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# Fusion Engineering and Design

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# Water distillation for coolant purification system of DEMO water-cooled lithium lead breeding blanket



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ARTICLE INFO	A B S T R A C T
<i>Keywords:</i> Water detritiation Primary coolant Tritium recovery Tritium inventory	During the DEMO pre-concept design phase, a review of existing and under development technologies suitable for the Coolant Purification System (CPS) of the Water-Cooled Lithium Lead (WCLL) Breeding Blanket (BB) was carried out and two tritium recovery strategies were individuated, named on-line and off-line. The on-line concept relies on the continuous by-pass and treatment of a certain primary coolant fraction into CPS whereas the off-line scheme foresees the discharge of the whole primary water inventory after one year of DEMO oper- ation, that will be treated in an external facility inside the tritium plant. Among these, the on-line strategy has been recently recommended as the most suitable for lowering the tritium inventory within BB and Primary Heat Transfer System (PHTS). Furthermore, such solution does not need high decontamination factor since treated water is not discharged in the environment, but it is redirected again into PHTS. In such condition, Water Distillation (WD) could be a promising technology with intrinsic simplicity and safety. The present work deals with the feasibility assessment of WD for DEMO WCLL BB CPS, investigating its capability to meet water CPS duty. A parametric study has been preliminary carried out, analysing the effect of selected parameters (i.e., tritium concentration at CPS inlet, tritium concentration in the enriched stream, efficiency of anti-permeation barriers, and operative conditions) on the WD sizing and operation. Then, a preliminary reference design has been individuated and verified with a dynamic model developed in MATLAB environment. Finally, the process flow diagram of the system has been derived and a preliminary sizing of auxiliary components is provided.

### 1. Introduction

Tritium permeation from Breeding Blanket (BB) into primary coolant is a relevant concern for DEMO operation. Once permeated into Primary Heat Transfer System (PHTS), tritium can migrate towards working areas and environment, due to permeation and leaks. To limit such possibility, two mitigation strategies are under investigation for DEMO, involving the use of specific coatings (i.e., anti-permeation barriers and/ or natural oxide layers) and the treatment of a certain portion of the primary flow rate in the Coolant Purification System (CPS), whose main function is to recover tritium permeated into PHTS keeping tritium concentration within primary coolant below design limits [1]. The present paper deals with the preliminary sizing of a CPS for the Water-Cooled Lithium-Lead (WCLL) BB variant, based on Water Distillation (WD) technology.

WCLL is one of the two BB concepts under investigation for DEMO, relying on pressurized water as coolant and Lithium-Lead (enriched at 90% in <sup>6</sup>Li) as breeder and neutron multiplier [2]. The function to remove power from WCLL BB is accomplished by two independent water-cooled systems: the Breeding Zone (BZ) PHTS and the First Wall (FW) PHTS [3].

The needs to operate the machine with efficient electricity production and to demonstrate the tritium self-sufficiency (i.e., large heat transfer areas and high tritium generation rate into BB, respectively) pose favourable conditions for tritium permeation towards PHTS. Such phenomenon is emphasized in the WCLL BB variant. Once permeated from BB into PHTS, tritium is trapped into primary coolant as HTO, due

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https://doi.org/10.1016/j.fusengdes.2023.113547

Received 11 October 2022; Received in revised form 10 January 2023; Accepted 4 February 2023 Available online 8 February 2023

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*Abbreviation*: BB, Breeding Blanket; BZ, Breeding Zone; CCWS, Component Cooling Water System; CECE, Combined Electrolysis Catalytic Exchange; CLM, Column; CND, Condenser; CNL, Canadian Nuclear Laboratories; CPS, Coolant Purification System; CVCS, Chemical and Volume Control System; ECO, Economizer; FW, First Wall; HCPB, Helium-Cooled Pebble Bed; HETP, Height Equivalent to one Theoretical Plate; ODE, Ordinary Differential Equation; PCS, Power Conversion System; PHTS, Primary Heat Transfer System; PRF, Permeation Reduction Factor; PWR, Pressurized Water-cooled Reactor; RBL, Reboiler; SG, Steam Generator; SPE, Solid Polymer Electrolyte; TER, Tritium Extraction and Removal System; TNK, Tank; VLE, Vapor-Liquid Equilibrium; WCLL, Water-Cooled Lithium-Lead; WD, Water Distillation; WDS, Water Detritiation System.

Nomenclature				
В	Bottom flow rate			
D	Distillate flow rate			
F	Feeding flow rate			
FT,CPS	CPS tritium removal rate			
FT,p	Tritium permeation rate			
K	Vapor-Liquid Equilibrium ratio			
L	Reflux flow rate			
Μ	Liquid holdup			
Р	Pressure, Pa			
R	Reflux ratio			
Т	Temperature			
V	Boil-up flow rate			
Greek sy	mbols			
α	Relative volatility			
η	CPS efficiency			

to isotopic exchange reaction between HT and  $H_2O$ . Therefore, HT partial pressure remains very low within the primary coolant, enhancing further tritium permeation. On the other hand, such phenomenon can be considered as a positive aspect in terms of safety, since primary coolant acts as tritium sink. Indeed, HTO does not permeate towards working area and external environment. The only quantifiable release from PHTS is due to leaks [1].

