E6-2008-71

S. A. Karamian[1], C. A. Ur[2], J. Adam[1,3], V. G. Kalinnikov[1],
N. A. Lebedev[1], G. K. Vostokin[1], C. B. Collins[4], I. I. Popescu[2]

SPALLATION AND FISSION PRODUCTS IN THE ($p+^{179}$Hf) AND ($p+^{\mathrm{nat}}$Hf) REACTIONS

Submitted to «Nuclear Instruments and Methods A»

[1] Joint Institute for Nuclear Research, Dubna, Russia
[2] H. Hulubei National Institute of Physics and Engineering and Induced Gamma Emission Foundation, Bucharest, Romania
[3] Institute of Nuclear Physics, Řež, Prague, Czech Republic
[4] Center for Quantum Electronics, University of Texas at Dallas, USA

Карамян С. А. и др. E6-2008-71
Продукты скалывания и деления в реакциях ($p + {}^{179}$Hf) и ($p + {}^{\rm nat}$Hf)

Экспериментально изучено образование высокоспиновых изомеров Hf и Lu в реакциях скалывания при промежуточной энергии протонов. Мишени из обогащенного ^{179}Hf (91 %) и $^{\rm nat}$Hf облучены на внутреннем пучке ускорителя фазотрон в Дубне при энергиях в интервале от 90 до 650 МэВ. Активационные выходы продуктов реакций измерены с применением методов γ-спектроскопии и радиохимии. Обнаружено, что сечения образования изомеров ^{179m2}Hf, ^{178m2}Hf и ^{177m}Lu сопоставимы с полученными ранее в реакциях скалывания на мишенях от Ta до Re. Таким образом, реакции эксклюзивной эмиссии только нескольких нуклонов, типа (p, p'), (p, $p'n$) и (p, $2pn$), способны передавать достаточный угловой момент продукту реакции и обеспечивать немалое сечение.

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

Препринт Объединенного института ядерных исследований. Дубна, 2008

Karamian S. A. et al. E6-2008-71
Spallation and Fission Products in the ($p + {}^{179}$Hf) and ($p + {}^{\rm nat}$Hf) Reactions

Production of the high-spin isomers of Hf and Lu has been experimentally studied in the spallation reaction with intermediate energy protons. The targets prepared of enriched ^{179}Hf (91%) and $^{\rm nat}$Hf have been exposed to protons with energy range from 90 to 650 MeV using the internal beams of the Dubna Phasotron accelerator. Activation yields of the reaction products are measured by methods of γ-spectroscopy and radiochemistry. The production cross sections for ^{179m2}Hf, ^{178m2}Hf and ^{177m}Lu have been found comparable to the values measured earlier through spallation of Ta to Re targets. Thus, the exclusive reactions with emission of only a few nucleons, like (p, p'), (p, $p'n$) and (p, $2pn$), can transfer enough angular momentum to the product and provide reasonable cross sections.

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

Preprint of the Joint Institute for Nuclear Research. Dubna, 2008

INTRODUCTION

As is known, the fragmentation of nuclei at intermediate energies serves as one of the most productive methods for the radionuclide synthesis, in particular, of the exotic species near the border of nuclear stability, i.e., nuclei with extreme proton, or neutron excess. During the latter decade, an interest has been arisen to production of isomers, especially those characterized by highest excitation energy, exotic structure and long half-life. This is motivated by the discussed possibility to use the long-lived isomeric material as an energy capacitor for the energy storage and release by demand. Quasiparticle K-hindered isomers of Lu and Hf at the mass range near $A = 180$ are characterized by the appropriate properties for that.

In Refs. [1, 2], the yields of high-spin isomers were measured in the spallation of the Ta to Re targets exposed to protons with energy range from 100 to 650 MeV. The enriched ^{186}W target was indicated as one of the most productive targets for accumulation of the 31-year-lived ^{178m2}Hf isomeric state located at excitation energy of $E_i = 2446$ keV and at spin $I^\pi = 16^+$. Unfortunately, an optimum cross section of about 0.5 mb can be reached at relatively high proton energy, as 400–600 MeV, and a high power accelerator is necessary to meet the requirement of highest productivity with the ^{186}W target. Then, the idea has been proposed to test the $^{\rm nat}$Hf and ^{179}Hf targets for optimization of the yield of such isomers as ^{177m}Lu, ^{178m2}Hf and ^{179m2}Hf. They should be accessible in reactions after emission of only a few nucleons, means using a proton accelerator to moderate energies, near by 100–150 MeV. Higher product cross section may also provide lower expensive irradiations. The cross section for a high-spin product must be significantly increased due to the initial spin, $I^\pi = 9/2^+$, of the ^{179}Hf target nucleus.

