Romanian Reports in Physics, Vol. 60, No. 3, P. 539–554, 2008

Dedicated to Prof. Ioan-Iovitz Popescu's 75th Anniversary

SPALLATION WITH INTERMEDIATE ENERGY PROTONS AS SOURCE OF LONG-LIVED ISOMERS

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(Received January 31, 2008)

Abstract. A systematic study of spallation reactions with intermediate energy protons and heavy targets was performed to understand the population of long-lived high-spin isomers in the mass region $A \approx 180$. Several target materials were considered: ^{nat}Ta, ^{nat}Re, ^{nat}W, ^{186}W and ^{nat}Hf. The irradiations were performed at several proton energies in the range 100–660 MeV. The experimentally determined yields were compared to the calculations performed with the LAHET code. Data and calculations were analyzed to determine the optimal combination of target material and proton beam energy for getting the higher absolute cross-section and the best isomer-to-ground state ratio.

Key words: high-spin isomers, protons, spallation, target activation, radionuclides, cross-sections, multistep model.

1. GENERAL REMARKS

Interest in studying spallation reactions induced by high energy protons impinging on heavy targets has been renewed nowadays due to their potentiality for the development of many modern applications. There are two main directions in which such reactions are of fundamental interest. One concerns the production of intense neutron beams by bombarding a thick heavy target with intense high energy proton beams to be used for material physics research or accelerator-driven systems for nuclear waste transmutation and subcritical reactors; we mention here only two main projects under development at the present time: the European Spallation Source (ESS) [1] and the Spallation Neutron Source (SNS) at Oak Ridge [2]. The other important use of the spallation reactions is production of exotic nuclei for nuclear physics research along with projectile fragmentation and fission techniques. There is currently an increased interest in developing beams of isomeric nuclei for the nuclear physics research [3, 4]. It was discussed [3] that by using high-spin isomeric nuclei as secondary beams in fusion reactions one can produce compound nuclei at higher angular momentum due to the contribution of

the intrinsic spin of the isomers. Other interesting uses of the isomeric beams concerns Coulomb excitation, static nuclear moments measurements, transfer reaction. One of the main issues related to the production of isomeric beams is their purification. Evidently, traditional mass separators cannot discriminate isomers from the ground state. Recent technical developments provided new solutions that are already in use or they will be shortly available. Laser resonance ionization, based on the energy differences of the atomic levels due to the hyperfine interaction of the electrons with the different nuclear spins of the ground state and the isomeric state, was successfully used at ISOLDE on-line isotope for the case of $^{68}Cu^{g}/^{68}Cu^{m}$ resulting in an increase of the isomer-to-ground state ratio by a factor of about 400 [5]. The first Coulomb excitation experiments with isomeric beams of $^{68,70}Cu$ produced through this method were recently reported [6]. Sometimes the laser resonance ionization cannot be used, as it is the case of the fragment separator FRS at GSI that can isotopically separate the secondary beams but cannot produce pure isomeric beams. It was shown in Ref. [7] that in this case a high resolution mass separation can be achieved by using cooled beams in a storage ring.

Our research interest focused on the use of spallation reactions as source of long-lived isomers. We investigated the production of high-spin isomers in the mass region $A \approx 180$ by using spallation of heavy targets with protons of intermediate energy. We devoted a particular attention to the production of the $^{178}Hf^{m2}$ isomer. This isomer is a singular case in the nuclear landscape; it is located at about 2.45 MeV excitation energy, it has spin and parity $J^{\pi} = 16^{+}$ and a half-life of 31 years, and decays through gamma-ray emission. Since its discovery in 1968 [8] a lot of experimental work was devoted to understand its structure and decay properties. The isomer was assigned a four quasi-particle (2 proton-type and 2 neutron-type) configuration [8, 9] being the band head of a high-K rotational structure [10, 11] with K = 16. The main decay (99.9%) goes through a highly converted 13 keV E3 gamma-ray transition to the 13^{-} state of the $K^{\pi} = 8^{-}_{1}$ band. A recent high sensibility measurement of the isomer decay [12] established precisely its location at 2446.0 keV excitation energy. The remarkable long lifetime of the isomer is the result of the combined effect of the K quantum number selection rules and location of the isomer at excitation energy lower than any other state with spin 14 ħ or higher, a characteristic of the yrast traps.

The long lifetime of the $^{178}Hf^{m2}$ isomer made it very attractive for the development of isomeric beams from the beginning [13]. The main difficulty is how to produce a pure isomeric beam under the conditions of a very low value of the isomer-to-ground state ratio from the producing reactions. Recent advances in the use of cooled beams in storage rings [7] show that one can attain the sensibility levels needed to separate $^{178}Hf^{m2}$ isomeric nuclei from the ^{178}Hf ground state nuclei ($\Delta M_{(m2\text{-}g)}/M_{g} = 1.5 \times 10^{-5}$). An important advantage of the $^{178}Hf^{m2}$ isomer over other high-spin isomers is the possibility to accumulate isomeric nuclei for long time and produce isomeric targets that can be stored for long time before use.

