

## THE $^{178m2}\text{Hf}$ ISOMER YIELD IN REACTIONS WITH DIFFERENT PROJECTILES

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The long-lived high-spin  $^{178m2}\text{Hf}$  K-isomer can be produced in nuclear reactions with different projectiles. The reaction yields and cross-sections have been measured in the series of experiments and the results are now overviewed. A new possibility of the  $^{178m2}\text{Hf}$  isomer production using 4.5 GeV Bremsstrahlung is explored and the new series of reactor irradiations are performed to obtain the reliable values of the production and destruction cross-sections for this isomer under thermal and resonance neutron irradiations. The systematics of isomer-to-ground state ratios are drawn and real production capabilities are estimated for the best reactions. Potential isomer applications have been earlier stressed in some publications with probably overestimated expectations.

### 1. Introduction

Nuclear isomers in the mass range close to  $A = 180$  are of special interest because they are characterized by unique combinations of high excitation energy, high spins and K-quantum numbers with long lifetimes. Such features make these isomers extremely attractive for applications to  $\gamma$ -ray pulsed sources because they may store the nuclear excitation energy for long time and also provide the high density of energy. In the classical example of the 31-year-lived  $^{178m2}\text{Hf}$  isomer, the energy density reaches 1.3 GJ/g.

Potential applications of the  $^{178m2}\text{Hf}$  isomer for energy storage and controlled release with the pulsed emission of  $\gamma$ -rays were extensively discussed in literature, Refs. [1 - 3]. Reactor irradiations are known to be most productive for accumulation of radioactive isotopes, thus the reactor yield of the  $^{178m2}\text{Hf}$  isomer has to be measured in reliable experiment.

On the other hand, the  $^{178m2}\text{Hf}$  production in first wall of a fusion reactor was discussed in Ref. [4]. Fusion neutrons should be productive for the synthesis of  $^{178m2}\text{Hf}$  in stellar conditions, as well. Let remind that the radioactive products of nucleosynthesis define the stable isotope abundances. For instance, the production and survival of the  $^{180m}\text{Ta}$  isomer in the cosmogenic radiation bath was described in [5] and in the references therein. The  $\gamma$ -radiation of radionuclides plays important role in the energy balance of stars. Astronomical manifestations of  $^{178m2}\text{Hf}$  are defined by its production and destruction cross-sections within photon and neutron radiation bath in stellar conditions.

An excited nuclear state manifests itself as a metastable isomer when its decay is significantly retarded due to some kind mismatch between the wavefunctions of initial and final states. Decay retardation, being useful for energy accumulation, at the same time is accompanied with the suppression of the isomer production cross-section because of the similar factor of the wavefunctions mismatch. In theory, the conservation of K-quantum number is not an absolute imperative because it is conserved until the axial symmetry of nuclear shape is perturbed. After the experimental studies, it was evident that the K-hindrance factor decreases with the excitation energy growth.

The long-lived states (isomers) are populated in nuclear reactions through the cascade of  $\gamma$ -quanta emitted by the excited reaction residue. At typical residual excitations, the K hindrance must be significantly diminished and the isomer yield should be fortunately increased. But yet, the isomer-to-ground state ratio in many cases remains not high,  $\sigma_m/\sigma_g \ll 1$ , as known from experiments. This is because of high spin of the isomeric state that is not populated well under conservation of total angular momentum.

The correlation of isomer cross-section  $\sigma_m$  with the spin deficit was qualitatively clear even before the experiments reviewed here. However, it does not mean that the isomer yields and m/g ratios could be reliably calculated in theory and used for practical estimations. In reality, the spin distribution of the residual nucleus can not be easily predicted for many reactions. Opposite way, the measured m/g ratio sometimes serves as a basis for estimates of the mean angular momentum of the residual nuclei, for instance, in the spallation reaction with the intermediate-energy protons. Another uncertainty is due to the structure peculiarities of the level scheme and the  $\gamma$ -cascade branching for some individual nucleus. Simplified statistical model calculations may not be very accurate, especially if they are applied to the excited levels below 3 MeV. So, as normally in nuclear physics, the experimental measurements are needed to get reliable values of the reaction cross-section and yield. Thus, the  $^{178m2}\text{Hf}$  production experiments carried out in JINR, Dubna are described below.

## 2. Neutron cross-sections for $^{178\text{m}2}\text{Hf}$

In a first series of experiments, metal Hf samples of about 20 mg were irradiated in an outer channel of the IBR-2 Dubna reactor of the Frank Laboratory of Neutron Physics, Joint Institute for Nuclear Research. The standard method of Cd difference was applied for the isolation of the separate effects of thermal and epi-Cd neutrons. The neutron spectrum at the location of the targets was well-known from previous experiments, but NiCr-alloy samples were nevertheless irradiated as spectators.

In measured spectra from the activated Hf samples,  $\gamma$  lines were observed and their peak areas quantitatively determined for the following radionuclides:  $^{175}\text{Hf}$ ,  $^{179\text{m}2}\text{Hf}$ ,  $^{180\text{m}}\text{Hf}$  and  $^{181}\text{Hf}$ . The bulk of the activity was due to  $^{175}\text{Hf}$  (70 d) and  $^{181}\text{Hf}$  (42.4 d) formed in  $(n, \gamma)$  reactions. Negligible activity was contributed from admixtures of other elements in the Hf material. Only Zr was present in a quantity of about 3 %, while the concentration of other elements was estimated to be on the level of less than 1 ppm. The detected yields of  $^{175}\text{Hf}$  and  $^{181}\text{Hf}$  were used as additional, intrinsic calibrators of the neutron fluxes and then the thermal cross-section  $\sigma_{\text{th}} = (0.44 \pm 0.02)$  b and resonance integral  $I_{\gamma} = (5.8 \pm 0.7)$  b were deduced for  $^{180\text{m}}\text{Hf}$  production in the  $^{179}\text{Hf}(n, \gamma)$  reaction. The resulting values were in good agreement with the tabulated data [6, 7], confirming the accurate calibration of the neutron flux in these irradiations.

