MEASUREMENTS OF THE ^{195m}Pt ISOMER YIELD IN IRRADIATIONS OF ^{193}Ir TARGETS WITH NEUTRON FLUX

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Abstract

The double neutron capture experiments are productive to deduce the cross sections for radioactive target isotopes, as well as for some applications. In particular, the ^{195m}Pt isomer activity achievable with double-neutron capture is known to be the best for radio-therapeutic use in a view of the convenient halflife, 4.01 *d*, and a great yield of soft gamma, conversion, and Auger-electron radiation at decay. The possibility of production due to the double-neutron capture was indicated in literature, but the details were yet unknown. We suppose the path from initial ^{193}Ir target to ^{195m}Pt through the following intermediate nuclides: ^{193}Ir $(n, \gamma)^{194}$Ir$(n, \gamma)^{195m}$Ir$\beta^- \rightarrow ^{195m}$Pt. The latter isomer yield is observed and quantitatively determined in the experiment on activation of the enriched ^{193}Ir target at IBR-2 reactor. High cross section is evident for the indicated above process.

1. Introduction

Studies of double neutron capture reaction look potentially productive for different issues including an extension of the neutron cross-section database for radioactive targets, a problem of nucleosynthesis of some isotopes at stellar conditions, and application of radio-nuclides and isomers for clinical therapy. The advantages for the medical use involve a possibility to produce the isomers emitting a soft radiation and providing no radioactive pollution past the decay. There is also an attraction to develop new options for production within a "generator" method which allows the chemical isolation of the therapeutic activity from the ballast target material. The most successful method of radiotherapy on today exploits the ^{99m}Tc isomer produced via ^{99}Mo in the generator scheme.

The ^{195m}Pt (4.01 *d*) isomer is another example of the species promising for the therapeutic uses. It is characterized by convenient properties, same as ^{99m}Tc, or even better because the ^{195m}Pt decay results in multiple generation of Auger electrons which are efficient for processing of the damaged area without the significant radiation dose onto healthy tissues. There are also known bio-molecules contained platinum. At the same time, the production methods are not as attractive. This nuclide is straightly produced in irradiations of stable isotopes of ^{194}Pt with thermal neutrons and ^{195}Pt, ^{196}Pt – with fast neutrons or bremsstrahlung. The moderate productivity could be reached, but the isomer of interest is accumulated in a bulk of the stable Pt material and it cannot be isolated by the chemical means.

Considering the above, the proposal [1] of an indirect method for accumulating of ^{195m}Pt as a result of double neutron capture seems of interest. The final product can be separated from the target substance (Ir) by chemical means. Such a way appears promising, and the proposed scheme should therefore be tested in a quantitative manner. The authors of [1] did not clarify the problem completely. The chain of processes leading to ^{195m}Pt is shown in Fig. 1. The radioactive decay properties are taken from Nuclear Data Tables including the branch of ^{195m}Pt feeding in β^- decay. The ^{195m}Pt $(13/2^+)$ isomer is populated with a probability of 40%

due to the decay of ^{195m}Ir (11/2$^-$) isomer [2], while the ground state of ^{195g}Ir (3/2$^+$) decays exclusively to the ground state ^{195g}Pt (1/2$^-$) because of spin selection regularities in the radioactive decays.

