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14. ABSTRACT

This report results from a contract tasking Induced Gamma Emission Foundation as follows: The contractor will investigate the production and accumulation of the nuclear isomer 178m2 Hafnium by spallation of Tungsten targets with high-energy protons. Targets made of the isotope 186 Tungsten will be designed using the LAHET computer code, and fabricated for experimental verification. The Hafnium yield will be assayed and compared to LAHET calculations.

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**Final Report
Item 0004**

to the
European Office of Aerospace Research and Development (EOARD)
describing progress in the research

*“New perspectives for the production and accumulation of $^{178m2}\text{Hf}$
isomers”*

Grant/ Cooperative Agreement Award No: **FA8655-02-1-3069**

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1. Introductory remarks

The experimental evidences on the possibility to trigger the release of the energy stored in the high-spin and high-energy isomeric state $^{178\text{m}2}\text{Hf}$ [1,2] has opened new perspectives and opportunities for the gamma-ray laser research. We know that the Hafnium isomers are the best choice as they act as real batteries do: one can charge them (exciting the isomeric states through some nuclear reaction mechanism) and the energy can be stored for a long time before use (the half life of the isomers is 31 years). The release of the energy can be switched on and off with the help of x-ray devices. Moreover, it was shown [3] that the energy of the x-rays needed to trigger the process is low; this fact has two major consequences: 1) the need of small devices at low cost to produce the triggering x-rays, and 2) huge energy gain (two orders of magnitude). All these features make the Hafnium isomers unique in the landscape of all the isomeric nuclei that can be produced through the nowadays experimental methods. To make the Hafnium isomers attractive for large scale applications one should answer some questions concerning the methods and easiness to produce them. Without a clear answer the whole problem will remain at an academic level. The existing inventory of these isomers is extremely low (**and it decays out continuously!**) and it resulted as a by-product of experiments oriented toward different goals. The IGE Foundation enrolled itself in the effort to answer these questions and during the last five years has brought many new results about the production of $^{178\text{m}2}\text{Hf}$ isomers through spallation with high energy protons. Clear conclusions could be drawn concerning the optimization of the isomer production for higher yields and low contamination background.

We initiated the study with targets in natural composition that did not imply high preparation costs. This was the case of the irradiation of natural Tantalum [4,6] and natural Rhenium [5,6] targets. These studies gave us the opportunity to refine the analysis methods and to improve significantly the computer code for the simulation of the spallation process. They showed without doubts that the production of $^{178\text{m}2}\text{Hf}$ isomers can be optimized for higher yields. Important know-how for the practical production process was developed. We concluded that we need to overcome two major difficulties in the isomer production process:

