Diagnostic and Therapeutic RI Generators

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1. Introduction

Different types of generators have been developed for the convenient use of ^{99m}Tc as the demand for this radioisotope is strong. Currently, the demand for ^{99m}Tc is more than 80 % of the total demand for medical isotopes in the world.

A ^{99m}Tc generator, in general, is composed of a column packed with ceramic adsorbent, tubing, eluent reservoir or vials, collection vials, and shielding. The key technology to develop a good generator is how to load ^{99}Mo as much as possible while maintaining the quality of eluted ^{99m}Tc as good as possible. The technology is well developed and already available commercially for the case of the fission ^{99}Mo/^{99m}Tc because loading of few curries of ^{99}Mo on a conventional adsorbent, i.e. alumina is not a serious task in the chemical point of view. However, the current infrastructure of the supply of ^{99}Mo to the world market is sturdy as the research reactors are getting aged. In this regard, alternative research activities to develop generators with (n,γ)^{99}Mo have been performed by different groups. To develop commercially viable (n,γ)^{99}Mo/^{99m}Tc generator, the generator column should have high adsorption capacity for molybdenum at least several fold higher than the fission ^{99}Mo generator column. To achieve such high adsorption capacity, gel generator, PZC, and other technologies have been developed. However, there are still many restrictions to apply these technologies for the commercial production.

As one of the latter approaches, several candidate adsorbents have been developed [1]. One of the new adsorbents developed at Korea Atomic Energy Research Institute shows a high adsorption capacity for Mo (~200 mg/g) and reasonable elution efficiency for ^{99m}Tc (60 ~ 80%). In addition, (n,γ)^{99}Mo can be loaded by a column operation just like fission Mo generator production.

2. Experiments

2.1 Adsorbent Preparation

The adsorbent is synthesized by the sol-gel processing. For the synthesis, aluminum tri-sec-butoxide 97%, fuming sulfuric acid are purchased from Aldrich chemical Co. and used without further purification. Other necessary chemicals and solvents used in the experiments are ACS grades.

Synthesized materials are grinded and sieved for the average particle sizes of 75 – 150 um.

2.2 Characterizations

To perform characterization experiments, a desired amount of molybdenum oxide (Aldrich Chemical Co.) is dissolved in a 0.4N sodium hydroxide solution and the solution pH is adjusted to 10.5. To this solution, a trace amount of ^{99}Mo solution is added to make 0.3*u*Ci/ml of the specific solution activity just before starting experiments.

Batch loading experiments are performed for 2 hours by shaking 0.5gram of the synthesized adsorbent in 25ml of 10,000mg/L molybdate solution at room temperature. The activities of the solutions before and after contact are measured by using a HPGe γ –ray detector. The activity difference is considered as loaded amount of molybdate on the adsorbent.

To determine elution efficiency for ^{99m}Tc from a molydate-loaded column, a column with 1.0 cm ID is packed with 2 grams of adsorbent. To this column, 50 ml of molybdate solution was introduced at the flow rate of 1.0 ml/min and washed by passing 25 ml of saline solution twice. Elution of ^{99m}Tc performed 23hours after the column loaded and every 23 hours thereafter by using each 20ml of saline solution. The activity of eluted ^{99m}Tc is measured at 140.5KeV by HPGe γ–ray detector.

3. Results and Discussion

The synthesized adsorbent is functionalized by sulfate groups. Introduction of sulfate, which is an inorganic ligand can be achieved by the impregnation in a highly concentrated sulfate solutions or by the immobilization of a preformed functional precursors on the adsorbent. In this research, the later approach is applied. Sulfated functional precursor is synthesized and reacted with an aluminum-tri-alkoxide by the sol-gel scheme.

The sulfate content of the synthesized adsorbent is approximately 3.4mmol/g. The maximum uptake capacity for molybdenum in the batch is approximately 210mg/g at pH=6 ~ 6.5. The uptake capacity in the batch means approximately 65% of the total sulfate are replaced by the molybdate as the reaction scheme is shown in Figure 1.

