o} \varepsilon \tag{3}$$

and a non-converted fraction C_R of

$$C_{\rm R} = C_0 (1 - \varepsilon) \tag{4}$$

The water fraction W_R will be adsorbed in the subsequent drier with the efficiency η , while the unconverted fraction C_R passes through the drier.

With the usual definition of DF (total tritium at inlet divided by total tritium at outlet), we obtain the "Total" detritiation factor

$$DF_T = \frac{W_o + C_o}{C_R + W_R (1 - \eta)}$$
(5)

or, using equations (2), (3) and (4)

$$DF_T = \frac{1}{1 - \eta(W_o + \varepsilon C_o)}$$
(6)

If we take into account the relative biological radiotoxicity of the outlet mixture, which is of major importance for personnel working in the immediate vicinity of the plant, we have to weigh the water (HTO) outlet contribution fully, but we now can omit the usual simplification to count all outlet tritium as HTO. This is justified by the fact that any tritiated species having passed the recombiner unconverted at 500°C can well be expected to have a biological effect not exceeding that of HT.

With this justification, we can define a "Biological" DF, where the effect of the non-converted species C_0 is suppressed by the radiotoxicity ratio of $\frac{\text{HTO}}{\text{HT}} \ge 10000$, which gives

$$DF_{Biol} = \frac{1}{1 - \eta(W_o + \frac{\varepsilon C_o}{10000})}$$
(7)

or - neglecting insignificant terms,

$$DF_{Biol} \approx \frac{1}{1-\eta}$$
 (8)

which is just the water retention efficiency of the plant.

This demonstrates that for plant personnel the ED water retention efficiency η is by far more important than the conversion efficiency ε . This was the main reason to propose a system with particularly efficient water retention. The benefit for humans at close range, however, should not divert us from looking for high efficiency in recombiners as well: eventually, all tritiated species may end up as HTO or organically bound tritium, albeit spread over a much larger area.

<u>Conclusions</u>

It has been demonstrated, that air detritiation systems - by implementing the recommended modifications of conventional elements and improved system layout - can meet the requirements of large DT fusion experiments : reliable "hot standby" operation with minimum power consumption, negligible secondary tritium release and water (including HTO) removal to 0.01 ppm (by volume). The high water retention efficiency is achieved by using two drier stages regenerated in counterflow with dry air. This minimises the requirements for isotopic swamping, thus reducing the amount of collected water and the size of further systems required for water reprocessing. With a maximum capacity designed for an event with low probability, this plant may also be used - as the JET ED system - for treating potentially contaminated effluents from a large number of minor sources. The actual plant size and layout, including the capacities of individual components will have to be determined on the basis of detailed safety studies covering all potential releases from the various next step device subsystems using the design principles presented as a guideline. It appears worthwhile to give a further example for the flexibility of this approach : assuming the safety study would predict a relatively frequent release of small batches with high tritium concentration (as might be expected from waste handling/treatment operations), plant performance for such occasions could be improved further by adding one or two small diameter/extended length adsorber bed(s). The sorbent in these beds could be refrigerated using the built-in cooling device for even higher water retention efficiency.

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Fig 1 Conventional recombiner system as used at JET [1]; thermal insulation (not shown) by lagging of individual components.



Fig 2 Improved recombiner design: counterflow heat exchanger integrated with catalyst and heater, process boundary at room temperature.



Fig 3 Conventional adsorber system as used at JET [1]; thermal insulation (not shown) by lagging of individual components.



Fig 4 Improved adsorber design: heating/cooling device embedded in sorbent, water condenser integrated, process boundary at room temperature.



Fig 5 Flow diagram of detritiation system based on improved component design, presented in matrix form. One out of 36 main process paths and one out of 10 regeneration loops is highlighted. In addition, choice of many other operating modes (eg recombiner recirculation, use of components or blowers in parallel for maximum flow rate) is evident.

ANNEX

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