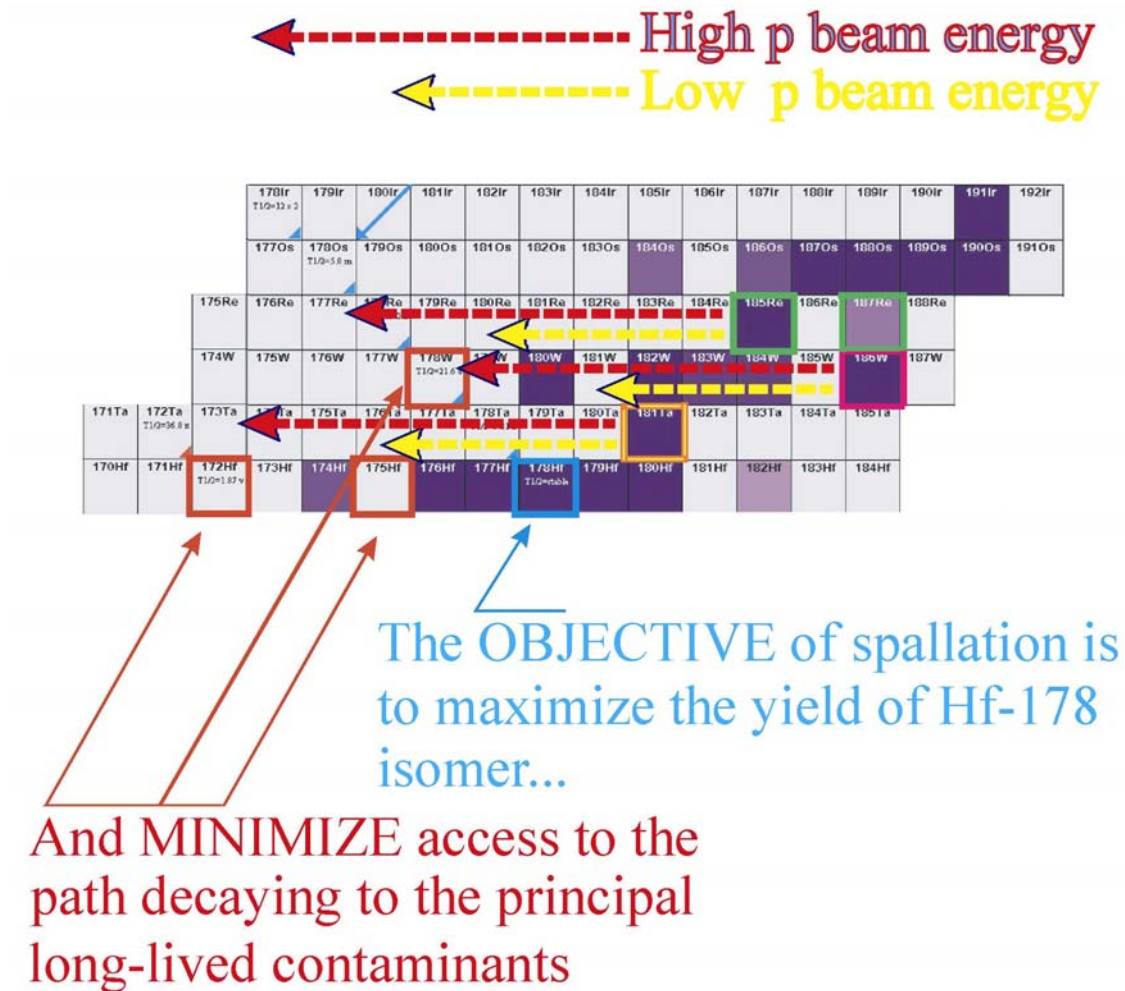


Figure 1. Schematic representation of the spallation process for different targets and incident proton energies. The mass regions populated are intuitively shown through arrows. The red squares indicate the main sources of contamination of the final Hafnium sample. The radionuclides ^{175}Hf ($T_{1/2} = 70$ d) and ^{172}Hf ($T_{1/2} = 1.87$ y) generate an intense gamma-radiation background for more than a decade leading to long cooling time of the samples. The ^{178}W through EC-decay ends in ground state ^{178}Hf reducing the isomer-to-ground state ratio.

- 1) the $^{178\text{m}2}\text{Hf}$ isomeric state is located at high excitation energy (~ 2.45 MeV) and high spin ($16\hbar$); as a consequence the total population cross section of ^{178}Hf nuclei is spread on many states with lower spin and excitation energy; the use of high energy beams will favor an increase of the population of states with high excitation energy (as it is the case of our isomer);

- 2) the ^{178}Hf nucleus is located near the beta stability line and the cross section for its population through spallation is low; the use of target nuclei as rich as possible in neutrons will populate better such nuclei.

The former studies of spallation with natural Tantalum [4,6] and natural Rhenium [5,6] targets showed us that we have to find the best compromise between the target material and the incident energy of the protons to favor production of the ^{178}Hf while the main sources of contaminants will be reduced. 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 further from the target mass. This is schematically shown in Fig. 1. 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. A summary of the spallation reactions studied by us is given in Table I.

Table I. Summary of the spallation reactions studied at the LNP Dubna synchrocyclotron for the production and accumulation of the $^{178m2}\text{Hf}$ isomers.

Reaction	Target Composition	Proton Beam Energy (MeV)	Reaction Channel Leading to ^{178}Hf
$p + \text{natTa}$	$^{180}\text{Ta} - 0.012\%$ $^{181}\text{Ta} - 99.988\%$	100, 200, 660	p2n 2p2n
$p + \text{natRe}$	$^{185}\text{Re} - 37.4\%$ $^{187}\text{Re} - 62.6\%$	150, 300, 450, 660	4p4n 4p6n
$p + \text{natW}$	$^{180}\text{W} - 0.13\%$ $^{182}\text{W} - 26.31\%$ $^{183}\text{W} - 14.28\%$ $^{184}\text{W} - 30.64\%$ $^{186}\text{W} - 28.64\%$	300, 450, 660	3p 3p2n 3p3n 3p4n 3p6n
$p + ^{186}\text{W}$	$^{186}\text{W} - 96.80\%$	300, 450, 660	3p6n

These were the main ideas that encouraged us to start the present contract. A survey of the available isotopes in the neighborhood of ^{178}Hf has indicated ^{186}W as the best target candidate. Its natural abundance is 28.6% and we had to buy enriched material at the level of 96.8% in order to eliminate the contribution from the other lighter W isotopes to

