^{122}Sb – A POTENTIAL RADIOTRACER: EVALUATION OF CYCLOTRON PRODUCTION VIA NOVEL ROUTES

by

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Antimony-122, having a half-life of 2.723 d and I_β = 97.59%, is an important radiotracer in studies of environmental contamination and food crops. For the work discussed in this paper, the production of ^{122}Sb was done via the ^{nat}Sn(p, xn)^{122}Sb nuclear reaction. Radiochemical separation was performed by silica gel column chromatography and liquid-liquid extraction methods. Excitation functions for the ^{122}Sb radionuclide, via ^{122}Sn(p, n)^{122}Sb, ^{nat}Sn(p, xn)^{122}Sb, ^{122}Sn(d, 2n)^{122}Sb, ^{nat}Sn(d, xn)^{122}Sb, ^{124}Sn(p, 3n)^{122}Sb and ^{124}Sn(d, 4n)^{122}Sb reactions, were calculated by ALICE/91, ALICE/ASH, and TALYS-1.2 codes and compared with existing data.

Key words: nuclear data, production, ALICE/ASH, silica gel, antimony-122

INTRODUCTION

Natural tin is an important component of several widely used alloys. For studying wear behavior of components containing Sn by thin layer activation (TLA), induced antimony activities can be used. In that case, the preferred isotopes would be ^{124}Sb ($T_{1/2}$ = 60.2 d) and ^{122}Sb ($T_{1/2}$ = 2.72 d) [1]. Radionuclide ^{122}Sb (I_β = = 97.59%) was used for the studies of environmental contamination and food crops [1-3]. The ^{122}Sb reaction has one long-lived ground state radionuclide, ^{122g}Sb ($T_{1/2}$ = 2.73 d), and two short-lived metastable states, ^{122m}Sb ($T_{1/2}$ = 5.3 10^{-4} s and $T_{1/2}$ = 4.19 min). The long-lived ground state of ^{122g}Sb was measured after the complete decay of the two short-lived metastable states to ^{122}Sb by a 100% internal transition process. Therefore, the production cross-section of the ^{122}Sb isotope is considered as the cumulative production of $^{122g+m}$Sb. An intense, independent γ-ray of 564.2 keV was used to determine the production cross-sections of ^{122}Sb [1-2].

Nuclear data play an important role in the choice of a radioisotope for various applications. Data concerning nuclear structure and decay determine the suitability of a radioisotope for possible applications in medicine and agriculture, while nuclear reaction data apply to the possibility of producing a radionuclide in pure form. The production of non-conventional radionuclides, however, demands detailed nuclear data research covering both experimental investigations and nuclear model calculations. Radioisotopes produced by charged-particle nuclear reactions can be widely used in medicine and agriculture [4-6].

There are four methods for the production of ^{122}Sb via cyclotron:
(1) ^{122}Sn(p, n)^{122}Sb, (2) ^{124}Sn(p, 3n)^{122}Sb,
(3) ^{122}Sn(d, 2n)^{122}Sb, and (4) ^{124}Sn(d, 4n)^{122}Sb [7-10].

In this work, ^{122}Sb was produced via proton bombardment of natural tin as a target. Also, the excitation functions of ^{122}Sb using ALICE/91, ALICE/ASH, and TALYS-1.2 codes for various reactions were calculated and compared with previously published experimental data.

MATERIALS AND METHODS

Calculation of the excitation function

Briefly, the ALICE/ASH code is a modified and advanced version of the ALICE code [11]. The geometry-dependent hybrid (GDH) model is used for the description of the pre-equilibrium particle emission from nuclei [12]. The TALYS-1.2 code (equilibrium and pre-equilibrium) is optimized for incident projectile energies ranging from 1 keV up to 200 MeV on target nuclei, with mass numbers between 12 and 339. It

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includes photon, neutron, proton, deuteron, triton, ^{3}He, and α-particles, as well as projectiles and ejectiles and single-particle and multi-particle emissions and fission [13]. Equilibrium and pre-equilibrium particle emissions during the decay process of a compound nucleus are very important for a better understanding of the nuclear reaction mechanism induced by medium energy particles. The highly excited nuclear system produced by charged particles' first decays by emitting fast nucleons at the pre-equilibrium (PE) stage and, later on, by the emission of low-energy nucleons at the equilibrium (EQ) stage.

