ACCELERATOR PRODUCTION AND NUCLEAR ASPECTS OF ^{88}Y: AN EFFICIENT RADIOTRACER

by

Milad ENFERADI [1*], ***Mahdi SADEGHI*** [2], ***and Fatemeh BOLOURINOVIN*** [2]

[1]Department of Nuclear Engineering, Science and Research Branch, Islamic Azad University, Tehran, Iran
[2]Agricultural, Medical & Industrial Research School, Nuclear Science and Technology Research Institute, Karaj, Tehran, Iran

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Yttrium-88 ($T_{1/2}$ = 106.6 d, $I_{\beta+}$ = 0.2% and I_{EC} = 99.8%) is used in mixed gamma efficiency calibration standards and also as a substitute for ^{90}Y to quantify the biodistribution of Y-pharmaceuticals in animals. Yttrium-88 and strontium-85 as gamma emitting radiotracers were produced via $^{nat}SrCO_3$(p, xn) and ^{nat}RbCl(p, xn) nuclear processes at AMIRS, with 18 and 15 MeV protons, respectively, at a current of 20 μA for 10 hours. The deposition of $^{nat}SrCO_3$ on the Cu backing was carried out by means of the sedimentation method. Yttrium-88 was separated in a 92 5% radiochemical yield using the precipitation technique by precipitating ^{nat}Sr as $SrSO_4$ (strontium-85 as a tracer) while radioyttrium passed through the filter paper. Also, a theoretical study of the nuclear reaction cross-sections for proton and deuteron induced reactions on ^{nat}Sr and ^{nat}Rb for the production of ^{88}Y and ^{85}Sr, respectively, was performed using the EMPIRE (version 3.1 Rivoli), TALYS-1.26 codes and the TENDL-2010 database.

Key words: accelerator production, ^{88}Y, ^{85}Sr, precipitation technique, EMPIRE

INTRODUCTION

Yttrium belongs to group IIIB of the periodic table. It has an atomic radius of 2.3 Å (1 Å = 10^{-10} m) and its electronegativity is 1.22 [1]. Several isotopes of yttrium can be produced in the cyclotron, such as: ^{86}Y ($T_{1/2}$ = 14.7 h, $I_{\beta+}$ = 34%, $E_{\beta+max}$ = 1.2 MeV), ^{87}Y ($T_{1/2}$ = = 79.8 h, I_{EC} = 99.8%, E_γ = 388.5 keV –82.1% and E_γ = = 484.8 keV –89.7%, and ^{88}Y ($T_{1/2}$ = 106.6 d, $I_{\beta+}$ = = 0.2%, I_{EC} = 99.8%, E_γ = 898 keV –92.7%, and E_γ = 1836 keV –99.4%), using a variety of nuclear reactions with particle energies between 5 and 70 MeV [1-4]. The longer-lived ^{88}Y is used in mixed gamma-efficiency calibration standards and also as a substitute for the chemical yield determination of ^{90}Y [1, 4]. Its mode of decay is predominantly by means of electron capture and the decay emissions include strong γ-rays [2, 5].

In cyclotron production of radionuclides, reaction cross-section data play an important role. One needs the full excitation function of the nuclear process to be able to calculate the yield with reasonable accuracy. Another important point is the number of competing reaction channels [6].

There are three principal routes of producing ^{88}Y in cyclotrons: (1) the bombardment of strontium with protons or deuterons, *i. e*., ^{88}Sr(p, n) or ^{88}Sr(d, 2n)-reactions [2, 4], (2) α- or ^{3}He-induced nuclear reactions on rubidium, *i. e*., the ^{87}Rb(^{3}He, 2n)^{88}Y, and ^{85}Rb(^{4}He, n)^{88}Y processes [7, 8], and (3) the ^{88}Zr/^{88}Y generator, *i. e*., the ^{89}Y(p, 2n)^{88}Zr ^{88}Y, ^{89}Y(d, 3n)^{88}Zr ^{88}Y, ^{nat}Mo(p, x)^{88}Zr ^{88}Y, and ^{nat}Sr(α, xn)^{88}Zr ^{88}Y reactions [9-12].