Important applications as the development of gamma-ray lasers (see [14] and references therein) are related to the possibility of triggering the release of the energy stored in isomeric states under the action of external radiation fields. A scenario in two steps is proposed: a) isomeric nuclei absorb the external radiation and are excited to gateway state(s); b) the gateway state(s) decay by emission of a cascade of fast gamma rays bypassing the isomeric state and reach the ground state. Such an effect was observed for the first time in ^{180}Ta [15]. The ground state of ^{180}Ta has $J^{\pi} = K^{\pi} = 1^{+}$ and a half-life of 8.15 h while the isomeric state $J^{\pi} = K^{\pi} = 9^{-}$ located at 77.1 keV excitation energy [16] has a lifetime of $> 1.2 \times 10^{15}$ years. Following the exposure of samples containing such long-lived isomers to an intense Bremsstrahlung radiation from a 6 MeV medical linac the characteristic X-rays of the ground state decay daughters (^{180}Hf – 86% EC and ^{180}W – 14% β^{-}) were detected. The result was later confirmed by several other measurements (see Refs. [17, 18] for example) and it was established that the minimum energy needed for inducing the decay of the isomer is about 1 MeV. While this case is of no practical interest given the largely unfavorable ratio between the energy needed to trigger the isomer decay and the net energy gain of the process, it was the first demonstration of the possibility to trigger emission of gamma rays from isomeric nuclei. A way more attractive case is the *m2* isomeric state of ^{178}Hf because of its high excitation energy and long lifetime. It was hypothised that in this case a similar triggering process can also be induced with much lower energy electromagnetic radiation. Measurements performed with a dental X-ray source [19] and at Spring8 synchrotron [20] brought evidence in favor of such a process ignited by X-rays of energy as low as ~ 10 keV. The results are still under debate as later measurements performed at the APS synchrotron [21, 22] failed to reproduce them.

Studies with ^{178}Hf^{m2} are strongly limited by the very low inventory of such isomers available around the world. A synthetic summary of the ^{178}Hf^{m2} isomer production schemes is presented in Ref. [23]. The main production methods investigated until now are:

i) *Fusion-evaporation reactions*. The ^{176}Yb(α, 2n)^{178}Hf^{m2} reaction was used at JINR Dubna [24]. The maximum cross-section for the formation of the isomer is about 9 mb and the isomer-to-ground state (σ_m/σ_g) ratio is about 5%. The number of byproducts is reduced. The method requires a cyclotron accelerator to produce the α beam;

ii) *Neutron induced reactions*. The reactions with thermal and low-energy neutrons as ^{177}Hf(n, γ) and ^{178}Hf(n, n$'\gamma$) have very low cross-sections of the order of 10^{-2}–10^{-3} mb with σ_m/σ_g ratio of the order of 10^{-9}–10^{-6} [25]. They require the use of a nuclear reactor for the irradiations and result in a large number of byproducts. The reaction ^{179}Hf(n, 2n) with protons of 14.5 MeV

has a cross section of about 7 mb that is at the level of the fusion-evaporation reactions and σ_m/σ_g ratio only one order of magnitude lower [26]. The number of byproducts is relatively small. With the latest development of compact neutron sources able to deliver a relatively high neutron flux at 14 MeV [27] this method might be revisited in the future.

iii) *Spallation reactions*. The process that produced the largest quantity of $^{178}Hf^{m2}$ isomer (10^{17}) until now was the spallation with a 400 μA proton beam at 800 MeV energy of a ~ 1 kg Ta beam dump at the Los Alamos National Laboratory LAMPF meson factory [28]. In this reaction the σ_m/σ_g ratio for the independent production of both isomeric and ground state nuclei is intermediate between the values for the fusion-evaporation and the (n, 2n) reactions. The main disadvantages of this method are the use of a huge accelerator to produce the high-energy proton beam and the extremely high activity of the irradiated samples.

The present paper summarizes our efforts during the last decade to optimize spallation of heavy targets with intermediate energy protons for the production of high-spin isomers in the mass region A ≈ 180. The main interest was focused on the production of $^{178}Hf^{m2}$ but also other high-spin isomers as $^{177}Lu^{m}$ (E_x = 970.2 keV, $J = 23/2\,\hbar$, $T_{1/2}$ = 160.4 *d*) and $^{179}Hf^{m2}$ (E_x = 1105.8 keV, $J = 25/2\,\hbar$, $T_{1/2}$ = 25.1 *d*) could be identified and measured.

