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COMPARATIVE ANALYSIS OF THE  $^{178m2}\text{Hf}$  YIELD  
AT REACTIONS WITH DIFFERENT PROJECTILES

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Карамян С. А. E15-2004-116  
Сравнительный анализ выходов изомера  $^{178m2}\text{Hf}$  в реакциях  
под действием различных частиц

Долгоживущий высокоспиновый  $K$ -изомер  $^{178m2}\text{Hf}$  может быть получен в ядерных реакциях под действием разных бомбардирующих частиц. Проведена серия экспериментов для измерения выходов и сечений реакций, результаты подытожены в настоящей работе. Построена систематика изомерных отношений и оценена реальная продуктивность лучших реакций. Такие итоги актуальны, поскольку исследования с изомерами важны как для развития ядерно-научных знаний, так и для возможных приложений. В популярной литературе ожидания явно переоценены в отношении потенциального использования изомеров. Реальные возможности ограничены, в частности, из-за недостаточного выхода при получении изомеров, а также из-за других ограничений.

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Karamian S. A. E15-2004-116  
Comparative Analysis of the  $^{178m2}\text{Hf}$  Yield at Reactions  
with Different Projectiles

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. The systematics of isomer-to-ground state ratios are drawn and real production capabilities are estimated for the best reactions. Such a summary is relevant to the significance of the isomer studies both for the nuclear-science knowledge and for possible applications. Potential isomer applications have been earlier stressed in popular publications with probably overestimated expectations. The real possibilities are restricted in part by the production yield and by other shortcomings as well.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR.

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## 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.

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. Such hindrances for the decay typically arise because of collective deformation of a nucleus in excited state, and either of the angular momentum step at the decay transition or of a structure inhibition. In the region of statically deformed nuclei, the structure hindrance may play an important role, in addition to the selection rules by the spin and parity ( $I^\pi$ ) for the multipole electromagnetic transitions. For instance, a change in the orientation of the angular momentum vector generates special structure inhibition known in literature as  $K$  hindrance. Remind that  $K$  is a quantum number of the  $I$  vector projection to the deformation axis. The axial symmetry of the nuclear deformation is a major assumption. Many  $K$ -hindered isomers are known, and their properties are described in literature (see, for instance, [1–4] and references therein).  $^{178m2}\text{Hf}$  is one of the clearly identified  $K$  isomers because the  $K$ -hindrance factor is as high as of about  $10^9$  in this case. Without the latter value, the isomeric state should decay fast, with half-life of 0.8 s, instead of real  $T_{1/2} = 31$  y. Thus, the methods available for the unique isomer production are of interest in different aspects, in particular, for extension of the exotic-nuclei phenomenology.

A field of the present work is intentionally narrowed to the nuclear reaction features only. The nuclear structure properties are touched as far as they influence the isomer production cross-section. The discussion of applications is excluded because we have no intention to support a «sensation» appeared in some newspapers and magazines, for instance, in the «Washington Post», «Der Spiegel», etc. One can find more professional and responsible approach in the review articles published in physical journals [1–5]. They contain the explanation that the isomer research is at a very basic stage, far from real use of the isomer sample as an energy or weapon unit. At the summary of the present work, the restrictions are discussed for the amount of isomer material that can be really produced.

Decay retardation, being useful for energy accumulation, is accompanied at the same time 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 experiments of Refs. [6, 7], 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 states. In  $\gamma$  cascades, the most probable are the transitions of low multipolarity,  $E1$ ,  $M1$ ,  $E2$ , and they cannot directly supply the required spin deficit if the isomer spin is much higher than the residual angular momentum. Many stretched transition in the cascade are needed and the probability is decreased. There is no possibility to violate the angular momentum conservation because it is an integral of motion and it should be conserved absolutely in both classical and quantum mechanics.

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 cannot 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 of below 3 MeV. So, as usual in nuclear physics, the experimental measurements are needed to get reliable values of the reaction cross-section and yield. The experiments are described below.

One introductory remark concerns also the discussion of the isomer application in a mode of the controlled source of energy and radiation, assumed that the isomer decay can be artificially stimulated (triggered) by the external radiation. Within the «up-conversion» scheme, a photon is absorbed and provides the transition from the isomer to some higher lying level. The latter one should decay fast to the ground state and the isomeric energy is released. But the efficiency of such a process should be again restricted by the wavefunction mismatch between the isomer and other levels. Triggering cross-section can be too small, even if so lucky intermediate level exists. Extensive experimental studies may clarify the triggering efficiency. In the present paper, triggering experiments are out of discussion, and the review of experimental attempts for triggering known up to date is given in Ref. [1].

## 1. $^{178m2}\text{Hf}$ PRODUCTION WITH BREMSSTRAHLUNG

Photon-induced nuclear reactions were systematically studied in [8, 9] at the irradiations of  $^{\text{nat}}\text{Ta}$ ,  $^{\text{nat}}\text{Hf}$  and  $^{178m2}\text{Hf}$  targets with bremsstrahlung at the end-point energy of 23.5 MeV. The activation technique 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 there were 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 easily populated 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 [8, 9]. But later, at the 22-MeV end-point bremsstrahlung irradiation, higher sensitivity of measurements was reached [10], 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 the highest yield of  $(\gamma, n)$  products at such an energy and because of the highest spin value of the  $^{179}\text{Hf}$  nuclei among the other stable Hf isotopes. The isomer-to-ground ratio was found to be

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

Respectively, the absolute yield should not be high, when one uses the 22-MeV bremsstrahlung for the accumulation of the  $^{178m2}\text{Hf}$  isomer. Assume that 10 g of isotopically enriched  $^{179}\text{Hf}$  is irradiated close behind the converter and the electron beam has an intensity of 100  $\mu\text{A}$ . The estimated yield of  $^{178m2}\text{Hf}$  of about  $4 \cdot 10^7$  atoms/s is out of interest for the production of the isomer in an amount needed for the target preparation.

