Transactions of the Korean Nuclear Society Spring Meeting
Jeju, Korea, May 10-11, 2007

Applicability of Sol-Gel Derived Adsorbent for the Production of (n,f) ^{99}Mo Generator

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1. Introduction

High performance adsorbents as the column material for (n,γ) ^{99}Mo/^{99m}Tc and ^{188}W/^{188}Re generators have been developed at KAERI. These adsorbents should have enough loading capacities to produce such generators with high activities as the loading isotopes are not carrier-free. In this regard, the adsorbents are synthesized by sol-gel processing in which the ligand density can easily be adjusted and maximized. The marginal capacity of such adsorbents should be higher than 200 mg/g for molybdenum. From the previous works [1], the sol-gel processing techniques are adequately applied to meet the criteria.

Further studies are being undertaken to employ the adsorbents for the production of high capacity (n,f) ^{99}Mo/^{99m}Tc. Domestically, a private company has lined up the production facility of the molybdenum generator in the activity range of 300 ~ 1,000 mCi/ea. The column elements are composed of manganese oxide doped on silica and alumina at the backup layer. In this column, the manganese oxide is the main reactive layer to retain (n,f)^{99}Mo.

The sol-gel derived adsorbent, in this study, is employed as the replacement of the manganese oxide doped silica in the column and tested for the loading efficiency of (n,f)^{99}Mo, elution of ^{99m}Tc, labeling property of the eluted ^{99m}Tc. The same loading and elution procedures as the commercial production are applied for the tests and quality controls.

2. Experiments

Detailed synthesis scheme for the adsorbent is described elsewhere [2]. In this study, adsorbent containing 3.4mmol/g sulfur is used.

A test column is prepared in a glass column that used for generator production at Sam Young Unitech Co. The sol-gel derived adsorbent (0.2g) is packed as shown in Figure 1. The same loading process as the regular production for the generators at Sam Young Unitech Co. is applied. The loading solution contains 2.0Ci of ^{99}Mo in 4 ml of nitrate solution at pH = 5 ~ 6. The loaded column is washed twice with each 20ml of saline solution. The column activity is equivalent to 500mCi with 5 reference dates.

Elution tests are performed every 24 hours from one day after the column loading. Elution of ^{99m}Tc is performed with solution vials containing 5~10ml of saline solutions and vacuum vials for the extraction.

Activities of ^{99m}Tc in eluted solutions are measured by a dose calibrator (Capintec, CRC-127R). Radionuclidic impurities in eluted solutions are measured by using a HPGe γ-ray detector (EG & G Ortec, Model # GEM 10175). Two samples of the eluted ^{99m}Tc solutions are labeled with Mebrofenine cold kit produced at KAERI. For this test, 10 ~ 20mCi ^{99m}Tc in 1.5 ~ 2.0ml of saline are injected to the cold vials, shaken well, and stored for 5minutes before the measurement of the loading efficiency. For the measurement, an ITLC scanner (Berthold, Trance master 20) is used. MEK is used as the developing agent on the Whatman #1 paper.

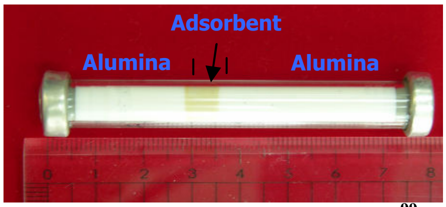

Figure 1. Column Prepared for (n,f)^{99}Mo/^{99m}Tc Generator; weights of adsorbent: 0.2g; alumina: 0.54g (shorter) and 1.56g (longer in the figure)

3. Results and Discussions

The sol-gel derived adsorbent is sulfate functionalized alumina containing approximately 3.4mmol of sulfur per gram of the adsorbent. The maximum adsorption capacity for the molybdenum in molybdate form (MoO_4^{2-}) is ~ 220mg/g.

The quantity of adsorbent used for the preparation of the column is 0.2g, which can adsorb maximum 44mg of molybdenum, which is equivalent 479Ci of ^{99}Mo. However the loading solution contains very little amount of molybdenum in the chemical point of view, the adsorbent can adsorb less quantity by the chemical equilibrium. To analyze the equilibrium adsorption capacity, an adsorption study was performed and found that the chemical equilibrium of the adsorption process could be described by a Langmuir isotherm:

$$q = \frac{1.22 \cdot C}{1 + 5.4 \times 10^{-3} \cdot C} , \quad (1)$$

where, q : adsorbed amount of molybdenum (mg/g)
C : concentration in loading solution (mg/L)

By using this equation, the expected loading amount in equilibrium with 2.0Ci ^{99}Mo is 0.253mg, which is equivalent to 121Ci on the column. Hence, it is possible to produce a generator with extremely high activity by using the developed adsorbent.

