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Production and Radiochemistry of Antimony-120m: Efforts Toward Auger Electron Therapy with ^{119}Sb

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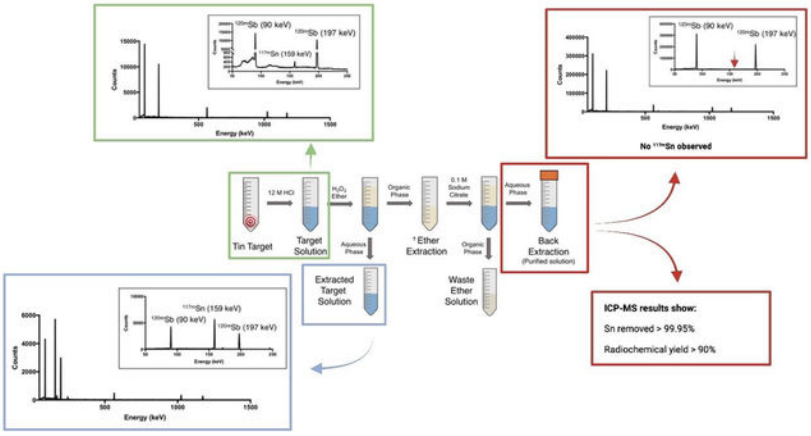
Abstract

Targeted Meitner-Auger Therapy (TMAT) has potential for personalized treatment thanks to its subcellular dosimetric selectivity, which is distinct from the dosimetry of β^- and α particle emission based Targeted Radionuclide Therapy (TRT). To date, most clinical and preclinical TMAT studies have used commercially available radionuclides. These studies showed promising results despite using radionuclides with theoretically suboptimal photon to electron ratios, decay kinetics, and electron emission spectra. Studies using radionuclides whose decay characteristics are considered more optimal are therefore important for evaluation of the full potential of Meitner-Auger therapy; ^{119}Sb is among the best such candidates. In the present work, we develop radiochemical purification of ^{120}Sb from irradiated natural tin targets for TMAT studies with ^{119}Sb.

Graphical Abstract:

[+]contributed equally to this work



Keywords

Antimony-119; Targeted Meitner-Auger Therapy; natural tin; proton irradiation; liquid-liquid extraction

1. Introduction

Targeted radionuclide therapy (TRT) has garnered attention in basic research and commercial sectors, accelerating the search for radionuclides with optimal therapeutic decay properties. TRT uses selective delivery systems (e.g. peptides, antibodies or other bio vectors) in combination with therapeutic radionuclides (β^-, α and Meitner-Auger electron emitters) for personalized and localized treatment [1,2]. While in recent years β^- and α emitters have gained much clinical popularity, notably ^{177}Lu and ^{225}Ac respectively, Meitner-Auger electron (MAE) emitters have been often overlooked as therapeutic candidates despite potential to offer the most selective treatment on the subcellular level [3–5].

MAE decay follows capture (EC) or isomeric transition. The energy released by this de-excitation of a higher electron energy state filling an inner electron shell vacancy is reinvested into the emission of an electron from the same atom, causing an additional electron vacancy. The process repeats, causing an electron cascade [3]. The kinetic energy of each ejected electron corresponds to the difference in energy between the energy released during electron shell vacancy filling and the ionization energy of the electron. Similarly, conversion electrons are emitted through an analogous process (called internal conversion) during nuclear de-excitations following radioactive decay. As the source of energy for electron emission is the (relatively small) difference in electron/nucleon shell energies, resultant kinetic energy for MAE's and CE's range from tens of eV to tens of keV [6] compared to β^- (0.1–2.2 MeV) and α emission (5–8 MeV). While the low energy makes it unlikely that membrane targeted MAEs will reach target cell DNA, MAE therapy does not suffer from dose range effects [7]. Due to high particle energy and low linear energy transfer (LET), β^- therapy is plagued with dose range effects, resulting in energy absorption within a large area (0.5–10 mm) surrounding decay location [8]. Alpha-emitting radionuclides have

a much shorter particle range (40–100 μm), with the high particle energy accompanied by very high LET. Unfortunately, large-scale production of α emitters is unestablished and requires reactors or high-energy cyclotrons for production, which are uncommon and in high demand. Available generators can produce only limited quantities and often require decades to produce [9–12]. The advantage of MAE therapy over β^- and α therapy is not only production capacity but also particle range and formation of multiple electrons (cascade) per decay. MAEs typically have electron ranges less than the length of one cell (<10 μm) [8], potentially enabling them to treat single cell metastases.

