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WPTFV-PR(17) 18065

B Butler et al.

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Preprint of Paper to be submitted for publication in
Fusion Engineering and Design



This work has been carried out within the framework of the EUROfusion Consortium and has received funding from the Euratom research and training programme 2014-2018 under grant agreement No 633053. The views and opinions expressed herein do not necessarily reflect those of the European Commission.

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Applicability of a Cryogenic Distillation System for D-T Isotope Rebalancing and Protium Removal in a DEMO Power Plant

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Abstract

The tritium plant at the future European DEMOnstration (DEMO) fusion power plant will require isotope separation systems for protium removal and isotopic rebalancing of the torus exhaust stream. Protium removal and isotopic rebalancing can occur within a single separation technology which is based upon the exploitation of isotopic effects; the minimum amount of exhaust gas processing can be calculated using a heuristic developed for this purpose. A model has been developed to quantify the size and separation ability of a cryogenic distillation system to perform the required separation duties. Comparison of a reference scenario and six alternative scenarios shows that the isotope imbalance ratio between deuterium and tritium has the largest effect on the size and separation ability of a cryogenic distillation system; provided that a trace amount of protium is acceptable within the gas introduction system into the torus. A cryogenic distillation system is found to be a suitable technology in terms of energy usage and isotope separation, but a single system size is not suitable to process a range of different compositions. This evaluation of the applicability of cryodistillation to isotope rebalancing and protium removal for a DEMO reactor, form a base case against which other hydrogen isotope separation technologies can be investigated and compared in terms of size, power requirements and safety¹.

Introduction

Cryogenic distillation (CD) is an established method of separating hydrogen isotopes for industrial processes. In the future demonstration fusion power plant (DEMO), providing ~2GW thermal power, if a burn-up efficiency of DT fuel of 1-2% is assumed, of the order of 100 kg of hydrogen isotopes are expected to be recycled through the reactor each day [CITATION But152 \l 2057]. Fusion will occur between deuterium (D) and tritium (T) isotopes, and this ratio of isotopes must be maintained to a set point to achieve optimal fusion parameters. However, isotopic effects or a requirement to vary the D-T ratio of gas injected into the torus may result in an imbalance of isotopes coming from the torus exhaust. Furthermore, unwanted protium may enter the exhaust stream through steel outgassing or air leaks.

Part of the role of the tritium plant that will support the torus will be to redress these imbalances and to limit any protium that enters the fuel cycle. These system blocks of the tritium plant are shown in Figure 1; isotope rebalancing and protium removal occur immediately after exhaust processing. The suitability of a CD system to fulfil the duties of these system blocks is investigated, and the investigation will be used to compare quantitatively other technologies against this established one.

