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Activation cross section measurement of alpha-particle induced reactions on natural neodymium

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Abstract

Excitation functions of alpha-particle induced reactions on ^{nat}Nd up to 50 MeV were measured at the RIKEN AVF cyclotron. To derive cross sections activation method, stacked target technique and gamma-ray spectrometry were adopted. Formations of ^{153,145}Sm, ^{151,150,149,148m,148g,144,143}Pm, and ^{149,147}Nd were investigated. The results were compared with the previous experimental data and the TENDL-2019 data. Discrepancies among most of them were found.

Keyword

Samarium-153, Samarium-145, Alpha irradiation, Neodymium target, Cross section

1. Introduction

Radionuclides of samarium have been studied for wide-range applications such as nuclear medicine. ¹⁵³Sm ($T_{1/2} = 46.3$ h) is a beta- and gamma-emitter, which can provide a relatively high dose-rate. This radionuclide is used for treatment of bone metastases as the EDTMP (ethylenediamino-tetrakis-methylenediphosphonic acid) chelate (Finlay et al., 2005). ¹⁴⁵Sm has a long half-life of about one year ($T_{1/2} = 340$ d) and emits low-energy X-rays. It was suggested that these properties can be applied for brachytherapy (Fairchild et al., 1987).

¹⁵³Sm and ¹⁴⁵Sm are generally obtained via neutron capture reactions on enriched ¹⁵²Sm and ¹⁴⁴Sm, respectively, in nuclear reactors (International Atomic Energy Agency, 2011; Uddin et al., 2014, 2008). However, the specific activity of the ¹⁵³Sm produced in reactors by the neutron capture reaction is rather low (Qaim et al., 2007; Tárkányi et al., 2014). For practical use of ¹⁵³Sm and ¹⁴⁵Sm in radiotherapy, other production ways with relatively high specific activities are required.

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These radionuclides can be generated in alpha-particle induced reactions on natural neodymium. Nevertheless, there has been only one previous experimental study for the $^{nat}\text{Nd}(\alpha,x)^{153}\text{Sm}$ reaction up to 26.2 MeV (Qaim et al., 2007) and no previous experimental study for the $^{nat}\text{Nd}(\alpha,x)^{145}\text{Sm}$ reaction was found in our literature survey based on the EXFOR library (Otuka et al., 2014). Therefore, we decided to perform an experiment on the $^{nat}\text{Nd}(\alpha,x)$ reactions up to the alpha-particle energy of 51 MeV.

2. Experimental

The experiment was performed at the RIKEN AVF cyclotron. The standard stacked-foil activation technique and gamma-ray spectrometry were adopted for this experiment. The target consisted of 21 ^{nat}Nd foils (purity: 99.0%, thickness: 16.68 mg/cm²; Goodfellow Co., Ltd., UK) and 14 ^{nat}Ti foils (purity: 99.6%, thickness: 2.35 mg/cm²; Nilaco Corp., Japan). The average thicknesses of these foils were derived by measuring their weights and surface areas. Those target foils were arranged in Nd-Nd-Nd and Ti-Ti pairs in the stack to handle the recoil effect. As the activity loss due to the recoil effect changes slowly with the particle energy we can suppose that the low energy foils of the pairs are recoil compensated. Namely, the same amount of reaction products was implanted into and recoiled out of the foil. Accordingly, only 14 Nd and 7 Ti foils, which correspond to the lower particle energy ones of each set, were used in the data analysis. The ^{nat}Ti foils were placed in the stack to monitor the beam parameters by using the $^{nat}\text{Ti}(\alpha,x)^{51}\text{Cr}$ monitor reaction and to monitor the energy loss of the beam throughout the target. The stacked target was irradiated with an alpha-particle beam in a target holder served as a Faraday cup for 1 h. The initial beam energy was measured by the time-of-flight method (Watanabe et al., 2014) and the average beam intensity was measured by charge integration on the Faraday cup. The measured initial values were 51.1 MeV and 172 nA, respectively. The beam spot was collimated down to 3 mm in diameter which is almost the same size of the gamma-ray calibration source. The beam energy degradation through the target stack was calculated by using the SRIM code (Ziegler et al., 2008).

After a cooling time of approximately 30 min, we disassembled the stacked target and the gamma-ray spectrometry of the activated foils was started. The gamma-ray spectra were acquired only for the recoil compensated downstream foils by an HPGe detector (ORTEC GMX30P4-70) without chemical separation. The recorded spectra were analyzed by the Gamma Studio (SEIKO EG&G) software. Determination of the geometry dependent detector efficiency was performed using a standard gamma-ray point-like source composed of a mixture of multiple radioisotopes $^{57,60}\text{Co}$, ^{85}Sr , ^{88}Y , ^{109}Cd , ^{113}Sn , ^{137}Cs , ^{139}Ce , ^{203}Hg and ^{241}Am . Measurements were performed several times applying different waiting times (0.7 h – 4.0 d) to follow the decay of the produced radionuclides. The distances between the foils and the detector were adjusted to keep the dead time less than 5%. The nuclear data of reaction products in this experiment were taken from the NuDat 2.8 data library (National Nuclear Data Center, 2019) and Qcalc (Pritychenko and Sonzogni, 2003), which are listed in Table 1.

