Fixed Bed Study for a Detritiation Adsorber

K. R. Kim, M. S. Lee, S. Paek, S. P, Yim, D. H. Ahn, H. Chung and M. H. Shim*

Korea Atomic Energy Research Institute, 150 Deokjin-dong, Yuseong-gu, Daejeon, 305-353, KOREA

* University of Science & Technology, 52 Eoeun-dong, Yuseong-gu, Daejeon, 305-333, KOREA

SUMMARY

A method of predicting the tritium concentration in the air leaving an atmospheric detritiation dryer was modeled for designing a fixed bed dryer and preparing an advanced dryer control. In order to quantify the bed utilization and the dynamic capacity against an inlet humidity and a flow rate, a series of quantitative tests based on the break-through behavior were carried out in an isothermal fixed bed of synthetic zeolites such type as molecular sieve 4A, 5A, 13X and mordenite. The amount of water vapor breaking during the adsorption was estimated to give a breakthrough capacity at the various inlet flow rates and humidity conditions. The molecular sieve 13X exhibited a better adsorption performance at a given bed height.

1. INTRODUCTION

The recovery of tritium from a waste gas stream is important with respect to a reduction of a tritium emission to the environment. A fusion research facility and a power plant will require large atmospheric detritiation systems to mitigate the tritium releases and to recover the heavy water vapor. The best available technology for these systems is the oxidized-and-adsorb process, where tritiated species are converted to tritium oxide (HTO) and adsorbed on an atmospheric detritiation dryer.[1,2,3,4]

Conventional atmospheric detritiation dryers use synthetic zeolites as the adsorbent and rely on a thermal–swing cycle. This permits a continuous detritiation of a gas by using multiple desiccant beds, each beds is regenerated following a period of moisture

removal, in order to maintain the gaseous tritium emission below the approved emission limits.[5]

In designing a fixed bed dryer and preparing an advanced dryer control, it is necessary to quantify the bed utilization and dynamic behavior against an inlet humidity and a flow rate. This study is aimed at obtaining an insight into the breakthrough behavior in small column experiments and a dynamic modeling for the operating performance of the atmospheric detritiation dryer.

2. MATHEMATICAL MODEL

An adsorber is considered as an isothermal fixed bed column with a constant void fraction(ε_b) arising from the gaps. The fixed bed is composed of cylindrical pellets of adsorbent with a uniform diameter(d_p). The adsorbent is a porous solid with a constant porosity(ε_p) as shown in Figure 1.

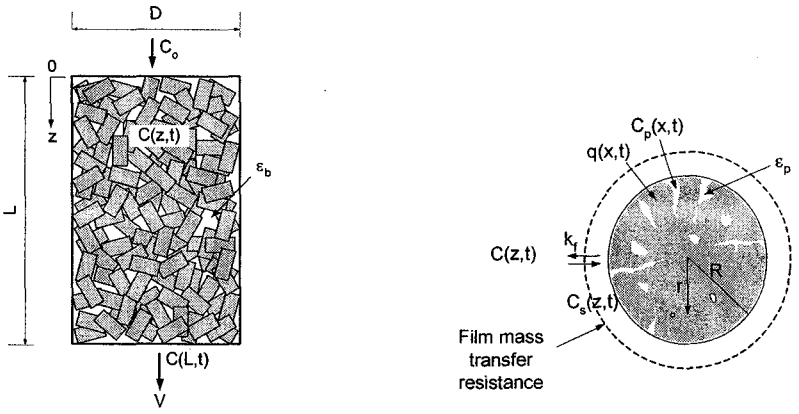


Figure 1. Model of the fixed bed column

The governing equation for the bulk-fluid phase of the adsorption column can be written as follows:

$$\frac{\partial C}{\partial t} = E_m \frac{\partial^2 C}{\partial z^2} - v\frac{\partial C}{\partial z} - \frac{2k_f}{R}\frac{1-\varepsilon_b}{\varepsilon_b}\left(C - \frac{q|_{r=R}}{q_m b - b\,q|_{r=R}}\right) \tag{1}$$

The boundary conditions for the inlet and the outlet of the column, and the initial condition are of the following form.

$$E_m \left.\frac{\partial C}{\partial z}\right|_{Z=0} = v(C - C_f), \quad \left.\frac{\partial C}{\partial z}\right|_{z=L} = 0, \quad C|_{t=0} = 0$$

Accumulation in the solid phase and a radial diffusion in the cylindrical particle are described by the following equation.

$$\frac{\partial q}{\partial t} = D_{ef}\left(\frac{\partial^2 q}{\partial r^2} + \frac{1}{r}\frac{\partial q}{\partial r}\right) \tag{2}$$