Current literature in MAE therapy primarily discusses application of commercially available candidates such as ^{111}In, ^{67}Ga, ^{125}I, and ^{99m}Tc, which have clinical precedence. While these γ-emitters do undergo MAE emission, their other properties (e.g. other emissions or half-life) may limit their application for TMAT. Despite these properties, studies yield encouraging therapeutic results, demonstrating antitumor efficacy and induction of double strand DNA breaks [13–17].

Antimony-119 (^{119}Sb, $t_{1/2}$ = 38.19 h, EC = 100%) is one of the most potent radionuclides for TMAT [3,18,19]. The half-life of ^{119}Sb is convenient for radiopharmaceutical preparation and administration. Decaying to stable ^{119}Sn, ^{119}Sb releases a high number (24) of Meitner-Auger and conversion (<50 keV) electrons, and importantly, the decay has a low photon-to-electron energy ratio of 0.9, expanding the therapeutic window [3].

It was shown previously that production of clinically relevant quantities of ^{119}Sb is efficiently possible with low-energy cyclotrons via ^{119}Sn(p,n)^{119}Sb [21,22]. Despite being a promising TMAT candidate, few ^{119}Sb research reports have focused on production, purification, and complexation [18]. Separation of a desired radionuclide from bulk target material often involves exploitation of chemical differences between the two species and can be achieved through a wide variety of techniques including solid-phase chromatography, liquid-liquid extraction, precipitation, complexation, and thermal diffusion, among others [23–27]. While each approach has advantages and downfalls, a robust separation technique requires reproducibility. Radioantimony/tin separations reported in the literature highlight consistency challenges. Sadeghi *et al.* report a technique [28] based on silica-gel column chromatography with HCl eluent; however, not only does this presumably lead to a relatively high concentration of silica in the final solution [29], but the technique is not reproducible [22]. Another study reported a more concrete method based on previously established anion-exchange chromatography [30]. Despite the successful separation, reported chromatograms are drastically different from those of the original paper, perhaps as a result of different anion-exchange solid phases [21,22]. It was also noted that timing between target dissolution and breakthrough have a “critical impact on the success of the separation” with increased time increasing tin breakthrough, likely as a result of changing of oxidation states or hydrolysis [21,22]. These consistency issues motivate development of a reliable separation method. Given historic success with liquid-liquid extraction [31,32], this method was selected for further study.

In the present work, we report irradiation of ^{nat}Sn to produce a broad array of antimony radioisotopes to study radiochemical separation. Anticipating biological use, liquid-liquid

extraction was developed to purify radioantimony from bulk tin target material. Furthering this separation technique can be adopted for production of isotopically pure ^{119}Sb by irradiation of enriched ^{119}Sn.

2. Materials and Methods

2.1 General

All solvents, reagents, and resins were purchased from commercial suppliers (Sigma-Aldrich, Fisher Scientific, TCI America, Alfa Aesar, AK Scientific, Fluka) and used as received. Water used was ultrapure (18.2 MΩ cm^{-1} at 298 K, Milli-Q, Millipore, Billerica, MA). Irradiations were performed using the University of Wisconsin-Madison GE PET trace (Madison, Wisconsin, USA) and the TR13 cyclotron at TRIUMF (Vancouver, Canada). At the University of Wisconsin-Madison, high purity germanium (HPGe) gamma spectrometry was conducted using an aluminum-windowed detector (Ortec, Knoxville, Tennessee) with Canberra (Concord, Ontario) Model 2025 research amplifier and multichannel analyzer. This system was energy and efficiency calibrated using ^{241}Am, ^{133}Ba, ^{152}Eu, ^{137}Cs, and ^{60}Co sources (Amersham PLC, Little Chalfont, UK). The full width at half maximum resolution is 1.8 keV at 1333 keV. At TRIUMF, an N-type co-axial HPGe gamma spectrometer from Canberra was fitted with a 0.5 mm beryllium window as previously described [33]. Detector energy, width, and efficiency calibrations were performed using a ^{152}Eu and ^{133}Ba source. Inductively coupled plasma mass spectrometry (ICP-MS) analysis was conducted at TRIUMF using an Agilent 8900 Triple Quadrupole ICP-MS. All measurements were acquired in helium mode.

