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Dedicated to Professor Dr. Cozar Onuc on His 70th Anniversary

THE PRODUCTION OF THE RADIOISOTOPES ^{165}Dy, ^{166}Ho, ^{171}Er, ^{124}Sb FOR BRACHYTHERAPY USE

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ABSTRACT. The thermal neutron cross-sections (σ) of the reactions ^{164}Dy(n,γ) ^{165}Dy, ^{165}Ho(n,γ)^{166}Ho, ^{170}Er(n, γ)^{171}Er, ^{123}Sb(n, γ)^{124}Sb were measured by the activation method. The powder samples were irradiated in an isotropic neutron field obtained from the ^{241}Am-Be and ^{239}Pu-Be neutron sources, moderated with paraffin wax. The γ-ray spectra from the irradiated samples were measured by high resolution γ-ray spectrometry with a calibrated n-type Ge detector. The thermal neutron cross-sections for the reactions studied has been determined to be: for ^{164}Dy(n,γ) ^{165}Dy reaction 2400±200 barns, for ^{165}Ho(n,γ)^{166}Ho reaction 55.3±12 barns, for ^{170}Er(n, γ)^{171}Er reaction 1.3±2 barns and for ^{123}Sb(n, γ)^{124}Sb reaction 3.5±0.5 barns. The thermal neutron cross sections for the reactions ^{164}Dy(n,γ) ^{165}Dy, ^{165}Ho(n,γ)^{166}Ho and ^{123}Sb(n, γ)^{124}Sb are in good agreement, within limits of uncertainty, with most of the values in the literature. The thermal neutron cross section for the reaction ^{170}Er(n, γ)^{171}Er was not in good agreement with the values in the literature, but it is a measurement that has been made, and it can be evaluated again in the future.

Keywords: *neutrons, gamma spectroscopy, capture reactions, effective section, isotopes radioactive*

1. INTRODUCTION

Brachytherapy dates back to 1901 and is commonly used as an effective treatment for cervical, prostate, breast, and skin cancer and can also be used to treat tumors in many other body sites. Radioactive seeds or sources are placed in or near the tumor itself, giving a high radiation dose to the tumor while reducing the radiation exposure in the surrounding healthy tissues. This work presents radioactive seeds that are obtained after a nuclear reaction with neutrons.

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Neutron activation cross-section data have become important for theoretical and experimental studies concerning the interaction of neutron with matter. There are some (n,γ) reactions that can be used to determine the thermal neutron cross-sections. The aim of this present work is to measure the cross-sections for dysprosium, holmium, erbium and antimony, respectively of the reactions: $^{164}Dy(n,\gamma)^{165}Dy$, $^{165}Ho(n,\gamma)^{166}Ho$, $^{170}Er(n,\gamma)^{171}Er$, and $^{123}Sb(n,\gamma)^{124}Sb$. An accurate determination of thermal neutron cross-section is important because it is generally used for reactivity control or adjustment in reactor cores and in-core flux measurements in very small distances of core lattices. We found in literature a number of experimental and evaluated data on the thermal neutron capture cross-sections for the reactions mentioned and we compared them with the results obtained in this work.

2. EXPERIMENTAL PROCEDURE

2.1. Neutron source

The irradiation of samples was performed by the neutrons from a combination of sources: a 33 Ci ^{239}Pu-^{9}Be source and a 5 Ci ^{241}Am-^{9}Be source which are immersed in paraffin moderator with (see Fig.1) a width of 3-4 centimeters in order to obtain 61.5 % thermal neutrons for a divergent flux. [1].

