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FORMATION OF THE HIGH-SPIN ^{179m2}Hf ISOMER IN REACTOR IRRADIATIONS

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Production cross-sections in neutron capture reactions with thermal neutrons are typically low for high-spin isomers with $I \geq 10$. The isomer ^{177m}Lu ($I^{\pi} = 23/2^-$) is an exception that confirms the general tendency, because the high spin of the target ^{176g}Lu ($I^{\pi} = 7^-$) nucleus provides a rather modest spin deficit $\Delta I = 4\hbar$ in the ^{176}Lu(n, γ)^{177m}Lu reaction. In contrast with neutron capture, fast neutron reactions supply additional possibilities. The production of high-spin isomers in micro- and milligram amounts would be important for using them in experiments on triggering and controlled release of energy, as is discussed in Ref. [1]. The properties of ^{179m2}Hf isomer ($T_{1/2} = 25.1$ d, $I^{\pi} = 25/2^-$, $E^* = 1.106$ MeV) make it one of the best candidates for triggering experiments, even in comparison with the ^{178m2}Hf isomer widely used during recent years in many studies.

The yields of high-spin ^{178m2}Hf and ^{179m2}Hf isomers were measured in reactions with neutrons [2-4], with bremsstrahlung photons [5,6], with a ^{4}He -ion beam [7,8] and with protons at intermediate energy [9,10]. The spallation reaction with protons shows [9,10] the best absolute productivity, ~$3 \cdot 10^{11}$/s, because a massive target can be irradiated with a high-current proton beam. However, many undesired radioactive nuclides are produced at intermediate energies and radiation-safety restrictions require a long "cooling" time of the target before chemical processing. This delayed processing is possible for the 31-years-lived ^{178m2}Hf, but not for the 25-days-lived ^{179m2}Hf. The spallation reaction is therefore not the best for ^{179m2}Hf production. The ^{176}Yb(α, n) reaction was used for the population of the ^{179m2}Hf state when it was originally discovered in Ref. [11]. The yield of the reaction was not discussed in [11], but later [8] when ^{179m2}Hf was detected from the same projectile-target combination (^{4}He + ^{176}Yb) in the accumulation of the ^{178m2}Hf isomer. It was shown that a rather low production yield, of about 10^7/s, can be reached for ^{179m2}Hf at an optimum energy and with the ^{4}He-ion beam current as high as 100 μA. This means that a method of ^{179m2}Hf production with high yield and efficiency has not been known until now. Obviously, neutron irradiation is potentially the method of highest productivity and the yield of ^{179m2}Hf in reactor irradiations should be examined.

Metal ^{nat}Hf foils of 1-mm thickness 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 that allowed a count rate up to 20 kCs/s with a reasonable dead time and conserving spectral resolution on the level of 1.8 keV for ^{60}Co lines. Standard test sources were used for energy and efficiency calibrations.

The neutron spectrum at the location of 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 fluences in each irradiation by the resulting ^{51}Cr and ^{58}Co activities. The Hf samples were irradiated with and without Cd shields and the method of Cd difference allowed the isolation of the effect of thermal neutrons and the deduction of the thermal cross-section.

In measured spectra of activated Hf, the γ lines were observed and quantitatively determined for the following radionuclides: ^{175}Hf, ^{179m2}Hf, ^{180m}Hf and ^{181}Hf. The bulk of the activity was defined by ^{175}Hf and ^{181}Hf formed in (n, γ) reactions. The contribution due to the activation of admixtures of other elements in the Hf material was negligible. Only Zr is present in a quantity of about 3%, while the concentration of other elements can be estimated on a level $\leq 10^{-5}$ g/g. As was expected, the self-absorption in the 1-mm Hf sample attenuated fluxes of both thermal and resonance neutrons, and the reduction factor was estimated by the detected yield of ^{175}Hf and ^{181}Hf. With such internal calibration of the fluxes, the deduced values of the thermal cross-section and the resonance integral for ^{180m}Hf isomer production in the ^{179}Hf(n, γ) reaction appear to be in good agreement with the tabular data [12]. Low intensity γ-lines of ^{178m2}Hf could not be found in the spectra because of much higher count rate of other nuclides listed above.

