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Half-life measurements of the ¹⁴⁴Pm isotope with γ -spectroscopy

C Yalçın^{1,2}, Gy Gyürky², Z Halász², T Szücs², R T Güray¹, G G Kiss², Zs Fülöp², N Özkan¹, E Somorjai²

[1] Kocaeli University, Department of Physics, Umuttepe 41380, Kocaeli, Turkey [2] Atomki, H-4001 Debrecen, POB.51., Hungary

E-mail: caner.yalcin@kocaeli.edu.tr

Abstract. The accurate half-life information for reaction products is very crucial in order to measure the cross section of a reaction via the activation method. Precise and accurate cross section measurement of reactions which are resulting in ¹⁴⁴Pm requires the precise half-life value of this isotope. The adopted half-life of ¹⁴⁴Pm has a relatively high uncertainty and conflicting data can be found in the literature. In order to mitigate this controversy and to reduce the uncertainty, the half-life of ¹⁴⁴Pm is measured with gamma spectroscopy. Experimental methods and preliminary results are presented.

1. Introduction

In the last decade, a number of cross section measurements were carried out using the activation method for the study of the astrophysical p-process (or γ -process) [1–12]. These cross sections are needed for two purposes: to supply data for the nuclear reaction network [13, 14] related to astrophysical p-process and also to improve our knowledge about theoretical cross section calculations [15, 16] by comparing them with the experimental ones. Experimental aspects of the reaction cross section measurements by the activation methods are discussed e.g. in Ref. [17]. A precise and accurate cross section measurement of a reaction using the activation method requires the precise half-life of the reaction product. One of the recent reaction cross section measurements is on the ¹⁴¹Pr(α ,n)¹⁴⁴Pm reaction from Sauerwein et al. from 2011 [7]. This cross section measurement requires precise half-life value of ¹⁴⁴Pm. In the literature the half-life of the ¹⁴⁴Pm isotope is controversial and has a relatively high uncertainty. Half-life values available for this isotope are 377 ± 16 d [18] and 349 ± 16 d [19], the uncertainty of these measurements are 4.24% and 4.58% respectively. The recommended half-life of ¹⁴⁴Pm is 363 ± 14 d [20, 21] which is the weighed average of the two values quoted above. In order to mitigate this controversy and to reduce the uncertainty, the half-life of the ¹⁴⁴Pm isotopes was determined in the present work based on the measurement of the radioactive decay curve of the sources with HPGe detectors. In this paper the experimental methods and preliminary results are presented.

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Table 1. Main radioactive isotopes in the source. The recommended half-life values are taken from [20, 21] and only the highest relative intensity gamma radiations are tabulated with their intensities in parentheses.

Isotope	Decay mode	Recommended half-life	Gamma energy (Intensity) [keV (%)]
143 Pm	ϵ : 100%	$265 \pm 7 \text{ d}$	741.98 (38.5)
$^{144}\mathrm{Pm}$	ϵ : 100%	$363\pm14~{\rm d}$	476.78(43.8), 618.01(98), 696.49(99.490)
$^{146}\mathrm{Pm}$	ϵ : 66.0%	5.53 ± 0.05 y	453.88(65.0), 735.93(22.5)
$^{146}\mathrm{Pm}$	β^- : 34.0%	5.53 ± 0.05 y	747.24(34.0)
$^{148}\mathrm{Pm}$	β^- : 100%	$5.368 \pm 0.002 \; \rm d$	550.27(22.0), 914.85(11.5), 1465.12(22.2)
148m Pm	β^- : 95.8%	$41.29 \pm 0.11 \text{ d}$	414.07(18.66), 550.27(94.9), 629.97(89.0),
			725.70(32.8), 915.33(17.17), 1013.81(20.3)
56 Co	ϵ : 100%	$77.236 \pm 0.026 \ {\rm d}$	846.77(99.94), 1037.843(14.05), 1238.288(66.46)