Several methods, such as co-precipitation [13],the electrochemical approach [14, 15], ion exchange chromatography [2, 16, 17], filtration [18], and liquid-liquid extraction [2, 19, 20], have been used in an attempt to separate radioyttrium from Sr-based targets. Purification methods using ion-exchange resin, the electrochemical approach and co-precipitation incorporate difficult and time consuming steps. The maximum reported yield for Y purification has been around 90% [13, 15].

The aim of this work was to produce ^{88}Y by coating $^{nat}SrCO_3$ via the sedimentation method, on a pure copper backing, with sufficient stability at high-power beam bombardment. Also, a much simpler separation technique was developed to achieve higher yields than via the previously reported methods. Accordingly, radioyttrium was separated from $^{nat}SrCO_3$ using a

* Corresponding author; e-mail: m.nferadi@srbiau.ac.ir

novel precipitation process in which $SrSO_4$ was precipitated and filtered.

MATERIALS AND METHODS

Excitation function

In brief, TALYS-1.26 is the latest version of the TALYS code that simulates nuclear reactions involving gammas, neutrons, protons, deuterons, tritons, ^{3}He, and α-particles in the incident energy range of 1 keV to 200 MeV for target nuclides with a mass 12 and above [21]. TENDL (TALYS-based evaluated nuclear data library) is a nuclear data library which provides the output of the TALYS nuclear model code system for direct use in both basic physics and applications. TALYS is a computer code system for the analysis and prediction of nuclear reactions [22]. The third version, TENDL-2010, is based on both default and adjusted TALYS calculations and data from other sources [21]. The EMPIRE (version 3.1 Rivoli), the latest version of the EMPIRE code, is a modular system of nuclear reaction codes comprised of various nuclear models and designed for calculations over a broad range of energies and incident particles. The system can be used for theoretical investigations of nuclear reactions, as well as for nuclear data evaluation. A projectile can be a photon or a nucleon, light or heavy ion. The energy range starts just above the resonance region when neutron projectiles are concerned, extending up to a few hundred MeV heavy ion-induced reactions [23]. The code accounts for major nuclear reaction models, such as the optical one, Coupled Channels and DWBA (ECIS06), Coupled Channels' Soft-Rotator (OPTMAN), Multi-step Direct (ORION+TRISTAN), NVWY Multi-step Compound, exciton model (PCROSS and DEGAS), hybrid Monte Carlo simulation (DDHMS), as well as for the full featured Hauser–Feshbach model, including the optical model for fission. The heavy ion fusion cross-section can be calculated within this simplified coupled channels approach (CCFUS) [23].

Calculation of required target thickness and theoretical yield

To obtain optimum physical dimensions of the target such as thickness, some estimation on the basis of the SRIM code (the stopping and range of ions in matter) was made [24, 25]. The recommended thickness of the target is 927 m for 90º geometry. To minimize the thickness of the carbonate strontium layer and increase heat transfer, a 6º geometry is preferred, in which case a 96.89 m $SrCO_3$ layer is recommended.

Nuclear reaction cross-section data are needed in radioisotope production programs mainly for the optimization of production routes, *i. e.*, to maximize the yield of the desired product and minimize the yields of radioactive impurities. From a given excitation function, the expected yield of the product for a certain energy range, *i. e.*, target thickness, can be calculated using the expression [6]

$$Y \quad \frac{N_L H}{M} I(1 \quad e^{\lambda t}) \int_{E_1}^{E_2} \frac{dE}{d(\rho x)} \quad ^{1} \sigma(E)\, dE \quad (1)$$

where Y is the activity (in Bq) of the product, N_L – the Avogadro number, H – the enrichment (or isotopic abundance) of the target nuclide, M – the mass number of the target element, I – the projectile current, $dE/d(\rho x)$, the stopping power, $\sigma(E)$ – the cross-section at energy E, λ – the decay constant of the product, and t – the time of irradiation. The calculated yield value represents the maximum yield that can be expected from a given nuclear process. It should be pointed out that the non-isotopic impurities produced can be removed by chemical separations, whereas the level of isotopic impurities can be suppressed only by using enriched isotopes as target material and/or by a careful selection of the particle energy range effective in the target [6, 20].