The yield of the high-spin ^{179m2}Hf isomer was not high and obviously originated from reactions with fast neutrons. Such a conclusion is definite, because the effect of thermal neutrons was found to be insignificant in that bare and Cd-shielded samples showed the same activation within the standard error. The possible reactions leading to ^{179m2}Hf are listed in Table 1. The upper limit on σ_{th} is the result of the present measurement. Formally, one may assume that the detected yield of ^{179m2}Hf reflects the resonance integral and then the value of 13 mb is deduced. But we suppose that the (n, γ) reaction with slow neutrons makes a completely negligible contribution and thus 13 mb is again the upper limit. Most favorable is the (n, $n'\gamma$) reaction, because the spin deficit ΔI is not so high in this case compared to the other reactions listed in Table 1. The ^{179m2}Hf isomer has an excitation energy of 1.106 MeV, thus one should assume an effective threshold value of about 1.5 MeV for its population in (n, $n'\gamma$) reactions. The number of neutrons with $E_n \geq 1.5$ MeV was determined using the mentioned-above calibration with the NiCr spectator and the known fast-neutron spectrum at the location of the target.

After all, the cross-section and isomer-to-ground state ratio σ_m/σ_g were determined for production of the ^{179m2}Hf isomer in the ^{179}Hf(n, $n'\gamma$) reaction. The latter values were found to be promising for the accumulation of ^{179m2}Hf in reactor irradiations. The relatively good ratio of σ_m/σ_g is understood because the modest spin deficit in the reaction is partially covered by the angular momenta of the bombarding and emitted neutrons. The effect of particle angular momenta should be stronger for the (n, 2n) reaction at 14.8 MeV, and indeed an even higher isomer-to-ground state ratio is deduced for the ^{180}Hf(n, 2n)^{179m2}Hf reaction, accordingly to the experimental results of Ref.[3].

Table 1. Production of the ^{179m2}Hf isomer in reactions with neutrons.

Reaction	^{178}Hf (n, γ) ^{179m2}Hf		^{179}Hf (n, n′γ) ^{179m2}Hf	^{180}Hf (n, 2n) ^{179m2}Hf
Energy	thermal	resonance	$E_n \geq 1.5$ MeV	$E_n = 14.8$ MeV
ΔI [ħ]	12	12	15/2	12
σ; I_γ [mb]	≤ 0.2	≤ 13	4.5 ± 0.5	25$^{*)}$
σ_m/σ_g	$\leq 2.4 \cdot 10^{-6}$	$\leq 7 \cdot 10^{-6}$	$1.6 \cdot 10^{-3}$	$7 \cdot 10^{-3}$

$^{*)}$ Ref. [3]. The value may include some contribution from the (n, n′γ) reaction.

In Ref. [4], it was proposed that the (n, n′γ) reaction could make a comparable contribution at 14.8 MeV compared with the (n, 2n) yield. But in the case of reactor irradiations, the production of ^{179m2}Hf due to the (n, 2n) reaction should be neglected, because the reactor spectrum falls exponentially and the effective threshold of the ^{180}Hf(n, 2n)^{179m2}Hf reaction is as high as about 10 MeV.

In Figure 1, the systematics are shown for the ^{179m2}Hf isomer-to-ground state ratio versus mean angular momentum of the reaction product. The latter parameter is determined from the standard recommendations of nuclear reaction physics. The spin difference, ΔI, of the entrance and exit channels of the reaction is also included. Fig.1 is plotted using the results of Dubna experiments [6-8,10], plus the measurement [3] for the (n, 2n) reaction. One can see a strong growth of the σ_m/σ_g values with the angular momentum and this is explained by the decrease in the ΔI value. The spin deficit ΔI should be covered by the cascade of statistical γ rays during the later stages of the reaction. Naturally, a low ΔI value can be reached easier than high values and the probability is correspondingly changed as shown in Fig.1. The mean angular momentum of the spallation product has not been known, and the measured σ_m/σ_g value is shown as a strip in Fig.1. The strip width reflects the standard errors of the measurements. The intersection of the strip with the regular curve of the systematics may serve to estimate the angular momentum of the spallation product. The discussed regularities are more or less typical for the production of high-spin isomers in nuclear reactions. In particular, similar manifestations were observed and discussed for ^{178m2}Hf in Refs. [7,13].

Using the production cross-section measured for the ^{179}Hf(n, n′γ)^{179m2}Hf reaction (see Table 1), one can estimate the rate of production and the absolute quantity that can be accumulated. Assuming that 1 g of enriched ^{179}Hf is exposed at the reactor core to a flux of about $5 \cdot 10^{14}$ n/cm^2·s during 1 month, more than 10^{16} isomeric ^{179m2}Hf nuclei should be produced. This quantity is definitely enough for the preparation of a ^{179m2}Hf target for use in triggering experiments.