A yield of the high-spin  $^{179\text{m}2}\text{Hf}$  isomer was detected and clearly originated from reactions with fast neutrons, since the effect of thermal neutrons was found to be insignificant for its production. This conclusion was definite because bare and Cd-shielded samples showed the same activity of  $^{179\text{m}2}\text{Hf}$  within the standard error. No  $\gamma$  lines from  $^{178\text{m}2}\text{Hf}$  were observed in this first series of irradiations, reflecting its low production yield and the presence of much higher activities of other nuclides.

A second series of experiments was performed in order to significantly improve the sensitivity of the measurements for detection of the low-yield isomers. At the same position in the outer channel, a larger Hf sample was placed for a longer duration. The sensitivity was improved by three orders-of-magnitude, but this was yet insufficient for observation of  $^{178\text{m}2}\text{Hf}$ . A strong increase in the neutron flux was then sought to increase the production of this isomer. In the reactor, an inner channel was available that allowed an irradiation near, but outside, the active core within a cylindrical region shielded by a 3-mm layer of  $\text{B}_4\text{C}$ . A Hf sample was exposed there for eighteen days, after which decay of the resulting activity was followed for two years. Finally, activity of  $^{178\text{m}2}\text{Hf}$  was successfully observed and its yield determined after the third series of irradiations.

At the inner location, the thermal flux was determined in earlier experiments to be about  $0.5 \cdot 10^{12}$  neutrons/( $\text{cm}^2 \text{ s}$ ), thus during the eighteen-day irradiation a fluence near  $10^{18}$  n/ $\text{cm}^2$  could be accumulated. In the present experiments, the value of flux was not used explicitly since all measurements were carried out in a relative mode by comparing the activities of  $^{178\text{m}2}\text{Hf}$ ,  $^{179\text{m}2}\text{Hf}$  and  $^{180\text{m}}\text{Hf}$  nuclei to those of spectators and intrinsic calibrators  $^{175}\text{Hf}$ ,  $^{181}\text{Hf}$  and  $^{95}\text{Zr}$  which were present within the targets. Such a method is reliable and accurate in the presence of shields. Data processing requires special care for high-sensitivity measurements of neutron-production cross sections. The burnup of isomeric nuclei produced during the irradiation must be taken into account when the neutron fluence exceeds  $10^{20}$  n/ $\text{cm}^2$ . In the present irradiations, however, even in the inner channel the fluence was 100 times lower. This is due to the fact that IBR-2 is a pulsed reactor constructed specially for time-of-flight spectroscopy, not to achieve a high mean power. With this fluence, burnup of high-spin hafnium isomers or other double neutron-capture reactions may be expected to have a probability below  $10^{-3}$  compared to the probability for single neutron capture.

Description of the data processing detail can be found in Ref. [8]. The production and destruction processes separately were characterized now for the  $^{178\text{m}2}\text{Hf}$  nucleus by thermal  $\sigma_{\text{th}}$  and resonance  $I_{\gamma}$  values.

Neutron capture cross-sections for Hf nuclides are summarized in Table 1 with literature data taken from Ref. [6]. The present results are given for the reactions of particular interest,  $^{177}\text{Hf}(n, \gamma)^{178\text{m}2}\text{Hf}$ ,  $^{178}\text{Hf}(n, \gamma)^{179\text{m}2}\text{Hf}$  and  $^{178\text{m}2}\text{Hf}(n, \gamma)^{179\text{g},\text{m}2}\text{Hf}$ . The latter reaction which is responsible for  $^{178\text{m}2}\text{Hf}$  destruction (burnup) may be especially exotic and is discussed in more detail below.

More than thirty years ago, activity of the  $^{178\text{m}2}\text{Hf}$  isomer was observed [9] from isotopically-enriched hafnium targets irradiated within a high-power reactor. Gamma spectra of this isomer were carefully studied using state-of-the-art techniques available at that time. However, the production cross section given in Ref. [9] may not be very accurate because it was determined by only considering the effect of thermal neutrons, neglecting the resonance neutron flux, and assuming that the destruction cross-section was at a rather low level of less than 20 b. It is well-known that a purely thermal flux does not exist in any

reactor near its active core and the presence of resonance neutrons changes the yields of reaction products. Also the  $^{178m2}\text{Hf}$  yield could be strongly reduced due to the burnup process at the fluences applied in the work of Ref. [9],  $\geq 10^{21}$  n/cm<sup>2</sup>. The degree of burnup definitely depends on the values of  $\sigma_{\text{th}}$  and  $I_\gamma$  for the isomeric nucleus, but the relatively low  $\sigma_{\text{th}}$  used in Ref. [9] was unexpected.

