III. 3. Production of $^{28}$Mg for Biological Studies

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Introduction

Magnesium plays an important role in the living system as an essential element. Many biological and biochemical functions of Mg, however, have not been fully understood, mainly because there is no other radiotracer suitable for biological study but $^{28}$Mg, a short-lived $\beta^-$ emitter with a half-life of 20.9 hr. The $^{27}$Al($\alpha$, 3p)$^{28}$Mg reaction was our choice for the production of this radionuclide due to high specific activity. Although a conventional method for the preparation of $^{28}$Mg from Al targets has already been reported 1), we have developed an alternate method for the production of no-carrier-added and salt-free $^{28}$Mg using only column chromatographic technique, suitable for routine preparation.

Materials and methods

Two stacked Al foils (20 x 20 mm, 100 $\mu$m thick; purity: 99.999%, Nilaco, Japan) were irradiated by a 50-MeV $\alpha$ particle beam from the AVF cyclotron of CYRIC. The calculated energies deposited in the target for the production of $^{28}$Mg ranged from 48.4 to 41.7 MeV. The one hour irradiations were usually carried out at an average current of 5 $\mu$A. After the irradiation the activated areas (usually 20 mm diameter; approx. 170 mg) were cut out of the target foils, and 20 ml of 2M HCl was then added to them in a flask with a few mg of copper(II) fluoride to facilitate the pure Al to dissolve in a diluted HCl solution by depositing copper and also to add a carrier for $^{18}$F. The flask was gently heated until the foils were completely dissolved, and the solution was then evaporated to dryness. A small portion of water was added to the residue and the evaporation was repeated to completely remove HCl.

The evaporated residue was dissolved in 4 ml of 2M NH$_4$SCN, and the resulting solution was passed through Sep-Pak Plus tC$_{18}$ cartridges (Waters) which had been treated with tri-n-butyl phosphate (TBP) and then washed with 2M NH$_4$SCN in advance $^{3,4}$. The cartridges were washed by repeating the above procedure, and the combined eluates were mixed with 20 ml of 0.5M oxalic acid.
The separation of $^{28}\text{Mg}$ from the target aluminum was carried out according to the method developed by Stelow and Van Ver Walt\(^2\). After the mixed solution had been applied onto the cation exchange resin, AG50W-X4 (H\(^+\) form, 100-200 mesh, Bio-Rad; 12 mm I.D. and \(\text{ca} \ 30 \ \text{cm} \) long), it was washed with 400 ml of 0.5M oxalic acid to elute aluminum ions and then with 40 ml of 0.01M HCl to remove oxalic acid from the column. The $^{28}\text{Mg}$ was finally eluted with 60 ml of 2M HCl. The last 40 ml portion of the eluate was collected and evaporated to dryness, and the residue was finally dissolved in an appropriate solvent such as saline for subsequent use in biological study.

**Results and discussion**

Chemical separation is carried out for removal of not only a target material but also concomitantly produced radionuclides from $^{28}\text{Mg}$. A separation method for providing no-carrier-added and salt-free $^{28}\text{Mg}$ is more preferable for biological tracer use. Table 1 lists possible radionuclides produced by a-irradiation of pure Al. In the present study, the formations of these radioisotopes were observed as shown in Fig.1, together with one rather long-lived positron emitter, which was identified as $^{18}\text{F}$ from the decay curve analysis of a 511 keV peak. Fluorine-18 was produced from the oxygen present in the target surface via the $^{16}\text{O}(\alpha,\text{pn})^{18}\text{F}$. Since $^{27}\text{Mg}$ and $^{29}\text{Al}$ decay out rapidly, only $^{7}\text{Be}$, $^{18}\text{F}$, $^{22}\text{Na}$ and $^{24}\text{Na}$, are the target radionuclides to remove.