Fortunately, the neutron irradiation of ^{179}Hf in a reactor leads only to the production of ^{179m2}Hf and ^{180m}Hf activities, and the latter one is short-lived, with $T_{1/2} = 5.5$ h. No significant background activities are created if the ^{179}Hf material is chemically purified and highly enriched. The admixtures of ^{174}Hf and ^{180}Hf should be suppressed, otherwise long-lived backgrounds from ^{175}Hf and ^{181}Hf will arise. Anyway, the use of pure isotopic ^{179}Hf target allows one to suppress the backgrounds and to avoid a strong activation.

However, this method of isomer production has some disadvantages. The feedstock ^{179}Hf material cannot be separated from the produced isomeric ^{179m2}Hf either by chemical processing or by isotope separation. For many experiments with isomeric targets, the presence of large amount of stable ground-state nuclei would create backgrounds much more intense than the useful signal. The separation of isomer from ground state is a technically difficult problem, but laser separation methods may be capable of solving it. In the literature [14] experiments are described along this direction and they are promising specifically for Hf isomer isolation from the ground state of the same nuclide.

Finally, let us discuss the problem of the "burn-up" of the produced isomeric nuclei in reactor irradiations. Thermal neutron capture is the most destructive process, because the cross-section σ_{th} can be as high as thousands barns for isomers. Unfortunately, they are not yet measured and this is a challenge for modern neutron experiments. Nevertheless, if the fluence of neutrons in reactor irradiation reaches a value near 10^{21} n/cm^2, then the accumulation process can be disturbed by destruction due to the thermal neutron capture reaction on the produced isomer. The same is also true for resonance neutrons, but their flux is typically lower than that of thermal neutrons. The feedstock isotopes are also in danger of useless depletion under high-fluence neutron irradiations because of their possible transmutation to the neighboring (A+1) isotope of the same element instead of $(n, n'\gamma)$ production of the desired isomer.

The restrictions due to the transmutation and "burn-up" processes are insignificant for the present experiment because the fluence values used are low, being of about 10^{16} n/cm^2. Even a 30-day irradiation at the flux of $5 \cdot 10^{14}$ n/cm^{2}s discussed above should not be extremely dangerous. But strictly speaking, this is dependent on the unknown σ_{th} and resonance integral I_γ values for the ^{179m2}Hf nuclei.

Attempts to produce ^{178m2}Hf in massive neutron irradiations, like the experiment of Ref. [2], should be definitely influenced by "burn-up" of the isomeric nuclei. The destruction of ^{178m2}Hf was tested in experiments [15] when its transmutation to ^{179m2}Hf was observed in the ^{178m2}Hf$(n, \gamma)^{179m2}$Hf reaction. The σ_{th} cross-section and I_γ value had been determined by the method of activation for this partial branch of the neutron capture reaction. And the yield of the stable ground state of ^{179}Hf could not be measured. However, for the "burn-up" the total capture cross-section is important, including both isomeric and ground states population. According to Ref. [16], the isomer and ground state can be populated with comparable cross-section after neutron capture to highly-excited compound states. Consequently, we may assume now that the total σ_{th} and I_γ values are just two times higher that ones measured for the ^{179m2}Hf final state.

In such an assumption, the fluence dependence of ^{178m2}Hf isomer accumulation is calculated and shown in Fig. 2. The transmutation functions are also given for stable feedstock isotopes. The cross-sections of neutron capture and I_γ values are accounted for following Ref. [12]. The ^{178m2}Hf production cross-section was taken from Ref. [2] and the destruction process is estimated as described above based on the results of Ref. [15]. One can see that at fluences above 10^{21}/cm^2, the accumulation curve deviates strongly from a linear function and even decreases. This is due to both the transmutation of the feedstock ^{177}Hf target nuclei and to the burn-up of the accumulated ^{178m2}Hf. The conclusion can be drawn that the destruction role is underestimated in Ref. [2] for ^{178m2}Hf. One note more: the stable ^{179}Hf isotope is long-lasting in the neutron flux, so it can be effectively used for the accumulation of the ^{179m2}Hf isomer within the irradiation time restricted by the isomer lifetime.