Concerning the water CPS, in the pre-conceptual design phase a review of existing and under development facilities for water detritiation was carried out, posing emphasis on size, complexity, and costs of such technologies. A relevant outcome of the analysis was the need for antipermeation barriers into BB region, in order to limit CPS flow rate within feasible range. Such a technology has been tested at laboratory scale, demonstrating the possibility to operate with a Permeation Reduction Factor (PRF, defined as the ratio between permeation rate through bare walls and permeation rate through walls equipped with anti-permeation barriers) up to 10 000 [4]. Nevertheless, its behaviour under neutron irradiation relevant for DEMO BB must yet be verified [5]. Furthermore, two strategies were investigated for the CPS: the on-line and the off-line schemes. The first one relies on a continuous by-pass of a certain fraction of the primary flow rate, that is routed and treated within the CPS. On the other hand, the off-line strategy, adopted in CANDU reactors, consists in the overall replace of the primary coolant with fresh water after a certain period. Discharged coolant is treated in a dedicated facility in the tritium plant [1]. Although the off-line approach could benefit of the overall simplification of the system, along with less stringent issues in the size of the CPS technologies, a sensitivity analysis has recently demonstrated the need to adopt an on-line CPS in order to reduce tritium inventory within BB and PHTS [5]. For this reason, the on-line strategy has been considered in the present work.

Among the available technologies for water detritiation, two processes have been recently reviewed for DEMO water CPS: the Water Distillation and the Combined Electrolysis Catalytic Exchange (CECE). WD is widely used in CANDU heavy water upgrader. Although presenting significant advantages (e.g., simplicity and safety), WD provides relatively low decontamination efficiency. These features make such solution promising for the on-line strategy, where high decontamination factor is not mandatory. On the other hand, CECE provides high decontamination efficiency, while relying on a more complex technology. Nowadays, the strategy is to adopt WD for the water CPS and CECE for the Water Detritiation System (WDS) [6].

#### 2. Review of outcomes from tritium permeation analysis

A proper design of the CPS starts from the update of tritium mass balance in the primary coolant. For this purpose, most recent outcomes of the tritium permeation analysis have been reviewed.

In this regard, Moreno and Urgorri performed a parametric study investigating the effect of some selected features on tritium permeation in WCLL and Helium-Cooled Pebble Bed (HCPB) BB concepts [5,7]. The possibility to adopt off-line or on-line strategy was firstly investigated for water CPS. A relevant outcome was that tritium permeation is not influenced by CPS strategy as well as by the amount of primary flow rate routed in CPS. As a matter of fact, tritium permeated into primary coolant is trapped as HTO, thus the HT partial pressure remains very low, allowing further permeation. In case of off-line solution, tritium concentration increases linearly until the complete water is discharged, whereas the on-line strategy allows to keep an equilibrium concentration when tritium permeation rate is balanced by CPS removal rate. The equilibrium value depends on the CPS efficiency as well as on the CPS mass flow rate.

Assuming a PRF equal to 100, tritium permeation rate from PbLi through BZ walls is calculated as 0.42 g d<sup>-1</sup>, whereas the one towards FW PHTS is evaluated to be 9.28 mg d<sup>-1</sup>. This latter considers only tritium coming from breeder; an additional contribution is foreseen from plasma chamber through the FW and Moreno and Urgorri hypnotized a first tentative value of 20 mg d<sup>-1</sup>, set as boundary conditions of their calculations. In any case, contribution of FW PHTS can be considered negligible compared to BZ PHTS.

Further parameters were the subjects of the parametric study [7]. PRF was varied between 1 and 1000, evaluating a linear dependence of permeation rate with such parameter. The efficiency of the Tritium Extraction and Removal System (TER) was also investigated, changing between 80 % and 95 % (assuming 82 % as reference). This parameter influences tritium concentration in PbLi and, thus, permeation rate towards PHTS. Nevertheless, the range of study is quite limited, and its effect is not very impactful in the results. The last parameter investigated is the H<sub>2</sub> addition into coolant. This is a common practice already adopted in Pressurized Water-cooled Reactors (PWRs) to compensate O2 formation in primary coolant due to radiolysis. H<sub>2</sub> addition presents two main effects: (i) the increase of tritium permeation by enhancing HT recombination on the wall surfaces (such phenomenon is limited by the presence of anti-permeation barriers), and (ii) the increase of the permeation towards the Power Conversion System (PCS), via Steam Generator (SG) walls. As a matter of fact, H<sub>2</sub> addition displaces chemical equilibrium towards HT and T2 molecules, leading to higher concentration of such species in primary coolant and, thus, higher permeation through SG walls.

## 3. Water distillation technology

Distillation is a separation method that involves vapor and liquid phases at the same pressure and temperature in the coexisting zone of the device. The contacting medium for the two phases can be plates, trays, or packings (random or structured), forming the distillation column. Feed flow rate is introduced at one or more levels along the column. By gravity, liquid moves downward and vapor upward through the column, providing a counter-current contact between the two streams. In such a flow pattern, liquid and vapor approach thermal, pressure, and composition equilibrium along the column, depending on the efficiency of the contacting device. The lighter (lower-boiling) components tend to concentrate in the vapor phase, thus in the upper part of the column (rectifying section), whereas the heavier (higher-boiling) components in the liquid phase, thus in the lower part (stripping section) [8]. Internal flow streams are kept by a reboiler and an overhead condenser. Reboiler vaporizes a certain fraction of the liquid flow, providing the boil-up in the column. The rest of the liquid flow rate is withdrawn as bottom product, enriched in the heavier components. In the total condenser

configuration, the overhead condenser condenses the whole vapor flow coming from the column's rectifying section. Part of the condensate is returned in the column as reflux and the rest is withdrawn as distillate, enriched in the lighter component.

