Cross sections of deuteron induced reactions on \( ^{nat} \text{Sm} \) for production of the therapeutic radionuclide \(^{145}\text{Sm} \) and \(^{153}\text{Sm} \)

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Abstract

At present, targeted radiotherapy (TR) is acknowledged to have great potential in oncology. A large list of interesting radionuclides is identified, including several radioisotopes of lanthanides, amongst them \(^{145}\text{Sm} \) and \(^{153}\text{Sm} \). In this work the possibility of their production at a cyclotron was investigated using a deuteron beam and a samarium target. The excitation functions of the \(^{nat} \text{Sm}(d,x)^{145,153}\text{Sm} \) reactions were determined for deuteron energies up to 50 MeV using the stacked-foil technique and high-resolution \( \gamma \)-ray spectrometry. The measured cross sections and the contributing reactions were analyzed by comparison with results of the ALICE, EMPIRE and TALYS nuclear reaction codes. A short overview and comparison of possible production routes is given.

Keywords: deuteron irradiation, natural samarium target, \(^{145}\text{Sm} \), \(^{153}\text{Sm} \), physical yield

1. Introduction

A large number of potential therapeutic radionuclides that emit low energy (conversion and Auger), intermediate and high energy electrons (-emitters), or \( \alpha \)-particles are known (Beyer, 2000; Neves et al., 2005; Qaim, 2001; Rosch, 2007; Uusijarvi et al., 2006a,b; Zalutsky, 2011). Among these, the presently investigated \(^{153}\text{Sm} \) is used for brachytherapy (Fairchild et al., 1987; Gowda et al., 2004; Meigooni and Nath, 1992) and \(^{153}\text{Sm} \) for treatment of bone pain (Bauman et al., 2005; Finlay et al., 2005; Ramamoorthy et al., 2002). Both above radioisotopes can be produced through neutron capture on samarium enriched in \(^{144}\text{Sm} \) and \(^{152}\text{Sm} \) respectively (IAEA, 2003, 2011). The production yields are high, but the products are carrier added with moderate specific activity. There is an effort to fulfill the requirements of nuclear medicine in medical radioisotope production without relying on nuclear reactors or at least without highly enriched uranium (Lakatong Foundation, 2010). The radio-lanthanides \(^{145}\text{Sm} \) and \(^{153}\text{Sm} \) can also be produced at accelerators by using charged particle or photonuclear reactions. In the frame of our systematic investigation of deuteron induced reactions on lanthanides we have measured the activation cross sections of many long-lived radio-products induced in samarium targets, among them \(^{145}\text{Sm} \) and \(^{153}\text{Sm} \). No earlier experimental data for their excitation functions were found in the literature. The results for production of \(^{145}\text{Sm} \) and \(^{153}\text{Sm} \) via deuteron induced reactions are compared. A discussion of the different production routes is also presented. Activation cross sections of the other investigated radioisotopes in \(^{nat} \text{Sm}(d,x) \) reactions will be published separately.

2. Experiment and data evaluation

The activation cross sections of the studied radioisotopes \((^{145}\text{Eu}, \quad ^{145}\text{Sm}, \quad ^{153}\text{Sm}) \) were measured, relative to the \(^{27}\text{Al}(d,x)^{22,24}\text{Na} \) and \(^{nat}\text{Ti}(d,x)^{48}\text{V} \) monitor reactions, by using a stacked foil activation technique and \( \gamma \)-ray spectrometry. More details of the experiment and the data evaluations are presented in Table 1, while the decay data used are shown in Table 2. The uncertainty on each cross-section point was determined in a standard way (of-Weights-and Measures, 1993) by taking the square root of the sum of the square of all relative individual contributions (except the nonlinear time parameters), supposing equal sensitivities of the different parameters appearing in the formula. The final uncertainties of the cross-sections contain uncertainties of the beam current measurement (7 %), the number of target
nuclei (5 %), the determination of activities and conversion to absolute number of the produced nuclei (1-15 %). The absolute values of the cross-sections are estimated to be accurate within 13 %. The uncertainty of the energy scale was estimated by taking into account the energy uncertainty of the primary beam (0.3 MeV), the possible variation in the target thickness and the effect of beam straggling. The energy uncertainty in the last foil was estimated to be 1.3 MeV.