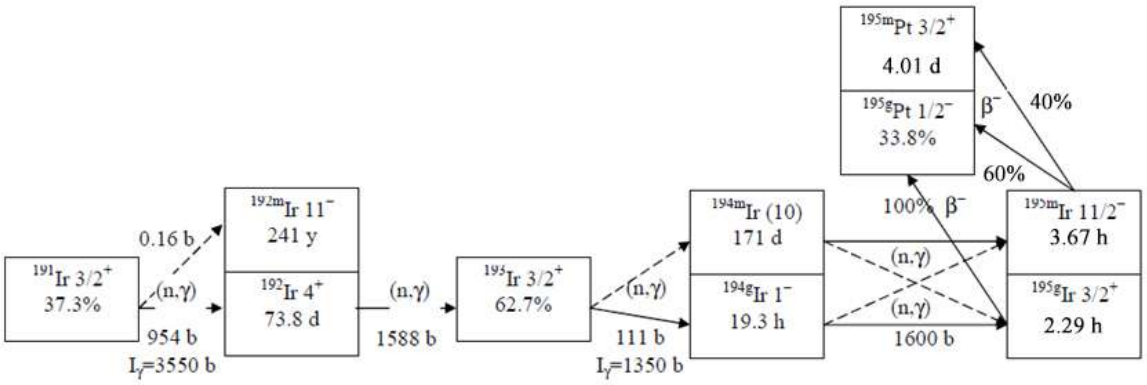

Fig.1. Producer scheme for the ^{195m}Pt isomer via double neutron capture. Cross sections are given according to [3].

The neutron cross sections are commonly used according the data compiled in [3-6]. But there are not enough data to describe the formation of ^{195m}Pt and this defines the most significant uncertainty of calculations on the ^{195m}Pt yield. Only two values of 6 important are known for thermal cross sections plus one value for the resonance integral. It would be impossible to evaluate the yield of ^{195m}Pt from such initial data, even at the case when the well-developed mathematical program is available. The results of [1] were obviously obtained using the theoretical cross sections though cross sections, resonance integral values, and isomer-to-ground state ratios could not be predicted by theory. One may hope for a fast progress of the neutron data, but the experiment on observation of ^{195m}Pt in neutron irradiations seems more direct and productive. The isomer must be activated under irradiating of ^{193}Ir with moderately high integral flux of slow neutrons as at the IBR-2 reactor of JINR, Dubna.

Preliminarily, one could define the most productive way to reach it among the branches shown in Fig. 1. It is clear that an abundant yield is expected for the ^{194m}Ir(n, γ)^{195m}Ir reaction since the irradiated nucleus possesses a relatively high spin: 10 or 11, exceeding the product spin. However, the accumulation of ^{194m}Ir at the first step of neutron capture is strongly suppressed due to a great spin difference. It would be difficult to expect that ^{193}Ir (3/2$^+$) is transformed after neutron capture to ^{194m}Ir (10 or 11). The cross section must be small because of $\Delta I \approx 9$. The ^{194g}Ir(n, γ)^{195m}Ir$\rightarrow$^{195m}Pt process is likely to be more efficient since ΔI (for the transition from 1$^-$ to 11/2$^-$) is twice lower. The latter reaction yield is under the scope in the present experiment.

2. Test activation of iridium at low neutron flux

For the first stage of the present experiments iridium foils of natural isotopic composition were irradiated with a modest neutron flux at the neutron irradiation port of the MT-25 accelerator. The MT-25 microtron of FLNR, JINR accelerates electrons to energies of 23 MeV with a beam current of up to 20 μA and a power of 0.46 kW. Observing the safety

regulations, the beam current was reduced to 10 μ*A* and it was converted to the bremsstrahlung flux used then for generation of fast neutrons and consequent moderation of them. The neutron irradiation port was available at the Dubna microtron. The products of neutron double capture could not be detected because low flux of about $3 \cdot 10^8$ *n/cm*^{2}*s* was not enough to activate the products of interest ^{195g,m}Ir and ^{195m}Pt. Other products of activation were successfully detected, and the cross sections were used for simulation of the higher-sensitivity experiment at IBR-2 reactor. According to the test experiment at MT-25, the epithermal neutron flux in overall was a 35 times lower than the thermal neutron flux. The hard bremsstrahlung radiation was suppressed by 4 orders of magnitude compared to the position near the converter.

