

WET SCRUBBER COLUMN FOR AIR DETRITIATION

A. N. PEREVEZENTSEV* and A. C. BELL *EURATOM/UKAEA Fusion Association
Culham Science Centre, Abingdon OX14 3DB, United Kingdom*

B. M. ANDREEV,† M. B. ROZENKEVICH, Yu. S. PAK, and A. V. OVCHAROV
D. Mendeleev University of Chemical Technology of Russia, Miusskaya Sq. 9, 125047, Moscow, Russia

Received September 12, 2008

Accepted for Publication February 23, 2009

This paper evaluates detritiation of air contaminated with tritium in the form of water vapor using a scrubber column filled with structured packing. This technique is based on isotopic exchange between tritiated water vapor and liquid water. In combination with a catalytic oxidizer operated at room or slightly elevated temperature, the scrubber column can also decontaminate air contaminated with tritiated molecular hydrogen. Mass transfer rates measured for structured packings made of stainless steel and copper alloy confirmed high efficiency of the detritiation process. Study of the effect of various operation parameters on column efficiency al-

lows optimization of column operation. It was demonstrated that this technique is competitive with the drying technique with respect to the decontamination factor (DF) provided and the amount of tritiated water to be generated. Benefits offered by the wet scrubber technology are based on the nature of the isotopic exchange process. No need for regeneration allows reduction in the number of units and as such decreases the capital cost of the facility for continuous operation. The DF can be controlled by changing the flow rate of feedwater.

KEYWORDS: air detritiation, tritium facility, packed column

I. INTRODUCTION

Facilities handling large quantities of tritium, for example, tritium plants of fusion reactors, such as Joint European Torus (JET), TFTR, and in the future ITER, employ means to detritiate air and other gases prior to their discharge to the environment.¹⁻³ Two main approaches have been in use for gas detritiation. In the first approach gaseous species are decomposed in a reaction with a chemical reagent, for example, a hot metallic getter, followed by collection of the released molecular hydrogen by adsorption or formation of metal hydrides with metallic hydrogen getters.⁴ Owing to the nature of the process and the formation of solid radioactive waste, this approach is limited to processing mostly inert gases that are not chemically active toward metallic getters and contain a relatively small amount of impurities that can react with the metallic getter.

The second approach is based on the catalytic conversion of all tritium-containing gaseous species into water

followed by its removal from the gas stream. Water then is disposed of as a waste stream or processed for tritium recovery.⁵ Several methods of tritiated water collection have been used, such as condensation, passing through water bubblers, and drying.² The latter method is most widely adopted in tritium-handling facilities because it enables the detritiation of a large flow of gas at a relatively low operational cost and should provide a nearly constant time decontamination factor (DF). Detritiation is based on moisture removal from the gas stream through adsorption or absorption by solid sorbents. However, the detritiation factor observed may not match the dehumidification factor^{6,7} (DHF). For “fresh” driers or driers with a low level of contamination, the DF can be larger than the DHF. For driers collecting highly tritiated water, the DF could be less than the DHF. Two main reasons can be given to explain this effect. First is incomplete regeneration of driers,⁶ which leads to the release of tritiated water retained in driers after regeneration. Second is the presence of hydrogen-containing species strongly adsorbed by the adsorbent. Hydrogen in these species can participate in reversible reactions of isotopic exchange with tritiated water vapor in air and then cause $DF > DHF$, if reactions occur with tritium moving into solid

*E-mail: Alexander.Perevezentsev@iter.org

†We regret to inform our readers that Prof. Boris M. Andreev passed away on January 9, 2008.

matrix, or $DF < DHF$, if reactions occur in opposite direction.

