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DOUBLE-NEUTRON CAPTURE REACTION AND NATURAL ABUNDANCE OF ^{183}W, ^{195}Pt, AND ^{199}Hg ISOTOPES

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Реакция двойного нейтронного захвата и распространенность изотопов ^{183}W, ^{195}Pt и ^{199}Hg

Данные по нейтронным сечениям многочисленны для стабильных изотопов всей последовательности атомных ядер, но не для радиоактивных нуклидов. Эксперименты по двойному нейтронному захвату могут дать дополнительные сведения. На импульсном реакторе ИБР-2 ОИЯИ для облучения образцов вблизи активной зоны достигается средний интегральный поток около $2{,}3 \cdot 10^{12}$ см$^{-2} \cdot$ с$^{-1}$, достаточный для того, чтобы детектировать продукты двойного нейтронного захвата методом активации. В настоящем эксперименте зафиксированы высокие сечения захвата медленных нейтронов радиоактивными ядрами ^{182}Ta и ^{194}Ir, играющими роль промежуточных ядер-мишеней. Эти значения, вместе с известным для ^{198}Au, доказывают вклад двойного нейтронного захвата в образование изотопов ^{183}W, ^{195}Pt и ^{199}Hg при звездном нуклеосинтезе.

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Double-Neutron Capture Reaction and Natural Abundance of ^{183}W, ^{195}Pt, and ^{199}Hg Isotopes

The database for neutron cross sections is well developed over the nuclide chart for stable isotopes and not as much for the radioactive species. Double-neutron capture experiments could be productive to supply more data. Time-integrated mean flux of slow neutrons reaches a value of $2.3 \cdot 10^{12}$ cm$^{-2} \cdot$ s^{-1} at the irradiation port near the active zone of the IBR-2 pulsed reactor of JINR. This is enough to detect the double-neutron capture products by the activation method. A high capture cross section is deduced in the present experiment for intermediate radioactive ^{182}Ta and ^{194}Ir target nuclides. Together with the known one for ^{198}Au, these values may prove an essential role of double-neutron capture process for nucleosynthesis of ^{183}W, ^{195}Pt, and ^{199}Hg isotopes at stellar conditions.

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

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INTRODUCTION

In double-neutron capture process, the relatively short-lived ^{194}Ir (19.28 h) is present as an intermediate target nucleus. The ^{195}Pt could be found among the final products of activation due to the following process: $^{194g}\mathrm{Ir}\,(n,\gamma)^{195m}\mathrm{Ir}\,\beta^- \rightarrow {}^{195m}\mathrm{Pt}\ (13/2^+)$. The latter nuclide is convenient for detection being 4.01 d-lived and also because of abundant feeding (40%) at the decay of 3.67 h-lived $^{195m}\mathrm{Ir}\ (11/2^-)$ isomer. The ground state of 2.29 h-lived $^{195}\mathrm{Ir}\ (3/2^+)$ is produced with a great yield in the reaction, but decays directly to the stable ^{195}Pt ground state and not to the isomer [1]. Such properties provide an advantageous option for production of ^{195m}Pt through the double-neutron capture reaction. In the case of $^{\mathrm{nat}}$Ta target, the consequent double-neutron capture leads to ^{183}Ta ($T_{1/2} = 5.1$ d) via the intermediate 114.4 d-lived ^{182}Ta isotope. In the present work, the ^{195m}Pt and ^{183}Ta activities were detected by the γ-spectroscopy method past irradiation of ^{193}Ir enriched (98.5%) and $^{\mathrm{nat}}$Ta targets at the IBR-2 reactor. The method of ^{195m}Pt production with double-neutron capture reaction was stressed in literature [2] as useful for the sake of clinical applications.

Commonly, there are recommended neutron cross sections in values reduced in [3–6]. But not enough data are available to describe the formation of ^{195m}Pt and this defines a most significant restriction for the yield calculating. Only two values out of six important are known for thermal cross section plus one value for the resonance integral. It would be impossible to evaluate the yield of ^{195m}Pt from such initial data, even in the case, when the well-developed computer program is available. The results of [2] were obviously obtained using estimated in theory cross sections, though, as known, the cross sections, the resonance integral values, and the isomer-to-ground state ratios could not be predicted in theory. One may hope for a fast progress in the neutron data, but the experiment on observation of ^{195m}Pt in neutron irradiations seems more direct and productive. The flux supplied by the IBR-2 reactor is enough to detect the activation of double-neutron capture products.