Two samples of natural Ir foils with weights of 0.25 *g* each were subjected to the irradiation by neutrons for activation under the above conditions over 14 hours. One sample was wrapped with the Cd metal foil of 0.55 *mm* thickness, while the other was not. Past the irradiation, the accumulated activities were studied with a standard HP Ge spectrometer for the periods of up to one year to detect of the short- and long-lived products. The decay γ lines emitted by products of Ir activation are listed in Table 1. It was easy to determine the number of atoms of such nuclides as ground states of ^{192g}Ir ($T_{1/2}$ = 73.8 days) and ^{194g}Ir ($T_{1/2}$ = 19.3 *h*). Due to their great activity, more long-lived isomeric states of ^{192m}Ir (241 *y*) and of ^{194m}Ir (171 *d*) were hardly observable. Only, the latter isomer yield could be estimated at a low level with the resonance neutrons. As is expected, the products of double neutron capture are invisible at the neutron flux generated by MT-25. The measured yield of ^{190}Ir serves to estimate the degree to which the hard bremsstrahlung flux was suppressed at the point of the sample activation.

Table 1. Gamma radiation emitted by the products of Ir neutron activation*)

Nuclide	**Spin, parity**	$T_{1/2}$	**Main E_γ, keV**	**Yield of 100 decays**	**Formed in the reaction:**
^{190}Ir	4^-	11.78 *d*	407.2	30.8	(γ, *n*)
			518.6	34.0	
			605.4	39.9	
^{192}Ir	4^+	73.83 *d*	296.0	28.7	(*n*, γ)
			308.5	29.7	
			316.5	82.7	
^{192m}Ir	1^-	241 *y*	168.1	0.092	(*n*, γ)
^{194}Ir	1^-	19.28 *h*	293.5	2.52	(*n*, γ)
			328.4	13.1	
			645.2	1.18	
^{194m}Ir	10 or 11	171 *d*	482.6	97	(*n*, γ)
			600.5	62	
			687.8	59	
^{195}Ir	$3/2^+$	2.29 *h*	98.9	9.73	(*n*, γ)+ (*n*, γ)
			129.7	1.25	
			211.3	2.40	
^{195m}Ir	$11/2^-$	3.67 *h*	364.9	9.31	(*n*, γ)+ (*n*, γ)
			432.9	9.40	
			684.9	9.40	
^{195m}Pt	$13/2^+$	4.01 *d*	98.9	11.4	β$^-$decay of ^{95m}Ir
			129.8	2.83	

*) Numerical values are given in accordance with Nuclear Data Sheets.

The standard method of the Cd-difference was applied to distinguish the contributions from thermal and epithermal neutrons in the activation of Ir. "Spectator" targets were used for numerical calibration. This function was performed by a pair of the Ta foils similar in weight and thickness, one of which was clad in Cd shielding to determine the effects of thermal and epithermal neutrons separately. The tabular value of the thermal cross section for neutron capture on ^{181}Ta is referred to the most probable energy of 0.0253 *eV* (at room temperature). Calibration by ^{181}Ta therefore yields the cross sections for Ir activation at the same energy of neutrons.

The results of test activation of Ir at low neutron flux are given and compared to the data of [3-6] in Table 2.There could be slight deviations due to the averaging over the Maxwell spectrum at different temperatures for individual experiments. However, random errors overwhelm the deviations due to the different room temperatures. We were also able to find the resonance integrals for radiative neutron capture by ^{191}Ir and ^{193}Ir targets. A comparison is also given in Table 2. Short-lived first isomeric levels *m*1 decay quickly to the ground states and their yields could not be determined separately from the ground *g* states in ^{192}Ir and ^{194}Ir. Table 2 contains the same total (*m*1 + *g*) values as in [3–6].

Table 2. Cross section of slow neutron capture by Ir and Ta isotopes

Target	**Product**	**σ_{th}, barn**	**I_γ, barn**	**Reference**
^{181}Ta*)	^{182}Ta	20.5±0,5	655±20	[3]
^{180}Ta	^{181}Ta	563±60	1346±100	[3]
^{191}Ir	^{192g}Ir+^{m1}Ir	954±10	3550±300	[3]
		955	3324	[4]
		920	–	[5]
		309	–	[6]
		550 ± 60	3560 ± 150	Present
	^{192m2}Ir	0,16	–	[3]
^{193}Ir	^{194g}Ir +^{m1}Ir	111±5	1350±100	[3]
		112	1376	[4]
		111	–	[5]
		111	–	[6]
		96 ± 12	1553 ± 90	present
	^{194m2}Ir	0,04	–	[5]
		$(1.0 \pm 2.8)10^{-3}$	0.15±0.05	present
^{194g}Ir	^{195g}Ir	1600	–	[5]