For nuclear reaction physics, an extension of the spallation studies to the experiments with Hf targets should bring the valuable information for comparative analysis of the isomeric yield in reactions with different projectiles and targets. A review of the methods and reactions have been presented in Ref. [3] being updated to 2004. In addition to the spallation experiments, the advanced studies have been reported and reviewed in [3] for isomer yields in the different reactions induced by bremsstrahlung, neutrons and ^{4}He ions.

For spallation reactions, the theoretical predictions are not available on the yields of high-spin isomers, because the angular momentum coordinate remains out of consideration in the known simulation codes while the isomer-to-ground state ratio, σ_m/σ_g, must restrict significantly the yield of concrete isomeric states. Simulation of the yields for Lu and Hf isomers in the Hf targets exposed to protons looks especially difficult also due to very specific type of the reactions, namely of the exclusive emission of a few nucleons: $(p,\ p')$; $(p,\ p'n)$; and $(p,\ 2pn)$. Thus, only experimental results can supply reliable data on the yield of isomers, and the present experiment seems to be important and intriguing for a new conclusion on the reaction mechanism.

1. EXPERIMENT AND RESULTS

In our previous experiments [1, 2] on the spallation of Ta to Re targets, metal samples were irradiated. Even enriched ^{186}W material was available [2] in a metal form and in enough amount. At the present series, metal Hf foils could be used as the $^{\rm nat}$Hf target, but the enriched ^{179}Hf has been commercially supplied in a form of the HfO_2 oxide. Our test with the natural HfO_2 powder shows not very good quality of layers prepared by the mechanical pressing it down to the Al backing plate. Thus, we ought to apply the chemical deposition method for the ^{179}Hf targets preparation.

The Hf oxide was transformed to the nitrate salt and the solutions of the Hf nitrate (hidrated) in acetone together with organics additions were pasted onto 6μ thick Al foil. The foil was then heated up and the hydrated salt was decomposed again to HfO_2 and was fixed on the Al surface as the mechanically tough layer. The organics additions were burned in this process. Repeating the deposition many times, one can reach 5 mg/cm^2 thickness of the Hf oxide layer that remains mechanically strong. Thicker layers are not as stable and make a risk of the material loss during the irradiation. All these operations were tested first using the $^{\rm nat}$Hf material.

Finally, this method was applied to the preparation of the enriched ^{179}Hf targets. A multiply folded sandwich of the $^{179}HfO_2$ layers on the Al foils was prepared and used for irradiation with the internal beam at the Dubna Phasotron (synchrocyclotron) at DLNP. Four sandwiches have been irradiated at different energies, each has contained of about 70 mg of ^{179}Hf enriched to the abundance of 91%. The sandwiches have a surface size of 10×15 mm. The target sandwich was installed onto a holder and inserted inside the cyclotron chamber at definite radius. The proton energy has been defined by the chosen radius value. Corresponding calibration function — energy versus radius — is known with good accuracy from the accelerator staff.

Internal beam circulates in cyclotron by almost circular orbits and it touches tangentially the target surface penetrating in it because of relatively long proton range as compared to the 10 mm target length along the beam. Due to the vertical oscillations, the beam has finite size, but not larger than the target height of 15 mm. Transmission of protons through the sandwich target is illustrated in Fig. 1.

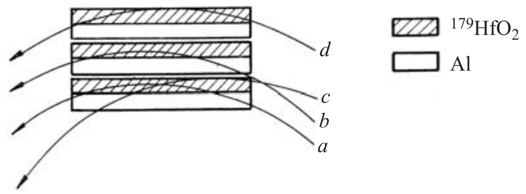

Fig. 1. Schematic illustration of the proton orbits transmitting through the multi-layer target at internal radius of the synchrocyclotron

A curvature of the proton orbits is relatively low in the cyclotron, for instance, a radius value of about 120 cm corresponds to the proton energy of 110 MeV. Particles are directed almost parallel to the target surface. But the thickness of layers in the target is very low as well, and even small curvature of the trajectory would provide the particle crossing through the layers. In Fig. 1, the curvature is enhanced for better illustration of the real scheme. One important peculiarity should be in account: the presence of radial oscillations of the orbits. As a result, the different-kind trajectories are possible, they are shown schematically in Fig. 1. This pattern allows one to calculate more reliably the mean energy losses for projectiles initiating the reactions in the target. Remind that linear energy loss dE/dx parameter for proton transmission in Al is lower than that in HfO_2 by a factor of about 2.5. Major set of trajectories crosses both Al and HfO_2 layers. The trajectories of type «d» in Fig. 1, when particle is going explicitly within the HfO_2 layer, appear not very often. More probable variant includes a half range in Al and half — in HfO_2.