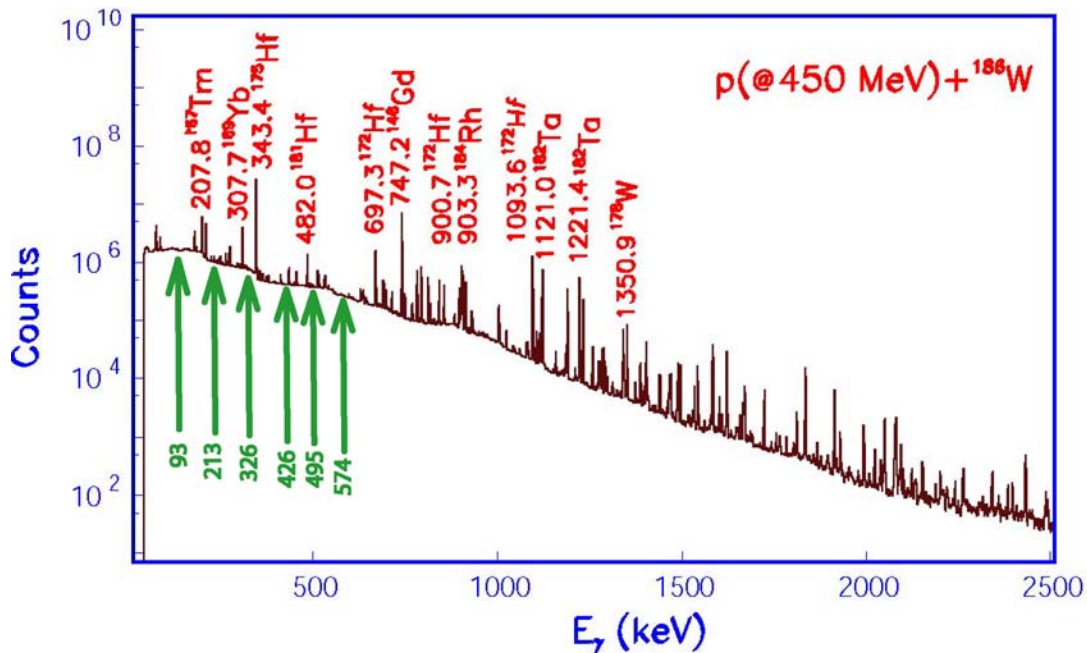
the final results. A preliminary study was performed from the analysis of some natural Tungsten samples irradiated in Dubna a couple of years ago. The results were affected by the complete decay out of the radionuclides with lifetimes shorter than 70 days but they allowed us to calibrate the Monte Carlo calculations and to get realistic estimates for the irradiation of the enriched Tungsten targets.

2. Experimental details

The enriched isotope was delivered in form of metallic pieces with thickness of about 1.5 mm that could be used without any preliminary preparation. Three similar targets were built. Each metallic foil of enriched isotope was placed on a 50 μm thickness ^{nat}W holder and fixed together on an Al backing. The Al backing was cooled in order to remove the heat released by the stopping of the beam in the target. The irradiation of the samples was performed at the internal beam of the synchrocyclotron of DLNP, JINR Dubna. 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. A direct comparison of the irradiation of the two materials could be performed at no supplementary cost. The irradiation of the ^{nat}W provided the missing information on the radionuclides that decayed out in the samples analyzed for the Second Item of the present Contract and allowed for a more accurate comparison between the irradiation of natural and enriched Tungsten targets.

After irradiation, the samples were very hot mainly due to short lived activity and a ‘cooling’ period of one month was needed before they could be safely handled. The gamma-ray activity of the samples was measured before and after cooling in order to estimate the short lived radionuclides production yield.



Acquired: 5/26/2003 9:30:17 AM Real Time: 84794.70 s Live Time: 71257.24 s.

Detector: #2 HPGGe CANBERRA

Channels: 8192

Figure 2. Sample gamma-ray spectrum recorded with a HP Ge detector after irradiation of the ^{186}W sample at 450 MeV proton beam energy. The count rate scale is logarithmic. The strongest channels are marked with red and with the energy of the lowest lying gamma-ray transitions. The green arrows indicate the places in the spectrum where we have to identify the lines belonging to the spontaneous decay of the $^{178\text{m}2}\text{Hf}$ isomer.