Caution!—Antimony and Sn radioisotopes are produced by proton irradiation of ^{nat}Sn targets as listed in Table 1. All radionuclides are gamma emitters. Proper radiation safety procedures and shielding were implemented when handling these radionuclides in laboratories approved for radioactive material.

2.2 Irradiation of Tin Target

Natural tin targets (3.0 cm diameter, 0.127 mm thick, acquired from Sigma-Aldrich) were irradiated with protons (entrance energy 16 MeV for GE PETtrace and 12.8 MeV for TR13) for 1 h with a current of 5 μA.

The foil was water cooled (GE PET trace) or cooled by a double helium-cooling window configuration (TR13) with one helium jet cooling the 25 μm thick aluminum foil separating the target from the cyclotron vacuum, and the other one the front of the tin target. Irradiated targets were left in the cyclotron target area for 2 hours following irradiation. The target was then removed from the cyclotron and transported to the shielded fume hood for further radiochemical processing. The foil was trimmed to remove non-irradiated portions and reduce tin mass.

2.3 Liquid-Liquid Extraction

Target foils (between 400–480 mg after trimming) were dissolved in 12 mL of HCl (12 M) and allowed to stand in a covered (not sealed) 16 mL borosilicate KIMAX tube at room

temperature overnight. 4 mL of the target solution was set aside, while 8 mL was transferred into a 50 mL Falcon centrifuge tube. A small volume of HCl (12 M, 800 μL) was used to rinse the KIMAX tube prior to adding 8 mL of the target solution. H_2O_2 (30% w/w, contains inhibitor, 800 μL) was added to the target solution to oxidize Sb(III) and Sn(II) to Sb(V) and Sn(IV). The solution was allowed to sit for approximately 30 minutes. Dibutyl ether (10 mL) was added to the target solution (8.8 mL), and the biphasic solution vortexed for 15 min. After the solution settled, phases were separated by pipetting organic phase from the Falcon centrifuge tube into a new 50 mL Falcon tube. Of the initial 10 mL dibutyl ether, 9 mL were collected to ensure no aqueous contamination. Next, two washing steps of the organic phase were conducted by adding 9 mL of HCl (10 M) to the dibutyl ether and vortexing the solution for 10 min before separating as previously described. For the first wash, no dibutyl ether was sacrificed (i.e., 9 mL were recovered upon separation). After the second wash, 1 mL of dibutyl ether was sacrificed, leaving 8 mL of dibutyl ether solution. Finally, back-extraction was completed by adding 8 mL of sodium citrate solution (0.1 M, pH 5.5) to the organic phase and vortexing for 30 min. Phases were separated as described above, obtaining 7 mL of citrate solution.

Gamma spectroscopy on six samples was carried out after separation (target solution, extracted target solution, HCl wash #1, HCl wash #2, extracted ether solution and final back-extracted solution). One milliliter of each solution was added to separate 2 mL glass vials to ensure counting geometry was compatible with calibration. Solutions were measured with gamma spectrometer positioned 15 cm away from detector perpendicularly in the measurement chamber, and counts were acquired until activity error reached below 10%.

ICP-MS analysis of six non-irradiated tin targets was performed on samples collected throughout the liquid-liquid extraction process. Targets were dissolved and treated as described above, without setting 4 mL of the target solution aside, starting with 12 mL of the target solution, reserving one milliliter of the target solution after each separation step, resulting in 9 mL of the citrate solution recovered. The samples were dried down overnight and converted to a 2% nitric acid matrix for analysis.

