

A $^{109}\text{Cd}/^{109\text{m}}\text{Ag}$ GENERATOR

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Sorption of Cd and Ag by a cation exchange resin has been studied at different molarities of nitric acid. The sorption capacity of Cd on a cation exchanger has been determined. A $^{109}\text{Cd}/^{109\text{m}}\text{Ag}$ generator is suggested, based on the sorption of Cd on AG 50W-X8 organic cation exchanger at 0.01M HNO_3 . $^{109\text{m}}\text{Ag}$ is eluted with 0.2M NaCl, physiologically compatible for human use.

INTRODUCTION

Ultrashort-lived radionuclides offer an advantage over those with longer half-lives for visualizing veins, chambers of the heart and certain organs with gamma-cameras and scanners. Because venograms and angiograms are usually completed within a minute after the injection of the

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radionuclide, the use of ultrashort-lived radionuclides is possible and has the advantage that repeat studies can be performed quickly because the radioactivity from a previous injection would decay away rapidly. The short half-life of the nuclide would further reduce the radiation exposure to the patient. Radionuclides with ultrashort half-lives could be used either with the bolus injection technique or with the continuous infusion technique.

Radionuclides with such short half-lives are most easily obtained by decay from a long-lived parent radionuclide, the daughter being separated from the parent by elution from a generator¹. Several routes for the production of ^{109}Cd have been reported in the literature, namely proton, neutron and α bombardment of Ag ^{2,3}, as well as proton bombardment of In ⁴. In a nuclear reactor ^{109}Cd is produced via the $^{108}\text{Cd}(n,\gamma)^{109}\text{Cd}$ reaction or $^{107}\text{Ag}(n,\gamma)^{108}\text{Ag} \xrightarrow[2.4 \text{ m}]{\beta^- 97\%} ^{108}\text{Cd}(n,\gamma)^{109}\text{Cd}$ ⁵. We have developed a preliminary method for obtaining $^{109\text{m}}\text{Ag}$ ($T = 39.6 \text{ s}$) from long-lived parent ^{109}Cd ($T = 453 \text{ d}$) radionuclide⁶.

EXPERIMENTAL

Reagents and instruments

Cadmium oxide and silver nitrate (E. Merck) of purity over 99.9% were used as target materials. AG 50W-X8 (50-100 mesh H^+ form) was obtained from Bio-Rad and the other reagents were of analytical grade. The radioactivity was measured by γ -ray spectrometry using a coaxial Ge-detector coupled to a Canberra Series 85 multichannel analyzer.

Irradiation

Known quantities of natural CdO targets were sealed in quartz ampoules and cold welded in aluminium cans. The irradiations were carried out inside the core of the 9 MW swimming pool type PARR-I reactor for a total of 30 d at a neutron flux of $\sim 2 \times 10^{14} \text{ cm}^{-2} \text{ s}^{-1}$. Short irradiations of a few hours were carried for the production of $^{110\text{m}}\text{Ag}$ by irradiating AgNO_3 in a similar manner as described above.

Sorption of Cd and Ag

Irradiated CdO was dissolved in 1M HNO_3 , evaporated to dryness and finally a solution was prepared in distilled water. 5 ml of the aqueous solution of known molarity of HNO_3 containing 1 mg Cd ml^{-1} was shaken at room temperature with 100 mg of resin for 60 min. The supernatant was separated after centrifugation and 1 ml was counted for ^{115}Cd (492 keV 10.2% and 527 keV 33%). Neutron irradiated AgNO_3 was dissolved in distilled water. The above mentioned procedure was adopted for measurement of the distribution coefficient of Ag, except that the concentration of Ag was kept at 0.06 mg ml^{-1} . $^{110\text{m}}\text{Ag}$ activity was determined by using peaks of 657 keV (95%), 764 keV (22%) and 885 keV (73%).

The distribution coefficient was calculated by the formula.

$$K_d = \frac{a}{a_0 - a} \cdot \frac{v}{m}$$

a = activity sorbed on the ion-exchange resin,

a_0 = initial activity in solution,

v = volume of the aqueous phase in ml,

m = mass of the ion exchange resin in g.

Ion-exchange capacity of the resin

About 3 g of AG 50W-X8 (50-100 mesh H^+ form) cation exchange resin was placed into a glass column of 1 cm diameter and conditioned with 5 bed volumes of the 0.001M HNO_3 solution. The solution containing 5 mg ml^{-1} of Cd was passed through the column and 1 ml portions of eluate were collected for the quantitative determination of cadmium breakthrough.

Preparation of $^{109}\text{Cd}/^{109\text{m}}\text{Ag}$ generator

A glass filter column (1 cm ϕ x 15 cm) was filled with 3 g AG 50W-X8 (50-100 mesh H^+ form) and washed with 0.01M HNO_3 . $^{109}\text{CdNO}_3$ (485 MBq g^{-1} Cd) was acidified to a molarity of 0.01M HNO_3 and fed onto the column. The column was washed with 10 ml of 0.01M HNO_3 . A total of 160 mg Cd was loaded onto the column. The generator of $^{109\text{m}}\text{Ag}$ was prepared after 20 d of the target irradiation. During this period nearly all of the ^{115}Cd activity had decayed.