Preliminarily, a test experiment has been performed [7] using relatively low neutron flux generated with MT-25 electron accelerator at FLNR, Dubna. A flux

Table 1. Measured cross sections for the slow-neutron capture reactions by the Ir isotopes. Comparison to the data of [3–6] is given in [7]

Target	Product	$T_{1/2}$	$\sigma_{\rm th}$, b	I_γ, b
^{191}Ir $(3/2^+)$	^{192g}Ir (4^+)	73.83 d	550±65	3560±150
	^{192m_1}Ir (1^-)	1.5 min		
	^{192m_2}Ir (11^-)	241 y	—	—
^{193}Ir $(3/2^+)$	^{194g}Ir (1^-)	19.28 h	98 ± 12	1550 ± 90
	^{194m_1}Ir (4^+)	32 ms		
	^{194m_2}Ir (10 or 11)	171 d	$(1.0 \pm 2.8) \cdot 10^{-3}$	0.15 ± 0.05

of about 10^8 cm$^{-2}\cdot$ s^{-1} was not enough to observe the products of double-neutron capture, but major products of the Ir isotopes activation were successfully detected and the corresponding $\sigma_{\rm th}$ and I_γ values were measured. The results are reduced in Table 1. One of the important conclusions follows that the most productive way to reach ^{195m}Pt is going through the consequence of ^{194g}Ir $(n,\,\gamma)$ ^{195m}Ir $\beta^- \rightarrow ^{195m}$Pt. The alternate branch with population of ^{194m}Ir (171 d) at the first step of neutron capture is inefficient because of low cross section measured now for this high-spin product as indicated in Table 1. A great spin-difference $\Delta I \approx 9$ between the initial ^{193}Ir $(3/2^+)$ and final ^{194m}Ir (10 or 11) nuclides suppresses the yield of a product in agreement with the systematic of [8] and with the results taken for isomers of Hf [9]. At the second step of neutron capture, the ^{195m}Ir product possesses a twice lower spin $11/2^-$ and it could be successfully populated in (n,γ) reaction, then it decays in 40% of events to ^{195m}Pt. Unfortunately, the ^{195}Ir ground state decays exclusively to the ground state of ^{195}Pt and there is no branch to the ^{195m}Pt isomer. So, the only efficient chain of processes leading to the ^{195m}Pt isomer is specified.

1. THE ^{195m}Pt YIELD DUE TO DOUBLE-NEUTRON CAPTURE (EXPERIMENT)

The numbers of radioactive atoms accumulated past irradiation time t as a result of single- and double-neutron capture N_1 and N_2, respectively, which could be expressed solving the linear differential equations for accounting of the accumulation and exponential decay low. The following symbols are used below: the decay constants are λ_1 and λ_2 for the products with mass-numbers $(A_t + 1)$ and $(A_t + 2)$, where $\lambda = \ln 2/T_{1/2}$ and A_t indicates the mass-number of the stable target. The yield of products is of course proportional to the number of target atoms N_0 and is defined by the flux F of neutrons per cm$^2\cdot$ s. To be definitive, let us assume that only thermal neutrons are productive and their cross sections are σ_1 and σ_2. At moderate neutron flux, we can neglect the target

material exhausting, as well as a burning-up of the $(A_t + 2)$ product due to the capture of the third neutron.

If necessary (at high fluxes), a factor of burning-up for the $(A_t + 1)$ product could be involved replacing λ_1 with $(\lambda_1 + \sigma_2 F)$. The resonance neutron contribution is described by similar equations with substitution of the resonance integral I_γ instead of σ. However, the resonance neutron flux F_r must be specified, and the Westcott parameter for definite activation product accounted

$$N_1(t) = \frac{N_0 \sigma_1 F}{\lambda_1} \left(1 - \mathrm{e}^{-\lambda_1 t}\right), \tag{1}$$

$$N_2(t) = \frac{N_0 \sigma_1 \sigma_2 F^2}{\lambda_1 \lambda_2} \left[1 - \frac{\lambda_1 \, \mathrm{e}^{-\lambda_2 t}}{(\lambda_1 - \lambda_2)} + \frac{\lambda_2 \, \mathrm{e}^{-\lambda_1 t}}{(\lambda_1 - \lambda_2)}\right]. \tag{2}$$

As mentioned above, the accumulation of ^{195m}Pt (4.01 d) happens through the radioactive β^-decay of 3.67 h-lived ^{195m}Ir. It means that longer-lived product is formed after decay of the short-lived predecessor. The half-life of ^{195m}Ir — 3.67 h, is much shorter than a typical irradiation time comparable to the half-life of a product. It is therefore logical to assume that ^{195m}Ir is transformed to ^{195m}Pt with no time delay and the parameter λ_2 corresponds to the decay of ^{195m}Pt (4.01 d). Definitely, the population efficiency $k = 0.40$ for the final product must also be included into Eq. (2) as a reducing factor. The cross section of ^{194g}Ir activation at the first step is known, but the branch leading to the ^{195m}Ir isomer at the second step remained uncertain until now. In the present experiment, the corresponding cross section and resonance integral were successfully determined using the fluxes at IBR-2 of about $2.3 \cdot 10^{12}$ and $2.0 \cdot 10^{11}$ cm$^{-2} \cdot$ s^{-1} for thermal and resonance neutrons, respectively.