2. EXPERIMENTAL DETAILS

We performed a systematic study of proton-induced spallation reactions at intermediate beam energies on several heavy targets: ^{nat}Ta and ^{nat}Re [29], ^{nat}W and ^{186}W [30], and ^{nat}Hf. The experimental measurements were performed at the Laboratory of Nuclear Problems of the Joint Institute for Nuclear Research Dubna. The proton beam was delivered by the 6 m Synchrocyclotron (phasotron). The maximum proton energy that can be achieved with this accelerator is 660 MeV; irradiations at lower energies can be performed with the internal beam at different radial positions. The proton beam intensity was about 2–3 μA. The targets and their holder were optimized for the efficient removal of the heat released by the beam in the targets. The targets were built in form of metallic foils and mounted on a cooled Al backing. The effective target thickness is defined by its size along the beam direction, as they are mounted tangent to the protons orbits. In this way, even thin foils result in ‘thick’ targets as the protons transit the whole width of the targets. The thickness mentioned in the paper refers to this dimension tangent to the beam direction. A schematic representation of the experimental setup for the samples irradiations is shown in Fig. 1.

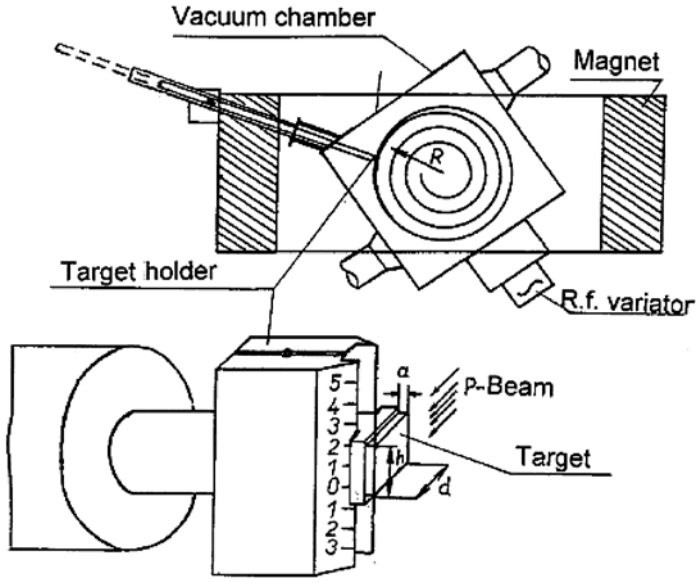

Fig. 1 – The 6m synchrocyclotron (phasotron) of LNP, JINR Dubna. Targets can be irradiated at different energies by inserting them at different radii inside the accelerator. The inset shows an enlarged image of the target holder [29].

After irradiation, the high activity produced by short-lived radionuclides, required the cooling of the targets during few weeks. This period of time sets the limit on the shortest lived radioisotopes that can be identified and measured through our method. To measure the activity following the decay of $^{178}Hf^{m2}$, that is very weak compared to the overall activity of the samples, chemical separation of the Hf fraction from the bulk material was needed. The chemical separation consists of dissolution of the samples in acid, isolation of the Hf fraction from the bulk matter and the major part of the other radionuclides, fine rectification of the Hf fraction, purification of the Hf fraction from Lu accumulated following the Hf decay, isolation of the individual fraction of other elements. Identification of the radionuclides produced during the irradiation and their yields are done through gamma-ray activity measurements. Based on the intensity of the gamma-ray lines, we determined the total number of produced atoms by taking into account the detector efficiency, the decay scheme and the individual spectroscopic properties of the nuclides.

Gamma rays emitted from the irradiated samples were measured with a standard gamma-ray spectroscopy chain consisting of a 20% HP Ge detector, a high-rate spectroscopy amplifier (ORTEC 973) and a multichannel buffer (ORTEC 921). This system can sustain counting rates of up to 20 kcounts/s without significant deterioration of the gamma-ray energy resolution. Since each sample had a different gamma-ray background, both as intensity and composition, for each

measurement we had to optimize the sample – detector distance and the absorbers (Pb, Cd and Cu) in front of the detector to maintain the counting rate of the detector below the limit of 20 kcounts/s. The absolute yields of the radionuclides were estimated from the analysis of the gamma-ray spectra and by using the standard equations for radioactive decay and accumulation. The detection efficiency was measured separately for each sample – detector distance and absorber thickness with standard gamma-ray calibration sources. Well known intense gamma-ray lines emitted by the samples themselves were also used for efficiency calibration; they have the advantage of taking into account in a natural way the self-absorption of the gamma rays in the target material.