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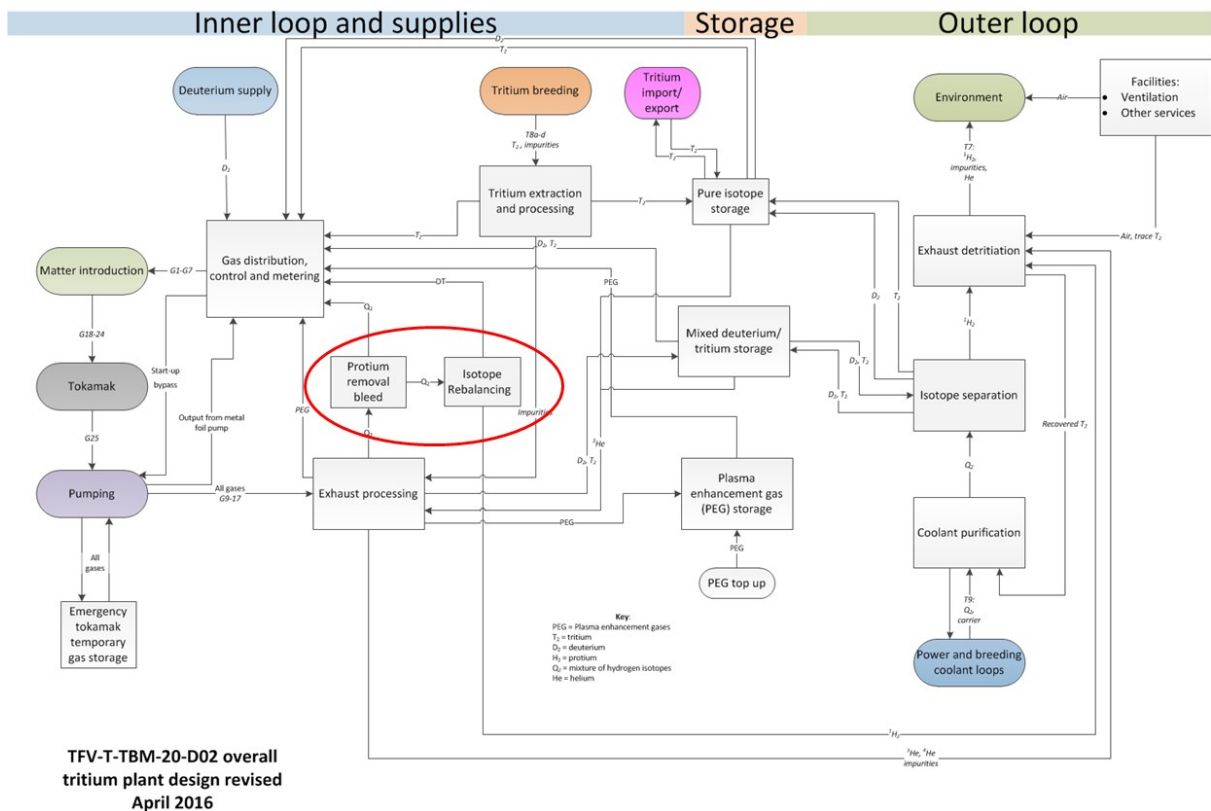


Figure 1: DEMO Tritium Plant Architecture [CITATION But15 \ | 2057]

During this work, it was found that the functions of isotope rebalancing and protium removal could be combined into one system, provided that less than approximately 10% of the DT from the tokamak exhaust is continuously processed. In this analysis, the combined model of the isotope rebalancing and protium removal system consists of two sub-systems:

- a) A gas flow splitter that directs a small fraction of the DT flow to the remainder of the system block which consists of:
- b) A combined arrangement of CD columns that both perform the function of substantively separating deuterium and tritium and also removal of protium

Outputs from the protium removal and isotope rebalancing systems are likely to comprise:

1. A route to the gas distribution, metering and control system – the majority of hydrogen isotopes recovered from the torus exhaust are directed along this path
2. An enriched deuterium or tritium feed to the gas distribution, metering and control system used to perform the isotope rebalancing function
3. A complementary enriched tritium or deuterium gas flow which will need to be directed to a storage facility for future use
4. A waste steam to the exhaust detritiation system (EDS) through which protium is removed from the gas stream

Over 90% of the matter processed by these systems will be returned as mixed isotopes to the gas distribution, control and metering system (GDCM). A small matter stream will be sent to EDS, where residual trace tritium will be removed and the remaining gas will be vented to atmosphere. There will be a stream of a pure isotope, either deuterium or tritium, that is sent to storage for introduction into the GDCM system at a future date; the order of magnitude of the purity of this stream will depend upon the

size of the separation system. Important quantifiable bounding parameters in developing feasible protium removal and isotope rebalancing systems include:

- The amount of protium removed from the exhaust stream. The rate of protium removal must equal, at a minimum, the rate at which it enters the torus and exhaust stream.
- The amount of tritium sent to the stream going to the EDS. The tritium plant will have limits on the amount of radioactivity that can be discharged to atmosphere, and the EDS will also have a certain detritiation factor (DF). The protium-rich stream must contain less tritium than the total allowable tritium input to the EDS.
- The amount of energy required to run the system. The overall efficiency of DEMO will depend upon the amount of energy required to keep it running; therefore, a system that requires prohibitive amounts of energy to separate isotopes will not be feasible.

Other criteria will also affect the choice of technology used for the protium removal and isotope rebalancing system. These criteria are not quantifiable at this stage of the DEMO tritium plant design, but comparisons can be made between different systems and technologies to identify a preferable option. These criteria include:

- The tritium stored within the system. Minimising tritium inventory is a main principle of the design of a DEMO tritium plant. Smaller systems contain less tritium. For a CD system, information on operating limits and column design decisions, such as packing types, is required to estimate a tritium inventory within the system. Tritium within CD systems is primarily stored as a liquid, and the size and design of the column reboiler and liquid sump can influence the tritium inventory by orders of magnitude.
- The radiological safety of the system. Technologies which retain or store tritium in a safe mode in accident or upset conditions are preferable to those in which a tritium release is possible.

