Tailoring medium energy proton beam to induce low energy nuclear reactions in $^{86}\text{SrCl}_2$ for production of PET radioisotope ^{86}Y

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Abstract

This paper reports results of experiments at Brookhaven Linac Isotope Producer (BLIP) aiming to investigate effective production of positron emitting radioisotope ^{86}Y by the low energy $^{86}\text{Sr}(p,n)$ reaction. BLIP is a facility at Brookhaven National Laboratory designed for the proton irradiation of the targets for isotope production at high and intermediate proton energies. The proton beam is delivered by the Linear Accelerator (LINAC) whose incident energy is tunable from 200 to 66 MeV in approximately 21 MeV increments. The array was designed to ensure energy degradation from 66 MeV down to less than 20 MeV. Aluminum slabs were used to degrade the proton energy down to the required range. The production yield of ^{86}Y (1.2 ± 0.1 mCi (44.4 ± 3.7) MBq/ $\mu\text{A}\cdot\text{h}$) and ratio of radioisotopic impurities was determined by assaying an aliquot of the irradiated $^{86}\text{SrCl}_2$ solution by gamma spectroscopy. The analysis of energy dependence of the ^{86}Y production yield and the ratios of radioisotopic impurities has been used to adjust degrader thickness. Experimental data showed substantial discrepancies in actual energy propagation compared to energy loss calculations.

Keywords: low energy reaction, BLIP, Yttrium-86, production, energy degradation

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1. Introduction

Production of radioisotopes using proton induced reactions (p,n), (p,2n) with low threshold energies is best accomplished in small low energy (medical) cyclotrons. The energy of the proton beam at such facilities is often continuously tunable which facilitates optimum production yield and radiopurity. Understanding and controlling the uncertainty in the proton energy is especially important when radio radionuclide(s) of the same element with half-life comparable or longer than that of the radioisotope of interest is produced through competing nuclear reactions. Isotopes, radioactive or stable, cannot be separated chemically and impurity to product ratio will not decrease with time. A suitable example is the production of positron emitter ^{89}Zr ($T_{1/2}=78.4$ h) using the $^{89}\text{Y}(p,n)$ nuclear reaction. Its excitation function peaks at about 15 MeV reaching about 800 mb (Khandaker et al., 2012). Meanwhile, the cross section of the competing $^{89}\text{Y}(p,2n)^{88}\text{Zr}$ ($T_{1/2}=83.4$ d) reaction is about 300 mb at 15 MeV. While the production energy < 16 MeV has been deemed acceptable (Holland et al., 2009) the product radioisotopic purity reportedly can be improved if the incident energy is reduced: $^{88}\text{Zr}/^{89}\text{Zr}$ ratio (%) decreases from 4 at 15 MeV to 0.249 at 12 MeV to 0.004 at 10 MeV (Walther et al., 2011). Therefore, having a few MeV uncertainty in the energy of the protons hitting the target may become detrimental for the product radioisotopic purity.

While always undesirable, the uncertainty in the proton energy may be viewed as somewhat less critical in the case of the low energy $^{86}\text{Sr}(p,n)$ reaction, used for production of positron emitting ^{86}Y ($T_{1/2}=14.6$ days). While the separation of excitation functions energy domains for the $^{86}\text{Sr}(p,xn)$, $x=1, 2$ reactions is quite similar to the $^{89}\text{Y}(p,xn)$ reaction in the example above, the ratio of half-lives of the corresponding products is different (Rosch et al., 1993a). The likely radioisotopic impurities such as $^{85\text{m}}\text{Y}$ ($T_{1/2}=4.86$ h) and $^{85\text{g}}\text{Y}$ ($T_{1/2}=2.68$ h) that are produced through the (p,2n) reaction have half-lives shorter than that of the ^{86}Y and therefore if produced can be minimized by adding necessary cool off time after the end of bombardment.

