Activation cross sections and isomeric ratios in reactions induced by 14.5 MeV neutrons on $^{152}$Sm, $^{154}$Sm and $^{178}$Hf

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Abstract. Cross sections for the reactions $^{152}$Sm(n, p)$^{152g}$Pm, $^{154}$Sm(n, p)$^{154g}$Pm, $^{178}$Hf(n, p)$^{178g}$Lu, $^{152}$Sm(n, d)$^{153}$Pm and $^{152}$Sm(n, α)$^{149}$Nd were measured at 14.5 MeV neutron energy by the activation method. On the basis of these cross sections, the associated isomeric ratios in $^{154}$Pm, $^{152}$Pm, $^{178}$Lu and the comparison with the predictions of different compound and precompound models, conclusions are drawn about the role of the preequilibrium processes in 14.5 MeV neutron induced reactions. Calculations for equal angular momentum removal by equilibrium and preequilibrium emitted particles better reproduced the experimental isomeric ratios, than for higher angular momentum removal in the preequilibrium phase. The isomeric ratios may be used as a source of additional information about the spin of the isomeric states in $^{152}$Pm and $^{154}$Pm for which the spectroscopic information is uncertain.

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1. Introduction

The continuous interest [1, 2] in activation cross section measurements with 14 MeV neutrons is due to the fact that they provide in a simple way, nuclear data necessary for a detailed understanding of the nuclear reaction mechanisms at low energies. The simple exit channels (n, p), (n, 2n), (n, α), (n, d), (n, γ), which dominate the cross section, provide valuable data for testing the predictive power of contemporary theoretical and empirical models [3–5]. Especially useful information can be obtained from such data for the compound and precompound emission mechanisms [6, 7]. Analysis of experimental data with 14 MeV neutrons was performed in [7] with the use of the excitation model for the preequilibrium (PE) emission and with the Weisskopf-Ewing evaporation model for compound nucleus (CN) emission processes. The part of the cross section for PE emission at 20 MeV excitation energy ($\approx 14.5$ MeV neutrons) was estimated in that work to be about 20% from data for different targets with mass numbers from 55 to 181. In the same work it was shown that in the PE phase the transition rates $\lambda_{\pm}$ between states with neighboring exciton configurations are independent of nuclear structure effects. According to the golden rule, the transition rates $\lambda_{\pm}$ are determined from the product $|M|^2g^3$, where $g = (6/\pi^2)a$ is the single particle state density and $|M|^2$ is the average squared matrix element, parameterized by the relation:

$$|M|^2 = FM A^{-3} E^{-1}. \quad (1)$$

Here FM (MeV$^3$, ≈ $K$ in [7]) is a normalization constant [4], $A$ the mass number and $E$ the excitation energy in MeV. When a structure dependent level density parameter $a (\pm A/8)$ was used for $g$, the fit to the experimental data required values for $|M|^2$, which almost completely compensated for the variations in $a$, keeping in this way approximately constant the transition rates $\lambda_{\pm} \sim |M|^2g^3$. It is interesting to check these results for other target nuclei and when alternative theoretical models are applied. Such possibility is presented by the code STAPRE [4], in which the angular momentum conserving Hauser-Feshbach model is complemented with the exciton model. For calculations with this code, a value of 500 for the normalizing constant FM was shown [4] to represent better the $^{56}$Fe (n, pγ) reaction than the value of 700 in the same nucleus deduced in [7] for calculations with a similar code. This parameter is important since it determines the PE fraction ($f_{PE} = \sigma_{PE}/\sigma_{R}$) of the cross section for fixed exciton numbers. The excitation functions around 14 MeV neutron energy for $^{115}$In were reproduced by STAPRE in [1], using a value of FM = 250 and level densities calculated by the back-shifted Fermi-gas formula [8]. Sufficient information still does not exist about the FM values to be used for an optimal predictive power of these codes for a variety of target nuclei.

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Another interesting feature of the reactions with 14.5 MeV neutrons is the isotopic dependence of the \((n, p)\) cross section. This dependence is shown [9, 10] to be determined by the systematic change of the \(Q\)-value for the \((n, p)\) reaction with the mass number. The slope increase of the isotopic dependence of the \((n, p)\) cross sections for \(^{74,76,77,78}\text{Se}\) with increasing neutron energy (13, 14.5 and 16.6 MeV) [2] due to decrease of the CN part of the cross section with increasing projectile energy. The change in the slope reflects the difference in the ways the \(Q\)-value affects the \((n, p)\) cross section for the CN and PE mechanisms [9]. In order to reproduce such data, FM values of 500, 450, 400 and 200 for the Te, Xe, W and Os isotopic sequences respectively were deduced in [9]. In confirmation of the statement [7] about structure independence of the transition rate \(\lambda \pm\), it was pointed out that if a structure dependent level density parameter \(a(\pm A/8)\) is used, the FM-values should vary from isotope to isotope in order to reproduce the isotopic dependencies.

