

BNL--36530

BNL 36530

DE85 012263

CONF-850507--44

NUCLEAR DATA FOR PRODUCTION OF ^{117m}Sn FOR BIOMEDICAL APPLICATION*

L.F. MAUSNER and S. MIRZADEH

Medical Department, Brookhaven National Laboratory, Upton,
New York, U.S.A.

T.E. WARD

Indiana University Cyclotron Facility, Bloomington, Indiana,
U.S.A.

Abstract The $^{117m}\text{Sn}(4+)\text{DTPA}$ has been shown to have high uptake in cortical bone with negligible soft-tissue deposition. The long half-life (14 d) of ^{117m}Sn and its emission of abundant short-range Auger and conversion electrons, make $^{117m}\text{Sn}(4+)\text{DTPA}$ an attractive therapeutic agent for bone tumors. The necessary nuclear data for production of clinical quantities of ^{117m}Sn using $\text{Sb}(p,2pxn)$ reactions at BLIP was obtained. The excitation functions for $^{121}\text{Sb}(p,2p3n)$ and $^{123}\text{Sb}(p,2p5n)$ reactions in the regions of $43 < E_p < 60$ MeV were measured and compared with predicted values obtained from semi-empirical formulae given by Silberberg and Tsao. ^{117m}Sn is routinely produced at ORNL by thermal neutron activation of ^{116}Sn . We have investigated the possibility of improving the specific activity of the reactor-produced ^{117m}Sn by irradiating enriched ^{116}Sn and ^{117}Sn in the core of the HFBR. Our results indicate that, due to a strong absorption resonance at $E_n \sim 110$ eV, better specific activity for reactor-produced ^{117m}Sn can be obtained by irradiating natural tin in the core of a reactor.

INTRODUCTION

It has recently been shown^{1,2} that the non-phosphate $^{117m}\text{Sn}(4+)\text{DTPA}$ has high affinity to normal bone with little soft-tissue deposition. Since the uptake is primarily in cortical bone, the long physical half-life (14 d) and the abundance of short range Auger and conversion electrons make $^{117m}\text{Sn}(4+)\text{DTPA}$ an attractive therapeutic agent for bone tumors. Compared to ^{32}P phosphates (with high energy β 's) this tin radiopharmaceutical offers similar bone dose but significantly lower bone marrow and whole body exposures. However, the low specific activity of

MASTER

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

reactor produced ^{117m}Sn is a limitation in this application.

To this end, a study has been made of the feasibility of producing clinical quantities of no-carrier-added ^{117m}Sn by irradiating a natural antimony metal target with protons using $\text{Sb}(p,2pn)^{117m}\text{Sn}$ reactions at the Brookhaven Linac Isotope Producer (BLIP)³. Antimony is a good target in that adequate yields of both ^{117m}Sn and ^{118}Te , also of interest to our program, can be produced simultaneously.

We have also investigated methods of improving the specific activity of ^{117m}Sn attainable with reactors. In a series of irradiations at different core positions in the BNL High Flux Beam Reactor (HFBR) employing enriched $^{116},^{117},^{118}\text{Sn}$, the corresponding cross-sections for (n,γ) , $(n,n'\gamma)$, and $(n,2n)$ reactions were obtained, respectively.