The world's largest facility for air detritiation [Exhaust Detritiation System (EDS)] is in operation at the JET thermonuclear experimental reactor. Its design capacity is 500 m³/h. The facility includes three driers each filled with ~400 kg of molecular sieve. The usual schedule of operation includes one drier in operation, a second in regeneration, and the third regenerated and kept in a standby mode. The next fusion reactor, ITER, would need air detritiation facilities having a capacity up to an order of magnitude larger than the facility at JET (Ref. 8). Driers would contain several tonnes of molecular sieve adsorbent. Such facilities comprise part of the tritium confinement system and therefore are classified at ITER as safety important components (SICs), which shall be available at any time they are required. Because of the nature of the process, driers need to be regenerated, and this requires cycling the operation of isolation valves and air heaters. The frequency of failure to operate for valves and heaters presents a major challenge to meet SIC requirements.^{9,10} For example, regular replacement of isolation valves appeared necessary after several years of EDS operation at the JET tritium plant.¹¹

Employing air detritiation based on driers presents a big challenge with respect to meeting the SIC requirements for systems having a capacity much larger than that of the JET EDS. The requirement to control and a strong limit for allowable tritium release into the environment impose a very restricted margin on the availability of air detritiation systems.

There is another technology for gas detritiation based on the reversible reaction of tritium isotopic exchange between water vapor in air (HTO_V) and liquid water (H_2O_L):



The process is arranged in counterflow mode in a packed column, as illustrated in Fig. 1 for air detritiation. Such columns, called here wet scrubbers, are well established in the chemical industry for the purification of gases.¹² Because of the nature of the process, the packed column can decontaminate only gases contaminated with tritiated water vapor. Other tritium-containing gaseous species shall need to be converted into water beforehand. It is worth mentioning that molecular hydrogen can be effectively converted into water using a hydrophobic catalyst operated at room or slightly elevated temperature.¹³ Remaining sources of tritium, such as tritiated gaseous hydrocarbons, present a very small fraction of contamination for the vast majority of applications at tritium-handling facilities and do not cause a noticeable contribution to tritium release to the environment.

This paper presents results from a study of the performance of structured packing materials for air detritiation using wet scrubber columns.

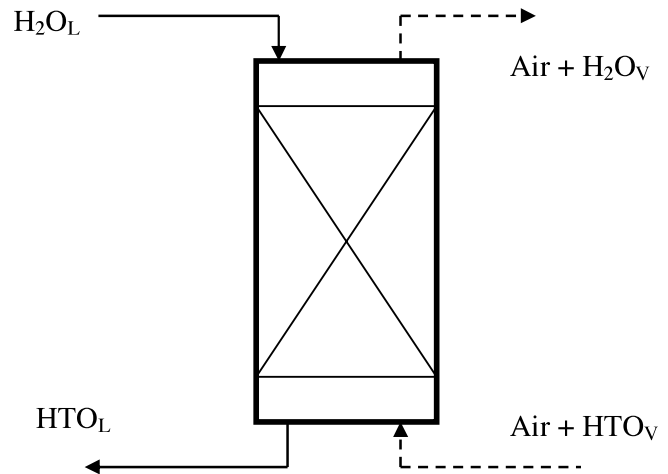


Fig. 1. Arrangement of water and air flows for tritium removal from air in wet scrubber column.

II. EXPERIMENT

II.A. Experimental Installation

The experimental installation is illustrated in Fig. 2. The air, saturated with tritium-free water vapor in the humidifier, was injected under the packed bed of the wet scrubber column. The accuracy of controlling the air flow rate was $\pm 1\%$. Tritiated water was fed onto the top of the wet scrubber packed bed. This arrangement allowed a reduction in the amount of tritiated water used in the facility. Feedwater streams to the humidifier and wet scrubber column were preheated to the required operating temperatures. The accuracy of water flow rate control was $\pm 5\%$. The temperatures of the wet scrubber and humidifier columns were also controlled by water circulation through thermally insulated water jackets. The residual content of the water vapor in the air after the condenser was determined from the temperature of the coolant, controlled by a thermostatic refrigerator in the range of 0 to 2°C. The following parameters were measured: gas temperatures after the humidifier and condenser, humidity of the gas at the inlet of the humidifier and the outlet of the condenser, and pressure drops in the humidifier and wet scrubber column. The isotopic compositions were measured in the water at the inlet and outlet of the wet scrubber column and the condensate from the condenser. The flow rates of the condensate and water at the outlet of the wet scrubber were also measured for tritium mass balance evaluation.