Target preparation of $^{nat}SrCO_3$

A $^{nat}SrCO_3$ (Aldrich, 99.995%) thick layer was deposited on the elliptical copper backing (11.69 cm^2 surface area) by means of the sedimentation method [26]. To prepare the target, a particular home-made system consisting of two plates (19 10^3 cm^3) made of teflon was constructed. The upper plate contains an elliptical window of 11.69 cm^2. The copper backing is fixed between these two plates; the upper part is sealed by an O-ring fitted window. The window's geometrical shape determines the actual target coating area. In this method, ethyl cellulose ($C_{20}H_{38}O_{11}$, Aldrich) was added to strontium carbonate. Then a suspension mixed in water-free acetone was obtained by stirring. The $SrCO_3$ suspension solution was stirred for 3-4 minutes and immediately upon that loaded into the cylinder of the upper disk. The window was covered by a teflon plate with a small hole. At room temperature, the solution (acetone) evaporated slowly through the hole after approximately 24 hours [20, 27]. Optimum conditions of target preparation were obtained through several repeated experiments with different amounts of ethyl cellulose and acetone. The best amount of ethyl cellulose is that of approximately 35% of strontium carbonate. A concentration of 52.4 mg/mL strontium carbonate in acetone solution is optimized.

Cyclotron production of ^{85}Sr for tracer studies

In order to develop the separation of ^{88}Y from either $^{nat}SrCO_3$, it was necessary to add ^{85}Sr as a tracer to

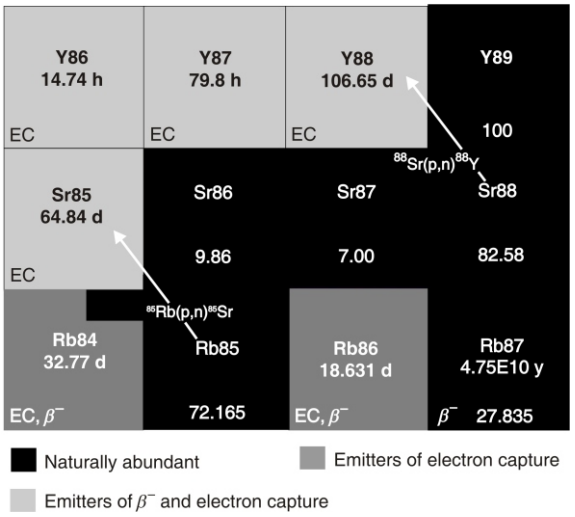

Figure 1. Possible routes of cyclotron production of ^{88}Y and ^{85}Sr in this work

the Sr target. According to fig. 1 [28], the best production routes of ^{88}Y and ^{85}Sr via proton beams are ^{88}Sr(p, n)^{88}Y and ^{85}Rb(p, n)^{85}Sr, respectively.

In order to produce strontium-85, the deposition of natural rubidium chloride on a copper backing was carried out via the sedimentation method. 520 mg RbCl (Aldrich, 99.99%), 208 mg ethyl cellulose (EC) and 4 mL acetone were used to prepare a layer of enriched rubidium chloride over a 11.69 cm^2 area and with a thickness of 62.2 mg/cm^2 [29]. The coated natural RbCl was introduced into the target holder and bombarded at the AMIRS (Agricultural, Medical and Industrial Research School) with 15 MeV protons at a current of 20 μA for 10 hours.

