The Production of High Specific Activities of Tin

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Radioactive substances of a high specific activity are very useful in nuclear spectroscopy and in the tracer technique. In this work the production of the two long-lived radioactivities of tin, ^{113}Sn and ^{117m}Sn, was studied by means of cyclotron bombardment. These nuclides can be obtained separately from targets of a natural isotopic abundance by means of a cyclotron with a fixed frequency, provided the two reactions, ^{113}In(d, 2n)^{113}Sn and ^{115}In(α, pn)^{117m}Sn, are aimed at respectively. The purposes of the present investigation are to determine the thick target yield for these reactions and to establish the procedure for preparing the high specific activities of tin from the irradiated target.

Maxwell et al.[1] have reported procedures for the preparation of high specific activities of tin by means of alpha particle-induced reaction on cadmium of a natural abundance. In this case ^{113}Sn and ^{117m}Sn are produced concomitantly from the reactions ^{110}Cd(α, n)^{113}Sn, ^{111}Cd(α, 2n)^{113}Sn and ^{114}Cd(α, n)^{117m}Sn. In the present work, the thick target yields for these nuclides have also been determined.

Experimental

Thick Target Yield.—*Bombardment.*—Indium and cadmium plates of about 150 mg./cm^2 were bombarded with 11.2 MeV. deuterons or 22.1 MeV. alpha particles from the 110 cm. cyclotron of Osaka University. The target materials were 99.9% chemically pure. They were soldered on a water-cooled block of copper and irradiated at the "external target" position. The incident particle energies were degraded with aluminum absorbers and estimated from the range-energy relation.[2] The beam current was kept as low as about 2 μ amp. to prevent the target materials from melting. Total activations of about 2 μ amp.$\times$0.5 hr.=3600 microcoulombs were performed.

The total numbers of incident projectiles were determined by the use of monitor foils, by, that is, chromium and silver foils for deuterons and alpha particles respectively. The excitation function for the ^{52}Cr(d, 2n)^{52}Mn reaction[3] and the ^{107}Ag+$\alpha\rightarrow$ ^{109}Cd reaction[4] were used for the calibration.*

Source Preparation.—The procedures of chemical separations before the measurement of the tin activities were as follows. The irradiated cadmium target was dissolved in aqua regia. About 20 mg. of a iron(III) carrier was added, and the tin activity was coprecipitated with ferric hydroxide from a 2 N ammonia solution. The precipitate was then redissolved in 2 ml. of diluted hydrochloric acid.

The irradiated indium target was treated with hydrochloric acid, and a 20 mg. carrier of tin was added. The tin activity was separated from a 0.5 N hydrochloric acid solution as a sulfide. The sulfide was dissolved in 2 ml. of concentrated hydrochloric acid.

Activity Measurement.—The tin activities produced were measured with a gamma ray scintillation spectrometer. After the interfering activities had decayed off after several days, the activities of the 2 ml. liquid sample mentioned above were measured spectrometrically in a 1(3/4)''$\times$2'' well-type NaI scintillator. The photopeaks of the 390 keV. gamma ray from ^{113}Sn$-^{113m}$In and the 160 keV. gamma ray from ^{117m}Sn were measured. When both activities were contained, the contribution of the Compton part of the 390 keV. gamma ray to the 160 keV. photopeak was subtracted.

The counting efficiencies of the photopeaks (counting rates per disintegration rate) for both activities were calculated from several data as shown in Table I.

The Obtained Thick Target Yield.—The Thick target yields of the In+d, In+α and Cd+α reactions at some incident particle energies are summarized in Table II.

The Excitation Function for the ^{115}In(α, pn)^{117m}Sn Reaction.—The low values of thick target yields for the ^{115}In(α, pn)^{117m}Sn reaction suggest that the studied region of alpha particle energies is too low for the high-yield production of the activity. In order to study further the energy dependency of the yields in the higher region, the excitation function up to 40 MeV. for this reaction was obtained

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* In the latter case two processes, ^{107}Ag(α, pn)^{109}Cd and ^{107}Ag(α, 2n)^{109}In$\rightarrow^{109}$Cd, are involved.