Water Distillation is a particular application of distillation technology, involving water as feeding. It is a promising technology for water detritiation purposes in applications where a very high decontamination efficiency is not required. This is due to the quite low relative volatility between light water (H<sub>2</sub>O) and lightly tritiated water (HTO), in the range of 1.11 and 1.03 between 278 K and 363 K [9]. For this reason, aiming at maximizing the separation factor, WD columns are operated under vacuum conditions (typically between 10 kPa and 30 kPa [10]). Thus, WD presents the safety advantage of low temperature and sub-atmospheric pressure, reducing the eventuality of tritium release [11]. Furthermore, WD does not require electrolyser, no need of pumps, no oxygen scrubbing, and no issues related to hydrogen explosion hazard. On the other hand, the main disadvantage is the larger number of stages (thus, larger dimensions) in comparison with more efficient technologies such as CECE.

#### 3.1. Sizing procedure

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The sizing methodology proposed in this work relies on assumptions summarized as follow. It is based on the graphical method proposed by McCabe and Thiele [12] for binary mixture. Feed stream is supposed to contains only H<sub>2</sub>O (the lighter species) and HTO (the heavier component), thus neglecting other species involving deuterium and tritium. It is considered an acceptable assumption since deuterium is not foreseen in the feeding stream, except for negligible concentration, and tritium content is so low that the amount of T<sub>2</sub>O can be neglected. The column is divided in multiple stages, following McCabe-Thiele method, and, for each stage, the Vapor-Liquid Equilibrium (VLE) is assumed for all the species. For the *i*<sup>th</sup> species the VLE ratio is defined as  $K_i = y_i/x_i$ , where  $x_i$ and  $y_i$  are mole fractions of  $i^{th}$  species for liquid and vapor phases, respectively. For an ideal mixture Raoult's law and Dalton's law are valid. Thus,  $K_i = P_i^0/P$ , where  $P_i^0$  is the vapor pressure of the pure  $i^{\text{th}}$ component and P is the total pressure, whereas the relative volatility between  $i^{\text{th}}$  and  $j^{\text{th}}$  components ( $\alpha_{ii}$ ), defined as the ratio of  $K_i$  and  $K_i$ , is  $\alpha_{ii} = P_i^0 / P_i^0$ . An empirical formula for the evaluation of H<sub>2</sub>O to HTO relative volatility was proposed by Van Hook [9]:

$$ln\alpha = ln\frac{P_{H_2O}}{P_{HTO}} = \frac{37\ 813.2}{T^2} - \frac{136.751}{T} + 0.124096\tag{1}$$

where T is temperature in K. For the  $i^{th}$  species, the composition of vapor  $y_i$  in equilibrium with liquid of composition  $x_i$  is:

$$y_i = \frac{\alpha x_i}{1 + (\alpha - 1)x_i} \tag{2}$$

The VLE could be considered a reasonable description of trayed column. A larger margin of error is expected for packed column [13]. Nevertheless, such calculation gives relevant information for the sizing which later will be refined with safety margins in terms of dimensions.

As presented in Eq. (1),  $\alpha$  depends on temperature and, thus, on the pressure. During operation, the reference pressure is kept at the top of the column. Due to pressure drops, pressure rises descending along the column and the amount of the increase depends on the selected contacting medium. A proper evaluation of the relative volatility is mandatory for a good sizing of the WD column, since  $\alpha$  is close to 1 for H<sub>2</sub>O-HTO system. For this reason,  $\alpha$  is evaluated at the top and the bottom of the column and an average value is assumed in the design procedure.

Other assumptions are constant liquid and vapor flow rate above and below the feeding level, adiabatic conditions, and negligible heat mixing and sensible heat effects.

Fig. 1 provides a schematic view of the WD column considered in this



Fig. 1. Layout of the WD column.

analysis. The column is divided in several stages (*N*), the inlet water flow (*F*) with a tritium concentration ( $x_{F, HTO}$ ) enters at the feeding stage ( $N_F$ ). Along the column, the liquid interacts with the vapour phase, therefore the tritium concentration in the liquid phase increases towards the bottom of the column while the one in the vapour phase decreases along the top. At the top, all vapour is condensed, of this a certain fraction is routed back into the column ( $L_{top}$ ) while the rest (*D*), having a tritium concentration  $x_{D,HTO}$ , is sent back to the PHTS. At the bottom, the liquid is collected in the reboiler, here a certain fraction is vaporized and sent back to the column ( $V_{bottom}$ ), whereas the rest, with a tritium concentration  $x_{B,HTO}$ , is extracted as liquid (*B*) and directed to the WDS.

According with the layout reported in Fig. 1, the overall mass balance and the  $i^{th}$  component mass balance of the WD column are described by the following equations:

$$F = D + B \tag{3}$$

$$Fx_{i,F} = Dx_{i,D} + Bx_{i,B}$$
(4)

where *F* is the feed flow rate, *D* the distillate flow rate, and *B* the bottom product flow rate. In the present work, feed stream is considered exclusively as liquid, thus  $x_{i,F}$  represents the total content of the *i*<sup>th</sup> component. *F*, *D*, and *B* compose the flow rates entering and leaving the column.

