LONGLIVING ^{178m2}Hf,^{172}Hf,^{150}Eu AND ^{133}Ba ISOTOPES PHOTOPRODUCTION STUDY

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The yield of the ^{178m2}Hf ($T = 31y$), ^{172}Hf(1.87), ^{150}Eu (36.9) and ^{133}Ba (10.5) isotopes was measured after irradiation of a tantalum target with 1.0 GeV Kharkiv linear accelerator electron beam. The integral and average cross-sections are obtained. The results are discussed and compared with another data. The number of produced ^{178m2}Hf atoms in tantalum target is $(2.7 \pm 0.3) \cdot 10^{11}$.

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1. INTRODUCTION

Photonuclear reactions starting from a high-spin isomer can be important for the study of giant resonances built on quasiparticle states as well as for the study of the amount of K-mixing at high excitation energies. The interest in such reactions may also be motivated by the possible applications, e.g. the search for efficient ways of γ-laser pumping [1].The Hf state is an example of a so-called K isomer. Not only the spin of the excited nucleus is very high (J = 16), but its projection on the nuclear symmetry axis, denoted by the quantum number K, is also 16. Selection rules for low-multiple electromagnetic decay severely inhibit transition that change K. That's why the isomer takes so long to venture the first decay step-down to a band of different J states with K = 8 (see Fig. 1). The bottom state of that band is another isomer, which a half-life of only 4 seconds, that briefly bars the way to the K = 0 ground-state band [2]. In 1999 feature results on supervision of triggered decay long-living isomer ^{178m2}Hf have been published, that, appear, tore off a way to creation of the nuclear laser [3]. This isomer is a fourquasiparticle, 16^+, long-lived (31 year) yrast trap. It was be considered as a unique object for such investigation. Experiences have been executed by group of researchers under the direction of Carl B. Collins, director of the Center of quantum electronics at Texas University in Dallas (USA) [4]. The essence of experiment has consisted in acceleration of isomer disintegration by influence on it by the radiation of dentist x-ray installation. This work has caused chain reaction of similar experiments which, basically, have not confirmed first optimistic results. The review of these works and the executed in 1998-2003 experiments are submitted and systematized in [5]. To study mechanisms by which a release of isomer decays triggering, a critical requirement is that of ^{178m2}Hf isomeric atoms must be available in milligram or even larger amount. In the nature this isomer does not exist, it can be created on the accelerators or in a reactor. Large quantity of ^{178m2}Hf was produced at Los Alamos by Ta irradiation with 800 MeV protons from a high-current accelerator (formerly LAMPF).

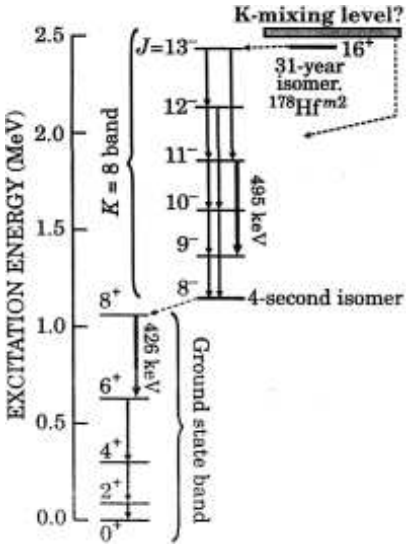

Fig.1. *The nuclear isomer ^{178m2}Hf, sitting 2.45 MeV above the hafnium-178 ground state, has projected angular momentum $K = 16$ on the nuclear symmetry axis. Thus its halflife for entering the $K = 8$ band of exited states is very long (31 years). From there, only a 4-second isomer keeps it from the $K = 0$ ground state band. Exited states are labeled by spin* J *and parity.* 495 *and* 426 keV *are gamma-rays energies for which counting was reportedly enhanced by x-ray triggering in an experiment at the University of Texas. The experimenter suggested that a putative* K *mixing level above the 31-year isomer allows the isomer prompt entry into the $K = 8$ band*

