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The production of ^{88}Y in the proton bombardment of $^{\text{nat}}\text{Sr}$: New excitation and separation studies

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abstract

The cyclotron production of ^{88}Y at iThemba LABS is performed via the reaction $^{88}\text{Sr}(p,n)^{88}\text{Y}$. The yields obtained were inconsistent with nuclear data obtained from the literature and the excitation function of the nuclear reaction was re-measured, using a differentiation of thick-target production rate measurements. Ion exchange chromatographic methods are described to separate ^{88}Y from $^{\text{nat}}\text{Sr}$ target material using AG MP-1 resin and AG 50W-X4 resins, respectively.

1. Introduction

^{88}Y ($T_{1/2}$ 106.6 d) can be effectively produced by means of cyclotron irradiation. With protons, it can be produced via the reaction $^{88}\text{Sr}(p,n)^{88}\text{Y}$. Its mode of decay is predominantly by means of electron capture and the decay emissions include the strong γ -rays of 898.0 keV (93.7%) and 1836.1 keV (99.2%), respectively (Firestone and Eckström, 2004). The radionuclide is formed in proton-induced reactions, using $^{\text{nat}}\text{Sr}$ as target material, only via the $^{88}\text{Sr}(p,n)^{88}\text{Y}$ reaction (Kettern et al., 2002), although $^{87}\text{Rb}(^3\text{He},2n)^{88}\text{Y}$ and $^{85}\text{Rb}(^4\text{He},n)^{88}\text{Y}$ routes have also been reported (Sachdev et al., 1967; Homma et al., 1980).

The separation of Y from Sr has been described in the literature by using two preferred methods, namely, by means of ion exchange chromatography (Saito, 1984; Grahek et al., 1999a, b, 2000; Kettern et al., 2002, Aardaneh et al., 2006) or by liquid extraction (Shikano et al., 1987). Electrochemical separation of their radioisotopes has also been described to obtain a product of high radiochemical and radionuclidic purity (Lange et al., 1957; Reischl et al., 2002). ^{88}Y has also been obtained as a product from $^{88}\text{Zr}/^{88}\text{Y}$ generators (Fassbender et al., 2004), separating ^{88}Zr from bombarded niobium capsules.

^{88}Y is used as small point sources in the calibration of instruments, as well as in the determination of mixtures containing Sr radionuclides (Lone et al., 1993), the accurate determination

of yttrium in superconductive oxide ceramics (Shikano et al., 1987; Katoh et al., 1987) and as a substitute for ^{90}Y (a β -emitter radionuclide used for therapy) to quantify the biodistribution of Y-pharmaceuticals in animals (Rösch et al., 1993a, b; Goodwin et al., 1994; Behr et al., 1999; Griffiths et al., 2003). It is used effectively as tracer for the chemical yield determination of ^{90}Y (Arzumanov et al., 2001).

The excitation function for the production of ^{88}Y via the (p,n) reaction on ^{88}Sr has been measured by several authors. Recently, Kettern et al. (2002) published new data up to 25 MeV and compared their results with the relevant older (and rather incomplete for this energy region) data (Blaser et al., 1951; Sachdev et al., 1967) measured in the 1950s and 1960s, as well as with a later data set by Levkovskii (1991). Discrepancies between the relevant data sets are rather large, almost a factor of 2 between the data of Kettern et al. and Levkovskii. It was, therefore, decided to re-measure the $^{88}\text{Sr}(p,n)^{88}\text{Y}$ excitation function up to an energy of nominally 20 MeV.

Metallic Sr reacts rapidly with oxygen in the atmosphere, while the available compounds of Sr are brittle. As a result, it proved to be difficult to exploit the conventional stacked-foil technique by preparing thin samples of a compound containing Sr. Although methods exist to obtain thin samples of brittle substances on metallic backing foils, such methods also have their disadvantages. In the case of the well-known sedimentation method (Rösch et al., 1993a, b), for example, the measured data are often plagued by very large scatter, probably due to deterioration of the targets during the course of those experiments. It was, therefore, decided to measure the thick-target production rate curve first, using thick targets, and to deduce the excitation function by means of a differentiation method (Vermeulen et al., 2007).