The proposed sizing procedure needs a set of boundary conditions that are: tritium permeation rate into coolant  $(F_{T,p})$ , tritium concentration in the feeding water column  $(x_{F,HTO})$ , and tritium concentration in the water leaving the column, the one at the bottom,  $(x_{B,HTO})$ . Tritium concentration in the feeding stream corresponds to the design value assessed in water primary coolant whereas the one at the bottom is linked to the WDS features. In these conditions, the CPS removal rate, calculated as *B* times  $x_{B,HTO}$ , must balance the tritium permeation rate towards PHTS.

Considering Eq. (4), the minimum *F* to be routed into CPS is calculated assuming  $x_{D,HTO}$  equal to zero, being  $x_{F,HTO}$ ,  $x_{B,HTO}$ , and *B* fixed. This represents the ideal conditions for which distillate flow rate is completely purified from HTO species or, in other words, the condition at which the tritium removal efficiency is 100 %. Such an operation would require a very large number of stages and, thus, extremely high columns. For this reason, the analysis shown in the following section presents results for different feed flow rates, higher than minimum *F*.

Once boundary conditions and external flow rates are defined, two methods are available to obtain desired separation: increase the number of stages of the column (i.e., increase column height) and increase reflux, L (i.e., increase energy consumption). Two extreme cases can be identified: infinite number of stages and infinite reflux. In the case of infinite number of stages, the reflux ratio (R) defined as L to D ratio, is minimum while in case of infinite reflux the number of stages is minimum. Optimal conditions usually derive from an economic analysis. For WD columns, actual reflux ratio is from 1.1 to 1.3 times the minimum R [14]. In the present work, the ratio between R and minimum R is assumed 1.2 and minimum R is calculated with Underwood's equations [15]. Reflux (L) and boil-up (V) represent the internal flow rates and are defined as follows:

$$V_{top} = V_{bottom}$$
(5)

$$L_{\text{bottom}} = L_{\text{top}} + F \tag{6}$$

$$D = V_{top} - L_{top}$$
(7)

$$\mathbf{B} = \mathbf{L}_{\text{bottom}} - \mathbf{V}_{\text{bottom}} \tag{8}$$

where Eqs. (5) and (6) are valid for feeding flow rate in liquid phase.

Once defined internal and external flow rates, along with bottom and distillate compositions, the number of stages needed for the separation and the axial location of feeding are evaluated with the graphical method proposed by McCabe and Thiele [12].

The number of stages corresponds to the number of plates for a trayed column. In case of packed column, the Height Equivalent to one Theoretical Plate (HETP) expresses the efficiency of packing material and the overall height of the column can be obtained as HETP times the number of stages. Packed column is preferable when high number of stages is required for the separation. This is the case of WD, characterized by low separation factor. A proper packing must ensure high efficiency of separation, together with low pressure drops. Several works [10,11,16] suggested CY Gauze Packing, by Sulzer [17], for WD purposes. A variety of correlations was developed for the evaluation of HETP, even if they usually depend on unknown geometrical features of the packing material [18]. In case of CY Gauze Packing, HETP and the characteristic linear pressure drops (pressure drops per unit length, are provided by Sulzer in two diagrams depending on the load factor  $F_c$  [17], defined as:

$$F_c = v_v \rho_v^{0.5} \tag{9}$$

where v is the superficial velocity,  $\rho$  is the density, and subscript v stands for vapor phase. In the proposed design procedure, a liner pressure drops of 3 mbar m<sup>-1</sup> is assumed and the operative  $F_c$  is derived from the SULZER diagram, whose corresponds a certain value of HETP. Several curves of linear pressure drops and HETP as function of  $F_c$  are provided by SULZER for different operative pressures. In the considered range of pressure (see section 3.1.1) the  $F_c$  goes from 1.5 to 1.6 Pa<sup>0.5</sup> (Sulzer recommends a value between 1.5 and 2 Pa<sup>0.5</sup> for CY Gauze Packing), and the associated HETP assumes the almost constant value of 0.11 m.

Column length is obtained as HETP times the required number of theoretical stages whereas the column diameter is derived from the column free area *A* calculated as:

$$A = \frac{V_{top}}{F_c \rho_v^{0.5}} \tag{10}$$

It is worth emphasizing that in this preliminary phase of the design, only the packed volume is considered in the column dimensioning. A complete design with selection and sizing of auxiliary equipment (e.g., support plates and liquid-distributing device) that can affect dimension of the column is postponed in a more consolidated design phase.

#### 3.1.1. Parametric study

In the present design phase, interfaces, requirements, and safety considerations are not yet consolidated for DEMO CPS. For this reason, a parametric study has been carried out to investigate the impact of some relevant parameters on the size of the WD column, such as: i) column operative pressure; ii) PRF in the BB side affecting the feeding water flow rate; iii) tritium concentration at the column outlet (bottom) to be sent to WDS, and iv) tritium concentration in the water primary coolant corresponding to column inlet concentration.

Four separated analyses have been carried out. Table 1 provides the ranges and the reference values of the parameters varied during the analysis. Based on the outcomes of the most recent tritium permeation activity [7], bare-wall tritium permeation rate in BB is assumed equal to 42.928 g d<sup>-1</sup>. Given such tritium permeation rate, the allowable tritium concentration in primary coolant of 5 Ci kg<sup>-1</sup>, a PRF of 100, and a CPS efficiency of 100 %, the resulting water CPS by-pass is 34.4 kg h<sup>-1</sup>.

The first analysis investigates the effect of the column operative pressure on its dimensions (height and diameter) as shown in Figs. 2 and 3 (please, refer to the online version of the paper for coloured figures).