It would be very probable that the yield must be higher with the electron beam of higher energy, and new 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-based collimation system and relatively thin converter and target samples were used for the better definition of experimental conditions. After a 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. The experiment is described in Ref. [11] in some detail.

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 than 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  $^{181}\text{Ta}(\gamma, p2n)^{178m2}\text{Hf}$  reaction can be written formally because at high energy not only nucleons but mesons are generated and emitted. Respectively, a variety of reactions leading to the same product arise. Photon absorption at  $E_\gamma \geq 200$  MeV involves the mechanism of meson generation and corresponding peaks are strongly manifested in the excitation function. Above 1200 MeV, the absorption cross-section reaches almost constant asymptotic value of about 0.12 mb/nucleon.

At low energies,  $E_\gamma \leq 200$  MeV, the quasi-deuteron mechanism provides 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. Thus, 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 ratio (2) that is comparable with the values determined in [12] for the Ta spallation by protons of intermediate energy of (600–300) MeV. At a few hundred MeV range the photon absorption transfers to the nucleus sufficient energy for the emission of many particles, as in the case of 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 be a rather thick sample of Ta.

The problem of optimization of such a unified assembly was solved analytically in [11]. As is known, the radiation energy losses of electrons in ultrarelativistic case are proportional to the electron energy

$$\left(\frac{dE}{dx}\right)_{\text{rad}} = c E_e. \quad (3)$$

The total yield of photonuclear reaction in the sample of thickness  $d$  exposed to the electron beam can be evaluated as following:

$$Y_{nr} \sim \int_0^d N_\gamma(t) \bar{\sigma} dt = \frac{\bar{\sigma} c}{\mu^2} (\mu d + e^{-\mu d} - 1), \quad (4)$$

where  $\bar{\sigma}$  is a mean cross-section of the reaction, and  $\mu$  is a linear attenuation coefficient for the bremsstrahlung photons. In Fig. 1, the absolute production yield of  $^{178m2}\text{Hf}$  is plotted as a function of the thickness of the Ta sample when it is irradiated with a  $100\ \mu\text{A}$  electron beam at 4.5 GeV. 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 20 mm thick, a number of  $^{178m2}\text{Hf}$  atoms reach the value of

$$Y = 3.2 \cdot 10^9 \text{ atoms/s} \cdot 100\ \mu\text{A}. \quad (5)$$

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 a factor should not be significant.

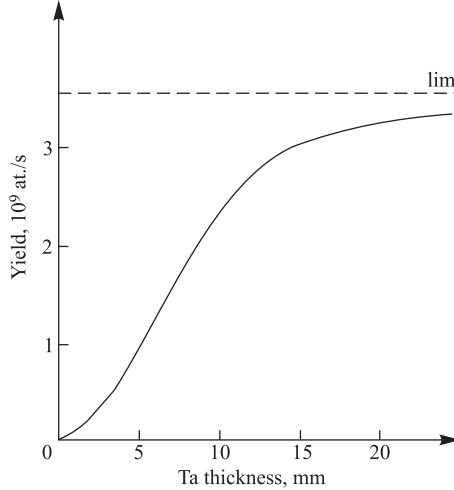


Fig. 1. Yield of the  $^{178m2}\text{Hf}$  as a function of thickness of the Ta sample exposed to  $100\ \mu\text{A}$  electron beam at 4.5 GeV

## 2. NEUTRON-INDUCED REACTIONS

Production cross-section in neutron capture reactions with thermal neutrons are typically low for high-spin isomers with  $I \geq 10$ . The isomer  $^{177m}\text{Lu}(I^\pi = 23/2^-)$  is an exception that confirms the general tendency, because the high spin of the target  $^{176g}\text{Lu}(I^\pi = 7^-)$  nucleus provides a rather moderate spin deficit  $\Delta I = 4\hbar$  in the  $^{176}\text{Lu}(n, \gamma)^{177m}\text{Lu}$  reaction. In contrast with neutron capture, fast neutron reactions provide additional possibilities.

The yield of high-spin  $^{178m2}\text{Hf}$  isomer in reactions with neutrons was tested in experiments [13–15]. Relatively high cross-section was found [14, 15] in the  $^{179}\text{Hf}(n, 2n)^{178m2}\text{Hf}$  reaction induced with 14.5-MeV neutrons. But a productivity is restricted by the neutron flux available when the  $\text{T}(d, n)^4\text{He}$  reaction is used for neutron generation. Much higher fluxes are created in reactors, but the spectrum is soft and needed energies of  $E_n \geq 10$  MeV have very low probability. Unfortunately, slow neutrons are not effective for the  $^{178m2}\text{Hf}$  production because the neutron capture  $^{177}\text{Hf}(n, \gamma)^{178m2}\text{Hf}$  reaction is characterized by very low isomer-to-ground state ratio  $\sim 0.5 \cdot 10^{-9}$ , accordingly [13].