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2. Experimental

2.1. Nuclear data

A brass target holder contained an aluminium degrader, a Cu monitor foil and a SrCl_2 target for each of a series of bombardments. All the targets had similar thicknesses of nominally 800 mg/cm^2 and were thick enough to stop the beam. Activations were performed using degraders of various thicknesses to cover the energy region from threshold up to 17.5 MeV, the maximum energy which could be obtained from the ATOMKI cyclotron in Debrecen, Hungary. The monitor foils were high purity Cu (99.99%, Goodfellow Metals, UK) with a thickness of 25 μm , for the accurate determination of the incident proton flux. The $^{65}\text{Cu}(p,x)^{65}\text{Zn}$ monitor reaction with IAEA recommended cross sections (Gul et al., 2005) were used for this purpose. The targets were irradiated inside the cyclotron vacuum at an average beam current of 50 nA, each bombardment lasting about 30 min.

After bombardment, each SrCl_2 target was dissolved in 3 mL of water in a standard serum vial. Once filled and sealed, these vials constituted appropriate counting sources. The liquid sources ensured a consistent counting geometry, as well as allowing calibration sources of the same geometry to be easily prepared.

2.2. Radiochemical separations

Analytical grade reagents were used throughout this work and were obtained from Merck (SA) Pty. Ltd. or Sigma Aldrich GmbH, which included Sigma, Aldrich, Fluka and Riedel de Haen products. The AG 50W-X4 and AG MP-1 ion exchange resins used in this work were obtained from BioRad Laboratories, Richmond, USA. Deionized water from a Millipore MilliQ reagent grade water system, to a conductivity of greater than 10 MO/cm , was used for all experimental and production work.

2.2.1. Experiment 1

A target of 8.0 g bombarded SrCl_2 was dissolved in 100 mL water. To it was added a 10 mL solution containing 50 mg of ammonium carbonate and stirred. The ^{88}Y was precipitated and the Sr solution was decanted such that the precipitate was left behind. The precipitate was dissolved in 2 mL 2.0 M HCl, evaporated to dryness and the salts dissolved in 200 mL water. The solution was loaded on to a 10 mL column containing AG MP-1 resin (treated with 50 mL 25% NH_3 and the column rinsed with 100 mL water prior to use). The remaining contaminant (Sr) was removed from the column by eluting it with 100 mL water. The final product (^{88}Y) was eluted from the resin column with 25 mL 6.0 M HCl and evaporated to dryness, before finally being picked up in 5 mL 0.1 M HCl.

2.2.2. Experiment 2

Bombarded SrCl_2 (8.0 g) was dissolved in 50 mL 0.5 M HCl and loaded on to a 10 mL column containing AG 50W-X4 cation exchange resin (equilibrated with 50 mL 0.5 M HCl). Any excess ^{85}Sr was removed from the resin column using 70 mL 1.2 M HNO_3 . The ^{88}Y was eluted using 50 mL 4.0 M HNO_3 and evaporated to dryness, before finally being picked up in 5 mL 0.1 M HCl.

3. Results and discussion

3.1. Nuclear data

The ^{88}Y activities were determined by off-line γ -ray spectroscopy using the 898.0 keV (93.7%) and 1836.1 keV (99.2%) γ -lines.

The statistical uncertainties were insignificant compared to the systematic uncertainty, except near the reaction threshold, the latter of which was estimated to be about 7%: beam current integration (4%), detector efficiency (5%), counting geometry (1%) and decay corrections (2%). The ^{65}Zn cross sections extracted from the Cu monitor foils were also found to be in excellent agreement with the IAEA recommended values, thus, the directly measured current integrator values were independently confirmed (see Fig. 1).