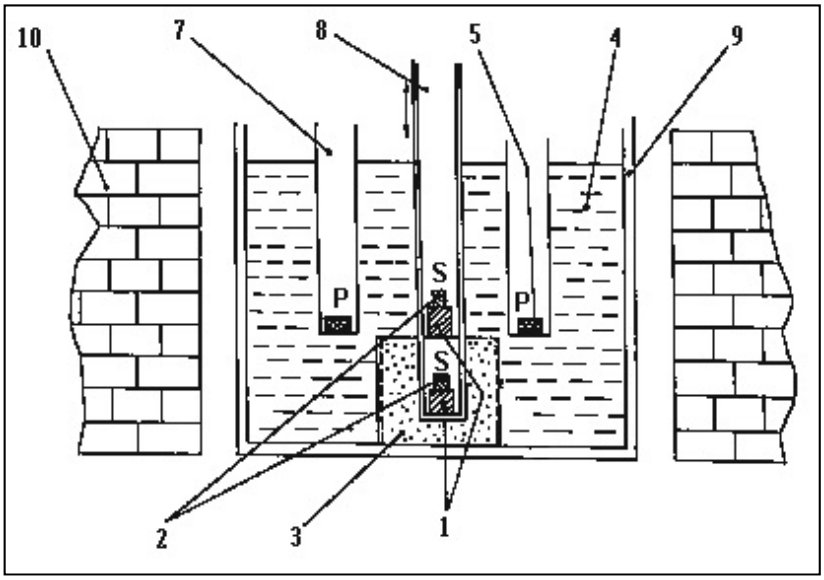

Fig. 1. Configuration of the neutron sources Am-Be and Pu-Be:
1) Am-Be source, 2) Pu-Be source, 3) boron paraffin, 4) pure paraffin for thermalization, 5) the sample introduced for irradiation, 7) irradiation channel, 8) central channel for fast neutrons irradiation, 9) Fe walls with paraffin, 10) boron paraffin bricks for protection.

2.2. Sample irradiation

The analysis is made on solid samples, transformed in powder: metallic Dy, erbium oxide powder (Er_2O_3), metallic Ho, metallic Sb. The samples are weighed out and the results are presented in Table 1.

Table 1. Weight of the samples

Sample	**Weight (g)**
Dy metalic	1.445
Ho metalic	1.1737
Er_2O_3	2.152 (1.882 Er_2)
Sb metalic	2.1529

After the thermalization of the neutrons in paraffin towards the irradiation channel, the neutrons are going to be captured by the samples, where the reactions ^{164}Dy(n,γ)^{165}Dy, ^{165}Ho(n,γ)^{166}Ho, ^{170}Er(n, γ)^{171}Er, ^{123}Sb(n, γ)^{124}Sb take place.

The samples were irradiated with a flux of $3.05 \cdot 10^5$ n/cm^2·s and the irradiation times for the reactions were chosen for a period greater than 3.5 half-lives, yielding enough activity to be measured in a γ-ray counting system (Table 2). [2]

Table 2. The half-lifes of the isotopes

The isotopes	**Half-life ($T_{1/2}$)**
^{165}Dy	2.3 h
^{166}Ho	26 h
^{171}Er	7.5 h
^{124}Sb	60 d

2.3. Measurement of activity

The induced gamma activities emitted from the activation samples were measured by using a high-resolution γ-ray spectrometer. The γ-ray spectrometer was a GC1019 type Canberra high-purity germanium (HPGe)-detector. The HPGe-detector was operated at liquid nitrogen temperature and was coupled with a 3106D Canberra type High-Voltage module, an Ortec amplifier and a computer-based multichannel analyzer. The detector was shielded by a lead armor NZ-138 type. The spectrum analysis was done using the Genie-2000 computer program. The detection efficiency for the γ-ray spectrometer was calibrated with a set of standard sources:

^{241}Am (59.541 keV), ^{22}Na (511.006 keV), ^{137}Cs (661.657 keV), ^{60}Co (1173.237 keV and 1332.501 keV). At the instant when the activation has been terminated, (t=0), the activity of the samples is given by the following expression:

$$\Lambda_0 = \frac{\sigma m N_A \phi \chi S}{M}, \quad (1)$$

where

Λ_0 [Bq] = the number of disintegrations per second of the element in the sample,
σ [cm^2] = cross-section of the reaction,
m [g] = mass of the target element,
N_A = Avogadro's number (6.023×10^{23} molecules/mole,
ϕ [neutrons/cm^2s] = neutron flux,
χ [%] fraction of the target isotope in the sample (isotopic abundance),
S = saturation factor, $1 - e^{-\lambda t}$ where $\lambda=0.693/T_{1/2}$, t = the irradiation time,
M = atomic weight of the element. [3]