At the same time, the isomer population in the  $^{178}\text{Hf}(n, n'\gamma)^{178m2}\text{Hf}$  reaction has never been experimentally tested. In addition, the low yield observed in  $(n, \gamma)$  reaction [13] can be a result of burnup of the produced isomeric nuclei. Burnup process may be significant at high fluence applied in [13], but its cross-section was not known. Even now, the data on the  $^{178m2}\text{Hf}$  burnup in reactor irradiations are not complete. Only the branch of the  $^{178m2}\text{Hf}(n, \gamma)^{179m2}\text{Hf}$  transmutation was experimentally characterized in Ref. [16], but the total  $^{178m2}\text{Hf}(n, \gamma)$  cross-section was not yet measured. In such a context, a new experiment has enough motivation to be performed testing both burnup and  $(n, n'\gamma)$  processes.

Metal  $^{\text{nat}}\text{Hf}$  foils 1 mm thick were activated in an external channel of the IBR-2 reactor at FLNP (JINR, Dubna) and were then studied using a 20% efficiency Ge gamma detector. This was accomplished by spectrometric electronics, which allowed a count rate up to 20 kCs/s with a reasonable dead time and conservation of spectral resolution. The neutron spectrum at the location of the target was known from previous experiments. But in addition, NiCr-alloy samples were used as spectators for the calibration of the thermal and fast neutron fluence. The Hf samples were irradiated with and without Cd shields and the method of Cd difference allowed isolating the effect of thermal neutrons and deduction of the thermal cross-section.

In measured spectra of activated Hf, the  $\gamma$  lines were observed and quantitatively determined for the following radionuclides:  $^{175}\text{Hf}$ ,  $^{179m2}\text{Hf}$ ,  $^{180m}\text{Hf}$  and  $^{181}\text{Hf}$ . The bulk of the activity was defined by  $^{175}\text{Hf}$  and  $^{181}\text{Hf}$  formed in  $(n, \gamma)$  reactions. They served for the intrinsic calibration of the thermal and resonance neutron fluxes in a presence of the flux attenuation due to the self-absorption in the 1-mm Hf samples.

In this way, the thermal cross-section  $\sigma_{\text{th}}$  and resonance integral  $I_{\gamma}$  values were figured out for the  $^{180m}\text{Hf}$  isomer formation and the results are in accordance with the tabular values [17]. The yield of the high-spin  $^{179m2}\text{Hf}$  isomer was newly obtained and attributed to the  $^{179}\text{Hf}(n, n'\gamma)^{179m2}\text{Hf}$  reaction with neutrons of fission spectrum. The isomer-to-ground state ratio  $\sigma_m/\sigma_g \approx 1.6 \cdot 10^{-3}$  does not contradict the systematics of [18].

The activity of  $^{178m2}\text{Hf}$  was too low, and it could not be distinguished and estimated even past long (1.5 years) «cooling» of the sample after irradiation. Only upper limit was established for the number of produced  $^{178m2}\text{Hf}$  nuclei. Respectively, a limit for the cross-section of the  $^{178}\text{Hf}(n, n'\gamma)^{178m2}\text{Hf}$  could be evaluated immediately. This reaction may be productive only at neutron energies of above 3 MeV because of the isomer excitation energy 2.45 MeV plus 0.5 MeV spent for the ejected neutron and gammas. A number of such neutrons were estimated using the known spectrum of fast neutrons at the location of the irradiated sample and the measured activity of  $^{58}\text{Co}$  in the NiCr spectator. As a result, the cross-section of the  $^{178m2}\text{Hf}$  production at  $(n, n'\gamma)$  reaction has been restricted



by the upper limit of  $\sigma_m \leq 0.04$  mb, and the corresponding  $\sigma_m/\sigma_g$  value has been found to be as low as  $\leq 1.5 \cdot 10^{-5}$ .

More complicated would be an estimation of the definite value of a limit for the neutron capture cross-section leading to the  $^{178m2}\text{Hf}$  isomer. Complications are due to the contribution of both thermal and resonance neutrons and due to the unknown burnup cross-section for the produced isomeric nuclei. The approach of Ref. [13] was obviously too simplified because rather low burnup cross-section  $\leq 20$  b was assumed, and the effect of resonance neutrons was neglected. At the present experiment with total fluence  $\leq 10^{18}$  n/cm<sup>2</sup>, the burnup can be completely neglected (but not at 3–4 orders of magnitude higher fluences as in [13]).

However, the isolation of individual yields induced by thermal and resonance neutrons is still a problem in our case, too. The Cd-difference method is not applicable, when the product yield is not really observed and only the upper limits restrict the numbers of produced nuclei. Thus we propose to operate with the effective cross-section  $\sigma_{\text{eff}}$ . As known, after the irradiation during  $t_{\text{irr}}$  in reactor, the product yield in linear approximation is proportional to

$$Y \sim t_{\text{irr}} \left( \sigma_{\text{th}} \cdot F_{\text{th}} + \frac{I_\gamma}{\ln(E_2/E_1)} F_{\text{res}} \right) = t_{\text{irr}} \cdot F_{\text{th}} \cdot \sigma_{\text{eff}}, \quad (6)$$

where  $F_{\text{th}}$  and  $F_{\text{res}}$  are the fluxes of thermal and resonance neutrons, respectively, and  $\sigma_{\text{eff}}$  is defined as follows:

$$\sigma_{\text{eff}} = \left( \sigma_{\text{th}} + \frac{I_\gamma}{c} \right), \quad (7)$$

$$c = \frac{F_{\text{th}}}{F_{\text{res}}} \ln(E_2/E_1). \quad (8)$$

$c$  is a constant for definite irradiation position at definite reactor.  $E_1$  and  $E_2$  define the «resonance» range of energies, and for heavy nuclei the typical value is known as

$$\ln \frac{E_2}{E_1} \approx (8 - 10). \quad (9)$$

The  $F_{\text{th}}/F_{\text{res}}$  ratio should be below unity, at least, for Dubna IBR-2 reactor, and the recommended value  $c = 5$  can be realistic in our case.

