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Production of As Isotopes in the Interaction of ^{nat}Ge with 14-30 MeV Protons

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Abstract**—**Cross sections of As radionuclides in the interaction of ^{nat}Ge with 14-30 MeV protons have been deduced by off-line γ-ray spectroscopy to find optimal reaction channels leading to radiotracers for positron emission tomography. The experimental results were compared with the previous results and those estimated by the compound nucleus reaction model.

Keywords**—**compound nucleus reaction model, off-line γ-ray spectroscopy, radionuclide.

I. INTRODUCTION

THE positron-emitting radionuclide such as ^{18}F has been used as imaging biomarkers in positron emission tomography (PET) [1]. Diverse radionuclides need be studied for PET. Since arsenic isotopes have various half-lives, some of them would be of use for this purpose. The half-life of ^{73}As is as long as 80.3 days, while that of ^{68}As is as short as 2.53 minutes [2]. The half-lives of ^{69}As, ^{70}As, ^{71}As, ^{72}As and ^{74}As are 15.2 minutes, 52.6 minutes, 65.28 hours, 26.0 hours and 17.77 days, respectively.

In order to produce the arsenic positron emitters, natural Ge foils could be bombarded by protons. However, since natural Ge consists of ^{70}Ge, ^{72}Ge, ^{73}Ge, ^{74}Ge and ^{76}Ge [3], various reactions occur together.

The cross sections for production of the radionuclides of interest are compared with those estimated by the compound nucleus reaction model [4]. In the model the total reaction cross section has been estimated using the cross section for forming the compound nucleus, its survival probability and its probability of decaying to the products of interest. In this work effective optimal conditions to produce radionuclides for PET have been examined in terms of projectile energy.

II. EXPERIMENTAL

Natural germanium foils of thickness of 133.1 μg/cm^2 were bombarded by 30 MeV protons. The protons were provided by the MC-50 cyclotron, Korea Cancer Center Hospital. The mean energies at the center of the target foils were reduced to 14 and 26 MeV, owing to energy degradation [5] in aluminum foils of 258 and 884 mg/cm^2 thickness, respectively, that were located upstream before the Ge target foils. Typical beam intensity was about 300 nA.

Following bombardment, the foils were assayed with a calibrated intrinsic γ–ray spectrometer. More than 20 spectra at each of the bombarding energies were stored and later analyzed with the code SAMPO [6]. Decay curves were analyzed with the CLSQ code [7]. Radionuclides were confirmed by using their γ-ray energies and half-lives in [8]. The results were compared with those estimated by the compound nucleus reaction model and the previous data from [9].

III. RESULTS AND DISCUSSION

Typical γ-ray spectra stored ~35 minutes and ~1 day after the bombardment are displayed in Fig. 1 and Fig. 2, respectively. In Fig. 1(a), 175, 252, 497, 595, 668, 745, 906, 1040 and 1114 keV γ-rays of ^{70}As ($t_{1/2}$=52.6min) and 630 and 834 keV γ–rays of ^{72}As ($t_{1/2}$=26.0hr) are predominant at 14 MeV. At 26 MeV,

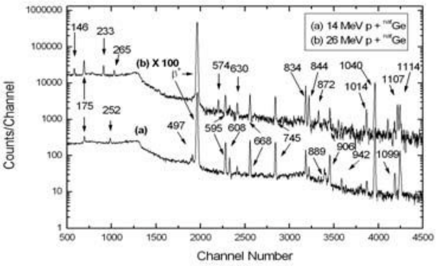

Fig. 1 Spectra taken ~35 minutes after the bombardment of 14 and 26 MeV protons

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146 and 233 keV γ–rays of short-lived ^{69}As ($t_{1/2}$=15.2min) in addition to all the γ–rays pertinent to ^{70}As and ^{72}As are shown in Fig. 1(b). 175 keV γ–ray has the contribution of ^{71}As ($t_{1/2}$=2.70d) from its corresponding half-life analysis. 574, 872 and 1107 keV γ–rays of ^{69}Ge ($t_{1/2}$=39.05hr) shown in Fig. 1(b) were emitted at the β$^+$-decay of ^{69}Ge from the ^{70}Ge(p,pn) ^{69}Ge reaction or the β$^+$-decay of ^{69}As produced in the ^{70}Ge(p,2n)^{69}As reaction. In Fig. 2(a), 630, 894 and 1051 keV γ–rays of ^{72}As, 596 and 635 keV γ–rays of ^{74}As ($t_{1/2}$=17.8d), and 559 and 657 keV γ–rays of ^{76}As ($t_{1/2}$=26.3hr) are dominant at 14 MeV. The γ–rays pertinent to ^{71}As and ^{69}Ge produced at 26 MeV are also shown in Fig. 2(b).