As shown in Fig. 2, the column height increases when the feeding flow rate approaches the value of  $34.4 \text{ kg h}^{-1}$ , at which a CPS efficiency of 100 % is required to guarantee the 5 Ci kg<sup>-1</sup> limit in the primary coolant. To avoid such unfeasible column dimensions, a lower efficiency must be considered. The efficiency reduction can be compensated by increasing the water feed flow rate into the column. In fact, for a water feed flow rate greater than 50 kg h<sup>-1</sup> the required column height is lower than 20 m for all the investigated operative pressure. It is worth noticing that a feasible maximum height for a distillation column can be considered 40 m. As a matter of fact, there is experience in employing columns of such dimensions for heavy water distillation [10].

Fig. 3 shows the variation of the column diameter as a function of *F* and pressure. It is worth specifying that, for CY Gauze Packing, the largest diameter supplied to date by Sulzer is 1.8 m [17]. Thus, such value is considered as upper limit and the maximum feed flow rate is obtained accordingly. As expected, the column diameter increases with *F* and the increasing rate is higher for lower pressure (higher volumetric vapor flow rates). Combining the effect of *F* on height and diameter of the column, the optimal condition in terms of packing volume is in the range of 40 and 50 kg h<sup>-1</sup> for every pressure considered. It is worth also mentioning that power consumption for boil-up and condensation increases linearly with *F* and much rapidly for higher pressure. At 50 kg

Table 1
Parametric study and reference case.

Parameter	Unit	Range	Reference value
Operative pressure	kPa	10 - 60	10
Permeation Reduction Factor	-	10 - 1000	100
Tritium concentration in bottom stream	Ci kg <sup>-1</sup>	10 - 1000	100
Tritium concentration in feed stream	${ m Ci}~{ m kg}^{-1}$	0.2 - 5	5



Fig. 2. Column height vs. feed flow rate for different operative pressure.



Fig. 3. Column diameter vs. feed flow rate for different operative pressure.

 $h^{-1}$  of feeding flow rate, the power required for boil-up (nearly the same value for condensation) ranging between 675 kW and 1223 kW, increasing operative pressure from 10 kPa to 60 kPa. For *F* equal to 100 kg  $h^{-1}$ , the power range is 1353-2435 kW.

The second analysis assesses the effect of anti-permeation barriers at BB walls on the column dimensions. Fig. 4 shows the overall packing volume as a function of feeding flow rate for several PRFs (from 10 to 1000). Obviously, the needed feed flow rate decreases by increasing the



Fig. 4. Overall packing volume vs. feed flow rate for different PRF.

PRF due to the reduction of tritium permeation rate at BB. The minimum *F* ranges between 344 kg h<sup>-1</sup> and 3.44 kg h<sup>-1</sup> for a PRF between 10 and 1000, respectively. For the assumed boundary conditions, a relevant advantage in terms of *F* is obtained increasing PRF from 10 to 300, whereas for higher PRF the benefit is less significant. The increase of the overall packing volume is strictly related to the flow rate, since the required flow area is larger for higher flows. Thus, keeping in mind the maximum column diameter of 1.8 m, the increase of *F* requires multiple columns in parallel. For PRF greater than 100, the operation with a single column is ensured keeping *F* below 112 kg h<sup>-1</sup>. The worst case is for PRF equal to 10, for which at least 11 columns are required. Regarding the column height, Fig. 5 shows that feasible dimension (lower than 40 m) is achievable per each PRF, conveniently increasing the feed flow rate. It is worth keeping in mind that the increase of *F* leads to higher energy consumption for boiling and condensation.

Another parameter investigated is the tritium concentration in the bottom stream ( $x_{B,HTO}$ ), responsible of some technological issues. Focusing on the WD column, high tritium concentration in the bottom part could lead to formation of an explosive mixture, due to radiolysis of tritiated water. An acceptable value for the WD technology can be considered 1000 Ci kg<sup>-1</sup> [10]. More stringent requirements could be related to the electrolyser of the CECE technology, foreseen for the WDS. As a matter of fact, bottom product coming from WD column will be an input stream for WDS. The long-term effect of highly tritiated water on a Solid Polymer Electrolyte (SPE) has been recently studied at the Canadian Nuclear Laboratories (CNL), highlighting some issues related to the loss of sufficient integrity of the material composing the electrolyser [19]. For the ITER WDS, it is assumed a tritium concentration at the electrolyser of 500 Ci kg<sup>-1</sup>, expecting a lifetime of 1.5 years for SPE membrane under these conditions [20]. For these reasons, a parametric study has been conducted in the present work on the  $x_B$ , within the range specified in Table 1.

The bottom flow rate reported in Table 2 is calculated to fulfil CPS task, assuming the defined composition. It is worth noticing that low tritium concentration in the bottom stream determines a higher flow rate to WDS. Thus, looking at the interface with the Water Detritiation System, a compromise between tritium content and mass flow rate must be assessed.

The bottom composition does not affect the minimum feeding flow rate but the lower is the separation requirement (i.e., the lower is tritium concentration in the bottom stream) the lower is the required column height (see Fig. 6). The optimal condition in terms of packing volume is in the range of 40-60 kg h<sup>-1</sup> of *F* (see Fig. 7). Under such conditions and for each analysed case, height and diameter of the WD column are well below 40 m and 1.8 m, respectively.

The last parameter analysed is the tritium concentration in the primary coolant and, thus, in the WD feeding stream. Based on the



Fig. 5. Overall packing height vs. feed flow rate for different PRF.

