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Production of Carrier Free ^{113}Sn by Deuteron Irradiation of Indium

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Summary

Experimental measured cross sections for the In(d,xn) ^{113}Sn reaction by irradiation of natural indium with deuterons are reported.
A stacked foil arrangement was used and the excitation function in the range between 27.5 MeV and the Coulomb threshold was obtained. Total flux measurements were performed with aluminium monitor foils.
The ^{113}Sn content was determined measuring the gamma rays emited by the daughter, ^{113m}In, in equilibrium. The overall error in absolute cross sections was estimated as $\pm 20\%$.
The excitation function for the ^{113}In (d,2n) ^{113}Sn reaction was compared with LANGE and MÜNZEL's semiempirical values. Position, shape and peak's half width agrees with these predictions, but the maximum of the experimental curve is higher than the predicted.
This divergency can possibly be explained because a proton magic number nucleus is built.
The yield of the conventional ^{112}Sn (n,γ) ^{113}Sn reactor production method and the studied accelerator yield were critically confronted and it was shown that the deuteron product will be prefered because higher relative yields and a carrier-free ^{113}Sn will be obtained.

Zusammenfassung

Querschnitte der In(d,xn) ^{113}Sn-Reaktion wurden experimentell bestimmt.
Bestrahlt wurden Folienpakete aus natürlichem Indium und Aluminium zwischen 27,5 MeV und der Coulombschwelle. Der integrale Deuteronenstrom wurde mit den Al-Monitor-Folien gemessen.
Die ^{113}Sn-Ausbeute wurde durch die Gamma-Strahlen der Tochter, ^{113}In, nach Einstellung des radioaktiven Gleichgewichts ermittelt. Der Gesamtfehler wurde zu etwa $\pm 20\%$ abgeschätzt.
Die Anregungsfunktion der ^{113}In (d,2n) ^{113}Sn-Reaktion wurde mit den halbempirischen Werten von LANGE und MÜNZEL verglichen. Form, Lage des Maximums und Halbwertsbreite sind etwa gleich, aber das Maximum der experimentellen Kurve ist vielfach größer.
Dieses Verhalten ist erklärbar, weil ein protonenmagischer Kern entsteht.
Die Ausbeute der konventionellen ^{112}Sn (n,γ) ^{113}Sn-Reaktorproduktionsmethode wurde mit der bei Deuteronenbestrahlung verglichen; es ergab sich, daß die Zyklotronbestrahlung vorzuziehen ist wegen der höheren relativen Ausbeute und weil ein trägerfreies Produkt entsteht.

Résumé

Les séctions efficases pour la réaction In(d,xn)^{113}Sn ont été mesurées.
Nous avons irradié des paquets contenants des couches d'indium et d'aluminium entre 27,5 MeV et le seuil coulombique. Le courant de deutons fut mesuré avec des moniteurs en aluminium. L'activité du ^{113}Sn a été mesurée par celle du ^{113}In à l'état d'équilibre radioactif. L'erreur est estimé à $\pm 20\%$.
La fonction d'excitation pour la réaction ^{113}In(d,2n)^{113}Sn a été calculé suivant la méthode de LANGE et MÜNZEL.
La position et la forme des maximum montrent un bon accord, mais les valeurs expérimentales des sections efficaces sont beaucoup plus élevées que celles qui résultent des calculs. Peut-être la différence s'explique par la formation d'un noyau à nombre de protons *magique*.
Les rendements de la méthode classique de production du ^{113}Sn par la réaction ^{112}Sn(n,γ)^{113}Sn et de l'irradiation de l'In par les deutons sont comparés. On trouve que la production dans le cyclotron est préférable à cause du rendement plus élevés et parce qu'on obtient un produit sans porteur.