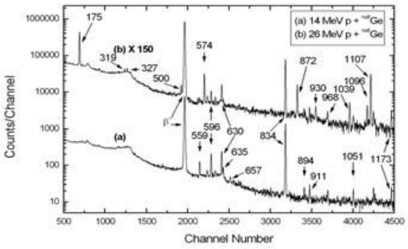

Fig. 2 Spectra taken ~1 day after the bombardment of 14 and 26 MeV protons

Typical half-life analysis for 834 and 595 keV γ–rays at 14 MeV is shown in Fig. 3 and Fig. 4, respectively. The decay curve of 834 keV line in Fig. 3 shows that it is emitted from ^{72}As. As shown in Fig. 4, the 595 keV line has two components: 595.2 keV of ^{70}As and 595.8 keV of ^{74}As. Similarly, the half-life analysis for 175 keV line observed at 26 MeV showed two components: 175.3 keV of ^{70}As and 174.9 keV of ^{71}As.

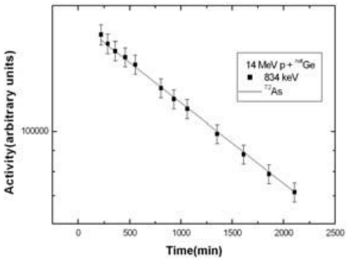

Fig. 3 Half-life analysis for 834 keV γ–ray observed at 14 MeV. Solid squares represent the data. The line represents the CLSQ fitting.

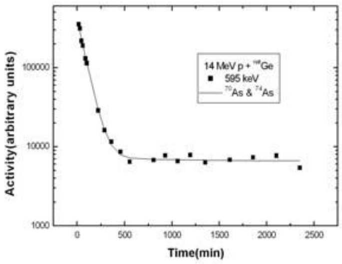

Fig. 4 Half-life analysis for 595 keV γ–ray observed at 14 MeV. Solid squares represent the data. The line represents the CLSQ fitting.

Since natural Ge consists of ^{70}Ge(21.23%), ^{72}Ge(27.66%), ^{73}Ge(7.73%), ^{74}Ge(35.94%) and ^{76}Ge(7.44%) [3], radionuclides of interest could be produced from several reactions. For example, ^{72}As could be produced by ^{72}Ge(p,n)^{72}As and ^{73}Ge(p,2n)^{72}As reactions, but the former reaction predominantly prevails at 14 MeV due partly to higher natural abundance in ^{72}Ge. The (p,n) reaction is more prevailing at the lower bombarding energy, while the (p.2n) reaction at the higher bombarding energy. Hence one could more accurately obtain the cross sections of ^{70}As and ^{76}As. The reaction cross sections for ^{70}Ge(p,n)^{70}As, ^{70}Ge(p,2n)^{69}As+^{70}Ge(p,pn)^{69}Ge, ^{72}Ge(p,2n) ^{71}As, ^{74}Ge(p,n)^{74}As and ^{76}Ge(p,n)^{76}As are shown in Table I with the previous data from [9]. They appeared to be systematically lower than those reported in [9]. This observance should be scrutinized more systematically.

TABLE I
REACTION CROSS SECTIONS

Projectile Energy (MeV)	Reaction	Cross Section (mb)	
		This work	Previous work [9]
13.8	^{70}Ge(p,n)^{70}As	300±30	575±58
13.8	^{74}Ge(p,n)^{74}As	326±23	513±51
13.8	^{76}Ge(p,n)^{76}As	111±10	182±18
26.1	^{70}Ge(p,2n)^{69}As + ^{70}Ge(p,pn)^{69}Ge	404±40	786±79[a]
26.1	^{72}Ge(p,2n)^{71}As	401±30	521±52[b]

[a] The value is the average of the accumulative cross sections for ^{70}Ge(p,2n)^{69}As and ^{70}Ge(p,pn)^{69}Ge measured at 25.7 and 26.6 MeV in [9].