Such an approach expressed in Eqs. (6)–(9) has the advantages that:

- 1)  $\sigma_{\text{eff}}$  value for the isomer can be deduced simply from the measured number of produced nuclei using the measured thermal neutron flux, and
- 2) the isomer-to-ground state ratio can be evaluated exploiting the tabular values of  $\sigma_{\text{th}}$  and  $I_\gamma$  for the production of the ground state nuclei. Equation (7) allows one to calculate  $\sigma_{\text{eff}}$  for the ground state products and to compare it with that determined in experiment for the isomer.

Thus, the results in the present experiment were deduced for slow neutrons. The upper limits for  $(n, \gamma)$  and  $(n, n'\gamma)$  production reactions are compared in Table 1 with cross-sections known from literature.

**Table 1. Cross-sections of the  $^{178m2}\text{Hf}$  production in reactions with neutrons**

Reaction	$^{177}\text{Hf}(n, \gamma)^{178m2}\text{Hf}$		$^{178}\text{Hf}(n, n'\gamma)^{178m2}\text{Hf}$	$^{179}\text{Hf}(n, 2n)^{178m2}\text{Hf}$
Projectile energy	Thermal	Resonance	$E_n \geq 3 \text{ MeV}$	$E_n = 14.5 \text{ MeV}$
$\Delta I(\eta)$	12	12	31/2	11
$\sigma_{\text{eff}}; I_\gamma(\text{mb})$	$1.1 \cdot 10^{-3} *$ $\leq 2 \cdot 10^{-2} **$	– $\leq 0.2 **$	$\leq 4 \cdot 10^{-2} **$	7.3 ***
$\sigma_m/\sigma_g$	$1.5 \cdot 10^{-9} *$ $\leq 1.1 \cdot 10^{-8} **$	– $\leq 2.8 \cdot 10^{-8} **$	$\leq 1.5 \cdot 10^{-5} **$	$3.5 \cdot 10^{-3} ***$
* Recalculated results of [13]. ** Limits established in the present work. *** Ref. [14].				

In order to specify the resonance integral  $I_\gamma$ , we apply another method for the result processing. The additional sample was irradiated at the straight channel of the reactor (behind the neutron mirror) within the container surrounded with the  $\text{CB}_4$  layer of 3 mm thick.

The thermal neutrons were completely screened out, and low-energy resonances at  $E_n \leq 10 \text{ eV}$  were suppressed too. After  $\gamma$ -spectra measurements, the number of  $^{178m2}\text{Hf}$  in the sample was restricted by the upper limit and the flux calibration was done by such products as  $^{95}\text{Zr}$ ,  $^{175}\text{Hf}$ ,  $^{181}\text{Hf}$  and  $^{182}\text{Ta}$ . The resonance integral values for such products were corrected taking into account the neutron spectrum after the  $\text{CB}_4$  filter. However, such a correction is not enough because at the Hf sample of 1 mm thick the self-absorption near the resonance energies may be significant. The self-absorption can be neglected for  $^{95}\text{Zr}$  and  $^{182}\text{Ta}$ , it appears for  $^{175}\text{Hf}$  and is significant for  $^{181}\text{Hf}$ . Remind, that  $^{95}\text{Zr}$  is produced due to the little admixture (3%) of Zr in the Hf material, and  $^{182}\text{Ta}$  is formed at the two-step capture process, the same as in the astrophysical  $s$ -process:  $^{180}\text{Hf}(n, \gamma)^{181}\text{Hf} \beta^- \rightarrow ^{181}\text{Ta}(n, \gamma)^{182}\text{Ta}$ . The  $^{181}\text{Hf}$  yield is measured directly and the result includes the self-absorption factor. In the second reaction, the yield of  $^{182}\text{Ta}$  is not influenced by self-absorption because of very low concentration of the  $^{181}\text{Ta}$  nuclei in the target during the irradiation. Finally, the real flux values are estimated using  $^{95}\text{Zr}$  and  $^{182}\text{Ta}$  activities, both are in agreement, while the calibrations by  $^{175}\text{Hf}$  and  $^{181}\text{Hf}$  show noticeable reduction of the flux. Using such quantitative measurements, we could estimate the resonance flux attenuation for the  $^{177}\text{Hf}(n, \gamma)$  reaction, as well. Finally, the upper limits for  $I_\gamma$  and for the

isomer-to-ground state ratio were deduced. They correspond to the production of  $^{178m2}\text{Hf}$  with resonance neutron capture and are also given in Table 1.

It seems that Ref. [13] provided higher sensitivity of measurements. But, as mentioned above, the burnup effect of the produced  $^{178m2}\text{Hf}$  could be strong at fluences above  $10^{21}$  n/cm<sup>2</sup> and was probably underestimated in [13]. In addition, the resonance flux was neglected, and that could not be correct for the irradiation inside any reactor. Even assuming the best thermalization, one has to use Eq. (7) for  $\sigma_{\text{eff}}$  with the numerical coefficient  $c \approx 20$ . On this basis we recalculated the results [13] in more realistic approximations including the resonance neutron contribution and the burnup effect.