*) The Ta sample was used as a "spectator" for calibrating of the σ_{th} and I_γ values

There was a problem to detect relatively low yield of ^{194m2}Ir isomer suppressed due to the high spin (10, or 11) of this product in presence of a great γ background from ^{192}Ir main activity. The measurements were continued during one year mainly searching for ^{194m2}Ir (171 *d*). It is well known that the isomer-to-ground state ratio falls sharply as the spin of reaction products rises [7, 8]. Table 2 gives the value measured for the resonance integral of ^{194m2}Ir. The thermal cross section is restricted by an upper limit that 1/10 the one used in [5]. This likely means that the ^{194m2}Ir isomer is mostly formed in reactions with resonance neutrons. The cross sections listed in Table 2 demonstrate moderate scattering of values even for well known products of activation, for instance for the ^{192}Ir nuclide.

From our test experiment described in more details in [11] one more general conclusion follows that the accepted literature sources [3–6] in some cases do not supply a high accuracy for the neutron cross-section data. Deviations are great enough, and not only from the present results, but also between each to other. Indeed, even for the main activation product of ^{192g}Ir, the scattering of results is evident. Our thermal cross section for ^{192g}Ir equals 550 *b*, and it has appeared just in mid-point between the cited results of the American [3] and Japan [6] groups: 954 and 309 *b*, correspondingly.

It was proposed in [9] that the ^{199}Hg(γ, α) reaction could be used to produce ^{195m}Pt with the 30 *MeV* electron accelerator. It would be possible to separate Pt by means of radiochemistry in this case as well. However, the (γ, α) reaction with a heavy target does not ensure [10] high yield since the emission of alpha particles is two orders of magnitude lower than the (γ, *p*) yield and four to five orders of magnitude lower than the yield of (γ, *n*) reactions. The path through the ^{196}Hg(γ,*p*)^{195}Au reaction is also closed because electron-capture decay of the ^{195}Au nuclide does not populate the ^{195m}Pt isomer.

3. 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, could be expressed solving the linear differential equations for account of the 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 $cm^2 s$. 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 third neutron.

$$N_1(t) = \frac{N_0 \sigma_1 F}{\lambda_1}(1 - e^{-\lambda_1 t}) \ ; \tag{1}$$

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

If necessary (at high fluxes), the burning-up of the (A_t + 1) product could be involved replacing λ_1 with ($\lambda_1 + \sigma_2 \cdot F$). 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 Westscott parameter for definite activation product accounted.

As mentioned above, the accumulation of ^{195m}Pt(4.01 *d*) happens through the radioactive β^- decay of 3.67 *h*-lived ^{195m}Ir. Means, longer-lived product is formed after decay of the short-lived predecessor. The half life of ^{195m}Ir – 3.67 *h* is much shorter compared to the accumulation time for the product of interest. 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. Definitely, the population efficiency *k* = 0.40 for the final product must also be included into Eq. (2) as a reducing factor.

The double neutron capture is going mainly through the population of ^{194g}Ir at first capture step, and the path through the ^{194m}Ir isomer is much less likely because of its high spin

(10 or 11). The cross section of ^{194g}Ir activation at the first step is known, but the branch leading to the ^{195m}Irisomer at the second step was 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 $2.3\cdot10^{12}$ and $2\cdot10^{11}$ n/cm^2s for thermal and resonance neutrons, respectively.

The method of Cd-difference was again 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. The 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 HP Ge detector is 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, ^{194}Ir(n, γ)^{195m}Ir$\rightarrow^{195m}$Pt, is characterized by the following values: σ_{th} = 5150 *b* and I_γ =295 *b*.

