Fusion Engineering and Design 133 (2018) 64–69

Contents lists available at ScienceDirect

Fusion Engineering and Design

journal homepage: www.elsevier.com/locate/fusengdes

Tritium separation performance of adsorption/exchange distillation tower packed with structured packing

Satoshi Fukada[a,*], Yoshiaki Miho[a,b], Kazunari Katayama[a]

[a] Department of Advanced Energy Engineering Science, Kyushu University, Kasuga-Koen, 816-8580, Japan
[b] Water Treatment Division, Sasakura Engineering Co. LTD., Takeshima, Osaka, 555-0011, Japan

ARTICLE INFO

Keywords:
Tritium
Water distillation
Separation
Wastewater
McCabe-Thiele diagram

ABSTRACT

Separation of tritium from tritiated water is analyzed based on the theoretical plate model comparatively for the three cases among a water distillation tower, the Girdler-Spevack (G-S) bithermal exchange process and the combined electrolysis chemical exchange (CECE) process. The McCabe-Thiele diagrams to design large-scale detritiation systems are drawn for each, and tritium concentration profiles in each system are compared. It is clarified how detritiation behavior in the distillation tower is enhanced by an increase in the separation factor. The number of theoretical stages and internal flow rates to achieve detritiation of the bottom-to-top concentration ratio of $x_B/x_D = 100$ in cases of the G-S and CECE processes, are estimated based on the equilibrium stage separation factor reported in the past.

A water distillation tower of 108 mm in diameter and 1000 mm in height packed with structured packing coated with Zeolite 13X are experimentally tested for detritiation of a large amount of wastewater to be exhausted from nuclear reactors. Separation performance between HTO and H_2O under reduced pressure is experimentally verified in the water distillation tower. Enhancement of the x_B/x_D ratio is experimentally proved under the total reflux condition as a function of evaporation rate. Activation on surfaces of adsorbent coated on the structured packings enhances the stage separation factor. The enhancement ratio is affected by liquid-gas dispersion in the column and flow instability in the packed tower.

1. Introduction

Deuterium (D) and tritium (T) are hydrogen isotopes, which are expected to be used for the fuel of fusion reactors. Around 150 ppm of D mainly as HDO in a molecular form is included in natural water, and it is separated for applications to neutron moderator of heavy water nuclear reactors. Although water distillation is a simple way because of no need of catalyst, the separation factor is small. Therefore its plant scale may become huge if as it is. Several economic and realistic D enrichment methods instead have been developed such as the chemical exchange between NH_3-H_2O, the Girdler-Spevack (G-S) bithermal exchange process between H_2S-H_2O [1] and the combined electrolysis chemical exchange (CECE) process using Pt catalytic exchange between H_2-H_2O, recombiner and electrolysis [2]. The first two exchange methods do not need catalyst. Therefore, it can be applied to large-scale ones. An efficient hydrophobic Pt catalyst has been developed for the last H_2-H_2O one [3].

On the other hand, T will be produced by neutron reaction with ${}_3Li^6$ compound breeders in fusion reactors or initial load for fusion reactors will be prepared in gas-cooled fission reactors. Separation between H_2O and HTO is needed in any cases. In addition, detritiation of coolant and wastewater from water-cooled fission rectors becomes necessary. A large amount of wastewater generated in the Fukushima 1F site is waiting for detritiation or radioactivity decrease.

In the analysis of the present study, we determine T concentration profiles in the three separation systems of water distillation, the G-S bithermal and the CECE, and we estimate each plant scale for the large-scale detritiation process of low-concentration tritiated wastewater. The target decontamination factor of wastewater is 100 in the top-to-bottom T concentration ratio in a similar way to the US estimation [4]. The legally allowed exhaust level of wastewater is 60 Bq/cm^3 in Japan, and the detritiation concentration is set lower than that. The total number of theoretical stages inside a detritiation tower and the T concentration profile are analyzed using the McCabe-Thiele diagram of the water distillation, G-S and CECE processes. The operating power and apparatus scale are compared among the three candidates.