The following equation was used to derive the cross section σ (cm²),

$$\sigma = \frac{\lambda}{nI_{\alpha}} \frac{C_{\gamma}}{\varepsilon_d \varepsilon_{\gamma} \varepsilon_t (1 - e^{-\lambda T_b}) e^{-\lambda T_c} (1 - e^{-\lambda T_m})}$$

where λ is the decay constant of each radionuclide (s⁻¹), n is the surface density of target atoms (cm⁻²), I_{α} is the alpha-particle-beam intensity (s⁻¹), C_{γ} is the net peak area of the gamma-line counts, ε_d is the detector efficiency,

ε_γ is the gamma-ray abundance, ε_t is the dead time correction, T_b is the irradiation time (s), T_c is the cooling or waiting time (s) and T_m is the acquisition time (s).

We investigated the whole excitation function for the $^{nat}\text{Ti}(\alpha,x)^{51}\text{Cr}$ monitor reaction up to 51 MeV. In case of properly measured beam intensity and initial energy all the experimentally determined cross section should fit with the recommended curve. If deviation is observed in the amplitude, the beam intensity can be adjusted for proper amplitude agreement. For deviations in the energy scale we have two options to tune the experimental parameters. One can fine-tune the initial bombarding energy or the thickness of the target foils. As the beam energy was determined accurately by the time-of-flight method the 51.1 ± 0.1 MeV initial particle energy was adopted. The minor correction in the energy scale calculated by using the SRIM code was possible by adjusting the thickness of the target foils. Since the ^{nat}Nd foils were about five times thicker than the ^{nat}Ti foils and the thickness of the Ti foils was determined with higher accuracy, we only adjusted the thickness of the ^{nat}Nd foils. Reducing the average thickness of the Nd foils, within its 2% uncertainty by 1.5% to 16.43 mg/cm^2 proper agreement was found for the energy scale. The cross sections derived for the monitor reaction are shown in Fig. 1 together with the IAEA recommended excitation function (Hermanne et al., 2018). As good agreement between our result and the recommended values was found no additional adjustment other than the thickness of the Nd foils was adopted for the data analysis to determine the cross sections of the $^{nat}\text{Nd}(\alpha,x)$ reactions.

The calculated energy thickness of the Nd foils is changing from 0.6 to 1.4 MeV. The total uncertainty of the energy associated with the Nd foils is increasing as the beam penetrated the stack from 0.2 MeV to 1.2 MeV, which were derived from the initial uncertainty of the incident projectile energy, the uncertainty of the foil thicknesses and stopping powers. The total uncertainty of the cross sections (8–28.6%) were derived as the square root of the quadratic sum of the propagating uncertainties: beam intensity (5%), gamma intensity (<5.8%), detector efficiency (5%), target thickness (1-2%), target purity (1%) and counting statistics (0.2–27.0%) of the net peak area of the particular gamma-ray.

3. Results and discussion

Activation cross sections for 11 radionuclides $^{153,145}\text{Sm}$, $^{151,150,149,148m,148g,144,143}\text{Pm}$, and $^{149,147}\text{Nd}$ were determined. The results including uncertainties are listed in Tables 2-4 for each radionuclide and graphically shown in Figs. 2-12 compared with the TENDL-2019 data (Koning et al., 2019) and previous experimental data if available. The TENDL-2019 values for enriched Nd targets were normalized using the natural abundance of neodymium. By fine tuning the input parameters one could estimate much properly the experimental data using the TALYS code. As we wanted to test the quality of the data in the TENDL-2019 library, which are widely used by research groups, we did not perform additional calculations with adjusted input parameters. Tables 2-4 present the mean energy with its uncertainty followed by the energy thickness of the corresponding foil in parentheses and the cross section followed by its total uncertainty.

3.1 The $^{nat}\text{Nd}(\alpha,x)^{153}\text{Sm}$ reaction

The production cross sections of ^{153}Sm ($T_{1/2} = 46.50$ h) were derived from the 103.18-keV interference free gamma-line ($I_\gamma = 29.25\%$) after a waiting time of 1.4-4.0 days. Since the detected gamma-ray energy was relatively low, we considered its attenuation in the metallic Nd foil. The attenuation rate was calculated using the X-ray mass attenuation coefficients (Hubbel and Seltzer, 2004) and the derived cross sections were corrected by +2.1% due to self-absorption effect. Only the $^{150}\text{Nd}(\alpha,n)^{153}\text{Sm}$ reaction contributes directly to the formation of ^{153}Sm . Indirect formation is also possible by decay of ^{153}Pm with much shorter half-life ($T_{1/2} = 5.25$ min), therefore the cross sections shown in Fig. 2 are cumulative and compared with the data of the only earlier work (Qaim et al., 2007) and the TENDL-2019 values (Koning et al., 2019). The earlier experimental data are not consistent with our experimental data. The TENDL data underestimate both experimental results. Qaim et al. (Qaim et al., 2007) used oxide powder targets ($^{nat}\text{Nd}_2\text{O}_3$), sedimentation method to prepare the samples, the usual stacked-foil technique and gamma-ray spectrometry. Although the half-life 1.95 d and the 103.2-keV gamma-line ($I_\gamma = 28.3\%$) they used for the analysis are slightly different from the nuclear data we adopted but that deviation cannot explain the observed difference in the cross sections.