The Langmuir type of an adsorption isotherm has been considered in this study:

$$q = \frac{a}{1-\varepsilon_b}\frac{bC}{1+bC} = q_m\frac{bC}{1+bC} \tag{3}$$

The following boundary conditions and initial condition are considered for the particle.

$$\left.\frac{\partial q}{\partial r}\right|_{r=0} = 0, \quad D_{ef}\left.\frac{\partial q}{\partial r}\right|_{r=R} = k_f\left(C - \frac{q|_{r=R}}{q_m b - b\,q|_{r=R}}\right), \quad q|_{t=0} = 0$$

The equations are written in a dimensionless form as reported in literature. [6]

We obtain a system for an ordinary differential equation for both the bulk-fluid and particle phase by applying the method of lines (space discretization of the governing PDEs). The proposed mathematical model is solved by MATLAB software.

3. EXPERIMENTAL

A series of regeneration/adsorption cycles was performed using a bench-scale adsorber. The adsorber was loaded with 1.6mm (nominal) of synthetic zeolites such type as 4A, 5A, 13X molecular sieves and mordenite. Each test consisted of regenerating a 10cm long by 1.1cm diameter bed to a known condition, followed by an adsorption using moisture stream with a constant humidity. The bed was vertical and gas flowed downward during an adsorption and a regeneration.

Constant humidity was used during each adsorption test. CEM (Control-Evaporation-Mixing, Bronkhorst Hi-Tec. Co.) system was applied to supply the carrier gas with a constant humid condition. The defined quantity of the liquid can be mixed with the carrier gas and vaporized in the CEM system as seen in Table 1. Capacitance–type hygrometers from Michell Instruments with Cermet sensors monitored the inlet and outlet of the dryer bed. The outlet humidities expressed as a dew-point temperature were automatically recorded through an interface with a computer.

Table 1. Humidity conditions by using the liquid delivery system with a vapor control.

Water flow (g/hr)	Dry air flow (SLPM)	Absolute humidity (gH_2O)/g dry air)	Relative humidity (%)			Dew point (℃)
			15℃	25℃	35℃	
0.1	1.0	0.0015	13.01	7.25	4.22	-11.02
0.2	1.0	0.0029	25.97	14.46	8.41	-3.06
0.3	1.0	0.0044	38.87	21.64	12.59	2.05
0.4	1.0	0.0058	51.72	28.79	16.75	6.11
0.5	1.0	0.0073	64.51	35.91	20.89	9.35
0.6	1.0	0.0088	77.24	42.99	25.01	12.05
0.7	1.0	0.0102	89.92	50.04	29.11	14.38
0.8	1.0	0.0117	-	57.06	33.19	16.42
0.9	1.0	0.0131	-	64.05	37.26	18.25
1.0	1.0	0.0146	-	71.01	41.3	19.9

4. RESULTS AND DISCUSSION

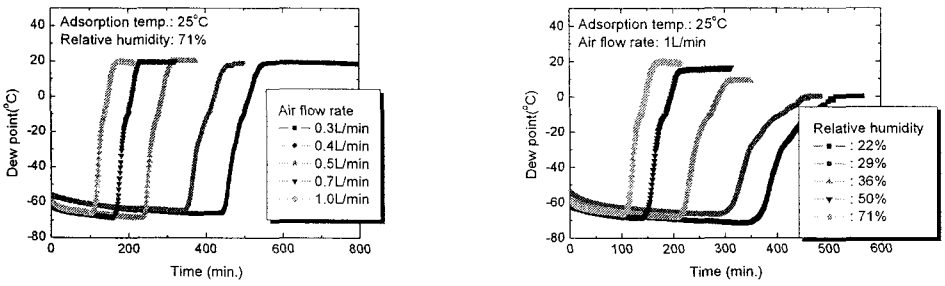
4.1 Numerical Simulation

The model equations are solved by using an explicit method of the finite difference technique with the appropriate initial and boundary conditions. 50 interior grid points for the axial domain in the fluid phase and 5 interior points for the radial domain in the adsorbent phase were selected, respectively, to solve the mathematical model in this study. The built-in function 'ode15s.m' of the MATLAB software was used as the ODE solver to obtain the solutions from a set of ordinary differential equations. The outlet concentration at a different time is determined and the breakthrough patterns are generated. The model parameters of a particle diffusion coefficient (D_{ef}), mass transfer rate (k_f) and axial dispersion coefficient (E_m) could be adjusted to give the best-fit of a curve.

