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THE PREPARATION OF MAGNESIUM-28 BY THE IRRADIATION IN HIFAR OF MIXTURES CONTAINING COMPOUNDS OF LITHIUM-6 AND MAGNESIUM-26

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ABSTRACT

An examination of the specific activities and yields of ^{28}Mg obtained by the irradiation in HIFAR of a number of mixtures of lithium carbonate with either magnesium carbonate or magnesium oxide showed that up to 0.25 mCi of ^{28}Mg at approximately 6 mCi g^{-1} of magnesium could be obtained. A separation procedure based on the precipitation of magnesium hydroxide is described and the results of a number of preparations by this method are presented.

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1. INTRODUCTION

On half life considerations ^{28}Mg (half life 21.5 hours) is the only nuclide of magnesium to be of value as a tracer. It has been used in medicine to investigate magnesium metabolism (Silver 1968) and also in studies of animal nutrition (Rogers 1965). However the availability of this nuclide is subject to some limitations. Carrier free ^{28}Mg can be obtained by nuclear reactions such as $^{37}Cl(p, 6p\ 4n)\ ^{28}Mg$ or $^{27}Al(^4He, 3p)\,^{28}Mg$ (Hudis 1957) using a particle accelerator; the $^{26}Mg(t, p)\ ^{28}Mg$ reaction can be used but will result in a lower specific activity as the nuclide is no longer carrier free. The last method has the advantages of a relatively high cross section (8 mb for 2.73 MeV tritons (Bishop 1962)) and lower accelerator cost (the triton beam can be obtained by means of an electrostatic accelerator (Mori et al. 1967)).

In a nuclear reactor ^{28}Mg can only be obtained by the secondary reaction brought about by the tritons arising from the irradiation of ^6Li. By using the sequence of reactions: $^6Li(n, \alpha)t$ and $^{26}Mg(t, p)\,^{28}Mg$ the production of ^{28}Mg was studied over a number of years at Brookhaven National Laboratory (e.g. Hillman 1963; Stang 1964). Methods were developed there to manufacture and irradiate an alloy containing only ^6Li and ^{26}Mg; the absence of nuclides not involved in the reactions enables the maximum efficiency to be attained for the secondary reaction in the system. In this way ^{28}Mg has been obtained at specific activities of 17.5 mCi g^{-1} (Mirza 1970) and 40 mCi g^{-1} (Stang 1964).

However, there are problems associated with the use of lithium-magnesium alloy. Not only is considerable heat generated in the $^6Li\ (n,\alpha)t$ reaction (cross section 950 barns) but also thin foils must be used so that there is less self attenuation of the neutron flux. Consequently the alloy must be cooled efficiently during irradiation and, because of its reactivity with air, the foil must be sealed in an inert atmosphere.

^{28}Mg produced in this way is not free from contaminating nuclides. Excluding very short half lived nuclides, where decay is virtually complete within one hour of removal from the reactor, the following contaminants remain: (i) tritium from the $^6Li\ (n,\alpha)t$ reaction, (ii) ^{18}F from the $^{16}O(t, n)\,^{18}F$ reaction produced between the oxide film and the alloy foil, (iii) ^{24}Na from the $^{25}Mg(t,\alpha)\,^{24}Na$ and $^{24}Mg(n, p)\,^{24}Na$ reactions if the ^{26}Mg abundance is less than 100% (Hillman 1969) and (iv) activation of any sodium impurities.

The irradiation of materials containing ^6Li in HIFAR (the AAEC's High Flux Australian Reactor) for periods of a few days, is probably best carried out in the 4H self service positions where there is a high thermal neutron flux (about 5×10^{13} neutrons $cm^{-2}\ s^{-1}$) and where the heat generated can be removed quickly. However it was considered unlikely that the available rig design could provide sufficient cooling to prevent ^6Li alloys from melting during irradiation. An alternative method involving the irradiation of mixtures of ^6Li as the carbonate with ^{26}Mg as either the oxide or carbonate was therefore studied. Although it was expected that both a lower specific activity of ^{28}Mg would result due to the dissipation of triton energy by atoms of the other elements present and larger quantities of radio contaminants would be produced, considerable advantages could also be anticipated. These were in the main related to the ready availability of ^6Li as lithium carbonate and ^{26}Mg as magnesium oxide as well as the stability of these compounds to heat and air; this would avoid the necessity of special target encapsulation in an inert atmosphere during irradiation.

2. EXPERIMENTAL

2.1 ^{28}Mg Yields and Specific Activities

Mixtures of lithium carbonate with magnesium carbonate at natural abundance were prepared by coprecipitation. Mixtures of lithium carbonate with magnesium oxide at either natural or 99.7% ^{26}Mg abundance were prepared by grinding the compounds together. ^6Li abundances from natural up to 96% were used. The mixtures were sealed into titanium capsules by welding and irradiated in a 4H self service facility (X33) in HIFAR.