II.B. Procedure of Mass Transfer Parameter Evaluation

The rate of mass transfer in a packed column can be characterized using several parameters adopted in chemical engineering,¹² for example, height equivalent to

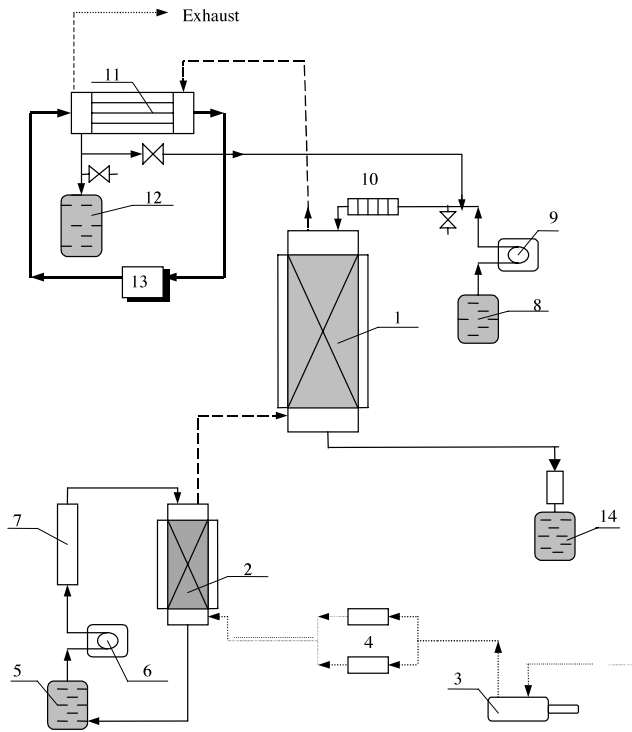


Fig. 2. Block diagram of test installation (1, wet scrubber column; 2, air humidifier; 3, air compressor; 4, flow controllers; 5, 8, 12, and 14, water buffer tanks; 6 and 9, water dosing pumps; 7 and 10, water heaters; 11, water vapor condenser; 13, thermostatically controlled refrigerator for condenser 11).

theoretical plate (*HETP*), height of mass transfer unit (*HTU*), and coefficient of mass transfer (*K*). The two latter parameters can be expressed using concentrations either in gas phase (*HTU_G*, *K_G*) or those in liquid phase (*HTU_L*, *K_L*). The parameters listed above are, however, linked to each other. The number of theoretical plates (*NTP*) (*N*) in the packed bed was determined using Eq. (2) (Refs. 12 and 13):

$$N = [\ln(\Delta Y_B / \Delta Y_T)] / \ln(\alpha / \lambda) = [\ln\{(X_B / \alpha) / [(X_T / \alpha) - Y_T]\}] / \ln(\alpha / \lambda) , \quad (2)$$

where

α = equilibrium separation factor for tritium isotopic exchange between liquid water and water vapor

λ = molar ratio of water vapor flow to flow of liquid water in the packed bed

Y_T, Y_B = tritium concentrations in water vapor in air at, respectively, the top and bottom of the packed bed ($Y_B = 0$ because air was saturated with tritium-free water vapor)

X_T, X_B = tritium concentrations in liquid water at the top and bottom of the packed bed, respectively

ΔY = difference between tritium concentration in vapor and concentration for vapor that is in equilibrium with liquid water at the same cross section of the packed bed

$\Delta Y_T, \Delta Y_B$ = values of ΔY for top and bottom of the packed bed, respectively.

The correctness of the parameters used in Eq. (2) was checked for all tests by evaluating the tritium mass balance

$$G_V \cdot (Y_T - Y_B) = L \cdot (X_T - X_B) , \quad (3)$$

where

G_V = flow rate of water vapor in air

L = flow rate of liquid water in the packed bed.

Necessary corrections were made to take into account the pressure drops in communication lines and the humidifier and wet scrubber column for calculating the water vapor pressure in air. The discrepancy between the amount of tritium that left the water and that gained in the air was within $\pm 7\%$ for all tests.