The method of Cd-difference was applied when two enriched ^{193}Ir (98.5%) targets of 20 mg weight each were exposed at the vertical channel of the IBR-2 reactor in FLNP, Dubna. The targets with and without Cd shielding were irradiated during the 17-d reactor run. Metal foils of Ta served as spectators. The Ir samples were dissolved with electrochemical method for consequent isolation of the Pt fraction applying the chromatography. Gamma spectroscopy with HPGe detector was used for the activity measurements. The dissolving yield was calibrated by the ^{192}Ir activity (present due to the ^{191}Ir admixture), while the Pt isolation method was tested elsewhere. Finally, the gamma lines of ^{195m}Pt decay have been measured with a good statistical accuracy, and the producer process, $^{194g}\mathrm{Ir}(n,\gamma)^{195m}\mathrm{Ir} \rightarrow {}^{195m}$Pt, is characterized by the following values: $\sigma_{\mathrm{th}} = 5150$ b and $I_\gamma = 295$ b, including the reduction factor due to the β^- decay branch.

The determined now cross sections are enough to evaluate the activity yield at a high neutron flux of about $2.5 \cdot 10^{15}$ cm$^{-2} \cdot$ s^{-1} like at the Oak Ridge reactor (USA). One must account in calculations that the N_1 intermediate product is

partially exhausted due to the second neutron capture. Then, the equilibrium activity of ^{195m}Pt could reach a level of 1.0 Ci per mg of the ^{193}Ir target material (more detail is given below). Such an activity satisfies the requirements for production of a great specific-activity needed for radiotherapy applications. Proper technical tools and methods must be of course developed for the chemical processing of intense β,γ-ray sources.

2. NUCLEAR AND ASTROPHYSICAL CONSEQUENCES

A high cross section is established for the double-neutron capture process: $^{193}\mathrm{Ir}(n,\gamma)^{194}\mathrm{Ir}(n,\gamma)^{195m}\mathrm{Ir}(3.67\ \mathrm{h}) \rightarrow^{195m}\mathrm{Pt}$. Accounting an efficiency of the β^- decay branch leading to ^{195m}Pt, one immediately deduces the values of $\sigma_{\mathrm{th}} = 12900$ b and $I_\gamma = 740$ b characterizing the constituent $^{194}\mathrm{Ir}(n,\gamma)^{195m}\mathrm{Ir}(11/2^-)$ reaction. The decay branch of low-spin $^{195g}\mathrm{Ir}(3/2^+)$ to $^{195m}\mathrm{Pt}(13/2^+)$ was not observed being negligible [1]. In the reaction, the cross section for this 2.29 h-lived species must exceed by an order of magnitude the observed one for the population of isomeric $^{195m}\mathrm{Ir}(11/2^-)$ state as follows from account of typical isomer-to-ground state ratios dependent on spin for (n,γ) products [9]. Therefore, a total capture cross section for the short-lived ^{194}Ir (19.28 h) target nuclide must be extremely high expressed in a value on the scale of 10^5 b, unlike the cross section assumed in [10]. This is a surprising result in itself.

In addition, a high cross section of 10^5 b in total must produce the great effect of exhausting for the intermediate ^{194}Ir nuclide with the suppression of the final ^{195m}Pt product yield. In particular, for calculation, one has to substitute in Eq. (2) the value of $0.40\,\sigma_2^m$ instead of σ_2 and $(\lambda_1 + \sigma_2^t\,F)$ instead of λ_1, where σ_2^m corresponds to the population branch of ^{195m}Ir past neutron capture and σ_2^t to a total cross section of neutron capture by ^{194}Ir. Within the indicated choice of values, the yield of ^{195m}Pt is calculated as a function of the irradiation time t and the neutron flux F. In Fig. 1, one can see that the saturation with time is reached earlier than in $2T_{1/2}$, and at lower N_2 values. Also, almost linear dependence of the yield versus flux takes place, instead of a flux square F^2 function predicted assuming a negligible burn-up effect.