Introduction

The reactions leading to ^{113}Sn are of actual interest because the main use of this radioisotope is at present the ^{113m}In generator which is applied in nuclear medicine. The daughter, ^{113m}In, desintegrates emitting only monoenergetic gamma radiation (see Fig. 1), easily detected with conventional rectilinear scanners. Also its half life (100 min) [1] and his chemical properties make it very suitable for marking molecules to be used in diagnostic and dynamic studies with gamma cameras.
The mother nuclide, ^{113}Sn, can be obtained through different nuclear reactions: in reactors by neutron irradiation of tin; in accelerators by irradiation of indium with protons or deuterons or by bombardement of cadmium with alpha particles.

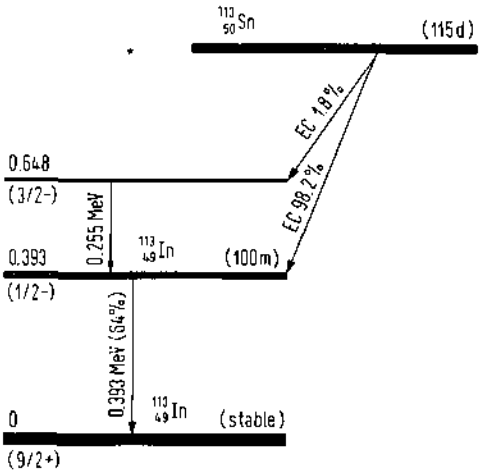

Fig. 1. Decay sheme and some nuclear properties from the Sn-In-113 chain (values taken from Ref. [1])

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1. C. M. LEDERER, J. M. HOLLANDER and I. PERLMAN, Table of Isotopes, 6th Edition, J. Wiley & Sons, Inc., New York 1968. — H. E. BOSCH, M. C. SIMON, E. SZIEHMAN, L. GATTO and S. M. ABECASSIS, Phys. Rev. 159, 1029 (1967).

Neutron irradiation of tin (generally enriched in ^{112}Sn) is however the conventional ^{113}Sn production method because it is apparently cheaper and easier.
The irradiation with charged particles offers the possibihty to produce carrier free ^{113}Sn. This property is desired for loading the mentioned generators, because they are based on absorption on inorganic ion exchangers. These have low capacity and, as it is necessary a high specific activity of the element that has to be fixed, carrier free will be the ideal condition.
The obtention of carrier free ^{113}Sn was mentioned in the literature, but excitation functions and irradiation yields were not given before [2].
The main purpose of this work was to measure the cross sections for the In(d,xn) ^{113}Sn nuclear reactions at different deuteron energies up to 27 MeV.
The knowledge of these values will be useful for the evaluation of an alternative ^{113}Sn production method and also will give some information concerning excitation function systematics and the factors affecting these, like the build up of a magic number nucleus.

Experimental Procedure

The cross sections for In(d,xn) ^{113}Sn reaction were experimentally determined by activation using the stacked foil technique.
The following nuclear reactions leading to ^{113}Sn should be expected when natural indium (4.28% ^{113}In and 95.72% ^{115}In) is irradiated with deuterons:

^{113}In (d,2n) ^{113}Sn $\quad Q = -\ 4.0$ MeV
^{115}In (d,4n) ^{113}Sn $\quad Q = -\ 20.3$ MeV.

The Q-values were calculated using MATTAUCH's mass tables [3]. The Coulomb threshold were calculated to be 7.2 MeV (FERMI's constant 1.6); but real threshold is some keV higher because recoil energy of the products and compound nucleus were not considered.
The reactions below threshold and those leading to isotopes other than ^{113}Sn were neglected in this work, including the isomer ^{113m}Sn ($T_{1/2} = 20$ min).

a) Target assembly

High purity indium (≥ 99.99) was rolled to get a uniform thickness between 40 and 110 mg · cm^{-2}; also high purity aluminium foils (≥ 99.999) were employed whose thickness was 2.7 to 5.4 mg/cm^{-2}.
Foils were cutted in 1×1 cm squares, degreased with solvents, washed with diluted hydrochloric acid, then with destilled water and finally dried in a desiccator.
Before assembling the targets each foil was identified and weighed. The foils were stacked in a holder and pressed together with a frontal thick copper colimator. This diaphragm has a circular opening (0.8 cm diameter), smaller than the deuteron beam, it ensures a centrally fixed beam spot on the foils.
The stacks were assembled as follows: the first foil of each target was a thin Al foil, then alternatively In and Al foils and the last one was a thick aluminium sheet (120 μm).