Brookhaven Linac Isotope Producer (BLIP) has been used for isotope production based on the nuclear reactions most probable at high and intermediate proton energies such as $^{\text{nat}}\text{Zn}(p,2p)^{67}\text{Cu}$ (200 MeV) (Mausner et al., 1998), $^{\text{nat}}\text{RbCl}(p,x)^{82}\text{Sr}$ (90→40 MeV) (Phillips et al., 2000). During routine operation several targets are positioned in array one after another separated by water gaps. Hence, every position in the array offers different energy range since protons lose energy as they pass through the targets and water gaps. The energy incident on the last target slot, positioned downstream of routinely irradiated RbCl targets is 34 MeV. It can be used for a number of reactions but uncertainty in the proton energy exiting RbCl hinders its utilization. The uncertainty in energy arises from proton straggle as well

as lack of data on the extent of melting of RbCl salt during irradiation and therefore its density. Therefore the usage of this slot is limited to nuclear reactions where the levels of the co-produced radioisotopic impurities are either acceptable or can be reduced by the decay after EOB. A good example of the former would be the $^{nat}\text{Ga}(p,x)^{68}\text{Ge}$ reaction that had been used at BLIP production of ^{68}Ge (Medvedev et al., 2009; Meinken et al., 2005). The radioimpurities ^{69}Ge ($T_{1/2}=39\text{h}$) and ^{71}Ge (decays with $T_{1/2}=11.7\text{ d}$ by electron capture accompanied by soft x-rays) are much shorter lived than ^{68}Ge ($T_{1/2}=270.95\text{ d}$)

In this paper we report the results of feasibility studies of the production of radioisotopes at BLIP at energies below 20 MeV. The $^{86}\text{Sr}(p,n)^{86}\text{Y}$ nuclear reaction was used as a test reaction partly due to favorable impurity/product excitation functions and half-lives ratios as mentioned above. Despite a highly energetic gamma ray emission profile, the radioisotope ^{86}Y has received considerable attention in the research community as a PET emitter for imaging paired with radiotherapeutic isotope ^{90}Y (Jodal et al., 2014; Price et al., 2014; Zhou et al., 2013). The commercial availability of this isotope is limited. The chemical separation of ^{86}Y from ^{86}Sr targets has been a subject of quite a few studies each of those addressed facility specific applications and limitations (Aardaneh et al., 2006; Avila-Rodriguez et al., 2008; Finn et al., 1999; Garmestani et al., 2002; Lukic et al., 2009; Park et al., 2004; Reischl et al., 2002; Rosch et al., 1993b; Sadeghi et al., 2010; van der Meulen et al., 2009). Since we also have reported the separation of Y from strontium chloride targets irradiated at BLIP previously (Medvedev et al., 2011; Medvedev et al., 2012b), this paper is only focused on production yields² and radioisotopic purity of the yttrium fraction as a function of energy incident on $^{86}\text{SrCl}_2$.

1 Experimental part

1.1 Target design and target material handling

Strontium chloride ($^{84}\text{Sr} - 8\%$, $^{86}\text{Sr} - 97.02\%$, $^{87}\text{Sr} - 0.78\%$, $^{88}\text{Sr} - 2.12\%$) was received from Oak Ridge National Laboratory, TN, USA. To ensure the salt was carbonate and hexahydrate free a portion of it (about 800-1000 mg per irradiation) was dissolved in 10-20 ml of 2.5 N HCl and the resulting solution was evaporated to dryness. The obtained $^{86}\text{SrCl}_2$ solid was heated at 250 °C for at least two hours, transferred to a desiccator and stored under vacuum over Drierite®.

The design of the capsule used for salt isolation for irradiation was similar to the one reported previously (Medvedev et al., 2012a). Briefly, the capsule comprised of two halves (Figure 1) made out of Aluminum alloy 6061: the downstream part is best described as a frustum of a cone positioned on a disk, with a cylindrical well ($d = 1.15\text{ inch}$) for target material machined on the top of the frustum. For the

²The Yield terminology throughout this manuscript is used according to the NNDC Memo CP-C/347McLane, V., 2004, Cross sections measured on thick targets. https://www-nds.iaea.org/nrdc/memo_cpc/cpc347.doc., accessed on January 12, 2015