Experimentally-observed neutron cross section deduced now for radioactive ^{194}Ir target is comparable to the highest thermal cross sections known over the nuclide chart and requires an appropriate interpretation, probably, due to a strong compound resonance exactly near the neutron binding energy in ^{195}Ir. Both m and g species of ^{195}Ir reach the ^{195}Pt ground state after decay, and the known abundance of stable ^{195}Pt isotope comprises the production through the double-neutron capture by ^{193}Ir.

In the same experiment, the spectator $^{\mathrm{nat}}$Ta targets were also irradiated and the second-step $^{182}\mathrm{Ta}(n,\gamma)^{183}\mathrm{Ta}$ reaction demonstrated values of $\sigma_{\mathrm{th}} = 25300$ b and $I_\gamma = 16600$ b exceeding enough the tabular data [3]. Meanwhile, the cross

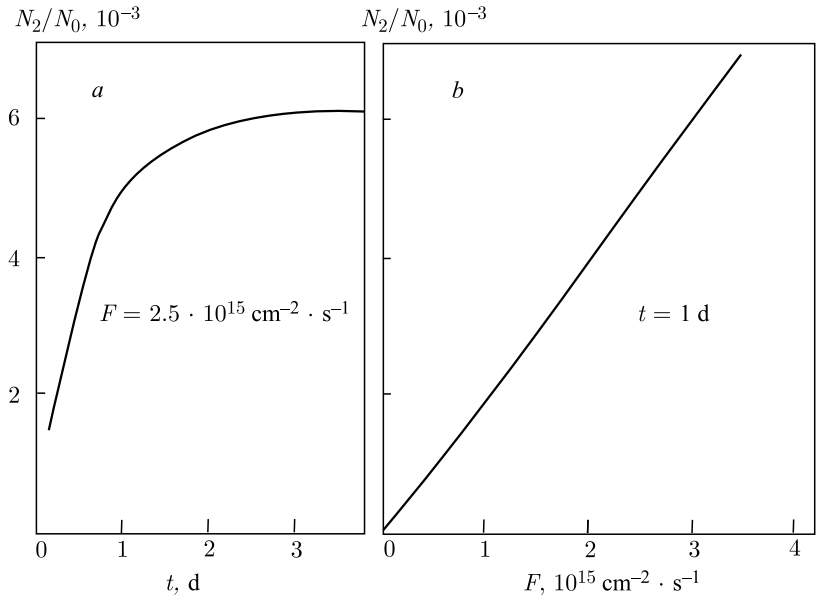

Fig. 1. Calculated number of produced ^{195m}Pt nuclei as a function of time (*a*) and of neutron flux (*b*)

section of 47000 b was reported for ^{182}Ta in old publication [11], unfortunately, not specified to distinguish $\sigma_{\rm th}$ and I_γ. Given here numerical values (except the estimate of 10^5 b for ^{194}Ir) are obtained with a standard inaccuracy of about 10%, including the errors due to the calibration and recalculations. Remind also a high value of $\sigma_{\rm th} = 25100$ b known [3] for the neutron capture by radioactive ^{198}Au with production of ^{199}Au and then ^{199}Hg after β^- decay. Measured cross sections for radioactive odd–odd nuclides, like ^{182}Ta, ^{194}Ir, and ^{198}Au, are reduced in Table 2. The natural abundance of ^{183}W, ^{195}Pt, and ^{199}Hg nuclides being correspondingly: 14.3, 33.8, and 16.9%, may include a contribution from the double-neutron capture at stellar nucleosynthesis. In general, the double-

Table 2. Parameters of the thermal and resonance-neutron capture reactions measured in the present work for radioactive odd–odd isotopes of ^{182}Ta and ^{194g}Ir, and known [3] for ^{198}Au

(n,γ) reaction	Neutron number	E^*, MeV	$\sigma_{\rm th}$, b	I_γ, b
^{182}Ta$\rightarrow^{183}$Ta	109$\rightarrow$110	6.934	25300	16600
^{194g}Ir$\rightarrow^{195m}$Ir	117$\rightarrow$118	7.232	12900	740
			$\approx 10^5$	$\approx 10^4$
^{198}Au$\rightarrow^{199}$Au	119$\rightarrow$120	7.584	25100	—

neutron capture way differs from the standard s- and r-processes. The second neutron capture happens prior to the β^-decay of the first capture product (unlike the s-process), while the capture of the third and further neutrons is improbable (unlike the r-process).