The measured ^{88}Y thick-target production rate curve for SrCl_2+p is shown in Fig. 2. The reaction threshold is near 4 MeV and the production rate for the full energy region, i.e. from threshold up to the maximum measured energy of 17 MeV, is about 1.6 $\text{MBq/}\mu\text{Ah}$. A standard polynomial function was fitted through the measured data using the code TABLECURVE 2D (1996).

The polynomial of Fig. 2 could be differentiated analytically, allowing the derivation of the ^{88}Y excitation function for $^{88}\text{Sr}+p$, shown in Fig. 3. The data of Kettem et al. (2002) and Levkovskii (1991)

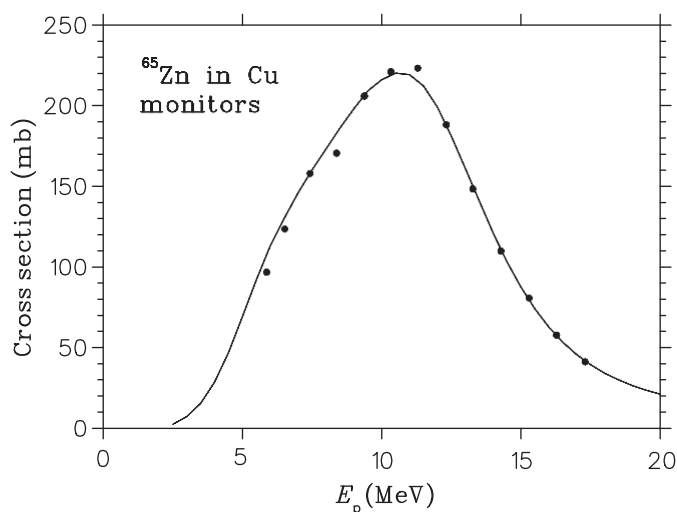


Fig. 1. Excitation function of ^{65}Zn in the reaction of protons with Cu monitor foils. The solid curve represents the values as recommended by IAEA (Gul et al., 2005), while the bullet symbols are the measured values for this work.

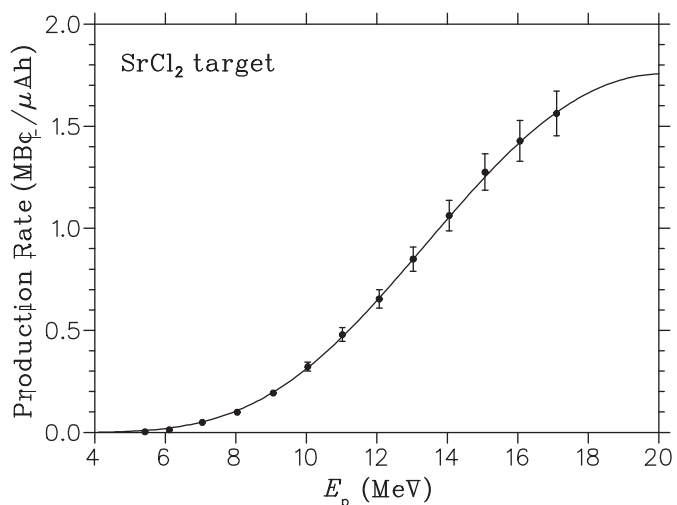


Fig. 2. Thick-target production rate curve of ^{88}Y produced in the proton bombardment of SrCl_2 . The solid symbols are the measured values of this work while the solid curve is a polynomial fit. Error bars are shown when they exceed the symbol size.

are also shown. Interestingly, the data of this work are lower than those of Levkovskii but higher than those of Kettern et al., falling just about half-way between those two data sets. The maximum of the excitation function is at about 12.5 MeV, thus, the energy region 4–20 MeV is ideal for the routine production of ^{88}Y .

Theoretical calculations were performed with the code ALICE-IPPE (Ignatyuk et al., 1993) in order to compare with the existing data and, if found to give a reasonable reproduction, to extrapolate the measured curves up to 20 MeV. A standard input prescription was used, the details of which have been summarized elsewhere (Vermeulen et al., 2007). The results are shown in Fig. 4.