According to the method of Banerjee *et al*. [30], it is possible to separate Y from the bulk of the Sr target material by solvent extraction, using 0.1 M $HClO_4$ and 0.004 M 18-crown-6 in nitrobenzene. The irradiated natural rubidium chloride-deposited target on the copper backing was dissolved with a liquid flow-through a stripper in an acetonic system with 100 mL acetone. The obtained solution was evaporated to near dryness and the residue dissolved in concentrated HCl (10 mL). Then, the obtained solution was filtrated with Whatman-41 filter paper in order to remove the ethyl cellulose (paste). In the solvent extraction experiment, a filtrate solution was used for loading. 5 mL of organic solution – 0.004 M 18-crown-6 (Aldrich, 99.5%) in nitrobenzene and 30 mL of aqueous perchloric acid ($HClO_4$) solution containing $SrCl_2$ and $RbCl_2$ were placed in a 100 mL separatory funnel, shaken for 15 minutes, and allowed to settle for 30 minutes. The rubidium extracted into the organic phase and strontium remained in the aqueous phase (washes 6 times, 5 6 mL). The characteristic γ-ray used for the identification of ^{85}Sr ($T_{1/2}$ = 64.9 d) was 514 keV.

Irradiation and Y/Sr radiochemical separation

The coated natural $SrCO_3$ was introduced into the target holder and bombarded with 18 MeV protons at a current of 20 μA for 10 hours. AMIRS operate a Cyclone-30 (IBA, Belgium). To protect the target material from reaching excessively high temperatures during irradiation, a jet of cooling water flows across the back of the copper backing in direct contact with it, so that the heat is removed efficiently from the backing. In order to improve thermal conductivity, the copper backing is indented with grooves at the back. No direct cooling is applied over the front of the deposited target [20, 31].

Sulfuric acid readily forms a solid sulfate with Sr during the chemical reaction ($SrCl_2(aq) + H_2SO_4(aq) = SrSO_4(s) + 2HCl(aq)$, $K_{ps}(SrSO_4) = 3.44\ 10^{-7}$), constituting a reliable precipitating agent for separating yttrium from strontium. Sulfuric acid was gradually added to the $YCl_2/SrCl_2$ solution under heating and stirring conditions, so as to increase the formation of Sr precipitation. Afterwards, the solution/precipitation was loaded on to a chimney supporting a Whatman-42 filter paper and filtered under vacuum. The majority of ^{88}Y went through the filter paper and the residue was efficiently eluted with H_2SO_4. Finally, the solution containing radioyttrium was evaporated to dryness and reconstituted with HCl [32].

RESULTS AND DISCUSSION

Calculation of the excitation function

In this study, excitation functions of the proton-induced reactions of ^{88}Y and ^{85}Sr were calculated using EMPIRE (version 3.1 Rivoli) and ALICE/ASH [33], TALYS-1.26 nuclear codes and the TENDL-2010 database and then compared to existing data. The codes were used simultaneously, so as to increase the accuracy of the calculations.

Excitation function of ^{nat}Sr(p, xn)^{88}Y and ^{88}Sr(p, n)^{88}Y reactions

The evaluation of the calculated results with TALYS-1.26 showed that the best range of energy, one favoring the ^{nat}Sr(p, xn)^{88}Y reaction, is from 16 to 6 MeV (see fig. 2). For this reaction, two cross-section measurements exist in literature: the experimental data reported by Michel *et al.* [34], and Kettern *et al*. [5]. There is a relatively good agreement between the nuclear computer codes. According to the EMPIRE (version 3.1 Rivoli) and TALYS-1.26 codes, the maximum excitation function for a ^{nat}Sr(p, xn)^{88}Y reaction is 782 mb (1 b = 10^{-28} m^2) and 815.12 mb at E_p = 14 MeV,