3. Model calculations

The updated ALICE-IPPE (Dityuk et al., 1998) and EMPIRE (Herman et al., 2007) codes (the modified versions for deuteron induced reactions ALICE-IPPE-D and EMPIRE-D) were used to analyse the present experimental results. These modifications were developed and implemented in the original codes at IPPE and more details can be found in our previous reports (Herman et al., 2009; Tarkanyi et al., 2007). In these codes a simulation of direct (d,p) and (d,t) transitions by the general relations for a nucleon transfer probability in the continuum is included through an energy dependent enhancement factor for the corresponding transitions. The phenomenological enhancement factor is based on systematics of experimental data of the related reactions (Ig-natyuk, 2011). The theoretical data from the TENDL-2012 (Koning et al., 2012) library (based on the most recent TALYS code (Koning et al., 2007) were also included in the comparison.

4. Results

According to Table 1, the $^{145}$Sm ($T_{1/2} = 340$ d) is produced directly and through the decay of $^{145}$Eu ($T_{1/2} = 5.93$ d). Both products were identified in our spectra. Possible contribution from long-lived $^{145}$Pm ($T_{1/2} = 17.7$ a) can be ignored in this experiment. The situation is similar for $^{153}$Sm ($T_{1/2} = 46.50$ h), but in this case the parent $^{153}$Pm ($T_{1/2} = 5.3$ min) was not detected due to the short half-life and the low predicted cross sections.

4.1. Excitation functions

The cross sections for all the reactions studied are shown in Figures 2, 5 and the numerical values are shown in Table 3. The results for the 40 and 20 MeV primary incident energies are shown separately to point out the agreement in the overlapping energy range. The reactions responsible for the production of the given activation products and their Q-values, obtained from the calculator of the Brookhaven Nat. Lab. (Pritychenko and Sonzogni, 2003), are given in Table 2.

4.1.1. $^{145}$Eu (direct production)

The radioisotope $^{145}$Eu ($T_{1/2} = 5.93$ d) is formed at low energies by the $^{144}$Sm(d,n) reaction (first maximum) and by $^{147-150}$Sm(d,4-7n) reactions above 20 MeV. The measured experimental data are shown in Fig. 2, together with the theoretical predictions. The results in TENDL-2012 slightly overestimate both the low and high energy experimental data. The overestimation is more significant in the case of EMPIRE-D and ALICE-D.
### Table 1: Main experimental parameters and methods of data evaluation

| Parameter                        | Value                                                                 | Data evaluation                                                                 | Method | Reference          |
|----------------------------------|-----------------------------------------------------------------------|---------------------------------------------------------------------------------|--------|--------------------|
| Reaction                         | Sm(d,x)                                                              | Gamma spectra evaluation                                                        |        |                    |
| Incident particle                | Deuteron                                                             | Determination of beam intensity                                                |        |                    |
| Method                           | Stepped flux                                                        | Decay data                                                                      |        |                    |
| Stack composition                | 147-149 Sm-AI, repeated 27 times                                     | Reaction Q-values                                                               |        |                    |
| Target and thickness             | 145 Sm foil, 25.14, 23.13, 24.20 mm                                   | Determination of beam energy                                                    |        |                    |
| Number of target foils           | 27                                                                  | Uncertainty of energy                                                           |        |                    |
| Isotopic abundance               | 145-149 Sm-AI                                                        | Uncertainty of energy                                                           |        |                    |
| Accelerator                      | Cyclotron 90 cyclotron of the Université Catholique in Louvain la Neuve (LLN) | Cross sections                                                                 |        |                    |
| Primary energy                   | 50 MeV                                                              | Uncertainty of cross sections                                                  |        |                    |
| Irradiation time                 | 66 min                                                               | Yield                                                                           |        |                    |
| Beam current                     | 120 nA                                                              |                                                                                  |        |                    |
| Monitor reactions [recommended values] | 147 Al(α,4n) + 109 Ag(α,4n) reactions                               |                                                                                  |        | Tarkanyi et al. 2001 |
| Monitor target and thickness     | 147-149 Sm foil, 10.9 mm                                            |                                                                                  |        |                    |
| Detector                         | HPGe                                                                |                                                                                  |        |                    |
| Cooling times                    | 7.5, 10.5, 9.5, 12.5 h                                              |                                                                                  |        |                    |