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4 irradiation the salt was ground in a mortar and hand packed into the well. The upstream part represents a
5 disk machined to mate with the downstream part. The isolation of the target material from the cooling
6 water is ensured by an Inconel c-ring, positioned in a groove encircling the bottom of the frustum. A
7 smaller diameter c-ring is situated on the top of the frustum thus forming a barrier to prevent any powder
8 or melted salt from escaping the well. The small c ring seals against the upstream window upon capsule
9 closing. When assembled, the parts are bolted together with eight screws located on the periphery of the
10 capsule.
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17 After irradiation the capsule was delivered to a shielded hot cell where it was opened by loosening
18 and removing the screws. The lower part was flipped over a beaker with 2.5 N HCl to collect the salt.
19 Any salt that remained adhering to the lower or upper part of the capsule was dissolved in a few drops of
20 18 MOhm water. The drops were recovered using a plastic pipette and combined with the main target
21 solution fraction. A total of 20 ml of 2.5 N HCl was used for each experiment. A quantitative aliquot was
22 removed from the target solution, diluted and assayed by gamma ray spectroscopy.
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28 Due to the high cost of $^{86}\text{SrCl}_2$ the salt was recycled. After the assay the target solution was stored
29 until the dose rate was below 10 mR/h at contact. At this point the solution was removed from the hot
30 cell, and evaporated in a HEPA filtered hood. The obtained solid was treated similar to the target material
31 described above: heated at 250 $^{\circ}\text{C}$ for at least two hours, transferred to a desiccator and stored under
32 vacuum over Drierite[®].
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37 1.2 Irradiation conditions: beam current and energy

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40 The target station at BLIP receives pulses from Linac which routinely operates at 117 MeV for ^{82}Sr
41 production. Linac proton energy is incrementally tunable at nominal energies of 37, 66, 93, 117, 139, 160,
42 181, and 200 MeV.. The beam of protons from Linac is directed through Beryllium (0.3048 mm),
43 AlBemet[®] (0.3048 mm), stainless steel (0.787 mm) windows, 2.67 mm of cooling water, stainless steel
44 target box window (0.508 mm) and a layer of cooling water before reaching the first target. As a result
45 the 37 MeV beam is degraded to <9 MeV by the time it reaches the first target. Therefore the next Linac
46 energy tune of 66 MeV was used in this experiment. The 66 MeV beam degrades down to 51 MeV when
47 it exits the stainless steel target box window. Since the optimum energy for $^{86}\text{Sr}(p,n)^{86}\text{Y}$ reaction is 14.5
48 MeV additional energy degradation was ensured using aluminum slab degraders of different sizes,
49 manufactured specifically for this work. The degrader and the capsule holder were designed such that the
50 water gaps between the upstream target box window, degrader, and face of a target capsule remain
51 constant at 2.54 mm. Energy degradation and straggling in the windows, cooling water, degrader, and
52 target material were calculated using TRIM subroutine in SRIM 2013 (Ziegler and Biersack, 2013).
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In each experiment, the target array was irradiated at the Linac energy tune of 66 MeV (Table 1). The actual incident proton energy was measured by a laser profile monitor. Total accumulated charge during target irradiation was measured using a beam current transformer located in the beam line. Since the beam spot area is larger than 1.15 inch diameter of the $^{86}\text{SrCl}_2$ puck, a radiochemical measurement of the proton beam was carried out. Two ^{nat}Cu foils ($d \times h = 1.15 \times 0.005$ inch and $d \times h = 2.5 \times 0.005$ inch) monitor foils were positioned upstream against target box window and irradiated together for 10 minutes before each $^{86}\text{SrCl}_2$ irradiation experiment. The foils were attached to Al backing slab. The calculated incident energy on the upstream monitor foil was 48.4 MeV. After irradiation the foils were dissolved individually and analyzed by assaying an aliquot of the solution by gamma spectroscopy for ^{62}Zn content. Activity values were decay corrected to the EOB. From the activity measurements, the beam current was calculated using the activation equation (Helus and Wobler, 1983) using the cross section data for the $^{nat}\text{Cu}(p,x)^{62}\text{Zn}$ nuclear reaction tabulated by International Agency for Atomic Energy (IAEA). To arrive to the tabulated values in 40-60 MeV range IAEA used cross sections measured separately by Mills (Mills et al., 1992), Michel (Michel et al., 1997), and Uddin (Uddin et al., 2004). The relative error in cross section values for the data sets varied between 7.1% and 9.5%, so the average value of 8.1 % was assumed for uncertainty estimations in this work.

