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Column Behavior of Molybdate (MoO_4^{2-}) on the (n,γ) $^{99}Mo/^{99m}Tc$ Generator

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1. Introduction

Among the radioisotopes that have been employed in the field of nuclear medicine, technetium-99m (^{99m}Tc) is considered as the most important radioisotope.

^{99m}Tc is, in general, produced from $^{99m}Tc/^{99}Mo$ generators, which are portable and simple to operate. Recently, Korea Atomic Energy Research Institute (KAERI) has developed a high performance adsorbent for $^{99}Mo/^{99m}Tc$ and $^{188}W/^{188}Re$ generator columns by the sol-gel processing [1]. The adsorbent has its capacity more than 200mg/g for molybdenum, which means the adsorbent can be employed for the (n,γ) $^{99}Mo/^{99m}Tc$ generator.

While developing the adsorbents, it is found that the pore geometry of the adsorbent is an important factor in column operation. When the adsorbent has smaller pore diameter and surface area, the adsorption capacity on the column is less than that expected. Hence, the synthesis procedures are varied to make adsorbent with different pore geometry to investigate the effects on the adsorption behavior on the column.

In this paper, the pore characteristics, the equilibrium adsorption capacity in the batch, adsorption in the column, and the distribution of molybdate along the loaded column will be discussed.

2. Experiments

Detailed synthesis schemes for the adsorbents are described elsewhere [2]. The adsorbent used in this study is in the particle size of 75 – 150μm having 3.4mmol/g of sulfate functional groups.

2.1 Pore Characterization

Average pore diameters and surface areas of the adsorbents (A) and (B) are measured by the nitrogen adsorption at 77 K with ASAP 2010.

2.2 Adsorption Isotherm

To study the equilibrium loading capacities with the adsorbent (B), a series of 25 ml molybdate solution in the concentration ranges of 50 – 5000 mg/L at pH = 10.5 are equilibrated with each 0.25g of the adsorbent by shaking for 3 hours. To measure the concentrations of the solutions before and after the contacts, 7.5μCi of ^{99}Mo is added to the solutions before conducting the experiments. All the solution concentrations are measured by using HPGe γ–ray detector (EG & G Ortec, Model # GEM 10175).

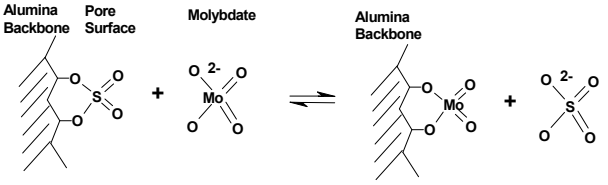
2.3 Column Operation and Radiography

To study the behavior of molybdate, columns in 1.0cm ID are packed with each 1.0 gram of the adsorbent between two layers of inert glass beads. To the columns, each 2.5 ~ 25ml of 10,000 mg/L molybdenum solution at pH=10.5 is fed at 1.0ml/min flow rate. After the loading, the columns are washed with 50ml of saline solutions. By using a TLC scanner (Bioscan, mini-scan B-MS-1000) and a collimator (2.0cm thickness, 0.1mm windows) the activities of adsorbed ^{99}Mo along the columns are measured. The measured data are splined for better presentation.

3. Results and Discussion

The adsorption equilibrium between the adsorbent (B) and molybdates are studied. By the chemical reaction between molybdate and the sulfate-functionalized surface sites shown below, the equilibrium can be described by a Langmuir isotherm:

Chemical Reaction;



Langmuir Isotherm; $$q = \frac{q_{\max} \cdot K \cdot C}{1 + K \cdot C} \qquad (1)$$

where, q is the equilibrium capacity of adsorbent (mg/g), q_{max} is the maximum uptake capacity(mg/g), K is the equilibrium constant(L/mg), and C is the equilibrium concentration of molybdenum(mg/L).

By a non-linear fitting to the experimental data, q_{max} and K are determined as 225.7 ± 7 mg/g and 0.0054 ± 0.0007L/mg, respectively as shown in Figure 1. The equilibrium could be well represented with these constants.

The adsorption capacities in the column are approximately the same or a little less than the capacities in batches from our previous experiments in most cases. However, the adsorption capacities of some adsorbents, which are synthesized at different conditions, are much smaller in the column than others (Figure 2). The adsorbents having low column capacities cannot be

used for the generators. To find the reason of the difference in the capacity, the pore characteristics and the uptake capacities are measured and compared between good and poor quality adsorbents, (A) and (B), respectively. It is obvious from the Table 1 that the adsorbent (B), which has larger pore diameter and volume has higher uptake capacity in the column than that of the adsorbent (A) having smaller pore diameter and volume. By this comparison, it is obvious that the diffusion resistance in the smaller pore causes the poor adsorption capacity in the column.

