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IDENTIFICATION OF THE DISCRETE GAMMA-RAYS FOLLOWING THE ^{104}Ru(p,n)^{104}Rh NUCLEAR REACTION

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Se masoara pentru prima data cu un spectrometru de înalta rezolutie radiatiile gama discrete produse în urma reactiei nucleare ^{104}Ru(p,n)^{104}Rh. Masuratoarea este de tipul "in-beam", înregistrându-se atât radiatiile prompte cât si cele întârziate. Sunt prezentate estimarile de model teoretic pentru functiile de excitatie ale sectiunilor eficace de reactie în domeniul energiei protonilor incidenti cuprins între pragul de reactie si 15MeV. Se trec în revista conditiile experimentale iar în final sunt prezentate valorile energiilor ($E_?$) si intensitatilor relative ($I_?$) ale radiatiilor gama masurate. Este mentionata utilitatea noilor date experimentale obtinute în aceasta lucrare pentru bazele de date nucleare de referinta.

For the first time discrete gamma-rays following the nuclear reaction ^{104}Ru(p,n)^{104}Rh are measured with a high resolution spectrometer. The type of experiment is "in-beam" one, recording both prompt and delayed gamma rays. There are presented theoretical model estimations of the cross-section excitation functions for the proton beam energies ranging from the reaction threshold up to 15MeV. Experimental set-up is described in some details. Complete table with the g - rays energies ($E_?$) and their relative intensities ($I_?$) is reported. The usefulness of the new experimental data obtained in this paper for the reference nuclear data base is mentioned.

Introduction

Nuclear decay data represent an important means for characterizing and quantifying radioactive isotopes, and also provide an important tool for understanding the nuclear structure. Our present knowledge on nuclear data is preserved and updated mainly in complex data-bases such those from NNDC (National Nuclear Data Center) at Brookhaven National Laboratory – USA [1] and IAEA (International Nuclear Energy Agency) in Vienna, Austria [2]. An on-line interrogation of these data-bases reveals a lack of measurements on gamma-

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ray production following $^{104}\mathrm{Ru}(p,n)^{104}\mathrm{Rh}$ nuclear reaction at proton energies above the threshold. This knowledge could be important for detailed evaluations of gamma-ray fields in complex situations like in the future generations of nuclear reactors or medical irradiations. It is the main goal of the present paper to report on the first measurement of the gamma rays following the bombardment of a ^{104}Ru target with 7 MeV protons. This experiment has been performed at the IFIN-HH ("Horia Hulubei" National Institute for Physics and Nuclear Engineering) Tandem Van de Graaff electrostatic accelerator. Gamma rays were recorded with a high resolution, high efficiency GeHP (High Purity Germanium) detector positioned at 55 degrees relative to the beam direction, in order to reduce the angular distribution effects in the line intensities. Careful reduction of the complex gamma-spectra allowed for identification of contaminants in the isotopic enriched target. Deconvolution of the peaks from the recorded spectra and efficiency corrections performed with standard calibration sources allowed a precise determination of the gamma lines intensities on a relative scale. The measurements were preceded by Hauser-Feshbach compound nucleus calculations in order to estimate the optimum beam energy leading to a clean and intense gamma spectra (a single dominant reaction channel and optimum intensity). This work is intended to be the first step in our endeavor to measure absolute cross sections for $^{104}\mathrm{Ru}(p,n\gamma)^{104}\mathrm{Rh}$ nuclear reaction.

1. The excitation functions

Based on the most recent compilation of the nuclear masses [3] we have estimated a Q-value of -1.923 MeV for the reaction of interest: $^{104}\mathrm{Ru}(p,n)^{104}\mathrm{Rh}$. Although we used in experiment highly isotopic enriched ^{104}Ru targets, traces from the other stable Ru isotopes (see ref. [4]) could last in the target composition. Therefore, we expect to observe in the high resolution gamma-spectra also lines originating from the beam interaction with ^{96}Ru and $^{98-102}$Ru nuclei. In order to obtain an estimation of their population strength we performed reaction model calculations for all stable Ru isotopes potentially present in the target. At the bombarding energies in the MeV range above the reaction threshold, the dominant reaction mechanism is the compound nucleus [5]. The mathematical formulation of this mechanism is currently developed in the Hauser-Feshbach theory [6]. This formalism in its most advanced form is coded in the EMPIRE-2 code system, available from the NNDC web site [7]. EMPIRE-2 is one of the most complete and easy-to-use reaction mechanism computation environments. The main parameters in the calculations are the transmissions coefficients $T_l^a(\varepsilon)$