#### Table 2

Flow rate of the bottom stream for different tritium concern	itration.





Fig. 6. Column height vs. feed flow rate for different tritium concentration in bottom stream.



Fig. 7. Packing volume vs. feed flow rate for different tritium concentration in bottom stream.

experience of CANDU reactors, a reference value of 5 Ci kg<sup>-1</sup> has been considered [1,5]. Although in the design activity for DEMO(J05) the limit assumed by JAERI was 10 Ci kg<sup>-1</sup> [21], the tritium concentration considered in the preliminary safety data list for the European DEMO has been 0.2 Ci kg<sup>-1</sup> [22]. Since evaluations about this requirement are still ongoing, for the preliminary assessment of WD as relevant technology for DEMO water CPS, the effect of the feeding composition on the column size has been studied in the range reported in Table 1.

Fig. 8 shows the required packing volume for different feeding compositions and flow rates. For low tritium concentrations, a drastic increase in the overall dimensions and in the feeding flow rates are observed, although the maximum height of 40 m is reachable for each case (see Fig. 9). Assuming the maximum diameter of 1.8 m, the operation with a single WD column is feasible in the range 5-2 Ci kg<sup>-1</sup>. For the case of 1 Ci kg<sup>-1</sup>, at least two parallel columns are needed for the assumed boundary conditions (i.e., operative pressure 10 kPa, PRF 100, and  $x_B 100 \text{ Ci kg}^{-1}$ ). The worst scenario is for 0.2 Ci kg<sup>-1</sup>; in this case a



Fig. 8. Overall packing volume vs. feed flow rate for different tritium concentration in feeding stream.



Fig. 9. Overall packing height vs. feed flow rate for different tritium concentration in feeding stream.

very large feeding flow rate (higher than 860 kg  $h^{-1}$ ) is required to cope with the CPS specifications. At least 6 columns should be required, making WD technology unattractive for CPS purposes. It is worth keeping in mind that, among the existing plants, Darligton Tritium Removal Facility is one of the biggest worldwide, with 360 kg h<sup>-1</sup> of processed tritiated water [1]. A possible solution to fulfil CPS role with WD technology, while keeping the requirement of  $0.2 \text{ Ci kg}^{-1}$  in primary coolant, should be the use of improved anti-permeation barriers. For example, a PRF of 900 at the BB walls would allow operation with a single column, keeping *F* equal to 100 kg  $h^{-1}$ .

Furthermore, it is worth mentioning that the presented design procedure considers steady state conditions. However, as a demonstration plant, DEMO is not foreseen to work continuously over the whole life cycle. Several short and long maintenance phases are expected, in which CPS could works without tritium permeation from BB.

#### 3.2. Verification of the reference design

The reference design of the WD column has been preliminary derived from the outcomes of the parametric study. To date, the boundary conditions assumed for the reference sizing are 10 kPa at the top of the column, the presence of anti-permeation barrier on BB walls with PRF equal to 100, 5 Ci kg<sup>-1</sup> in the PHTS, and 100 Ci kg<sup>-1</sup> in the bottom stream. The main dimensions and the most relevant operative parameters of the reference design are summarized in Table 3. Furthermore, a preliminary design verification has been carried out with Sulcol<sup>TM</sup> 3.5, an open-source code provided by Sulzer [17]. The liquid holdup along the column has been verified to be 5.1 % of the packing volume, corresponding to a tritium inventory of ~0.825 g (around 7938 Ci). Most of this is collected in the bottom part of the column, where water is enriched in tritium.

The final verification of the preliminary design has been carried out by developing a staged dynamic modelling of the column. The rationale is to verify the behaviour of the technology under different operative conditions, following the considerations on DEMO operational phases (see section 3.1.1).

The model is based on the following assumptions: binary mixture involved in the process (i.e.,  $H_2O$  and HTO), constant and uniform pressure along the column, constant and uniform relative volatility, constant and uniform molar flows above and below the feeding stage, no vapor holdup in the packing, and VLE in all stages. Furthermore, the total condenser operation is supposed, that is all the vapor stream exiting the top of the column is condensed [13].

Based on these assumptions, the modelling solves the overall and the components' material balance equations in each stage. Eq. (2) is used as closure law. For the  $i^{\text{th}}$  stage the overall and the  $j^{\text{th}}$  component's mass balance equations are, respectively:

$$\frac{dM_i}{dt} = L_{i+1} - L_i + V_{i-1} - V_i$$
(11)

$$\frac{d(M_{i}x_{i,j})}{dt} = L_{i+1}x_{i+1,j} - L_{i}x_{i,j} + V_{i-1}y_{i-1,j} - V_{i}y_{i,j}$$
(12)

where *M* is the liquid holdup whereas *L* and *V* are defined by Eqs. (5), (6), (7), and (8). The set of Ordinary Differential Equations (ODEs) are solved with the ode15s solver in MATLAB. Since experimental data from industrial scale water distillation device are not available in literature, the modelling implementation has been verified using experimental data from the campaign conducted by Yamamoto and Kanagawa on a laboratory scale WD column [23]. The experimental apparatus consists of a random-packed column 100 cm long and 1.6 cm large, equipped with a boiler, a condenser, and a reflux regulator. The experimental campaign was conducted in total reflux mode: once wetting the packings with distilled water and feeding an initial charge in reboiler (tritiated water of  $1 \times 10^{-8}$  Ci cm<sup>-3</sup>), the column was operated without feeding flow rate and without sampling (except for monitoring) until the steady state. Reboiler and condenser ensured a reflux flow rate of 6.5 g min<sup>-1</sup> under the atmospheric pressure and 100°C. According with Yamamoto and Kanagawa [23], 20 theoretical stages are considered for the modelling. The steady state comparison between experiment and simulation is summarized in Table 4 in terms of tritium concentration at the top of the column and of the ratio between bottom and top tritium concentration.