3.2 The $^{nat}\text{Nd}(\alpha,x)^{145}\text{Sm}$ reaction

The long lived ^{145}Sm radionuclide ($T_{1/2} = 340$ d) can be produced in (α,xn) reactions on the stable isotopes of $^{142,143,144,145,146}\text{Nd}$. Major contribution can be expected from the (α,n) and the $(\alpha,3n)$ reactions on the relatively abundant isotopes of ^{142}Nd and ^{144}Nd , respectively. Practically only one gamma-line, which can be considered as interference free, was used to determine the activity of ^{145}Sm . Gamma-ray measurements at $E_\gamma = 61.2265$ keV ($I_\gamma = 12.15\%$) were performed to derive the cross sections for the $^{nat}\text{Nd}(\alpha,x)^{145}\text{Sm}$ reaction after a cooling time of 1.4-4.0 days. Considering the low energy of this gamma-line the derived cross sections were corrected by +8.4% because of the attenuation of the gamma intensity due to the self-absorption effect. The results are shown in Fig. 3 in comparison with the TENDL-2019 data (Koning et al., 2019). The TENDL-2019 theoretical values are almost consistent with our experimental data. No data published earlier were found in the literature survey.

3.3. The $^{nat}\text{Nd}(\alpha,x)^{151}\text{Pm}$ reaction

The radionuclide ^{151}Pm ($T_{1/2} = 28.40$ h) can be formed on the ^{148}Nd and ^{150}Nd target isotopes. It was identified and analyzed by using the gamma-line at $E_\gamma \approx 340.1$ keV, which is the sum of the $E_\gamma = 340.08$ keV ($I_\gamma = 22.5\%$) and the $E_\gamma = 341.0$ keV ($I_\gamma = 0.074\%$) gamma-lines. In principle decay contribution from decay of the short lived ^{151}Nd ($T_{1/2} = 12.44$ min) is possible above 16 MeV therefore the deduced cross sections are considered as cumulative ones. The cooling time was 1.4-4.0 days and the dead time was less than 2%. The result is shown in Fig. 4 with the TENDL-2019 data (Koning et al., 2019). The theoretical values underestimate the experimental data in the whole investigated energy region. No data published earlier were found in the literature survey.

3.4. The $^{nat}\text{Nd}(\alpha,x)^{150}\text{Pm}$ reaction

The radionuclide ^{150}Pm can be formed only in reactions on the ^{148}Nd and ^{150}Nd target isotopes. For formation of this radionuclide no contribution by decay is possible. The production cross sections of ^{150}Pm ($T_{1/2} = 2.698$ h) were

measured using the interference free gamma-line at $E_\gamma = 333.92$ keV ($I_\gamma = 68\%$). The applied cooling time was about 0.7-3.4 hours due to its short half-life and the dead time was 0.6-4.3%. Figure 5 shows our experimental result in comparison with the TENDL-2019 values (Koning et al., 2019). The TENDL data underestimate our experimental data in the measured energy region. No data published earlier were found in the literature survey.

3.5. The $^{nat}\text{Nd}(\alpha,x)^{149}\text{Pm}$ reaction

Reactions on the $^{146,148,150}\text{Nd}$ stable target isotopes can contribute to the formation of the ^{149}Pm radionuclide. Indirect formation by decay of ^{149}Nd is also possible. Cross sections of the $^{nat}\text{Nd}(\alpha,x)^{149}\text{Pm}$ process were determined by using the net peak area of the interference free gamma-ray at $E_\gamma = 285.95$ -keV ($I_\gamma = 3.10\%$). The applied cooling time was more than 1.4 days. Due to the short half-life of the parent radionuclide ^{149}Nd ($T_{1/2} = 1.728$ h), cumulative cross sections were deduced. The result is shown in Fig. 6 in comparison with the TENDL-2019 data (Koning et al., 2019). The theoretical values show a peak at around 28 MeV, however, the derived cross sections increase monotonically with the energy. Beside the shape difference the TENDL data underestimate our experimental result above 35 MeV. No data published earlier were found in the literature survey.

3.6. The $^{nat}\text{Nd}(\alpha,x)^{148m}\text{Pm}$ reaction

The radionuclide ^{148m}Pm can be formed on the target isotopes of $^{145,146,148,150}\text{Nd}$, accordingly several independent reaction may contribute to the activation cross section of the $^{nat}\text{Nd}(\alpha,x)^{148m}\text{Pm}$ process. Contribution from decay is not possible for this case. The production cross sections of ^{148m}Pm ($T_{1/2} = 41.29$ d) were derived by using the $E_\gamma = 629.97$ -keV gamma-line ($I_\gamma = 89.0\%$). Gamma-spectra collected after a cooling time longer than 1.4 days were used for data analysis, for which the associated dead time was less than 2%. The applied long cooling time assured total decay of ^{149}Nd ($T_{1/2} = 1.728$ h) with decay gamma at 630.237 keV within the detector resolution. The cross sections obtained from the interference free net counts of the gamma-line are shown in Fig. 7 in comparison with the experimental data measured on ^{146}Nd enriched targets (Aumann and Guckel, 1977) normalized to the natural abundance (17.19%), and the TENDL-2019 values (Koning et al., 2019). The deviation from the ^{146}Nd enriched target data shows contribution from the other target isotopes. The theoretical calculation remarkably underestimates the population of this high spin meta-stable state by comparing to our experimental result.