Table 3. The molecular masses of the target elements and the isotopic abundance in nature of the initial radionuclides

Element	The molecular mass (M)	Isotop	Isotopic abundance χ (%)
Dy	162.5	^{164}Dy	28.18
Ho	164.9	^{165}Ho	100
Er	167.26	^{170}Er	14.91
Sb	121.76	^{123}Sb	42.64

In order to find out the cross-section σ, we have to determine the induced activity Λ_0. For this we will use the absolute activity using a thorium (Th) sample with his activity known. We use another formula:

$$\varepsilon_f = \frac{N_i}{\varepsilon_g \cdot \Lambda \cdot p_i \cdot t}, \quad (2)$$

where

N_i = the area under the photopeak,
ε_g = geometrical efficiency (the probability that a certain radiation falls on detector),
Λ = source activity (absolute),
p_i = decay fraction of the unknown activity, which is the fraction of the total disintegrations in which the measured gamma is emitted,
t = the measuring time of the spectrum [4]; for the measuring time we chose a value of 6 hours, in order to obtain all the photopeaks (radioisotopes) of Th.

Using some important photopeaks from the Th spectrum with the greatest p_i, we can obtain an efficiency curve that would be useful to find out the induced activity for whatever photopeak from the spectrum of the radioisotopes ^{165}Dy, ^{166}Ho, ^{171}Er, ^{124}Sb. (Figure 2, 3, 4 and 5)

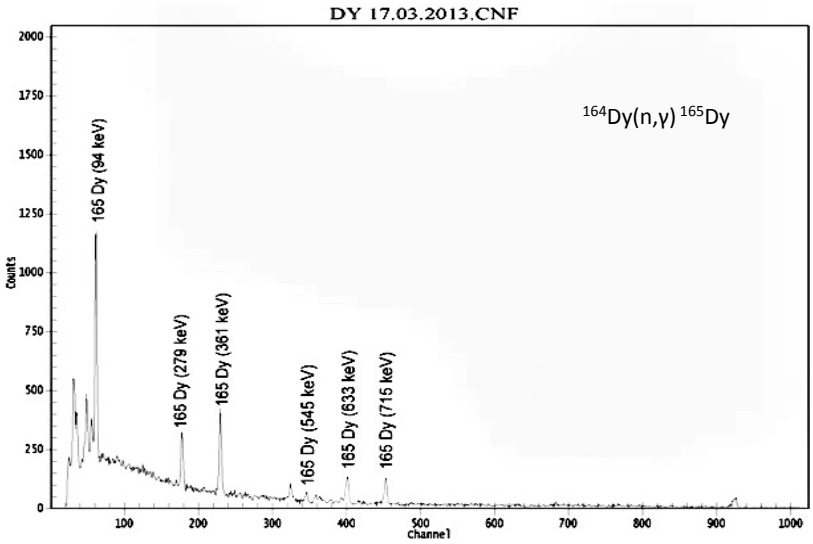

Fig. 2. The spectrum of ^{165}Dy showing the main radionuclides and their γ-energies in keV