b) Irradiations

Targets were bombarded with the deuteron beam on the synchrocyclotron of the Atomic Energy Commission, at Buenos Aires, Argentina.
Four stacks were exposed to the 27,5 MeV external beam with different absorber thickness to overlap the experimental points.
Before each irradiation the beam focus was checked with glass plates. If necessary the focalisation was properly set adjusting the magnetic lenses, then the sample holder was situated in irradiation position, with the target normal to the inciding beam.
The integrated beam current was measured with the first Al foil, using the well known ^{27}Al(d,αp)^{24}Na excitation function [4].
Al foils stacked between the indium foils were used as absorbers to degrade the energy of the deuterons and collect the back escaping recoil atoms.
The last thick Al sheet was used to estimate the contribution of ^{27}Al(n,γ)^{24}Na reaction, induced by fast break-up neutrons. This foil was stacked well behind the range of deuterons with energies over the threshold of the (d,αp) reaction.
A current monitor was also connected to the holder to give a rough indication about flux constancy, variations during irradiations of 6 to 7 hrs were small enough to be neglected.
Beam intensity was around ten nanoampere and the total charge about 0.2 Coulomb at each stack.

c) Activity measurements

Twelve hours after the end of the irradiation each target was disassembled. The first and last Al monitor foils of each stack were counted immediately, using a calibrated 3×3 inch INa(Tl) scintillation cristal and a pulse heigh multichannel analyser for measuring the total ^{24}Na content.
From these results we calculated the integrated deuteron flux that incided on the target and the small break-up neutron flux that were built up on it. Gamma rays spectra of these foils showed that only ^{24}Na was present.
To calculate the cross sections for ^{113}Sn built up, through d,2n and d,4n reactions, we determined the ^{113m}In content in each In and Al foil. For these countings, done 1 month later, a high resolution Ge/Li detector and a multichannel spectrometer were used. No chemical separation was required and possible error sources thus avoided.

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2. S. W. BARNES, Phys. Rev. **56**, 414 (1969). — R. K. GORGIS and R. VON LIESHAUT, Physics **24**, 672 (1958). — K. D. COLEMAN and M. L. POOL, Phys. Rev. **72**, 1070 (1947).
3. J. H. E. MATTAUCH, W. THIELE and A. N. WAPSTRA, Nucl. Phys. **67**, 1 (1965).
4. U. MARTENS and G. W. SCHWEIMER, KFK Report 1083 (1969).

The ^{113m}In activity found in each Al catcher foil was added to that of the indium foil situated immediately before in the stack. The contribution was low and thus generally could be neglected.

In order to obtain the absolute ^{113}Sn activity some In foils were dissolved carefully and counted against standards.

The 2.75 MeV ^{24}Na and the 0.393 MeV ^{113m}In peaks were used respectively to determine the desired activity. The areas under the peaks in the spectra were obtained using COWELL's integration method.

Gamma spectra of the most active Al and In foils were taken at different times during some months to follow the decay and check the purity.

Results and Discussion

The total production cross sections (σ_E) for ^{113}Sn formation, through (d,2n) or (d,4n) reactions depending of the deuteron energy, were calculated applying the following modified activation formula:

$$\sigma_E = \frac{A_0\,^{113}\mathrm{Sn}}{\Phi_d\, N\, y\, \lambda\, t_i}\ [\mathrm{cm}^2]$$

$A_0\,^{113}$Sn: Tin-113 activity produced on the In plus Al catcher foils, corrected to the end of the irradiation (des/sec).
Φ_d : Integrated deuteron flux (d/sec) calculated from ^{24}Na content in the Al monitor foil.
N : Number of In atoms per unit area of the foil.
y : Isotopic abundance of the true target nucleus at given deuteron energy.
λ : Tin-113 decay constant = $2.51 \cdot 10^{-4}$ hrs^{-1}.