Calculation methods of the equilibrium reaction

Equilibrium emission is calculated according to the Weisskopf-Ewing (WE) model [14], neglecting the angular momentum. During evaporation, the basic parameters are binding energies, inverse reaction cross-section, pairing and level density parameters. The reaction cross-section for incident channel a and exit channel b can be written as

$$\sigma_{ab}^{WE} \quad \sigma_{ab}(E_{inc}) \frac{\quad b}{\quad b} \quad b \qquad (1)$$

where E_{inc} is the incident energy,
$_b \quad (2s_b \quad 1)/\pi^2\hbar^2\mu_b \; d\varepsilon\sigma_b^{inv}(\varepsilon)\varepsilon\omega_1(U)/\omega_1(E)$,
U – the excitation energy of the residual nucleus, μ_b – the reduced mass, and s_b – the spin and the total single-particle level density is taken as

$$\omega_1(E) \quad \frac{1}{\sqrt{48}} \frac{\exp[2\sqrt{\alpha(E \quad D)}]}{E \quad D}; a \quad \frac{6}{\pi^2} g \qquad (2)$$

where σ_b^{inv} is the inverse reaction cross-section, E – the excitation energy of the compound nucleus, D – the pairing energy, and g – the single particle level density [15].

Basic calculation methods of pre-equilibrium reactions

The excitation functions for pre-equilibrium calculations were calculated using exciton models, a two-component exciton model, one-component exciton model, and a photon exciton model. Pre-equilibrium processes cover a sizable part of the reaction cross-section for incident energies between 10 to (at least) 200 MeV. Pre-equilibrium reactions have been modeled both classically and quantum mechanically and are included in TALYS [12].

GDH model and hybrid model

In calculations of the hybrid and GDH model, code ALICE/ASH was used. This code can be applied for the calculation of excitation functions, energy, and angular distribution of secondary particles in nuclear reactions induced by nucleons and nuclei with energy up to 300 MeV. A generalized superfluid [16] has been applied for nuclear level density calculations in the ALICE/ASH code. ALICE/91 and ALICE/ASH codes use the initial exciton number as n_0 = = 3. But, in these models, different alpha (α), deuteron (d) and proton (p) exciton numbers are used in pre-equilibrium GDH model calculations. The other code model parameters can be found in detail in reference [13]. In the ALICE/ASH code, the hybrid and GDH models for pre-compound emissions and the Weisskopf-Ewing model for compound reactions are selected [15].

Possible routes to ^{122}Sb

According to fig. 1 [17], four routes can be used to produce ^{122}Sb with conventional cyclotrons with deuteron or proton beams:
(1) ^{122}Sn(p, n)^{122}Sb, (2) ^{124}Sn(p, 3n)^{122}Sb,
(3) ^{122}Sn(d, 2n)^{122}Sb and (4) ^{124}Sn(d, 4n)^{122}Sb.

Despite the lower natural abundance of ^{122}Sn compared to ^{124}Sn, the ^{122}Sn(p, n)^{122}Sb nuclear reaction appears to be the most suitable route for the production of ^{122}Sb. To summarize the strengths of this reaction [9]:

- the most common type of cyclotron may be used,
- low stopping power of the incident particles high production yields,
- low bombardment energy less demanding target cooling requirements,
- reasonably high natural abundance of the target material lower cost,
- radionuclidic impurities can be avoided, and
- high specific activities can be reached.