RESULTS AND DISCUSSION

The sorption of Cd and Ag from 0.001M - 10M HNO_3 solutions on a cation-exchange resin is shown in Fig. 1. The adsorbability of Cd(II) decreases with increasing molarity of HNO_3 . At high concentration of HNO_3 between 5-10M, the distribution coefficient is 1. Maximum K_d value was measured at the lowest molarity (0.001M) of HNO_3 used. Sorption behavior of Ag(I) is quite similar as observed in the case of Cd(II). The present results essentially confirm earlier measurements⁷ carried out in 0.1M - 4M HNO_3 solutions.

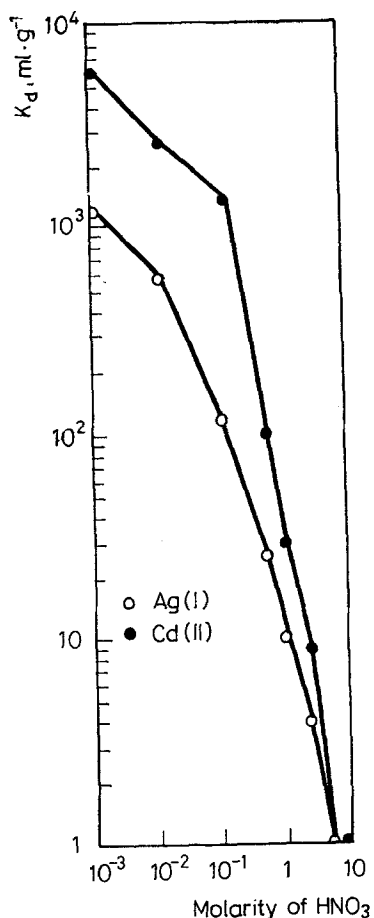


Fig. 1. Sorption of Cd(II) and Ag(I) on AG 50W-X8 (50-100 mesh) cation exchanger

The exchange capacity of the cation exchanger as determined in the present work for Cd at 0.001M HNO_3 was 230 mg Cd g^{-1} of the resin. Strong sorption of Cd on the cation-exchange resin was observed when it was loaded in 0.001M HNO_3 solution. However, the elution yield of $^{109\text{m}}\text{Ag}$ was very poor, hence a 0.01M HNO_3 solution was used for the loading of Cd on the column, which gave reasonably good separation of $^{109\text{m}}\text{Ag}$ from ^{109}Cd . The

elution of $^{109\text{m}}\text{Ag}$ was tested with distilled water, dilute HCl and HNO_3 solutions. Desorption of $^{109\text{m}}\text{Ag}$ was not possible with the above mentioned solutions, however, NaCl solutions were successfully employed for the elution of $^{109\text{m}}\text{Ag}$, leaving ^{109}Cd on the column. The solubility of AgCl in solution containing chlorine ions has been the subject of several investigations^{8,9}. As the $^{109\text{m}}\text{Ag}$ grows, it is dissolved in NaCl and thus eluted. $^{109\text{m}}\text{Ag}$ comes off carrier-free as AgCl or AgCl_2^- in a saline solution physiologically compatible for human use. A reasonably good yield of $^{109\text{m}}\text{Ag}$ and minimum leakage of ^{109}Cd was observed by using 0.1M NaCl. At an elution rate of 4 ml min^{-1} , the yield of $^{109\text{m}}\text{Ag}$ varied from 40-70%. The leakage of ^{109}Cd was checked after 30 min of elution, when all of the eluted $^{109\text{m}}\text{Ag}$ activity had decayed and the parent-daughter equilibrium of $^{109}\text{Cd}/^{109\text{m}}\text{Ag}$ was attained. Leakage of Cd was also confirmed by measuring the 933 keV (1.3%) γ -energy of $^{115\text{m}}\text{Cd}$ ($T = 44.6 \text{ d}$). Long durations of counting were practiced for the determination of Cd contamination by measuring $^{109\text{m}}\text{Ag}$ or $^{115\text{m}}\text{Cd}$. More than 3 liters of 0.1M NaCl was passed through the column for the desorption of $^{109\text{m}}\text{Ag}$ in a period of 2 months. The amount of Cd in the eluate increased after passing 500 ml of eluent. To keep the leakage of Cd up to a level of $10^{-4} \text{ \%}/4 \text{ ml}$, a scavenger column of 300 mg AG 50W-X8 (50-100 mesh H^+) was attached in tandem with the generator. The scavenger column was replaced by a new one after the passage of 200 ml of eluent. The performance of the generator described would be more useful, if no-carrier-added ^{109}Cd parent were used, and this would also eliminate any risk of cadmium metal poisoning.

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