Double-neutron capture reaction and natural abundance of $^{183}$W, $^{195}$Pt and $^{199}$Hg isotopes

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Abstract. There are much data on neutron cross sections over the chart of nuclides for stable isotopes and not as much for the radioactive ones. Double neutron capture experiments could be fruitful to provide more data. Time-integrated mean flux of slow neutrons reaches the value of 2.3·10$^{12}$ n/cm$^2$ s 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 obtained in the present experiment for intermediate radioactive $^{182}$Ta and $^{194}$Ir target nuclides. Together with the known data for $^{199}$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.

Keywords: neutron flux, radioactive products, double capture, isotope abundance

1. Introduction

In a 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: $^{194}$Ir (n, $\gamma$)$^{195m}$Ir $\beta^-$ $^{195}$Pt (13/2$^+)$). The latter nuclide is convenient for detection being 4.01 d–lived and the main channel (40%) in the decay of 3.67 h–lived $^{195m}$Ir (11/2$^-$) isomer. The ground state of $^{195}$Ir (3/2$^-$) (2.29 h) is produced with a large yield in the reaction, but it decays directly to the ground state of the stable $^{195}$Pt, 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 $^{182}$Ta target, the consequent two-neutron capture leads to $^{185}$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 $^{185}$Ta activities were detected by the $\gamma$-spectroscopy method after irradiation of $^{194}$Ir (98.5% enriched) and $^{182}$Ta targets at the IBR-2 reactor. The method of $^{195m}$Pt production by double neutron capture reaction was stressed in [2] as useful for clinical applications.

In fact, there are recommended neutron cross section values given in [3 – 6]. However, there are not enough data available to describe the formation of $^{195m}$Pt, which puts the most significant restriction on calculation of the yield. Only two of six important values 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 theoretically estimated cross sections, though it is known that the cross sections, the resonance integral values and isomer-to-ground state ratios cannot not be predicted in theory. One may hope for fast progress in neutron data, but the experiment on observation of $^{195m}$Pt in neutron irradiations seems more direct and fruitful. The flux supplied by the IBR-2 reactor is enough to detect the activation of double neutron capture products.

Initially, a test experiment [7] has been performed using relatively low neutron flux generated by the MT-25 electron accelerator at FLNR, JINR. The flux about 108 n/cm$^2$ s was not enough to observe the products of double neutron capture, but major products of Ir isotopes activation were successfully detected and the corresponding $Q_n$ and $J_f$ values were measured. The results are given in Table 1. One of the important conclusions is that the most efficient way to obtain $^{195m}$Pt is through the process $^{194}$Ir (n, $\gamma$)$^{195m}$Ir $\beta^-$ $^{195}$Pt. The alternative branch with the population of $^{194m}$Ir (171 d) at the first step of neutron capture is inefficient because of the low cross section measured for this high-spin product (see Table 1). A large spin-difference $\Delta I \approx 9$ between the initial $^{193}$Ir (3/2$^+$) and the final $^{194}$Ir (10 or 11) nuclides suppresses the yield of the product in agreement with the systematics [8] and the results obtained for the Hf isomers [9]. At the second step of neutron capture, the...
$^{195}\text{Ir}$ product has a twice-lower spin $11/2^-$ and may be successfully populated in $(n, \gamma)$ reaction, it then decays in 40% events to $^{195}\text{Pt}$. Unfortunately, the ground state of $^{195}\text{Ir}$ decays only to the ground state of $^{195}\text{Pt}$ and there is no branch to the $^{195}\text{mPt}$ isomer. Therefore, the only efficient sequence of processes leading to the $^{195}\text{mPt}$ isomer is obvious.

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

| Target   | Product   | $T_{1/2}$ | $\sigma_{\text{fs, barn}}$ | $I_\gamma \text{, barn}$ |
|----------|-----------|-----------|----------------------------|--------------------------|
| $^{191}\text{Ir}(3/2^-)$ | $^{192}\text{Ir}(4^-)$ | 73.83 d | 550±65 | 3560±150 |
|          | $^{192}\text{mIr}(1^-)$ | 1.5 min | - | - |
|          | $^{193}\text{mIr}(11^-)$ | 241 y | - | - |
| $^{191}\text{Ir}(3/2^-)$ | $^{194}\text{Ir}(1^-)$ | 19.28 h | 98±12 | 1550±90 |
|          | $^{194}\text{mIr}(4^-)$ | 32 ms | - | - |
|          | $^{194}\text{mIr}(10 \text{ or } 11)$ | 171 d (1.0±2.8) $10^{-3}$ | 0.15±0.05 |

2. The $^{195}\text{mPt}$ yield due to double neutron capture (experiment)

The numbers of radioactive atoms accumulated after irradiation time $t$ as a result of single and double neutron capture $N_1$ and $N_2$, respectively may be obtained by the solution of linear differential equations taking into account the accumulation and exponential decay law. The following notations are used below: the decay constants $\lambda_1$ and $\lambda_2$ for the products with mass numbers $(A_t + 1)$ and $(A_t + 2)$, where $\lambda = \ln2/T_{1/2}$, $A_t$ is the mass number of the stable target. Obviously, the yield of products is proportional to the number of target atoms $N_0$ and is determined by the flux $F$ of neutrons per cm$^2$.s. Let us assume that only thermal neutrons are involved and their cross sections are $\sigma_1$ and $\sigma_2$. At moderate neutron flux we may neglect the target material exhausting as well as 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 may be introduced replacing $\lambda_1$ with $(\lambda_1 + \sigma_2 F)$. The resonance neutron contribution is described by similar equations substituting the resonance integral $I_\gamma$ instead of $\sigma$. However, the resonance neutron flux $F_r$ and the Westcott parameter for the activation product must be specified.