3.7. The $^{nat}\text{Nd}(\alpha,x)^{148g}\text{Pm}$ reaction

The half-life of the ground state of the ^{148g}Pm ($T_{1/2} = 5.368$ d) is shorter than the half-life of the ^{148m}Pm ($T_{1/2} = 41.29$ d) meta-stable state. Beside direct formation the ground state also populated by IT decay (4.2%) of the ^{148m}Pm meta-stable state, while the β^- decay of ^{148m}Pm (95.8%) populates the same ^{148}Sm daughter nuclei and may result in the same gamma-lines as decay of ^{148g}Pm . For data assessment the net peak area of the $E_\gamma = 1465.12$ -keV independent gamma-line ($I_\gamma = 22.2\%$) was used from the gamma spectra collected after a cooling time of 1.4-4.0 days. As decay of ^{148m}Pm does not populate every energy levels of the ^{148}Sm daughter nuclei this gamma-line can be considered interference free for ^{148g}Pm . To derive the production cross sections of ^{148g}Pm only the IT decay contribution of ^{148m}Pm had to be subtracted. By knowing the formation cross section of ^{148m}Pm its IT decay contribution can be calculated and subtracted. The independent cross sections were thus derived.

Figure 8 shows our experimental data together with the data normalized from those measured on ^{146}Nd targets (Aumann and Guckel, 1977) and the TENDL-2019 values (Koning et al., 2019). The comparison of the experimental data shows that the normalized data measured on ^{146}Nd targets (Aumann and Guckel, 1977) are only a partial contribution of the data can be measured on natural neodymium targets. The TENDL estimation provides different shape for the excitation function, a monotone increasing curve, while our data exhibit a plateau above 35 MeV.

3.8. The $^{\text{nat}}\text{Nd}(\alpha, x)^{144}\text{Pm}$ reaction

The radionuclide ^{144}Pm can be produced on all stable isotopes of Nd. Due to the large number of contributing individual reactions with different threshold energies the expected excitation function is a monotone increasing curve without strong structure. ^{144}Pm has a half-life of 363 days and accordingly the gamma-ray measurements were started after a long cooling time. For data assessment the gamma-line at $E_{\gamma} = 696.49$ keV ($I_{\gamma} = 99.49\%$) was selected. This gamma-line can be considered as an interference free gamma-line, although ^{149}Nd has a similar energy decay radiation, but due to its much shorter half-life ($T_{1/2} = 1.728$ h) it did not contribute to the peak at the time of the measurement. The possible co-production of ^{144}Pr did not contribute to the peak since its half-life ($T_{1/2} = 17.28$ h) is much shorter and by the time the gamma-measurements were started ^{144}Pr decayed completely. The derived cross sections of the $^{\text{nat}}\text{Nd}(\alpha, x)^{144}\text{Pm}$ process are shown in Fig. 9 in comparison with the TENDL-2019 data (Koning et al., 2019). The theoretical excitation function exhibits some structure and the predicted curve slightly overestimate the experimental result at lower energy region, whereas underestimates above 40 MeV. No data published earlier were found in the literature survey.

3.9. The $^{\text{nat}}\text{Nd}(\alpha, x)^{143}\text{Pm}$ reaction

The radionuclide ^{143}Pm can be formed in principle on all stable isotopes of natural Nd except for ^{150}Nd and also by decay of the co-produced short-lived $^{143\text{m.g}}\text{Sm}$. The half-life of ^{143}Pm is 265 days and gamma-spectra collected after the longest cooling time were used for data analysis. The cross sections of the $^{\text{nat}}\text{Nd}(\alpha, x)^{143}\text{Pm}$ process were determined based on measurements of the $E_{\gamma} = 741.98$ -keV ($I_{\gamma} = 38.5\%$) gamma-line. The possible co-production of ^{143}Pr did not contribute to the peak because of the negligible gamma intensity of $E_{\gamma} = 742.10$ keV ($I_{\gamma} = 1.2 \times 10^{-6}\%$). Contribution from decay of ^{143}Sm is included, therefore our experimental data are cumulative cross sections and shown in Fig. 10 and compared with the TENDL-2019 data (Koning et al., 2019). The TENDL data are roughly consistent with our result. No data published earlier were found in the literature survey.

3.10. The $^{\text{nat}}\text{Nd}(\alpha, x)^{149}\text{Nd}$ reaction

The radionuclide ^{149}Nd is formed only on the $^{148,150}\text{Nd}$ target isotopes directly and by decay of ^{149}Pr indirectly. Considering the half-life of ^{149}Nd ($T_{1/2} = 1.728$ h), the first series of gamma spectra with few hours cooling time were used for data analysis. The most intense gamma-line at $E_{\gamma} = 211.309$ keV ($I_{\gamma} = 25.9\%$) from the ^{149}Nd decay was selected to derive the cross sections of the $^{\text{nat}}\text{Nd}(\alpha, x)^{149}\text{Nd}$ process. The obtained cross sections are cumulative as they include decay contribution of ^{149}Pr . Our results are compared with the TENDL-2019 data (Koning et al., 2019) as shown in Fig. 11. Their behaviors are completely different from each other in the whole energy region. No data

published earlier were found in the literature survey.