It would be clear that the radiative neutron capture is most destructive process, because the cross-section  $\sigma_{\text{eff}}$  can be as high as thousands barn. Respectively, at the fluence range of  $\Phi \geq 10^{21}$  n/cm<sup>2</sup> the isomer burnup can be manifested. Other nuclear processes in reactor irradiations are characterized by much lower cross-sections, but due to the neutron capture, even the feedstock (target) isotopes are in danger of useless depletion. The transmutation functions for stable Hf isotopes are calculated and shown in Fig. 2.  $\sigma_{\text{th}}$  and  $I_\gamma$  values are taken from [17], and they are combined to get the unified parameter  $\sigma_{\text{eff}}$  using Eq. (7) and numerical coefficient  $c = 20$ . The latter choice should be adequate to the conditions of Ref. [13] experiment. One can see that  $^{177}\text{Hf}$  is most unstable in neutron flux among other Hf nuclei.

In presence of burnup of the target and isomer nuclei, the isomer accumulation function is expressed as follows:

$$N_m(\Phi) = \frac{N_0 \sigma_{mp}}{(\sigma_{tb} - \sigma_{mb})} \{ [\exp - (\sigma_{mb} \Phi)] - \exp - (\sigma_{tb} \Phi) \}, \quad (10)$$

where  $N_m$  and  $N_0$  are the numbers of the isomer and target atoms,  $\Phi$  is fluence,  $\sigma_{mp}$  is the isomer production cross-section,  $\sigma_{mb}$  and  $\sigma_{tb}$  are the burnup

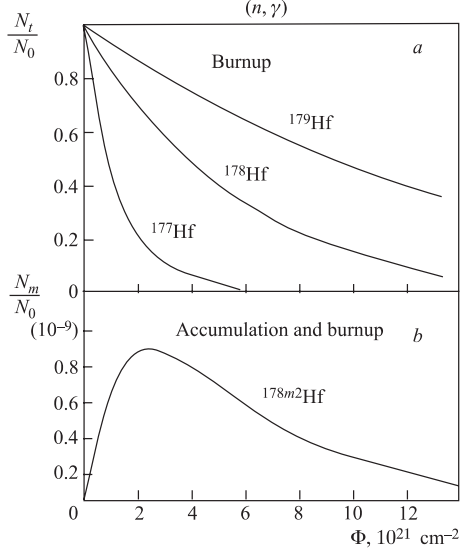


Fig. 2. *a* — calculated transmutation functions for the Hf target stable isotopes in  $(n, \gamma)$  reaction; *b* — the  $^{178m2}\text{Hf}$  accumulation. For the stable isotopes, burnup cross-sections are taken from [17] and for the isomer are estimated using the results of [16]. Condition of irradiations corresponds to that in Ref. [13]

cross-sections of the isomer and target. In (10) we use again  $\sigma_{\text{eff}}$  quantities. The production and destruction  $\sigma_{\text{eff}}$  values are needed for the calculation of the accumulation function for  $^{178m2}\text{Hf}$ . One can estimate the destruction cross-section using the results of experiment [16]. The values of  $\sigma_{\text{th}} = 45$  b and  $I_\gamma = 1070$  b were obtained for the partial branch of the  $^{178m2}\text{Hf}(n, \gamma)$  reaction with the population of the  $^{179m2}\text{Hf}$  isomeric state. Combined together with  $c = 20$  they lead to  $\sigma_{\text{eff}} \approx 100$  b. But total destruction cross-section should also include that for the  $(n, \gamma)$  branch leading to the  $^{179g}\text{Hf}$  ground state. In rough approximation, we took  $\sigma_{\text{eff}} = 200$  b as the total destruction cross-section for  $^{178m2}\text{Hf}$  due to  $(n, \gamma)$  capture.

With such a choice, the processing of experimental results [13] should be revisited and finally, the production cross-section is increased significantly. More intense destruction requires respectively the higher production probability to get the same number of produced atoms (see Eq.(10)). Recalculated cross-section is given in Table 1 and the corresponding  $\sigma_m/\sigma_g$  value, as well. They characterize the  $(n, \gamma)$  reaction observed in [13] and evaluated in the present work in more realistic assumptions. In addition, the accumulation function is shown in Fig. 2 for the  $^{178m2}\text{Hf}$  isomer as is calculated in similar approach with the same numerical parameters. One can see that at fluences above  $10^{21}$  n/cm<sup>2</sup>, the accumulation curve deviates strongly from linear function and then decreases. This is due to both the transmutation of the  $^{177}\text{Hf}$  target nuclei and to the burnup of accumulated  $^{178m2}\text{Hf}$ .

The experiment of present work also does not promise much higher yield of the  $^{178m2}\text{Hf}$  isomer. Finally, the conclusion follows that the reactor irradiation cannot serve as a high efficiency method for the  $^{178m2}\text{Hf}$  production.

### 3. 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 (Refs.[12, 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 us 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.