EXPERIMENTAL

For excitation function measurements, thin targets of $^{121},^{123}\text{Sb}$ (typically $1-3 \mu\text{g}\cdot\text{cm}^{-2}$) were prepared by evaporating enriched metals (99%) onto $\sim 10 \text{ mg}\cdot\text{cm}^{-2}$ copper support discs. These copper-supported antimony samples were placed between copper cover foils to avoid recoil losses of tin isotopes. Then the samples were sandwiched between copper degrader foils ($276 \text{ mg}\cdot\text{cm}^{-2}$) and were irradiated for 3.5 h with 143.5 nA of $59.8 \pm 0.1 \text{ MeV}$ protons at the Indiana University Cyclotron Facility. After irradiation, the individual samples (antimony with copper backing) were dissolved in the presence of antimony and tellurium carriers. Tellurium was isolated by reduction to metal by SO_2 , then tin was separated by retention on anion-exchange resin from 2 M HCl , and subsequently was eluted with 3 M HClO_4 . Samples for non-destructive neutron activations were prepared by encapsulating 2-10 mg of $^{116},^{117},^{118}\text{Sn}$ (as SnO_2 with enrichments of 95.60, 84.23, and 97.07%, respectively) in low-Na quartz ampules. These samples together with flux monitoring samples were irradiated for an hour in three different core positions of the HFBR. The fast neutron flux was monitored using the following standards: $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$, $^{58}\text{Ni}(n,p)^{58}\text{Co}$, and $^{208}\text{Tl}(n,2n)^{202}\text{Tl}$ with cross-sections of 0.725 ± 0.045 , 113 ± 7 ,⁴ and $4.00 \pm 0.32 \text{ mb}$,⁵ respectively. The *reaction* $^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$ with $\sigma = 4.1 \text{ b}$ was used to monitor the thermal flux.

RESULTS AND DISCUSSION

In order to select optimal irradiation conditions at the BLIP, the determination of relevant thin target excitation functions from 22-75 MeV on ^{121}Sb and ^{123}Sb are underway. Data from 43-60 MeV are complete and show the $^{121}\text{Sb}(p,2p3n)^{117m}\text{Sn}$ reaction cross-sections increasing from 2.1 mb to 5.2 mb in this energy

NUCLEAR DATA FOR PRODUCTION OF ^{117m}Sn FOR BIOMEDICAL APPLICATION

range, while the $^{123}\text{Sb}(p,2p5n)^{117m}\text{Sn}$ reaction cross-sections are essentially constant at 5 mb.

These measurements together with the theoretical calculations of excitation functions are shown in Figure 1. The calculations were performed using a modified version of code SPALL⁶, employing semi-empirical formulae given by Silberberg and Tsao for peripheral interactions⁷. Our experience with Silberberg's formulae indicates that the standard deviation between predicted and experimental values of cross-sections is about 50% at 100 MeV and improves at higher energies. However, at energies below 100 MeV the standard deviation increases rapidly and is a factor of 2 or 3 at 20-50 MeV, as indicated in Figure 1.

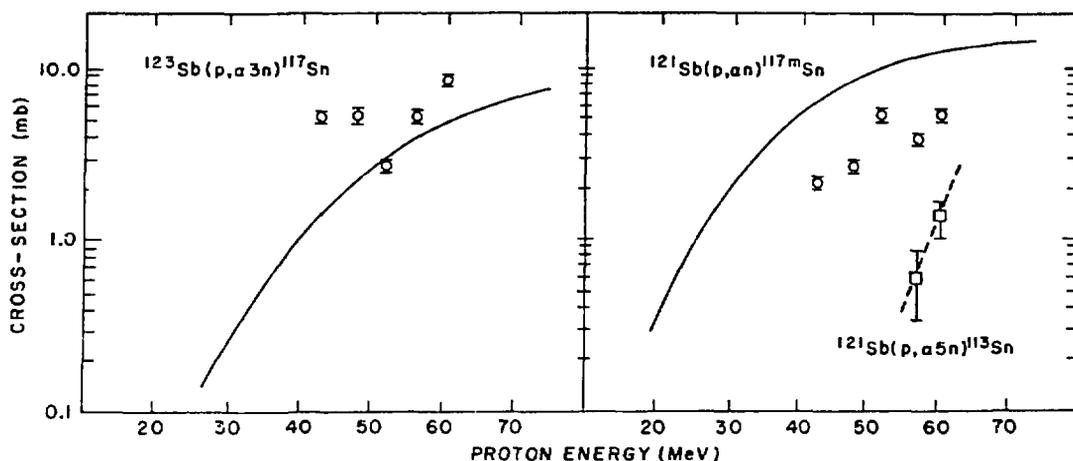


Figure 1. Excitation Functions; Points are experimental values of cross-sections and lines are predicted ones.