Values of *HETP*, *HTU* (*HTU_G*), and *K_G* were calculated¹² as

$$HETP = H / N , \quad (4)$$

$$HTU = HETP \cdot (1 - \lambda / \alpha) / \ln(\alpha / \lambda) , \quad (5)$$

and

$$K_G = G_V / (S \cdot HTU) , \quad (6)$$

where *H* and *S* are the height and cross-section area of the packed bed.

III. RESULTS AND DISCUSSION

Commercial CY-type structured packings from SULZER Chemtech, Switzerland, made of stainless steel and copper alloy were tested. This type of packing was selected because of its good mass transfer parameters and high throughput. Structured packings are known to operate at low resistance for gas to flow at a superficial gas velocity up to ~ 2 m/s. Packings of 62-mm diameter were used in the tests. Packing made of stainless steel was additionally acid etched to improve its wetting properties for water.

The tests were carried out with either the return of condensate directed to the top of the wet scrubber column or without it. In the first case liquid water flow along the column was larger than flow of feedwater (*L_{FEED}*). Because no effect of the return of condensate on the

TABLE I
Results of Tests with Packing Made of Stainless Steel*

Test Number	T (K)	W (m/s)	G_V (mol/h)	L (mol/h)	L_{FEED} (mol/h)	λ	$HETP$ (m)	HTU (m)	K_G (mol/m ³ ·s)
1	313	1.10	39.4	69.4	46.7	0.57	0.42	0.31	12.5
2	313	1.10	39.4	62.2	35.0	0.63	0.48	0.40	9.8
3	317	0.55	28.3	51.7	29.4	0.55	0.33	0.24	11.7
4	303	1.10	23.1	74.2	66.7	0.31	1.24	0.70	3.2
5	303	2.0	38.1	86.7	75.0	0.44	1.27	0.78	4.8
6	305	0.55	12.6	62.8	58.3	0.20	1.33	0.71	1.8
7	313	0.55	20.8	43.9	31.1	0.47	0.40	0.28	7.4
8	313	0.55	20.8	78.1	65.6	0.27	0.46	0.25	8.2
9	313	1.90	63.9	92.2	49.4	0.69	0.60	0.48	13.0
10	333	0.55	64.4	97.2	39.4	0.66	0.32	0.26	24.6
11	332	1.10	115.6	136.7	35.0	0.85	0.44	0.39	28.9

* W is superficial air velocity in packed bed at operation temperature; G_V is flow of water vapor in air entering scrubber column; L is water flow rate along the scrubber column; L_{FEED} is feedwater flow rate; $\lambda = G_V/L$.

TABLE II
Results of Tests with Copper Alloy Packing

Test Number	T (K)	W (m/s)	G_V (mol/h)	L (mol/h)	L_{FEED} (mol/h)	λ	$HETP$ (m)	HTU (m)	K_G (mol/m ³ ·s)
1	294	0.55	6.5	7.4	7.4	0.88	0.049	0.048	13.3
2	294	1.10	12.9	12.6	12.6	1.03	0.081	0.078	16.3
3	293	1.10	12.1	13.0	13.0	0.93	0.085	0.078	15.2
4	293	1.10	12.1	18.6	18.6	0.61	0.093	0.070	17.0
5	304	0.55	12.3	14.6	4.2	0.84	0.070	0.061	19.8
6	303	1.10	22.3	25.0	7.6	0.90	0.076	0.069	31.8
7	313	0.55	20.8	23.3	6.9	0.89	0.045	0.041	49.8
8	313	1.80	62.2	82.8	30.6	0.75	0.087	0.073	83.8
9	323	0.55	36.8	35.8	8.3	1.03	0.088	0.080	45.2
10	324	1.10	74.4	82.8	16.9	0.90	0.083	0.076	96.3
11	323	1.10	70.3	77.2	23.6	0.91	0.071	0.066	105
12	323	1.80	108.1	112.8	23.9	0.96	0.116	0.110	96.7

values of $HETP$, HTU , and K_G was observed, the return of condensate is not mentioned in Tables I and II.