4.2 Adsorption Tests

Figure 2(a) shows that the breakthrough appears earlier at a higher air flow rate containing a constant moisture content. On the other hand, the breakthrough curves are a little steeper at a higher air flow rate. In general, if the flow rate is decreased or the bed length increased, the breakthrough curve becomes steeper. The driving force for an adsorption, as well as a regeneration, is the difference in the water vapor pressures in air and at a desiccant surface. The amounts of water vapor passed during each adsorption step to reach a breakthrough were estimated to be about 0.2kg H_2O/kg adsorbent by time-integrating the outlet water vapor content and multiplying by the air flow-rate.

Figure 2(b) shows that the breakthrough appears earlier and the breakthrough curves are steeper at a higher humid condition. This is to say that the breakthrough zone causing a mass transfer from a fluid to an adsorbent becomes narrower due to a higher driving force.



(a) (b)

Figure 2. Breakthrough curves for a water vapor adsorption on molecular sieve 13X bed at a different (a) air flow rate and (b) inlet humidity.

The breakthrough appears earlier at a higher temperature condition as seen in Figure 3. This tendency is thought to be because the increasing adsorption temperature increases the temperature difference of the air stream through the adsorption column. The average amount of water vapor breaking during an adsorption through the fixed bed was estimated to be about 20 wt.% as a breakthrough capacity at the normal humidity conditions.

Adsorption performance for the tritiated water vapor is determined by the breakthrough point or the dynamics of reaching that point. Figure 4 shows that the breakthrough for the molecular sieve 13X appears later than the others at a constant humidity. Among the synthetic zeolites tested in this study, it is shown that molecular sieve 13X has a higher dynamic capacity.

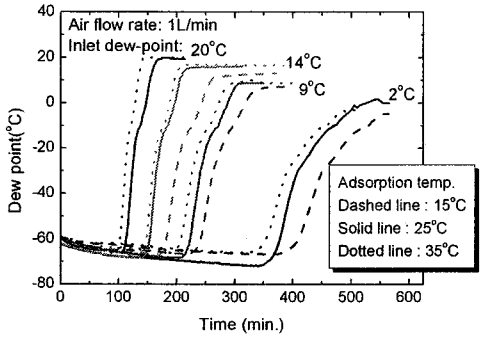


Figure 3. Breakthrough curves for a water vapor adsorption on molecular sieve 13X bed at different humidity and adsorption temperature.

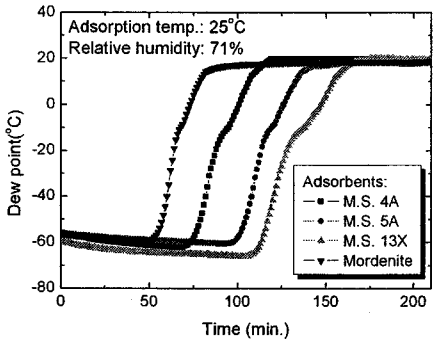


Figure 4. Breakthrough curves for a water vapor adsorption on molecular sieves and mordenite beds at constant humidity.

5. CONCLUSIONS

The mathematical model of an isothermal adsorption in a fixed detritiation bed has been formulated and solved by using a finite difference explicit scheme. A small scale technique was used to study the adsorption characteristics of an atmospheric detritiation dryer. In an isothermal fixed bed adsorption system for a constant inlet humidity and

flow rate of the air stream, the breakthrough patterns were obtained to quantify the adsorption performance of the water vapor on the synthetic zeolites.

ACKNOWLEDGEMENTS

This project has been carried out under the Nuclear R&D Program by MOST.

REFERENCES

1. P. J. Allsop, and C. C. Barfoot, "Long-term Performance of Atmospheric-detrtiation Dryers," Fusion Technology, Vol.28, p.1445 (1995)
2. P. J. Allsop, L. L. Deschenes, B. M. MacDonald, and J. A. Senohrabek, "Predicting Detriation-dryer Performance at Low Activities," Fusion Technology, Vol.28, p.1451 (1995)
3. C. Malara, I. Ricapito, R.A.H. Edwards, and F. Toci, Evaluation and Mitigation of Tritium Memory in Detritiation Dryer, Journal of Nuclear Materials, Vol.273, p.203 (1999)
4. F. Toci, A. Viola, R.A.H. Edwards, T. Mencarelli, and V. Forcina, "Sorbent Materials for Fusion Reactor Tritium Processing," Fusion Engineering and Design, Vol.28, p.373 (1995)
5. Canadian Standard Association, "Guidelines for Calculating Derived Released Limits for Radioactive Materials in Airborne and Liquid Effluents for Normal Operation of Nuclear Facilities," CAN/CSA–N288.1-M87 (1987)
6. D. M. Ruthven, "Principles of Adsorption and Adsorption Process," John Wiley & Sons, Inc, New York, p.208 (1984)