The accumulated charge values (current $\times$ time) given in Table 1 were calculated as follows:

$$\Delta\mu A_h(^{86}\text{SrCl}_2) = \frac{[\text{radiochemical } \Delta\mu A_h(\text{Cu}_{1.15}^{\text{foil}})]}{[\text{beamline transformer } \Delta\mu A_h(\text{Cu foils})]} \times [\text{beamline transformer } \Delta\mu A_h(^{86}\text{SrCl}_2)]$$

where $\text{radiochemical } \Delta\mu A_h(\text{Cu}_{1.15}^{\text{foil}})$ – accumulated charge calculated from radiochemically measured beam current based on the activity in 1.15 inch foil, multiplied by irradiation time;

$\text{beamline transformer } \Delta\mu A_h(\text{Cu foils})$ – accumulated charge measured by the beam line transformer during Cu monitor foils irradiation

$\text{beamline transformer } \Delta\mu A_h(^{86}\text{SrCl}_2)$ - accumulated charge measured by the beam line transformer during $^{86}\text{SrCl}_2$ target irradiation.

The beam current and accumulated charge obtained from larger foil ($d=2.5$ inch) was mainly used for additional beam diagnostics and was not part of the above equation.

1.3 Determination of theoretical and experimental production yield of ^{86}Y

Theoretical yield Since the excitation function for $^{86}\text{Sr}(p,n)^{86}\text{Y}$ reaction varies substantially between 14.5 and 10 MeV, the total predicted activity of ^{86}Y was determined as a sum of the activity values produced in each of the n layers of the $^{86}\text{SrCl}_2$ puck.

$$A_{total} = \sum_{i=1}^n A_i$$

Activity in the individual layer of thickness Δx_i was determined using activation equation (Helus and Wobler, 1983). All parameters were converted to SI units; the conversion coefficients are omitted for clarity:

$$A_i = I \times \frac{\rho \times N_a}{M} \times \Delta x_i \times \sigma_i(\bar{E}_i) \times \left(1 - e^{-\frac{0.693}{T_1} \times t} \right)$$

where, I – beam current, prot/sec, ρ – density of $^{86}\text{SrCl}_2$, g/cm³, M – molecular weight of $^{86}\text{SrCl}_2$, Δx_i – thickness of the i -th layer, cm, $\sigma_i(\bar{E}_i)$ – cross section value for the $^{86}\text{Sr}(p,n)^{86}\text{Y}$ reaction for energy for the energy of the protons on the i -th layer. To obtain the expression for $\sigma(E)$, the cross section data (Figure 2) was fitted in the Sigma Plot 12.5 software (see Supplemental information S1).

The entrance energy on each layer was determined as follows:

$$E_{i+1} = E_i + dE/dx(E_i) \times \Delta x_i$$

Where $dE/dx(E_i)$ – is the stopping power for the protons having the energy i -th layer. The energy dependent stopping power $dE/dx(E)$ for protons in $^{86}\text{SrCl}_2$ was calculated using SRIM subroutine of SRIM 2013 software. The generated data in the output file was fitted using Sigma Plot[®] (Systat software, Inc, version 12.5, San Jose, CA) software (see Supplemental information S2). The algorithm for yield calculations was realized in Microsoft Excel[®] 2010.

Further, the theoretical target yield of ^{86}Y can be calculated dividing the total activity by the accumulated charge:

$$Y = \frac{A_{total}}{\Delta \mu A \cdot h}$$

Experimental production yield of yttrium isotopes To determine experimental production yield the irradiated salt was assayed for ^{86}Y and other co-produced radioimpurities (Table 2) using gamma spectroscopy. In a typical experiment the target can was opened within 3 hours after EOB and the salt was dissolved in a total of 20 ml of 2.5 N HCl. The exact volume of the solution was determined by weight taking into account the density of the solution. An aliquot of the solution was assayed on the ORTEC HPGe detector (AMETEK[®], Oak Ridge, TN). GammaVision-32 software, version 6.01 (AMETEK[®], Oak