Te122 0+ 2.603	Te123 10^{13} year ½ + EC 0.908	Te124 0+ 4.816	Te125 ½+ 7.139	Te126 0+ 18.95
Sb121 5/2+ 57.36	Sb122 2.7238 d 2– EC, β–	Sb123 7/2+ 42.64	Sb124 60.20 d 3– β–	Sb125 2.76 year 7/2+ β–
Sn120 0+ 32.59	Sn121 27.06 h 3/2+ β–	Sn122 0+ 4.63	Sn123 129.2 d 11/2– β–	Sn124 0+ 5.79
In119 2.4 m 9/2+ β–	In120 3.08 s 1+ β–	In121 23.1 s 9/2+ β–	In122 2.5 s 1+ β–	In123 5.98 s 9/2+ β–

Figure 1. The nuclide region around ^{122}Sb. Several nuclear reaction routes are possiible to form the ^{122}Sb-radioisotope (□→ naturally aboundant, ■→emitters of β^-, and electron capture, □ →emitters of β^-)

RESULTS AND DISCUSSION

Excitation function

The excitation functions of ^{122}Sn(p, n)^{122}Sb, ^{124}Sn(p, 3n)^{122}Sb, ^{122}Sn(d, 2n)^{122}Sb, ^{124}Sn(d, 4n)^{122}Sb, ^{nat}Sn(p, xn)^{122}Sb, and ^{nat}Sn(d, xn)^{122}Sb reactions were calculated using ALICE/91, ALICE/ASH, and TALYS-1.2 codes and compared to existing data [18-20]. To increase the accuracy of the calculations, the codes were used simultaneously

^{122}Sn(p, n)^{122}Sb and ^{124}Sn(p, 3n)^{122}Sb reactions

Theoretical antimony production cross-sections for ^{122}Sn(p, n)^{122}Sb, and ^{122}Sn(d, 2n)^{122}Sb reactions have been illustrated in fig. 2. There is no experimental data for these reactions in literature, therefore only theoretical calculations are shown in fig. 2. There is a relatively good agreement between the prediction of the excitation functions made by ALICE/91, ALICE/ASH and TALYS-1.2 codes. The evaluation of the acquired data has shown that the best ranges of energy for ^{122}Sn(p, n)^{122}Sb and ^{124}Sn(p, 3n)^{122}Sb reactions are 19 8 and 13 7 MeV, respectively.

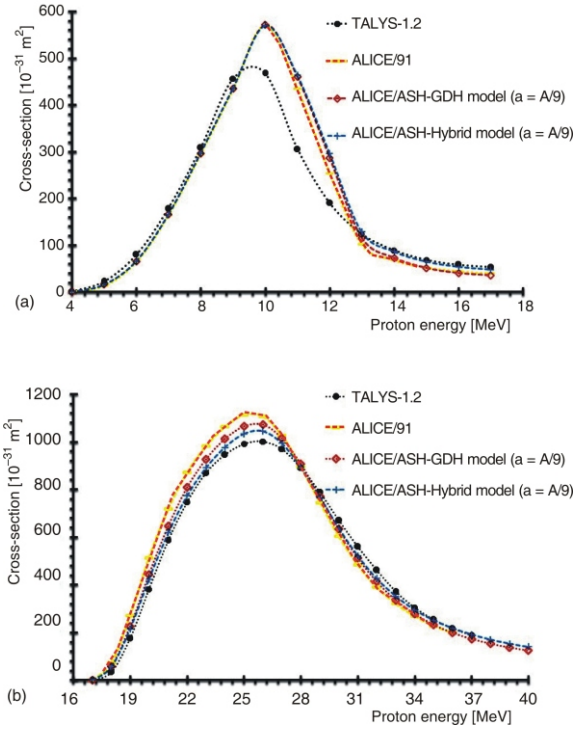

Figure 2. Excitation functions of (a) ^{122}Sn (p, n)^{122}Sb, and (b) ^{124}Sn(p, 3n)^{122}Sb reactions by ALICE/91, ALICE/ASH, and TALYS-1.2 codes

^{122}Sn(d, 2n)^{122}Sb and ^{124}Sn(d, 4n)^{122}Sb reactions

^{122}Sn(d, 2n)^{122}Sb and ^{124}Sn(d, 4n)^{122}Sb reactions are used to produce ^{122}Sb. Figure 3 shows a comparison between the calculated cross-sections with TALYS-1.2, ALICE/ASH, and ALICE/91 codes. The best range of incident energy for ^{122}Sn(d, 2n)^{122}Sb and ^{124}Sn(d, 4n)^{122}Sb reactions was assumed to be 18 9 and 40 27 MeV, respectively. According to the TALYS-1.2 code, the maximum cross-section for a ^{122}Sn(d, 2n)^{122}Sb reaction is 687.943 10^{-31} m^2 at E_d = 13 MeV.