The effect of solution pH on the equilibrium uptake is studied at 3 different initial molybdenum concentrations (0.2, 1, 5mg/ml) by differing the initial solution pH's. As shown in Figure 2, the equilibrium uptake is not much different by the solution pH, when the initial concentration of molybdenum is low. However, the uptake capacity is greatly affected by the solution pH when the adsorbent becomes saturated by the molybdates.

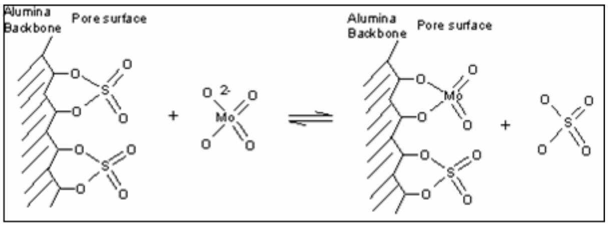

Figure1. Reaction between functional group and molybdate

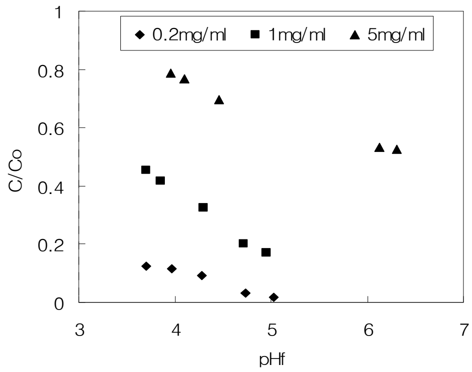

Figure2. Mo uptake capacity exchange at different pH (Each symbol for the different initial concentration, ◆: 0.2 mg/ml; ■: 1.0 mg/ml; ▲: 5 mg/ml)

The uptake capacity of the adsorbent in the column experiment is 195 mg/g, which differs only 7 % from the batch capacity. It is observed that approximately 5% of molybdate in the feed solution is not loaded on the column from the beginning (Figure 3). This phenomenon can be understandable by the ion-exchange reaction between sulfate and molybdate. A high sulfate concentration produced by the exchange reaction between molybdate and sulfate from the upper part of the column may interfere the exchange between the two species at the low part of the column. Further study on this exchange reaction will be performed in the future.

From the column experiment, it is noted that the column was packed with dried adsorbent before passing the feed solution. Other research works, i.e. PZC from Japan, have developed generator materials, which have similar adsorption capacities in batch contacts [2]. However, molybdenum loading on a column is performed a sequent process such as a batch contact to adsorb molybdenum and then by particle loading to a column. However such process is a great challenge if one deals with highly radioactive powders. Hence, it is much valuable to develop adsorbents, which adsorb radioactive molybdenum in a column as much as in a batch.

Elution study has been performed by the molybdenum-loaded column. From the given condition, the elution efficiency for ^{99m}Tc is approximately 75%. To estimate precise elution efficiency, series of elution experiments should be performed with stronger ^{99}Mo activities.

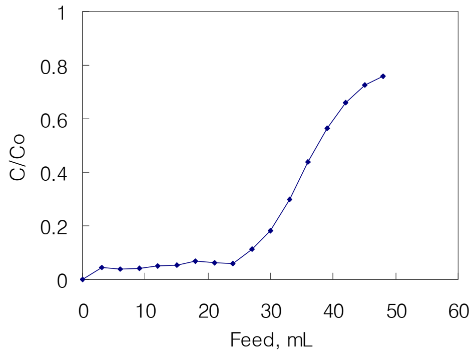

Figure3. Breakthrough curve in column at room temperature (Co: feed concentration, 10,000 mg/L)

3. Conclusion

A sulfated alumina adsorbent is developed as the column material for a (n, γ)^{99}Mo/^{99m}Tc generator. The maximum molybdenum uptake capacities are approximately 210 mg/g and 195 mg/g in the batch and the column contacts, respectively. This new adsorbent system has much advanced characteristics compared to other adsorbents as this adsorbent can be pre-packed in a column to load radioactive molybdenum. Hence, serious handling of powdered radioactive materials can be avoided in the production process of the generator columns.

Also, the high capacity for the molybdenum promises that this adsorbent can be used as the column materials for 188W/188Re generators. Further studies have been conducted and will be published in scientific journals.

REFERENCES

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