#### 4.1.2. 145Sm (cumulative)

The cumulative cross sections for 145Sm (T1/2 = 340 d) production are shown in Fig. 3. In this experiment 145Sm is produced directly via (d,pxn) reactions and from decay of the shorter-lived 145Eu (T1/2 = 5.93 d) discussed above. The cross sections were deduced from spectra measured after 5 half-lives of the 145Eu parent decaying completely with EC to 145Sm (97 %). The missing part was corrected on the basis of the measured 145Eu cross sections. As stated earlier, the possible contribution from the long-lived 145Pm (T1/2 = 17.7 a) can be ignored. The first peak of the excitation function at 10 MeV is the sum of the 144Sm(d,n) and 144Sm(d,p) reactions. The higher energy part is the sum of 147−150Sm(d,n) and 147−150Sm(d,p3-6n) reactions. There is a good agreement between the cumulative production of 145Sm and the results of TENDL-2012 at higher energies. At energies below 15 MeV, as a result of the known underestimation of (d,p) [Hermanne et al. 2009] and compensation by overestimation of (d,n) reaction, the TENDL discrepancy is finally only a few percent. In case of ALICE-D and EMPIRE-D at low energies the description of the (d,p) is better, but the high energy part is significantly overestimated.

![Figure 3: Experimental and theoretical excitation functions for Sm(d,x)145Sm](image-url)
Table 2: Decay characteristics of the investigated activation products and Q-values of contributing reactions

| Nuclide | Decay path | Half-life | Eγ (MeV) | Iγ (%) | Contributing reaction | Q-value (MeV) |
|---------|------------|-----------|----------|--------|-----------------------|---------------|
| 145Eu   | ε: 100 %   | 5.93 d    | 653.512  | 1658.53| 144Sm(d,n)            | -1008.5       |
|         |            |           |          |        | 147Sm(d,4n)           | -20424.02     |
|         |            |           |          |        | 148Sm(d,5n)           | -28565.4      |
|         |            |           |          |        | 149Sm(d,6n)           | -34435.74     |
|         |            |           |          |        | 150Sm(d,7n)           | -42422.43     |
|         |            |           |          |        | 152Sm(d,9n)           | -56276.58     |
|         |            |           |          |        | 154Sm(d,11n)          |               |
| 145Sm   | ε: 100 %   | 340 d     | 61.2265  | 12.15  | 144Sm(d,p)            | 4532.534      |
|         |            |           |          |        | 147Sm(d,p3n)          | -16981.99     |
|         |            |           |          |        | 148Sm(d,p4n)          | -25123.37     |
|         |            |           |          |        | 149Sm(d,p5n)          | -30993.72     |
|         |            |           |          |        | 150Sm(d,p6n)          | -38980.41     |
|         |            |           |          |        | 152Sm(d,p8n)          | -52834.55     |
|         |            |           |          |        | 145Eu decay           | -1090.5       |
|         |            |           |          |        | 145Pm decay           | -15583.55     |
| 153Sm   | β−: 100 %  | 46.28 h   | 103.18012| 29.25  | 152 Sm(d,p)           | 3643.834      |
|         |            |           |          |        | 154Sm(d,2pn)          | -10191.36     |
|         |            |           |          |        | 153Pm decay           | -11320.47     |

When complex particles are emitted instead of individual protons and neutrons the Q-values have to be decreased by the respective binding energies of the compound particles: np-d, +2.2 MeV; 2np-t, +8.48 MeV; n2p-3He, +7.72 MeV; 2n2p-a, +28.30 MeV.