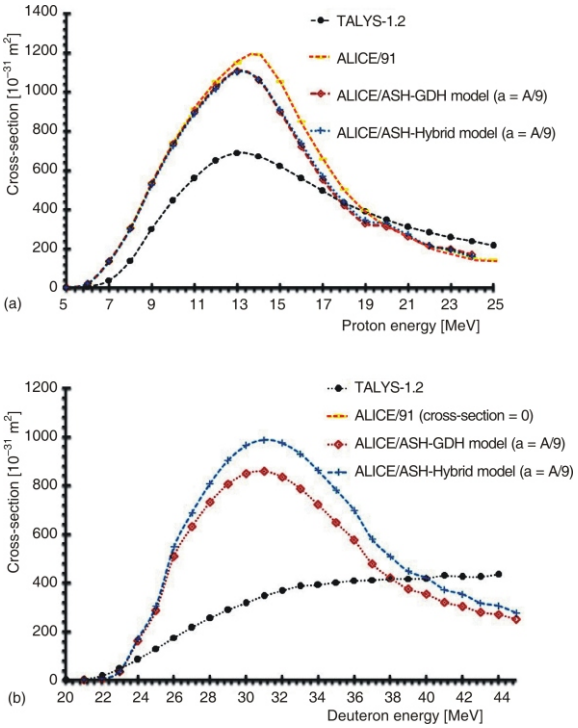

Figure 3. Excitation functions of (a) ^{122}Sn (d, 2n)^{122}Sb, and (b) ^{124}Sn (d, 4n)^{122}Sb reactions by ALICE/91, ALICE/ASH, and TALYS-1.2 codes

^{nat}Sn(p, xn)^{122}Sb and ^{nat}Sn(d, xn)^{122}Sb reactions

Khandaker *et al*. (2009) [2] used stacks of a high purity metallic form of tin with a thickness of 50 5 μm to measure independent and cumulative cross-sections of radioisotopes produced as a function of incident proton energy in the range of 6 up to 42 MeV. Copper and aluminum – ^{nat}Cu(p, x)^{62}Zn, ^{27}Al(p, x)^{24}Na, ^{27}Al(p, x)^{22}Na – were used as monitors to measure the beams' intensity and degradation of beam energy, while high purity germanium was used for detection. Also, using the stacked-foil activation technique, Hermanne *et al*. (2006) [1] were determined cross-sections of proton induced reactions on natural Sn up to 67 MeV. In this

study, we considered the same process as described by Khandaker *et al*. [2] and Hermanne *et al*. [1], with nuclear model calculations (TALYS-1.2, ALICE/91, and ALICE/ASH codes). In general terms, the three codes validate experimental data well (see fig. 4a). So far, there is no experimental data in literature for the ^{nat}Sn(d, xn)^{122}Sb reaction, thus nuclear model calculations could play an important role in finding the maximum excitation function of ^{nat}Sn(d, xn)^{122}Sb reaction (fig. 4b).