Calculation Methods

Protium removal and isotope rebalancing can be combined into a single isotope separation system by assuming a bounding argument by one of the system blocks. The amount of gas sent through the protium removal and isotope rebalancing systems affects the size and design of each of the systems. The flows can be approximated in an ideal system, *i.e.* without taking into account separation efficiencies or isotopologue effects, by considering the exhaust isotope amounts and the desired ratio of those isotopes flowing back through the matter injection system into the torus. The most efficient fusion occurs when there are equal amounts of deuterium and tritium [CITATION But15 \ | 2057]; therefore, a base case was considered where the feed into the main GDCM system consisted of equal amounts of the two isotopes.

Isotopic imbalance may occur in a number of ways that would result in the exhaust gas being enhanced in one isotope. Deuterium may be injected into the torus in order to control the edge of the plasma or to mitigate disruptions [CITATION Kru15 \ | 2057]. It is possible, although less likely, that tritium would be used instead of deuterium. Other potential sources of pure deuterium include pellet injection and neutral beam injection. Estimates from Butler *et al.* indicate that over 90% of the gas entering the GDCM system will be an equal mix of deuterium and tritium isotopes [CITATION But151 \ | 2057].

An idealised formula for calculating the amount of gas sent through the protium removal and isotope rebalancing systems can be found by considering the composition of the exhaust stream, S , in $\text{mol}\cdot\text{h}^{-1}$, at steady state. The stream will contain $X \text{ mol}\cdot\text{h}^{-1}$ of isotope A, and slightly more of the enhanced isotope B, denoted by $(X + \delta)$. Protium in the stream is designated by α . Therefore:

$$S = 2X + \delta + \alpha$$

The exhaust stream is then split into two streams, a feed stream F and a recycle stream R . The excess amount of isotope B (δ) must be removed by the isotope rebalancing system. Therefore:

$$x_B F = \delta$$

Given that the exhaust stream will comprise gases, the whole stream can be assumed to be well mixed. The mol fraction of B in the exhaust stream is calculated by considering the relative amount of isotope B in the entire exhaust stream:

$$x_B = \frac{S_B}{S} = \frac{X + \delta}{2X + \delta + \alpha}$$

As such, the minimum flow of the feed stream is calculated by considering that δ must be the product of the isotopic mole fraction and the flow of gas through the isotope rebalancing system, designated by F :

$$F = \frac{\delta}{x_B} = \frac{\delta}{X + \delta} (2X + \delta + \alpha)$$

Figure 2 shows how the flows are calculated and split for this idealised separation system; an excess deuterium scenario can be modelled as easily as an excess tritium scenario. The figure shows the flowrates of each H , D , and T in the exhaust stream, the feed stream, and the recycle stream.

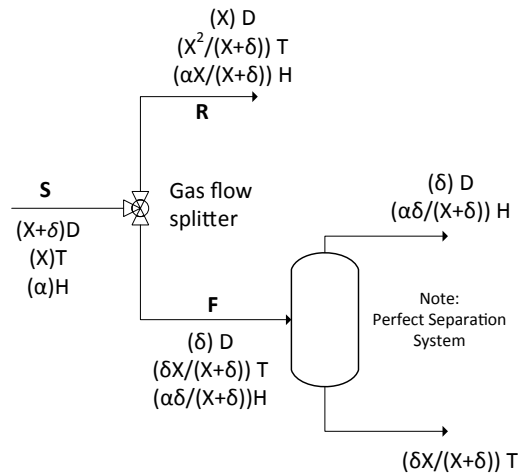


Figure 2: Split before Separation Method for Excess Deuterium

There must be a sufficiently high flowrate of F such that the amount of protium in the stream exceeds the amount that enters the torus, β :

$$x_H F = \frac{\alpha\delta}{X + \delta} \geq \beta$$

This equation was used to calculate the concentration of protium within the system, α , which is higher than β , as well as to check that the designed isotope separation system removes sufficient amounts of protium. It has been assumed that there is no upper limit for α ; all scenarios showed that α was over two orders of magnitude smaller than the flows of deuterium and tritium. It was found in all scenarios that isotope imbalance was the bounding calculation case.