2.1. THE p + ^{nat}Ta REACTION

Three ^{nat}Ta targets with a thickness of 33.3 g/cm^2 (about 2 cm thick along the beam direction) were irradiated with protons of 660, 200 and 100 MeV, respectively, at internal positions. A fourth irradiation was performed using the extracted 660 MeV proton beam and a target consisting of a stack of two thin foils of Ta (156 mg/cm^2) and Al (30.6 mg/cm^2). This way the two foils were exposed to the same beam fluence. The cross-sections for the production of ^{7}Be and ^{22}Na through the p + Al reaction were measured with high accuracy in Ref. [31] and they were used to calibrate the cross-section of the products. Natural Ta contains 99.988% of ^{181}Ta and 0.012% of the long-lived isomer ^{180}Ta^m (> 1.2×10^{15} years) so that we could safely consider the samples as being mono-isotopic (^{181}Ta) for all the considerations. The yield of the radionuclide ^{178}W produced in the reaction ^{181}Ta(p, 4n)^{178}W is known with high accuracy [32] and, since the gamma rays associated with its decay are strong, it could also be used for cross-section calibration purposes.

2.2. THE p + ^{nat}Re REACTION

In the case of the ^{nat}Re irradiation, the targets were of 21 g/cm^2 thickness (about 1 cm thick). Four targets were exposed to the internal beam at different incident energies, 150, 300, 450 and 660 MeV, respectively. As in the case of Ta irradiation, a stack of two foils of ^{nat}Re and Al of comparable thickness (0.56 mg/cm^2) were irradiated with the 660 MeV extracted beam for cross-section calibration purposes. After irradiation, the targets were left for 3 weeks to cool down before the first activity measurement was performed. As a result radionuclides with half-lives shorter than 2 days could not be detected. The weakest activity determined with our method corresponds to a production cross-section as low as 1 μb. The ^{nat}Re contains two components, ^{185}Re and ^{187}Re with abundancies of 37.4% and 62.6%, respectively. Intense gamma rays corresponding

to ^{185}Os and ^{183}Re were identified in the measured spectra; they are populated through the ^{187}Re(p, 3n)^{185}Os and ^{187}Re(p, p4n)^{183}Re reactions. Similar reactions with ^{181}Ta target were accurately measured [32].

2.3. THE p + ^{nat}W AND p + ^{186}W REACTIONS

Three targets of enriched ^{186}W (96.8%) in metallic form were built. Each target was about 7 mm thick. The ^{186}W targets were placed on a 50 μm ^{nat}W backing and, then fixed on the cooled Al holder. The irradiations of the samples were performed at the internal beam of the synchrocyclotron. The position of the targets inside the accelerator was chosen to provide incident beam energies of 650, 450 and 300 MeV, respectively. The particular design of the targets allowed for the simultaneous irradiation of the enriched ^{186}W material and of the ^{nat}W holder foil under identical conditions allowing for a direct comparison of the results for both materials. The natural abundance of the ^{186}W isotope is 28.6%. After irradiation the samples were cooled for 1 month so that short-lived radionuclides could not be measured by our method.

2.4. THE p + ^{nat}Hf REACTIONS

Four targets of 13.3 g/cm^2 thickness (1 cm thick) were irradiated with protons at 650, 450, 250 and 120 MeV. The activity of the targets immediately after irradiation was rather high due to the use of massive targets and high beam intensities and, hence, cooling of the targets for one month was necessary before performing gamma-ray activity measurements. Afterwards, the targets were left to cool down for another two and a half months. Since the background gamma-ray activity was still too high to allow identification of the weak activity associated with the decay of the ^{178m2}Hf isomer we performed the chemical purification of the material from other elemental compounds . The chemical procedure was modified to account for the fact that in this case the bulk material was Hf itself. The isotopic composition of ^{nat}Hf is: ^{180}Hf – 35.08(16)%, ^{179}Hf – 13.62(2)%, ^{178}Hf – 27.28(7)%, ^{177}Hf – 18.60(9)%, ^{176}Hf – 5.26(7)%, ^{174}Hf – 0.16(1)%. Therefore, when using a ^{nat}Hf target, the ^{179}Hf^{m2} and ^{178}Hf^{m2} isomers can be produced exclusively through the following reactions: ^{180}Hf(p, p′n)^{179}Hf^{m2}, ^{180}Hf(p, p′2n)^{178}Hf^{m2} and ^{179}Hf(p, p′n) ^{178}Hf^{m2} and inelastic scattering of protons.