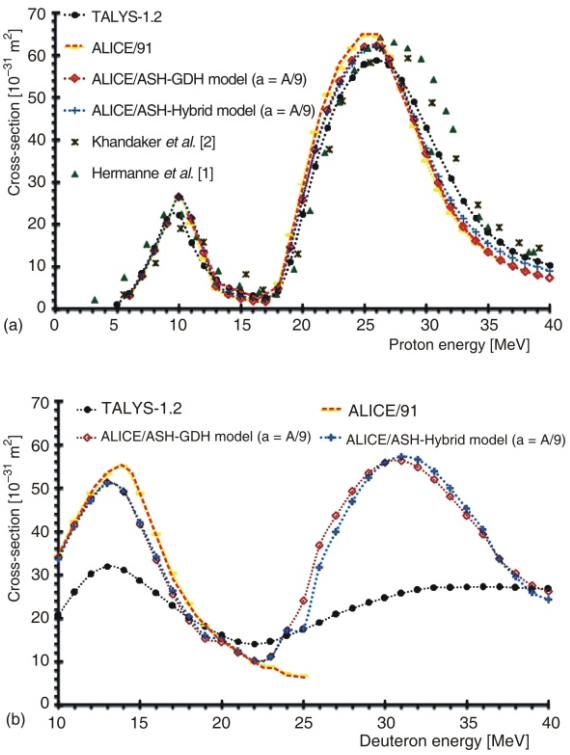

Figure 4. Excitation functions of (a) ^{nat}Sn(p, xn)^{122}Sb, and (b) ^{nat}Sn(d, xn)^{122}Sb reactions by experimental data, ALICE/91, ALICE/ASH, and TALYS-1.2 codes

Cyclotron production

The required thickness of the target was calculated according to the SRIM (The stopping and range of ions in matter) code [21]. The physical thickness of the target layer is chosen in such a way that for a given beam/target angle geometry (90°), the incident beam is excited from the target layer with predicted energy. To minimize the thickness of the target layer, 6° geometry is preferred (so that the required layer thickness is lesser for a coefficient of 0.1), in which case a 17.14 μm tin layer is recommended. Using the electrodeposition method [22], natural tin (Merck, 99.9%) containing ^{112}Sn (0.97%), ^{114}Sn (0.65%), ^{115}Sn (0.34%), ^{116}Sn (14.53%), ^{117}Sn (7.68%), ^{118}Sn (24.23%), ^{119}Sn (8.59%), ^{120}Sn (32.59%), ^{122}Sn (4.63%), and ^{124}Sn (5.79%) isotopes, formed a metallic layer on the copper backing over an area of 20.5 cm^2. Electrodeposition experiments were carried out using potassium stannate trihydrate ($K_2Sn(OH)_6$, 115 g/L, Aldrich 99.9%) and potassium hydroxide (KOH, 20 g/L). Optimal conditions for the electrodeposition of tin were as follows: 40 g/L ^{nat}Sn, a temperature of 75 °C and DC current density of 50 mA/cm^2 [8]. Due to the formation of a tin-copper junction with a strong metal-metal interaction, heat produced during the bombardment is effectively transferred to the copper backing and, finally, to the cooling water running at the back of the target.

The prepared target was irradiated with 180 μA proton beams of 26.5 MeV for 20 min, in a Cyclone30 machine (installed at the Agriculture, Medical and Industrial Research School, Tehran), at a glancing angle of 6 degrees, so as to achieve a higher production yield. At this angle, the effective thickness of the target is about 10 times its actual thickness. After the irradiation, the target was dissolved with 15 mL of concentrated HCl containing 5 mL of H_2O_2. Solvent extraction and ion exchange chromatography proved to be the best methods for the separation of antimony radionuclides from Sn target solutions [23-28].

Liquid–liquid extraction method

After the dissolution of the irradiated ^{nat}Sn in conc. HCl and H_2O_2, the obtained solution was evaporated to near-dryness and the residue dissolved in concentrated HCl (30 mL) (fig. 5). Then, 10 mL of organic solution, *n*-butyl ether ($C_8H_{18}O$), and aqueous solution containing tin, antimony, copper, and zinc were placed in a 100 mL separatory funnel, shaken for 15 min and allowed to settle for further 30 min. Following this, the solution was shaken for another 15 min (6 times) [26]. The radioantimony extracted into the organic phase and tin, copper, and zinc remained in an aqueous phase. The organic phase was then transferred into the separatory funnel and washed with a 40 mL concentration of HCl, with an acid-to-organic solvent volume ratio of 2:3. For this purpose, the mixture was stirred and allowed to stand for

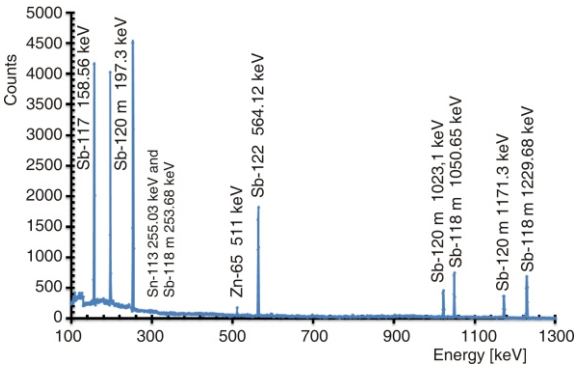

Figure 5. Non-destructive γ-ray spectrum of proton irradiated ^{nat}Sn taken 24 hours after end of bombardment