For deuterons with kinetic energy up to 20 MeV Indium-113 reacts giving Tin-113, hence the abundance used was $y = 4.28\,\%$. Over the threshold of the (d,4n) reaction the lighter isotope, ^{115}In, reacts giving ^{113}Sn and thus $y = 95.72\,\%$ was used for calculations.

The deuteron energy at the middle of each foil calculated by using WILLIAMSON, BOUJOT and RICHARD's range tables [5] and used as abscissa in Fig. 2. The horizontal bars at this figure are energy indetermination ($\leq$0.5 MeV) plus thickness of the foil where the particular cross section was determined. To obtain the range in indium, graphic interpolation between tin and cadmium range curves were taken. With the cross sections given at Table 1 the excitation function for the reactions In(d,xn)^{113}Sn was drawn (for natural Indium target), see Fig. 2. The vertical bars at this figure show the overall error for the measured cross sections, composed of square root of the number of counts, uncertainity of counting efficiency plus errors in the monitor cross section, half live of the products, foil thickness and all time measurements.

On Fig. 3 the excitation function for the ^{113}In(d,2n)^{113}Sn reaction was drawn. This curve exhibits the classical shape for reaction occurring through the mechanism of compound nucleus and evaporation of neutrons. A tentative curve is drawn (dashed) in the lower part of Fig. 3 using the characteristic values given by LANGE and MÜNZEL [6].

Shape and width at half height of the maximum of the (d,2n) excitation function agree with the semiempirical estimations given by these authors for this reaction, but the experimental curve is much higher.

This divergency could be explained because Indium (49 proton nucleus) captures protons or deuterons with higher efficiency than neighbor nuclei; the 50 proton shell will be fulfilled in this case and a magic compound nucleus results from this nuclear reaction.

LANGE and MÜNZEL remarked in his work that particular deviations from the general estimation method should be expected when a magic nucleus arises in the reaction. For the (d,4n) reaction we get unsufficient

Table 1. *Experimental determined cross sections for the reactions* In (d,xn) ^{113}Sn *as function of deuteron energy*

Deuteron energy (MeV)	Foil thickness (mg/cm^2)	Isotope abundance (%)	Measured cross section (milibarn)
Reaction: Indium (d,2n) ^{113}Sn			
7.5	111.70	4.28	292
8.8	49.64	4.28	463
10.9	43.80	4.28	966
12.9	53.29	4.28	1096
14.3	45.80	4.28	955
15.8	52.37	4.28	1012
16.2	51.11	4.28	944
17.3	51.10	4.28	973
18.7	51.10	4.28	962
19.2	111.69	4.28	681
Reaction: Indium (d,4n) ^{113}Sn			
21.2	47.45	95.72	38
21.8	101.47	95.72	45
22.3	53.29	95.72	27
23.6	47.45	95.72	24
24.2	114.61	95.72	29
24.6	43.80	95.72	28
25.6	119.64	95.72	35
26.3	114.61	95.72	99
26.5	113.88	95.72	128

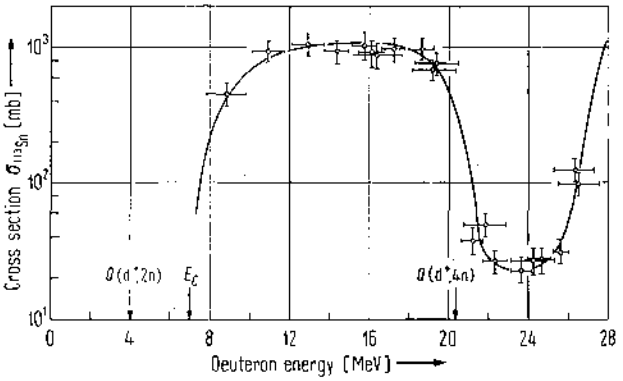

Fig. 2. Experimental cross sections for In (d,xn) ^{113}Sn reactions as function of the energy of the deuterons; target: natural Indium (^{113}In = 4.28%, ^{115}In = 95.72%)

5. C. F. WILLIAMSON, J. P. BOUJOT and J. PICARD, CEA Report 3042 (1966).
6. J. LANGE and H. MÜNZEL, KFK Report 767 (1968).