3. Results and Discussion

3.1 Target Irradiation

Though ideal for targeted radionuclide therapy application, the low energy photon emissions of ^{119}Sb are difficult to quantify via standard HPGe detector γ-spectroscopy. From a financial perspective, radionuclidically pure ^{119}Sb via (p,n) nuclear reaction requires expensive isotopically enriched ^{119}Sn (~8.6% natural abundance—necessitating material recovery beyond the scope of this work). Because of these reasons, reported experiments used radionuclides produced via proton bombardment of a natural tin target (Figure 1) which is ideally suited for developing radiochemical procedure and chelation.

According to SRIM calculations, the beam deposits 1.7 MeV into the foil (for TR13). The 5 μA beam increases the temperature of the target to no more than 160°C, far away from the tin melting point of 232°C. Due to the diverse isotopic makeup of natural tin and their moderate (p,x) cross-sections for low energy protons,[28] several radionuclides are produced

(Table 1). Following a suitable cooling period (>28 h), ^{120m}Sb ($t_{1/2}$ 5.76 d) and ^{117m}Sn ($t_{1/2}$ 14.0 d) activity remains to be used as tracers. Decay corrected to EOB for 12.8 MeV incident beam energy, (1.42 ± 0.08) MBq of ^{120m}Sb and (28.5 ± 1.1) kBq of ^{117m}Sn were produced (n = 3), and were slightly higher than TENDL calculation – 1.11 MBq of ^{120m}Sb and 25.0 kBq of ^{117m}Sn [35]. The presence of these two radiotracers allows monitoring of radiochemical separation using non-destructive γ-spectroscopy.

3.2 Purification of ^{120m}Sb by Liquid-Liquid Extraction

Figure 2 illustrates the technique used to separate radioantimony from bulk tin target material. It is crucial that H_2O_2 is added to the target solution shorty before performing the separation (as opposed to during target dissolution) otherwise the yield will be drastically reduced. We hypothesize this is necessary to ensure all antimony is oxidized to the pentavalent oxidation state. Once this has occurred, [^{120m}Sb]Sb(V) can be selectively extracted from the aqueous solution with dibutyl ether - only 10 minutes of vortexing is required for (near) quantitative extraction. In strong HCl, the species present in solution are $[SnCl_6]^{2-}$ and $[SbCl_6]^-$; thus selective uptake of the latter into ether is likely a result of more diffuse charge distribution, resulting in a more hydrophobic species that prefers organic solvation [36]. Following separation of phases, two washing steps with 10 M HCl (not shown in diagram) help remove minute amounts of Sn(IV) that may have partitioned into the organic phase. Lastly, since radiolabeling is most desirably performed from aqueous solution, the Sb(V) is back-extracted into a 0.1 M sodium citrate solution, whose pH (5.5) is essential. The citrate is not only required for successful back-extraction but also mitigates Sb(V) hydrolysis, as is done for Sb(III) [37]. The success of this procedure is quantified in Table 2 and can be seen in Figure 3, where γ-ray spectra of the target solution, extracted target solution and back extracted solution are shown. The target solution most notably shows ^{120m}Sb ($E\gamma$ = 90 keV, 197 keV) and ^{117m}Sn ($E\gamma$ = 159 keV) with activities 1.42 MBq and 28.5 kBq, respectively. The extracted target solution has retained only a small amount of ^{120m}Sb (2.4%) and vast majority of ^{117m}Sn (>95%), as can be qualitatively noted by the relative intensities of peaks. Finally, the back extracted solution contains no observable ^{117m}Sn. The elemental concentration of stable tin in the final solution was determined to be lower than 18 ppm (<170 μg in total), showing that more than 99.95% of the bulk tin target is removed in the separation process. Net yield of ^{120m}Sb in the final solution (relative to target solution) was (69 ± 2) %. When correcting for sacrificed volume, the yield is >90% (Table 2). The amount of stable antimony present in the final solution was determined to be <40 μg. Experiments negating sacrificial volume were conducted providing radiochemical yield (94.6 ± 4.0) % (N=3) with ^{117m}Sn activity below limit of detection 480 Bq, which sets an upper limit of <0.1% original target Sn mass remaining in final solution.