In the latter section, we introduce our experimental results of water distillation assisted with adsorption/exchange, and how the T

---

* Corresponding author.
E-mail address: sfukada@nucl.kyushu-u.ac.jp (S. Fukada).

https://doi.org/10.1016/j.fusengdes.2018.05.051
Received 10 September 2017; Received in revised form 23 April 2018; Accepted 15 May 2018

separation is enhanced by adsorption/desorption/isotopic exchange in a distillation tower packed with structured packing having an ability of surface adsorption/exchange. The enhancement mechanism of the isotope separation is discussed in short.

2. Analysis

2.1. Water distillation

The isotope separation factor between H_2O and HTO in a unit of the stage number *i* for a water distillation tower is defined as follows:

$$\alpha_{H-T}=\frac{x_{HTO,i}y_{H_2O,i}}{y_{HTO,i}x_{H_2O,i}}=\frac{x_{HTO,i}(1-y_{HTO,i})}{y_{HTO,i}(1-x_{HTO,i})}\quad i=1\,\text{to}\,N, \tag{1}$$

where *N* is the total number of theoretical stages and $y_{\mathrm{HTO},i}$ and $y_{\mathrm{H2O},i}$ are the HTO and H_2O molar fractions in vapor, and $x_{\mathrm{HTO},i}$ and $x_{\mathrm{H2O},i}$ are those in condensate. HTO is enriched in the condensed liquid phase in water distillation process, *i.e.*, $\alpha_{\mathrm{H\text{-}T}} > 1$. Then the equilibrium line combining the vapor molar fraction y_i ($=y_{\mathrm{HTO},i}$) and condensate one x_i ($=x_{\mathrm{HTO},i}$) for each stage *i* inside a distillation tower is described as follows:

$$y_i=\frac{x_i}{\alpha_{H-T}-(\alpha_{H-T}-1)x_i}\quad i=1\,\text{to}\,N. \tag{2}$$

When water is supplied from or extracted to an adiabatic tower with a condensate feed flow rate F ($=D+B$) (mol/s), a detritiated distillate flow rate *D* (mol/s) and an enriched bottom flow rate *B* (mol/s) under a steady-state condition except for the bottom heater and top condenser, the stage flow rates in the upper depleted and lower enriched sections are given as *L* and $L+F$, respectively. The T molar fractions leaving a stage *i* for the upper depleted and lower enriched sections are described as follows:

$$y_{i+1}={}^{(R_Dx_i+x_D)}\!\big/_{(R_D+1)}\quad i=0\,\text{to}\,N_F, \tag{3}$$

$$y_{i+1}={}^{(R_Bx_i-x_B)}\!\big/_{(R_B-1)}\quad i=N_F+1\,\text{to}\,N, \tag{4}$$

where N_F is the stage number in the upper section above the feed point, and x_D and x_B are the T molar fractions in the distillate and bottom flows. R_B and R_D are the reflux ratios of the bottom and distillate flows, which are defined as follows:

$$R_D={}^{L}\!\big/_{D}, \tag{5}$$

$$R_B={}^{(L+F)}\!\big/_{B}. \tag{6}$$

The following relation is held between R_D and R_B:

$$R_B={}^{(DR_D+F)}\!\big/_{B}. \tag{7}$$

When $R_D=R_B=\infty$, which corresponds to the total reflux condition, the following relation called the Fenske's equation is obtained [5]:

$$\left({}^{x_i}\!\big/_{1-x_i}\right)=\alpha_{H-T}{}^{i}\left({}^{x_D}\!\big/_{1-x_D}\right)\quad \mathrm{i}=1\,\mathrm{to}\,\mathrm{N}. \tag{8}$$

Tritium molar fractions in the distillation tower are determined using the relations of Eqs. (2)–(4) from the top distillate, x_D, through an arbitral stage position, x_i, to the bottom condensate, x_B, for any given conditions of R_D and R_B. Fig. 1 shows a calculation example for the T molar fraction in the distillation tower to separate T from $x_D = 0.1$ to $x_B = 0.9$ at the conditions of $\alpha_{\mathrm{H\text{-}T}} = 1.05$, $R_D = 50$ and $D/F = 0.5$. The diagram is called the McCabe-Thiele one. The chain line is the equilibrium line of Eq. (2), and the operating line of Eq. (3) or (4) by a solid line. The tie lines combining the HTO molar fractions in the condensate and vapor flows are drawn by broken lines.