In total, as many as about 70 radionuclides were identified, and as a result the mass distribution of fragmentation products could be plotted. For example, it is shown in Fig. 3 for the case of  $p+^{186}\text{W}$  reaction at 630 MeV. Master tables of the radionuclide yields and cross-sections are given in Refs. [12, 20] for different targets and energies. Many details of the  $\gamma$ -spectroscopic measurements, the calibration and evaluation procedures, etc. are also given there. Figure 3 charac-

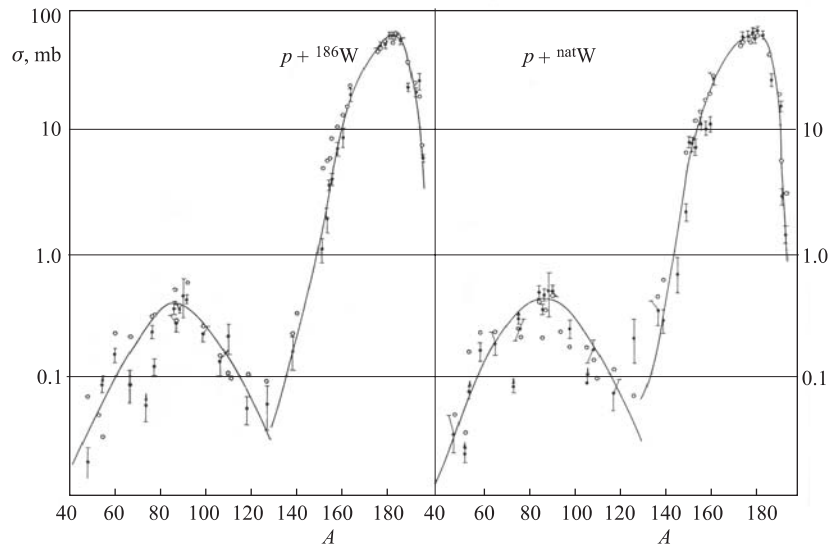


Fig. 3. Mass-distribution of the nuclides produced after the irradiation of  $^{186}\text{W}$  (a) and  $^{\text{nat}}\text{W}$  (b) targets with protons of 630 MeV mean energy. Circles with error bars correspond to experimental results, while without bars — to the calculation using the LAHET code. Lines guide the eyes

terizes 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 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  $^{177m}\text{Lu}$ ,  $^{178m2}\text{Hf}$  and  $^{179m2}\text{Hf}$ . The cross-sections and isomer-to-ground state ratios  $\sigma_m/\sigma_g$  are compared in Table 2 for the production of isomers with 650-MeV proton beam using different targets. The cross-sections of neighboring radionuclides with strong gamma activity are also reported in Table 2. They define the radioactive contaminations of the final isomeric material. It results that background isotopes are produced with very similar cross-sections when different targets are used while the productivity of isomers has large variations. As expected, the best material for isomers production is  $^{186}\text{W}$  target; its use leads to the increase of the cross-section by a factor of about 2.5 for the high-spin isomers of Hf and Lu. This is valid not only at 650-MeV beam energy but at 450 MeV, as well.

**Table 2. Cross-sections and isomer-to-ground state ratios for the formation of high-spin isomers after the spallation of different targets at proton beam energy of 650 MeV. Some products with high gamma activity are also listed for the comparison**

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

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  $10\hbar$  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. For instance, the total cross-section for  $^{178}\text{Hf}$  decreases from Ta to Re while the  $\sigma_m/\sigma_g$  ratio is, on the contrary, increasing. With the  $^{186}\text{W}$  target, the total cross-section for the  $^{179}\text{Hf}$ ,  $^{178}\text{Hf}$  and  $^{177}\text{Lu}$  nuclides formation is higher than with Re while the  $\sigma_m/\sigma_g$  ratio is much better than with the Ta target. Thus, the enriched  $^{186}\text{W}$  is the best target because both  $\sigma_m/\sigma_g$  and total cross-section ( $\sigma_m + \sigma_g$ ) are optimal in this case. The experimental results [12, 20] also show that  $\sigma_m$  and  $\sigma_m/\sigma_g$  values are flatly reduced with the proton energy decrease. Such a weak dependence allows one to use Table 2 results for calculations of the isomeric yield in the case of thick targets when the proton energy is noticeably degraded within the target. The highest productivity irradiation with the intermediate energy protons requires the massive target [19]. However, a few kg amount of the enriched  $^{186}\text{W}$  material would be too expensive. Instead, let us estimate the maximum production yield of  $^{178m2}\text{Hf}$  in the regular Ta target. From Table 2 it follows that the cross-section with Ta should be lower by a factor of 0.65 than with  $^{186}\text{W}$ , but such a reduction is reimbursed due to the gain in the material price.

For estimation, assume now the most powerful proton beam of the Los Alamos accelerator with energy of 800 MeV and with the intensity up to 1 mA. Passing through a 10-cm Ta target, a proton losses the energy from 800 to 560 MeV. The  $^{178m2}\text{Hf}$ -formation cross-section, should be of about 0.3 mb (Table 2). Thus, one can figure out immediately the yield value for  $^{178m2}\text{Hf}$ :

$$Y_{\max} \approx 10^{12} \text{ nuclei/s} \cdot 1 \text{ mA}, \quad (11)$$

i.e. of about 10 milligram of the isomeric material per year of effective irradiations with the high-power beam. Such a brute-force approach is hardly realistic, but the estimated value is near the absolute maximum of production with the known in literature facilities for irradiations. A factor-2 gain can be reached assuming a target 20 cm thick and taking into account the reactions induced by the secondary particles. But this leads to very large amount of Ta for chemical processing and to severe radiation safety conditions under irradiation and processing. At present, the cardinal increase of the production yield much above (7) is probably impossible, unless new facilities supply some fantastically high fluxes of accelerated particles, or (and) kg amounts of the enriched exotic  $^{180m}\text{Ta}$  are available as a target material.