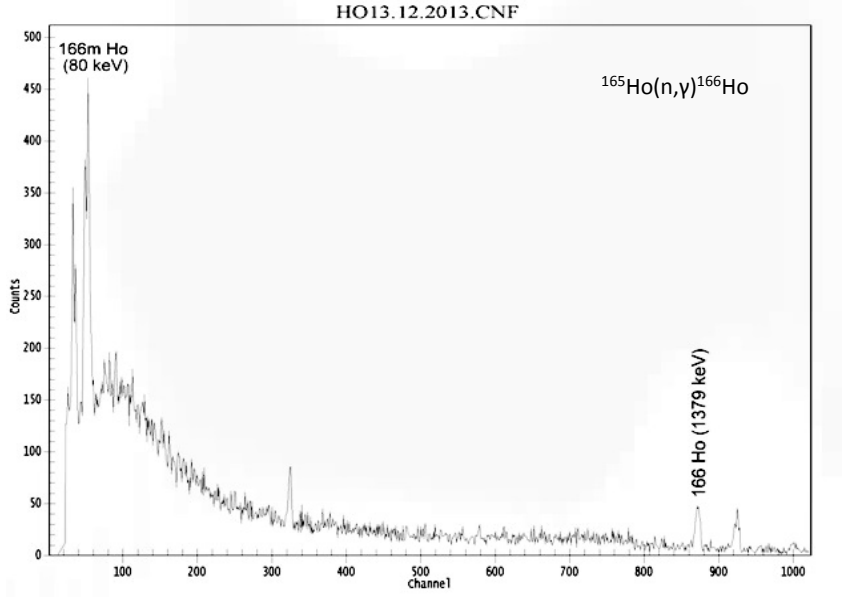

Fig. 3. The spectrum of ^{166}Ho showing the main radionuclides and their γ-energies in keV

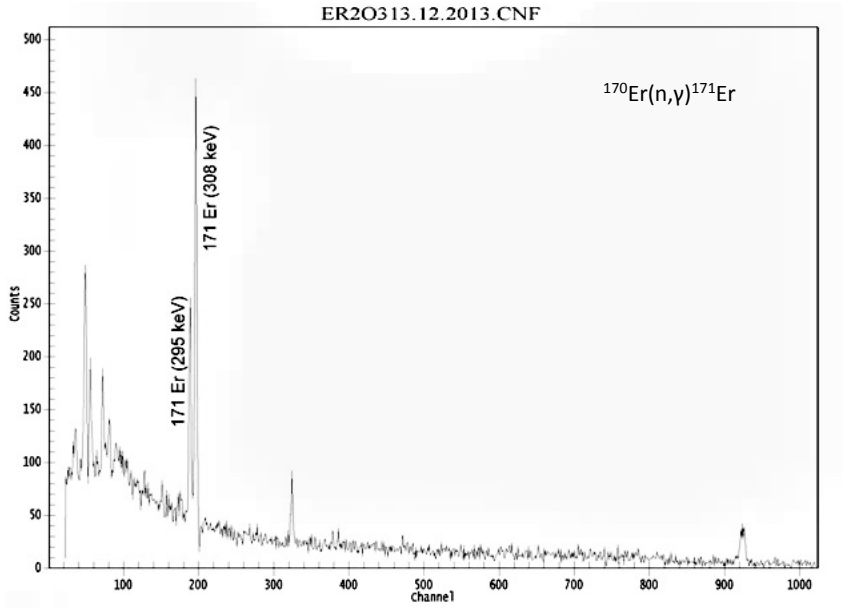

Fig. 4. The spectrum of ^{171}Er showing the main radionuclides and their γ-energies in keV