Four sandwich targets were exposed to protons in the Phasotron at radiuses corresponded to 255, 155, 130 and 110 MeV. Calculated mean energy values are 242, 139, 112 and 90 MeV, respectively. Below, we use the latter mean energies to attribute the obtained experimental results. The target length is relatively short, and the observed yield of a product should be proportional to the mean cross section because secondary reactions may be neglected. The presence of known amount of Al in the exposed target allows one to calibrate the whole set of measured cross sections in absolute values referring to the detected yield of ^{22}Na. The cross sections for ^{22}Na are known from literature.

Another advantage of the chemically prepared HfO_2 target at Al substrate as compared to metal Hf samples appears due to the absence of the Zr admixture in ^{179}Hf after mass-separation and chemical isolation. The Zr spallation did create the background production of radioactive nuclides in the mass-number range corresponded to the fission fragment products of the ($p + {}^{\rm nat}$Hf) reaction. With enriched ^{179}Hf the Zr contamination is excluded, however a large amount of Al creates the additional γ-background in the wide range of γ energies due to the intense long-lived activities of ^{22}Na, $E_\gamma = 1274.5$ and 511 keV, and of ^{7}Be, 477.8 keV. Compton continuum covers the range of γ energies at $E_\gamma \leqslant$ 1274.5 keV and disturbs the observation of low intensity lines corresponded to some fission fragments and isomers. In addition, the yields of ^{22}Na and of ^{7}Be nuclides in the ($p + {}^{179}$Hf) reaction remain indefinite because they are produced with much higher cross section in Al.

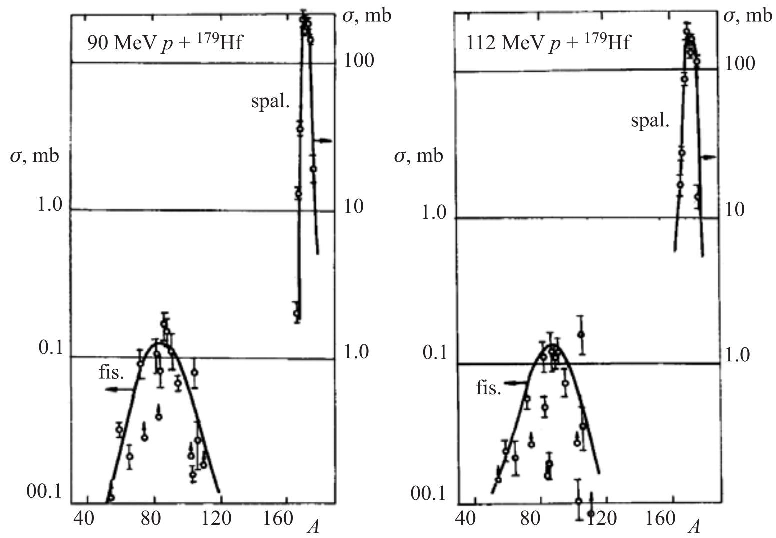

Fig. 2. Fragmentation-product mass distributions as measured for the ^{179}Hf target exposed to 90 and 112 MeV protons

For comparison, the experiment with metal $^{\rm nat}$Hf foils have been performed at similar conditions in the internal beam at the Phasotron accelerator. Radius values have been selected corresponding to four mean-energy values of the projectile protons: 100, 235, 440 and 640 MeV.

Spectra of gamma activity have been measured for all $^{\rm nat}$Hf and ^{179}Hf targets several times during 5 months past irradiation. The standard «Canberra» 20% coaxial HP Ge and better resolution planar X-ray spectrometer both were used in the measurements. Typical methods of the efficiency calibration were applied using the standard sources and by intrinsic γ lines. The spectra were accumulated within the «Maestro» code format and they were analyzed then applying the

«Deimos» code. After a γ-line fit, its half-width, energy position, area and standard deviations were determined with the best reliability and accuracy as possible following the spectral data.

The individual γ-line identification and decomposition of the spectra were performed basing on known properties of radionuclides contained at the nuclear data base and in tables of isotopes. An area of some identified γ line allows one to deduce (applying the decay factors) a number of atoms for the corresponded radionuclide and to determine the relative yield of this product at the irradiation run. Finally, the production cross sections are determined for many radioactive nuclei. In general, the system of data is schematically similar to the results published in [1, 2] for Ta to Re targets. The representative spallation peak is strongly manifested for isotopes with mass-numbers near and below the atomic weight of the target material. The fission-fragment yield provides the second peak positioned at $A \leqslant 110$. Numerical values of the measured cross sections are listed in Tables 1 and 2.