The gamma-ray decay of the $^{178\text{m}2}\text{Hf}$ isomer is weak compared to the activity of other radionuclides. This is illustrated in Fig. 1 where the green arrows indicate where we have to identify the strongest gamma-ray peaks following the decay of the $^{178\text{m}2}\text{Hf}$ isomer. It is obvious that to get a good statistical accuracy for the yield of $^{178\text{m}2}\text{Hf}$ we need to reduce the radiation background. This was achieved through chemical isolation of the Hafnium fraction. The chemical separation consisted of the following operations:

1. Dissolution of the W samples in concentrated hydrofluoric acid with addition of HNO_3 ;

2. Isolation of the Hf and W fractions from the bulk matter and from major part of other radionuclides (mostly rare-earth metals);
3. Fine rectification of the Hf fraction from the remaining W substances.
4. Final purification of the Hf fraction from the Lu accumulated as a daughter of Hf radionuclides decay.
5. Isolation of the individual fractions of other elements.

Permanent monitoring of the γ -ray activity controlled the transmission of different elements through the chemical separation processes.

The gamma-ray activity was recorded with a 20% HP Ge detector. The signals from the detector were processed with high quality electronic modules that permitted us to record good quality spectra at high counting rates (the energy resolution of the gamma peaks at 1332 keV was of 1.8 keV). The dead-time of the spectroscopic chain was kept within moderate limits (below 20%) even at counting rates as high as 20 kCounts/s. The geometry of the measurement (distance and absorber in front of the detector) was optimized for each sample. Energy and efficiency calibration were performed with standard gamma-ray sources and with well known internal gamma-ray lines (the intense γ -lines of ^{172}Hf and ^{175}Hf). The efficiency of the detector as a function of the γ -ray energy was measured separately for each source-detector distance and absorber thickness. The accuracy on the efficiency values was better than 5%.

3. Data analysis and calculations

The yield of a reaction product per one bombarding proton, is defined as:

$$Y = \int_{E_{\min}}^{E_{\max}} \sigma(E) \left(\frac{dE}{dx} \right)^{-1} dE, \quad (1)$$

where E_{\max} and E_{\min} define the proton energy range in the target and dE/dx is the energy dependent stopping power of protons in the target material expressed in $\text{MeV/at}\cdot\text{cm}^{-2}$ if E

is in MeV and σ in cm^2 . The mean cross-section is connected with the yield through the expression:

$$\bar{\sigma} = Y \left[\int_{E_{\min}}^{E_{\max}} \left(\frac{dE}{dx} \right)^{-1} dE \right]^{-1}. \quad (2)$$

From the analysis of the recorded spectra we measured the yields for about 70 radionuclides produced following the fragmentation of the Tungsten target. The yield of each nuclide was determined based on the measured intensity of its characteristic γ -ray lines, corrected for the detector efficiency and the individual spectroscopical properties of nuclides [7].