The results of ^{99m}Tc elution are summarized in Table 1. The elution efficiency on the next day of loading is approximately 10% with 5 ml of saline solution. On the following Monday, three consecutive elution are performed by each 5 ml. The elution efficiency is the highest in the second portion of the elution. Hence, the elution on Friday was not performed with enough amount of saline. After then, the elution efficiency is relatively high. The low elution efficiency at the beginning is possibly due to the un-reacted sites after ^{99}Mo loading on the adsorbent. These sites may interact with technetium during the elution. Hence, it would be valuable to study the interaction between the adsorbent and technetium. Also, less strong adsorbent can be synthesized for the (n,f) ^{99}Mo/^{99m}Tc generator.

^{99}Mo quantity in the eluted solution is an important criterion in determining the quality of a ^{99}Mo/^{99m}Tc generator. Allowable quantity of ^{99}Mo is less than 150ppm, and commercial generators normally show the quantity less than 50ppm. In this study, the amount of ^{99}Mo is higher than the standard at the beginning and becomes less after then. This higher leaching of ^{99}Mo is possibly due to the impurity on the adsorbent, which is used without washing treatment.

To assure the quality of eluted ^{99m}Tc, an alumina content test is performed for the solution on the first day. As shown in Figure 2, the eluted solution contains alumina much less than the 10 ppm standard solution by comparing the color and the size of the spots.

Table 1. Results of ^{99m}Tc from the Loaded Column (Lot. No.: SY0807-60)

Date	^{99}Mo Generator, mCi	Elution Efficiency % (ml)	Mo/Tc Ratio, ppm	Remarks
Thurs	2000			Loading
Fri	1554	10.6 (5)	877	Alum. Test
Mon	729	17.5 (5) 37.9 (5) 6.9 (5)	249 121	Labeling Labeling
Tue	567	65.2 (5)	58.8	
Wed	440	94.8 (10)	57.0	
Thurs	342	101 (10)	49.4	Ref. Day
Fri	266	65.4 (15)	90.8	10+5.5ml
Sat	206	96.7 (5)	17.4	
Mon	124	90.6 (5)	15.8	
Wed	75	88.7 (5)	14.4	

Mo-99 Loading: 4.0 mL x 500 mCi/mL = 2000 mCi
Elution: everyday 9:00 AM

Labeling test with Mebrofenine cold kit is performed to see the quality of eluted ^{99m}Tc. Mebrofenine is successfully labeled with the eluted ^{99m}Tc solution as shown in Figure 3.

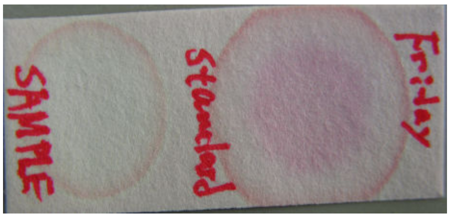

Figure 2. Results of Alumina Content Test: Alumina Standard Contains 10 ppm (Dark Color)

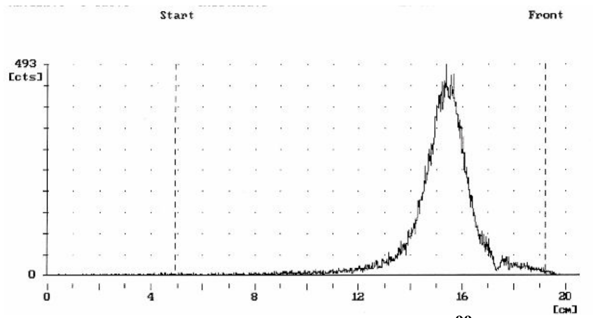

Figure 3. Results of ITLC Scan for the ^{99m}Tc-labeled Mebrofenine

4. Conclusions

A high performance adsorbent derived by the sol-gel processing is tested to use it as the (n,f)^{99}Mo/^{99m}Tc generator column. Various tests such as capacity, alumina content, and labeling efficiency show that the adsorbent has reasonable ability as the column material. The amount of ^{99}Mo in the eluted solution is not satisfactory at the beginning due to the use of unwashed adsorbent. The elution efficiency is also unsatisfactory at the beginning, which is possibly due to the remaining of large amount of non-reacted sites. In the future, the interaction between the adsorbent and ^{99m}Tc will be studied. Putting aside this future study, problem can be solved technically, either by reducing the capacity of the adsorbent or by altering the column washing process to remove the non-reacted sites. By resolving this problem, the development of a high activity generator, even higher than 10Ci, can be achievable.

REFERENCES

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