Table 1. Neutron capture cross-sections for hafnium isotopes

Target	$J^\pi_t$	Product	$J^\pi_p$	$\sigma_{\text{th}}$ , b	$I_\gamma$ , b	$\sigma_m/\sigma_g$	Reference
$^{174}\text{Hf}$	$0^+$	$^{175}\text{Hf}$	$5/2^-$	561	436	—	[6]
$^{176}\text{Hf}$	$0^+$	$^{177}\text{Hf}$	$7/2^-$	23.5	880	—	[6]
$^{177}\text{Hf}$	$7/2^-$	$^{178g}\text{Hf}$	$0^+$	373	7,173	—	[6]
		$^{178m1}\text{Hf}$	$8^-$	0.96	—	$2.6 \cdot 10^{-3}$	[6]
		$^{178m2}\text{Hf}$	$16^+$	$2.6 \cdot 10^{-6}$	$5 \cdot 10^{-5}$	$(7 \pm 2) \cdot 10^{-9}$	Present
$^{178}\text{Hf}$	$0^+$	$^{179g}\text{Hf}$	$9/2^+$	84	1,950	—	[6]
		$^{179m1}\text{Hf}$	$1/2^-$	53	—	0.63	[6]
		$^{179m2}\text{Hf}$	$25/2^-$	$\leq 2 \cdot 10^{-4}$	$\leq 1.3 \cdot 10^{-3}$	$\leq 2.4 \cdot 10^{-6}$	Present
$^{178m2}\text{Hf}$	$16^+$	$^{179g}\text{Hf}$	$9/2^+$	190	4,500	$(0.24 \pm 0.07)$	Present*
		$^{179m2}\text{Hf}$	$25/2^-$	$45 \pm 5$	$1,060 \pm 60$	—	[23]
$^{179}\text{Hf}$	$9/2^+$	$^{180g}\text{Hf}$	$0^+$	41	630	—	[6]
		$^{180m}\text{Hf}$	$8^-$	0.45	6.9	$1.1 \cdot 10^{-2}$	[6]
$^{180}\text{Hf}$	$0^+$	$^{181}\text{Hf}$	$1/2^-$	13.04	35.0	—	[6]

\* Obtained by comparison of present results with those of Ref. [9].

Recently, the burnup cross-section due to thermal neutrons was measured in Ref. [10] for the  $^{177m}\text{Lu}$  isomer and a value of  $\sigma_{\text{th}} = 590$  b was obtained, including both neutron capture and superelastic scattering components. Noting that  $^{177m}\text{Lu}$  also possesses high spin and excitation energy similar to  $^{178m2}\text{Hf}$ , one may expect them to have comparable burnup cross-sections. This does not contradict the known correlation of  $\sigma_{\text{th}}$  and  $I_\gamma$  values with the c.n. resonance density.

Such arguments motivated the present independent measurements of the  $^{178m2}\text{Hf}$  production cross section in reactor irradiations and an evaluation of the role of burnup which depends on neutron flux. The burnup cross-section is deduced using the following method. Observed in [9] yield of  $^{178m2}\text{Hf}$  has been simulated applying the production  $\sigma_{\text{th}}$  and  $I_\gamma$  values determined in the present experiment. The burnup cross-section has been found to be much higher than assumed in [9]. Accordingly, the production cross-section was underestimated in [9] because they almost neglected the burnup process significant at high-fluence irradiations.

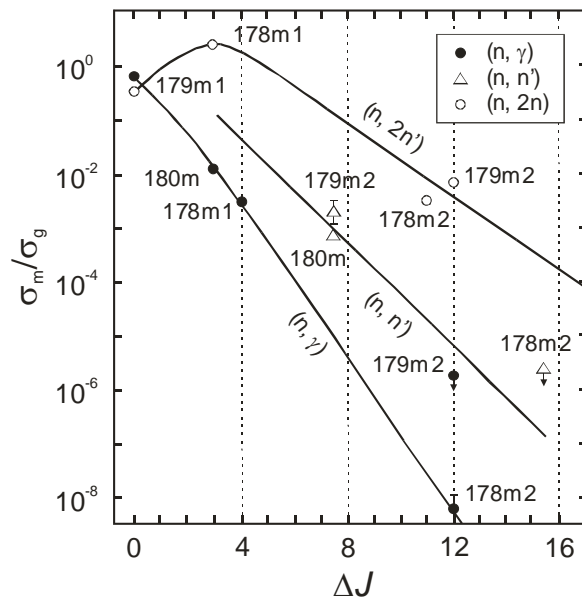


Fig. 1. Measured values of  $\sigma_m/\sigma_g$  ratio versus spin deficit ( $\Delta J$ ) parameter for Hf isomers produced in neutron-induced reaction.

The  $\sigma_m/\sigma_g$  ratios could also be deduced from the present results. They are reduced in Table 1, and shown in Fig. 1 as a function of the spin deficit that corresponds to the population of the definite isomer in the definite reaction. The results for reactions with fast neutrons are taken from Refs. [4, 6, 11] and also are displayed in Fig. 1. The reaction mechanism details are discussed in Ref. [8], and in particular, it is indicated there that the superelastic scattering may contribute significant part in the total cross-section of the thermal neutron interaction with isomeric nuclei. Such conclusion seems in accordance with Refs. [10, 12, 13].

### 3. $^{178m2}\text{Hf}$ production with Bremsstrahlung

Photon-induced nuclear reactions were systematically studied in [14,15] at the irradiations of  $^{nat}\text{Ta}$ ,  $^{nat}\text{Hf}$  and  $^{178m2}\text{Hf}$  targets with Bremsstrahlung at the end-point energy of 23.5 MeV. The activation techniques was applied,

and as many as 18 yields of the  $(\gamma, \gamma)$ ;  $(\gamma, n)$ ;  $(\gamma, p)$ ;  $(\gamma, 2n)$  and  $(\gamma, \alpha)$  reactions were successfully measured. Among them, took place the reactions leading to the population of isomeric and ground states, thus the isomer-to-ground state ratios were deduced. Most original was the observation of reactions with the isomeric  $^{180m}\text{Ta}$  and  $^{178m2}\text{Hf}$  nuclei and the demonstration that the high-spin isomers are populated easy in the reaction product when the target nucleus is also a high-spin isomer. This does not contradict the correlation of yield with the nuclear spin difference of the target and product.