The results of the tests with packing made of stainless steel are given in Table I. Figure 3 shows that the value of HTU at a temperature of 303 K is independent of air velocity. At temperatures of 313 and 333 K, similar values of HTU were observed, which rise with an increase of air velocity.

Changing the operating temperature of the wet scrubber column leads to a change of water vapor pressure and subsequently to variation in parameters such as gas viscosity, density, and tritium diffusion coefficients. Therefore, Table I presents apparent values of K_G . In Table III the values K_G evaluated for different temperatures are normalized to the same temperature of 303 K. The partial

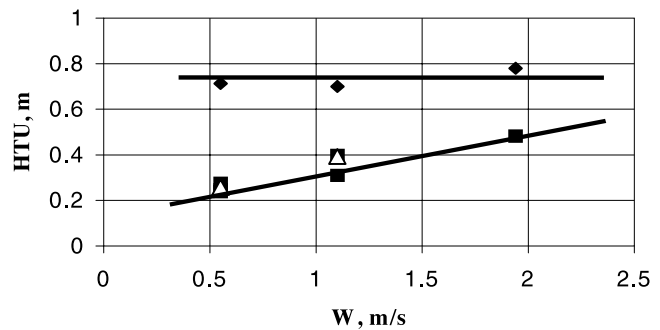


Fig. 3. Dependence of HTU for stainless steel packing on superficial air velocity in packing bed (◆, 303 K; ■, 313 K; △, 333 K).

TABLE III
Normalized Coefficients of Mass Transfer for Stainless Steel Packing

	Test Number										
	1	2	3	4	5	6	7	8	9	10	11
W (m/s)	0.55	0.55	0.55	0.55	0.55	1.1	1.1	1.1	1.1	2.0	1.9
T (K)	305	313	313	317	333	303	313	313	332	303	313
K_G (mol/m ³ ·s)	1.8	8.2	7.4	11.7	24.6	3.2	12.5	9.8	28.9	4.8	13.0
$(K_G)_{NORM}$ (mol/m ³ ·s)	1.7	5.3	4.8	5.5	5.3	3.2	7.3	5.7	6.5	4.8	7.5
	1.7	Average $(K_G)_{NORM} = 5.2$				3.2	Average $(K_G)_{NORM} = 6.5$			4.8	7.5

pressure of water vapor at 303 K has been chosen as a reference pressure for normalizing K_G values. Values of $(K_G)_{NORM}$ were calculated by multiplying the experimental values by a factor of $[P_{H_2O}(303\text{ K})/P_{H_2O}(T)]$. The dependence of the normalized K_G on the air velocity is given in Fig. 4.

Figures 3 and 4 show the dependences of HTU and K_G on the air velocity in the packed bed operated at different temperatures. At a temperature of 303 K, there is a linear rise of K_G with increasing air flow, and this results in a constant value of HTU according to Eq. (6). Values of $(K_G)_{NORM}$ are similar for temperatures of 313 and 333 K. Their rate of increase diminishes as the air velocity rises. These two factors lead to similar values of HTU for temperatures of 313 and 333 K and their increase with air velocity in Fig. 3. A change in the dependence of K_G on the gas velocity at a temperature increase above 303 K might result from the increased rate of mass transfer in the gaseous phase due to the rise in the water vapor pressure and the decreased contribution of this process to the overall mass transfer rate, which becomes controlled by diffusion in liquid.

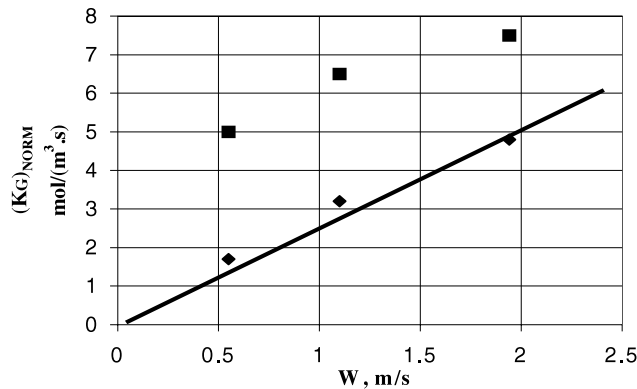


Fig. 4. Dependence of normalized K_G on superficial air velocity for stainless steel packing (◆, 303 K; ■, 313 to 333 K).