Separation efficiencies and isotopologue effects result in incomplete separation by any isotope separation system. Separation within a CD system is driven by the vapour-liquid equilibrium within the columns. The flows from these calculations can be used as a starting point for more detailed calculations using calculated or experimentally derived separation factors.

This heuristic is applied to simple modelling which is commensurate with the present level of knowledge of the DEMO tritium system. The method is not limited to simple modelling and can be applied to both other technologies and other methods of modelling. The simple calculations can be applied to other technologies, such as gas chromatography (GC) or thermal cycling adsorption processes (TCAP) to allow for cross comparison between potential separation systems. An initial estimate of flowrates and concentrations also can be vital when undertaking detailed modelling using specialist software.

Protium and Tritium Removal Limits

The rate of protium ingress into the exhaust gas stream was estimated from literature values of steel outgassing rates and scaled measured values of air ingress from the Joint European Torus (JET). Metal outgassing rates depend upon many factors including metal type, finishing, thickness, treatment, and temperature. Avdiaj and Erjavec estimate average outgassing rates from stainless steel in vacuum pressures between 10^{-11} and 10^{-13} mbar.l.s⁻¹.cm⁻² [CITATION Avd12 \ | 2057]. Given an estimated surface area of the torus of 1084 m²[CITATION Dou13 \ | 2057], and using a conservative estimate of 10^{-11} mbar.l.s⁻¹.cm⁻², the amount of protium entering the system from steel outgassing was estimated at **$1.08 \cdot 10^{-5}$ Pa.m³.s⁻¹, or $1.60 \cdot 10^{-5}$ mol.h⁻¹**. Similar hydrogen outgassing rates have been recorded for tungsten [CITATION KBa15 \ | 2057]. Water entering the DEMO torus due to air ingress was estimated to be is **$1.64 \cdot 10^{-3}$ Pa.m³.s⁻¹, or $2.42 \cdot 10^{-3}$ mol.h⁻¹** by scaling from known JET tokamak leak rates[CITATION Dou13 \ | 2057], considering conservative conditions of 30 °C heat with 80% humidity and improvements in welding technology over time. It is assumed that all of the protium within the water becomes gas; the change in deuterium or tritium due to this exchange is considered negligible given the overall flow rates of those isotopes. Combination of these values gives a total reference protium ingress rate of **$1.64 \cdot 10^{-3}$ Pa.m³.s⁻¹, or $2.44 \cdot 10^{-3}$ mol.h⁻¹**; this is the minimum amount that must be removed by the protium removal system.

The maximum amount of tritium allowed to leave the stream is dictated by the DEMO plant safety requirements document (PSRD) [CITATION Joh15 \ | 2057]. The PSRD states that a maximum of 333 TBq.y⁻¹ of tritiated water would be allowed to be sent to atmosphere. Although the EDS is expected to have a detritiation factor of over 1000, the design of the tritium plant must assume a much lower detritiation factor in order to comply with ALARP principles and to prepare for potential accident scenarios when the EDS may not operate to its full capacity. Therefore, a detritiation factor of ten is assumed for this work; calculations show that a total of 3330 TBq.y⁻¹ of tritiated water would be allowed to be sent to atmosphere. Assuming 97.4 TBq.g⁻¹ HTO [CITATION Shm95 \ | 2057], the maximum amount of tritium sent to EDS was estimated to be about **$1.8 \cdot 10^{-4}$ mol/hr**.

Scenarios and Modelling

The calculation method shown in the previous section was used to calculate the minimum required matter flows through a CD system to remove protium and rebalance deuterium and tritium isotopes. These calculations assume perfect separation; although unfeasible, they were used as an initial starting point against which ProSim iterated to find a feasible solution.

A reference scenario was developed based upon the most likely estimate of the torus exhaust gas stream flow and composition [CITATION But15 \ | 2057]. Six alternative scenarios were also considered and

compared to the reference scenario in order to evaluate the robustness of the system and to identify the parameters to which the system is most sensitive. Table 1 shows the reference scenario and alternative scenarios with the minimum required flowrates for each isotope as calculated by Equation 2. These values are used within ProSim to give an initial estimate of the CD system duties for each scenario.