The product-mass distributions for $(p + {}^{179}\mathrm{Hf})$ reaction at 4 values of the proton energy E_p are plotted in Fig. 2 and 3. For comparison, the results taken with the ${}^{\mathrm{nat}}$Hf targets at $E_p = 100$ and 400 MeV are presented in Fig. 4. The enriched ^{179}Hf and ${}^{\mathrm{nat}}$Hf targets demonstrate schematically similar mass distribution curves. However, the spallation yield of the Lu and Hf isomers is strongly different, this is discussed below.

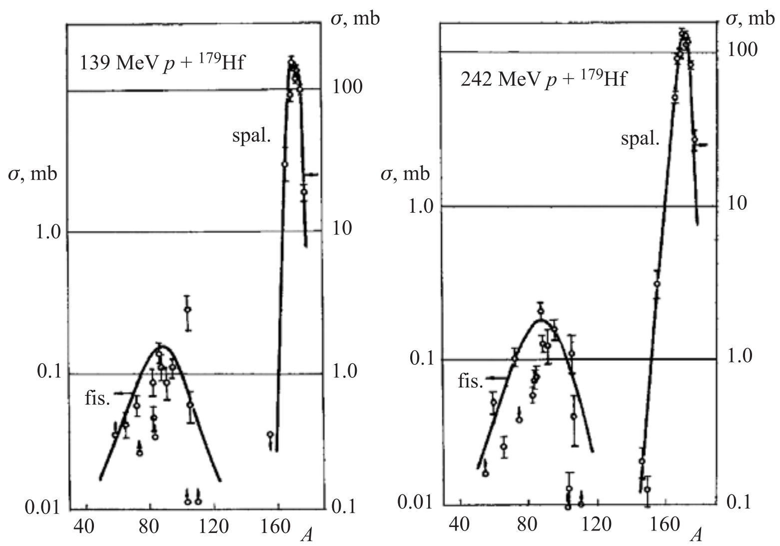

Fig. 3. Mass distributions taken at proton energies of 139 and 242 MeV with the ^{179}Hf target

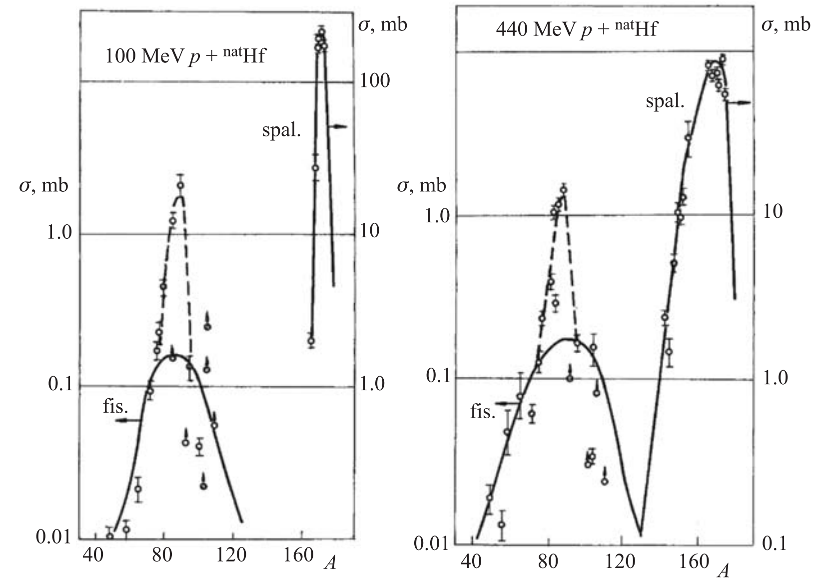

Fig. 4. The same as Fig. 3, but taken with the $^{\rm nat}$Hf targets exposed to 100 and 440 MeV protons

The individual mass-yield at spallation is determined by the cumulative yield of the end-point radionuclide for the isobaric chain of definite A. Such a nuclide recuperates almost a whole yield of the chain because the spallation products are typically neutron deficient and shifted far from the β-stability line. For fission products, the mass yields are estimated with restricted accuracy due to the incomplete cumulativity of many products in addition to standard deviations defined by the measurement procedure.

As is known, the isomeric yield is suppressed due to the isomer-to-ground state ratio σ_m/σ_g and because of the N/Z ratio for an isomer that is not optimum for production via the spallation. The ^{178m2}Hf activity is in addition lowered just due to its long half-life, 31 y. The γ lines of ^{177m}Lu and ^{179m2}Hf have been observed very clearly in the direct spectra, but the ^{178m2}Hf lines were not well observed in presence of intense γ-background. Chemical purification of the Hf material was need for detection of ^{178m2}Hf. The chemical treatment in this case should provide purification of the bulk Hf material from all radioactive micro-admixtures that is not the same task, as previously developed standard procedure of the selective chemical isolation of Hf fraction from the bulk amount of other elements (Ta, W, Re). But after extensive works, the satisfactory purification factor was obtained, and the ^{178m2}Hf yield was measured reliably.