The results of tests with a packing of copper alloy are given in Tables II and IV and in Figs. 5 and 6.

Tests 2, 3, and 4 in Table II demonstrate that a change of water load does not affect the rate of mass transfer (increasing parameter λ by a factor of 1.5 at a given gas velocity did not lead to a change in the HTU value). There is no apparent effect of operating temperature on the rate of mass transfer in the packing made of copper alloy.

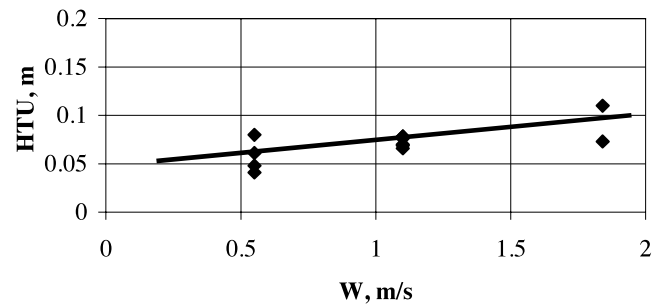


Fig. 5. Dependence of HTU for copper alloy packing on superficial air velocity in packing bed (◆, 293 to 313 K).

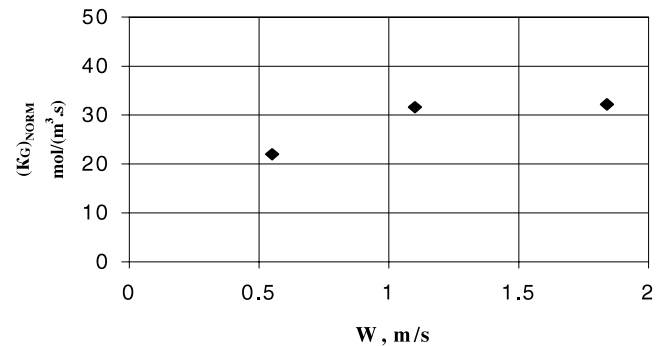


Fig. 6. Dependence of normalized K_G on superficial air velocity for copper alloy packing (◆, 293 to 313 K).

TABLE IV
Normalized Coefficients of Mass Transfer for Copper Alloy Packing

	Test Number											
	1	2	3	4	5	6	7	8	9	10	11	12
W (m/s)	0.55	0.55	0.55	0.55	1.1	1.1	1.1	1.1	1.1	1.1	1.8	1.8
T (K)	293	303	313	323	293	293	293	303	323	323	313	323
K_G (mol/m ³ ·s)	13.3	19.8	49.8	45.2	16.3	15.2	17.0	31.8	105	96.3	83.8	96.7
$(K_G)_{NORM}$ (mol/m ³ ·s)	24.2	19.8	28.6	15.5	30.0	27.7	30.9	31.8	36.1	33.1	31.2	32.2
	Average $(K_G)_{NORM} = 22.0$				Average $(K_G)_{NORM} = 31.6$						Average $(K_G)_{NORM} = 32.2$	

The rate of mass transfer observed for packing made from copper alloy is six to ten times greater than that for stainless steel. A wet scrubber column filled with copper alloy packing can operate effectively at large superficial gas velocity up to 2 m/s. For example, an air detritiation facility of 1000 m³/h throughput would need a column of 0.6-m inner diameter. To achieve a detritiation factor of 1000, such a column should require a packing of ~5-m height. This demonstrates that air detritiation using a wet scrubber column could be arranged in a compact device that can operate without regenerations and can therefore easily meet safety requirements.