Ridge, TN) was used for spectra acquisition and processing. The gamma rays given in the Table 2 were used to determine activity of the corresponding isotopes. The radioisotope ^{85m}Y was not measured as its peak corresponding to the gamma ray at 231.7 keV (22.8%) overlaps with the 231.65 keV (84%) gamma ray peak of ^{85}Y . The activity of ^{85}Y was measured using 504 keV photopeak in which case a 1.5% abundant contribution from corresponding peak of ^{85m}Y was considered nilpotent. Activity of ^{88}Y was measured several days after EOB, when ^{86}Y has decayed substantially. Activity values were decay corrected to the EOB including the activity of ^{87}Y which was created from reactions on $^{87,88}\text{Sr}$ impurities in the target material as well as indirectly generated by the decay of ^{87m}Y . The efficiency and energy calibration of the detector was carried using NIST traceable multinuclide gamma standard. The reproducibility of activity values was verified using NIST traceable Eu-152 standard and was found to be within 3% for 1408 and 344 keV peaks. The standards were purchased from Eckert and Ziegler® (Atlanta, GA). The overall error in this measurement (5.3%) was estimated as a combination of the error in dilution (1-2%) and error in activity measurement (3-5%). The overall error in the yield value included uncertainty from the beam current measurement (8.1%) and was therefore 9.7%.

2 Results and discussion

2.1 Target opening and $^{86}\text{SrCl}_2$ recovery

The target capsule used in this work has proven to be quite adaptable to irradiation of various materials in a form of foils, disks and pressed powder. It can be sealed and opened remotely and reliably isolates the content from cooling water. Thermal analysis of the target irradiation has predicted the $^{86}\text{SrCl}_2$ temperature rising above its melting point during irradiation. Upon target opening regions of molten salt having darker color were observed. While most of the salt readily dissolved in 2.5 N HCl, the molten pieces took somewhat longer. Since the recovery of the target material was carried out after the dose rates fallen off substantially no remote handling was required.

2.2 Dependence of the production yield on the degrader size - uncertainty in energy calculation

Initially the size of the degrader was chosen such that TRIM-calculated energy incident on SrCl_2 was 14.5 MeV. However, we did not observe the expected products of the salt activation upon target solution assay. The only product found was ^{87m}Sr in microCurie quantities which was most likely due to secondary neutron activation of the ^{86}Sr nuclei. Since no beam missteering was suspected, it was concluded that the proton beam prematurely degraded to the energies below 8 MeV. Hence, in the course of the consequent experiments the thickness of the degraders was decreased incrementally. The incremental decrease in the

degrader size resulted in the increase of the yield of the activation products (Table 3). The activation products for the experiments 1-3 are not shown in the Table 2: run 1 revealed no activation, while 2 and 3 showed ^{86}Y present in microCurie quantities. From experiment 4 to 5 a substantial increase in ^{86}Y yield did occur, while ^{85}Y still remained undetected. In run 6 the yield of ^{86}Y almost doubled compared to run 5, however the yield of ^{85g}Y has increased as well. This pattern is consistent with the excitation functions for this reaction (Figure 2), from which it is clear that the impurity of ^{85g}Y will increase as energy increases, while the yield of ^{86}Y will go down. This is demonstrated in the run 6 as the energy moved up from the (p,n) cross section peak. Run 7 is manifested by substantial increase in $^{87m}\text{Y}/^{86}\text{Y}$ ratio which is most likely due to the proton energy being closer to the $^{88}\text{Sr}(p,2n)^{87m}\text{Y}$ reaction peak at 24 MeV. Overall, the change in radioisotopic purity of was consistent with the adjustment of energy degradation.

Calculated energy profiles on the $^{86}\text{SrCl}_2$ salt (E_{in}) for all the experiments are shown in Figure 3. As expected, E_{in} increases as the degrader becomes thinner and the calculated proton straggling is more pronounced when energy is degraded to a lower value. What was not expected is the 10-12 MeV difference between the desired $E_{in}=14.5$ MeV, and the energy that is calculated for the run 5, where the yield and radio purity ^{86}Y was the best.

The uncertainty in the calculated E_{in} stems from two reasons; one is the uncertainty in the input data, and the other is the uncertainty in the code prediction.