#### 4. REACTION WITH $^4\text{He}$ IONS AT LOW ENERGY

A method of the  $^{178m2}\text{Hf}$  isomer production using the  $^{176}\text{Yb}(^4\text{He}, 2n)$  reaction was proposed and studied (see Refs. [21, 22]). A high-purity isomeric material was accumulated in the extensive irradiations with high-current  $^4\text{He}$ -ion

beam at the 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 of about  $1 \mu\text{g}$   $^{178m2}\text{Hf}$  past high-intensity long irradiations with  $^4\text{He}$  beam. Absolute productivity is not high because the energy losses of  $^4\text{He}$  ions in matter allow using only of about 0.2 g of  $^{176}\text{Yb}$ -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  $^{178m2}\text{Hf}$ , reviewed in [16].

The excitation function of  $^{178m2}\text{Hf}$  in the  $^{176}\text{Yb}(^4\text{He}, 2n)$  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  $^4\text{He}$  ion from 36 to 28 MeV in  $\text{Yb}_2\text{O}_3$  corresponds to the layer of  $70 \text{ mg/cm}^2$  thick.

The targets made of superenriched  $^{176}\text{Yb}_2\text{O}_3$  material were prepared and exposed to the  $^4\text{He}$ -ion beam at long irradiations for the  $^{178m2}\text{Hf}$  accumulation. The U-200 cyclotron at Dubna was modified for operation in the mode of high-intensity  $\alpha$ -particle beam. The extracted beam current of 36 MeV  $^4\text{He}^{++}$  ions reached  $100 \mu\text{A}$ , i.e. a beam power was 1.8 kW. Because of short range of the 36 MeV  $\alpha$ -particles, a volume density of power was high, and the Yb oxide layer could be unstable under the beam.

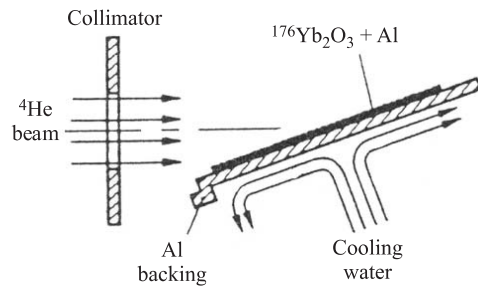


Fig. 4. Schematic draw of the target design for irradiations of the  $^{176}\text{Yb}$  enriched material with a 36 MeV  $^4\text{He}$ -ion beam of high intensity

The target construction was specially designed to prevent losses of the Yb oxide material; a scheme is shown in Fig. 4. The layer of  $\text{Yb}_2\text{O}_3$  was pressed down onto the Al backing; thin Al foil above the layer was also applied to improve a stability of the target. The  $\text{Yb}_2\text{O}_3$  material was distributed over a larger area than the beam cross-section, and the target was inclined to the beam direction. Thus, a heat removal due to heat conductivity was improved. The Al backing was cooled down with the water flow from the rear side of the backing plate. Targets of such a design

could resist the  $^4\text{He}^{++}$  ion current of  $100 \mu\text{A}$ , while at higher intensities some losses of the  $\text{Yb}_2\text{O}_3$  target material occurred.



The produced  $^{178m2}\text{Hf}$  material was chemically isolated past irradiation and then mass-separated, see in more detail [21, 22]. Only one important remark concerns a purity of materials used in the target construction. To prevent some ballast  $^{\text{nat}}\text{Hf}$  contamination at the isolated Hf fraction, it was necessary to take care about chemical purity of the target, backing and chemical compounds used at the extraction of a product material. The commercially supplied 96% enriched  $^{176}\text{Yb}$  was additionally enriched using the electromagnetic mass-separator up to the purity of 99.998%. This expensive operation was needed for the suppression of the yield of the  $^{175}\text{Hf}(T_{1/2} = 70 \text{ d})$  and  $^{172}\text{Hf}(1.87 \text{ y})$  background activities that could disturb some experiments with the produced  $^{178m2}\text{Hf}$  samples.

Under described conditions, the absolute yield of the  $^{178m2}\text{Hf}$  nuclei reached a value of

$$Y = 5 \cdot 10^8 \text{ nuclei/s} \cdot 100 \mu\text{A} \quad (12)$$

that should be compared with the yield of the other reaction.

## 5. OTHER REACTIONS AT LOW ENERGIES

Relatively high cross-section of the  $^{176}\text{Yb}(\alpha, 2n)^{178m2}\text{Hf}$  reaction leads to the idea of possible use of reactions like  $^{181}\text{Ta}(p, \alpha)$ ;  $^{178}\text{Hf}(\alpha, \alpha')$ ;  $^{179}\text{Hf}(\alpha, \alpha'n)$  and  $^{176}\text{Lu}(^7\text{Li}, \alpha n)$  at low energies. Production of  $^{178m2}\text{Hf}$  in these reactions was not studied yet, but it was known from the nuclear-reaction phenomenology that all of them are more or less probable processes at energy well above the interaction barrier. It means that the total cross-section should be about hundreds millibarn, and reasonably high angular momentum of the products provides not very low isomer-to-ground state ratio. Therefore, the  $^{178m2}\text{Hf}$  production cross-section is expected to be comparable with that known for the  $^{176}\text{Yb}(^4\text{He}, 2n)$  reaction, though not much more preferable.

Specially attractive 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, the reactions indicated above 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 strength of the irradiation. For instance, a beam current is chosen to be of  $100\ \mu\text{A}$  for all accelerators, and the same target thickness is assumed unless it is physically restricted due to 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 comparison. The quantities of enriched target isotopes are restricted by the value of 10 g because of high price of such substances.