and level densities parameters ρ_c. It was not the goal of the present paper to perform a fine-tuning of the parameters. For our purposes, the default parameters of the code (taken as the average values in the region) were accepted. For the nuclear reaction induced by the projectile "*a*", leading to the ejectile "*b*", each compound nucleus state contributes at the total cross section with a partial cross section given by the relation [7]:

$$\sigma_b(E,J,\pi) = \sigma_a(E,J,\pi)\frac{\Gamma_b(E,J,\pi)}{\sum_n \Gamma_n(E,J,\pi)}$$

where the index "*n*" counts the compound nucleus states and $\sigma_a(E,J,\pi)$ is the compound nucleus formation cross section, calculated in terms of the transmission coefficients $T_l^a(\varepsilon)$, by expression

$$\sigma_a(U,J,\pi) = \frac{\pi}{k^2}\frac{2J+1}{(2I+1)(2i+1)}\sum_{S=|I-i|}^{I+i}\sum_{l=|J-S|}^{J+S} f(l,\pi)T_l^a(\varepsilon)$$

where: k – wave number of the target-projectile relative motion, (i, I, J, S) – spins of the projectile, target, compound nucleus angular momentum and channel spin, respectively, l – orbital angular momentum of the projectile "*a*", π - compound nucleus parity and (ε, U) stand for the projectile and compound nucleus energy respectively. The quantity $f(l,\pi)$ ensures parity conservation and is unity for $p * P * (-1)^l = \pi$ and zero otherwise (p, P are the parities of the projectile and target respectively). In order to obtain the total cross section these partial cross sections have to be summed over spin J and parity π, and integrated over excitation energy E. The particle decay with (in our case one neutron) of the compound nucleus state (E,J,π) by the emission of the particle "*c*" has the expression [7]:

$$\Gamma_c(E,J,\pi) = \frac{1}{2\pi\rho_{NC}(E,J,\pi)}\sum_{J'=0}^{\infty}\sum_{\pi'}\sum_{j=J-J'}^{J+J'}\int_0^{E-B_c}\rho_c(E',J',\pi')T_c^{i,j}(E-B_c-E')\,\mathrm{d}E'$$

where B_c is the binding energy of the particle c in the compound nucleus, ρ_c – the level density in the compound nucleus and $T_c^{l,j}(\varepsilon)$ - transmission coefficient for particle c having channel energy $\varepsilon = E - B_c - E'$ and orbital angular momentum l. Cross sections have been computed for several energies above threshold, obtaining a so-called *excitation function* for the cross section, quantity that indicates the expected yields for different reaction channels as a function of the incident energy.

We performed excitation function calculations for the following nuclear reactions: $^{99-102,104}\mathrm{Ru}(p,\gamma)^{100-103,105}\mathrm{Rh}$, $^{99-102,104}\mathrm{Ru}(p,n)^{99-102,104}\mathrm{Rh}$ and

$^{99-102,104}\mathrm{Ru}(p,2n)^{98-101,103}\mathrm{Rh}$ for proton energies ranging from threshold to 15MeV and default EMPIRE-2 input parameters. The results of these calculations for the absolute cross sections of the $^{104}\mathrm{Ru}(p,\gamma)^{105}\mathrm{Rh}$ and $^{104}\mathrm{Ru}(p,n)^{104}\mathrm{Rh}$ reactions are presented in Figure 1. The first observation from this figure is that at a given incident energy, the cross section of the reaction $^{104}\mathrm{Ru}(p,n)^{104}\mathrm{Rh}$ is at least three orders of magnitude higher than of the proton capture reaction. Therefore we do not expect to observe in the experimental spectra gamma-rays originating from the $^{105}\mathrm{Rh}$ nucleus. Moreover the calculations indicate that by choosing the proton beam energy at $E_{beam} = 7\,\mathrm{MeV}$ the $(p,2n)$ reaction channels on all stable *Ru* isotopes are closed. Typically, the threshold energies for these reactions with two neutrons in the exit channel are located at energies above 9 MeV. In this respect, 7 MeV can be considerate a “safe” energy in order to open only a reaction channel with one neutron evaporated. In Table I are present the absolute values of the calculated cross sections at the incident energy of 7 MeV, for the open channels in all stable *Ru* isotopes. At this beam energy can be observed that the reaction of interest for this paper ($^{104}\mathrm{Ru}(p,n)^{104}\mathrm{Rh}$) has the highest yield.