3. RESULTS AND DISCUSSION

The use of spallation reactions to produce high-spin isomers as ^{178}Hf^{m2} has to face several problems concerning the reaction mechanism and the characteristics of

the isomer. Spallation populates predominantly neutron-deficient nuclei that in turn decay through EC and β^+ processes towards stable isobars. The ^{178}Hf nucleus is located on the β-stability line and its direct or independent population from spallation has a low cross-section; it results mainly in the ground state as cumulative decay of higher Z isobars. On the other hand, the ^{178}Hf^{m2} isomer, due to its high excitation energy and spin, can result exclusively from independent population. The cross-section of the independent population is spread over many states of lower spin and excitation energy and this reduces the cross-section for the isomer production. Practically, the independent-to-cumulative ratio for ^{178}Hf and the isomer-to-ground state ratio define the obtained yield of ^{178}Hf^{m2}. Both ratios are difficult to be predicted by calculations and our experimental results bring valuable information for the calibration of the computer code simulations.

The use of high energy proton beams will obviously favor direct population of states with high excitation energy but at the same time it will move the mass distribution of the spallation products towards emission of a larger number of nucleons. It is clear that production of the high-spin isomer ^{178}Hf^{m2} is subjected to a very delicate balance between the target material and proton beam energy. Low incident energies will favor production of nuclei in the vicinity of the target while high energies will move the peak of the mass distribution away from the target mass. Spallation at moderate and high proton energies favor population of nuclei emitting many neutrons and only few protons meaning that the target material has to be only few protons away from Hafnium but as rich as possible in neutrons.

To characterize the different combinations of proton beam energy and target material we identified some quality indicators: a) the absolute yield of the ^{178}Hf^{m2} isomer; b) the isomer-to-ground state ratio; c) the yield of the long-lived contaminants; d) the cooling time of the samples. One has to find the optimal combination of these parameters to identify the best reaction for the isomer production.

When we started this work, almost one decade ago, we realized that the experimental cross-section measurements through reactions induced by protons with energy in the range of 0.1–1.0 GeV are not yet well developed. Data were fragmentary, as reported in Ref. [33], and many of the known values were measured mostly in the period 1960–1970 with limited accuracy.

We started with the study of the p + ^{nat}Ta reaction for historical reasons. This was the reaction that produced the largest quantity of ^{178}Hf^{m2} isomer (10^{17}) until now. Production of ^{178}Hf^{m2} isomer in this case resulted as a byproduct of the operation of the LAMPF accelerator [28] and was never optimized for this purpose. The irradiation of a massive beam dump of ^{nat}Ta with a 400 μA proton beam at 800 MeV produced highly radioactive samples that were left to cool down for a long period of time before they could be safely handled. Many Hf isotopes were produced in this reaction with yields orders of magnitude larger than ^{178}Hf^{m2}; two

of them ^{175}Hf ($T_{1/2}$ = 70 d) and ^{172}Hf ($T_{1/2}$ = 1.87 y) produced an intense gamma radiation background for more than a decade so that the chemically separated Hf fraction could be used only 20 years after the irradiation. We proceeded with short irradiations of thinner samples at several proton energies. Then we completed the analysis with the irradiation of ^{nat}Re samples.

A detailed description of the experiments, data analysis and results is given in Ref. [29]. The left panel of Fig. 2 shows the mass-distribution of the cumulative yields for the two reactions at 660 MeV beam energy. The experimental results are compared with model calculations performed with the LAHET code [34] and for all cases the agreement is within a factor 2. The cumulative yields include the most probable products of spallation and the good agreement with the calculations confirms that the model reproduces well the spallation cross-sections near the maximum of the Z number distribution. Nuclei produced as independent yield are located on the tail of the Z number distribution on chains of isobars, far from the most probable charge and correspond to low yields. This is the case of the high-spin isomers of interest like ^{177}Lu^{m}, ^{178}Hf^{m2} and ^{179}Hf^{m2}. The calculated independent yields for two isobar chains are shown in the right panel of Fig. 2. It is clear that ^{178}Hf in the p + ^{187}Re reaction is located on the tail of the Z number