Conclusions

A natural tin target was irradiated with 12.8 MeV or 16 MeV protons to produce a variety of antimony radioisotopes. Following a two-day waiting period, the primary radionuclides of interest were ^{120m}Sb and ^{117m}Sn. A radiochemical purification method to separate radioantimony from tin using liquid-liquid extraction was developed and provided >90% radiochemical yield (corrected for sacrified volume) with separation factor of >1700.

Proposed separation method can be further applied for separation of ^{119}Sb from proton irradiated ^{119}Sn target.

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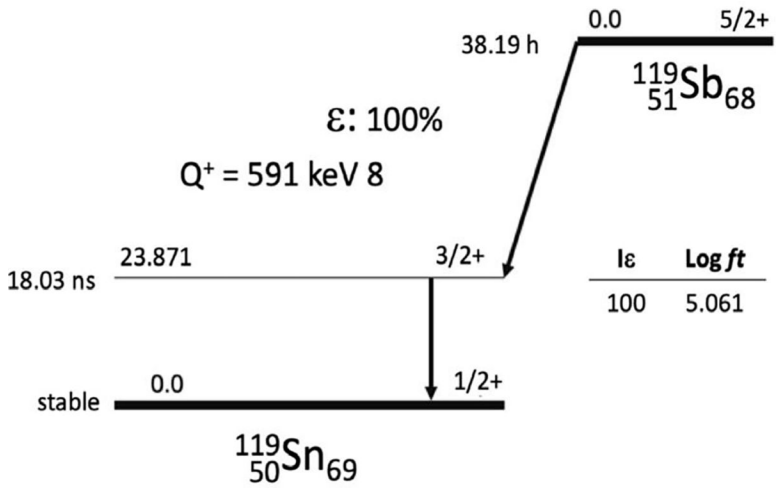

Figure 1.
Decay scheme of ^{119}Sb [20].

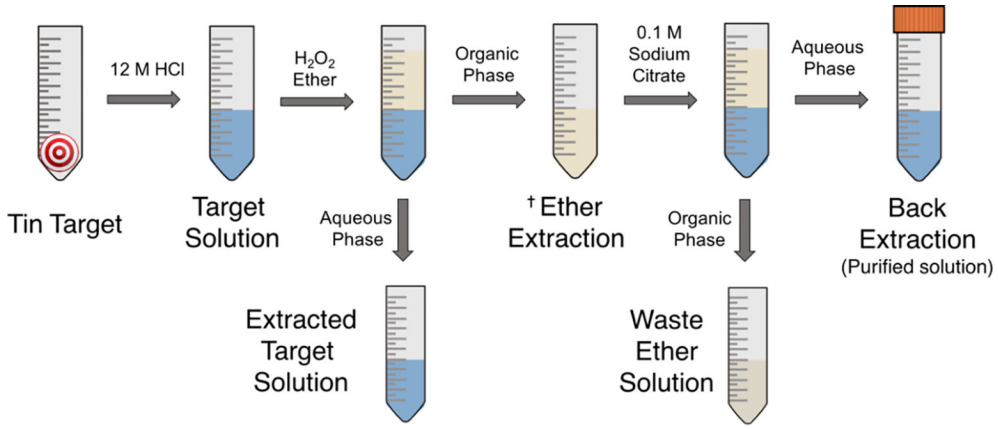

Figure 2.

Visual representation of radioantimony purification.†Wash ether with equal volume 10 M HCl (x2) prior to back extraction.

