PRODUCTION OF CARRIER-FREE GERMANIUM - 68 BY ALPHA-PARTICLE BOMBARDMENT ZINC CYCLOTRON TARGET

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1 INTRODUCTION

Production of ^{68}Ge is required for making a ^{68}Ga radionuclide generator for positron emission tomography. Radionuclide ^{68}Ge ($T_{1/2}$=288 days) decays to ^{68}Ga ($T_{1/2}$ = 68.3 min) by electron capture, and ^{68}Ga emits positrons with a branching ratio of 90% and weak γ - rays. The Germanium-68 can be produced with both ^{66}Zn (α, 2n) ^{68}Ge and ^{69}Ga (p, 2n) ^{68}Ge nuclear reactions. The solvent extraction technique using a carbon tetrachloride is known to be one of the most widely used methods for carrier-free ^{68}Ge preparation.

In the present report, a separation method of carrier-free ^{68}Ge from the zinc target by extraction chromatography using a mixture of carbon tetrachloride and xylene (1 : 4) as stationary phase on a column of polytetrafluoroethylene has been developed.

2 EXPERIMENTAL

Carbon tetrachloride, benzene, xylene were used as extractants. All chemicals used were of analytical reagent grade.

The radioactive tracer technique was used both for the determination of the distribution coefficients and to monitor of the separation processes. No-carrier-added ^{68}Ge was used for determination of the distribution coefficients for germanium-68. Radioactivity measurements were accomplished with a NP424 L type four-channel nuclear analyzer using a NaI(Tl) scintillator. To monitor the chemical yield of ^{68}Ge as separated from zinc target solution, the activities at each step of separation were measured using a Ge(Li) detector with 53 cm^3 volume and 5.0 KeV resolution at 1332 KeV line of ^{60}Co coupled to a ICA-70 analyzer. The ^{68}Ge activity was determined after 24 hrs of the end of equilibration by counting the ^{68}Ga activity in equilibrium with ^{68}Ge.

Equal volumes (1 ml each) of the aqueous and organic phases were shaken for 15 min in 10 ml stoppered test tubes. Preliminary experiments showed that the equilibrations were complete within 15 min. After extraction the aliquots of both phases (0.5 ml each) were pipetted to determine the distribution coefficient (the ratio of activities of germanium-68 in organic phase to that in aqueous phase, D_{Ge}. To prevent of losses of ^{68}Ge due to its volatilization the aqueous phase was cooled as small as -10°C before addition of carrier free ^{68}Ge. Aliquot was also kept in bath with a mixture of snow and sodium chloride before start of measurements of their activities.

The mixture of carbon tetrachloride and xylene with the volume ratio (1 : 4) was used as the stationary phase for preparation of chromatographic columns. The sorbent was prepared in the following way: a three grams of polytetrafluoroethylene powder of 150 - 200 mesh was moistened with 9 ml of ethanol to form a homogeneous mass. Then 1.5 ml of a mixture of CCl_4 and xylene (1:4) was poured onto this mass and the obtained mixture carefully mixed. Afterwards the excess of 1 M HCl solution was added to mixture under stirring to remove ethanol. The suspension was washed several times with 1 M HCl until the particles obtained was well settled to the bottom of glass. After the aqueous phase was decanted and the suspension obtained was transferred into the chromatographic column filled with 1 M HCl and packed by sedimentation method. Thus a chromatographic column filled with a 3 g of polytetrafluoroethylene with 1.5 ml a mixture of CCl_4 and xylene (1 : 4) was used (sorbent layer height H - 15cm; diameter ∅ - 0.8 cm; free volume V_0 - 3 ml).