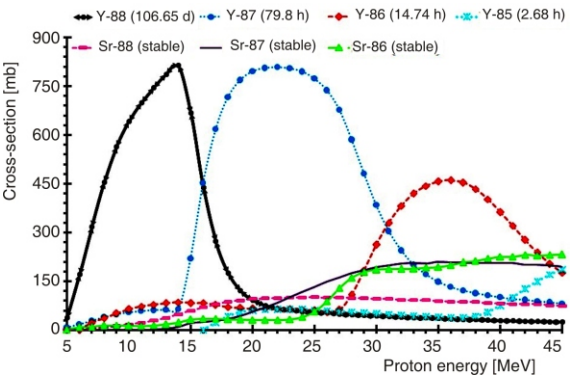

Figure 2. Excitation function of the ^{nat}Sr(p, x) reaction by TALYS-1.26 code

respectively. As shown in fig. 3(a), the data from refs. [5] and [34] exhibit lower values than nuclear model calculations of the codes. Also, to produce ^{88}Y with enriched strontium, Sachdev *et al*. [35] obtained nineteen data points up to 85 MeV, demonstrated to be the maximum cross-section of 960 mb at 15 MeV. Later, Levkovskij [36] and van der Meulen *et al*. [3] reported the maximum cross-section of 1070 and 894.78 mb at E_p = 12.8 MeV and 12.5 MeV, respectively. Theoretical yttrium-88 production cross-sections for ^{88}Sr(p, n)^{88}Y reactions have been illustrated in fig. 3(b).

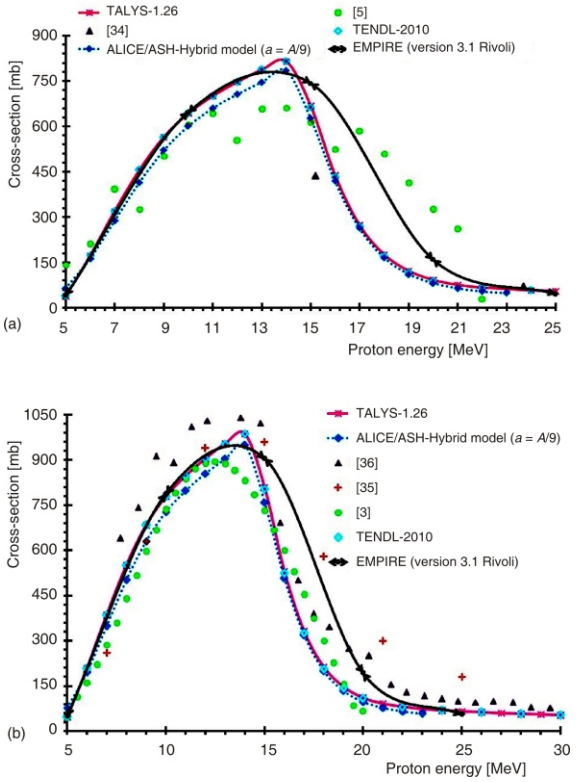

Figure 3. Excitation function of (a) ^{nat}Sr(p, xn)^{88}Y, and (b) ^{88}Sr(p, n)^{88}Y reactions by EMPIRE (version 3.1 Rivoli), ALICE/ASH, and TALYS-1.26 codes, the TENDL-2010 database and experimental data. The level density parameter (a) is equal to $a = A/y$. The default value of y is 9

Excitation function of the ^{nat}Sr(d, xn)^{88}Y reaction

Figure 4 shows a comparison between calculated cross-sections of the ^{88}Sr(d, xn)^{88}Y reaction. Up to now, no experimental data for this reaction were to be found in literature, meaning that nuclear model calculations might have an important role in finding the maximum excitation function of the said reaction. The best range of incident energy was assumed to be that of 25 to 10 MeV, and according to the EMPIRE (version 3.1 Rivoli) and TALYS-1.26 codes, the maximum cross-section for this reaction is 850 mb and 871 mb at E_d = 17 MeV, respectively.

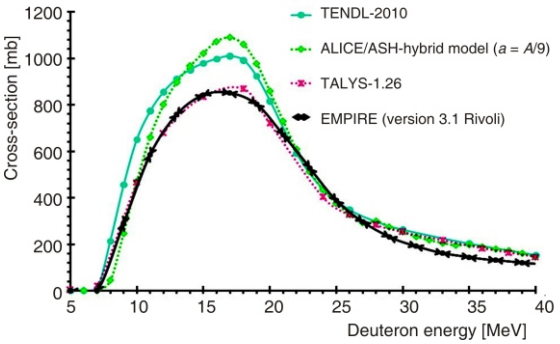

Figure 4. Excitation function of ^{nat}Sr(d, xn)^{88}Y by EMPIRE (version 3.1 Rivoli), ALICE/ASH, and TALYS-1.26 codes, TENDL-2010 database and experimental data