At the attempts of the  $^{178m2}\text{Hf}$  production with Bremsstrahlung, its low activity could not be distinguished in the presence of other radionuclides induced in the irradiated  $^{\text{nat}}\text{Hf}$  target [14, 15]. But later, at the 22 MeV end-point Bremsstrahlung irradiation, higher sensitivity of measurements was reached, Ref. [16], and the yield of  $^{178m2}\text{Hf}$  was observed and attributed to the  $^{179}\text{Hf}(\gamma, n)$  reaction. The latter reaction was most productive because of highest yield of the  $(\gamma, n)$  products at such energy and because of highest spin value of the  $^{179}\text{Hf}$  nuclei among other stable Hf isotopes. The isomer-to-ground ratio was found to be

$$Y_m/Y_g = (3.5 \pm 1.0) \cdot 10^{-5}. \quad (1)$$

The estimated yield of  $^{178m2}\text{Hf}$  of about  $4 \cdot 10^7$  atoms/s in optimum condition is out of interest for the production of the isomer in an amount needed for the isomeric target preparation.

It would be very probable that the yield must be higher with the electron beam of higher energy, and the experiment has been performed using a 4.5 GeV electron beam at the Yerevan synchrotron. A stack of Ta foils was exposed to the Bremsstrahlung generated in the W converter. The long-base collimation system and relatively thin converter and target samples were used for the better definition of experimental conditions. After long cooling time, the activity of Ta foils was measured with the 20 % efficiency Ge gamma-spectrometer. Only long-lived products have been survived to the time of measurements such as  $^{178m2}\text{Hf}$ ,  $^{172}\text{Hf}$ ,  $^{150}\text{Eu}$  and  $^{133}\text{Ba}$ , and they could be quantitatively determined. In more details, the experiment is described in Ref. [17].

The yield of  $^{178m2}\text{Hf}$  has been measured and the isomer-to-ground state ratio is found to be

$$Y_m/Y_g = (0.032 \pm 0.010), \quad (2)$$

i.e. much higher the value given above, (1), for the reaction induced by Bremsstrahlung at 22 MeV. The transmutation of  $^{181}\text{Ta}$  into  $^{178m2}\text{Hf}$  requires the emission of proton and two neutrons, however the reaction  $^{181}\text{Ta}(\gamma, p2n)^{178m2}\text{Hf}$  can be written only formally because at high energy, mesons are generated and emitted, not only nucleons. Respectively, a variety of reactions arises, all leading to the same product. Photon absorption at  $E_\gamma \geq 200$  MeV involves the mechanism of meson generation and corresponded peaks are strongly manifested in the excitation function. Above 1200 MeV, the absorption cross-section decreases and reaches almost constant, asymptotic value of about 0.12 mb/nucleon.

At low energies,  $E_\gamma \leq 200$  MeV, the quasi-deuteron mechanism makes the highest contribution, and even lower – the tails of E1 and E2 giant multipole resonances are of importance. The yield of  $^{181}\text{Ta}(\gamma, p2n)$  reaction at  $E_\gamma \leq 50$  MeV should be deteriorated by the energy deficit. So that, one may conclude that in the irradiations with 4.5 GeV Bremsstrahlung, the  $^{178m2}\text{Hf}$  isomer is produced mostly due to the absorption of photons in the range from 50 to 1200 MeV. After understanding of this, it would be easy to accept the ratio (2) that is comparable with the values determined in [18] for the Ta spallation by protons of intermediate energy, (600 - 300) MeV. At few hundred MeV range, the photon absorption transfers to the nucleus enough energy for the emission of many particles, like to the proton-induced spallation.

The estimated m/g ratio also leads to the conclusion that a reasonably high angular momentum is acquired by the residual nucleus in the reactions with high energy photons, not only with protons. Measured yield of  $^{178m2}\text{Hf}$  allows estimating the maximum achievable productivity of the reaction with 4.5 GeV Bremsstrahlung. In a view of optimization, thicknesses of the target and the converter should be enlarged. In this way, more quanta can be created and used for nuclear reactions, although obviously the absorption will also be increased. A reasonable compromise would be to unify both converter and target which would then to be a rather thick sample of Ta. The problem of optimization of such a unified assembly was solved analytically in [11] and the result is given in Fig. 2.

In addition to the major production by Bremsstrahlung, the reactions induced directly by electrons and the secondary processes are taken into account in the simplified approximation. With the Ta sample

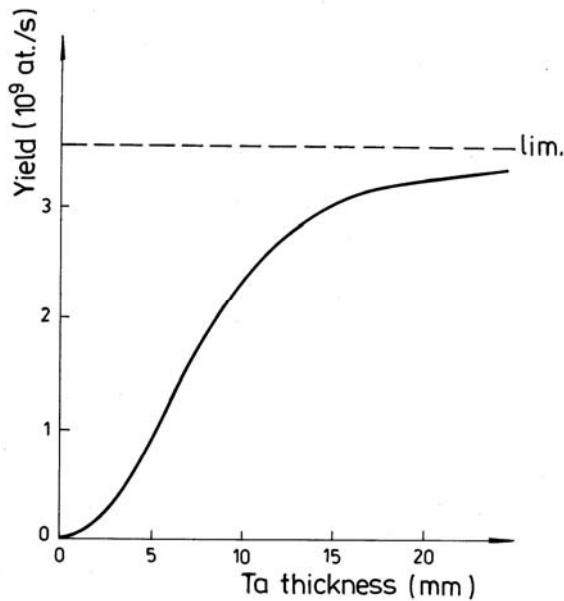


Fig. 2. Yield of the  $^{178m2}\text{Hf}$  as a function of thickness of the Ta sample exposed to  $100\ \mu\text{A}$  electron beam at  $4.5\ \text{GeV}$ .