The accuracy of the most of the input parameters is assured by secondary measurement (incident Linac energy) or with the documentation for the components of the target array. The incident energy uncertainty was measured to be ± 0.02 MeV, while most of the dimensions of the array are designed and built to ± 0.005 inch (± 0.127 mm) tolerance. Deviation in either of these parameters is not likely to cause the observed large discrepancy in energy. While the target array is designed to ensure that water gaps do not change the clearance water gap (2.667 mm by the design) between the target box and the beamline window may have inherited uncertainty. The box is lowered into the cooling water on chains and its horizontal position may be affected by beam pulses and water flow. To estimate the effect of the uncertainty in the clearance gap on a calculated E_{in} we varied the value of the water gap in the TRIM input file for the run 5 (Figure 4). As evident from the Figure 5, ~ 4 -4.5 ml of additional water in front of the array is required to “make up” for the “lost energy loss” in the run 5 array. We believe however that a deviation of the target box by more than 1-2 mm from the designed position is unlikely and therefore cannot explain the observed discrepancy.

Similar pattern in energy degradation calculations was observed by the researchers at the Isotope Production Facility (IPF) located within Los Alamos National Laboratory, NM, USA (Nortier, 2014). In

their experiment the energy was degraded down to below 20 MeV to irradiate $^{nat}\text{WO}_3$ targets to produce ^{186g}Re via the $^{186}\text{W}(\text{p},\text{n})$ nuclear reaction (Shigeta et al., 1996) The discrepancy between the proton energy calculated based on Anderson and Ziegler approach (Andersen and Ziegler, 1977) (26.3 MeV) and the practical proton energy estimated from radioisotope yield ratios (15.6 MeV) was 10.6 MeV (Fassbender et al., 2013). In this work we observe the difference of ~ 12 MeV between TRIM predicted E_{in} and optimum energy. Based on these observations, the discrepancy in energy seems to point to inaccuracy in the code predictions. Calculated energy values using G4beamline code (Roberts, 2013) were nearly identical to those obtained with the TRIM subroutine. The reason for the difference between calculated and actual energy, determined based on the ^{86}Y yield and impurity ratios remains unknown at this point.

2.3 Experimental production yield of ^{86}Y and its radioisotopic purity

Based on the radiopurity and yield values trend the actual proton energy for the run 5 was the most favorable for the production of ^{86}Y (Table 5) and most likely close to 15 MeV. The level of other yttrium radioisotope impurities was comparable to the data obtained in small cyclotrons by other groups (See table 5 for references). Further increase in energy resulted in elevation of the level of the $^{85m+g}\text{Y}$. The half-life of these isotopes are shorter than ^{86}Y so decay during shipment can be a way to reduce their level. However, both of the isotopes decay to a longer lived ^{85}Sr ($T_{1/2}=64.85$ d) which may present a challenge in further medical use of the ^{86}Y .

Theoretical thin target production yield of ^{86}Y was calculated based on the experimental cross section data for the energy range 14.4→10.2 MeV reported previously (Levkovskij, 1991; Rosch et al., 1993a). For the run 5 irradiation parameters (beam current, time of irradiation, and target thickness) the yield of ^{86}Y was 2.6+/-0.3 mCi (96.2+/-11.1 MBq) per $\mu\text{A}\cdot\text{h}$. The uncertainty associated with this number (12.9%) stems from the uncertainty in the cross section data for the $^{86}\text{Sr}(\text{p},\text{n})$ reaction ($\sim 10\%$) as well as uncertainty in the beam current measurement (8.1%). The regression that describes the excitation function for $^{86}\text{Sr}(\text{p},\text{n})$ reaction rests within 10% error bars for the data points from 15 to 10 MeV.

The production yield obtained in this work was 46% of the theoretical number (Table 2, run 5). Substantial discrepancy can be explained by the proton energy straggle phenomena. In the theoretical estimation of the yield the beam incident on a target was assumed to be monoenergetic, in practice it is not the case. The TRIM calculated energy straggle upon degradation of 66 MeV down to $E_{in}=15$ MeV is substantial: FWHM of the energy distribution is ~ 5 MeV (Figure 3). This means that a certain fraction of the beam do not produce the isotope at the assumed rate either because the energy is too high and off the cross section peak, or it is too low, even below threshold energy for this reaction. Moreover, upon further degradation through the salt the energy spread widens reducing effective beam current even more (Figure

5). It can be seen from Figure 5 that substantial fraction of the beam is below 10 MeV as it passes through the salt. Therefore it is reasonable to expect such a substantial deviation between calculated and experimental target yields obtained in this work. Other parameters, such as partial melting of the SrCl_2 salt can also contribute to the discrepancy. However we believe that proton straggle is the main reason. Proton straggle makes yield and purity prediction a very daunting task, especially if the cross section peaks have to be resolved so that the production of the impurities is minimized.