Excitation function of ^{nat}Rb(p, xn)^{85}Sr and ^{85}Rb(p, n)^{85}Sr reactions

Natural rubidium consists of ^{85}Rb (72.16%) and ^{87}Rb (27.84%). The natural rubidium target can be used to produce ^{85}Sr throughout accelerator proton bombardment. According to code results, the beneficial energy range of the proton particle to produce ^{85}Sr from ^{nat}Rb target is 16 MeV to 7 MeV. The results of nuclear model calculations by the three codes with measurements by Sakamoto *et al*. [37] and Buthelezi *et al.* [38] are shown in fig. 5(a). There is a relatively good agreement between the codes up to 13 MeV, whereas the results from refs. [37] and [38] exhibit lower values than the codes. Using the ^{85}Rb(p, n)^{85}Sr reaction to produce ^{89}Zr, the best range of incident energy was assumed to be 15 MeV to 7 MeV. The maximum cross section by the EMPIRE (version 3.1 Rivoli) code is 798 mb (E_p = 10.5 MeV), see fig. 5(b). For this reaction, two cross-section measurements exist in literature. Experimental data reported by Kastleiner *et al.* [39] and Levkovskij [36].

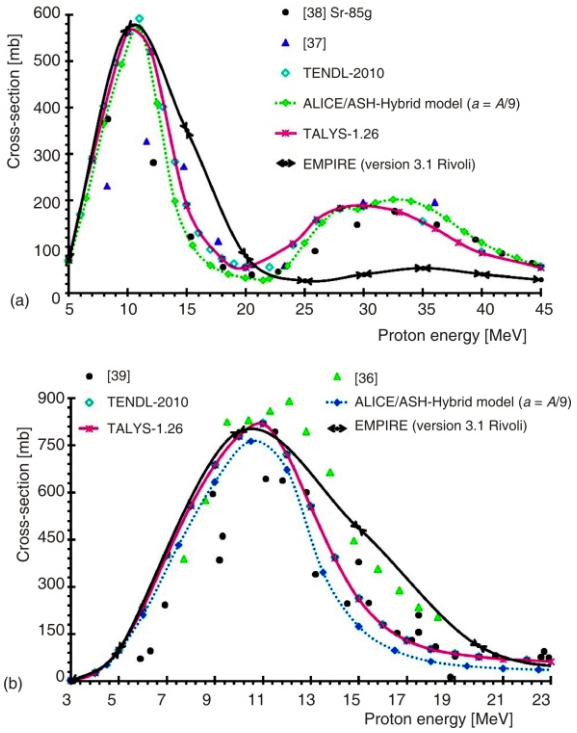

Figure 5. Excitation function of (a) ^{nat}Rb(p, xn)^{85}Sr and (b) ^{85}Rb(p, n)^{85}Sr reactions by EMPIRE (version 3.1 Rivoli), ALICE/ASH and TALYS-1.26 codes, TENDL-2010 database, and experimental data

Radiochemical separation

In order to develop the Y/Sr radiochemical separation, it was necessary to add ^{85}Sr and ^{83,82m}Rb as tracers to the Sr- and Rb-targets, respectively [2]. The production of ^{85}Sr and ^{81,82m,83}Rb is carried out via the ^{nat}Rb(p, xn)^{85}Sr and ^{nat}Kr(p, xn)^{81,82m,83}Rb reactions with 15 and 26.5 MeV protons, respectively [40]. The γ-ray spectrum of ^{85}Sr after radiochemical separation is presented in fig. 6. Yields of about 1.14 MBq ^{85}Sr per μAh were experimentally obtained.