2. Experimental methods

2.1. Source Preparation

The source was produced by the ¹⁴³Nd(p, γ)¹⁴⁴Pm and ¹⁴⁴Nd(p,n)¹⁴⁴Pm reactions. For these reactions, a natural neodymium oxides target was prepared onto thin Al foil via physical vapor deposition (PVD) method by using electron beam. The target thickness was determined with weight measurement as 295 ± 15 µg/cm². The thickness of the target was also checked by alpha energy loss measurement using the ThiMeT code [22, 23], the results of the two methods were in agreement. The target was irradiated at the cyclotron accelerator of Atomki in Debrecen, Hungary with a 7.5 MeV proton beam of 2.5 µA intensity for about 5 h. Before the gammacounting the target was cooled for about 63 days. Because of the natural Nd used for the target preparation, other isotopes of Pm were also produced via (p, γ) and (p,n) reactions. The composition of the source can be seen in Table 1. The main contaminant beside the Pm isotopes is ⁵⁶Co. This isotope is produced via the ⁵⁶Fe(p,n)⁵⁶Co reaction on the iron impurities in the Al backing foil.

2.2. Gamma counting and data analysis

In order to decrease the systematic uncertainties of the half-life measurement, the source was counted with two independent measurement systems. The first measurement system is composed of a Canberra HPGe detector with 100% relative efficiency (model GR10024), Canberra High Voltage power supply (model 3105D), Ortec amplifier (model 572A) and Ortec MCA (model ASPEC-927) at Atomki. The second measurement system located at the Nuclear Research Laboratory of Kocaeli University, Turkey is based on an Ortec HPGe detector with 25% relative efficiency (model GEM25P4-70) and an Ortec DSPEC jr 2.0 which incorporates all necessary electronics. Both detectors were covered with 10 cm lead shield to minimize natural background.

The first counting was carried out with the Atomki system. The measurement started 63 days after the irradiation, therefore most of the short-lived contaminants had already decayed out. The spectra were saved every 6 h collecting about 500 spectra in total. A typical gamma spectrum in the relevant energy region is shown in Figure 1. The most prominent peaks in the spectra are from the decay of 143,144,146,148 Pm isotopes and 56 Co. The dead time of the counting system was less than 0.3% from the beginning of the counting. The same source was measured with the Kocaeli system. The measurement there started 261 days after the irradiation, lasted for 92 days and the spectra were stored in every 6 hours.

Spectra were analyzed custom-made peak integration codes as well as with the GammaVision

software and a job file prepared for to obtain net peak areas. Half-life values of ¹⁴⁴Pm were determined from the analysis of three gamma peaks (see Table 1) by fitting a linear curve on semi-logarithmic plot of the net areas of the γ -peaks as a function of time. There were no special method used for the data treatment and none of the data points were excluded for fit. The statistical uncertainty of the half-lives was determined from the fit parameter. The analysis of the systematic uncertainties as well as the comparison of the results at the two counting facilities is still in progress.



Figure 1. Relevant energy region of a typical spectrum.

3. Results and Conclusion

According to the preliminary analysis of the currently available data, the obtained half-life for ¹⁴⁴Pm is compatible with previous measurement of Bunney et al. 1964 [19] but significantly lower than the previous measurement of Pagden et al. 1963 [18] and also lower than the recommended value in the literature [20, 21]. The uncertainty of the half-life measurement is about one order of magnitude smaller than that of the previous measurements. For this reason, the ¹⁴¹Pr(α ,n)¹⁴⁴Pm reaction cross section results [7] should be revised when the final half-life of ¹⁴⁴Pm becomes available. As a consequence of the shorter half-life value, the cross sections will need to be corrected down by several percent, but taking into account the total uncertainty of the cross sections, the main conclusions of [7] will not change.

The counting of the source is still in progress at the Nuclear Research Laboratory of Kocaeli University, therefore further details about the experiment and the full analysis will be presented in a forthcoming publication.

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