3.11. The $^{nat}\text{Nd}(\alpha, x)^{147}\text{Nd}$ reaction

The radionuclide ^{147}Nd ($T_{1/2} = 10.98$ d) is formed on the target isotopes of $^{145,146,148,150}\text{Nd}$. Formation by decay of ^{147}Pr ($T_{1/2} = 13.4$ min) is also possible. Spectra collected after a few days of waiting time were used for data analysis. The complete decay of ^{147}Pr can be considered by the time the gamma-measurements were started. The excitation function of the $^{nat}\text{Nd}(\alpha, x)^{147}\text{Nd}$ reaction was determined using the most intense gamma-line at $E_\gamma = 91.105$ keV ($I_\gamma = 28.1\%$). Several other co-produced isotopes have similar energy gamma-lines, but fortunately their half-lives are shorter and/or their gamma-intensity are much lower, therefore their effect on the ^{147}Nd decay gamma count is negligible. The derived cumulative cross sections were corrected by + 3.0% considering the attenuation of the low energy gamma-photons in the Nd target foil due to the effect of self-absorption. The results are shown in Fig. 12 together with the TENDL-2019 values (Koning et al., 2019). No data published earlier were found in the literature survey.

4. Summary

Activation cross sections of the alpha-particle induced reactions on ^{nat}Nd were measured up to 50 MeV at the RIKEN AVF cyclotron. We adopted the stacked-foil activation technique and gamma-ray spectrometry in this experiment. The excitation functions for 11 radioisotopes $^{153,145}\text{Sm}$, $^{151,150,149,148m,148g,144,143}\text{Pm}$, and $^{149,147}\text{Nd}$ were determined and compared with data published earlier and the TENDL-2019 data. Cross sections for formation of the ^{145}Sm , $^{151,150,149,148m,148g,144,143}\text{Pm}$, and $^{149,147}\text{Nd}$ were measured for the first time. The ^{153}Sm production cross section determined by us is in conflict with the cross section measured by Qaim et al. (2007). Our measured cross sections are considerably smaller than those of Qaim et al. (2007), accordingly the expected production yield should be also less than the predicted values by Qaim et al (2007). Due to lack of the detailed information on the experimental and analytical procedures, the observed difference was not explained satisfactorily. An independent confirmation is necessary to solve the situation. TENDL-2019 predicts our ^{145}Sm production cross section rather well, but the agreement with our cross sections is not satisfactory for other productions in general.

Declaration of Competing Interest

The authors have no conflicts of interest in this paper.

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References

Aumann, D.C., Guckel, W., 1977. Absolute cross sections and isomeric cross-section ratios for the $^{148}\text{Nd}(d,2n)$,

$^{148}\text{Nd}(p,n)$, and $^{146}\text{Nd}(\alpha,pn)$ reactions producing the isomeric pair $^{148}\text{Pm}^{\text{m.g}}$. *Phys. Rev. C* 16, 160–166.

<https://doi.org/10.1103/PhysRevC.16.160>

Fairchild, R.G., Kalef-Ezra, J., Packer, S., Wielopolski, L., Laster, B.H., Robertson, J.S., Mausner, L., Kanellitsas, C., 1987. Samarium-145: A new brachytherapy source. *Phys. Med. Biol.* 32, 847–858. <https://doi.org/10.1088/0031-9155/32/7/005>

Finlay, I.G., Mason, M.D., Shelley, M., 2005. Radioisotopes for the palliation of metastatic bone cancer: a systematic review. *Lancet Oncol.* 6, 392–400. [https://doi.org/10.1016/S1470-2045\(05\)70206-0](https://doi.org/10.1016/S1470-2045(05)70206-0)

Hermanne, A., Ignatyuk, A. V., Capote, R., Carlson, B. V., Engle, J.W., Kellett, M.A., Kibédi, T., Kim, G., Kondev, F.G., Hussain, M., Lebeda, O., Luca, A., Nagai, Y., Naik, H., Nichols, A.L., Nortier, F.M., Suryanarayana, S. V., Takács, S., Tárkányi, F.T., Verpelli, M., 2018. Reference Cross Sections for Charged-particle Monitor Reactions. *Nucl. Data Sheets* 148, 338–382. <https://doi.org/10.1016/j.nds.2018.02.009>

Hubbel, J.H., Seltzer, S.M., 2004. X-Ray Mass Extinction Coefficients [WWW Document]. NIST Stand. Ref. Database 126. <https://doi.org/https://doi.org/10.18434/T4D01F>

International Atomic Energy Agency, 2011. Nuclear Data for the Production of Therapeutic Radionuclides. Tech. Reports Ser. 473, 395.

Koning, A.J., Rochman, D., Sublet, J., Dzysiuk, N., Fleming, M., Marck, S. van der, 2019. TENDL: Complete Nuclear Data Library for Innovative Nuclear Science and Technology. *Nucl. Data Sheets* 155, 1–55. <https://doi.org/10.1016/j.nds.2019.01.002>

National Nuclear Data Center, 2019. Nuclear structure and decay data on-line library, Nudat 2.8 [WWW Document]. URL <http://www.nndc.bnl.gov/nudat2/>