2.4 Implications for the isotope production at the low energy slot

It has been mentioned previously that the low energy slot at BLIP (downstream of RbCl targets) offers a proton energy range 34→0 MeV during routine operation. The utilization of this slot is somewhat limited for low energy reactions due to uncertainty in energy exiting RbCl targets. While the energy beam spread is not as substantial when degraded to 34 MeV as it is when to 15 MeV, the uncertainty in RbCl density is a more important factor. The Gaussian shape of the beam shape results in uneven thermal load of the RbCl target and therefore melting of the target material in the center with a large density reduction. Additional uncertainty in exit energy arises after beam interruptions, when the salt solidifies and when a new set of targets is irradiated, when the salt is initially a fresh pressed puck. This may inevitably and uncontrollably effect incident energy on the targets downstream of RbCl and hence influence the yield, radiopurity of the product. Often the integrity of the target can be compromised due to the unpredictable thermal load fluctuations. In this work we minimized this uncertainty by using solid degraders in order to see if energy straggling can be managed. We also have demonstrated that respectable ^{86}Y thin target production yield and product purity can be achieved in this manner if BLIP is tuned to 66 MeV.

With implementation of a beam raster system presently underway at BLIP it will be possible to achieve homogeneous melting of RbCl targets and minimize the uncertainty in exit energy arising from RbCl density fluctuations. This will open the opportunity for more accurate energy prediction.

3 Conclusions

The energy was degraded at BLIP from 66 MeV down to less than 20 MeV to produce ^{86}Y using the low energy $^{86}\text{Sr}(p,n)$ reaction. The optimum degradation array was determined experimentally by varying the degrader thickness and benchmarking it against the radionuclides irradiation yield and ratio, taking into account excitation functions for the reaction. The calculated energy using TRIM and G4beamline codes for the optimum degrader was found to be 10-12 MeV higher than experimentally observed. Similar difference in energy propagation calculations has been observed by other researchers and attributed to code prediction inaccuracies at low energy. Experimental yield for the optimum degradation

array was 1.2 mCi (44.4 MBq) of $^{86}\text{Y}/\mu\text{A}\cdot\text{h}$. This is 46% of theoretical value and can be attributed to energy spread due to proton energy straggling. Radionuclidic purity was comparable to the products obtained in a smaller cyclotron. Accessing low energy by degradation can be used for isotope production; however substantial experimental energy degradation optimization is required.

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Tables

Table 1. Irradiation parameters of $^{86}\text{SrCl}_2$ target at incident Linac energy of 66 MeV

Run	Al degrader, mm	Time, min	$^{86}\text{SrCl}_2$ mass, mg	$\Delta\mu A_h(^{86}\text{SrCl}_2)$,
1	8.30	60	757	not measured
2	8.00	30	1020	17.1
3	7.49	30	1020	17.3
4	6.99	30	1094	27.7
5	5.99	31	1060	27.7
6	5.03	30	821	14.1
7	4.01	30	822	12.8

Table 2. Summary of the nuclear data for yttrium radionuclides and cross section data for corresponding nuclear reactions on ^{86}Sr and isotopic impurities