As mentioned above, the accumulation of $^{195}\text{mPt}$ (4.01 d) proceeds through the radioactive $\beta^-$ decay of 3.67 h-lived $^{195}\text{mIr}$. This means that the longer-lived product is formed after the decay of the short-lived predecessor. The half-life 3.67 h of $^{195}\text{Ir}$ is much shorter than a typical irradiation time comparable to the half-life of a product. Therefore, it is logical to assume that $^{195}\text{mIr}$ is transformed to $^{195}\text{mPt}$ with no time delay and the parameter $\lambda_2$ corresponds to the decay of $^{195}\text{mPt}$ (4.01 d). Definitely, the population efficiency $k = 0.40$ for the final product must also be introduced into Eq. (2) as a reducing factor. The cross section of $^{195}\text{mIr}$ activation at the first step is known, but the branch leading to the 195mIr isomer at the second step remained uncertain until now. In the present experiment, the corresponding cross section and the resonance integral were successfully determined at IBR-2 using the fluxes about $2.3\cdot10^{12}$ and $2.0\cdot10^{13}$ n/cm$^2$s for thermal and resonance neutrons, respectively.
The method of Cd-difference was applied when two enriched 193Ir(98.5%) targets of 20 mg weight each were exposed at the vertical channel of the IBR-2 reactor, FLNP, JINR. 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 by electrochemical method for consequent isolation of the Pt fraction applying the chromatography. Gamma spectroscopy with HP Ge detector was used for the activity measurements. The dissolving yield was calibrated by the 192Ir activity (present due to the 191Ir admixture), while the Pt isolation method was tested elsewhere. Finally, the gamma lines of $^{195}_{\text{m}}$Pt decay were measured with a good statistical accuracy, and the production process $^{194}\text{Ir} (n, \gamma) \rightarrow ^{195}_{\text{m}}\text{Ir}$ is characterized by the following values: $\sigma_\text{th} = 5150 \text{ b}$ and $I_\gamma = 295 \text{ b}$ including the reduction factor due to the $\beta$– decay branch.

The cross sections determined now are enough to evaluate the activity yield at the high neutron flux about $2.5 \times 10^{15} \text{ n/cm}^2\text{s}$, as at the Oak Ridge reactor. In calculations one must take into account that the $N_1$ intermediate product is partially exhausted due to the second neutron capture. Then, the equilibrium activity of $^{195}_{\text{m}}\text{Pt}$ may reach 1.0 Ci per mg of the 193Ir target material (more details are given below). This value of activity satisfies the requirements for production of great specific-activity solutions necessary for radiotherapy applications. Of course, proper technical tools and methods must be developed for the chemical processing of intense $\beta,\gamma$-ray sources.

3. Nuclear and astrophysical consequences

A high cross section is obtained for the double-neutron capture process $^{193}\text{Ir} (n,\gamma)^{194}\text{Ir} (n,\gamma)^{195}_{\text{m}}\text{Ir} (3.67 \text{ h}) \rightarrow ^{195}_{\text{m}}\text{Pt}$. Taking into account the efficiency of the $\beta$– decay branch leading to $^{195}_{\text{m}}\text{Pt}$, one immediately deduces the values of $\sigma_\text{th} = 12900 \text{ barns}$ and $I_\gamma = 740 \text{ b}$ characterizing the constituent $^{194}\text{Ir}(n, \gamma)^{195}_{\text{m}}\text{Ir}(11/2^-)$ reaction. The decay branch of low-spin $^{195}_{\text{m}}\text{Ir}(3/2^+)$ to $^{195}_{\text{m}}\text{Pt}(13/2^+)$ was not observed being negligible [1]. In reaction the cross section for the 2.29 h-lived products must exceed by an order of magnitude the observed one for the population of isomeric $^{195}_{\text{m}}\text{Ir}(11/2^-)$ state as follows from the typical isomer-to-ground state ratios depending on the spin for $(n, \gamma)$ products [9]. Therefore, a total capture cross section for the short-lived $^{194}\text{Ir}(19.28 \text{ h})$ target nuclide must be extremely high (105 barns) unlike the cross section assumed in [10]. This is a surprising result in itself. In addition, a high cross section of $10^5 \text{ barns}$ must result in the great effect of exhausting for the intermediate $^{194}\text{Ir}$ nuclide with the suppression of the final $^{195}_{\text{m}}\text{Pt}$ product yield. In particular, for calculation one has to substitute in Eq. (2) the value $0.40\sigma_\gamma$ instead of $\sigma_\gamma$ and $(\lambda_1 + \sigma_\gamma F)$ instead of $\lambda$, where $\sigma_\gamma$ corresponds to the population branch of $^{195}_{\text{m}}\text{Ir}$ after neutron capture and $\sigma_\gamma$ to a total cross section of neutron capture by $^{194}\text{Ir}$. Within the indicated choice of values the yield of $^{195}_{\text{m}}\text{Pt}$ is calculated as a function of the irradiation time $t$ and neutron flux $F$. From Figure 1 it can be seen that the saturation with time is reached earlier, than in $2T_{1/2}$, and at lower values of $N_2$. In addition, almost linear dependence of the yield on the flux is observed, instead of the flux square $F^2$ function predicted assuming a negligible burn-up effect.