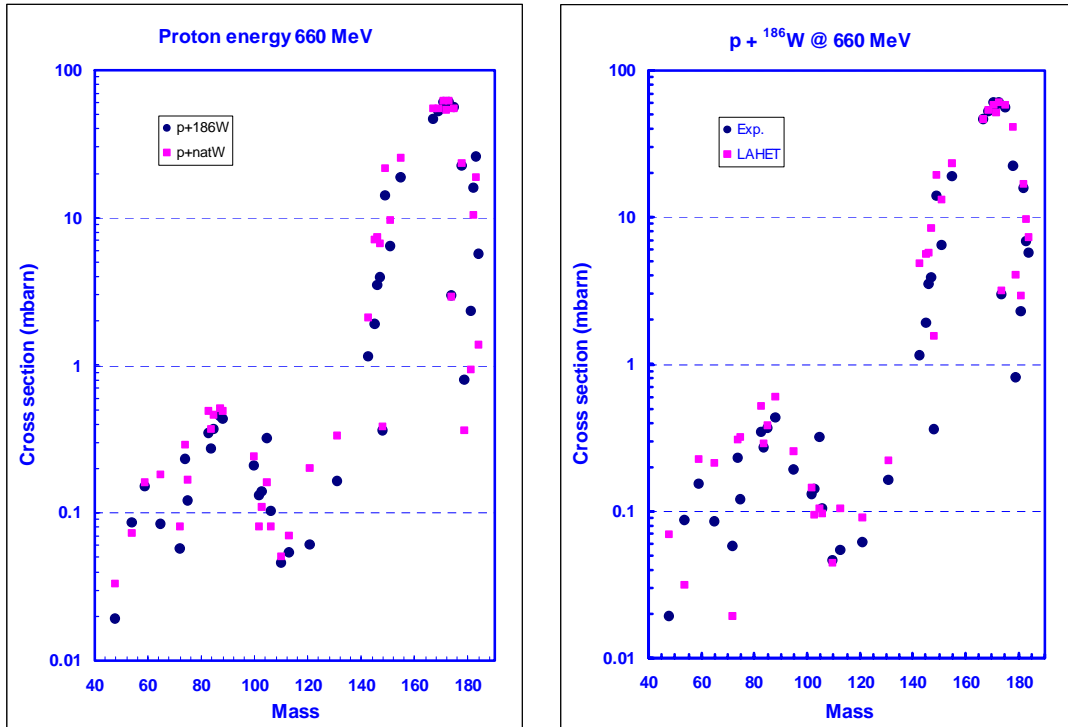


Figure 3. Mass distribution following the Tungsten fragmentation after irradiation with 660 MeV protons. The two mass distributions correspond to spallation (centered around mass 175-178) and to fission (centered around mass 80-90).

The statistical accuracy of the measured yield values is rather good for the products with high yields while in the case with low yield values statistical errors go up to 30%. The

final error is affected by systematic errors as the errors on the absolute calibration of the yields, the inaccuracy of the measured efficiency of detectors and the errors on the tabulated standards like the quantum yields of characteristic γ -ray lines belonging to the individual nuclides. Finally, the errors vary from 10% in the best cases to 40% for the cases of low intensity γ -ray lines in the spectra.

The results of the measurements are summarized in the Tables II – IV for proton energies 660 MeV, 450 MeV and 300 MeV, respectively.

Calculations were performed with the Monte-Carlo simulation LAHET code. In the present series of simulations, one nucleus produced in fragmentation corresponds to the cross-section of 3 μ barn, and this defines the statistical accuracy of the Monte-Carlo procedure. The calculated values are reported together with the experimental ones in Tables II – IV.

Since we irradiated also the natural Tungsten holder we made the analysis of these samples too and the results are reported together with those for the enriched target so that we have a direct comparison of the values for both targets when irradiated under identical conditions. This comparison is more accurate than using the results from the analysis of the old natural Tungsten reported at the Second Item of the present Contract. Moreover, we could make the comparison for all three irradiation energies.

The comparison of the measured and calculated cross-sections shows a general good agreement but, as already discussed in the previous reports, discrepancies appear in some cases. The LAHET code calculations underestimate the fission product yields and the discrepancy becomes more accentuated at lower projectile energies (see Fig. 3 right panel for the case of 660 MeV proton beam).

Table II. Measured and calculated (LAHET code) cross-section values (in mbarn) for the fragmentation products at 660 MeV proton energy with the ^{nat}W and ^{186}W (96.8%) targets.

Isotope	Type of yield	^{186}W target		^{nat}W target	
		experiment	calculation	experiment	calculation
1	2	3	4	5	6
^{184}Re	Indep.	4.500	} 7.300	1.100	} 2.800
^{184m}Re	Indep.	1.200		0.260	
^{183}Re	EC cum.	6.800	9.600	2.700	5.200
^{183}Ta	β^- cum.	19.000	-	16.000	-
^{182}Ta	Indep.	15.800	16.600	10.400	17.500
^{181}Hf	β^- cum.	2.300	2.900	0.930	1.880
$^{179m2}Hf$	Indep.	0.800	4.000	0.360	2.860
^{178}W	EC cum.	21.800	34.800	23.000	38.100
$^{178m2}Hf$	Indep.	0.480	5.700	0.180	4.300
^{177m}Lu	Indep.	0.260	1.160	0.130	0.750
^{175}Hf	EC cum.	55.600	57.400	55.000	56.900
^{174}Lu	Indep.	1.400	} 3.160	1.300	} 2.110
^{174m}Lu	Indep.	1.5500		1.600	
^{173}Lu	EC cum.	60.000	60.500	61.000	58.900
^{172}Hf	EC cum.	57.400	50.800	53.500	53.800
^{171}Lu	EC cum.	60.000	58.200	61.000	58.500
^{169}Yb	EC cum.	52.000	53.500	55.000	55.000
^{168}Tm	Indep.	1.000	0.850	0.730	0.420
^{167}Tm	EC cum.	46.500	46.000	55.000	48.500
^{156}Tb	Indep.	0.270	0.560	0.230	0.450
^{156}Eu	β^- cum.	0.120	0.003	0.100	-
^{155}Tb	EC cum.	18.700	23.100	25.000	26.200
^{151}Gd	EC cum.	6.400	13.000	9.600	15.900
^{149}Gd	EC cum.	6.900	9.100	10.500	12.400
^{149}Eu	EC cum.	7.000	10.200	11.000	13.400
^{148}Eu	Indep.	0.340	1.520	0.360	1.270
^{148m}Pm	Indep.	0.020	0.016	0.030	0.009
^{147}Eu	EC cum.	3.900	8.300	6.700	11.600
^{146}Gd	EC cum.	3.500	5.700	7.300	8.600
^{145}Eu	EC cum.	1.900	5.600	7.000	7.900
^{144}Pm	Indep.	0.078	0.560	0.150	0.520
^{143}Pm	EC cum.	1.130	4.800	2.100	6.300
^{140}Ba	β^- cum.	0.002	-	0.003	-
^{131}Ba	EC cum.	0.162	0.220	0.330	0.440
^{126}I	Indep.	0.006	-	0.007	-
^{121}Te	EC cum.	0.061	0.090	0.200	0.068
^{113}Sn	EC cum.	0.054	0.103	0.070	0.111