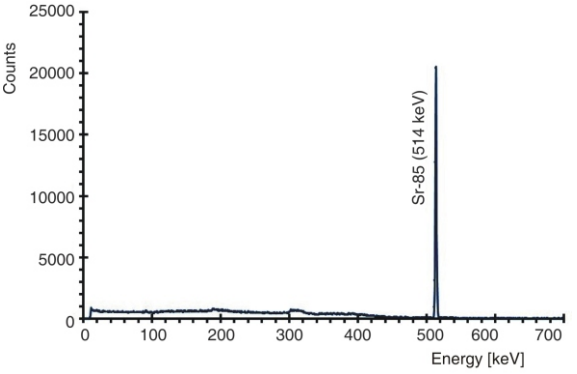

Figure 6. HPGe spectrum of ^{85}Sr after radiochemical separation; nother peaks have been detected in the γ-spectrum

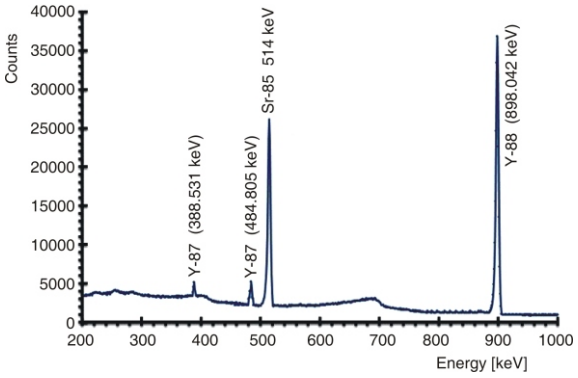

Figure 7. Non-destructive γ-ray spectrum of proton-irradiated $^{nat}SrCO_3$, taken 18 day after the end of bombardment (EOB)

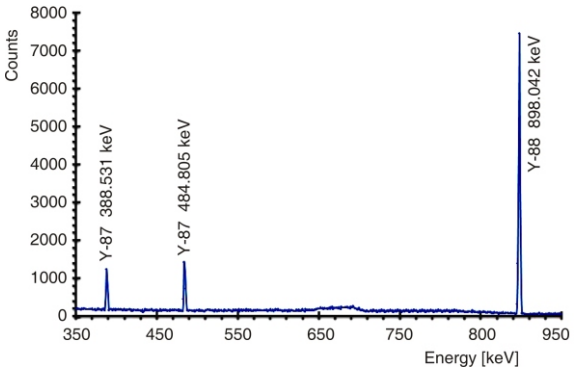

Figure 8. HPGe spectrum of radiochemically separated radioyttrium radionuclides, taken 19 day after the end of bombardment; no other peaks have been detected in the γ-spectrum

The total of the irradiated strontium carbonate-deposited target on the copper backing was dissolved with a liquid flow-through stripper in an acetonic system with 50 mL acetone. The obtained solution was evaporated to near dryness and the residue dissolved in 12 M HCl (10 mL). Then, the obtained solution was filtrated with a Whatman-41 filter paper, so as to remove ethyl cellulose. A filtrate solution was used for loading in the precipitation technique.

The separation of Yttrium from Sr was done by the precipitation method. The dissimilarity magnitudes of solubility product equilibrium constants for strontium sulfate and yttrium sulfate allow an efficient and rapid separation of Y from Sr by precipitating Sr as $SrSO_4$. The 2 M H_2SO_4 was the acid condition of choice to precipitate Sr with an approximate 92 5% overall yield efficiency over 30-40 minutes [32]. The filtration procedure reported by Avila-Rodriguez *et al*. [18] is faster and simpler, but the separation yield in that case less than 90%.

Table 1. Cyclotron production methods of ^{88}Y

Reaction	Beam energy [MeV]	Target enrichment	Yield at EOB [MBq/μAh]	Reference
^{nat}Sr(p, xn)^{88}Y	14 9	$^{nat}SrCO_3$ (99.99%)	1.75	[5]
	20 4	$^{nat}SrCl_2$ (99.99%)	1.6	[3]
	16 6	$^{nat}SrCO_3$ (99.99%)	1.02	[2]
	18 8	$^{nat}SrCO_3$ (99.99%)	1.326	This work
^{88}Sr(p, n)^{88}Y	18 8	$^{88}SrCO_3$ (99.99%)	1.605 (extrapolated)	This work
^{nat}Sr(d, xn)^{88}Y	25 10	$^{nat}SrCO_3$ (99.99%)	2.915 (theoretical)	This work
^{88}Sr(d, 2n)^{88}Y	25 10	^{88}RbCl (99.99%)	3.53 (theoretical)	This work
^{nat}Rb(α, n)^{88}Y	26 5	^{nat}RbCl (99.99%)	0.06	[2]
	26 5	^{nat}RbCl (99.99%)	0.08	[41]
^{nat}Sr(a, xn)^{88}Zr ^{88}Y	26 20	$^{nat}SrCO_3$ (99.99%)	8.2	[41]
^{nat}Mo(p, x)^{88}Zr ^{88}Y	800	Mo foil (99%)	0.116-0.049	[9]