[b] The value is the average of the values measured at 25.7 and 26.6 MeV in [9].

The cross sections of radionuclides produced in the interaction of Ge with protons could be explained in the compound nucleus reaction model [4]. In the model the total reaction cross section is calculated as follows:

$$\sigma = \sigma_{CN} P_{eq} P_{xn}, \qquad (1)$$

where σ_{CN} is cross section for forming the compound nucleus, P_{eq} is probability that the compound nucleus will survive pre-equilibrium decay, and P_{xn} is probability that the compound nucleus will emit neutrons and decay to the nucleus of interest. The cross section for forming the compound nucleus is obtained from the relation

$$\sigma_{CN} = \lambda\!\!\!^{-}{}^{2}_{ent} \frac{2I_c + 1}{(2I_p + 1)(2I_t + 1)} \frac{\Gamma_{ent}\Gamma}{(E - E_0)^2 + (\Gamma/2)^2}, \qquad (2)$$

where $\lambda\!\!\!^{-}$ is the relative wavelength in the entrance channel, I_c, I_p and I_t are nuclear spins of the compound nucleus, projectile and target, respectively, and E_0 is the center-of-mass energy at which resonance occurs. Γ_{ent} is the partial decay into the entrance channel while Γ is the total width of the exit channel and can be represented by the approximate relation $\Gamma=\Gamma_n+\Gamma_p+\Gamma_\gamma$, where Γ_n, Γ_p and Γ_γ are the partial widths for neutron emission, proton emission and γ–ray emission, respectively.

Assuming that Γ_γ is small compared to Γ_n and Γ_p,

$$P_{xn} = \frac{\Gamma_n}{\Gamma} = \frac{\Gamma_n}{\Gamma_n + \Gamma_p} = \frac{\Gamma_n / \Gamma_p}{\Gamma_n / \Gamma_p + 1}. \qquad (3)$$

If P_{eq} is close to unity, the reaction cross section can be rewritten as follows:

$$\sigma = ƛ_{ent}^{2} \frac{2I_c + 1}{(2I_p + 1)(2I_t + 1)} \frac{\Gamma_{ent}\Gamma}{(E - E_0)^2 + (\Gamma/2)^2} \frac{\Gamma_n / \Gamma_p}{\Gamma_n / \Gamma_p + 1} \qquad (4)$$

Internuclear barrier is taken from the Bondorf, Sobel and Sperber (BSS) coulomb potential [10]

$$V_{BSS}(r) = \begin{cases} \dfrac{Z_p Z_t e^2}{r} & for\ r \geq R_c \quad (R_c = R_p + R_t) \\ V_0 - Kr^n & for\ r < R_c \end{cases} \qquad (5)$$

where Z_p and Z_t are Z's of the projectile and target, respectively, R_p and R_t are the radii of the projectile and target, respectively. V_0, n and K are as follows:

$$V_0 = 0.6e^2 \left[\frac{(Z_p + Z_t)^2}{(R_p^{1/3} + R_t^{1/3})^3} - \frac{Z_p^2}{R_p} - \frac{Z_t^2}{R_t} \right]$$

$$n = \frac{e^2 Z_p Z_t}{R_c (V_0 - e^2 Z_p Z_t / R_c)} \qquad (6)$$

$$K = \frac{V_0 - e^2 Z_p Z_t / R_c}{R_c}$$

Bass proximity potential [11] is estimated by

$$V_{Bass}(r) = V_{coul} + \frac{\hbar^2 L^2}{2\mu r^2} - a_s A_p^{1/3} A_t^{1/3} \frac{d}{R_c} e^{-\frac{r - R_c}{d}} \qquad (7)$$

where L is the total angular momentum of the system and μ is the reduced mass. A_p and A_t are mass numbers of the projectile and target, respectively. d is the range parameter while as is the surface term in the liquid drop model mass formula. The V_{coul} term in BSS potential can be replaced by V_{BSS}

$$V_{Bass}(r) = V_{BSS} + \frac{\hbar^2 L^2}{2\mu r^2} - a_s A_p^{1/3} A_t^{1/3} \frac{d}{R_c} e^{-\frac{r - R_c}{d}} \qquad (8)$$

The radii used in the calculation are obtained from the relation $r_0 A^{1/3}$ and the empirical values for V_0, r_0 and d are -67 MeV, 1.06 fm and 0.6 fm, respectively. The cross sections were estimated under the condition that Γ_n is approximately equal to Γ_p.