20 mm in thickness, a number of  $^{178m2}\text{Hf}$  atoms reaches the value of

$$Y = 3.2 \cdot 10^9 \text{ atoms/s} \cdot 100\ \mu\text{A}. \quad (3)$$

The photon-conversion efficiency and, respectively, the nuclear reaction yield must be increased by some factor at channeling conditions, when electrons are directed along the crystal axes or planes. But with thick sample, such factor should not be significant.

#### 4. Spallation by intermediate energy protons

It is established that the largest quantity of  $^{178m2}\text{Hf}$  was produced at Los Alamos with 800 MeV protons from a high-current accelerator (formerly LAMPF). The advantage of this method was the ability to accumulate the isomer as a by-product within a massive Ta beam dump during the operation of the accelerator for other experiments. The shortcoming was due to a very high activity of other radionuclides produced in Ta fragmentation. The yield of  $^{178m2}\text{Hf}$  was reported

in Ref. [19], but the experimental details were described schematically and the productivity was only estimated. Recently, the reactions of proton-induced spallation were systematically studied for the Ta, W and Re targets of natural isotopic composition and for enriched  $^{186}\text{W}$  target, as well, Refs. [18, 20], using the 660 MeV synchrocyclotron at Dubna. The yields of the long-lived high-spin isomers of  $^{179m2}\text{Hf}$ ,  $^{178m2}\text{Hf}$  and  $^{177m}\text{Lu}$  are quantitatively determined and the measured values can be used for the productivity optimization in some future irradiations.

Let's characterize briefly these recent experiments. The metal foil targets fixed to the cooled Al backing were inserted for irradiation into the internal beam of protons of the Dubna synchrocyclotron (phasotron). Choosing the position of a target inside the accelerator, it was possible to vary the beam energy from 100 to 650 MeV on the basis of known calibrations.

Gamma-spectroscopy measurements of the irradiated samples were performed after a "cooling" period of 1 month because of the high activity of short-lived radionuclides accumulated during the activation. After the measurements, the samples were dissolved for chemical processing and isolation of the Hf fraction. The decay activity of the long-lived  $^{178m2}\text{Hf}$  state is rather low as compared to other nuclides activity and the chemical isolation was necessary to achieve good accuracy for the  $^{178m2}\text{Hf}$  yield. The gamma spectra were measured for the chemically isolated elemental fractions, in addition to the full activity spectra measured before.

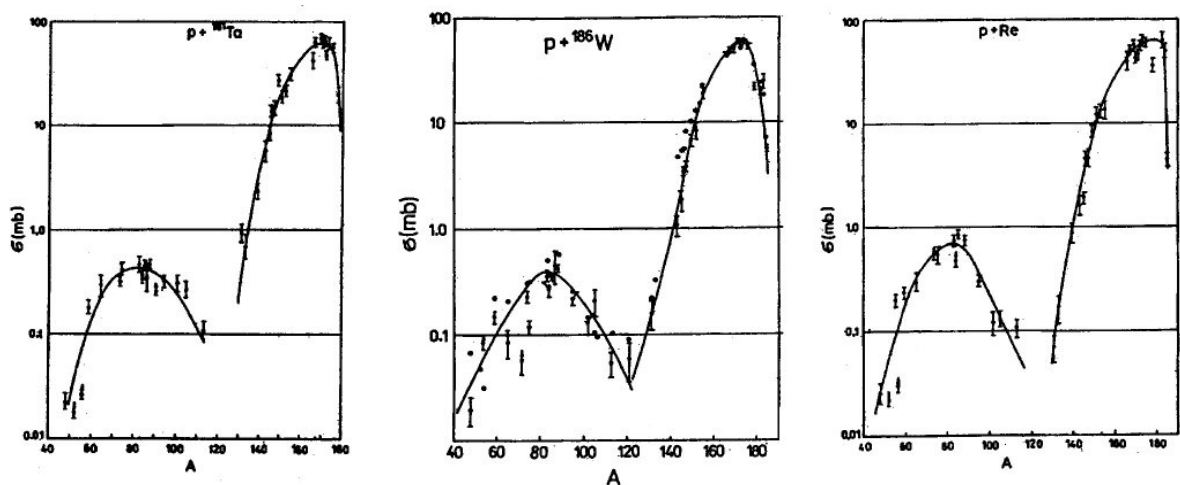


Fig. 3. Mass-distribution of the nuclides produced after the irradiation of  $^{\text{nat}}\text{Ta}$ ,  $^{186}\text{W}$  and  $^{\text{nat}}\text{Re}$  targets with protons of 630 MeV mean energy.

**Table 2. Cross-sections and isomer-to-ground state ratios for the formation of high-spin isomers after the spallation of different targets**

Nuclide	Target			
	<sup>nat</sup> Ta	<sup>nat</sup> W	<sup>186</sup> W	<sup>nat</sup> Re
Cross-section $\sigma$ , mb				
<sup>179m2</sup> Hf	0.52	0.36	0.80	0.12
<sup>178m2</sup> Hf	0.31	0.18	0.48	0.13
<sup>177m</sup> Lu	0.15	0.13	0.26	0.04
<sup>178</sup> W	5.9	23	21.8	36
<sup>175</sup> Hf	56	55	55.6	59
<sup>172</sup> Hf	47	53.5	57.4	55
<sup>173</sup> Lu	61	61	60	61
$\sigma_m/\sigma_g$ ratio				
<sup>179m2</sup> Hf	0.040	0.14	0.25	0.24
<sup>178m2</sup> Hf	0.021	0.044	0.092	0.14
<sup>177m</sup> Lu	0.103	0.21	0.29	0.40

The cross-sections of neighboring radionuclides with strong gamma activity are also reported in Table 2 because they define the radioactive contaminations of the final isomeric material. In Table 2, the  $\sigma_m/\sigma_g$  values of the order of (0.1 - 0.2) are the highest known in literature for the production of the high-spin isomers. Consequently, the angular momentum of the spallation residues cannot be low; it is probably as high as 10h or more. The dependence of the  $\sigma_m/\sigma_g$  values on the target mass number indicates a growth of the residual spin as the number of emitted nucleons increases.