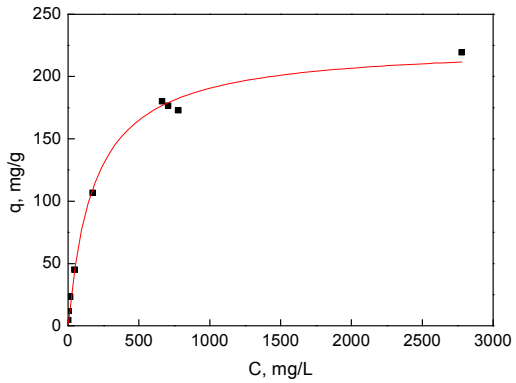


Figure 1. Langmuir Isotherm of Molybdenum Adsorption on Adsorbent (B)

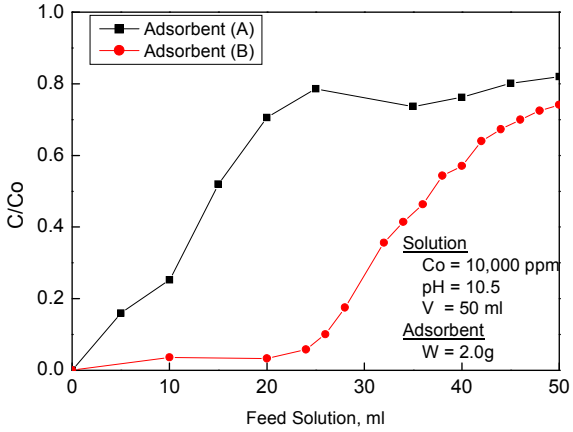


Figure 2. Breakthrough Curves in the Column

Table 1. Pore and Adsorption Properties of Adsorbent (A) and (B)

		Ads. (A)	Ads. (B)
Pore Property	Avg. Diameter(Å)	59.1	107.1
	Surf. Area (m^2/g)	60.4	69.1
	Volume (cm^3/g)	0.09	0.22
Uptake Capacity	Batch (mg/g)	200	217
	Column (mg/g)	101	195

The distribution of adsorbed molybdenum along the column is measured for different loading quantities with adsorbent (B) by the radiography experiments as shown in Figure 3. Molybdenum is only adsorbed at the front of the column when 15% of functional sites on the adsorbent are reacted. In the case of 100% saturation (195mg/g loaded), the molybdenum distributed throughout the column. This means the reaction between the molybdate and the reactive sites are in equilibrium and the diffusion resistance is minimal when the pore diameter is approximately 100Å.

By this study, an important parameter is found for the synthesis of a better adsorbent. The discussion for the relationship between the synthesis parameter and the adsorption properties will be discussed by other publication.

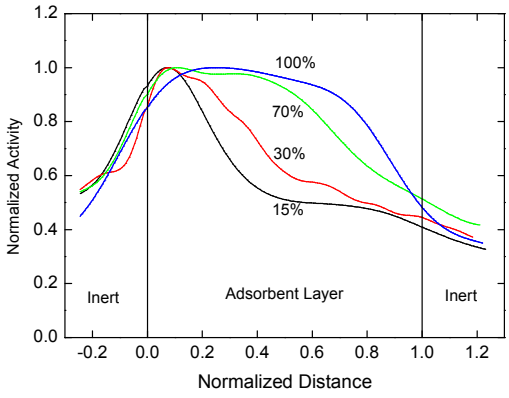


Figure 3. Radiography for the Distribution of Adsorbed Molybdenum on a Column of Adsorbent (A)

4. Conclusion

It is found that the chemical reaction between molybdate and the sulfate functionalized adsorbent is fast enough to be considered as in equilibrium when the pore is large enough. The diffusion of the molybdate through the pores is an important parameter for the column operation because the diffusion resistance becomes greater as the pore becomes smaller. From this study, it is also found that a synthesis parameter, which will be discussed with other publication, is important to control the pore geometry. By controlling the pores, it is possible to synthesize adsorbents with good pore characteristics as well as the high uptake capacity (> 200mg/g, Mo).

REFERENCES

[1] J.S. Lee et al. Sol-Gel Processed Ceramics for ^{99}Mo/^{99m}Tc Generator Materials, 5th International Conference on Isotopes, International Proceedings, Medimond, pp. 21-26, April 25-29, 2005, Brussels, Belgium.

[2] Jun Sig Lee, Jongsup Lee, et al. Absorbent for radioactive element and preparation method thereof, patent pending, 08 Nov. 8, 2006, application No. 10-2006-0109983