TABLE I. COUNTING EFFICIENCIES FOR ^{113}Sn AND ^{117m}Sn

Nuclide	Measured gamma ray MeV.	Photopeak efficiency	Total internal conversion coefficient	Intensity of the gamma ray per one disintegration	Counting efficiency
^{113}Sn	0.393	0.28	0.54	0.65	0.19
^{117m}Sn	0.159	0.95	41.0	0.79	0.74
	0.161	0.95	0.31		

TABLE II. THICK TARGET YIELDS* IN μc./μamp. hr. FOR ^{113}Sn AND ^{117m}Sn FROM THE REACTIONS In+d, In+α AND Cd+α

In+d reaction		In+α reaction		Cd+α reaction		
E_d (MeV., LAB)	^{113}Sn	E_α (MeV., LAB)	^{117m}Sn	E_α (MeV., LAB)	^{113}Sn	^{117m}Sn
7.1	$1.1(0.7)\times10^{-2}$	16.6	$3(4)\times10^{-4}$	11.5	—	$1.7\ (0.2)\times10^{-3}$
9.1	$9.1(0.6)\times10^{-2}$	19.1	$7(1)\times10^{-3}$	14.8	$2.3(0.2)\times10^{-2}$	$2.0(0.3)\times10^{-1}$
10.9	$2.6(0.2)\times10^{-1}$	20.8	$4.7(0.5)\times10^{-2}$	20.8	$3.2(0.2)\times10^{-1}$	$1.1(0.1)\times10^{0}$

* The values in the brackets are the estimated standard deviations.

by the use of the stacked foil method.

Indium was electrolytically deposited in a 10 mg./cm^2 thickness on a silver plate of 25 mg./cm^2, and the indium foil was stacked together with the backing. The excitation function was measured in the energy region of 40~22 MeV. with alpha particles of 40 MeV. from the 160 cm. cyclotron of the Institute for Nuclear Study, the University of Tokyo, and in the 22~12 MeV. region with alpha particles of 22 MeV. from the 110 cm. cyclotron of Osaka University. The incident energy for each foil was estimated from the range-energy relationship in silver.[2] After the appropriate chemical isolation of tin, the radioactivity of ^{117m}Sn was measured by the same method. The observed cross section was corrected for the straggling effect, the foil thickness and the initial beam energy spread.[4] The correction amounts, for example, to about 13% at 17 MeV.

The cross sections thus obtained for the ^{115}In-(α, pn)^{117m}Sn reaction are given in Table III and are expressed as the excitation function in Fig. 1. The thick target yield curve for this reaction calculated from the excitation function and the range-energy relation is shown in Fig. 2, together with the values of the thick target yields already given in Table II. Figure 2 shows that the yield at 40 MeV. is about twentyfold that at 20 MeV.

TABLE III. MEASURED CROSS SECTIONS FOR THE REACTION $^{115}In(\alpha, pn)^{117m}Sn$

E_α (MeV., LAB)	Cross section (mb.) with standard deviation
11.9	0.01 ± 0.02
14.9	0.04 ± 0.01
17.7	0.64 ± 0.06
22.1	$5.9\ \pm0.8$
27.1	$14\ \ \pm2$
31.5	$16\ \ \pm2$
35.4	$16\ \ \pm2$
39.1	$13\ \ \pm2$