Table 1. Cross-section values (in mb) for the fragmentation products with the $^{\mathrm{nat}}$Hf and ^{179}Hf targets measured at mean proton energies of 235 and 242 MeV, respectively. Errors are given in brackets

Isotope	$T_{1/2}$	E_γ, keV	Type of yield	$^{\mathrm{nat}}$Hf + 235 MeV p	^{179}Hf + 242 MeV p
^{179m2}Hf	25.1 d	453.7; 362.3	Indep.	0.33(0.06)	0.44(0.03)
^{178m2}Hf	31 y	574.2; 495.0	Indep.	0.15(0.03)	0.39(0.06)
^{175}Hf	70 d	343.4; 432.8	ε-cum.	80(3)	76(3)
^{172}Hf	1.87 y	1093.6; 900.7	ε-cum.	69(3)	75(3)
^{177g}Lu	6.65 d	249.7	Indep.	14(3)	25(4)
^{177m}Lu	160.4 d	413.7; 418.5	Indep.	0.49(0.06)	0.90(0.09)
^{174g}Lu	3.31 y	1241.8	Indep.	13.2(1.5)	13,0(0.6)
^{174m}Lu	142 d	992.1	Indep.	7.4(1.5)	9.9(1.5)
^{173}Lu	1.37 y	272.0	ε-cum.	115(7)	111(4)
^{172}Lu	6.7 d	1093.6; 900.7	Indep.	24(3)	27.9(0.6)
^{171}Lu	8.22 d	739.8; 667.4	ε-cum.	124(8)	122(5)
^{170}Lu	2 d	985.1	ε-cum	–	128(13)
^{169}Yb	32.0 d	307.7	ε-cum.	98(7)	91(3)
^{166}Yb	2.36 d	2052.4	ε-cum.	90(15)	48(6)
^{168}Tm	93.1 d	447.5; 720.3	Indep.	1.5(0.2)	2.3(0.3)
^{167}Tm	9.25 d	207.8	ε-cum.	101(7)	88(5)
^{160}Tb	72.3 d	1177.9	Indep.	0.015(0.003)	0.024(0.006)
^{156}Tb	5.35 d	534.3	Indep.	3.4(0.2)	0.20(0.05)
^{155}Tb	5.32 d	367.4	ε-cum.	20(8)	3.0(0.6)
^{149}Gd	9.3 d	748.2	ε-cum.	1.0(0.07)	0.09(0.03)
^{146}Gd→ ^{146}Eu	48.3→4.5 d	633.7	ε-cum.	0.15(0.005)	0.021(0.005)
^{156}Eu	15.2 d	2097.7	β^--cum.	0.2(0.05)	–
^{149}Eu	93 d	277.0	ε-cum.	1.05(0.07)	–
^{148}Eu	54.5 d	550.3	Indep.	0.03(0.006)	–

Table 1 (continuation)

Isotope	$T_{1/2}$	E_γ, keV	Type of yield	$^{\rm nat}$Hf + 235 MeV p	^{179}Hf + 242 MeV p
^{110m}Ag	249.9 d	884.7	Indep.	0.025(0.008)	0.007(0.001)
^{106m}Ag	8.3 d	1045.8	Indep.	0.058(0.015)	0.041(0.015)
^{105}Ag	41.3 d	443.4; 644.5	ε-cum.	0.17(0.01)	0.108(0.031)
^{102m}Rh	207 d	475.1	Indep.	0.024(0.009)	0.010(0.003)
^{103}Ru	39.4 d	497.1	β^--cum.	0.026(0.012)	0.014(0.004)
^{95m}Tc	60 d	582.1	Indep.	0.035(0.010)	0.021(0.010)
^{95}Nb	34.97 d	765.7	Indep.	0.058(0.004)	0.081(0.009)
^{91m}Nb	60.9 d	1204.8	Indep.	0.21(0.04)	0.12(0.03)
^{95}Zr	64.0 d	756.7; 724.2	β^--cum.	0.048(0.003)	0.011(0.003)
^{88}Zr	83.4 d	392.9	β^+-cum.	1.16(0.12)	0.073(0.010)
^{88}Y	106.6 d	898.1; 1836.0	Indep.	0.53(0.06)	0.051(0.006)
^{87}Y	3.35 d	388.4	β^+-cum.	–	0.20(0.03)
^{85}Sr	64.8 d	514.0	β^+-cum.	1.14(0.12)	–
^{82}Sr	25.3 d	776.5	β^+-cum.	0.44(0.06)	0.056(0.006)
^{84}Rb	32.8 d	881.6	Indep.	0.16(0.02)	0.075(0.015)
^{83}Rb	86.2 d	520.4	β^+-cum.	0.95(0.07)	0.071(0.010)
^{75}Se	119.8 d	264.6	β^+-cum.	0.16(0.03)	–
^{72}Se	8.5 d	834.0	β^+-cum.	0.12(0.02)	0.10(0.01)
^{74}As	17.77 d	595.8	Indep.	0.14(0.03)	0.039(0.006)
^{65}Zn	244.3 d	1115.5	β^+-cum.	0.04(0.01)	0.026(0.004)
^{60}Co	5.27 y	1332.5	Indep.	0.016(0.005)	–
^{56}Co	78.8 d	2598.6	ε-cum.	0.006(0.001)	–
^{59}Fe	44.5 d	1291.6;1099.5	β^--cum.	0.028(0.006)	0.051(0.009)
^{54}Mn	312.2 d	835.9	Indep.	0.048(0.011)	0.017(0.005)
^{48}V	16 d	1312.1	ε-cum.	0.035(0.007)	–
^{22}Na	2.6 y	1274.5	β^+-cum.	0.023(0.004)	–
^{7}Be	53.3 d	477.8	Indep.	0.058(0.009)	–