After irradiation the mixture was dissolved in acid and the magnesium quantitatively isolated by precipitation with 8-hydroxyquinoline (Vogel 1957). The product was analysed by gamma spectrometry, and found to be free from gamma emitting radio contaminants. The ^{28}Mg activity was measured in a calibrated 4π gamma ionisation chamber. ^{28}Mg yields and specific activities are shown in Table 1.

2.2 Method of ^{28}Mg Separation

2.2.1 Removal of ^{18}F

After irradiation a lithium carbonate-magnesium oxide mixture was dissolved in dilute hydrochloric acid and the solution passed through a 5 mm diameter column containing 1 g of hydrous zirconium oxide (Bio-Rad HZO-1, 50-100 mesh). Although the removal of ^{18}F in the column was substantial it was not complete and the amount absorbed was not increased by the prior addition of 10 mg of sodium fluoride to the solution.

A large improvement in ^{18}F removal was achieved by adding 2-3 ml of water followed by 0.5 ml of hydrofluoric acid and 2 ml of concentrated sulphuric acid to the irradiated lithium carbonate-magnesium oxide mixture. Most of the ^{18}F was then removed by fuming to dryness. By repeating the fuming process the ^{18}F contamination could usually be reduced to below 2% of the ^{28}Mg activity.

2.2.2 Separation of ^{28}Mg on a Chelating Resin

A preliminary tracer experiment showed that magnesium was effectively removed from neutral or ammoniacal solution by a column of Chelex 100 (Bio-Rad Laboratories) and could be recovered by elution with approximately 2 M hydrochloric acid. A mixture of 250 mg natural magnesium oxide with 450 mg lithium carbonate (97% ^{6}Li) irradiated for 48 hours was dissolved in 15 ml of about 1 M hydrochloric acid. After removal of most of the ^{18}F, 1 g of sodium chloride and 3 g of ammonium chloride were added and the solution was neutralised with ammonia then diluted to 100 ml. Passage of this solution through a 20 mm diameter column containing 20 g of Chelex 100, 50–100 mesh, resulted in almost complete absorption of the ^{28}Mg. The column was washed with sodium chloride solution and water then the ^{28}Mg was eluted in 100 ml of approximately 2 M hydrochloric acid. However it was found by gamma spectrometry that the separation from ^{24}Na was incomplete and the ^{28}Mg activity was contaminated in excess of 5% by ^{24}Na.

2.2.3 Separation of ^{28}Mg by Precipitation of Magnesium Hydroxide

A mixture of 125 mg natural magnesium oxide with 225 mg lithium carbonate (97% ^{6}Li) was irradiated. The ^{18}F was removed by fuming twice with sulphuric acid and the residue diluted to 20 ml with water. Magnesium hydroxide was precipitated by the addition of sodium hydroxide solution and separated in a centrifuge. The precipitate was found to be contaminated with ^{24}Na but this contamination was reduced by (i) the addition of 1 g of sodium chloride 'holdback' carrier each time before precipitating magnesium hydroxide and (ii) washing the precipitate a number of times with sodium chloride solution.

After two precipitations and washing each precipitate four times with sodium chloride the contamination with ^{24}Na was consistently $<$ 5% of the ^{28}Mg activity. Final ignition of the hydroxide precipitate at 1000°C for 10 minutes reduced contamination with ^{18}F and ^{3}H. The residual tritium was less than 10 μCi per 0.2 mCi of ^{28}Mg. Approximately 90% of the magnesium was recovered in this procedure.

^{28}Mg has been produced on a number of occasions and isolated by this method. The residue of magnesium oxide from ignition was dissolved in either acetic of hydrochloric acid as required. In Table 2 the yields, specific activities and contamination with ^{24}Na and ^{18}F are shown for these preparations.

3. DISCUSSION

The specific activity of ^{28}Mg obtained by irradiating a mixture of ^{26}MgO with $^{6}Li_2CO_3$, as expected, was much lower than 40 mCi g^{-1} obtained by the irradiation of lithium-magnesium alloy at a lower neutron flux (Stang, 1964). However it can be seen (Table 2) that ^{28}Mg with a specific activity of about 6 mCi g^{-1} can be regularly obtained by this method in quantities useful for tracer experiments. Even if magnesium oxide at natural abundance was used about 40 μCi of ^{28}Mg could be obtained although at a much lower specific activity.