The highest production cross-sections for Hf and Lu isomers have been found at the case of enriched <sup>186</sup>W target, but in practice, it must be very expensive if one uses kilograms of the isotopically enriched substance as a target. The reasonable substitution would be the <sup>nat</sup>Ta target, not expensive and also characterized with rather good cross-sections for the isomer production. The absolute maximum of the <sup>178m2</sup>Hf yield in p + Ta irradiation is estimated to be as high as 10<sup>12</sup> nuclei/s due to extremely high beam current achievable at Los Alamos, of about 1 mA, and assuming thick target of 10 cm. This corresponds to accumulation of about 10 mg of the isomer per year of effective irradiation and can be estimated as a maximum of production rate using existing and known in literature accelerator facilities.

### 5. Reaction with <sup>4</sup>He ions and other reactions at low energy

A method of the <sup>178m2</sup>Hf isomer production using the <sup>176</sup>Yb(<sup>4</sup>He,2n) reaction was proposed and studied in Refs. [21, 22]. A high-purity isomeric material was accumulated in the extensive irradiations with high-current <sup>4</sup>He-ion beam at Dubna U-200 cyclotron. The amount of the isomeric substance was enough to prepare the targets for investigation of the nuclear reactions with the high-spin exotic isomer and some of them were successfully observed and studied. However, the general deficit of the material amount has restricted a development of such studies. In total, it was possible to accumulate only about 1  $\mu$ g <sup>178m2</sup>Hf past high-intensity long irradiations with <sup>4</sup>He beam. Absolute productivity is not high because the energy losses of <sup>4</sup>He-ions in matter allow using only of about 0.2 g the <sup>176</sup>Yb-target material and not more at each irradiation. Some details of this method are described below taking into account that it was productive for the performance of many experiments with <sup>178m2</sup>Hf, reviewed in [23].

The excitation function of <sup>178m2</sup>Hf in the <sup>176</sup>Yb(<sup>4</sup>He,2n) reaction was measured and the cross-section showed a peak near  $E_\alpha = 32$  MeV. The optimum energy range of (28 - 36) MeV was deduced with mean cross-section of about 7 mb. Respectively, the mean isomer-to-ground state ratio was estimated to be  $\sigma_m/\sigma_g \approx 0.05$ . The stopping of <sup>4</sup>He ion from 36 to 28 MeV in Yb<sub>2</sub>O<sub>3</sub> corresponds to the layer thickness of 70 mg/cm<sup>2</sup>. The target construction was specially designed to prevent losses of the Yb oxide material under <sup>4</sup>He<sup>++</sup> ion beam current up to 100  $\mu$ A. The absolute yield of the <sup>178m2</sup>Hf nuclei reached a value of  $Y = 5 \cdot 10^8$  nuclei/(s·100  $\mu$ A) that should be compared with the yield of other reaction.

Relatively high cross-section of the <sup>176</sup>Yb( $\alpha$ , 2n)<sup>178m2</sup>Hf reaction leads to the idea of possible use such reactions as <sup>181</sup>Ta(p,  $\alpha$ ); <sup>178</sup>Hf( $\alpha$ ,  $\alpha'$ ); <sup>179</sup>Hf( $\alpha$ ,  $\alpha'$ n) and <sup>176</sup>Lu(<sup>7</sup>Li,  $\alpha$ n) at low energies. Production of <sup>178m2</sup>Hf in these reactions was not yet studied, but it was known from the nuclear-reaction phenomenology that all

In total, as many as about 70 radionuclides were identified, and the mass distribution of fragmentation products could be plotted, as a result. For example, it is shown in Fig. 3 for the cases of <sup>nat</sup>Ta, <sup>186</sup>W and <sup>nat</sup>Re targets at 630 MeV. Master tables of the radionuclide yields and cross-sections are given in Refs. [18, 20] for different targets and energies. Many details of the  $\gamma$ -spectroscopic measurements, the calibration and evaluation procedures, etc. are also given there. Fig. 3 characterizes here only basic properties common for the variety of studied fragmentation reactions. Two peaks in Fig. 3 correspond to the fission and spallation reaction mechanisms. For our purpose now, most important are the production cross-section for nuclides and isomers near and below A = 180.

We focus, in the following, on the discussion of the spallation yield of the long-lived high-spin isomers of <sup>177m</sup>Lu, <sup>178m2</sup>Hf and <sup>179m2</sup>Hf. The cross-sections and isomer-to-ground state ratios  $\sigma_m/\sigma_g$  are compared in Table 2 for the production of isomers with 630 MeV proton beam using different targets.

of them are more or less probable processes at energy well above the interaction barrier. Means, the total cross-section should be of about hundreds millibarn, and reasonably high angular momentum of the product provides not very low isomer-to-ground state ratio. So that, the  $^{178m2}\text{Hf}$  production cross-section is expected to be comparable with the known for the  $^{176}\text{Yb}(^4\text{He}, 2n)$  reaction, though not much more preferable.