Table 1: Scenarios

Name	Description	F _H (mol/hr)	F _D (mol/hr)	F _T (mol/hr)
Reference Scenario (DRef_NoSplit_RefH)	This is the reference scenario defined in this work. It uses the average expected flow through the DEMO fuel cycle [CITATION But151 \l 2057], which results in an excess of deuterium.	0.002	0.887	0.882
No input split (DRef_NoSplit_RefH)	The same overall flows are considered as in the reference scenario, but the entire flow is sent through the system rather than just the feed side stream.	0.502	184.2	183.2
Excess Tritium Reference TRef_Split_RefH	An excess of tritium is considered rather than an excess of deuterium, making this the tritium reference scenario. The same proportion of excess tritium is considered as excess deuterium is for the reference scenario.	0.002	0.882	0.887
Cryostat Environment (DRef_Split_NoAir)	The reactor is considered to be kept under a cryostat environment , therefore no air and humidity can leak into the gas stream. This assumption results in a lower flow of protium in the system.	2.14*10 ⁻⁵	0.887	0.882
High Deuterium Flow (DHigh_Split_RefAir)	A high deuterium flow , 10% excess, is considered in this scenario.	0.002	18.3	16.7
Higher Overall flows (DRef_Split_HighAll)	10% higher overall flows of gas through the system is considered in this scenario, but the relative amounts of each isotope are the same as for the reference scenario.	0.003	0.975	0.971
High Deuterium and Protium Flows (DHigh_Split_HighH)	Both a 10% excess of deuterium and protium through the system are considered in this scenario.	0.003	18.3	16.7

In all scenarios, there is a high amount of deuterium and tritium compared to protium going through the gas stream. The reference scenario redresses an isotopic imbalance of less than 1%, whereas two alternate scenarios investigate the effect of a 10% isotopic imbalance. The increase in imbalance has an immediate effect in the minimum flows required to be sent to the separation system.

One scenario assumes the torus is kept in a cryostatic environment. In this case, the only protium ingress will come from the outgassing rates of stainless steel. One other scenario considers the effects of having outgassing rates of 10⁻¹⁰ mbar.l/(s.cm²), *i.e.* an order of magnitude higher than the maximum estimated outgassing rate. The input flows show that the overall change to the feed stream from this change is negligible.

ProSim was used to model the CD system; a bespoke thermodynamic package for hydrogen isotopes based on Souers' model was incorporated into the package [CITATION Pro14 \l 2057]. This package previously has been used to develop and design two CD columns for hydrogen isotope separation as part of the new Water Detritiation System (WDS) at JET [CITATION Lef15 \l 2057].

Results

Reference Scenario

A single CD column was found not to give sufficient separation of the isotopes. A system design consisting of three CD columns in series, as shown in Figure 3, was developed.