1	2	3	4	5	6
^{110m} Ag	Indep.	0.046	0.044	0.050	0.025
^{106m} Ag	Indep.	0.103	0.096	0.080	0.093
¹⁰⁵ Ag	EC cum.	0.316	0.103	0.160	0.133
¹⁰³ Ru	β- cum.	0.140	0.094	0.110	0.065
¹⁰² Rh	Indep.	0.130	0.143	0.080	0.164
¹⁰⁰ Pd	β+ cum.	0.056	0.031	-	0.052
^{95m} Tc	Indep.	0.019	-	0.030	-
⁹⁵ Nb	Indep.	0.140	0.197	0.170	0.130
⁹⁵ Zr	β- cum.	0.050	0.059	0.040	0.037
^{91m} Nb	Indep.	0.021	0.225	0.047	0.228
⁸⁸ Zr	EC cum.	0.190	0.324	0.180	0.270
⁸⁸ Y	Indep.	0.240	0.275	0.310	0.220
⁸⁷ Y	β+ cum.	0.450	-	0.510	-
⁸⁵ Sr	β+ cum.	0.366	0.380	0.460	0.350
⁸⁴ Rb	Indep.	0.272	0.284	0.370	0.204
⁸³ Rb	β+ cum.	0.345	0.515	0.490	0.410
⁷⁵ Se	β+ cum.	0.120	0.318	0.166	0.240
⁷⁴ As	Indep.	0.230	0.306	0.290	0.250
⁷² Se	β+ cum.	0.057	0.019	0.080	0.030
⁶⁵ Zn	β+ cum.	0.084	0.209	0.180	0.234
⁵⁹ Fe	β- cum.	0.152	0.224	0.160	0.234
⁵⁶ Co	EC cum.	0.007	0.0156	0.019	0.022
⁵⁴ Mn	Indep.	0.086	0.0312	0.073	0.160
⁵² Mn	β+ cum.	0.007	0.047	0.022	0.034
⁴⁸ V	EC cum.	0.019	0.069	0.033	0.049
²² Na	β+ cum.	0.087	-	0.122	-
⁷ Be	Indep.	0.330	-	0.560	-

Table III. The same as Table II but at 450 MeV proton energy.