Nuclear reaction	$\sigma(mb)_{E=7\,\mathrm{MeV}}$
$^{99}\mathrm{Ru}(p,\gamma)^{100}\mathrm{Rh}$	0,5
$^{99}\mathrm{Ru}(p,n)^{99}\mathrm{Rh}$	270,2
$^{100}\mathrm{Ru}(p,\gamma)^{101}\mathrm{Rh}$	1,6
$^{100}\mathrm{Ru}(p,n)^{100}\mathrm{Rh}$	280,0
$^{101}\mathrm{Ru}(p,\gamma)^{102}\mathrm{Rh}$	0,6
$^{101}\mathrm{Ru}(p,n)^{101}\mathrm{Rh}$	292,1
$^{102}\mathrm{Ru}(p,\gamma)^{103}\mathrm{Rh}$	0,2
$^{102}\mathrm{Ru}(p,n)^{102}\mathrm{Rh}$	300,5
$^{104}\mathrm{Ru}(p,\gamma)^{105}\mathrm{Rh}$	0,1
$^{104}\mathrm{Ru}(p,n)^{104}\mathrm{Rh}$	308,9

Table I. Nuclear cross sections for the competitive reactions

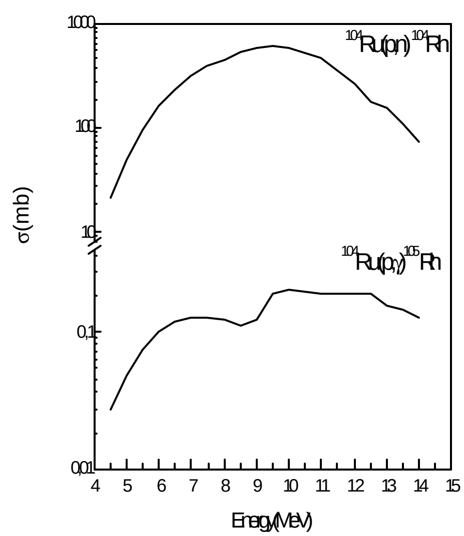


Fig. 1. Excitation functions for the reactions of interest $^{104}\mathrm{Ru}(p,\gamma)^{105}\mathrm{Rh}$ and $^{104}\mathrm{Ru}(p,n)^{104}\mathrm{Rh}$

2. Experimental set-up

The measurements described in this work have been performed at the Tandem Van de Graaff electrostatic accelerator from IFIN-HH. Thin Ru layers on thick Bi backings were prepared by vacuum evaporation techniques. The isotopic enrichment of the ^{104}Ru target was 98.72% according to the manufacturer certificate. The energy loss of the beam on the target estimated with SRIM package [9], was in the same order of magnitude with the accelerator beam resolution, allowing differential measurements for this work. Beam intensities of the order of 10 nA produced counting rates in the detector in the range 5…9 KHz, acceptable for the employed GeHP gamma spectrometer. The components of the spectrometer are presented schematically in Figure 2. All analog electronic modules are in the NIM standard, while MCA – PC is based on a CAMMAC crate.

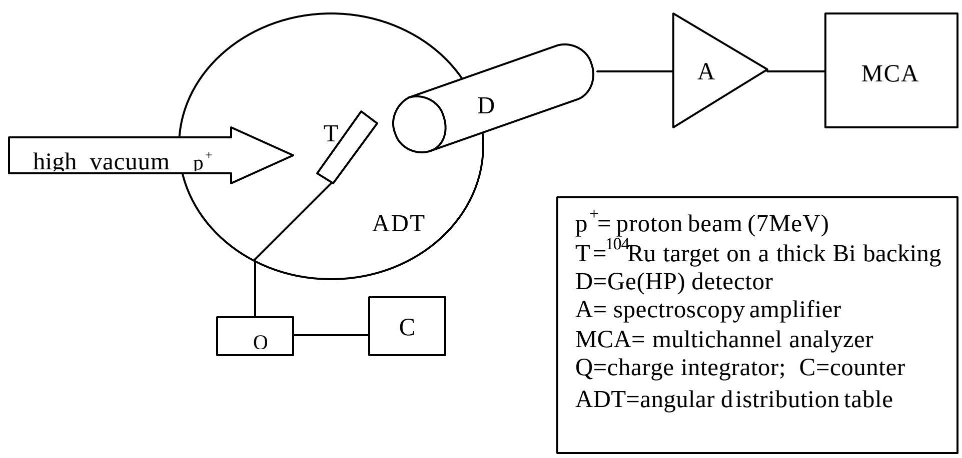


Fig. 2. Schematic representation of the experimental setup used for the measurements reported in this work