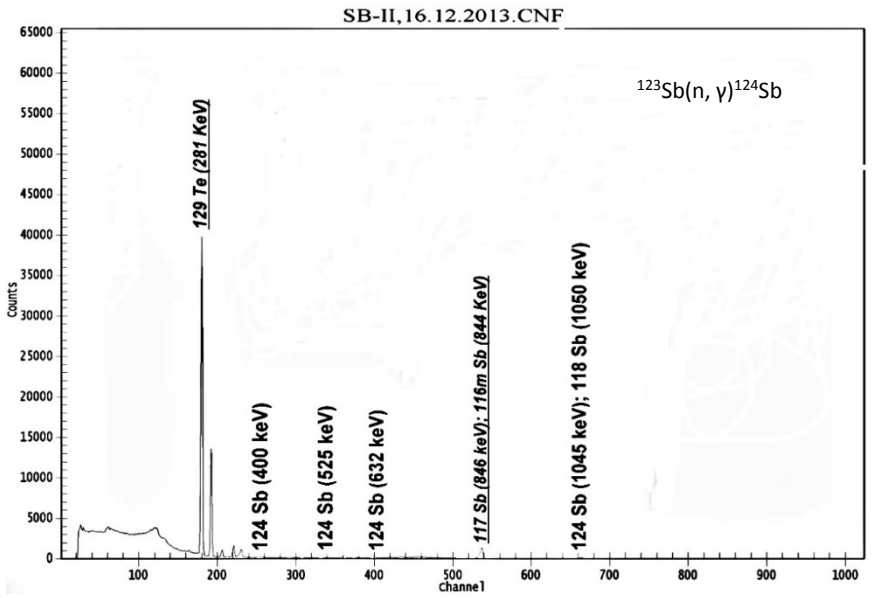

Fig. 5. The spectrum of ^{124}Sb showing the main radionuclides and their γ-energies in keV

To measure the activities of ^{164}Dy(n,γ)^{165}Dy, ^{165}Ho(n,γ)^{166}Ho, ^{170}Er(n, γ)^{171}Er, ^{123}Sb(n, γ)^{124}Sb reactions, we have chosen the γ-ray peaks with high intensity (p_i), well-separated, and relatively low background (Table 4).

Knowing the values from Table 3 too, we could calculate the cross-sections of the reactions using the formula (1).

Table 4. Nuclear data decay used for determination of the induced activity

Reaction	Main γ-rays		Activity
	Energy (keV)	Intensity p_i (%)	
^{164}Dy(n,γ)^{165}Dy	94	3.5784	146.0865504
	361	0.8400	
^{165}Ho(n,γ)^{166}Ho	80	12.7050	686.4544856
	1379	0.93	
^{170}Er(n, γ)^{171}Er	295	28.9	16.89549826
	308	64.4	
^{123}Sb(n, γ)^{124}Sb	253	99	750.5485954
	666	99.6	

3. RESULTS AND DISCUSSION

The thermal neutron cross-section for the ^{164}Dy(n,γ)^{165}Dy reaction given in Table 5 together with other literature values is 2400±200 barns. This value is close to within 4.16-4.76 % with the values obtained by RNAL, NGATLAS, JEF 2.2., Heft and Sehgal et al., but disagrees with the measurements of Johnsrud et al., Goldhaber and Muehlhause, Alstad et al. by 12.5-16.66 %.

Table 5. Thermal neutron cross-section for ^{164}Dy(n,γ) ^{165}Dy reaction

Year	References other works	Thermal neutron cross-section σ (barn)
2014	This work	2400±200
2005	Yucel [5]	2672±104
2001	Cho et al. [5]	2656±98
2000	RNAL [5] [55]	2520
1999	Holden [5]	2700
1998	ENDF/B-VI [5]	2651.63
1997	NGATLAS [5]	2520
1996	JEF 2.2 [5]	2520
1996	NuDat [5]	2650±100
1993	Griffin et al. [5]	2650±100

Year	References other works	Thermal neutron cross-section σ (barn)
1984	Simonits et al. [5]	2660±133
1984	Mughabghab [5]	2650±278
1978	Heft [5]	2300±200
1977	Lucas et al. [5]	2695
1976	Erdtmann [5]	2700±300
1974	Rzves and Zieba [5]	2700±200
1973	BNL [5]	2700±75
1972	Alstad et al. [5]	2800±110
1972	Fawcett et al. [5]	2700±200
1970	Vertebnyj et al. [5]	2740±45
1968	Goldman et al. [5]	2600
1968	Holden and Walker [5]	2600
1967	Scoville [5]	2600±410
1964	Esch and Feiner [5]	2700±200
1959	Sehgal et al. [5]	2490±300
1959	Johnsrud et al. [5]	2100
1958	House and Frost [5]	2750±150
1956	Walker [5]	2630±200

The thermal neutron cross-sections for the $^{170}Er(n, \gamma)^{171}Er$ reaction given in Table 6 together with other literature values is 1.3±2 barns. This value disagrees with all the values given by 69.7-89.33 %.