Table 2. The same as Table 1, but taken at mean proton energies of 139, 112 and 90 MeV with the ^{179}Hf target

Isotope	^{179}Hf + 139 MeV p	^{179}Hf + 112 MeV p	^{179}Hf + 90 MeV p
^{179m2}Hf	0.50(0.04)	0.56(0.04)	0.79(0.04)
^{178m2}Hf	0.44(0.09)	0.62(0.10)	0.68(0.06)
^{175}Hf	98(3)	111(4)	153(5)
^{172}Hf	100(4)	115(4)	165(5)
^{177g}Lu	16(3)	12.5(2.5)	18(4)
^{177m}Lu	0.75(0.08)	0.73(0.10)	0.67(0.08)
^{174g}Lu	10.9(0.6)	11.6(0.6)	9.6(0.6)
^{174m}Lu	8.5(1.5)	7.8(1.6)	7.1(1.8)
^{173}Lu	133(4)	150(4)	200(5)
^{172}Lu	22.1(0.6)	17.6(0.6)	12.7(0.4)
^{171}Lu	147(5)	157(8)	210(7)
^{170}Lu	151(25)	178(31)	212(30)
^{169}Yb	90(4)	84(5)	35(2)
^{166}Yb	30(8)	16.3(2.5)	2.0(0.3)
^{168}Tm	0.88(0.11)	0.60(0.10)	0.31(0.05)
^{167}Tm	60(3)	26.3(1.5)	13(1)
^{160}Tb	0.019(0.006)	0.023(0.006)	–
^{156}Tb	0.09(0.03)	0.05(0.01)	–
^{155}Tb	0.36(0.06)	–	–
^{110m}Ag	0.012(0.004)	0.007(0.002)	0.018(0.004)
^{106m}Ag	0.060(0.015)	0.038(0.013)	0.027(0.011)
^{105}Ag	0.28(0.08)	0.16(0.05)	0.08(0.02)
^{102m}Rh	–	0.030(0.005)	0.021(0.005)
^{103}Ru	0.013(0.004)	0.011(0.003)	0.016(0.002)
^{95m}Tc	0.045(0.009)	0.028(0.005)	0.013(0.003)
^{95}Nb	0.058(0.006)	0.041(0.007)	0.044(0.008)
^{91m}Nb	0.09(0.03)	0.125(0.025)	0.11(0.03)

Table 2 (continuation)

Isotope	^{179}Hf + 139 MeV p	^{179}Hf + 112 MeV p	^{179}Hf + 90 MeV p
^{95}Zr	0.007(0.002)	0.009(0.003)	0.012(0.002)
^{88}Zr	0.076(0.014)	0.095(0.015)	0.134(0.015)
^{88}Y	0.036(0.012)	0.016(0.005)	0.025(0.008)
^{87}Y	0.014(0.03)	0.125(0.038)	0.17(0.03)
^{85}Sr	–	0.021(0.005)	–
^{82}Sr	0.088(0.019)	0.11(0.02)	0.104(0.025)
^{84}Rb	0.036(0.007)	0.018(0.004)	0.08(0.02)
^{83}Rb	0.049(0.010)	0.053(0.008)	0.039(0.012)
^{72}Se	0.060(0.010)	0.061(0.013)	0.09(0.02)
^{74}As	0.028(0.006)	0.029(0.006)	0.028(0.009)
^{65}Zn	0.044(0.010)	0.023(0.006)	0.021(0.004)
^{59}Fe	0.036(0.006)	0.026(0.004)	0.032(0.003)
^{54}Mn	–	0.016(0.05)	0.011(0.04)