Emitted γ rays were detected in a 100 cm^3 hyperpure germanium detector placed at 55 degree with respect the proton beam, in order to minimize angular distribution effects on the intensities. The energy calibration of the spectrometer was performed with a ^{152}Eu γ radioactive source, covering the energy range from 122 keV to 1408 keV. Pulse-height data acquisition has been done on 4096 channels, fine enough to exploit the 2.0 keV energy resolution (at 1 MeV) of the γ spectrometer. In order to estimate the relative efficiency calibration curve of the spectrometer, we measured the photopeak areas of the ^{152}Eu γ lines, corrected for their intensities and finally fit the set of points with an analytical function given for example in references [10,11]. The experimental data for the relative efficiency presented in Figure 3 have been obtained with the relation

$$\varepsilon_{det}^{(i)} = A_\gamma^{(i)} / I_\gamma^{(i)}$$

where $A_\gamma^{(i)}$ represents the area of the photopeak with energy $E^{(i)}$ and $I_\gamma^{(i)}$ is the relative intensity of the γ ray with the same energy of the ^{152}Eu source (tabulated in Gamma–Ray Energy and Intensity standards [12]). The efficiency points were interpolated by a the following curve:

$$y = A_2 + \frac{A_1 - A_2}{1 + \exp[(x - x_0)/\Delta]}$$

where $x_0 = 6.17$, $\Delta = 0.57$, $A_1 = 5.70$, $A_2 = 3.51$ and $(x, y) = (\ln E, \ln \varepsilon_{det})$.

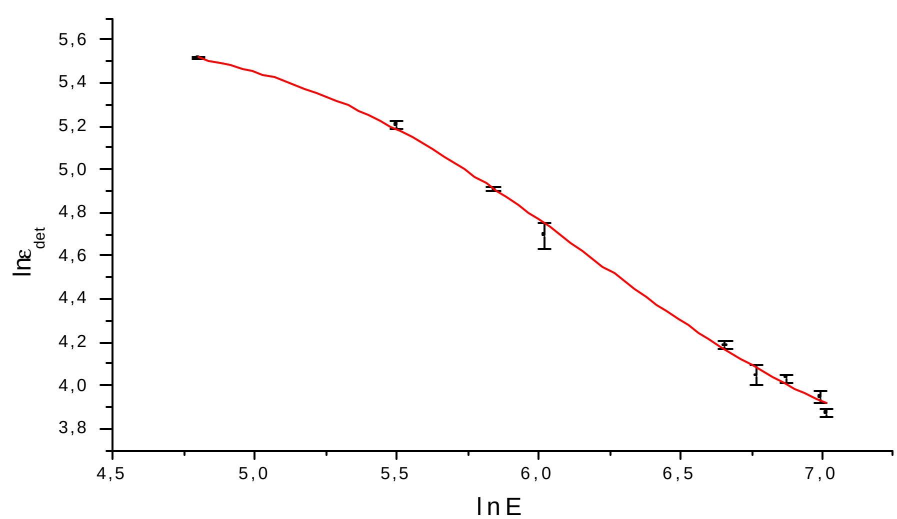
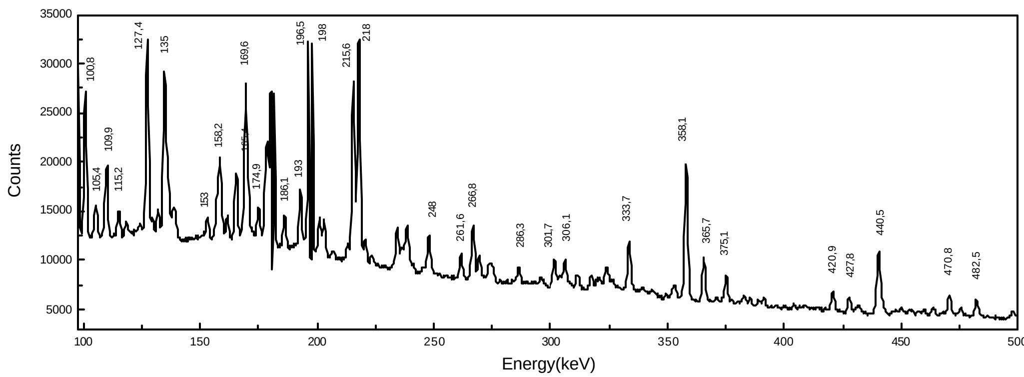