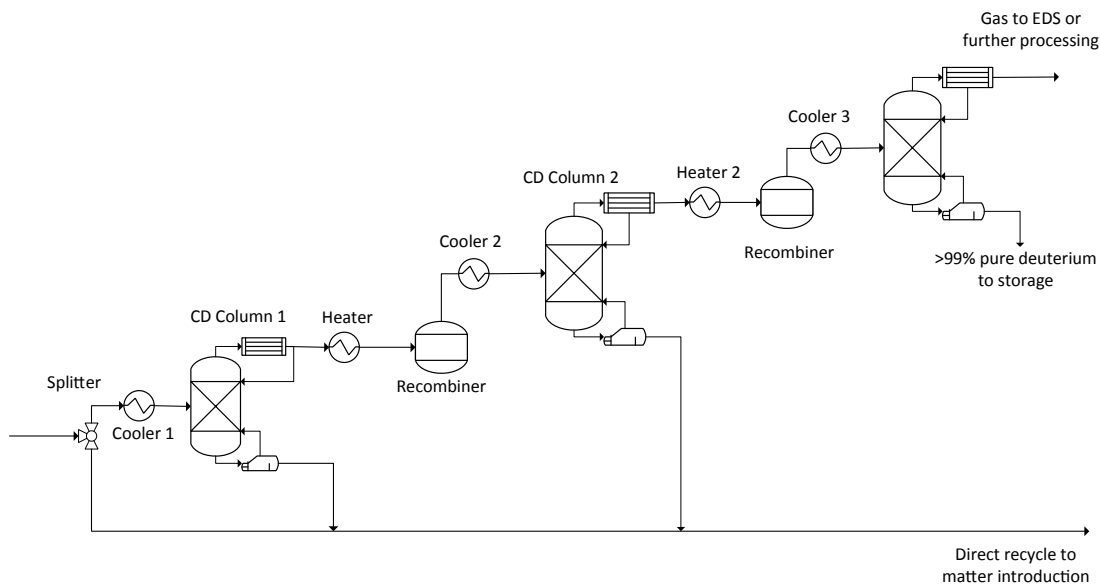


Figure 3: Three Column Design for Excess Deuterium Scenarios

Between the columns are catalytic reactors at room temperature which promote reactions to create single-isotope isotopologues (*i.e.* H_2 , D_2 , and T_2 instead of HD , HT , and DT) [CITATION Bai99 \l 2057].

It was not possible to investigate the sizes and tritium holdup of the distillation columns at this stage of preliminary design. Size estimation occurs when more detailed parameters such as packing material and operating ranges have been fixed; however, column parameters such as height and reflux ratio were minimised in order to decrease the expected size of each column.

At steady state, the majority of the required energy for the system goes into cooling the gas to cryogenic temperatures; therefore, the amount of energy required is directly proportional to the input to each distillation column. Table 2 shows the set of parameters used to gather baseline information on the energy requirements and separation efficiencies of this reference scenario.

Table 2: Optimal Operating Parameters for Reference Scenario Design

Variable	Value in Optimum Design
Reflux ratio of column 1	17
Reflux ratio of column 2	7.5
Reflux ratio of column 3	6
Number of ideal plates in column 1	60

Variable	Value in Optimum Design
Number of ideal plates in column 2	70
Number of ideal plates in column 3	60
Feed stage in column 1	30
Feed stage in column 2	48
Feed stage in column 3	50

The resultant amounts of energy required by the system, as well as the amount of tritium and protium removed to EDS, are shown in Table 3. The total power required is calculated by translating the amount of power required to create one watt of liquid helium cooling power and adding this amount to the electrical energy required by the heaters. A baseline conversion of approximately 200W/1W(He) was calculated.

Table 3: Criteria Values for Reference Scenario

Criteria	Value in Optimum Design	Units	Description
Cooling Power (provided at 20K)	-26.3	W	Cryogenic cooling power required
Total electric Power	5.3	kW	Overall energy required
Protium Removal	6.8×10^{-03}	mol/hr	Protium removed from system
Tritium Removal	2.1×10^{-05}	mol/hr	Tritium removed from system

The results in Table 3 show that the amount of tritium removed from the system is approximately one tenth that which is allowable from the entire tritium plant. Furthermore, a total of 5.3kW is estimated to be required to give a suitable separation of all three isotopes; 5.3kW is less power than existing or developing CD systems used for large scale hydrogen isotope separation. The process CD system at JET requires nearly 200W of cryogenic cooling power [CITATION Bai99 \ | 2057], and the CD column in development for the JET WDS requires approximately 100W of cryogenic cooling power [CITATION Lef151 \ | 2057]. This decrease in power requirement compared to existing columns can be attributed to two main points:

- By treating the input to the system as a bleed stream from a larger cycle, the amount of gas going through the entire system is minimised.
- An exact composition of the bottom products from the first two columns in series is not needed. Excess deuterium has been removed, but supplies from pure or mixed isotope storage facilities can be used to balance the isotope ratios before the stream is re-injected into the torus.

Alternative Scenarios

The majority of the alternative scenarios focussed upon different amounts of excess deuterium going through the isotope rebalancing and protium removal system. Expected fuelling scenarios indicate that pure deuterium may be used in pellet injection and neutral beam systems; the use of excess tritium would contravene the principle of minimising the overall amount of tritium used at DEMO. In this work, one scenario considers excess tritium removal in order to investigate the potential changes in processing design and procedure; a different system configuration is required as shown in Figure 4. This configuration is only

used for the excess tritium scenario, whereas the configuration shown in Figure 3 is used for all excess deuterium scenarios. This required layout change shows that a single layout of a CD system cannot be used for different excess reagent scenarios.