4.1.3. 153Sm

The radioisotope 153Sm (T₁/₂ = 46.50 h) is formed directly by the 152Sm(d,p) and 154Sm(d,2pn) reactions and from the decay of short-lived 153Pm (T₁/₂ = 5.25 min). The experimental and theoretical results are shown in Fig. 4. The 153Pm is produced via the 154Sm(d,2pn) low cross section reaction. The TENDL-2012 data underestimate the (d,p) reaction part and overestimate the (d,p2n). In case of ALICE-D and EMPIRE-D the agreement for low energy (d,p) part is good, but the (d,p2n) is underestimated.

4.2. Thick target yields

Thick target yields (integrated yield for a given incident energy down to the reaction threshold) were calculated from curves fitted to our experimental cross section data. The results for physical yields (Bonardi, 1987) are presented in Fig. 5. No earlier experimental thick target yield data were found in the literature.

5. Comparison of the production routes

The two radio-lanthanides discussed (145,153Sm) can be produced via various reactions. Among these competitive production routes we discuss below only the
Table 3: Measured cross sections of the $^{145}$Eu and $^{145,153}$Sm reactions and estimated uncertainties

| Energy (MeV) | $^{145}$Eu (mb) | $^{153}$Sm (mb) | $^{145}$Eu (mb) | $^{153}$Sm (mb) |
|--------------|----------------|----------------|----------------|----------------|
| 10.0         | 0.00           | 0.00           | 0.00           | 0.00           |
| 10.4         | 0.50           | 0.50           | 0.50           | 0.50           |
| 10.8         | 0.50           | 0.50           | 0.50           | 0.50           |
| 11.2         | 0.50           | 0.50           | 0.50           | 0.50           |
| 11.6         | 0.50           | 0.50           | 0.50           | 0.50           |
| 12.0         | 0.50           | 0.50           | 0.50           | 0.50           |
| 12.4         | 0.50           | 0.50           | 0.50           | 0.50           |
| 12.8         | 0.50           | 0.50           | 0.50           | 0.50           |
| 13.2         | 0.50           | 0.50           | 0.50           | 0.50           |
| 13.6         | 0.50           | 0.50           | 0.50           | 0.50           |
| 14.0         | 0.50           | 0.50           | 0.50           | 0.50           |
| 14.4         | 0.50           | 0.50           | 0.50           | 0.50           |
| 14.8         | 0.50           | 0.50           | 0.50           | 0.50           |
| 15.2         | 0.50           | 0.50           | 0.50           | 0.50           |
| 15.6         | 0.50           | 0.50           | 0.50           | 0.50           |
| 16.0         | 0.50           | 0.50           | 0.50           | 0.50           |
| 16.4         | 0.50           | 0.50           | 0.50           | 0.50           |
| 16.8         | 0.50           | 0.50           | 0.50           | 0.50           |
| 17.2         | 0.50           | 0.50           | 0.50           | 0.50           |
| 17.6         | 0.50           | 0.50           | 0.50           | 0.50           |
| 18.0         | 0.50           | 0.50           | 0.50           | 0.50           |
| 18.4         | 0.50           | 0.50           | 0.50           | 0.50           |
| 18.8         | 0.50           | 0.50           | 0.50           | 0.50           |
| 19.2         | 0.50           | 0.50           | 0.50           | 0.50           |
| 19.6         | 0.50           | 0.50           | 0.50           | 0.50           |
| 20.0         | 0.50           | 0.50           | 0.50           | 0.50           |