Geometrical parameters (e.g., number of stages, height and diameter), initial and boundary conditions (i.e., operative pressure, initial

column.

Table 3			
Preliminary	design	of Water	Distillation

Parameter	Unit	Value
Column height	m	11.55
Column diameter	m	0.97
Feeding flow rate	kg $h^{-1}$	45
Bottom flow rate	$kg h^{-1}$	1.72
Distillate flow rate	kg $h^{-1}$	43.28
Distillate composition	$\overline{\text{Ci}} \text{ kg}^{-1}$	1.23
Boil-up	kg $h^{-1}$	927.8
Reflux	$kg h^{-1}$	884.4
Pressure drops	kPa	3.47
Boiling power	kW	613
Condensing power	kW	616

#### Table 4

Verification of the	e modelling with	experimental	data

Parameter	Unit	Experiment [23]	This work
Tritium concentration at the top of column	Ci cm <sup>-3</sup>	$\approx 7\times 10^{-9}$	$7.4 imes$ $10^{-9}$
Ratio between bottom and top tritium concentration	-	pprox 1.75	1.75

holdup, initial composition, external and internal flows, feeding composition, and relative volatility) are derived from the sizing calculations (see Table 3). Moreover, feeding flow rate and tritium concentration in the bottom stream are kept constant, allowing a simplified operation of the WD column and the WDS, respectively, and tritium concentration at the feeding point is changed from 1 to 5 Ci kg<sup>-1</sup>. To do this, a proportional control system has been implemented in the modelling, assuming the *LV*-configuration. Such a control system regulates the internal flow rates (i.e., *L* and *V*) to cope with the operational specifications. Distillate and bottom flow rates change accordingly.

The most relevant steady state results are summarized in Table 5. The designed column shows capabilities to ensure a fixed tritium concentration in the bottom stream, suitably regulating internal flow rates. Bottom flow rate and CPS tritium removal rate ( $F_{T,CPS}$ ) rise with the increase of the tritium content in the feeding stream. With 5 Ci kg<sup>-1</sup>,  $F_{T,CPS}$  balances the tritium permeation at the BB (assuming a PRF equal to 100). Furthermore, Fig. 10 shows the CPS efficiency as a function of the feeding composition, where CPS efficiency is defined as:

$$\eta = \frac{x_{F,HTO} - x_{D,HTO}}{x_{F,HTO}} 100 \tag{13}$$

As expected, WD technology does not allow extremely high decontamination efficiency (instead CECE allows an efficiency close to 100 %) but, as mentioned in section 1, this is not required for an on-line CPS. Thus, Fig. 10 shows that CPS efficiency increases with  $x_{F,HTO}$ , approaching the maximum at the design point of 5 Ci kg<sup>-1</sup>.

#### 3.2.1. Process Flow Diagram

This section describes the preliminary process flow diagram (see Fig. 11) and define the most relevant interfaces of the Coolant Purification System. As presented in Fig. 11, the CPS is not directly linked to the PHTS. BZ and FW PHTS are equipped with a shared Chemical and Volume Control System (CVCS) to which the CPS is directly connected. For this reason, the CPS must fulfill detritiation of both BZ and FW PHTS at the same time.

The CPS processes a small quantity of the flow rate routed within the CVCS. Once redirected into CPS, water stream passes through the economizer (ECO), where it is cool down to the operative temperature of the WD column. Then, a pressure reducer completes the expansion to the operative pressure and the water stream is introduced into WD column (WD CLM). Upstream the feeding point, a line to the vacuum pump is foreseen to evacuate the column down to its operative pressure. As mentioned in section 3, the column is equipped with a condenser (CND) and a reboiler (RBL). The distillate is completely condensed and collected within a dedicated tank (TNK), before to be partially introduced in the column as reflux and partially redirected to the ECO. Here,

Table	5
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Bottom flow rate, tritium content in bottom stream, and CPS tritium removal rate for different tritium content in feeding stream.

$x_F$ (Ci kg <sup>-1</sup> )	B (kg h <sup>-1</sup> )	$x_B$ (Ci kg <sup>-1</sup> )	$F_{T,CPS}$ (g d <sup>-1</sup> )
1	0.177	98.41	0.043
2	0.565	98.83	0.139
3	0.953	99.25	0.236
4	1.339	99.68	0.333
5	1.721	100.3	0.429



Fig. 10. CPS efficiency vs. tritium content in feeding stream.

water stream is warmed accordingly with the CVCS interface. The CND is cooled by water coming from a Component Cooling Water System (CCWS).

Fig. 11 depicts the process flow diagram of the solution relying on a Kettle reboiler. In such a configuration, the whole bottom stream is led into the RBL, where it partially boils. The vapor generated moves in natural circulation towards the column whereas the liquid water is drawn by a pump and sent to the WDS. On the hot side, the RBL is fed with hot water intercepted from the CVCS, just upstream the letdown heat exchanger of such system, and then given back downstream the same heat exchanger.