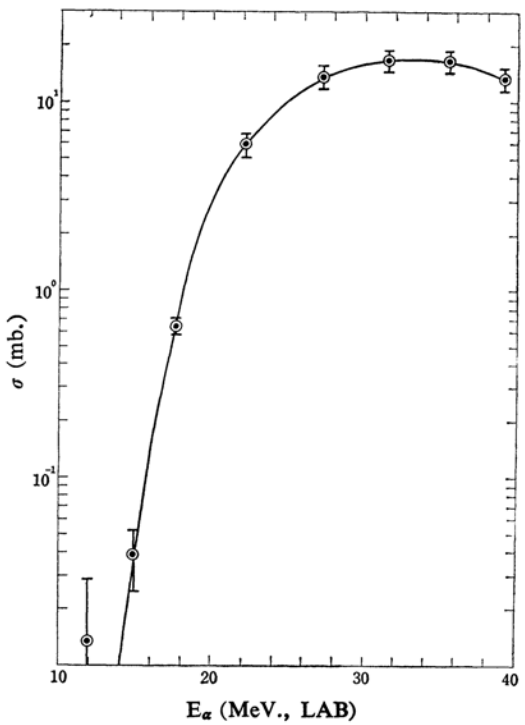

Fig. 1. Excitation function of the reaction $^{115}In(\alpha, pn)^{117m}Sn$.
Points: observed cross sections
Flags: standard deviations accompanied with the observed points
Solid curve: excitation fuction

The Preparation of High Specific Activities of Tin.—*Carrier-free Isolation.*—The irradiated indium target was dissolved in concentrated hydrochloric acid. After the addition of 20 mg. each of copper and of cadmium carriers, the tin activity was coprecipitated with copper sulfide by hydrogen sulfide

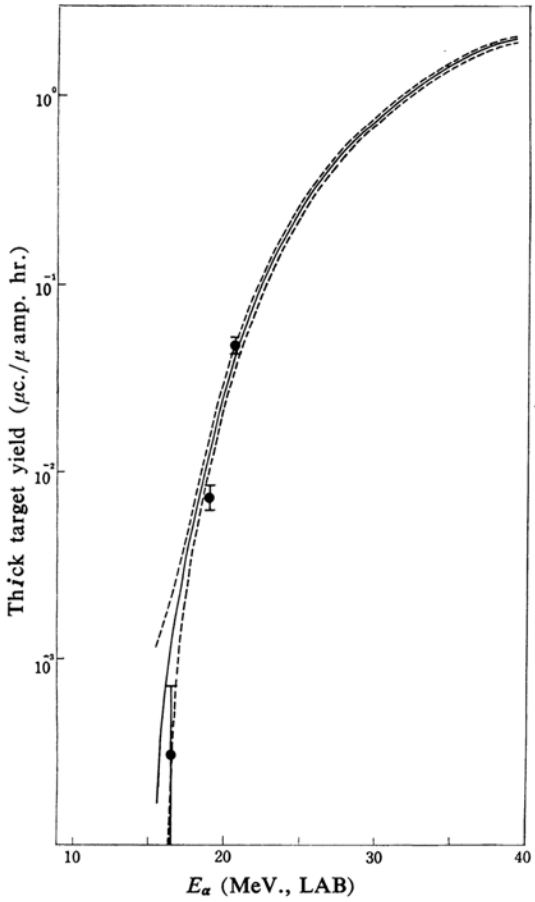

Fig. 2. Thick target yield curve for the reaction $^{115}In(\alpha, pn)^{117m}Sn$.
Solid curve: thick target yield curve calculated from the excitation function
Broken curves: upper and lower limits of the standard deviations
Closed circles: experimental values as shown in Table I
Flags: standard deviations in experimental values

from a 0.5 N hydrochloric acid solution. The sulfide was dissolved in a small amount of aqua regia, and the solution was evaporated almost to dryness. The residue was dissolved in 0.5 N hydrochloric acid. The tin activity was then again coprecipitated with copper sulfide from the solution containing 10 mg. each of indium and of cadmium carriers, After three coprecipitations, the copper sulfide with the tin activity was dissolved in a small amount of aqua regia, and then with water and hydrochloric acid; 10 ml. of a 2 N hydrochloric acid solution was obtained. The solution was passed through a column, 8 mm. in diameter, containing 3 ml. of Amberlite CG-400. After a complete elution of the copper by 30 ml. of 2 N hydrochloric acid, the carrier-free tin activity was eluted out with 25 ml. of 2 N nitric acid.

The irradiated cadmium target was dissolved in aqua regia. The solution was diluted with water, and 20 mg. of a iron(III) carrier was added. The tin activity was coprecipitated with ferric hydroxide from a 2 N ammonia solution. The reprecipitated ferric hydroxide was dissolved in hydrochloric acid, and 20 mg. each of indium and of copper carriers were added. Hydrogen sulfide was passed into the 0.5 N hydrochloric acid solution, and the tin activity was coprecipitated with copper sulfide. From this precipitate the carrier-free activity in a nitric acid solution was obtained by the same procedures as in the case of the indium target mentioned above.