Special attraction is the  $^{176}\text{Lu}(^7\text{Li}, \alpha n)$  reaction because  $^{176}\text{Lu}$  is a unique case of the high-spin (7) target. Respectively, the  $\sigma_m/\sigma_g$  ratio can reach a level of 50 % in this reaction, i.e. to be 10 times higher as compared to  $^{176}\text{Yb}(\alpha, 2n)$ . But at the same time, a maximum current of the  $^7\text{Li}$  ions is restricted due to the higher density of energy released in the target layer. In total, a factor of (3 - 5) can be the gain if one uses the high-current  $^7\text{Li}$  beam and the 90 % enriched  $^{176}\text{Lu}$  target of the best design in the sense of heat removal. A few orders of magnitude higher productivity is yet invisible. Nevertheless, indicated above reactions should be experimentally studied in order to operate with the reliable results, instead of some realistic estimations.

## 6. Comparison of different reactions

In Table 3, the absolute productivities of the reactions induced by different projectiles are compared for the  $^{178m2}\text{Hf}$  isomer, following the measurements discussed above. The comparison is somewhat conventional, because the absolute yield depends on the beam intensity and on the appropriate amount of the target material. Despite that, we want to get some ranking of reactions; therefore they should be compared at similar conditions in respect to input parameters characterizing a strength of irradiation. For instance, a beam current is chosen to be 100  $\mu\text{A}$  for all accelerators, and the same target thickness is assumed unless it is physically restricted due to the flux absorption, or the target material price. The chosen parameters are absolutely real, i.e. already reached at the facilities described in literature and remaining in operation today. No extraordinary powerful systems are involved in the comparison. The quantities of enriched target isotopes are restricted by the value of 10 gram because of high price of such substances.

Table 3. Quantitative parameters characterizing the different methods of the  $^{178m2}\text{Hf}$  isomer production

Projectile	Photons, $\gamma$		Neutrons, $^1_0\text{n}$		Protons, $^1_1\text{H}^+$		Alphas, $^4_2\text{He}^{++}$
$E_{\text{max}}$ , MeV	22	4500	thermal	14	650		36
Intensity	100 $\mu\text{A}$	100 $\mu\text{A}$	$5 \cdot 10^{14}$	$10^{13}$	100 $\mu\text{A}$		100 $\mu\text{A}$
Target	$e^-$	$e^-$	$\text{n/cm}^2\text{s}$	$\text{n/cm}^2\text{s}$			
Amount	$^{179}\text{Hf}$	Ta	$^{177}\text{Hf}$	$^{179}\text{Hf}$	Ta	$^{186}\text{W}$	$^{176}\text{Yb}$
$\sigma_m$ (mb)	10g (total)	33 g/cm <sup>2</sup>	1g (total)	10g (total)	33 g/cm <sup>2</sup>	5 g/cm <sup>2</sup>	0.07 g/cm <sup>2</sup>
$\sigma_m/\sigma_g$	-	-	$2 \cdot 10^{-3}$	7.3	0.3	0.5	7
Productivity, atoms/s	$3 \cdot 10^{-5}$	0.03	$7 \cdot 10^{-9}$	$3.5 \cdot 10^{-3}$	0.02	0.09	0.05
Rank	$4 \cdot 10^7$	$3 \cdot 10^9$	$3.4 \cdot 10^5$	$2.5 \cdot 10^9$	$2 \cdot 10^{10}$	$5 \cdot 10^9$	$5 \cdot 10^8$
	6	3	7	4	1	2	5

Remark:  $\sigma_m$  is not given for the Bremsstrahlung induced reactions because of the continuous spectrum of photons. The yield ratio was measured.

The ranks in Table 3 reflect the absolute yield of the reaction at comparable conditions. The p+Ta spallation is the most productive and its first rank could be expected. A productivity of  $2 \cdot 10^{10}$  atoms/s is given in Table 3 as the best, but in the condition comparable with other reactions. The absolute maximum has been estimated above assuming that the beam current can be as high as 1 mA with the target thickness of 10 cm. Even so, the production of  $^{178m2}\text{Hf}$  is restricted by mg amounts, while effective applications require kilograms. The latter amount is out of reality, at least at modern status of experimental physics. Despite such orders of magnitude mismatch, the results reviewed at the present report and summarized in Table 3 are of importance. They give a real basis for some speculations and estimations and also stimulate a nuclear-science progress in understanding of the processes with high-spin nuclear states.

For nuclear reaction theory, even more significant are the isomer-to-ground state ratios, in addition to the production yields. In  $\sigma_m/\sigma_g$  ratio, the scale factors in the reaction cross-section are excluded, and the ratio value has eventually strong implications for study of the nuclear reaction mechanism. In particular, mean angular momentum of the reaction residue has strong influence on the  $\sigma_m/\sigma_g$  ratio. Fortunately,  $\sigma_m/\sigma_g$  is measurable, and for some reactions, the residual spin can be figured out in theory. Thus, the correlation between  $\sigma_m/\sigma_g$  and the reaction-product spin can be verified after the measurements. Such

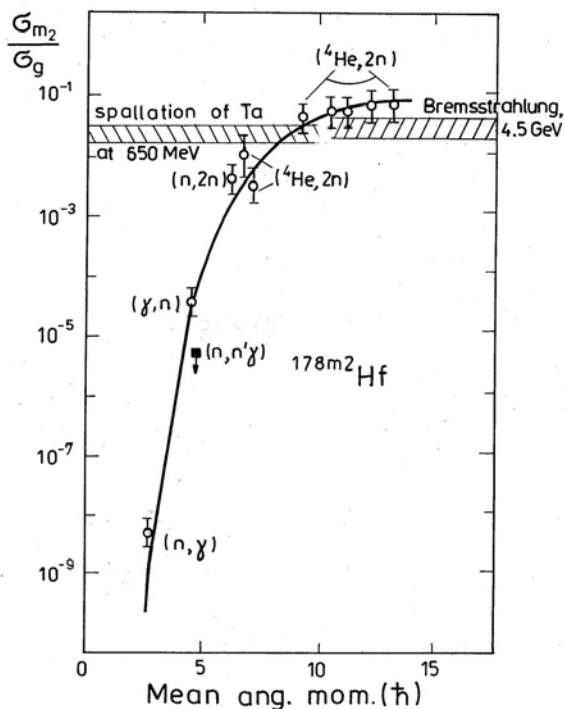


Fig. 4. Systematics of the isomer-to-ground state ratio versus the reaction product spin for the  $^{178m2}\text{Hf}$  isomer as is measured in reactions with different projectiles.