Whilst the cost of production of ^{28}Mg in this way is increased by the quantities of enriched nuclides used, a saving is effected both by not having to prepare a lithium magnesium alloy and also by not having to specially encapsulate the mixtures for irradiation. The disadvantage found in using compounds of lithium and magnesium in place of the elements lay in the production of much larger amounts of contaminating nuclides. In spite of the large excess of ^{18}F also produced, the effective removal of this nuclide was readily obtained by fuming with sulphuric acid and was virtually complete by ignition of magnesium hydroxide at 1000°C. As a period of some hours was usually required for shipment of the isolated ^{28}Mg to the user,the level of contamination by ^{18}F was further reduced because of the more rapid radioactive decay of this nuclide (half life 110 minutes).

Removal of ^{24}Na by precipitation of magnesium hydroxide proved more difficult to achieve. This difficulty could be attributed to retention of sodium on the hydroxide precipitate by an ion exchange mechanism. In contrast, the precipitation of magnesium with 8-hydroxyquinoline gave an excellent separation from both ^{18}F and ^{24}Na. This is a well known and reliable method for the quantitative isolation of magnesium. However it was used here only in the assay of the amount and the specific activity of ^{28}Mg arising in the irradiation samples because the ^{28}Mg produced in these experiments was required for pharmaceutical applications where freedom from organic residues is essential. Although precipitation with 8-hydroxyquinoline is potentially capable of leading to radioisotopically purer ^{28}Mg, further work is required to determine the conditions under which ^{28}Mg might be recovered from an oxide precipitate free from 8-hydroxyquinoline and organic degradation products. However because a satisfactory yield and purity is obtained by precipitation of magnesium hydroxide and the demand for this nuclide is limited, this further investigation is not yet warranted.

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TABLE 1

YIELD AND SPECIFIC ACTIVITY OF ^{28}Mg FROM THE IRRADIATION OF MIXTURE OF LITHIUM AND MAGNESIUM COMPOUNDS (5×10^{13} n cm^{-2} sec^{-1} approx.)

Mixture				Method of Mixing	Irradiation		^{28}Mg Produced		
Lithium		Magnesium							
% by Weight	% Abundance ^{6}Li	% by Weight	% Abundance ^{26}Mg		Weight of Mixture (mg)	Period (hr)	Activity (μCi)	Specific Activity (mCi g^{-1})	Remarks
26	60	8.7	Natural (11.0)	coprecipitated carbonate	350	6	4.5	0.15	melted on irradiation
21	60	7.2	〃	〃	350	6	2.6	0.10	–
15.5	97	3.5	〃	Li_2CO_3 + MgO milled together	156		3.1	0.7	–
13.5	97	10.5	〃		178	6	7	0.4	–
10.5	97	21.5	〃	〃	180	6	15	0.4	–
8.0	97	32	〃	〃	147	6	18	0.4	–
6.5	97	37	〃	〃	163	6	18	0.3	–
5.7	97	39.5	〃	〃	151	6	18	0.3	–
10.7	97	14	〃	Li_2CO_3+ MgO ground together	309	48	35	1.0	fused in can
9.5	97	17	〃		325	48	67	0.86	free flowing
8.3	97	20	〃	〃	326	48	80	0.90	〃
7.1	97	23	〃	〃	324	48	87	0.83	〃
5.9	97	26	〃	〃	324	48	95	0.83	〃
9.5	96	17	99.7	〃	193	48	260	6	–
12.5	96	10	〃	〃	288	48	220	7	–
5.8	96	26	Natural (11.0)	〃	350	48	38	0.44	lumps formed in irradiation
5.8	96	26	〃	〃	350	48	30	0.3	–
12.5	96	15	99	〃	200	48	170	6	–

TABLE 2

YIELD AND SPECIFIC ACTIVITY OF ^{28}Mg PRODUCED ROUTINELY BY HYDROXIDE PRECIPITATION

All material irradiated at 5 x 10^{13} neutrons cm^{-2} s^{-1} (approx.) for 48 hours

Weight MgO (99% ^{26}Mg) (mg)	Weight Li_2CO_3 (96% ^{6}Li) (mg)	^{28}Mg Activity (ex reactor) (mCi)	^{28}Mg Specific Activity (ex reactor) (mCi g^{-1})	Efficiency of MgO Recovery (%)	^{24}Na Content of ^{28}Mg (%)	^{18}F Content of ^{28}Mg (%)
50	150	0.175	6.3	96	5	5
60	180	0.175	5.4	90	5	2
60	180	0.21	6.4	88	3	2
60	180	0.143	4.9	78	4	1
60	180	0.194	5.6	93	10	2
60	180	0.154	5.9	70	1	1
60	180	0.28	7.5	99	3	1
60	180	0.173	5.9	78	5	1
60	180	0.31	9.3	90	2	5
60	180	0.189	6.5	88	5	1
60	180	0.23	6.3	99	7	1
60	180	0.149	4.6	87	1	1
60	180	0.20	6.5	85	9	1
450 *	250	0.076	0.44	65	5	2

* Mg at natural abundance