The ranks in Table 3 (column 10) reflect the absolute yield of the reaction (column 6) at comparable conditions. The  $p + \text{Ta}$  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 above, in Section 3 we discussed much higher yield achievable at this reaction. There is no contradiction, because 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 in the present paper 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, the latter parameter 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 a dependence is plotted in Fig. 5 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. 5.

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. 5 can be used for estimation of the  $\bar{I}_r$  parameter basing on the measured  $\sigma_m/\sigma_g$  ratio. In 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 us discuss a somewhat fantastic idea of using the  $^{180m}\text{Ta}$  material as a high-productivity target. Because of high spin ( $9^-$ ) of 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  $^{\text{nat}}\text{Ta}$  target.

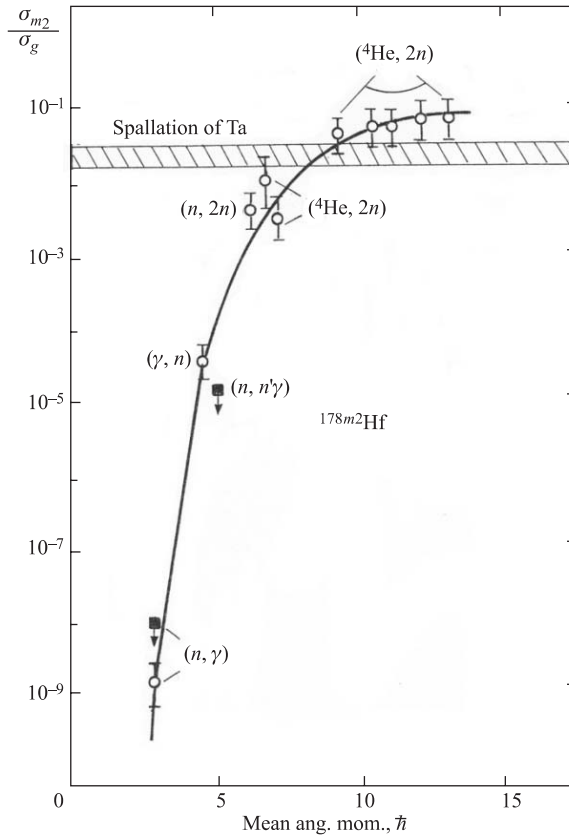


Fig. 5. Systematics of the isomer-to-ground state ratio versus the reaction product spin for the  $^{178m2}\text{Hf}$  isomer as is measured in reactions with different projectiles

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

Projectile	$E_{\text{max}}$ , MeV	Intensity	Target	Amount	Productivity, atoms/s	$\sigma_m$ , mb	$\sigma_m/\sigma_g$	Ref.	Rank
1	2	3	4	5	6	7	8	9	10
Photons, $\gamma$	22 4500	100 $\mu\text{A}^*$ 100 $\mu\text{A}^*$	$^{179}\text{Hf}$ Ta	10g (total) 33 g/cm <sup>2</sup>	4 · 10 <sup>7</sup> 3 · 10 <sup>9</sup>	– –	3 · 10 <sup>-5</sup> 0.03	[10] [11]	6 3
Neutrons, $^1_0n$	thermal 14	5 · 10 <sup>14</sup> /cm <sup>2</sup> ·s 10 <sup>13</sup> /cm <sup>2</sup> ·s	$^{177}\text{Hf}$ $^{179}\text{Hf}$	1g (total) 10g (total)	3.4 · 10 <sup>5</sup> 2.5 · 10 <sup>9</sup>	2 · 10 <sup>-4</sup> 7.3	0.5 · 10 <sup>-9</sup> 3.5 · 10 <sup>-3</sup>	[13] [15]	7 4
Protons, $^1_1\text{H}^+$	650 650	100 $\mu\text{A}$ 100 $\mu\text{A}$	Ta $^{186}\text{W}$	33 g/cm <sup>2</sup> 5 g/cm <sup>2</sup>	2 · 10 <sup>10</sup> 5 · 10 <sup>9</sup>	0.3 0.5	0.02 0.09	[12] [20]	1 2
Alphas, $^4_2\text{He}^{++}$	36	100 $\mu\text{A}$	$^{176}\text{Yb}$	0.07 g/cm <sup>2</sup>	5 · 10 <sup>8</sup>	7	0.05	[21]	5

Remark:  $\sigma_m$  is not given for the bremsstrahlung induced reactions because of the continuous spectrum of photons. The yield ratio was measured and given in column 8.

\* Electron beam intensity.

This follows from the systematics of Fig. 5. 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 clear yet.

Ignoring the cost arguments, one can deduce the absolute maximum of the productivity, as following:

$$Y_{\max} = 10^{13} \text{ atoms/s}, \quad (13)$$

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. Therefore, about 100 mg  $^{178m2}\text{Hf}$  can be accumulated in one-year effective irradiation run.

## SUMMARY

The known experimental results are reviewed for the production cross-sections of the  $^{178m2}\text{Hf}$  exotic isomer. 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 applications discussed in literature 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  $^{180m}\text{Ta}$  and  $^{176}\text{Lu}$  isotopes.

The measured isomer-to-ground state ratios are systematized, as well, because they define the quality characteristics of the accumulated  $^{178m2}\text{Hf}$  material. In addition, such systematics is significant in the nuclear-reaction phenomenology and can be used for the prediction of productivity in the case of unstudied reactions.

**Acknowledgments.** 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. [10–12, 18, 20–22]. These results are used in the present paper.

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