this exotic nucleus, the  $^{178m2}\text{Hf}$  high-spin isomer can be produced in the spallation reaction with much higher isomer-to-ground state ratio. The productivity can be enhanced by a factor of 10 using such a target, as compared to the regular  $^{nat}\text{Ta}$  target. This follows from the systematics of Fig.4. However, a kilogram amount of the 90 % enriched  $^{180m}\text{Ta}$  material is out of reality today. Creation of a special facility for the  $^{180m}\text{Ta}$  separation and the accumulation it in large amount should be extremely expensive, and even technical restrictions for that are not yet clear.

Ignoring the cost arguments, one can deduce the absolute maximum of the productivity, as following:  $Y_{\max} = 10^{13}$  atoms/s, if a 1 kg target made of 90 % enriched  $^{180m}\text{Ta}$  is exposed to the 800 MeV protons at a beam current of 1 mA. In this way, of about 100 mg  $^{178m2}\text{Hf}$  can be accumulated in one-year effective irradiation run.

## 7. Summary

Known experimental results are reviewed for the production cross-sections of the  $^{178m2}\text{Hf}$  exotic isomer. The isomer-to-ground state ratios are systematized. The productivity of different reactions is compared and they are ranked in an order of decreasing yield. Respectively, the values are estimated for the  $^{178m2}\text{Hf}$  material amount that can be accumulated in irradiations with different projectiles. Realistic parameters of existing experimental facilities restrict the production of large amount, while the discussed in literature applications require by orders of magnitude higher quantities. A thinkable maximum of productivity is estimated in assumption that the parameters of irradiations can be significantly enlarged using new facilities specially constructed for such irradiations and new isotope separator for the preparing of a kg amount of the  $^{180m}\text{Ta}$ ,  $^{179}\text{Hf}$  and  $^{176}\text{Lu}$  isotopes.

Experiments on the  $^{178m2}\text{Hf}$  isomer production could be carried out only within collaborations under the definite financial support and the corresponding acknowledgments are expressed in Refs. [8, 16 - 18, 20 - 22]. These results are used in the present review article.

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dependence is plotted in Fig.4 for the  $^{178m2}\text{Hf}$  isomer production. When the reaction product spin  $I_r$  is increasing, the spin-deficit parameter  $\Delta I$  is respectively decreasing, and the probability of isomer population is growing up. Such natural behaviour is experimentally confirmed and quantitatively characterized in Fig. 4.

It would not be easy to calculate in theory the  $\bar{I}_r$  value for the reactions with the intermediate-energy protons or with high-energy Bremsstrahlung. In such cases, the systematics of Fig. 4 can be used for estimation of the  $\bar{I}_r$  parameter basing on the measured  $\sigma_m/\sigma_g$  ratio. This way, the unique information is deduced confirming that the reaction residue receives rather high spin, like  $\bar{I}_r \sim 10 \hbar$ , both in proton-induced spallation and in the reaction of photon absorption at GeV energies. In addition, the systematics can be used in application to other processes for estimation of the production possibilities with not yet studied reactions.

At the end, let's discuss a somewhat fantastic idea of using the  $^{180m}\text{Ta}$  material as a high-productivity target. Because of high spin (9<sup>+</sup>) of

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## ВЫХОД ИЗОМЕРА $^{178m2}\text{Hf}$ В РЕАКЦИЯХ С РАЗЛИЧНЫМИ НАЛЕТАЮЩИМИ ЧАСТИЦАМИ

С. А. Карамян

Долгоживущий высокоспиновый К-изомер  $^{178m2}\text{Hf}$  может быть получен в ядерных реакциях с различными частицами. В серии экспериментов определялись сечения и выход реакций, дается обзор полученных результатов. Исследована новая возможность получения  $^{178m2}\text{Hf}$  с помощью тормозного излучения при энергии 4,5 ГэВ, а также проведена новая серия реакторных облучений. Определены надежные значения сечений получения и „сжигания” этого изомера под действием тепловых и резонансных нейтронов. Систематизированы изомерные отношения и оценена реальная продуктивность лучших реакций для накопления  $^{178m2}\text{Hf}$ . В некоторых публикациях были явно переоценены потенциальные возможности практического применения данного изомера.

## ВИХІД ІЗОМЕРУ $^{178m2}\text{Hf}$ У РЕАКЦІЯХ З РІЗНИМИ НАЛІТАЮЧИМИ ЧАСТИНКАМИ

С. А. Карамян

Довгоіснуючий високоспіновий К-ізомер  $^{178m2}\text{Hf}$  може бути отриманий у ядерних реакціях з різними частинками. У ряді експериментів визначались перерізи та вихід реакцій, наведено огляд одержаних результатів. Досліджено нову можливість одержання  $^{178m2}\text{Hf}$  за допомогою гальмівного випромінювання з енергією 4.5 ГеВ, а також виконано нову серію реакторних опромінювань. Визначено надійні значення перерізів одержування та „спалювання” цього ізомеру при дії теплових та резонансних нейтронів. Систематизовано ізомерні відношення та оцінено реальну продуктивність кращих реакцій для накопичення  $^{178m2}\text{Hf}$ . У деяких публікаціях було явно переоцінено потенційні можливості практичного застосування даного ізомеру.