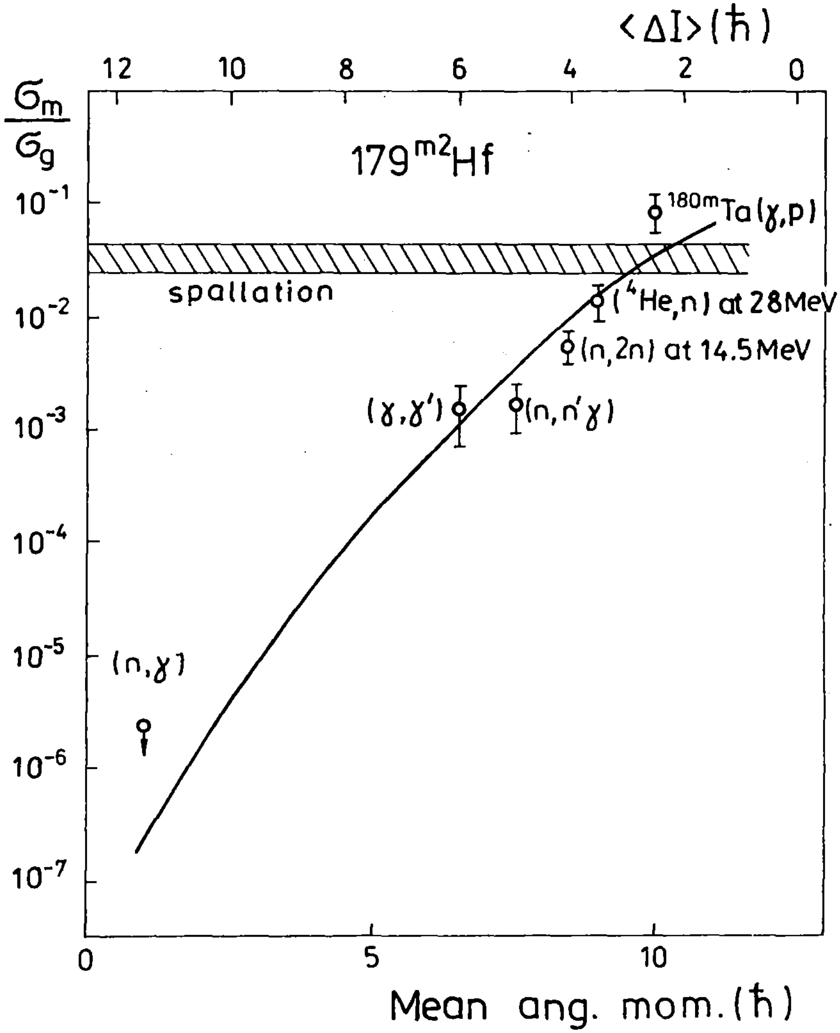

Fig.1. Systematics of the isomer-to-ground state ratios for the production of the ^{179m2}Hf isomer in different reactions. The solid curve is used to guide the eye.