Also shown in Fig. 4 is a rescaling of the measured cross-section values of Levkovskii according to a recommended prescription by Takács et al. (2002). The prescription was recommended after it was determined that Levkovskii's data were 20% too high, as a result of the older, less accurate, monitor reaction data used for beam flux determination. The renormalized values of Levkovskii are in good agreement with the values of this work. The overall agreement with the ALICE-IPPE prediction is

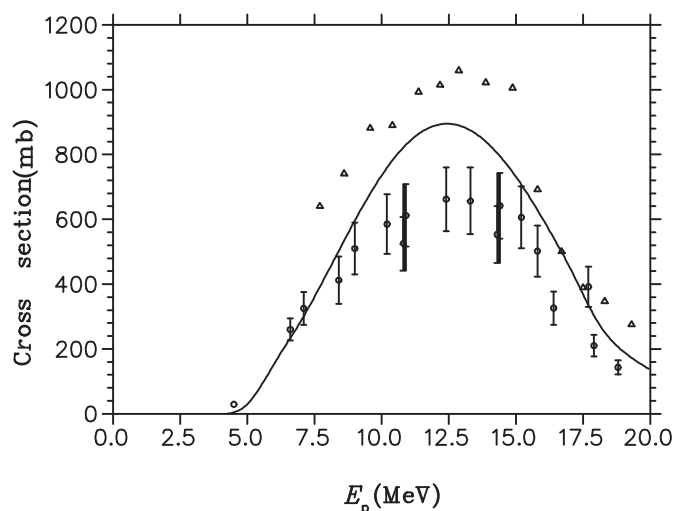


Fig. 3. Excitation function of ^{88}Y formed in the reaction of protons with ^{88}Sr . The solid curve was derived from the measured thick-target production rate data of this study (see Fig. 2). The open triangles are the data of Levkovskii (1991) and the open circles the data of Kettern et al. (2002).

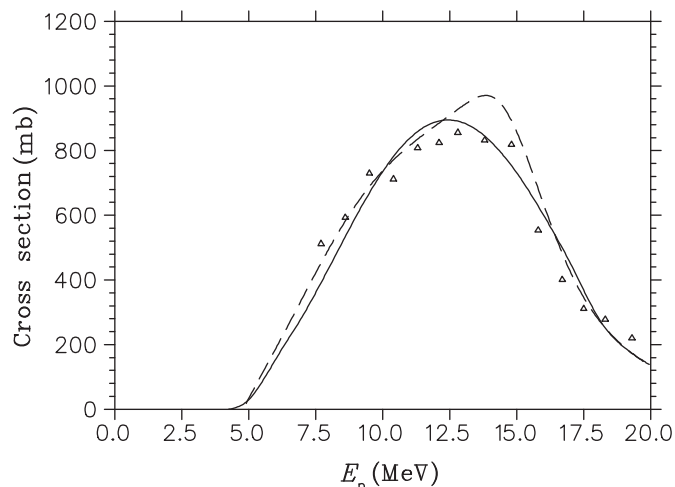


Fig. 4. Excitation function of ^{88}Y formed in the reaction of protons with ^{88}Sr . The solid curve was derived from the measured thick-target production rate data of this study (see Fig. 3). The open triangles are the re-scaled values of Levkovskii (see text) while the broken curve is a theoretical prediction by means of the computer code ALICE-IPPE.

also acceptable, although the shape is somewhat skewed towards a maximum at an energy about 1.5 MeV or higher.

3.2. Radiochemical separations

3.2.1. Experiment 1

Approximately 90% of the total Sr dissolved was removed by decanting. From the results obtained, the separation of Sr from Y was performed successfully (with yield of ^{88}Y ranging between 96.21% and 97.04%) and this experiment proved to be reproducible. The use of a more concentrated acid for the elution of ^{88}Y was to ensure that any iron in the production process would remain on the resin column. An elution curve demonstrating the separation of ^{88}Y from Sr is shown in Fig. 5.