2. DISCUSSION

The isomers are produced with the ^{179}Hf target after exclusive emission of a few nucleons, the reactions of ^{179}Hf(p, p')^{179m2}Hf, ^{179}Hf(p, $p'n$)^{178m2}Hf and ^{179}Hf (p, $2pn$)^{177m}Lu are only effective. For $^{\rm nat}$Hf, the set of possible reactions is wider since the content of stable $^{177-180}$Hf isotopes in the target, but let discuss now the ^{179}Hf case. Emission of (1 to 3) nucleons is very special mode among other spallation channels, and its properties must significantly deviate from the more regular case of emission of (6–10) nucleons. Remind that the latter reactions are productive for isomers of interest with W targets [2]. For Hf spallation, one may expect significant gain in their cross sections at low energies near 100 MeV, while the isomer-to-ground state ratio can be decreased. It would be impossible to predict in semi-quantitative estimations the final enhancement or reduction factor for isomers as compared to the Ta and W targets. Thus, measured cross sections are the only informative at the case of (p + Hf) reactions.

Present results show that the ^{179}Hf target provides the best cross sections for the ^{177m}Lu isomer production as compared to other targets [1, 2]. For ^{179m2}Hf, the best is still ^{186}W target, but at higher proton energies. In general, the Hf targets provide more abundant production of the Hf and Lu high-spin isomers at low proton energies, near 100 MeV. The advantages of enriched ^{179}Hf target, as compared to the ^{nat}Hf for isomer production are clearly manifested. The yields of ^{177m}Lu and ^{179m2}Hf in the ^{179}Hf target are higher, but not much. The best enhancement by a factor of about 1.8 can be reached at optimum proton energy.

For ^{178m2}Hf, in accordance with expectations, the production cross section is increased significantly by a factor of about 3, as compared to the ^{nat}Hf target at all energies of protons. However, the level of cross sections about 1–1.5 mb is not reached, only near 0.5–0.7 mb at the lowest energies. Still a positive gain due to the enriched ^{179}Hf application is evident both in increased cross section for the accumulation of the isomers, type of ^{178m}Hf, and in a possibility to reduce the energy of protons down to 100–150 MeV, as compared to 400–600 MeV recommended earlier for ^{186}W.

The isomer-to-ground state ratio, σ_m/σ_g, would be a physical parameter carrying a «finger print» of the reaction inherent essence. Independent yields of both m and g states have experimentally been measured for ^{174}Lu and ^{177}Lu nuclides. Respectively, the σ_m/σ_g and total cross-section values are deduced for them. These results supply also some basis for the estimation of total independent cross section $(\sigma_m + \sigma_g)$ for the ^{178}Hf and ^{179}Hf nuclides. The σ_m/σ_g parameter for the latter nuclides has been estimated using the measured σ_m and extrapolated $(\sigma_m + \sigma_g)$ values. The results are reduced in Table 3. As is expected, the σ_m/σ_g value at the case of ^{174}Lu nuclide appears to be by an order of magnitude higher than that for other isomers. This is due to the relatively low spin of ^{174m}Lu, $I^\pi = 6^-$. Thus, an influence of the angular-momentum deficit in the described reactions is clearly manifested, but not as drastically, as it could be supposed within some model expectations.

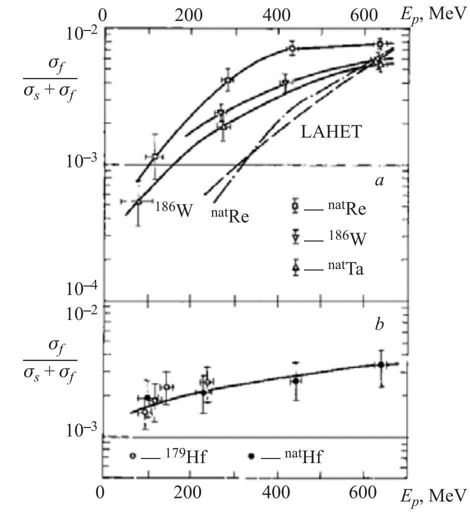

Fig. 5. Fission-to-spallation ratio as a function of the proton energy

Finally, the conclusion follows up that reactions with emission of 1, or 2, or 3 nucleons supply the individual channel cross section of about 10–15 mb and enough spin for population of high-spin isomers. Probably, such properties correspond to the peripheral type of interaction. Earlier, for spallation of Ta–Re targets, the mean spin of the residual nucleus was deduced [4] to be of about 8–10 h with wide distribution near the mean value. Surprisingly, similar properties may correspond also to the case of exclusive emission of a few nucleons in spallation of the Hf target.