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The advantage of this method was the ability to accumulate the isomer asby-product within a massive tantalum beam dump during the operation of the accelerator for other experiments [6]. However, the isomeric material produced in this way contain a very high activity of other radionuclides and the corresponding γ background remain high even after chemical isolation of the hafnium fraction. The 1.87-years-lived isomer ^{172}Hf is the most problematic contaminant. The productivity of ^{178m2}Hf in spallation reactions with protons at intermediate energies was systematically studied in 2002-2004 for Ta, W and Re targets at Dubna using $660\,MeV$ synchrocyclotron [7]. However, no scheme was proposed by which to overcome the basic disadvantage presented by the accompanying production of high contaminant activities. Better quality sources of ^{178m2}Hf were successfully produced by the ^{176}Yb(^{4}He, $2n$) reaction using a $36\,MeV$ ^{4}He -ion beam and subsequent chemical processing of the irradiated Yb target [8]. It is known, that the production reaction ^{179}Hf$(n, 2n)^{178m2}$Hf has a resonably good cross-section for $14\,MeV$ incident neutrons. The absolute yield is, however, restricted by practical limitation of neutron generators exploiting the d+T reaction. An attempt was made to observe the production of ^{178m2}Hf in reactor irradiation of ^{nat}Hf target. Is was assumed that some reasonable yield could be obtained due to the ^{178}Hf$(n, n)^{178m2}$Hf reactions with the neutrons in the fission spectrum. Possibility of ^{178m2}Hf isomer producing was investigated using $4.5\,GeV$ bremsstrahlung from an Yerevan electron synchrotron [9]. The tantalum target was irradiated in 1986 for other purposes and fortunately it was saved up to now, providing the best possible condition for the detection of the 31-year-lived ^{178m}Hf isomer. After spectrum measurements and the analysis of the received results authors calculated the productivity under optimum conditions. The maximum yields of ^{178m}Hf was found to be of about $3 \cdot 10^9$ atoms$/(s \cdot 100\mu A)$.

2. MEASUREMENTS AND RESULTS

Results of searches and researches of the induced activity on the tantalum radiator of the linear electron accelerator are described here. Kharkiv electron accelerator was started to operate in 1966. Archives investigation evidences that in the beginning this accelerator worked as bremsstrahlung source. Calendar duration of the tantalum radiator irradiation was 5 years (1966-1970). The working electron beam current was 0.15μA, beam energy was adjusted within $1\,GeV$. In this regime accelerator worked 10000 hours (2000 per year). Cooling time from bremsstrahlung intensive experiments to present radiation measurement is 35 years (1970-2006). In October, 2006 the tantalum radiator has been taken off and installed on gamma detector for spectrum measurement. The sample activity was measured using a Ge(Li) spectrometry with working volume 40 cm^3 and the $2.5\,keV$ energy resolution. The sample was installed on the Ge(Li) crystal in close contact geometry. The spectrum was analyzed using special code and accurate numerical values were obtained for the line energy and area. The source intensity is so high that measurement time was only 100 minutes (in [9] it was two weeks!) and the typical statistical errors deduced from the areas of the γ lines were less then $3-5\%$. No deviations were found in the relative intensity of the γ lines as compared to the tabular values for ^{178m2}Hf. The γ line widths were in agreement with those expected. These meant that no overlap occurred between the lines of interest and those from background contribution. Natural background γ lines were definitely present in the spectrum, but it did not disturb the detection of ^{178m2}Hf ($T = 31y$). In addition, the lines of ^{172}Hf, ^{150}Eu and ^{133}Ba were found in the spectrum, being long lived enough to have survived while other radionuclides decayed completely after such a long cooling time. The area of corresponding γ-line allowed a determination of the production yield for each given isotope after taking into account the decay factors, detector efficiency and quantum yield of the line. The latter parameters were taken from the tabular data [10]. It was found that the numbers of nuclei determined from different γ lines are practically identical. The measured γ spectrum is complicated for representation. It contains more than 100 peaks. In Fig.2 the spectrum fragment on which ^{178m2}Hf lines are concentrated is represented. In the spectra 13 from 15 cascade lines of the^{178m2}Hf decay [10] are presented. Two lines 274.4 and $309.5\,keV$ due to law decay probability (1.38% and 0.019% respectively) were not detected in the spectrum. Two pare of lines - $(88.9-93.2)$ and $(213.4-216.7)\,keV$ due to low detector energy resolution are partly overlapped. This did not confuse the line identification and the γ lines areas definition. The 13 independent yields was averaged and standard deviation was evaluated as less than 3%. In Fig.2 the single ^{172}Hf decay line with energy $125.812\,keV$ and relative probability decay 0.11% is present. With help of this line the number of ^{172}Hf nuclei in the irradiated Ta target was determined. In the γ spectra the short activities were also detected. One of them is ^{172}Lu which has living time 8.7 days. This isotope is the daughter product of long living ^{172}Hf $(1.87\,y)$ decay. In Fig.1 ^{172}Lu lines are seen. Those lines was used for ^{172}Lu nucleus amount in Ta target determination. In the spectrum the lines of the elements, which are situated too distant in N-Z diagram from Ta are presents. They are ^{150}Eu and ^{133}Ba. The Ta nucleus has large enough fissility with broad fission fragments mass distribution. Surely the ^{150}Eu and ^{133}Ba nuclei was produced in Ta photo fission reaction [11]. In Fig.2 the decay γ-lines of ^{150}Eu and ^{133}Ba are presented. In Fig.2 one can see level scheme of the ^{178m2}Hf decay. The ^{172}Hf and ^{133}Ba nuclides accumulated the total yield of the appropriate isobaric chain, while‘ ^{178m2}Hf and ^{150}Eu were produced independently in the reactions [9].