Otuka, N., Dupont, E., Semkova, V., Pritychenko, B., Blokhin, A.I., Aikawa, M., Babykina, S., Bossant, M., Chen, G., Dunaeva, S., Forrest, R.A., Fukahori, T., Furutachi, N., Ganesan, S., Ge, Z., Gritzay, O.O., Herman, M., Hlavač, S., Kato, K., Lalremruata, B., Lee, Y.O., Makinaga, A., Matsumoto, K., Mikhaylyukova, M., Pikulina, G., Pronyaev, V.G., Saxena, A., Schwerer, O., Simakov, S.P., Soppera, N., Suzuki, R., Takács, S., Tao, X., Taova, S., Tárkányi, F., Varlamov, V. V., Wang, J., Yang, S.C., Zerkin, V., Zhuang, Y., 2014. Towards a More complete and accurate experimental nuclear reaction data library (EXFOR): International collaboration between nuclear reaction data centres (NRDC). *Nucl. Data Sheets* 120, 272–276. <https://doi.org/10.1016/j.nds.2014.07.065>

Pritychenko, B., Sonzogni, A., 2003. Q-value Calculator (QCalc) [WWW Document]. URL <http://www.nndc.bnl.gov/qcalc/>

Qaim, S.M., Spahn, I., Kandil, S.A., Coenen, H.H., 2007. Nuclear data for production of ^{88}Y , ^{140}Nd , ^{153}Sm and ^{169}Yb via novel routes. *Radiochim. Acta* 95, 313–317. <https://doi.org/10.1524/ract.2007.95.6.313>

Tárkányi, F., Hermanne, A., Takács, S., Ditrói, F., Csikai, J., Ignatyuk, A. V., 2014. Cross sections of deuteron induced reactions on $^{\text{nat}}\text{Sm}$ for production of the therapeutic radionuclide ^{145}Sm and ^{153}Sm . *Appl. Radiat. Isot.* 91, 31–37. <https://doi.org/10.1016/j.apradiso.2014.05.007>

Uddin, M.S., Afroze, N., Datta, T.K., Hossain, S.M., Zakaria, A.K.M., Islam, M.A., Naher, K., Shariff, M.A., Yunus, S.M., Islam, S.M.A., 2014. Experimental cross section for the $^{152}\text{Sm}(n,\gamma)^{153}\text{Sm}$ reaction at 0.0334 eV. *Radiochim. Acta* 102. <https://doi.org/10.1515/ract-2013-2172>

Uddin, M.S., Chowdhury, M.H., Hossain, S.M., Latif, S.A., Islam, M.A., Hafiz, M.A., Mubin, S.H., Zakaria, A.K.M.,

Yunus, S.M., Azharul Islam, S.M., 2008. Thermal neutron capture cross sections for the $^{152}\text{Sm}(n,\gamma)^{153}\text{Sm}$ and $^{154}\text{Sm}(n,\gamma)^{155}\text{Sm}$ reactions at 0.0536eV energy. Nucl. Instruments Methods Phys. Res. Sect. B Beam Interact. with Mater. Atoms 266, 4855–4861. <https://doi.org/10.1016/j.nimb.2008.07.032>

Watanabe, T., Fujimaki, M., Fukunishi, N., Imao, H., Kamigaito, O., Kase, M., Komiyama, M., Sakamoto, N., Suda, K., Wakasugi, M., Yamada, K., 2014. Beam energy and longitudinal beam profile measurement system at the RIBF, in: Proceedings of the 5th International Particle Accelerator Conference (IPAC 2014). pp. 3566–3568.

Ziegler, J.F., Biersack, J.P., Ziegler, M.D., 2008. SRIM: the Stopping and Range of Ions in Matter [WWW Document]. URL <http://www.srim.org>

Tables

Table 1. Nuclear data of reaction products in this experiment.

Radionuclide	Decay mode	Half-life	E_γ (keV)	I_γ (%)	Contributing reactions	Q-value (MeV)	Threshold energy (MeV)
^{153}Sm	β^- : 100%	46.50 h	103.18012	29.25(22)	$^{150}\text{Nd}(\alpha, n)$	-6.8	6.9
^{145}Sm	ε : 100%	340 d	61.2265	12.15(10)	$^{146}\text{Nd}(\alpha, 5n)$	-38.2	39.3
					$^{145}\text{Nd}(\alpha, 4n)$	-30.6	31.5
					$^{144}\text{Nd}(\alpha, 3n)$	-24.9	25.6
					$^{143}\text{Nd}(\alpha, 2n)$	-17.1	17.5
					$^{142}\text{Nd}(\alpha, n)$	-10.9	11.3
^{151}Pm	β^- : 100%	28.40 h	340.08	22.5(9)	$^{150}\text{Nd}(\alpha, t)$	-12.8	13.2
					$^{148}\text{Nd}(\alpha, p)$	-8.9	9.1
^{150}Pm	β^- : 100%	2.698 h	333.92	68(4)	$^{150}\text{Nd}(\alpha, tn)$	-20.7	21.2
					$^{148}\text{Nd}(\alpha, d)$	-14.5	14.9
^{149}Pm	β^- : 100%	53.08 h	285.95	3.10(20)	$^{150}\text{Nd}(\alpha, t2n)$	-26.3	27.0
					$^{148}\text{Nd}(\alpha, t)$	-13.9	14.2
					$^{146}\text{Nd}(\alpha, p)$	-9.7	10.0
					Decay of ^{149}Nd		
$^{148\text{m}}\text{Pm}$	β^- : 95.8% IT: 4.2%	41.29 d	629.97	89.0(9)	$^{150}\text{Nd}(\alpha, t3n)$	-33.7	34.6
					$^{148}\text{Nd}(\alpha, tn)$	-21.3	21.8
					$^{146}\text{Nd}(\alpha, d)$	-14.9	15.3
					$^{145}\text{Nd}(\alpha, p)$	-9.6	9.8
$^{148\text{g}}\text{Pm}$	β^- : 100%	5.368 d	1465.12	22.2(5)	$^{150}\text{Nd}(\alpha, t3n)$	-33.6	34.4
					$^{148}\text{Nd}(\alpha, tn)$	-21.1	21.7
					$^{146}\text{Nd}(\alpha, d)$	-14.8	15.2
					$^{145}\text{Nd}(\alpha, p)$	-9.4	9.7
					Decay of $^{148\text{m}}\text{Pm}$		
^{144}Pm	ε : 100%	363 d	696.49	99.490(20)	$^{148}\text{Nd}(\alpha, t5n)$	-48.9	50.2
					$^{146}\text{Nd}(\alpha, t3n)$	-36.2	37.2
					$^{145}\text{Nd}(\alpha, t2n)$	-28.7	29.5
					$^{144}\text{Nd}(\alpha, tn)$	-22.9	23.6
					$^{143}\text{Nd}(\alpha, t)$	-15.1	15.5
					$^{142}\text{Nd}(\alpha, d)$	-15.2	15.7
^{143}Pm	ε : 100%	265 d	741.98	38.5(24)	$^{146}\text{Nd}(\alpha, t4n)$	-42.8	43.9
					$^{145}\text{Nd}(\alpha, t3n)$	-35.2	36.2
					$^{144}\text{Nd}(\alpha, t2n)$	-29.5	30.3
					$^{143}\text{Nd}(\alpha, tn)$	-21.6	22.2