low energy, light charged particle induced routes, presented in Table 4 in more detail ($E_{particle} < 100$ MeV). Table 4 does not contain the $^3$He induced reactions from practical reasons, taking into account the availability and cost of the high intensity $^3$He irradiations. The related excitation functions are shown in Figs. 6-11. Except for the $^{nat}Sm(p,x)^{145}Eu,^{145}Sm$ reaction, investigated by Blue et al. (Blue, 1988) (yields) and the present data on deuterons, no experimental data are available for cross sections and yields for charged particle production routes. The $(n,y)$ cross sections and yields were taken from [Mizadze et al. 1992, Pritychenko and Mughabghab 2012]. For comparison of production routes of charged particle induced reactions we have used TTY (thick target yield) derived from the theoretical data of TENDL, ALICE and EMPIRE which are normalized to the measured experimental data, in case of existing experimental data. Otherwise, the theoretical data were selected after comparing with the systematics of the experimental data in the same mass region. As for many practical applications, also for medical isotope production there are several factors determining the optimal production route. The first group of factors is connected to requirements in the patient related application: level of specific activity, radionuclidic purity, carrier added or carrier free product, large scale routine application, small scale research work route. The second group of factors is connected to the technology used: minimal required production yields, natural or highly enriched targets, target mass, target cooling problems, recovery and radioactive waste problems, the available beam properties (particle type, current, energy), possibility of parallel irradiations, etc. The third group of factors is determined by the continuously changing day to day requirements: the new competing radio-products and application methods, the new regulations and trend in nuclear technology. Nowadays the largest part of routinely used therapeutic radioisotopes is produced at nuclear research reactors. There is however a significant effort to transfer the production to accelerators by improving beam characteristics and production technology and/or introducing new accelerator produced isotopes with characteristics similar to reactor products.

5.1. Production of $^{145}$Sm

According to Table 4, the $^{145}$Sm(n,$\gamma$)$^{145}$Sm production route is the method of choice from point of view of yield and irradiation technology if a carrier added, relatively low specific activity $^{145}$Sm end product is satisfactory. The route to get carrier free, high specific activity $^{145}$Sm is possible only with accelerators through the decay of $^{145}$Eu or through the direct production from alpha induced nuclear reactions on natural or enriched neodymium. Indirect production through $^{145}$Eu obtained from irradiation of Sm targets requires two chemical separations, while in the case of direct production one chemical separation is needed to produce a high-specific-activity and high radionuclidic purity $^{145}$Sm end product. Out of them the $^{147}$Sm(p,3n)$^{145}$Eu-145Sm and the $^{147}$Sm(p,x)$^{145}$Sm are the most productive ($^{147}$Sm 15% in natural) and requires a 30 MeV cyclotron (or accelerator). The product is contaminated with long lived $\alpha$-emitter $^{146}$Sm, but with very low activity. The radionuclidic impurity is lower in the case
of the $^{144}\text{Sm}(d,n)^{145}\text{Eu}-^{145}\text{Sm}$ ($^{144}\text{Sm}$ 3.1%, low energy irradiation to avoid (d,3n) and contamination with long-lived $^{143}\text{Pm}$) and $^{142}\text{Nd}(\alpha,n)^{145}\text{Sm}$ reactions (low energy to avoid ($\alpha,2n$) and ($\alpha,3n$)). In case of other p-, d-, and $\alpha$ induced reactions the simultaneously produced $^{146,147}\text{Sm}$ do not cause serious impurity due to their extremely long half-lives. In all cases highly enriched targets are required. The excitation functions for the main reactions based on our ALICE-IPPE, EMPIRE results and on data in TENDL-2012, are shown in Figs. 6-9.

5.2. Production of $^{153}\text{Sm}$

According to the Table 4, it is difficult to compete in efficiency with the production of $^{153}\text{Sm}$ in reactors due to the large production cross section and also because the products of the proton and deuteron induced reactions are carrier added. The only way to get a no carrier added product is through the low yield $^{150}\text{Nd}(\alpha,n)^{153}\text{Sm}$ reaction (5.1% abundance). In all cases highly enriched targets are required. The proton and deuteron induced reactions can however have importance for local use due to the relatively short half-life of $^{153}\text{Sm}$ ($T_{1/2} = 46.50$ h) and due to the unused beam time of charged particle accelerators. The production requires highly enriched targets. The cross sections of the most important reactions are shown in Figs. 10-12.