The method developed by Weinreich et al.\(^1\) was mainly based on the separation by precipitation, which includes the filtration of the Al(OH)\(_3\) gel and consequently this procedure takes long time and sometimes results in serious loss of $^{28}\text{Mg}$. On the other hand, the present method using the cation exchange column originally developed by Stelow and Van Der Walt\(^2\) is very convenient and offers an excellent separation of a trace amount of Mg from Al. Among the undesirable radionuclides shown above, $^{18}\text{F}$, $^{22}\text{Na}$ and $^{24}\text{Na}$ can be removed with this column. In fact, radioactive sodium was observed to gradually elute from the cation exchange column over 300 ml of the solvent. On the other hand, $^{7}\text{Be}$ behaves quite similarly to $^{28}\text{Mg}$ with this column. Therefore, the TBP-silica gel column was successfully introduced to remove $^{7}\text{Be}$ before the cation exchange column. The $^{7}\text{Be}$ was efficiently retained by the four combined TBP-silica gel columns, prepared from a commercially available column of reverse phase silica gel. Accordingly no $^{7}\text{Be}$ contaminated the final product solution as demonstrated in Fig. 1 (b). In addition, a trace quantity of $^{48}\text{V}$, which might be derived from a Ti window of the cyclotron beam transfer duct, was sometimes found on the TBP-silica gel.

As seen in Fig. 1 no other $\gamma$-ray except those derived from $^{28}\text{Mg}$ or its daughter radionuclide, $^{28}\text{Al}$, was observed on the spectrum of the finally purified sample. The overall recovery yield from the irradiated target was $2.0 \pm 0.5 \ \mu\text{Ci}/\mu\text{Ah}$, $85 \pm 5\%$, based on the production yield measured on the target. This value was very consistent with the theoretical value, estimated from the excitation curve in the literature\(^5\). As $^{28}\text{Mg}$ was eluted with 2M
HCl from the cation exchange column, practically salt-free $^{28}$Mg could be finally obtained. The overall purification procedure usually took less than 2 hrs. Consequently, it can be concluded that the present method has simplified the recovery and separation of no-carrier-added $^{28}$Mg from a pure Al target and is hence suitable for biological tracer use.

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References


Table 1. Radionuclides produced by $\alpha$-irradiation of pure Al.

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half-life</th>
<th>Decay mode</th>
<th>Nuclear reaction</th>
<th>Q-value</th>
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<tr>
<td>$^{22}$Na</td>
<td>2.602 y</td>
<td>$\beta^+$.EC</td>
<td>$^{27}$Al($\alpha$, $2\alpha$)$^{22}$Na</td>
<td>-22.5 MeV</td>
</tr>
<tr>
<td>$^{22}$Na</td>
<td>15.03 h</td>
<td>$\beta^-$</td>
<td>$^{27}$Al($\alpha$, $\alpha$)+$^{24}$Na</td>
<td>-31.4 MeV</td>
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<tr>
<td>$^7$Be</td>
<td>53.29 d</td>
<td>EC</td>
<td>$^{27}$Al($\alpha$, $^7$Be)$^{24}$Na</td>
<td>-22.1 MeV</td>
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<tr>
<td>$^{27}$Mg</td>
<td>9.46 m</td>
<td>$\beta^-$</td>
<td>$^{27}$Al($\alpha$, 3$\alpha$)$^{27}$Mg</td>
<td>-30.1 MeV</td>
</tr>
<tr>
<td>$^{28}$Mg</td>
<td>20.93 h</td>
<td>$\beta^-$</td>
<td>$^{27}$Al($\alpha$, 3$\beta$)$^{28}$Mg</td>
<td>-21.6 MeV</td>
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<tr>
<td>$^{28}$Al</td>
<td>2.24 m</td>
<td>$\beta^-$</td>
<td>$^{27}$Al($\alpha$, 2$\beta$)$^{28}$Al $^{28}$Mg $\rightarrow$ $^{28}$Al</td>
<td>-20.6 MeV</td>
</tr>
<tr>
<td>$^{29}$Al</td>
<td>6.56 m</td>
<td>$\beta^-$</td>
<td>$^{27}$Al($\alpha$, 2p)$^{29}$Al</td>
<td>-11.1 MeV</td>
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Fig. 1. Gamma-ray spectra of (a) irradiated Al target and (b) separated Mg-28.