Chemical and radiochemical purity control

The solutions from the separation of Y from Sr in non-radioactivity tests were analysed by ICP-AES. The amount of Sr impurity detected for 2 M H_2SO_4 was 8.2 ppm. The results demonstrate that increasing H_2SO_4 molarity diminishes Sr impurity. However, the separation yield for yttrium enhanced when the acid molarity increased and reached a maximum value of 97% at 2 M acid concentration [32].

Radionuclidic purity control

The identification and assay of gamma ray-emitting radionuclides was carried out using γ-ray spectroscopy with a high purity germanium (HPGe) detector (Canberra^{TM} model GC1020-7500SL). As shown in fig. 7, there is impurity in the ^{85}Sr tracer in the dissolved sample. After separation (fig. 8), the obtained radioyttrium purity was more than 99%. Yields of about 1.326 MBq ^{88}Y per μAh were experimentally obtained. The theoretical thick target for $^{nat}SrCO_3$(p, xn)^{88}Y and $^{88}SrCO_3$(p, n)^{88}Y reactions gives a yield of 2.482 and 3.008 MBq/μAh, respectively. An overview of some of the methods for the production of ^{88}Y is given in tab. 1.

CONCLUSION

Cyclotron production of ^{88}Y at the AMIRS was performed via the $^{nat}SrCO_3$(p, xn)^{88}Y reaction, with 18 MeV protons at a current of 20 μA for 10 hours. The deposition of $^{nat}SrCO_3$ on Cu backing was carried out by means of the sedimentation method. Yttrium-88 was separated in a 92 5% radiochemical yield, using the precipitation technique by precipitating ^{nat}Sr as $SrSO_4$, while radioyttrium passed through the filter paper. This quick method, lasting less than 1 hour, is suitable for the purification of radioyttrium. Also, strontium-85 as a gamma emitting radiotracer was produced by irradiation of sedimented ^{nat}RbCl.

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Милад ЕНФЕРАДИ, Махди САДЕГИ, Фатемех БОЛУРИНОВИН

ПРОИЗВОДЊА У АКЦЕЛЕРАТОРУ И НУКЛЕАРНА СВОЈСТВА РАДИООБЕЛЕЖИВАЧА ИТРИЈУМА-88

Итријум-88 ($T_{1/2} = 106.6$ d, $I_{\beta+} = 0.2\%$ и $I_{EC} = 99.8\%$) користи се у мешовитим стандардима за калибрацију гама ефикасности, а такође и као замена за итријум-90 у одређивању биодистрибуције итријумских фармацеутикала у животиња. Радиообележивачи итријум-88 и стронцијум-85, гама емитери, производе се посредством нуклеарних реакција $^{nat}SrCO_3$(p, xn) и ^{nat}RbCl(p, xn) на акцелератору AMIRS, са протонима енергија од 18 MeV и 15 MeV, респективно, при струји од 20 μA у току 10 часова. Депоновање $^{nat}SrCO_3$ на бакарну подлогу извршено је седиментационим поступком. Итријум-88 сепариран је од стронцијума као 92 5% радиохемијски принос, преципитацијом ^{nat}Sr у виду $SrSO_4$. Такође, употребом програма EMPIRE (верзија 3.1 Риволи), TALYS-1.26 и базе података TENDL-2010, проучени су пресеци за нуклеарне интеракције изазване протонима и деутеронима на ^{nat}Sr и ^{nat}Rb ради производње итријума-88 и стронцијума-85, респективно.

Кључне речи: производња у акцелератору, итријум-88, стронцијум-85, преципитациона техника, EMPIRE програм