6. Summary and conclusion

From a series of radioisotopes produced in deuteron induced nuclear reactions on natural samarium the medically important $^{145}\text{Sm}$ and $^{153}\text{Sm}$ as well as $^{145}\text{Eu}$ as
Table 4: Production routes of $^{145}$Sm and $^{153}$Sm

| Nuclide | Reaction | $\sigma_{\text{max}}$ (b) | Energy range (MeV) | Yield (MBq/μg) | Reference |
|---------|----------|--------------------------|-------------------|----------------|-----------|
| $^{145}$Sm | $^{144}$Sm($n,\gamma$)$^{145}$Sm | 1.64 | thermal | 200 MBq/μg* | Mirzadeh et al. [1992] |
| $^{145}$Sm | $^{144}$Sm($d,n)$- $^{145}$Eu-$^{145}$Sm | 0.26 | 20-8 | 32 | this work (exp) |
| $^{144}$Sm | $^{145}$Sm($d,x$)$^{145}$Sm | 0.68 | 20-8 | 97 | this work (exp) |
| $^{147}$Sm | $^{144}$Sm($d,4n$)$^{145}$Eu-$^{145}$Sm | 0.89 | 50-25 | 713 | TENDL 2012 |
| $^{147}$Sm | $^{145}$Sm($d,x$)$^{145}$Sm | 0.99 | 50-25 | 914 | TENDL 2012 |
| $^{147}$Sm | $^{144}$Sm($p,3n$)$^{145}$Eu-$^{145}$Sm | 1.05 | 50-20 | 64854 | this work (EMPIRE) |
| $^{147}$Sm | $^{145}$Sm($p,x$)$^{145}$Sm | 1.18 | 50-20 | 81906 | this work (EMPIRE) |
| $^{142}$Nd | ($\alpha,n$)$^{145}$Sm | 0.31 | 24-14 | 289 | this work (ALICE) |
| $^{143}$Nd | ($\alpha,2n$)$^{145}$Sm | 1.05 | 35-19 | 2500 | this work (ALICE) |
| $^{153}$Sm | $^{152}$Sm($n,\gamma$)$^{153}$Sm | 206 | thermal | 105 MBq/μg* | Mirzadeh et al. [1992] |
| $^{153}$Sm | $^{152}$Sm($d,p$)$^{153}$Sm | 0.25 | 24-7 | 10057 | this work (exp + EMPIRE) |
| $^{154}$Sm | ($d,p2n$)$^{153}$Sm | 0.3 | 50-20 | 54639 | this work (exp + EMPIRE) |
| $^{154}$Sm | ($d,pn$)$^{153}$Sm | 0.14 | 100-20 | 145550 | this work (EMPIRE) |
| $^{150}$Nd | ($\alpha,n$)$^{153}$Sm | 0.020 | 40-15 | 60 | this work (EMPIRE) |

* irradiation time = half-life; neutron flux = 1.10$^{14}$n/cm$^2$/s; 100 % enriched target
# irradiation time = 180 d
parent of $^{145}$Sm were selected for detailed study. Experimental excitation functions of both isotopes were accurately determined and compared with the results of three nuclear reaction codes. The agreement with the theoretical results and the present experiments was discussed for each isotope separately, because the different codes gave different quality estimations in the case of the above three radioisotopes. From the measured excitation functions yield curves were also calculated in the investigated energy range. Since both samarium isotopes are important for medical applications, a comparison was also made between all possible routes. It turned out, as expected, that the neutron induced production in reactors is the most economical way for mass production of these isotope in carrier added form. However, it should be mentioned that for no carrier added end product and small scale or research production charged particle induced reactions can be used as an alternative.

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