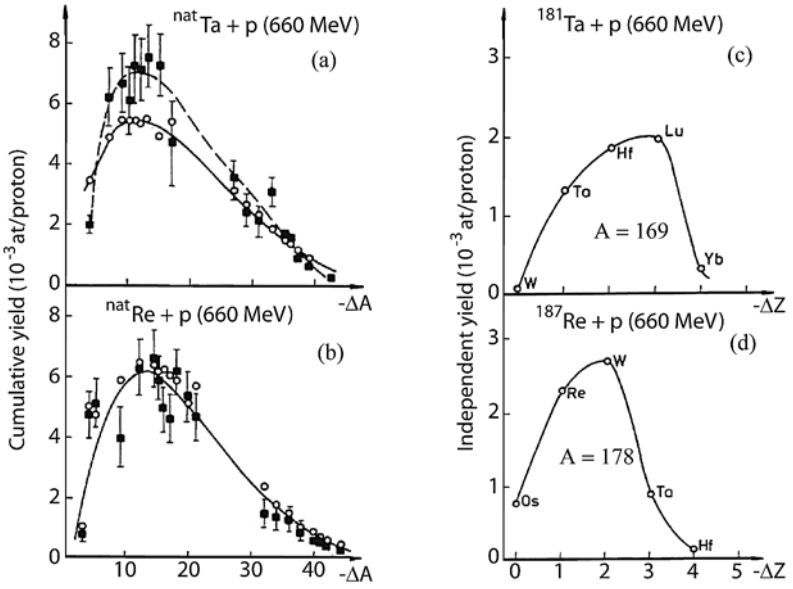

Fig. 2 – Left panel: mass-distributions of the cumulative yields for nuclides produced via the p + ^{nat}Ta (a) and p + ^{nat}Re (b) reactions at 660 MeV. Right panel: isobaric charge distributions for A = 169 nuclides produced in p + ^{181}Ta reaction (c) and A=178 in p + ^{187}Re reaction (d) at 660 MeV as calculated with the LAHET code. Black squares stand for the experimental results while the opened circles are calculated values.

distribution while at the maximum of the distribution we find the neutron deficient isotope ^{178}W. The ^{178}W ($T_{1/2}$ = 21.6 d) isotope will EC decay to ^{178}Ta that in turn will decay to the ground state of ^{178}Hf contributing to the degradation of the isomer-to-ground state ratio for the ^{178}Hf^{m2} isomer. To avoid such degradation effect the ^{178}W isotope has to be eliminated from the samples as soon as possible after irradiation.

The calculations are not able to distinguish between the independent production of isomers and ground state nuclei. Our experimental results bring valuable information for a quantitative treatment of the problem. In Ref. [35] it was shown for reactions induced by low energy nucleons that the isomer-to-ground state ratio is mostly determined by the angular momentum of the reaction product. Unfortunately, the presently available calculations cannot predict with accuracy the angular momentum distribution of the reaction residues.

A qualitative discussion [29] have shown that, in the case of reactions induced by high energy protons, the average angular momentum distribution of the residual nuclei has a maximum of about 10 h for residual nuclei with 10–15 nucleons less than the target material. Based on such arguments we performed a new round of measurement with ^{nat}W and ^{186}W. The measured cross-sections for 660 MeV protons are shown in Fig. 3. The experimental results are compared with the LAHET calculations and with the values from the previous measurements with ^{nat}Ta and ^{nat}Re targets. One can notice the wide peak located around mass 80–85; it corresponds to the fission fragments. The larger scattering of the points corresponding to the fission products as compared to the spallation ones is due in part to the lower yields and consequently higher statistical errors but also to the incomplete accumulation for some of the yields.

One can notice the general good agreement between the calculated and experimental results. The agreement becomes remarkably good near the maximum of the spallation products distribution. The total yield of the spallation reaction and the yields for the cumulative products are well reproduced. The agreement is worse for the fission products but we have to consider that the calculation of fission depends on several parameters that have still to be calibrated from the comparison with experimental data.

The available data allowed us to compare the isomer-to-ground state ratio and absolute cross-section for the population of high spin isomers with high energy protons impinging on heavy targets. The results for the irradiations at 650 MeV and 660 MeV are summarized in Table 1. The cross-sections for the main contaminants are also given.

For the stable isotopes we could not determine experimentally the ground state and total yields through our measuring method. For the calculation of the isomer-to-ground state ratio we had to use the calculated values. It can be seen that the cross-sections for the production of background isotopes are more or less constant for the cases considered but there are large variations for the production of