Focusing on the tritium inventory, together with the WD column, the reboiler represents one of the most critical components, being fed with enriched tritiated water coming from the WD CLM. To assess the inventory of the whole system, a preliminary design of the RBL has been carried out. For this purpose, two configurations widely adopted in WD technology have been considered: the Kettle reboiler and the horizontal thermosyphon. For the sizing, a 10 % tube plugging and a fouling

resistance of  $5 \times 10^{-5} \text{ m}^2 \text{ K W}^{-1}$  have been assumed. The main dimensions, along with the tritium inventory, are summarized in Table 6. It is worth noticing that both the solutions lead to a tritium inventory higher than the WD column. Among the two concepts, the horizontal thermosyphon allows smaller encumbrance and around half the tritium inventory of the Kettle reboiler. On the other hand, in case of horizontal reboiler the level difference with the bottom of the column must be carefully accounted to allow natural circulation. In case of too high distance, water circulation could be ensured with a dedicated pump.

# 4. Conclusions and future perspectives

The work presented in this paper has dealt with a preliminary assessment of the water distillation as a suitable technology for DEMO water CPS. During the pre-conceptual design phase, tritium permeation from BB to PHTS was recognized as a critical issue for WCLL BB concept. In that phase of the design, technologies for water detritiation were reviewed and two strategies (i.e., on-line and off-line) individuated. Among these, the on-line concept allows lower tritium inventory and the use of technologies characterized by lower detritiation factor. In this view, WD can be considered a promising technology due to its intrinsic simplicity and safety.

To date, several interfaces and boundary conditions of the CPS are not yet consolidated. For this reason, a parametric study has been proposed in the present work, aiming at investigating the effect of some relevant parameters on the design of the WD column. They are operative pressure of the column, PRF of anti-permeation barriers on BB walls, tritium concentration in the bottom product and in the feeding stream.

Parameter	Unit	Kettle RBL	Hor. Thermosyphon
Shell diameter	m	0.51	0.27
Tubes length	m	2.68	2.08
Number of tubes	-	66	72
Tritium inventory	g	2.83	1.25



Fig. 11. Process Flow Diagram ..

The main outcomes of this parametric study can be summarized as follows. In general, an optimum range for the feeding flow rate can be individuated, minimizing the required packed volume while keeping reasonable power for reboiler and condenser. Lowering operative pressure leads to higher relative volatility between H<sub>2</sub>O and HTO and, thus, to more compact units. Furthermore, low operative pressure needs lower power for reboiler and condenser, even if higher energy is required for vacuum. Moreover, the efficiency of anti-permeation barriers, expressed in terms of PRF, drastically impacts the design of the WD column. For the assumed reference conditions, a relevant benefit is highlighted by increasing PRF in the range of 10-300. Lower is the PRF, higher could be the number of parallel columns needed for detritiation purposes. Bottom stream composition represents a relevant interface with the WDS. Within the analyzed range, the required composition in the bottom stream does not affect greatly the size of the column. Regarding the interface with WDS, it is worth noticing that lower tritium concentrations at the bottom determines higher flow rates to WDS. On the other hand, the requirement on the allowable tritium concentration into PHTS leads to relevant effects on the column sizing. The considered range, based on the literature review, is guite large. With the reference boundary conditions (i.e., operative pressure 10 kPa, PRF 100, and  $x_{\rm R}$ 100 Ci kg<sup>-1</sup>), a tritium concentration higher than 2 Ci kg<sup>-1</sup> would allow the operation of a single WD column to fulfill the desired detritiation. For 1 Ci kg<sup>-1</sup> at least two 20 m long and 1.5 m large columns would be needed. The worst scenario is for an admissible concentration into PHTS of 0.2 Ci kg<sup>-1</sup>, for which unfeasible dimensioning of the system would be needed, unless improving efficiency of anti-permeation barrier (PRF around 900).

Starting from the outcomes of the parametric study and the definition of relevant boundary conditions, a reference design of the WD column has been derived and verified with a dynamic modelling developed in MATLAB environment. The behavior of the column has been verified under selected reference conditions (i.e., 5 Ci kg<sup>-1</sup> into PHTS) and for lower tritium concentration into primary coolant, following considerations about DEMO operational phases. Assuming an LV-configuration for the control system, the column can provide quite constant tritium content in the bottom product for different concentrations at the feeding. Finally, the preliminary process flow diagram of the CPS has been presented, focusing on relevant interfaces and the individuation of most critical components in terms of tritium inventory.

In conclusion, a feasibility study of WD as relevant technology for DEMO water CPS has been carried out. Although interfaces and boundary conditions must be still consolidated, a preliminary design of the WD column has been obtained. In the following activities, a strong interaction with design teams of the relevant interfaces is foreseen, aiming at the consolidation of the water CPS conceptual design. Furthermore, dynamic simulations will be carried out to investigate capabilities of the system under operational conditions relevant for DEMO (e.g., alternating plasma and maintenance phases). Such simulations could provide useful information in terms of tritium concentration achievable into PHTS.

#### **Declaration of Competing Interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

### Data availability

No data was used for the research described in the article.

#### Acknowledgment

This work has been carried out within the framework of the

EUROfusion Consortium, funded by the European Union via the Euratom Research and Training Programme (Grant Agreement No 101052200 — EUROfusion). Views and opinions expressed are however those of the author(s) only and do not necessarily reflect those of the European Union or the European Commission. Neither the European Union nor the European Commission can be held responsible for them.

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