Table 6. Thermal neutron cross-section for $^{170}Er(n,\gamma)^{171}Er$ reaction

Year	References	Thermal neutron cross-section σ (barn)
2014	This work	1.3±2
2007	Yucel [7]	7.99±0.56
2003	De Corte [7]	8.86±0.35
1997	Knopf and Waschkowski [7]	15±1
1989	De Corte and Simonits [7]	8.85±0.27
1978	Heft [7]	6.0±1
1972	Glomset and Pappas [7]	5.8±0.3
1968	Vertebnyj et al. [7]	12±5
1967	Gilette [7]	5.70±0.15
1963	Mangal and Gill [7]	4.3±0.65
1954	Barnes [7]	8.72±1.78
2007	NuDat [7]	5.8±0.3
2006	ENDFB-VII [7]	8.85
2005	JEFF 3.1 [7]	5.776
2003	Kolotov and De Corte [7]	8.86±0.35

Year	References	Thermal neutron cross-section σ (barn)
2002	JENDL 3.3 [7]	5.776
1999	Holden [7]	6±1
1984	Mughabghab [7]	5.8±0.3
1976	BROND 2.2 [7]	5.8±0.12

The thermal neutron cross-sections for the ^{165}Ho(n,γ)^{166}Ho reaction given in Table 7 together with other literature values is 55.3±12 barns. This values is close to within 5.17-9.83 % with the values obtained by Holden, De Corte, Rajput et al., Nguyen, Yucei et al., but disagrees with the measurements of ENDF/B-VII.0, Mughabghab, Danon et al., JFF2.2, IAEA, Scoville and Rogers by 17.18-17.91 %.

Table 7. Thermal neutron cross-section for ^{165}Ho(n,γ)^{166}Ho reaction

Year	References	Thermal neutron cross-section σ (barn)
2014	This work	55.3±12
2010	Nguyen [8]	59.7±2.5
1997	S. I. Kafala [6]	61.2
2009	Rajput et al. [8]	58.98±2.1
2006	ENDF/B-VII.0 [8]	64.67
2005	Yucei et al. [8]	59.2±2.5
2003	De Corte [8]	58.5±1.3
2003	Mughabghab [8]	64.7±1.2
1999	Holden [8]	58
1998	Danon et al. [8]	64.4±2.8
1997	Katala et al. [8]	61.2±0.8
1994	JFF2.2 [8]	66.59
1989	De Corte [6]	58.1
1987	IAEA [6]	64.7
1987	Gryntakis et al. [8]	61.2±1.1
1984	Mughabghab [8]	61.2±1.1
1984	Simonits et al. [8]	61.2±3
1978	Heft [8]	61.4±1.0
1976	Erdtmann [8]	63±3.3
1974	Ryves and Zieba [8]	61.2±1.1
1972	Steinnes [8]	65±2
1969	Walker [8]	63
1968	Scoville and Rogers [8]	67
1967	THAI-AEC-10 [8]	64
1967	Zimmerman et al. [8]	60±2
1967	Stephenson [8]	67
1962	Keisch and Faler [8]	64±6
1951	Pomerance [8]	64±3

The thermal neutron cross-sections for the $^{123}Sb(n, \gamma)^{124}Sb$ reaction given in Table 8 together with other literature values is 3.5 ± 0.5 barns. This value is close to within 7.4-20.63 % with all the values given.

Table 8. Thermal neutron cross-section for $^{123}Sb(n,\gamma)^{124}Sb$ reaction

Year	References	Thermal neutron cross-section σ (barn)
2014	This work	3.5 ± 0.5
1997	S. I. Kafala [6]	4.41
1989	De Corte [6]	4.08
1987	IAEA [6]	4.156
1978	Heft [6]	3.78

4. CONCLUSIONS

The thermal neutron cross-sections for the $^{164}Dy(n,\gamma)^{165}Dy$, $^{165}Ho(n,\gamma)^{166}Ho$, $^{170}Er(n,\gamma)^{171}Er$, $^{123}Sb(n, \gamma)^{124}Sb$ reactions have been measured using the activation method and are almost all in good agreement with the values from literature.

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