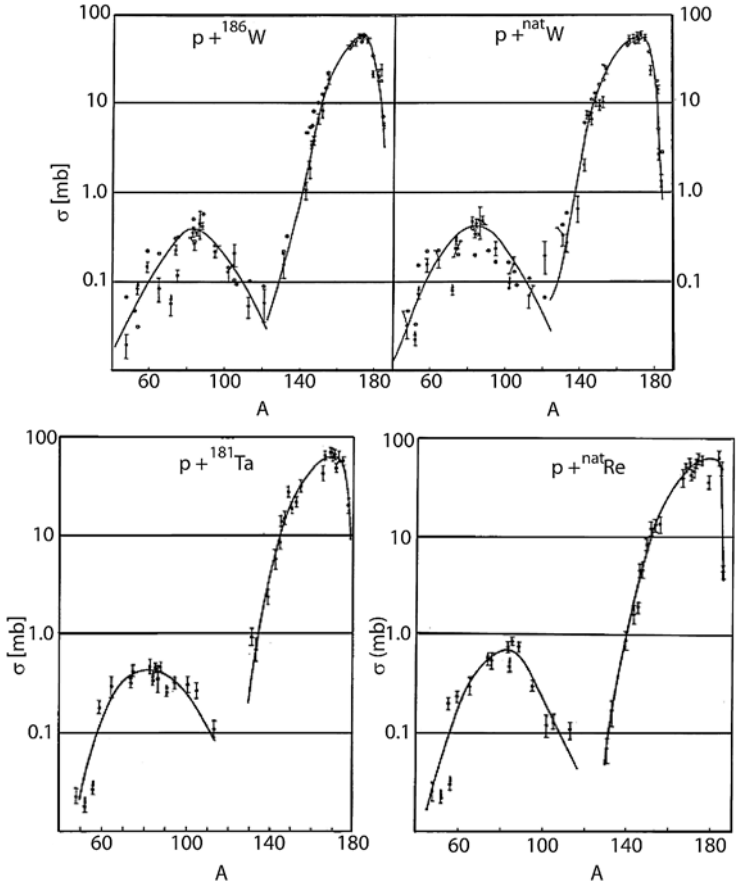

Fig. 3 – Upper panel: mass-distributions of the nuclides produced after the irradiation of ^{186}W and ^{nat}W targets with protons of 650 MeV. Lower panels: mass-distributions of the nuclides produced after the irradiation of ^{nat}Ta (left) and ^{nat}Re (right) targets with protons of 660 MeV. Experimental results are shown as black circles with error bars while the ones calculated with LAHET are represented with opened circles.

high-spin isomers. Similar results are obtained also for the reactions with 450 MeV protons. It results that ^{186}W is the best material for producing the high-spin isomers of interest through spallation with intermediate energy protons.

In the experiments with ^{nat}Re targets [29] we noticed that the ^{184m}Re high-spin isomer ($T_{1/2}$ = 169 *d*, J$^{\pi}$ = 8$^{(+)}$, E_x = 188 keV) was produced with high yield at proton energy as low as 100 MeV. The population of this isomer is achieved without net loss of protons. The possible reactions populating this isomer are: ^{185}Re(p, p′n)^{184m}Re and ^{187}Re(p, p′3n)^{184m}Re. It means that one can get high angular momentum even in residual nuclei with masses close to the target, after spallation with low energy protons. To investigate this possibility for populating

Table 1

Summary of the cross-sections and isomer-to-ground state ratios for the high-spin isomers of interest produced in the spallation reactions under investigation. The values for the ground state population cross-section are calculated. The cross-sections for the main contaminants producing high gamma-ray background are given for comparison

Nuclide	Reaction			
	p + ^{nat}Ta @ 660 MeV	p + ^{nat}W @ 650 MeV	p + ^{186}W @ 650 MeV	p + ^{nat}Re @ 660 MeV
Cross-section σ (mb)				
^{179}Hf^{m2}	0.52	0.36	0.80	0.12
^{178}H ^{m2}	0.31	0.18	0.48	0.13
^{177}Lu^{m}	0.15	0.13	0.26	0.04
^{178}W	5.90	23.00	21.80	36.00
^{175}Hf	56.00	55.00	55.60	59.00
^{172}Hf	47.00	53.50	57.40	55.00
^{173}Lu	61.00	61.00	60.00	61.00
Isomer-to-ground state ratio σ_m/σ_g				
^{179}Hf^{m2}	0.040	0.140	0.250	0.240
^{178}Hf^{m2}	0.021	0.044	0.092	0.140
^{177}Lu^{m}	0.103	0.210	0.290	0.400

high-spin isomers as ^{178}Hf^{m2} and ^{179}Hf^{m2} we performed the irradiation of ^{nat}Hf targets with protons of different energies.

The p + ^{nat}Hf reaction has two main benefits: it eliminates the production of ^{178}W that contributes to the production of useless ground state ^{178}Hf and it results in a lower yield of the long-lived contaminant ^{172}Hf. The cross-sections measured from the irradiation of the ^{nat}Hf sample at 650 MeV are shown in Fig. 4. The reported values can be directly compared with the results from the other measurements reported in this paper. The samples of ^{nat}Hf were contaminated with Zr in proportion of about 3% and this explains the appearance of the products from Zr spallation.