The \(\sigma_{n,p}\) isotopic dependencies for 10 elements ranging from Fe to Hg are presented in [11] in comparison with empirically and theoretically derived formulae [12, 13]. The straight lines drawn in this way are close to the data and allow the author of [11] to infer, that “no systematic variations” other than even-odd effects due to changes in \(Q\)-values in \(\sigma_{n,p}\) data “can be observed in the region of closed proton shells either in the target nuclei or in the residual nuclei.” In fact odd-even effects might be the reason for the undiscussed deviation of the \(^{77}\text{Se}\) cross section from the straight line isotopic dependence in [2].

The aim of the present work is to supply new data for the activation cross sections and the isomeric cross-section ratios in 14.5 MeV neutron induced reactions on \(^{152}\text{Sm}, ^{154}\text{Sm}, ^{178}\text{Hf}\) and to study the role of CN and PE mechanisms in these reactions using different models. It seemed also desirable to extend the isotopic dependence for the Sm isotopes and to compare it with theoretical model calculations. It is of separate interest to answer the question of whether the PE emitted protons carry the same or more angular momentum than the evaporated protons in 14.5 MeV neutron induced reactions.

### 2. Experimental procedure and data evaluation

The target samples were prepared from \(\text{Sm}_2\text{O}_3\) and \(\text{HfO}_2\) powders. Samarium targets enriched to 99.0\% \(^{152}\text{Sm}\) and to 99.2\% with \(^{154}\text{Sm}\) were made. The \(\text{HfO}_2\) targets were enriched to 92.4\% in mass 178. \(\text{Fe}_2\text{O}_3\) powder was added and mixed with the target material for the reactions with \(^{152}\text{Sm}\) and \(^{178}\text{Hf}\), in precisely measured weight ratio, in order to monitor the cross sections. The prepared mixtures (all about 200 mg each) were placed and sealed in polyethylene capsules in the form of cylinders 10 mm in diameter and few mm high and were irradiated at 0\text{°} from the beam axis at a commercial open tube type neutron generator “NA-4” (U\text{max} = 120 kV, \(I_{\text{max}} = 1.2\) mA, up to \(5 \times 10^{10}\) n/s). The neutrons being produced by the \(^{3}\text{H}(\text{H},n)^{4}\text{He}\) reaction with a beam of deuterons of about 1 mA, impinging on a Ti-T rotating target. Due to the small thickness of the scattering material between the tritium target and the sample, more than 96\% of the neutron beam remained unscattered and the mean energy of the neutron spectrum was taken from a previous determination [14]: 14.54 ± 0.24 MeV. The 846.8 keV \(\gamma\)-line from the reaction \(^{56}\text{Fe}(n,p)^{56}\text{Mn}\) was used as a monitor for the reactions with \(^{152}\text{Sm}\) and \(^{178}\text{Hf}\). The cross section for this reaction under our experimental conditions (i.e. for 14.54 MeV neutron energy) has previously been determined to be 110.9 ± 2.8 mb [14, 15]. The cross section of 1.9 ± 0.4 mb [22] for \(^{154}\text{Sm}(n,\alpha)^{151}\text{Nd}\) was used as a monitor for the reactions with \(^{154}\text{Sm}\).

The variation of the neutron flux with time was monitored every 60 sec during the irradiation by a scintillation counter. This procedure reduces errors in the determination of the activation cross sections originating from the instability of the neutron flux. Such flux changes, which usually are connected with fluctuations in the working regime of the neutron generator and with target depletion, might lead to serious deviations in the estimation of the initial activity especially for isotopes with half-life shorter than the irradiation time. To obtain enough activity for determining the cross sections, which in some of the cases were less than 1 mb, the irradiations lasted from 5 to 15 min. For this reason, we have modified the time factor \(F\) in the standard relation for determination of unknown cross sections \(\sigma\), in the case of “mixed powder” technique:

\[
\sigma = \frac{G_M A_x F_x S_x G_F}{G_y A_M F_M S_y A_x f_y f_x}
\]

Here \(x\) and \(M\) denote the isotope under investigation and the monitor, \(G\) is the weight of the isotopes, \(A\) is the atomic weight, \(S\), \(f\) and \(e\) are the peak area, the absolute detector efficiency and the corresponding detector efficiency. We have devised the irradiation time \(t_i\) into \(n\) intervals, each of duration \(\Delta t\), during which we assume that the flux was constant and have replaced the usual form of the time factor \(F\) [14] by the expression:

\[
F = \frac{\lambda}{\left(\sum_{i=1}^{n} \Phi_i e^{-(\lambda t_i + (n-i)\Delta t)}\right) (1 - e^{-\lambda \Delta t}) (1 - e^{-\lambda t_i})}
\]

where \(\lambda\) is the decay constant of the reaction product, \(t_0\) and \(t_3\) are the cooling and measurement times, \(\Phi_i\) is the mean relative neutron flux during the \(i\)-th interval. Presentation of \(F\) in the form (3) is valid only, when the sample and the monitor substances are exposed to the same neutron flux, as is in the case of the “mixed-powder technique” employed in this work.

One to three 4096 channel \(\gamma\)-ray spectra were obtained by using a high purity Ge detector. The samples were placed directly on the housing can of the detector. The detector efficiency was determined experimentally with an error of 6\%. The energy resolution of the overall detecting system was 2.2 keV FWHM at 1.33 MeV. The common cooling time before the first spectrum was 1 min. The accumulated spectra were transferred from the pulse-