![Figure 1](image-url). Calculated number of produced $^{195}_{\text{m}}\text{Pt}$ nuclei as a function of time (a) and neutron flux (b).
Experimentally observed neutron cross sections deduced here for radioactive $^{194}$Ir target is comparable to the highest thermal cross sections known over the nuclide chart and it requires an appropriate interpretation, probably, due to a strong compound resonance exactly near the neutron binding energy in $^{195}$Ir. Both $m$ and $g$ products 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 $^{195}$Ir.

In the same experiment the spectator $^{194}$Ta targets were also irradiated and the second-step $^{192}$Ta($n, \gamma$)$^{193}$Ta reaction demonstrated values of $\sigma_n = 25300 \, \text{b}$ and $I_\gamma = 16600 \, \text{b}$ substantially exceeding the tabular data [3]. Meanwhile, the cross section 47000 $\text{b}$ was reported for $^{182}$Ta in the publication [11] not specifying $\sigma_n$ and $I_\gamma$. The numerical values given here (except the estimate $10^5 \, \text{b}$ for $^{194}$Ir) were obtained with the standard deviation about 10% including the errors due to the calibration and recalculations. A high value of $\sigma_n = 25100 \, \text{b}$ was obtained in [3] for the neutron capture by radioactive $^{198}$Au with production of $^{199}$Au and then $^{199}$Hg after $\beta^-$ decay. Measured cross sections for radioactive odd-odd nuclides, such as $^{182}$Ta, $^{194}$Ir, and $^{198}$Au are given in Table 2. The natural abundances of $^{183}$W, $^{195}$Pt, and $^{199}$Hg nuclides are correspondingly 14.3, 33.8, 16.9%, and may include a contribution from the double neutron capture at stellar nucleosynthesis. In general, the double neutron capture way differs from the standard $s$- and $r$-processes. The second neutron capture occurs prior the $\beta^-$ 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).

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

| ($n, \gamma$) reaction | Neutron number | $E^*, \text{MeV}$ | $\sigma_{th}, \text{b}$ | $I_\gamma, \text{b}$ |
|-------------------------|----------------|------------------|----------------------|---------------------|
| $^{182}$Ta$\rightarrow^{183}$Ta | 109$\rightarrow$110 | 6.934 | 25300 | 16600 |
| $^{194}$Ir$\rightarrow^{195}$Ir | 117$\rightarrow$118 | 7.232 | $m$ | 12900 |
| | | | total $\approx 10^5$ | $\approx 10^4$ |
| $^{198}$Au$\rightarrow^{199}$Au | 119$\rightarrow$120 | 7.584 | 25100 | |

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

Our conclusion about the productive role of the double neutron capture seems doubtful, but this verdict is not absolute. Indeed, the Maxwell distribution for neutron kinetic energies involves the 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 factor $\sigma \sim E^{-1/2}$. Therefore, the reaction yield in the general case remains almost constant at low energies. The high thermal cross section at laboratory conditions may arise only due to the presence of a strong resonance at the neutron energy about zero. This resonance changes the asymptotics of $\sigma$ providing a strong enhancement and the significant reaction yield even despite a high temperature of the moderator in stellar conditions. Our measured cross sections for $^{182}$Ta and $^{194}$Ir together with the known data [3] for $^{198}$Au are just the cases of strong enhancement of cross sections by the resonance at $E \to 0$. The possibility of synthesis of $^{183}$W, $^{195}$Pt, and $^{199}$Hg isotopes is illustrated in Figure 2.

4. Summary

Production and chemical isolation of $^{195m}$Pt isomeric activity is of interest for radiotherapy of patients. The efficiency of double neutron capture reaction for accumulation of $^{195m}$Pt is now proved by the experiment on irradiation of the $^{193}$Ir enriched target at the IBR-2 reactor. A high cross section is obtained for the neutron capture reaction by radioactive $^{194}$Ir nuclide. The $^{194}$Ir targets were also irradiated over the experiment as spectators and they demonstrated a high cross section for the second neutron capture by radioactive $^{198}$Au. The cross sections of neutron capture by the odd-odd radioactive targets, such as $^{194}$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 high probability of the double-neutron capture process, in particular, for the production of $^{183}$W, $^{195}$Pt, and $^{199}$Hg isotopes.

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