Isotope	Type of yield	¹⁸⁶ W target		^{nat} W target	
		experiment	calculation	experiment	calculation
1	2	3	4	5	6
¹⁸⁴ Re	Indep.	6.100	} 11.440	1.930	} 4.550
^{184m} Re	Indep.	1.500		0.500	
¹⁸³ Re	EC cum.	10.000	14.400	3.500	8.000
¹⁸³ Ta	β ⁻ cum.	23.000	-	14.000	-
¹⁸² Re	EC cum.	8.000	7.850	10.000	5.500
¹⁸² Ta	Indep.	18.200	16.000	12.600	15.800
¹⁸¹ Hf	β ⁻ cum.	2.500	2.200	1.080	1.170
^{179m2} Hf	Indep.	0.870	3.400	0.390	2.370
¹⁷⁸ W	EC cum.	32.600	52.000	36.300	53.300
^{178m2} Hf	Indep.	0.520	4.900	0.210	3.400
^{177m} Lu	Indep.	0.260	0.870	0.110	0.380
¹⁷⁵ Hf	EC cum.	78.000	78.300	79.500	77.500
¹⁷⁴ Lu	Indep.	1.300	} 2.700	1.200	} 1.370
^{174m} Lu	Indep.	1.600		1.400	
¹⁷³ Lu	EC cum.	76.000	76.500	78.000	77.300
¹⁷² Hf	EC cum.	69.000	63.900	78.000	70.800
¹⁷¹ Lu	EC cum.	77.000	66.900	85.000	73.00
¹⁶⁹ Yb	EC cum.	62.000	55.300	69.000	60.900
¹⁶⁸ Tm	Indep.	0.900	0.600	0.600	0.230
¹⁶⁷ Tm	EC cum.	40.000	42.900	57.600	49.000
¹⁵⁶ Tb	Indep.	0.330	0.162	0.630	0.170
¹⁵⁶ Eu	β ⁻ cum	0.100	-	0.080	-
¹⁵⁵ Tb	EC cum.	10.800	7.160	12.400	10.500
¹⁵¹ Gd	EC cum.	1.840	2.220	3.750	3.830
¹⁴⁹ Gd	EC cum.	1.580	1.270	3.070	2.420
¹⁴⁹ Eu	EC cum.	1.590	1.420	3.170	2.600
¹⁴⁸ Eu	Indep.	0.130	0.206	0.150	0.220
^{148m} Pm	Indep.	0.027	0.006	0.025	0.009
¹⁴⁷ Eu	EC cum.	0.710	0.850	1.700	1.760
¹⁴⁶ Gd	EC cum.	0.610	0.440	1.600	1.180
¹⁴⁵ Eu	EC cum.	0.290	0.360	0.630	0.880
¹⁴⁴ Pm	Indep.	-	0.037	-	0.071
¹⁴³ Pm	EC cum.	0.030	0.247	0.120	0.450
¹⁴⁰ Ba	β ⁻ cum.	0.0009	-	0.001	-
¹³¹ Ba	EC cum.	-	0.047	-	0.031
¹²¹ Te	EC cum.	-	0.053	-	0.037
¹¹³ Sn	EC cum.	0.027	0.040	0.038	0.037
^{110m} Ag	Indep.	0.039	0.025	0.050	0.017
^{106m} Ag	Indep.	0.052	0.037	0.072	0.040

1	2	3	4	5	6
¹⁰⁵ Ag	EC cum.	0.150	0.041	0.206	0.052
¹⁰³ Ru	β ⁻ cum.	0.120	0.062	0.084	0.028
¹⁰² Rh	Indep.	0.160	0.056	0.089	0.059
¹⁰⁰ Pd	β ⁺ cum.	-	0.016	-	0.012
^{95m} Tc	Indep.	0.014	-	0.012	-
⁹⁵ Nb	Indep.	0.150	0.103	0.160	0.080
⁹⁵ Zr	β ⁻ cum.	0.063	0.040	0.050	0.027
^{91m} Nb	Indep.	0.024	0.069	0.037	0.080
⁸⁸ Zr	EC cum.	0.087	0.103	0.115	0.154
⁸⁸ Y	Indep.	0.194	0.084	0.258	0.090
⁸⁷ Y	β ⁺ cum.	0.330	-	-	-
⁸⁵ Sr	β ⁺ cum.	0.240	0.153	0.355	0.117
⁸⁴ Rb	Indep.	0.230	0.103	0.312	0.065
⁸³ Rb	β ⁺ cum.	0.250	0.190	0.360	0.151
⁷⁵ Se	β ⁺ cum.	0.080	0.112	0.130	0.059
⁷⁴ As	Indep.	0.170	0.078	0.245	0.108
⁷² Se	β ⁺ cum.	0.030	0.0125	0.050	0.012
⁶⁵ Zn	β ⁺ cum.	0.070	0.084	0.080	0.074
⁵⁹ Fe	β ⁻ cum.	0.120	0.109	0.141	0.077
⁵⁶ Co	EC cum.	0.011	0.015	0.015	0.0062
⁵⁴ Mn	Indep.	0.064	0.031	0.107	0.043
⁵² Mn	β ⁺ cum.	0.005	0.022	0.009	0.015
⁴⁸ V	EC cum.	0.007	0.0125	0.012	0.022
²² Na	β ⁺ cum.	0.086	-	0.102	-
⁷ Be	Indep.	0.290	-	0.350	-

Table IV. The same as Table II but at 300 MeV proton energy.