Cyclotron target is represented a copper backing plate with the bent edges. For the best cooling by flowing water the plate had ribs on the back side. The first the nickel sublayer up to a thickness of 10 μm was electrodeposited from a sulphate bath on the front surface of copper plate. Then the zinc layer was deposited from a sulphate bath onto copper/nickel backing plate up to a thickness of 300 μm. The weight of electrodeposited of zinc amounted to 5.5 g. The area of zinc layer was 8×3.2 cm^2. The zinc target was bombarded by α-particles with a energy of 36 MeV for 96 - 110 hrs at U - 150 cyclotron. Thus six targets were prepared and bombarded.

3 RESULTS AND DISCUSSION

As a rule, determination of the best conditions for separation of carrier free radionuclide is depended on the properties of the medium, which a cyclotron target is dissolved. Taking this into consideration extraction of

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Table 1. Distribution coefficients of ^{68}Ge.

Composition of aqueous phase	Extractant		
	Benzene	Xylene	CCl_4
8 M HCl + 0.5 M HNO_3	25,2 ± 2.0	21,1 ± 1.45	18,3 ± 1.4
8 M HCl + 1.0 M HNO_3	34,1 ± 2.4	37,1 ± 3.0	35,2 ± 2.8
8 M HCl + 2.0 M HNO_3	69,4 ± 5.2	86,3 ± 6.9	39,3 ± 3.1
8 M HCl + 3.0 M HNO_3	80,3 ± 6.4	85,7 ± 6.2	39,4 ± 2.7
8 M HCl + 4.0 M HNO_3	82,3 ± 5.6	87.4 ± 6.5	39,4 ± 3.2

carrier-free ^{68}Ge was studied from mixed of HCl and HNO_3 solutions by benzene, xylene, carbon tetrachloride. The results obtained are presented in Table 1. In the all systems the distribution coefficients of ^{68}Ge rise with increasing total hydrogen concentration in mixture of acids to the maximum, which occur at hydrogen concentration of 10 to 12 M. Thus, it was found that the mixture of 8 M HCl and 2 M HNO_3 is the optimal mobile phase for separation of carrier-free ^{68}Ge. Therefore a mixture of carbon tetrachloride and xylene with the volume ratio (1:4) is selected as a stationary phase for further work. The radiochemical separation of ^{68}Ge is carried in a lead-shielded cell a month after the end of irradiation, when relatively short-lived radionuclides (^{69}Ge, ^{57}Ni, ^{66}Ga, ^{67}Ga) have decayed. The production yield of ^{68}Ge measured after irradiation of zinc target with a 36 MeV α-particles current of a 50μA is amounted to 0.87 μCi/μA·h.

The radioactive layer of cyclotron zinc target is dissolved in 25 ml concentrated nitric acid and then the target solution is filtered through a fine filter paper. After the filter paper is washed with 5 ml of 14 M nitric acid. The solution obtained is represented nitric acid solution of 6 to 8 M, containing radionuclides of ^{68}Ge, ^{65}Zn, ^{57}Co and macro amounts of zinc, copper, nickel. Afterwards a portion of target solution (2 ml each) cooled as small as 4°C is diluted by a 4 ml of preliminarily cooled concentrated HCl acid as small as - 21°C. Then the solution obtained is passed through the column (prepared as describe above) at flow rate of 0.5 ml·min^{-1}. After loading of all target solution the flask is washed twice with 5 ml portions of cooled 10 M HCl, which is also passed through column. Then the column is washed with 10 ml of 8 M HCl. Carrier free ^{68}Ge is eluted with 4 ml of 0.1 M HCl. The eluate is collected by portions (0.5 ml each) into test tubes. The 98 % of ^{68}Ge is eluted in volume equal to a half of the column's free volume. The ^{68}Ge obtained was very pure.

4 CONCLUSION

Thus, we have developed the method for the production and separation of carrier free^{68}Ge from zinc cyclotron target. The measurement of material balance shows that the overall yield of ^{68}Ge is amounted to 86 %. The main losses are connected with adsorption of ^{68}Ge on glass surface of the applied equipment. The losses due to volatilization are estimated to be.2-3 %.