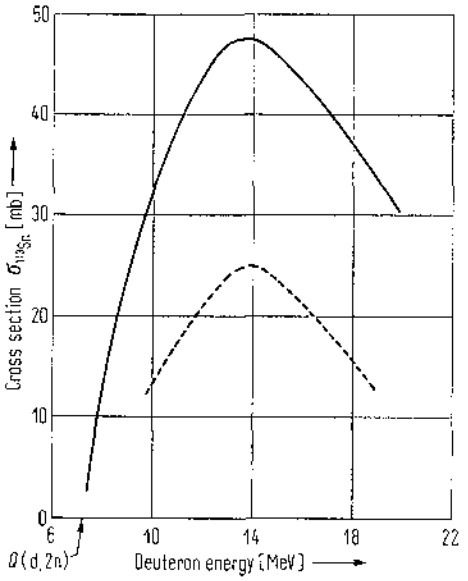

Fig. 3. Excitation function for ^{113}In (d,2n) ^{113}Sn reaction; target: ^{113}In. Dashed curve is the excitation function drawn with the semiempirical characteristic values taken from Ref. [6]

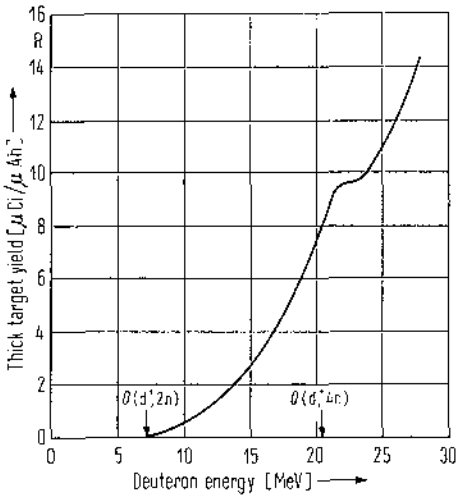

Fig. 4. Calculated thick target yield for an irradiation of indium targets, as function of the energy of the deuterons

experimental data and unfortunately there were no previously similar excitation functions published in the region of $Z = 50$; a general trend is not in sight to venture an extrapolation. Only a few data about (d,3n) reactions for $Z = 32$ [7] and for $Z = 65$ (d,4n) reactions excitation functions were mentioned [8] in the available literature.

The few values we have for the $^{116}In(d,4n)^{113}Sn$ reactions indicates however that an equal mechanism and proportional increase in the heigh of the excitation function curve should be expected using the same argument that before.

Thick target and reactor yields

Thick target yield for the production of ^{113}Sn by irradiation of metallic indium targets were calculated by numerical integration, as a function of the energy of inciding deuterons, using the cross sections measured in this work (up to 27 MeV).

The results are quoted in Table 2 and with these values Fig. 4 was drawn. The shoulder on the curve around 22–23 MeV represents the begining of the influence of the d,4n reaction.

A similar experience done at Hammersmith's Hospital Cyclotron [9] gives results that agree quite well with ours; in this experience a thick target yield of 2.1 μCi/μAh was found and a mean cross section of about 755 ± 18 mb for the 4 to 15.6 MeV deuteron range was calculated.

These yield figures are the highest production rate for a thick Indium target and are valid only for ideal conditions.

In later experiments, by internal beam irradiation we were not able to reproduce these yield values. Our explanations are that in this case firstly the beam is not so well focuses, secondly the internal target holder

7. A. Karpeles, Radiochim. Acta **12**, 212 (1968) and CNEA Report EN-14/29 (1970).
8. L. Nicola Siri and S. J. Nassiff, Radiochim. Acta **14**, 159 (1970).
9. S. Water, Private Communication, Hammersmith Hospital, Radiochemistry Section.