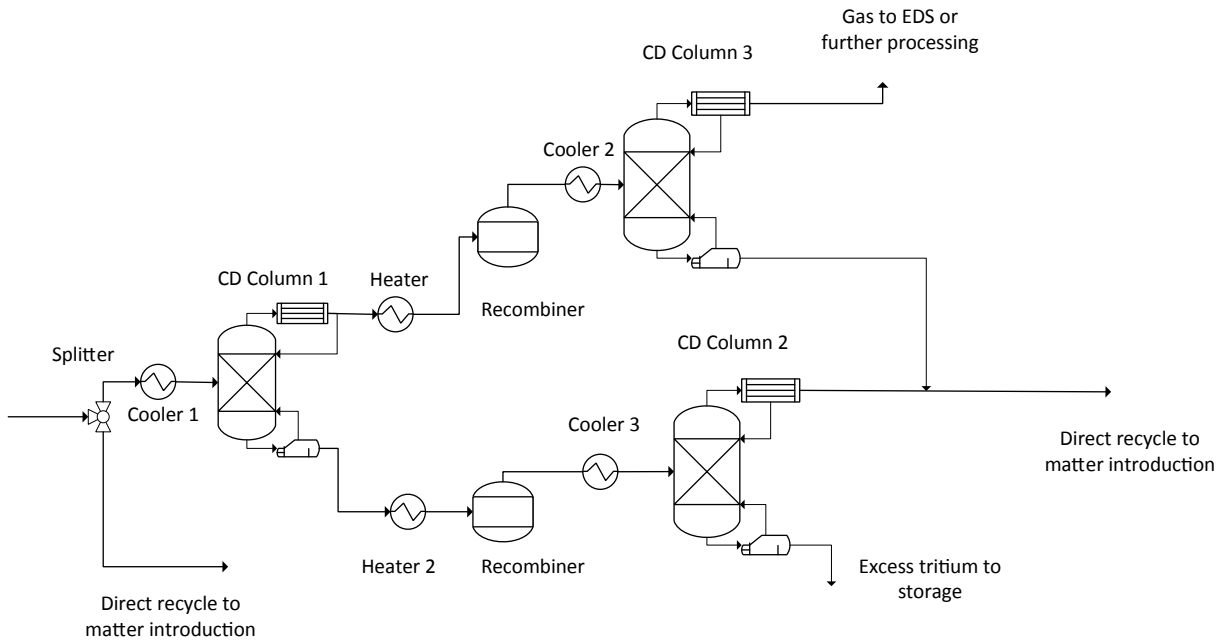


Figure 4: Three Column Design for Excess Tritium Scenarios

Sensitivity analyses were performed on the design parameters shown in Table 3 in order to investigate the suitability of the design parameters of the reference scenario on the alternative scenarios. For those in which the feed flow into the system was similar, the same column parameters could be used to achieve suitable criteria values. However, for the alternative scenarios in Table 1 in which the feed flows are significantly greater than those in the reference one, the column parameters were not suitable. Given that the reference parameters were not suitable for all alternative scenarios, parameters were chosen in terms of optimisation for each individual scenario.

Table 4: Quantifiable Criteria Values for Alternative Scenarios

Scenario	Net Cryogenic Cooling Power (W)	Total Required Power (kW)	Tritium Removal to EDS (mol/h)	Protium Removal to EDS (mol/h)
Reference Scenario (DRef_NoSplit_RefH)	-960	190	1.3×10^{-3}	0.05
Excess Tritium Reference (TRef_Split_RefH)	-28.2	5.6	6.5×10^{-5}	4.8×10^{-3}
Cryostat Environment (DRef_Split_NoAir)	-23.3	4.7	1.4×10^{-4}	4.1×10^{-5}
High Deuterium Flow (DHigh_Split_RefAir)	-432.7	86.5	1.3×10^{-4}	5.8×10^{-4}
Higher Overall flows (DRef_Split_HighAll)	-32.1	6.4	3.1×10^{-5}	8.7×10^{-3}
High Deuterium and Protium Flows (DHigh_Split_HighH)	-432.7	86.5	1.3×10^{-4}	6.5×10^{-4}

Table 4 shows that the amount of cooling power required is a strong function of the isotopic imbalance rather than the required protium removal. The scenarios in which the isotopic imbalance was greater resulted in significantly higher input feed flows and, therefore, input cooling requirements. The table also shows that the amount of tritium removed in the no-split scenario exceeds the amount that is an allowable input into the DEMO EDS.