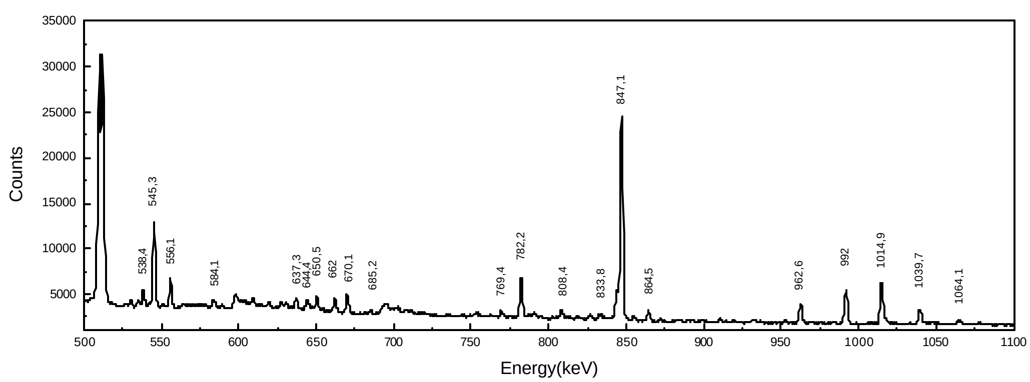
Fig. 3. Efficiency curve for the Ge(HP) detector. Standard gamma rays energies and relative intensities of ^{152}Eu have been used

3. Spectra analysis

In the Figures 4a,b (see below) we present examples of the gamma ray spectrum obtained with the GeHP detector placed at the angle of 55 degrees relative to the beam direction. The gamma ray relative intensities are calculated using the aforementioned efficiency curve by correcting the peak areas of the Figures 4a,b with the efficiencies from Figure 3. The results are presented in Table II on a relative scale, normalized to the intensity of the 135 keV gamma-ray transition (I_{135} = 100).



a)



b)

Fig. 4 a,b. Gamma ray spectra following ^{104}Ru(p,n) ^{104}Rh nuclear reaction

Table II.

Gamma rays following ^{104}Ru(*p,n*)^{104}Rh nuclear reaction at 7MeV bombarding energy, arranged in order of their energy ($E_?$). Their intensities I_γ are normalized to I_{135} = 100.

The uncertainty in the g-peak energies is ± 0.1keV.

E_γ(keV)	I_γ	E_γ(keV)	I_γ	E_γ(keV)	I_γ
100,8	77,9(11)	261,6	21,8(7)	637,3	30,3(13)
105,4	19,5(5)	266,8	56,3(12)	644,4	34,4(13)
109,9	40,6(8)	286,3	14,7(6)	650,5	31,1(13)
115,2	13,4(5)	301,7	30,1(9)	662	42,4(15)
127,4	101,4(13)	306,1	26,5(8)	670,1	57,7(18)
135	**100,0(13)**	333,7	56,6(13)	685,2	10,4(8)
153	12,2(5)	358,1	176,0(23)	769,4	19,3(11)
158,2	80,7(12)	365,7	54,7(13)	782,2	136,2(29)
165,4	42,2(9)	375,1	30,9(10)	808,4	27,6(13)
169,6	121,3(15)	420,9	28,5(10)	833,8	31,2(14)
174,9	19,1(6)	427,8	23,8(9)	847,1	755,9(71)
186,1	23,6(7)	440,5	109,9(20)	864,5	40,0(16)
193	37,1(9)	470,8	29,2(11)	962,6	86,1(25)
196,5	53,8(10)	482,5	29,3(11)	992	153,2(34)
198	59,1(11)	538,4	35,8(13)	1014,9	197,7(39)
215,6	132,4(17)	545,3	182,7(29)	1039,7	69,7(23)
218	160,8(18)	556,1	66,2(17)	1064,1	27,8(15)
248	33,3(9)	584,1	26,5(11)		

Conclusions

For the first time gamma rays from the $^{104}\mathrm{Ru}(p,n)^{104}\mathrm{Rh}$ reaction have been measured by high resolution spectroscopy. Model estimations of the cross-section for the reaction of interest and all competitive channels have been performed with a statistical model. Complete tables with gamma-ray energies and intensities are reported. The original data reported in this paper are expected to be of interest for the international data bases. Further work on the excitation functions and angular distributions of the γ rays emitted in the $^{104}\mathrm{Ru}(p,n)^{104}\mathrm{Rh}$ reaction is underway.

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