The determined now cross sections are enough to evaluate the activity yield at a high neutron flux of about $2.5\cdot10^{15}$ n/cm^2s like at the Oak Ridge reactor. One must account that the N_1 intermediate product (^{194}Ir) is strongly exhausted due to the second neutron capture with a great cross section of about 10^5 barn in total. For calculating, one has to substitute in Eq. (2) the value of $0.4\sigma_2{}^m$ instead of σ_2 and $(\lambda_1 + \sigma_2{}^t F)$ instead of λ_1 where $\sigma_2{}^m$ corresponds to the population branch of ^{195m}Pt isomer past neutron capture and $\sigma_2{}^t$ to the total cross section of neutron capture by ^{194}Ir. With indicated numerical values, one can find that the saturation of looked for activity 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 function. Both deviations from Eq. (2) are due to the intense burn-up of ^{194}Ir. Then, the equilibrium activity of ^{195m}Pt past (2 -3) days irradiation at Oak Ridge reactor may reach a level of 1.0 *Ci* per *mg* of the ^{193}Ir target material. Such an activity satisfies the requirements for production of a great specific-activity solutions needed for radiotherapy applications. Proper technical tools and methods must be of course developed for the chemical processing of intense β,γ-ray sources.

4. Nuclear and astrophysical consequences

A high cross section is established for the double-neutron capture process: ^{193}Ir(n,γ)^{194}Ir(n, γ)^{195m}Ir(3.67 h)$\rightarrow^{195m}$Pt. Accounting an efficiency of the β^- decay branch leading to ^{195m}Pt, one immediately deduces the values of σ_{th} = 12900 barn and I_γ = 740 *b* characterizing the constituent ^{194}Ir(n, γ)^{195m}Ir($11/2^-$) reaction. The decay of low-spin ^{195g}Ir($3/2^+$) contributes nothing to the ^{195m}Pt($13/2^+$) yield despite is produced with a great cross-section. The branch to this 2.29 h-lived species must exceed by orders of magnitude the observed now population of the isomeric ^{195m}Ir($11/2^-$) state due to the known spin restrictions for the yields of (n, γ) products. Thus, a total capture cross section for the relatively short-lived ^{194}Ir (19.28 *h*) appears to be extremely high and is expressed in a magnitude on the scale of 10^5 barn unlike to the magnitude assumed in [12]. A new value for ^{194}Ir is comparable to the greatest thermal cross sections known over the nuclide chart and it requires an appropriate interpretation, probably, due to the presence of 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 must include the production through the double-neutron capture by ^{193}Ir.

Table 3. 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*		σ_{th}, *b*	I_γ, *b*
^{182}Ta$\rightarrow^{183}$Ta	109→110	6.934		25300	16600
^{194}Ir$\rightarrow^{195m}$Ir	117→118	7.232	*m*	12900	740
			total	$\approx 10^5$ *b*	$\approx 10^4$ *b*
^{198}Au$\rightarrow^{199}$Au	119→120	7.584		25100	–

At the same experiment, the spectator ^{nat}Ta targets were also irradiated and the second-step ^{182}Ta$(n, \gamma)^{183}$Ta reaction demonstrated values of σ_{th}= 25300 *b* and I_γ= 16600 *b* exceeding enough the tabular data [3]. Meanwhile, in an old publication [12] the cross section of 47000 *b* was reported for ^{182}Ta, unfortunately not specified to distinguish σ_{th} and I_γ. Given here numerical values (except the estimate of 10^5 *b* for ^{194}Ir) are obtained within a standard inaccuracy of about 10% including the errors due to the calibration and recalculations. Remind also a high value of σ_{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 3. 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-neutron capture way differs from the standard *s*- and *r*-processes. The second neutron capture happens prior the β^- decay of a first capture product (unlike to *s*-process), while the capture of the third and further neutrons is improbable (unlike to *r*-process).