The rate of tritiated water production is an important factor for gas detritiation systems. This is because tritiated water that is collected requires further processing or disposal.⁵ The rate of tritiated water production by a wet scrubber column can be the same or slightly smaller than that for a dryer. The wet scrubber column will humidify the air until it reaches saturation at the column operating temperature. The air containing detritiated water vapor can be then discharged. If, however, the discharged air is cooled to allow condensate to be collected, this condensate is not to be returned to the column. With no condensate return to the column, the amount of tritiated water produced is independent of the initial humidity of gas to be detritiated and is determined by factor λ only. Simple mathematical consideration shows that for $\lambda = 1$, the tritiated water product flow rate would be equal to the amount of water vapor in the gas to be detritiated. This is the amount of water that would be collected by driers. A wet scrubber column operated at $\lambda > 1$ will produce less tritiated water than a drier for detritiating the same air. The larger the factor λ , the smaller the rate of tritiated water production would be. Therefore, the factor λ is an operational parameter that is a subject for optimization at the stage of the wet scrubber column design. The value of λ can be chosen as a trade-off between the quantity of tritiated water waste to be produced and the height of the packed bed required. This is illustrated in Fig. 7. The height of the packed bed needed to provide a required air detritiation factor ($DF = Y_B/Y_T$) is determined by the

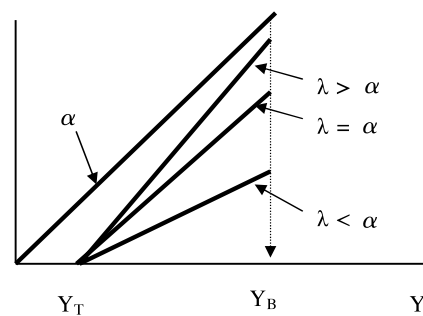


Fig. 7. McCabe-Thiele diagram to illustrate NTP needed to provide the same air detritiation at different factors λ .

number of steps (this is the NTP) that can be drawn between the equilibrium line (α) and operation line (λ) in the McCabe-Thiele diagram in Fig. 7. Figure 7 indicates that an increase of λ will result in the need for a larger NTP and, consequently, a longer packed bed to provide the same air detritiation factor ($DF = Y_B/Y_T$). As a general recommendation and to be competitive with driers in terms of the quantity of tritiated water produced, λ for the wet scrubber column should be selected in the range between 1 and the equilibrium separation factor α for tritium isotopic exchange between liquid water and water vapor.

Figure 7 indicates also that the detritiation factor to be provided by the wet scrubber column can be controlled by changing λ . For example, Fig. 8 illustrates the effect of selecting ratio λ on the design length of the packed bed (expressed in the NTP) in the wet scrubber column, which shall provide a detritiation factor of 1000. The smaller ratio λ is chosen as the design value; the shorter packed bed will be needed. Another side operation of the wet scrubber at a smaller ratio λ will result in an increased rate of tritiated water production. This gives the possibility for a more flexible design of the air detritiation facility based on a wet scrubber column than that based on a drier, the detritiation factor of which is fixed by the humidity of air to be detritiated.

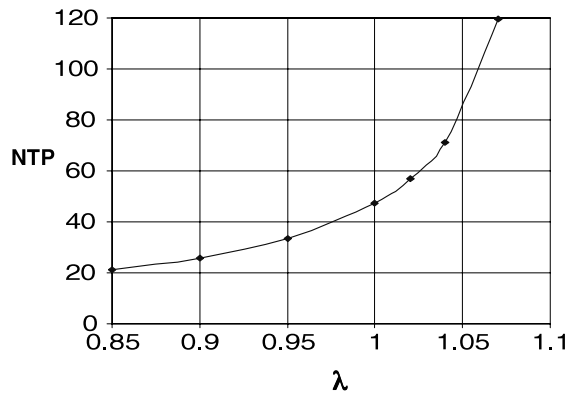


Fig. 8. Effect of ratio λ on length of packed bed needed to provide detritiation factor of 1000 (operation temperature of 293 K).