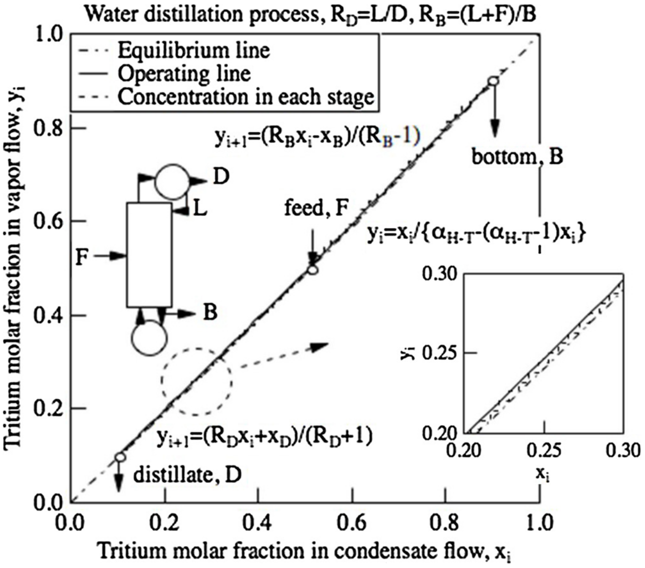

Fig. 1. McCabe-Thiele diagram for water distillation.

One can get a set of analytical solutions for finite values of R_D and R_B, when the T molar fraction is much smaller than unity. The solutions are derived from the original material balance equations. The T concentrations for each stage *i* in the upper depleted and lower enriched sections of the distillation tower are obtained as follows:

$$\frac{x_i}{x_D}=1+(\alpha_{H-T}-1)\sum_{j=0}^{i-1}\left(\frac{\alpha_{H-T}R_D}{R_D+1}\right)^j\quad i=1\,\text{to}\,N_F, \tag{9}$$

$$\frac{x_i}{x_B}=1-(\alpha_{H-T}-1)\sum_{j=1}^{N-i}\left(\frac{R_B-1}{\alpha_{H-T}R_B}\right)^j\quad i=N_F+1\,\text{to}\,N-1. \tag{10}$$

The number in the subscript *i* in x_i corresponds to the stage number. The T concentration ratio of the bottom flow to the distillate one is described as follows:

$$\frac{x_B}{x_D}=\frac{1+(\alpha_{H-T}-1)\sum_{j=0}^{N_F-1}\left(\frac{\alpha_{H-T}R_D}{R_D+1}\right)^j}{1-(\alpha_{H-T}-1)\sum_{j=1}^{N-N_F}\left(\frac{R_B-1}{\alpha_{H-T}R_B}\right)^j}. \tag{11}$$

As seen in Fig. 1 and Eq. (9), the reflux flow should satisfy the minimum condition of $R_D > 1/(\alpha_{H-T}-1)$ ($=R_{D,min}$)for T enrichment. Fig. 2 shows the stage number necessary to achieve the top-to-bottom T concentration ratio of $x_B/x_D = 100$ as a function of the stage separation factor and the reflux ratio under the distillate flow cut of $D/F = 0.9$. The reflux ratio decreases drastically with the increase of the separation factor. The condition of $\alpha_{\mathrm{H\text{-}T}} = 1.05$ can be achieved under the

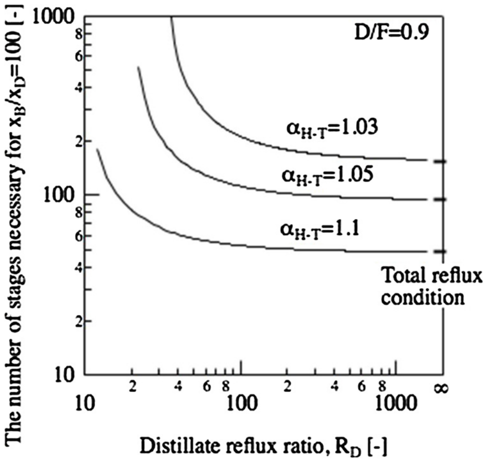

Fig. 2. The number of stages necessary to achieve the total tritium separation of $x_B/x_D = 100$.