Speaking on the nucleosynthesis, one must realize that 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 $T \geq 10^6$ *K* approximately corresponds to the neutron energy of $E_n > 100$ *eV*. The room-temperature thermal cross sections determined in a laboratory 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 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 $E \rightarrow 0$. 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 cross section at 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 in stellar conditions. Our measured cross sections for ^{182}Ta and ^{194}Ir together with the known [3] for ^{198}Au are just the cases of strong enhancement of cross sections by resonances at $E \rightarrow 0$. Possible synthesis of ^{183}W, ^{195}Pt, and ^{199}Hg isotopes is illustrated in Fig. 2.

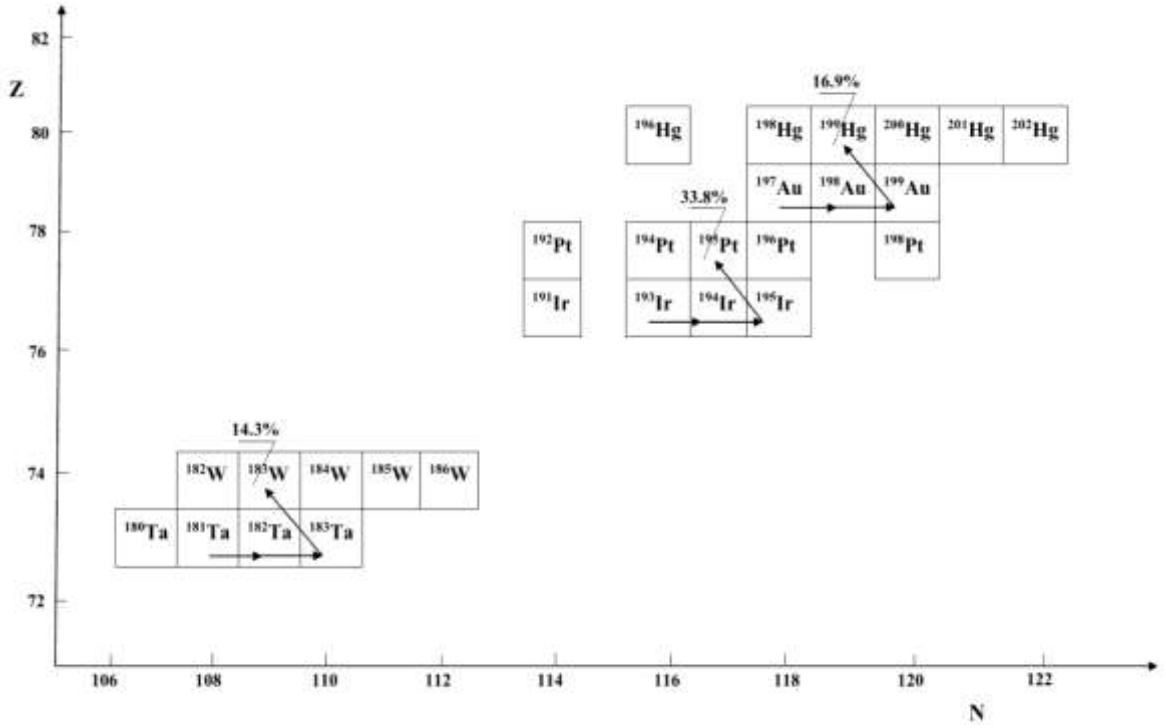

Fig. 2. Processes leading to the synthesis of ^{183}W, ^{195}Pt, and ^{199}Hg isotopes through the double neutron capture reaction, in addition to the standard *s*-process way.

5. 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 in the experiment on irradiation of the ^{193}Ir enriched target at IBR-2 reactor. Great cross section is revealed for the neutron capture reaction by radioactive ^{194g}Ir nuclide, and the detected yield of ^{195m}Pt isomer looks very promising for therapy applications. The latter method is advantageous also because of a possibility to apply the isolation of ^{195m}Pt by chemical means. The ^{nat}Ta targets were irradiated over the experiment as spectators, and they by chance have demonstrated also high 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 with nuclear reaction theory, as well as for studies of the processes in Universe responsible for the natural isotope abundances.

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