Discussion

The results from the reference and alternative scenarios show the effect of processing a fraction of the exhaust stream rather than the entire amount. When the system acts as a bleed stream from the main fuel cycle rather than an integrated step, there is a reduction in the amount of cooling power required and also in the amount of tritium sent to the EDS by nearly two orders of magnitude for the reference scenario. The scenarios in which a 10% isotopic imbalance is considered show a higher cooling power requirement than those with 1% or lower isotopic imbalances.

The difference in criteria values for these scenarios is greater than the cryostat environment scenario. Although this scenario shows a decrease in energy requirement, the total amount of tritium removed to EDS is increased over fivefold. In terms of tritium release and safety, this result shows that operating the torus in a cryostat environment causes a deficiency of protium such that the output to EDS has an increased amount of tritium present. By allowing a small amount of protium into the stream, separation between deuterium and tritium becomes more effective due to the wider number of isotopologues that form within the system. Therefore, the tritium in the protium stream to EDS decreases. It may therefore be necessary to inject a small amount of protium into the input of the CD system in order to ensure a minimum amount of tritium reaches the EDS.

The results from all scenarios highlight the fact that different levels of isotopic imbalance result in largely different gas flows for processing. In the cases of different overall flowrates with a similar imbalance ratio, the variation in required cooling power and system feed input is small. If isotope imbalances of greater than approximately 3 or 4% are expected in DEMO, a single CD system is unlikely to be suitable. Cryogenic distillation columns have a fixed range of flows over which they can operate; low flows result in plate weeping whilst high flows can result in column flooding, which results in uneven gas flows or a pressure in the column due to nonstandard flows.

The modelling focused on quantifiable criteria against which the individual scenarios could be compared. These quantifiable criteria give an indication as to how to compare the currently non-quantifiable criteria of tritium hold-up and radiological safety. Minimising the flow through the combined isotope rebalancing and protium removal system will both decrease the tritium hold-up and increase radiological safety.

Conclusions

A generic CD column system has been designed within ProSim in order to estimate separation efficiencies and energy requirements of different scenarios for an isotope rebalancing and protium removal system within a DEMO tritium plant. A method has been developed to minimise the amount of gas going through the separation system based upon permitted tritium release levels and expected protium ingress rates. For a reference scenario of excess deuterium and average air ingress rates into the torus volume, a system using three separate distillation columns and 5.3 kW of total power to cool and separate the isotopes can be designed to meet all separation requirements. Sensitivity analyses from alternative scenario development show that energy requirements are more sensitive to the ratio of isotope imbalance than the

amount of protium ingress. However, the removal of protium from the system results in higher amounts of tritium being sent through the EDS, implying that if the torus is operated in a cryostat environment, injection of additional protium into the CD system input will be required.

The amount of energy and input feed into the system is dependent upon the level of isotopic imbalance within the system. The results show that if large swings of isotopic imbalance are expected during DEMO operation, a single CD system is unlikely to manage the required separation. A single CD system also could not manage variation between excesses of tritium or deuterium, although processing practices can mitigate this issue.

The modelling of the scenarios put forward in this work is an initial investigation to assess the basic feasibility and approximate size of a CD system; the details given in the reference and alternative scenarios have been used to compare the scenarios to each other, but these details can also be used to compare the CD technology as a whole against other similar technologies, such as GC or TCAP separation methods. The current available modelling techniques for such similar technologies are still in simplified forms.

Future work will focus on applying different technologies to these scenarios and comparing the resultant criteria values. In particular, gas chromatography (GC) and thermal cycling absorption processes (TCAPs) are considered viable alternatives to CD systems. Both of these technologies work on the basis of temperature swing separation, which can result in large energy costs. CD systems rely upon cryogenic cooling, whereas GC & TCAP technologies are considered to have a lower risk of tritium release as the tritium is stored as a solid at atmospheric conditions. This work shows that CD is a feasible though inflexible option for isotopic rebalancing and protium removal at DEMO, and it also gives a baseline against which other technologies can be compared.

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