Efficiencies of the carrier-free isolations of the tin activity are shown in Table IV. The nitric acid solution thus obtained has about 1 mg. of residue.

TABLE IV. EFFICIENCIES OF CARRIER-FREE ISOLATIONS OF THE TIN ACTIVITY

Target	Chemical yield of Sn activity, %	Decontamination factor of	
		In	Cd
In	87	$>10^6$	$>10^3$
Cd	94		

Electrolytic Deposition.—In order to obtain a sample for beta ray spectrometry, the activity was collected by means of an internal electrolysis from a 0.3 N hydrochloric acid solution. The apparatus for the internal electrolysis shown in Fig. 3. The anode is a zinc plate, while the cathode is a thin layer (10 μg./cm²) of gold deposited in vacuo on a thin "Mylar" tape (500 μg./cm², 2 mm.×15 mm.). About 80% of the activity was deposited in 20 hr. It must be noted that the tin activity is dissolved back into the hydrochloric acid solution in a moment if the external circuit is cut off.

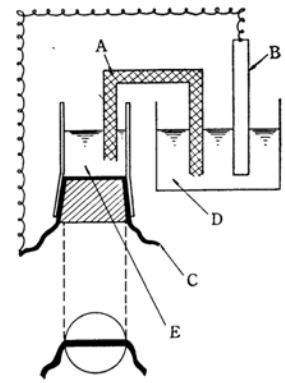

Fig. 3. Apparatus for the internal electrolysis.
A: Salt bridge (3% agar-agar, KCl satd.)
B: Zn-anode
C: Au-cathode (deposited in vacuo on "Mylar")
D: Anodic electrolyte (0.1 N $ZnCl_2$)
E: Cathodic electrolyte (0.3 N HCl, carrier-free tin activity, 0.2 ml.)

Discussion

If either ^{113}Sn or ^{117m}Sn is required separately, the In+d or In+α reactions respectively

may be used. However, if these nuclides can co-exist, the Cd+α reaction is preferable because of high thick target yield in the studied energy range and because of the higher melting point of cadmium than of indium.

The values of the thick target yield observed directly for the $^{115}In(\alpha, pn)^{117m}Sn$ reaction are in good agreement with the values from the excitation function at the energies lower than 21 MeV. If the values for the higher energies are necessary, the calculated values are available.

The separation procedure for the carrier-free activity by coprecipitation and ion exchange herein presented is efficient despite its simplicity in comparison with the distillation method.[1]

The tin activity was deposited internal-electrolytically on a gold film on "Mylar" foil (2 mm.×15 mm.) and was used as a sample source for the beta ray spectrometer of Osaka University.[5] The L and M conversion electrons of the 159 keV. gamma ray were resolved separately.[6] Thus, the specific activity of the sample was sufficiently high so as to let the spectrometer exhibit its resolving power (0.2%) fully, even at the low energy region.

Summary

The production of high specific activities of tin, ^{113}Sn and ^{117m}Sn, with a cyclotron had been studied. Thick target yields for the reactions (A) $^{113}In(d, 2n)^{113}Sn$, (B) $^{115}In(\alpha, pn)^{117m}Sn$ and (C) $^{110}Cd(\alpha, n)^{113}Sn+^{111}Cd(\alpha, 2n)^{113}Sn+^{114}Cd(\alpha, n)^{117m}Sn$ were determined for several incident particle energies. The reactions A and B are available for the production of the single tin activity. Reaction C is preferable to the others in the total yields of tin activities for the energy range studied, i.e., up to the energy of about 5 MeV. per nucleon of the incident particle. A thick target yield for reaction B has also been obtained from the excitation function up to the alpha particle energy of 40 MeV. The procedures for the carrier-free isolation of the tin activity from indium and cadmium targets by coprecipitation and ion exchange have also been described.

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