16-18 hours. Under this condition, Sb(V) back-extracted completely into the aqueous phase. The concentrations of Zn and Cu in the solution after separation were determined by polarography and colorimetric assays. The amount of Cu and Zn were found to be 5 ppm and 0.2 ppm, respectively. Standard copper concentrations were complexed by dithizone forming a pinkish complex [29]. About 90 5% of antimony radionuclides were extracted. The productions yield was ^{122}Sb 3.61 MBq/μAh. After separation, the obtained antimony purity was above 99% [8].

Silica gel column chromatography method

A column of a size of Ø1.0 × 10 cm was used, packed with silica gel (100-200 mesh, 99%, Aldrich). According to the methods of Khalid *et al*. [25] and Thisgaard *et al*. [27, 28], it is possible to separate Sb from the bulk of the Sn target material by pretreating the silica gel with 6 M HCl for 24 hours prior to the separation. The pretreated silica gel washed off and packed on the column and conditioned with 1 M HCl. The dissolved target solution (12 M HCl) diluted to 1 M HCl with deionized water and eluted through the column, followed by the washing of the column with 30 mL 1 M HCl, so as to remove any traces of Sn. Subsequently, the Sb-fraction eluted with 20 mL 6 M HCl (eluent temperature: 70-80°). Using silica gel chromatography, antimony radionuclides separated in a 80 5% radiochemical yield.

CONCLUSIONS

In this paper, possible production routes to the ^{122}Sb isotope (^{122}Sn(p, n)^{122}Sb, ^{nat}Sn(p, xn)^{122}Sb, ^{122}Sn(d, 2n)^{122}Sb, ^{nat}Sn(d, xn)^{122}Sb, ^{124}Sn(p, 3n)^{122}Sb, and ^{124}Sn(d, 4n)^{122}Sb reactions) have been discussed and the development of a production procedure for the most suitable route using a low energy cyclotron described. A reasonable agreement with experimental and theoretical excitation functions was obtained. Radionuclide ^{122}Sb was produced by the irradiation of electroplated tin natural on a copper substrate, using an alkali solution (potassium stannate trihydrate and potassium hydroxide). The solvent extraction of no-carrier-added ^{122}Sb from the irradiated tin natural target hydrochloric solution was investigated using di-n-butyl ether ($C_8H_{18}O$). Also, radioantimony separated in a 80 5% radiochemical yield was obtained using silica gel column chromatography.

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Махди САДЕГИ, Милад ЕНФЕРАДИ

ПРОИЗВОДЊА У ЦИКЛОТРОНУ НА НОВ НАЧИН

Антимон-122 са временом полураспада од 2.723 дана и $I_{\beta-}$ = 97.59%, значајан је радиообележивач коришћен за проучавање загађења животне средине и прехрамбених житарица. У овом раду разматрана је производња ^{122}Sb остварена нуклеарном реакцијом ^{nat}Sn(p, xn)^{122}Sb. Радиохемијска сепарација изведена је помоћу силицијумдиоксидне гел хроматографије и течно-течних екстракционих метода. Екстракционе функције ^{122}Sb радионуклида, добијених посредством ^{122}Sn(p, n)^{122}Sb, ^{nat}Sn(p, xn)^{122}Sb, ^{122}Sn(d, 2n)^{122}Sb, ^{nat}Sb(d, xn)^{122}Sb, ^{124}Sn(p, 3n)^{122}Sb и ^{124}Sn(d, 4n)^{122}Sb реакција, рачунате су програмима ALICE/91, ALICE/ASH и TALYS-1.2 и упоређене са постојећим подацима.

Кључне речи: нуклеарни подаци, циклотронска производња, ALICE/ASH програм, силицијумдиоксидни гел, ^{122}Sb