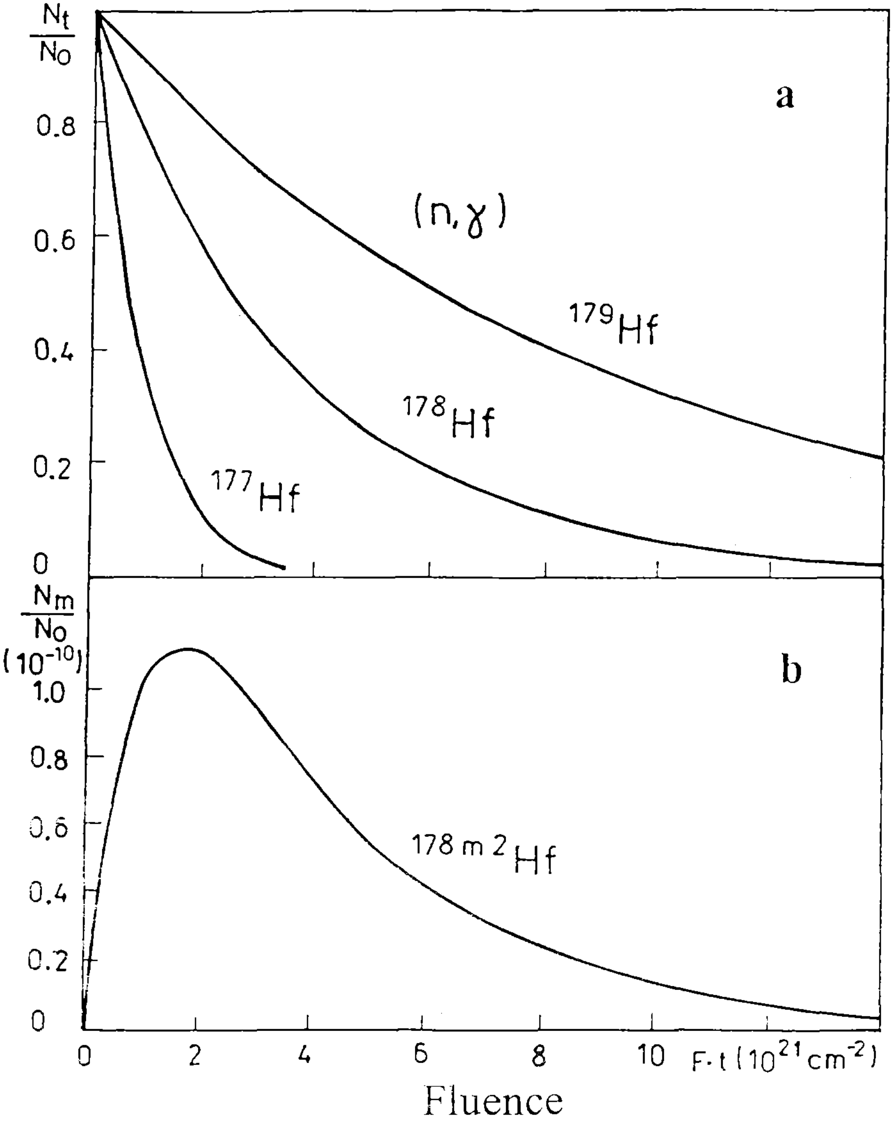

Fig.2. Fluence dependences for the transmutation of the stable Hf isotopes (a) and for the accumulation of the ^{178m2}Hf isomer in (n, γ) reactions (b).

In summary, the ^{179}Hf(n, n$'\gamma$)^{179m2}Hf production reaction is shown to be a method of accumulation of the ^{179m2}Hf high-spin isomer. It can be stored in an amount of 10^{16} atoms for a reasonably low cost, observing the radiation safety conditions in standard reactor irradiations. Other neutron-induced reactions with Hf nuclides are also discussed regarding aspects of production and transmutation of their isomers.

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Карамян С. А. и др. E15-2003-180
Образование высокоспинового изомера $^{179\mathrm{m}2}$Hf
в облучениях на реакторе

Второй изомер в ^{179}Hf ($T_{1/2}=25{,}1$ сут) представляет интерес (среди некоторых других ядерных состояний) с точки зрения возможности стимулированного высвобождения «чистой» ядерной энергии, поскольку запасенная изомером удельная энергия составляет около 0,5 МДж/мг. Выход данного изомера в ядерных реакциях ограничен в связи с его высоким спином ($I=25/2$). Продуктивность известных методов получения была достаточна для исследовательских работ, но не для применений. Показано, что облучения потоком быстрых нейтронов делительного спектра обладают продуктивностью, достаточной для накопления $^{179\mathrm{m}2}$Hf в количестве 10^{16} ядер. В эксперименте, проведенном на реакторе ИБР-2 в Дубне, были измерены выход, сечение σ_m и изомерное отношение σ_m/σ_g для реакции $^{179}\mathrm{Hf}(n,n'\gamma)\,^{179\mathrm{m}2}\mathrm{Hf}$.

Обсуждается также систематика значений σ_m/σ_g, построенная на основе имеющихся экспериментальных данных.

Работа выполнена в Лаборатории ядерных реакций им. Г. Н. Флерова ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна, 2003

Karamian S. A. et al. E15-2003-180
Formation of the High-Spin $^{179\mathrm{m}2}$Hf Isomer in Reactor Irradiations

The second isomer ($T_{1/2}=25.1$ d) in ^{179}Hf is one of a number of nuclear states that is interesting from the point of view of triggering a release of «clean» nuclear energy because it stores a specific energy of about 0.5 MJ/mg. The yield of this isomer in nuclear reactions is restricted due to its high spin, $I=25/2$. The productivity of previously known methods was enough for the creation of experimental amounts but not for potential applications. We show that irradiations in a reactor by the fast neutron flux within the fission spectrum are useful for the accumulation of $^{179\mathrm{m}2}$Hf in an amount of 10^{16} nuclei. In an experiment performed at the Dubna IBR-2 reactor, the yield, cross-section σ_m and isomer-to-ground state ratio σ_m/σ_g were measured for the $^{179}\mathrm{Hf}(n,n'\gamma)\,^{179\mathrm{m}2}\mathrm{Hf}$ reaction. The systematics of the σ_m/σ_g values deduced from the experimental data available for this isomer are discussed.

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

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