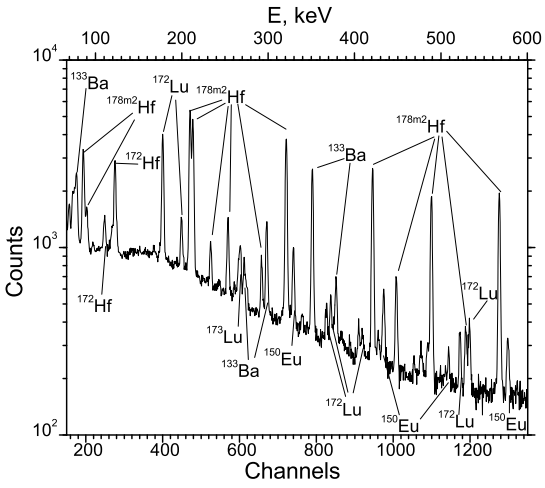

Fig.2. *The spectrum fragment on which ^{178m2}Hf lines are concentrated is represented*

In the Table 1 the number of the ^{178m2}Hf, ^{172}Hf, ^{150}Eu and ^{133}Ba atoms that were created under the action of the bremsstrahlung in this and Yerevan-Dubna [9] experiments are shown. The atoms numbers are given for irradiation ending time.

Table 1.*Isotopes amounts in the Ta target today*

Nucleus	T(y)	Type of yield	Kharkiv
^{178m2}Hf	31.1	Independent	$(2.7 \pm 0.1) \cdot 10^{11}$
^{172}Hf	1.87	Cumulative	$(2.6 \pm 0.1) \cdot 10^{8}$
^{150}Eu	36.9	Independent	$(5.2 \pm 0.015) \cdot 10^{9}$
^{133}Ba	10.51	Cumulative	$(1.2 \pm 0.04) \cdot 10^{11}$

One can see that in this work the yields of ^{178m2}Hf and other nuclei are more then three order bigger then in [9]. This can be explained by big intensity and irradiation length. In table 2 one can see the atomic amount in Ta sample today.

Table 2. *Isotopes amounts in the Ta target after irradiation*

Nucleus	Kharkiv	Dubna
^{178m2}Hf	$(5.9 \pm 0.2) \cdot 10^{11}$	$(1.57 \pm 0.08) \cdot 10^{8}$
^{172}Hf	$(1.4 \pm 0.1) \cdot 10^{14}$	$(2.3 \pm 0.2) \cdot 10^{10}$
^{150}Eu	$(1.0 \pm 0.05) \cdot 10^{10}$	$(8 \pm 2) \cdot 10^{7}$
^{133}Ba	$(1.2 \pm 0.05) \cdot 10^{12}$	$(8.7 \pm 0.9) \cdot 10^{8}$

The number of ^{178m2}Hf nuclei comparably with table 1 data decreased by twice. It reflect the fact, that from end of the sample radiation passed time interval equaled to half decay time.

The amount of ^{172}Hf decreased strongly due to relative short half decay time - 1.87 year. Having determined the number of the atom produced, it is possible to evaluate the production cross-section based on the known number of projectiles reaching the target. We evaluate the reaction yield in term of cross-section per "equivalent" photons. In work [9] the integral and average cross sections of $^{181}\mathrm{Ta}(\gamma, p2n)^{178m2}$Hf reaction were determined. Forth below we shall describe the method of those cross section determination was used in this work. Our method sufficiently different from used in work [9], because in our work tantalum sample was irradiated by electron beam when in [9] by bremsstrahlung. Equivalent photons number n_q determined as:

$$\mathrm{n}_q = (\mathrm{n}_e \cdot \mathrm{t})/2. \quad (1)$$

Here n_e is total initial electron number, which was $3.25 \cdot 10^{19}$, t is tantalum thickness in radiation units equal 0.073. Nuclear reaction cross section per equivalent photon (yield) determined as:

$$\sigma_q = \mathrm{N}_{at}/(\mathrm{n}_q \cdot \mathrm{n}_t), \quad (2)$$

where: σ_q - cross section per equivalent photon, N_{at}- number of isomer atoms produced in target, n_q - equivalent photons number, n_t - tantalum target thickness in nucleus per cm^2. We use bremsstrahlung spectra in so called "rectangular' approximation":

$$n_\gamma \sim 1/E_\gamma. \quad (3)$$

Photonuclear reactions cross-sections was taking as a constant in energy range from the threshold E_{th} until bremsstrahlung end point energy E_0. E_{th} was accepted as $100\,MeV$ and E_0 as $1100\,MeV$. In this case photonuclear cross-section may be determined from the yield with help of equation:

$$\sigma_\gamma = \sigma_q/(\ln \mathrm{E}_0 - \ln \mathrm{E}_{th}). \quad (4)$$

The integrated cross sections ICS and the average cross-section, $\bar{\sigma}$, are given in Table 3.

Table 3. *Comparison of the Kharkiv and Yerevan measured integral cross sections (ICS) and mean cross sections ($\bar{\sigma}$)*

Nucleus	$\sigma_q[cm^2]$ Kharkiv	ICS$[MeV \cdot b]$ Kharkiv	ICS$[MeV \cdot b]$ Yerevan	$\bar{\sigma}[cm^2]$ Kharkiv	$\bar{\sigma}[cm^2]$ Yerevan
^{178m2}Hf	$1.8 \cdot 10^{-28}$	0.03	0.0255	$3.0 \cdot 10^{-29}$	$2.55 \cdot 10^{-29}$
^{172}Hf	$0.42 \cdot 10^{-25}$	4.2	3.74	$4.2 \cdot 10^{-27}$	$3.74 \cdot 10^{-27}$
^{150}Eu	$0.3 \cdot 10^{-28}$	0.011	0.013	$1 \cdot 10^{-29}$	$1.3 \cdot 10^{-29}$
^{133}Ba	$0.36 \cdot 10^{-27}$	0.09	0.141	$0.9 \cdot 10^{-28}$	$1.41 \cdot 10^{-28}$

The important size for understanding of the mechanism of nuclear reaction is the isomer ratio σ_m/σ_g In work [9] this ratio has been determined by an indirect route and it appeared equal

$$\sigma_m/\sigma_g = 0.032. \quad (5)$$

The basic result of this work will be, that we find out and investigate the sample of tantalum containing $\sim 10^{11}$ nucleus of ^{178m2}Hf which further can be used for carrying out of experiments on deexcitation of this high spin and long-living isomer.

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ИССЛЕДОВАНИЕ ФОТООБРАЗОВАНИЯ ДОЛГОЖИВУЩИХ ИЗОТОПОВ $^{178m2}Hf$, ^{172}Hf, ^{150}Eu И ^{133}Ba

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Выход $^{178m2}Hf$ ($T = 31y$), ^{172}Hf (1.87), ^{150}Eu (36.9) и ^{133}Ba (10.5) изотопов измерен после облучения танталовой мишени пучком 1.0 ГэВ Харьковского линейного ускорителя электронов. Получены интегральные и усредненные сечения. Результаты обсуждаются и сравниваются с данными других авторов. Количество образовавшихся атомов $^{178m2}Hf$ в танталовой мишени составляет $(2.7 \pm 0.3) \cdot 10^{11}$.

ВИВЧЕННЯ ФОТОУТВОРЕННЯ ДОВГОЖИВУЧИХ ІЗОТОПІВ $^{178m2}Hf$, ^{172}Hf, ^{150}Eu И ^{133}Ba

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Вихід $^{178m2}Hf$ ($T = 31y$), ^{172}Hf (1.87), ^{150}Eu (36.9) і ^{133}Ba (10.5) ізотопів поміряно після опромінення танталової мішені пучком 1.0 ГеВ Харківського лінійного прискорювача електронів. Отримані інтегральні та усереднені перерізи. Результати обговорюються та порівнюються з даними інших авторів. Кількість утворених атомів $^{178m2}Hf$ у танталовій мішені складає $(2.7 \pm 0.3) \cdot 10^{11}$.