Several preliminary thick target bombardments at nonoptimal energies were performed. A saturation yield of $0.61 \text{ mCi} \cdot \mu\text{A}^{-1} \cdot \text{g}^{-1}$ was obtained in the region of $38 \leq E_p \leq 55 \text{ MeV}$, which is 80% of the theoretical value (based on our cross-sections). At this energy range for a 14-day irradiation, the fraction of ^{113}Sn is about 0.02. Although the yield of ^{117m}Sn is somewhat higher at 190 MeV, $1.1 \text{ mCi} \cdot \mu\text{A}^{-1} \cdot \text{g}^{-1}$ at saturation, there is an unacceptable amount (36%) of ^{113}Sn produced. The specific activity is wholly dependent on the tin impurity in the antimony target and is calculated to be $30 \text{ mCi} \cdot \mu\text{g}^{-1}$ for each ppm of tin impurity in the target.

Table 1 summarizes the data on a series of irradiations at different core positions in the HFBR on enriched $^{116}, ^{117}, ^{118}\text{Sn}$. Each value represents the mean of 2-6 determinations. The uncertainty of the listed cross-sections is +15%. This includes uncertainties due to counting statistics (~1%) detector

efficiencies (~5%), and cross-sections of the standards (~10%). Effective cross-sections of 166 and 195 mb were obtained for the $^{116}\text{Sn}(n,\gamma)^{117\text{m}}\text{Sn}$ and $^{117}\text{Sn}(n,n'\gamma)^{117\text{m}}\text{Sn}$ reactions, respectively. The significant increase in the cross-section of (n,γ) reaction is due to a strong neutron absorption resonance at $E_n \sim 110$ eV. The highest specific activity of $^{117\text{m}}\text{Sn}$, 8.0 mCi/mg, at saturation, is obtained by irradiating ^{117}Sn at position V-15 in the HFBR. This value is a factor of two higher than $^{117\text{m}}\text{Sn}$ available from ORNL, despite a thermal neutron flux only 0.08 times that available at ORNL.

An accurate value of the $^{117\text{m}}\text{Sn}$ half-life was also obtained by carefully following the decay of 14 samples of $^{117\text{m}}\text{Sn}$ from three separate runs. The weighted average $t_{1/2}$ obtained for the 158.6 keV γ -ray (86.4%) is 14.01 ± 0.03 d which is in good agreement with the previous value of 14.0 ± 0.3 d.

Table 1. Effective Cross-Sections for Production of $^{117\text{m}}\text{Sn}$ with Fission Neutrons.

Position in the Reactor	Neutron Flux		Cross-section, mb		
	Thermal	Fast. >1MeV	^{116}Sn (n, γ)	^{117}Sn (n,n' γ)	^{118}Sn (n,2n)
Reflector. V-11	1.5×10^{14}	9.0×10^{10}	5.4	14 ⁵	NI
Core-edge. V-14	8.25×10^{14}	6.4×10^{13}	21	16 ⁵	NI
In-core. V-15	1.95×10^{14}	3.0×10^{14}	166	195	9.7×10^{-2}

REFERENCE:

1. S. C. Srivastava, et al., Proc. Third World Congr. on Nucl. Med. Biol., edited by C. Raynaud, Vol. II (Pergamon Press, Paris, 1982), p. 1635.
2. S. C. Srivastava, et al., Int. J. Appl. Med. Biol. (in press).
3. L.F. Mausner and P. Richards, IEEE Trans. Nucl. Sci., **NS-30**, 1793 (1983).
4. A. Calamand, "Cross-Sections for Fission Neutron Spectrum Induced Reactions," in Handbook on Nuclear Activation Cross-Sections, Technical Report Series No. 156, IAEA, Vienna (1974).
5. G. J. J. Gerbino, I. M. Cohen, R. O. Korob and G. B. Baro, J. Inorg. Nucl. Chem., **38**, 1387 (1976).
6. J. T. Routti and J. V. Sandberg, Comput. Phys. Commun., **23**, 411 (1981).
7. R. Silberberg and C. H. Tsao, Ap. J. Suppl., **25**, 315 (1973).

*Supported under U.S. Department of Energy Cont. DE-AC02-76CH0016.