Speaking on the nucleosynthesis, one must realize that the room-temperature conditions differ strongly from the typical ones in the Space. A great variety of conditions exists in the Universe, but neutrons are generated within dense and hot sites. Produced neutrons are moderated to reach a temperature of the comprising matter. The temperature value of $T \geqslant 10^6$ K approximately corresponds to the neutron energy of $E_n > 100$ eV. The room-temperature thermal cross sections determined in laboratory conditions are not applicable to the evaluation of nucleosynthesis at stellar conditions. Even the resonance integral for heavy nuclei is supplied mostly by the neutron energies like $(1-10)$ eV, lower than the thermal energies in the Universe.

Our conclusion about productive role of the double-neutron capture seems doubtful, but this verdict is not absolute. Indeed, the Maxwell distribution for neutron kinetic energies involves an asymptotic $W(E) \sim E^{1/2}$ at $E \to 0$. At the same time, the neutron absorption cross section in geometrical approximation is expressed with the reversed factor of $\sigma \sim E^{-1/2}$. Therefore, the reaction yield, in general case, remains almost constant at low energies. The great thermal

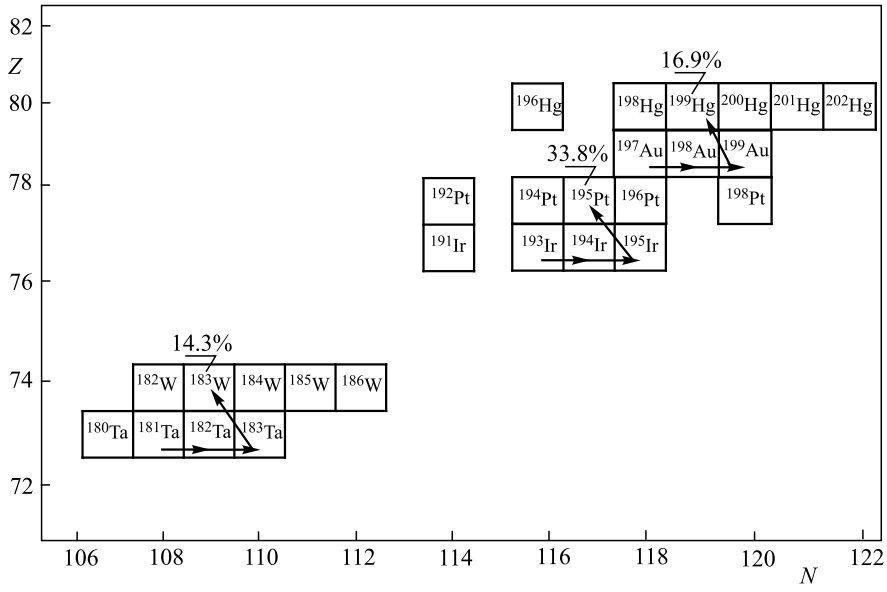

Fig. 2. Processes leading to the synthesis of ^{183}W, ^{195}Pt, and ^{199}Hg isotopes through the double-neutron capture reaction in addition to the standard s-process way

cross section in laboratory conditions could arise only due to the presence of a strong resonance at the neutron energy of about zero. This resonance changes the asymptotic of σ with a strong enhancement of it and provides the significant reaction yield even despite a high temperature of the moderator at stellar conditions. Our measured cross sections for ^{182}Ta and ^{194}Ir together with the known one [3] for ^{198}Au are just the cases of strong enhancement of cross sections by the resonance at $E \to 0$. Possible synthesis of ^{183}W, ^{195}Pt, and ^{199}Hg isotopes is illustrated in Fig. 2.

SUMMARY

Production and chemical isolation of ^{195m}Pt isomeric activity is of interest for radiotherapy of patients. The productivity of double-neutron capture reaction for accumulation of ^{195m}Pt is proved now past the experiment on irradiation of the ^{193}Ir enriched target at the IBR-2 reactor. Great cross section is revealed for the neutron capture reaction by radioactive ^{194g}Ir nuclide. The $^{\rm nat}$Ta targets were also irradiated over the experiment as spectators, and they by chance have demonstrated a great cross section for the second neutron capture by radioactive ^{182}Ta. The cross sections of neutron capture by the odd–odd radioactive targets, like ^{194g}Ir, ^{182}Ta, and ^{198}Au, exceeding 10^4 b, are of importance for understanding within the nuclear reaction theory. On the other hand, the natural isotope abundances are influenced due to the observed great probability of the double-neutron capture process, in particular, for production of ^{183}W, ^{195}Pt, and ^{199}Hg isotopes.

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