¹⁴⁹ Nd	β^- : 100%	1.728 h	211.309	25.9(14)	¹⁴² Nd(α ,t)	-15.5	16.0
					¹⁵⁰ Nd(α , α n)	-7.4	7.6
¹⁴⁷ Nd	β^- : 100%	10.98 d	91.105	28.1(7)	¹⁴⁸ Nd(α , ³ He)	-15.5	16.0
					¹⁵⁰ Nd(α , α 3n)	-19.7	20.3
					¹⁴⁸ Nd(α , α n)	-7.3	7.5
					¹⁴⁶ Nd(α , ³ He)	-15.3	15.7
⁵¹ Cr	ε : 100%	27.7025 d	320.0824	9.910(10)	¹⁴⁵ Nd(α ,2p)	-15.4	15.9
					⁴⁷ Ti(α , γ)	8.9	0.0
					⁴⁸ Ti(α ,n)	-2.7	2.9
					⁴⁹ Ti(α ,2n)	-10.8	11.7
					⁵⁰ Ti(α ,3n)	-21.8	23.5

Table 2. Cross sections of the $^{nat}\text{Nd}(\alpha,x)^{153,145}\text{Sm}$ reactions. The mean energy with its uncertainty followed by the energy thickness of the corresponding foil in parentheses and the cross section followed by its total uncertainty are presented.

E (MeV)	Cross section (mb)	
	^{153}Sm	^{145}Sm
49.3±0.2 (0.6)	0.220±0.020	262±20
48.1±0.2 (0.6)	0.238±0.021	262±20
45.1±0.2 (0.7)	0.292±0.024	293±22
43.8±0.2 (0.7)	0.317±0.025	323±24
40.6±0.3 (0.7)	0.346±0.027	350±26
39.2±0.3 (0.7)	0.404±0.031	341±25
35.7±0.4 (0.7)	0.549±0.042	346±26
34.2±0.4 (0.8)	0.586±0.044	314±23
30.4±0.5 (0.8)	0.731±0.055	216±16
28.7±0.5 (0.9)	0.846±0.064	185±14
24.2±0.7 (1.0)	0.963±0.072	161±12
22.2±0.8 (1.0)	0.963±0.072	173±13
16.9±1.0 (1.2)	1.17±0.09	36.1±3.0
14.3±1.2 (1.4)	0.405±0.030	2.41±0.22

Table 3. Cross sections of the $^{nat}\text{Nd}(\alpha, x)^{151,150,149,148m,148g,144,143}\text{Pm}$ reactions. The mean energy with its uncertainty followed by the energy thickness of the corresponding foil in parentheses and the cross section followed by its total uncertainty are presented.