The production of the ^{179}Hf^{m2} and ^{178}Hf^{m2} isomers is enhanced only at the lowest irradiation energy (120 MeV). For the production of ^{178}Hf and ^{179}Hf nuclei only the heavier isotopic components of the targets, with masses 179 and 180, can contribute. In the case of ^{179}Hf^{m2} only the ^{180}Hf component provides real yield through the ^{180}Hf(p, p′n)^{179}Hf^{m2} reaction. The ^{178}Hf^{m2} isomer is most probably produced mainly in the ^{179}Hf(p, p′n) reaction because of its advantageous spin-deficit $\Delta I = 11\,\hbar$ because the ground state spin of the ^{179}Hf is $9/2\,\hbar$. The other possible reaction ^{180}Hf(p, p′2n)^{178}Hf^{m2} is characterized by a higher $\Delta I = 31/2\,\hbar$ value because the ground state spin of the ^{180}Hf is 0 h and it should result in a much lower contribution to the population of the 16^+ isomer ^{178}Hf^{m2}. Based on such

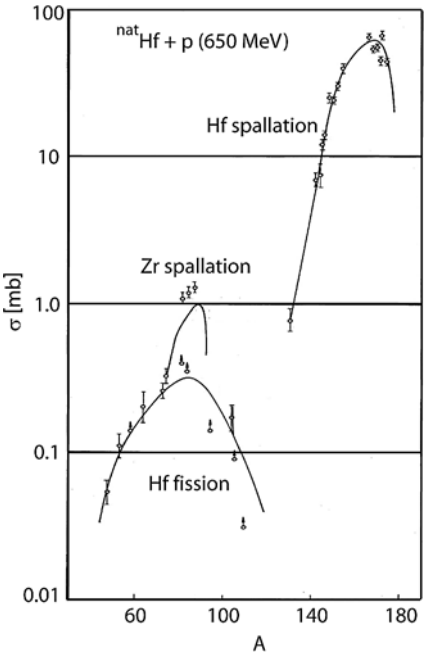

Fig. 4 – Cross-section of the products populated in the p + ^{nat}Hf reaction at 650 MeV. The mass distributions corresponding to the spallation of Hf and Zr and fission of Hf are evidenced.

qualitative analysis, one may conclude that $^{178}Hf^{m2}$ and $^{179}Hf^{m2}$ are both produced through the same type of reaction (p, p′n). The two reactions have similar spin-deficit parameter $\Delta I = 11$ and $12\,\hbar$, respectively. This means that, in first approximation, the ratio of the isomer yields $^{178}Hf^{m2}/^{179}Hf^{m2}$ should be primarily determined by the ratio of the isotopic abundance of the two Hf isotopes, 179 and 180. The experimental ratio of the isomers mean cross-sections: $^{178}Hf/^{179}Hf^{m2}$ = = 0.44 is indeed very close to the ratio of the ^{179}Hf and ^{180}Hf isotopic abundances (= 0.39). To verify the validity of such intuitive reasoning we plan to perform the irradiation of enriched ^{179}Hf targets with low energy proton beams.

4. CONCLUSIONS

We performed a systematic study of the spallation reactions induced by protons with energies in the range of 100–660 MeV on several heavy targets: ^{nat}Hf, ^{nat}Ta, ^{nat}Re, ^{nat}W and ^{186}W. The yields of the radionuclides produced in these reactions were measured using the activation method. The sensibility of the measurements was high and cross-sections down to 1 μb could be identified. For each measurement up to 60–70 radionuclides were measured. We evaluated the reactions from the point of view of the population of high-spin isomers with special interest for the $^{177}Lu^{m}$, $^{178}Hf^{m2}$ and $^{179}Hf^{m2}$ isomers. We compared the absolute

cross-section of the isomers, the isomer-to-ground state ratio, gamma-ray background activity for each combination of proton energy and target material. The experimental results were compared with Monte Carlo calculations performed with the LAHET code. The calculations are in rather good agreement with the experimental results and they were used for reliable estimates of the ground state yields of the nuclei close to the β-stability line. Spallation of ^{186}W with high energy protons resulted in the highest cross-section for the mentioned isomers and the estimated isomer-to-ground state ratios are among the highest reported in the literature. A striking result was the possibility to populate high-spin isomers with protons of about 100 MeV and a reduced number of neutron evaporation. Preliminary results from the irradiation of ^{nat}Hf samples support this conclusion.

Acknowledgments. The authors are grateful for the ongoing support and assistance provided by the IGE Foundation. Special thanks are owed to Academician Professor Ioan Iovitz Popescu, Director of the IGE Foundation, for his kind support, valuable advices and continuous encouragement to perform this research work.

Effort sponsored by the Air Force Office of Scientific Research, Air Force Material Command, USAF, under grants number F61775–99–WE030, F61775–00–WE056, FA8655–02–13065 and FA8655–04–1–3046. The U.S. Government is authorized to reproduce and distribute reprints for Government purpose notwithstanding any copyright notation thereon. The views and conclusions contained herein are those of the authors and should not be interpreted as necessarily representing the official policies or endorsements, either expressed or implied, of the Air Force Office of Scientific Research or the U.S. Government.

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