Examples of mass-distribution are shown in Figs. 2–4. Similar results are available also for other proton energies indicated above. The cross section integrated over the fission-product peak provides a fission-to-spallation ratio, σ_f/σ_s, that numerically characterizes a fission probability for the spallation residue arising after fast stage of the reaction. Fission can be activated only within long time-scale, after non- and pre-equilibrium emission of nucleons. Thus, the results obtained in the present work might throw some light on the fission properties of excited nuclei at a range of Z^2/A near 30. This range until now was covered in major by the studies of heavy-ion induced reactions, that was influenced with specific manifestations of high angular momentum, $I > 50$ h, and of original mechanisms, like quasi-fission.

Table 3. Isomeric cross section and isomer-to-ground state ratio values determined in the spallation of ^{179}Hf and $^{\rm nat}$Hf* targets by protons

Target	Proton energy, MeV	Cross sections, σ_m (mb)			
		^{174m}Lu	^{177m}Lu	^{178m2}Hf	^{179m2}Hf
^{179}Hf	90	7.1	0.67	0.68	0.79
	242	9.9	0.90	0.39	0.44
$^{\rm nat}$Hf	100	3.6	0.78	0.22	0.49
	440	6.8	0.54	0.13	0.31
		σ_m/σ_g ratios			
^{179}Hf	90	0.74	0.06	0.042*	0.08*
	242	0.76	0.036	0.025*	0.06*
$^{\rm nat}$Hf	100	0.41	0.04	0.02*	0.04*
	440	0.52	0.04	0.02*	0.05*

* The values are estimated using extrapolated independent yields of the ground-state nuclei.

*Cross sections are reduced to one Hf nucleus in the multi-isotope target

Fission probability for the spallation residue is displayed in Fig. 5 as a function of proton energy according to experimental results of Refs. [1, 2] and of the present work. Relatively flat function in Fig. 5 contradicts the predictions of the advanced computer code: LAHET, as well as the simplified statistical model calculations. Additional analysis of this problem might be necessary.

SUMMARY

Yields and cross sections are measured for the fragmentation products formed in irradiations of the $^{\rm nat}$Hf and ^{179}Hf targets by protons at 90 to 650 MeV. The product distribution covers a wide range of mass numbers and includes the spallation and fission peaks. Moderately abundant production of the high-spin ^{177m}Lu, ^{178m2}Hf and ^{179m2}Hf isomers is observed despite they are formed in specific reactions of exclusive emission of only a few nucleons from 1 to 3. Reaction mechanisms and optimization of the isomeric nuclei production methods are discussed.

Acknowledgements. This experiment was supported by US Air Force EOARD under the Grant No. FA8655-04-1-3046. The authors are grateful to Yu. A. Medvedchuk for her assistance during the manuscript preparation.

REFERENCES

1. *Karamian S. A., Adam J., Filossofov D. V., Henzlova D., Henzl V., Kalinnikov V. G., Lebedev N. A., Novgorodov A. F., Collins C. B., Popescu I. I., Ur C. A.* Accumulation of the ^{178m2}Hf isomeric nuclei through spallation with intermediate-energy protons of tantalum and rhenium targets // Nucl. Instr. Meth. A. 2002. V. 489. P. 448.

2. *Karamian S. A., Adam J., Chaloun P., Filossofov D. V., Henzl V., Henzlova D., Kalinnikov V. G., Korolev N. A., Lebedev N. A., Novgorodov A. F., Collins C. B., Popescu I. I., Ur C. A.* Yield of radionuclides and isomers produced in the fragmentation of $^{\rm nat}$W and ^{186}W (97%) targets with protons at 630, 420 and 270 MeV // Nucl. Instr. Meth. A. 2004. V. 527. P. 609.

3. *Karamian S. A.* Comparative analysis of the ^{178m2}Hf yield at reactions with different projectiles // Phys. At. Nucl. 2005. V. 68. P. 1765

4. *Karamian S. A., Adam J.* Estimation of the spallation-residue angular momentum in $p+$ Ta, Re reactions // Czech. J. Phys. 2003. V. 53. P. B381.

Received on May 20, 2008.

Корректор *Т. Е. Попеко*

Подписано в печать 10.07.2008.
Формат 60 × 90/16. Бумага офсетная. Печать офсетная.
Усл. печ. л. 1,18. Уч.-изд. л. 1,67. Тираж 300 экз. Заказ № 56229.

Издательский отдел Объединенного института ядерных исследований
141980, г. Дубна, Московская обл., ул. Жолио-Кюри, 6.
E-mail: publish@jinr.ru
www.jinr.ru/publish/