E (MeV)	Cross section (mb)						
	^{151}Pm	^{150}Pm	^{149}Pm (cum)	^{148m}Pm	^{148g}Pm	^{144}Pm	^{143}Pm
49.3±0.2 (0.6)	5.35±0.46	4.69±0.51	12.8±1.0	7.32±0.55	1.29±0.11	46.1±3.8	429±32
48.1±0.2 (0.6)	5.16±0.44	3.83±0.41	11.4±0.9	6.95±0.52	1.28±0.11	41.3±3.4	426±32
45.1±0.2 (0.7)	4.67±0.40	3.18±0.31	10.5±0.8	6.16±0.46	1.25±0.10	34.4±2.8	406±30
43.8±0.2 (0.7)	4.65±0.40	2.84±0.28	10.1±0.79	6.08±0.46	1.28±0.10	32.4±2.6	397±30
40.6±0.3 (0.7)	3.68±0.31	2.17±0.21	7.91±0.61	5.61±0.42	1.20±0.10	25.5±2.1	304±23
39.2±0.3 (0.7)	3.15±0.27	1.99±0.19	7.04±0.54	5.27±0.40	1.20±0.10	23.9±2.0	254±219
35.7±0.4 (0.7)	2.35±0.20	1.82±0.18	5.67±0.44	4.87±0.37	1.32±0.11	18.4±1.5	135±10
34.2±0.4 (0.8)	1.91±0.16	1.46±0.14	4.80±0.37	4.26±0.32	1.26±0.10	14.5±1.2	70.8±5.4
30.4±0.5 (0.8)	1.21±0.10	0.744±0.072	3.12±0.25	2.14±0.17	0.791±0.067	6.95±0.61	4.38±0.47
28.7±0.5 (0.9)	1.03±0.09	0.491±0.048	2.74±0.22	1.48±0.12	0.588±0.051	4.35±0.40	1.88±0.27
24.2±0.7 (1.0)	0.511±0.044	0.0636±0.0064	1.11±0.09	0.325±0.030	0.166±0.019	0.434±0.093	0.584±0.146
22.2±0.8 (1.0)	0.243±0.022	0.0113±0.0015	0.482±0.047	0.108±0.014	0.0649±0.0107		
16.9±1.0 (1.2)	0.0157±0.0029	0.00154±0.00044	0.0676±0.0142				
14.3±1.2 (1.4)	0.0113±0.0013	0.00153±0.00036	0.0510±0.0070				

Table 4. Cross sections of the $^{nat}\text{Nd}(\alpha,x)^{149,147}\text{Nd}$ reactions. The mean energy with its uncertainty followed by the energy thickness of the corresponding foil in parentheses and the cross section followed by its total uncertainty are presented.

E (MeV)	Cross section (mb)	
	^{149}Nd	^{147}Nd
49.3±0.2 (0.6)	4.68±0.57	6.95±0.55
48.1±0.2 (0.6)	4.68±0.51	6.34±0.50
45.1±0.2 (0.7)	3.83±0.32	5.16±0.41
43.8±0.2 (0.7)	3.82±0.31	4.80±0.38
40.6±0.3 (0.7)	2.99±0.24	3.59±0.28
39.2±0.3 (0.7)	2.69±0.22	3.10±0.24
35.7±0.4 (0.7)	2.11±0.17	2.42±0.19
34.2±0.4 (0.8)	1.57±0.12	1.95±0.15
30.4±0.5 (0.8)	0.782±0.061	0.959±0.077
28.7±0.5 (0.9)	0.498±0.040	0.638±0.052
24.2±0.7 (1.0)	0.117±0.009	0.127±0.012
22.2±0.8 (1.0)	0.0601±0.0050	0.0691±0.0080
16.9±1.0 (1.2)	0.0477±0.0039	0.0629±0.0079
14.3±1.2 (1.4)	0.0439±0.0035	0.0575±0.0052

Figure captions

Fig. 1. Excitation function of the $^{nat}\text{Ti}(\alpha,x)^{51}\text{Cr}$ monitor reaction compared with the IAEA recommended values (Hermanne et al., 2018).

Fig. 2. Excitation function of the $^{nat}\text{Nd}(\alpha,x)^{153}\text{Sm}$ reaction compared with the previous experimental data (Qaim et al., 2007) and the TENDL-2019 data (Koning et al., 2019).

Fig. 3. Excitation function of the $^{nat}\text{Nd}(\alpha,x)^{145}\text{Sm}$ reaction compared with the TENDL-2019 data (Koning et al., 2019).

Fig. 4. Excitation function of the $^{nat}\text{Nd}(\alpha,x)^{151}\text{Pm}$ reaction compared with the TENDL-2019 data (Koning et al., 2019).

Fig. 5. Excitation function of the $^{nat}\text{Nd}(\alpha,x)^{150}\text{Pm}$ reaction compared with the TENDL-2019 data (Koning et al., 2019).

Fig. 6. Excitation function of the $^{nat}\text{Nd}(\alpha,x)^{149}\text{Pm}$ (cumulative) reaction compared with the TENDL-2019 data (Koning et al., 2019).

Fig. 7. Excitation function of the $^{nat}\text{Nd}(\alpha,x)^{148m}\text{Pm}$ reaction compared with the previous experimental data on ^{146}Nd (Aumann and Guckel, 1977) normalized to natural ratio and the TENDL-2019 data (Koning et al., 2019).

Fig. 8. Excitation function of the $^{nat}\text{Nd}(\alpha,x)^{148g}\text{Pm}$ reaction compared with the previous experimental data on ^{146}Nd (Aumann and Guckel, 1977) normalized to natural ratio and the TENDL-2019 data (Koning et al., 2019).

Fig. 9. Excitation function of the $^{nat}\text{Nd}(\alpha,x)^{144}\text{Pm}$ reaction compared with the TENDL-2019 data (Koning et al., 2019).

Fig. 10. Excitation function of the $^{nat}\text{Nd}(\alpha,x)^{143}\text{Pm}$ reaction compared with the TENDL-2019 data (Koning et al., 2019).

Fig. 11. Excitation function of the $^{nat}\text{Nd}(\alpha,x)^{149}\text{Nd}$ reaction compared with the TENDL-2019 data (Koning et al., 2019).

Fig. 12. Excitation function of the $^{nat}\text{Nd}(\alpha,x)^{147}\text{Nd}$ reaction compared with the TENDL-2019 data (Koning et al., 2019).