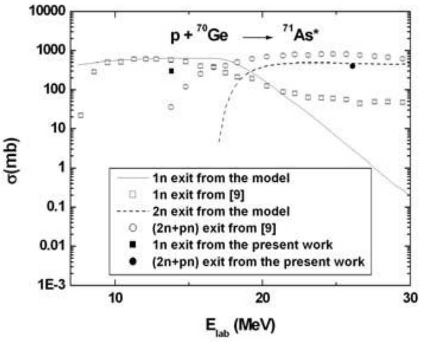

Fig. 5 Solid and dashed lines refer to the cross sections for ^{70}Ge(p,xn)$^{71-x}$As reactions with x=1-2 estimated by the compound nucleus reaction model. Open symbols refer to the data from [9] and the closed ones to the present work.

The calculated cross sections are shown for ^{70}Ge(p,n)^{70}As and ^{70}Ge(p,2n)^{69}As in Fig. 5, where the present results are displayed along with the previous data from [9]. Fig. 5 shows that the previous data for the (p,n) reaction in the projectile energy ranging from 9 to 22 MeV are in good agreement with those estimated by the compound nucleus reaction model. The calculated cross sections for the (p.2n) reaction could not be compared with its pertinent empirical data because they are not available. Similarly, the measured and calculated cross sections for ^{72}Ge(p,2n)^{71}As are shown in Fig. 6. Fig. 6 shows good agreement between them. The compound nucleus reaction model calculation indicates that ^{70}As could be produced effectively from the ^{70}Ge(p,n)^{70}As reaction in the vicinity of 15 MeV while ^{69}As could be produced in the vicinity of 23 MeV. However, the previous data from [9] revealed that the cross section for ^{70}As production peaked around 12 MeV that is lower than the value estimated by the compound nucleus reaction model.

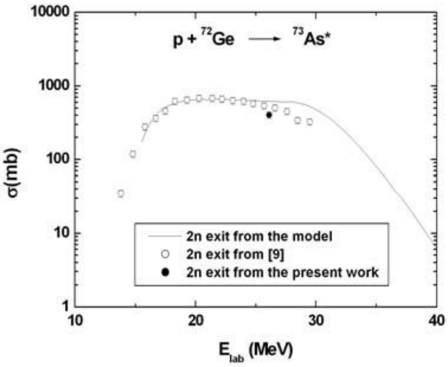

Fig. 6 Solid line refers to the cross sections for ^{72}Ge(p,2n)^{71}As estimated by the compound nucleus reaction model. Open symbols refer to the data from [9] and the closed one to the present work.

IV. CONCLUSION

The cross sections for ^{69}As, ^{70}As, ^{71}As, ^{74}As and ^{76}As produced by (p,n) or (p,2n) reactions have been measured by off-line γ–ray spectroscopy. The results were compared with those estimated by the compound nucleus reaction model and the previous data.

The cross sections previously measured for ^{72}Ge(p,2n)^{71}As were observed to be in good agreement with its calculated value in the energy region ranging from 14 to 30 MeV, while there was fair agreement between them for ^{70}Ge(p,n)^{70}As in the energy region from 9 to 22 MeV. Similar results for ^{72}Ge(p,2n)^{71}As and ^{76}Ge(p,n)^{76}As were obtained in this energy region, implying that the compound nucleus reaction model could be of use in estimating the pertinent reaction cross sections in proton-induced reactions.

The result shows that the radionuclides for PET such as ^{69}As and ^{70}As could be effectively produced by ^{70}Ge(p,2n)^{69}As in the energy region ranging from 20 to 25 MeV and $^{70}Ge(p,n)^{70}As$ in the energy region from 9 to 18 MeV, respectively.

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