3.2.2. Experiment 2

The procedure used for this experiment produced excellent results. All of the impurities were removed with the respective elution steps, while the ^{88}Y yield was 99.1%. When the experiment was repeated, the results with regard to the product yield were similar, although the percentage impurities (^{85}Sr) removed differed from the previous

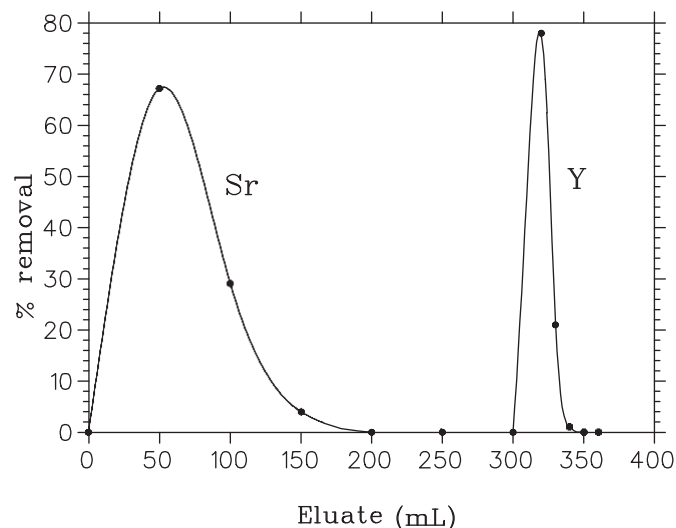


Fig. 5. Elution of ^{85}Sr and ^{88}Y from AG MP-1 using water and 6.0M HCl, respectively.

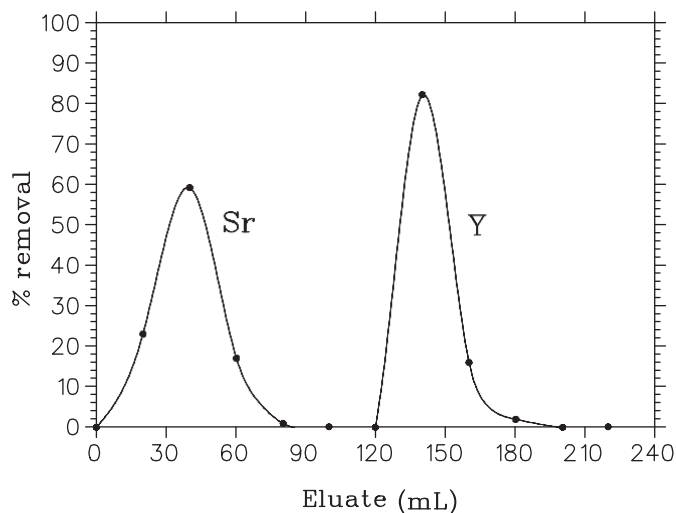


Fig. 6. Elution of ^{85}Sr and ^{88}Y from AG 50W-X4 using 1.2M HNO_3 and 4.0M HNO_3 , respectively.

experiment. They were all effectively removed from the resin column, however. From the experiments and productions performed, elution curves of ^{85}Sr and ^{88}Y were generated and are shown as Fig. 6.

4. Conclusion

A thick-target production rate curve was measured for ^{88}Y produced in the proton bombardment of SrCl_2 , from which the $^{88}\text{Sr}(p,n)^{88}\text{Y}$ excitation function was derived using a differentiation method. Good agreement was found with one set of literature data once those values were renormalized according to more recent monitor reaction data.

An effective separation between ^{88}Y from the Sr target material could be successfully obtained when using AG MP-1 macroporous anion exchange resin at a pH 4.7 or when performing a production using AG 50W-X4 cation exchange resin. The use of a co-precipitation step, using ammonium carbonate, proved to be very effective in the removal of the bulk of strontium from yttrium and can be used in both chromatographic methods.