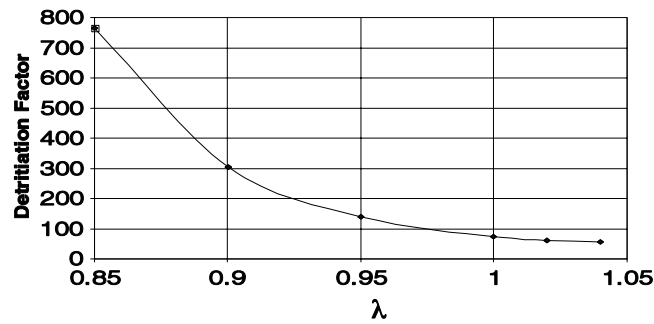


Fig. 9. Effect of ratio λ on detritiation factor that can be provided by wet scrubber column of 20 theoretical plates.

Other features of air detritiation by means of a wet scrubber column are the possibility to change the detritiation factor provided by a packed column of a given height and the simplicity of design for continuous operation. The first is illustrated in Fig. 9, which shows a change of the detritiation factor, which can be provided by the wet scrubber column of a given length, in response to a change of ratio λ . The simplification of design, compared with technology based on water vapor adsorption, is based on there being no need to change the gas pathway, as is required for adsorbent regeneration. The smaller quantity of valves needed for the scrubber column shall increase the facility availability.⁹

IV. CONCLUSION

Results of experimental tests demonstrate that a method of isotopic exchange between liquid water and water vapor can be employed for detritiation of air and other gases contaminated with tritium in the form of water vapor. A facility that is both powerful and flexible in operation can be created using a wet scrubber column filled with a structured packing material. Owing to the

nature of the process, no stop of operation is needed in contrast to the existing method of employing air driers, which require periodic regeneration. The wet scrubber column benefits from producing a lower quantity of tritiated water waste than that from a drier.

ACKNOWLEDGMENT

This paper is dedicated to Prof. Boris M. Andreev. Professor Andreev was one of the highest Russian experts in the field of separation of light element isotopes. For 26 years he headed the Department of Isotope Separation Technology of the Mendeleev University of Chemical Technology, Russia. The department was acknowledged in 1996 as a leading scientific school in the field of light element isotope separation.

REFERENCES

1. R. LAESSER et al., "Overview of the Performance of the JET Active Gas Handling System During and After DTE1," *Fusion Eng. Des.*, **47**, 173 (1999).
2. R. S. WILLMS et al., "Mathematical Comparison of Three Tritium System Effluent HTO Cleanup Systems," *Fusion Sci. Technol.*, **41**, 974 (2002).
3. R. H. HSU, "Confinement and Stripping Systems for APT Tritium Processing," WSRC-RP-97-00887, Savannah River Technology Center (1997).
4. A. NOBILE et al., "Design Optimisation of Metal Getter Reactors for Removing Tritium from Flowing Gas Streams," *Fusion Technol.*, **28**, 1558 (1995).
5. A. N. PEREVEZENTSEV and A. C. BELL, "Development of Water Detritiation for JET," *Fusion Sci. Technol.*, **53**, 816 (2008).
6. P. J. ALLSOP et al., "The Effects of Residual Tritium on Air-Detritiation Drier Performance," *Fusion Technol.*, **21**, 599 (1992).
7. F. SABATIER et al., "Assessment of Performance of the JET Exhaust Detritiation System," *Fusion Eng. Des.*, **54**, 547 (2001).
8. "Atmosphere Detritiation System," ITER document N32 DDD 39 01-08-06 R0.1, ITER International Organization.
9. S. OHIRA and T. HAYASHI, "Review of Ventilation Systems for ITER Nuclear Systems," N32TD17FJ (ITA-32-11), ITER International Organization (2007).
10. D. MURDOCH et al., "ITER Design Review: Tritium Issues," *Fusion Sci. Technol.*, **54**, 3 (2008).
11. P. D. BRENNAN et al., "Maintenance of the JET Active Gas Handling System," *Fusion Eng. Des.*, **69**, 71 (2003).
12. *Chemical Engineering*, Vol. 2, "Particle Technology and Separation Process," J. M. COULSON and J. F. RICHARDSON, Eds., Butterworth Heinemann, Oxford (1991).
13. M. ROZENKEVICH et al., "Development and Improvement of Devices for Hydrogen Generation and Oxidation in Water Detritiation Facility Based on CECE Technology," *Fusion Sci. Technol.*, **48**, 124 (2005).