Table 2. *Thick target yield as function of energy of the deuterons for* In (d,xn) ^{113}Sn *reaction at different irradiation times*

Deuteron energy range [MeV]	Target thickness [mg/cm²]	Cross section [mbarn]	Saturation activity (mCi/μA]	Σ Saturation activity[a] [mCi/μA]	$\Sigma A_{T/10}$[b] [mCi/μA]	ΣA_{1h}[c] [μCi/μAh]
27.5–25.0	144	75	9.6	9.6	0.6	2.4
25.0–22.5	126	28	3.1	12.7	0.8	3.2
22.5–20.0	119	200	21.1	33.7	2.3	8.5
20.0–17.5	110	815	3.4	37.1	2.5	9.3
17.5–15.0	97	1090	4.0	41.1	2.8	10.3
15.0–12.5	88	1120	3.7	44.9	3.0	11.4
12.5–10.0	77	920	2.7	47.6	3.2	12.0
10.0– 7.5	66	420	1.0	48.6	3.3	12.3

[a] Long-thick-target-yield or saturation activity for 1 μA (in milicurie).
[b] Short-thick-target-yield for an 11.5 days irradiation (in milicurie).
[c] Thick-target-yield for an irradiation of 1 hr (in microcurie).

Note: Below 20.3 MeV only the (d,2n) reaction on the lighter stable indium isotope ($y = 4.28\%$) yields ^{113}Sn.

absorbs a considerable amount of the beam (incorrect high current measurement) and finally that the heat dissipation in internal targets are not good enough and some activity could be lost by evaporation of the target (indium's melting point 156°C; boiling point 2000°C [10]).

It is known that the heat dissipation for internal targets, rather than beam intensity, limits the commercial production rate of radioisotopes in accelerators and obligates the accurate experimental determination of the thick target yield in each individual case. Also special thight target holders must be designed to allow irradiations of indium and other low melting point substances in vacuum chambers.

Tin-113 can also be produced irradiating with thermal and epithermal neutrons; a low specific activity product results because the natural abundance of the ^{112}Sn isotope is noly 0.96% and the cross section for the ^{112}Sn (n,γ) ^{113}Sn reaction is 0.71 barn [11] (older published values fluctuate between 0.6 and 1.3 barn).

To increase the production rate generally the Sn-In-113m generator manufacturers irradiate expensive enriched ^{112}Sn targets, but charges grows proportionally to the enrichment.

Final Considerations

The present work demonstrates that the estimations of excitation functions obtained using the compilation of LANGE and MÜNZEL's semiempirical curves [6] gives to low values when the studied reaction lead to a magic number nucleus and that the experimental determination of the cross sections are compulsory in this case. A sistematic series of measurements of cross sections with deuterons with energies up to 50 MeV should be undertaken to get the complete (d,4n) reaction excitation function and also to obtain more theoretical information and data about yields of ^{113}Sn production for thick targets.

With our data it was calculated that the ^{113}Sn production yield with medium energy deuterons in a 10-hr irradiation of a thick indium target, at 100 μA, yields the same amount of activity as in 3,5 month reactor irradiation of 1 g natural tin at a flux of $1 \cdot 10^{13}$ n/cm^2 sec thermal neutrons.

Resuming we can say that the production of ^{113}Sn in a cyclotron can be justified because the reactor product has allways lower specific activity. We believe that in the future when the irradiation technology of thick targets with high current is managed, a competition between both methods will arise, if generator loading is in mind; proton or deuteron irradiation will be prefered because carrier free ^{113}Sn is obtained as accelerator product.

The author is indebted to the Cyclotron staff who performed the irradiations, to Mr. M. RIVERO who collaborated in the assembly of the targets and wishes to acknowledge the help of Prof. Dr. W. SEELMANN-EGGEBERT, Prof. Dr. H. MÜNZEL, Dra. S. J. NASSIFF and Dr. R. RADICELLA in discussing the results and for reading the manuscript.

10. K. SVOBODA, UJV Report 2258. Ch.
11. R. S. TILBURY and H. H. KRAMER, Nucl. Sci. Eng. **31**, 545 (1968).