Isotope	Half life	E_γ , keV (I, %)	Production pathway	Eth, MeV	Energy for max cross sections, +/- 1 MeV	Reference for cross sections
^{86}Y	14.74 h	1076.63 (82.5) 1153.05 (30.5) 627.72 (32.6) 777.37 (22.4) 443.13 (16.9) 703.33 (15.4)	$^{86}\text{Sr}(\text{p},\text{n})$ $^{86\text{m}}\text{Y}(\text{IT}) \rightarrow ^{86}\text{Y}$	6.09	14	(Levkovskij, 1991; Rosch et al., 1993a)
$^{86\text{m}}\text{Y}$	48 min	208.1 (93.7)	$^{86}\text{Sr}(\text{p},\text{n})$	6.09	14	(Levkovskij, 1991; Rosch et al., 1993a)
^{85}Y	2.68 h	504.4 (60)	$^{86}\text{Sr}(\text{p},2\text{n})$	15.7	24	(Levkovskij, 1991; Rosch et al., 1993a)
$^{85\text{m}}\text{Y}$	4.86 h	231.7 (22.8)	$^{86}\text{Sr}(\text{p},2\text{n})$	15.7	24	(Levkovskij, 1991 Rosch et al., 1993a)
^{87}Y	79.8 h	484.8 (82.2)	$^{87}\text{Sr}(\text{p},\text{n})$ $^{88}\text{Sr}(\text{p},2\text{n})$ $^{87\text{m}}\text{Y}(\text{IT}) \rightarrow ^{87}\text{Y}$	2.7	13	(Levkovskij, 1991; Sachdev et al., 1967)
				13.9	19	
$^{87\text{m}}\text{Y}$	13.37 h	380.79 (76.06)	$^{87}\text{Sr}(\text{p},\text{n})$ $^{88}\text{Sr}(\text{p},2\text{n})$	2.7	13	(Levkovskij, 1991; Sachdev et al., 1967)
				13.9	24	
^{88}Y	106.63 d	898.04 (93.7) 1836.2 (99.2)	$^{88}\text{Sr}(\text{p},\text{n})$	4.5	12	(van der Meulen et al., 2009; Levkovskij, 1991)

Table 3. Comparison of experimental (this work^a) and literature data for isotopic yields of Yttrium isotope using ⁸⁶Sr(p,n) reaction.

run	TRIM calculated <i>E_{in}</i> , MeV	^{86g} Y yield, mCi (MBq)/μA- h	Yttrium fraction composition, % of ^{86g} Y				
			^{86m} Y	^{85g} Y	^{87m} Y	^{87g} Y	^{88g} Y
4	21.8	0.02 (0.74)	Not detected	Not detected	n/d	n/d	n/d
5	26.5	1.2 (44.4)	225	n/d	1.04	0.37	0.013
6	30.3	2.2 (81.4)	255	15.6	1.0	0.31	0.012
7	34.1	0.8 (29.6)	372	598	5.8	1.6	0.08
Literature data							
(Yoo et al., 2005) ^b	14.5	4.5 (166.5)	220	Not reported	0.43	0.27	0.024
(Garmestani et al., 2002)	13.8+/-0.5	1.9 (70.3)	210	Not reported	1.42	0.4	0.11
(Rosch et al., 1993a)	14	4.2 (155.4)	280	<0.1	1.5-3.0	0.4-0.5	0.02-0.03
(Reischl et al., 2002)	15.1	1.3 (48.1)	400	Not reported	Not reported	0.5	0.03
(Avila- Rodriguez et al., 2008)	11	0.95 (35.15)	186	Not reported	2.5	0.27	0.37
(Lukic et al., 2009)	14.2	3.9 (144.3)	200	0.01	1.6	0.2	0.06

^a-9.7% uncertainty in the yield numbers ^b – SrO target

Figure Captions

Figure 1. A photograph of upstream (a) and downstream (b) parts of the bolted capsule with c-ring positioned in the grooves

Figure 2. Excitation functions plots for the $^{86}\text{Sr}(p,n)$ reaction ($\bullet$, Levkovskij 1991 (Levkovskij, 1991), $\blacktriangle$, $\circ$ Rosch 1993 (Rosch et al., 1993a)) and the $^{86}\text{Sr}(p,2n)$ reaction ($\blacksquare$, $\square$, Levkovskij 1991). The trend lines are shown by solid lines.

Figure 3. TRIM calculated proton energy profile incident on $^{86}\text{SrCl}_2$ salt calculated for various degrader thicknesses.

Figure 4. TRIM calculated E_{in} as a function of size an additional to 2.668 mm water layer between the beam line window and target box. The data is calculated for the run 5.

Figure 5. G4beamline simulation of proton energy distribution upon propagation through $^{86}\text{SrCl}_2$ target: the data combined for energy incident, in the middle and exiting $^{86}\text{SrCl}_2$ salt in run 1.

Figures

a)

b)

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