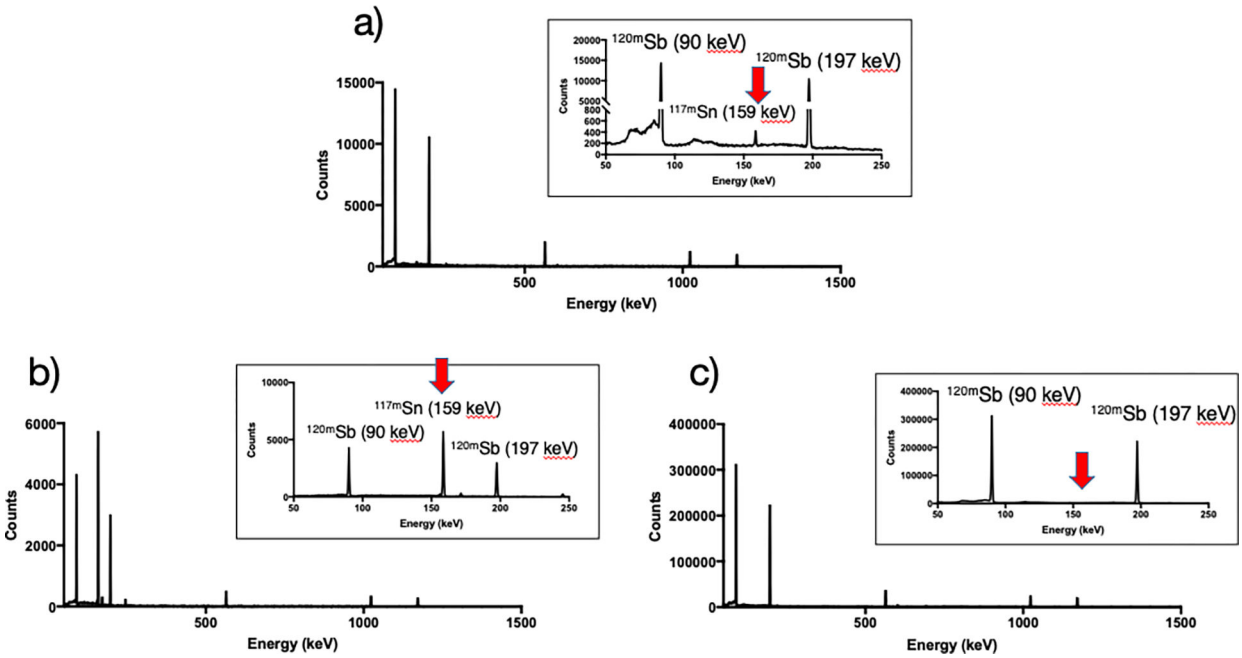

Figure 3.

γ-ray spectra of a) target solution b) extracted target solution c) back extracted solution.

Table 1.

Abundance of natural tin isotopes and their produced (p, n) and (p, p*) radionuclides.

Isotope	Natural Abundance (%)[a]	Radionuclide Produced via proton induced nuclear reactions
^{117}Sn	7.68	^{117m}Sn ($t_{1/2}$ = 14.0 d)
^{118}Sn	24.22	^{118}Sb ($t_{1/2}$ = 3.6 min)
^{119}Sn	8.59	^{119}Sb ($t_{1/2}$ = 38.2 h)
^{120}Sn	32.58	^{120m}Sb ($t_{1/2}$ = 5.7 d)
^{122}Sn	4.63	^{122}Sb ($t_{1/2}$ = 2.7 d)

[a]Ref [34].

Table 2.

Activities of ^{120m}Sb and ^{117m}Sn expressed as a fraction over total target solution activity.[a]

Solution	%^{120m}Sb Activity (100/initial activity)	% ^{117m}Sn Activity (100/initial activity)
Extracted Target Solution	2.4% ± 0.4%	98% ± 4%
HCl Wash #1	0.38% ± 0.07	N.D
HCl Wash #2	0.28 ± 0.05	N.D
Extracted Ether	0.07%[b]	N.D
Final Citrate Solution (net total)[c]	69% ± 2%	N.D
Final Citrate Solution (corrected)[d]	95% ± 2%	N.D

[a]Values calculated using activity concentration (Bq/mL) and volume, without correcting for sacrificed volume unless otherwise specified. Activity then divided by initial activity of purified target solution. Reported error is standard deviation (n = 3). N.D. = not detected;

[b]Due to N.D. in two trials, n = 1.

[c]Percent isolated activity over starting activity;

[d]Percent activity corrected for sacrificed volume over starting activity.