Isotope	Type of yield	¹⁸⁶ W target		^{nat} W target	
		experiment	calculation	experiment	calculation
1	2	3	4	5	6
¹⁸⁴ Re	Indep.	11.300	} 19.400	3.350	} 7.600
^{184m} Re	Indep.	2.600		0.800	
¹⁸³ Re	EC cum.	13.000	24.500	5.700	13.600
¹⁸³ Ta	β ⁻ cum.	25.000	-	14.000	-
¹⁸² Re	EC cum.	16.000	13.200	-	9.700
¹⁸² Ta	Indep.	20.000	14.400	11.800	12.500
¹⁸¹ Hf	β ⁻ cum.	1.900	1.400	0.820	0.670
^{179m2} Hf	Indep.	0.720	2.040	0.300	2.370
¹⁷⁸ W	EC cum.	56.800	78.900	54.600	79.800
^{178m2} Hf	Indep.	0.380	3.000	0.170	2.200
^{177m} Lu	Indep.	0.160	0.440	0.080	0.190
¹⁷⁵ Hf	EC cum.	95.700	93.200	103.200	97.000
¹⁷⁴ Lu	Indep.	1.200	} 1.600	1.000	} 0.820
^{174m} Lu	Indep.	1.100		1.300	
¹⁷³ Lu	EC cum.	71.700	80.700	93.600	90.900
¹⁷² Hf	EC cum.	76.600	64.400	79.400	81.000
¹⁷¹ Lu	EC cum.	73.300	61.000	85.500	74.300
¹⁶⁹ Yb	EC cum.	38.000	42.100	57.500	54.000
¹⁶⁸ Tm	Indep.	0.320	0.3000	0.200	0.105
¹⁶⁷ Tm	EC cum.	18.000	26.000	36.400	37.300
¹⁵⁶ Tb	Indep.	0.130	0.030	0.180	0.027
¹⁵⁶ Eu	β ⁻ cum.	0.100	-	0.090	-
¹⁵⁵ Tb	EC cum.	5.500	0.320	8.200	0.980
¹⁵¹ Gd	EC cum.	1.100	0.017	0.950	0.140
¹⁴⁹ Gd	EC cum.	-	0.012	-	0.049
¹⁴⁹ Eu	EC cum.	-	0.015	-	0.064
¹⁴⁸ Eu	Indep.	0.033	-	0.022	0.003
^{148m} Pm	Indep.	0.012	-	-	-
¹⁴⁷ Eu	EC cum.	-	0.012	-	0.015
¹⁴⁶ Gd	EC cum.	-	-	0.021	0.023
¹⁴⁵ Eu	EC cum.	-	0.0036	-	0.014
¹⁴⁴ Pm	Indep.	-	0.0030	-	0.0062
¹⁴³ Pm	EC cum.	-	-	-	0.009
¹³¹ Ba	EC cum.	-	0.012	-	0.0062
¹²¹ Te	EC cum.	-	0.0062	-	0.022
¹¹³ Sn	EC cum.	0.0074	0.0062	0.019	0.025
^{110m} Ag	Indep.	0.016	0.003	0.014	0.009
^{106m} Ag	Indep.	0.023	0.016	0.023	0.019
¹⁰⁵ Ag	EC cum.	0.120	0.012	0.190	0.028

1	2	3	4	5	6
¹⁰³ Ru	β ⁻ cum.	0.076	0.0019	0.066	0.012
¹⁰² Rh	Indep.	0.123	0.016	0.110	0.012
^{95m} Tc	Indep.	0.02	-	0.020	-
⁹⁵ Nb	Indep.	0.101	0.031	0.110	0.030
⁹⁵ Zr	β ⁻ cum.	0.058	0.0094	0.060	0.0054
^{91m} Nb	Indep.	0.04	0.0062	-	0.037
⁸⁸ Zr	EC cum.	0.05	0.019	0.045	0.019
⁸⁸ Y	Indep.	0.085	0.022	0.130	0.030
⁸⁷ Y	β ⁺ cum.	0.2	-	0.240	-
⁸⁵ Sr	β ⁺ cum.	0.18	0.022	0.200	0.031
⁸⁴ Rb	Indep.	0.127	0.016	0.180	0.012
⁸³ Rb	β ⁺ cum.	0.10	0.041	0.160	0.065
⁷⁵ Se	β ⁺ cum.	0.028	0.016	0.060	0.016
⁷⁴ As	Indep.	0.059	0.022	0.110	0.009
⁷² Se	β ⁺ cum.	0.017	-	0.028	0.003
⁶⁵ Zn	β ⁺ cum.	0.019	0.019	0.027	0.0123
⁵⁹ Fe	β ⁻ cum.	0.052	0.044	0.069	0.028
⁵⁶ Co	EC cum.	0.01	-	0.015	0.006
⁵⁴ Mn	Indep.	0.04	0.006	0.044	0.022
⁵² Mn	β ⁺ cum.	0.006	0.006	0.006	0.006
⁴⁸ V	EC cum.	0.008	-	0.008	-
²² Na	β ⁺ cum.	0.06	-	0.050	-
⁷ Be	Indep.	0.17	-	0.150	-

4. Discussion

The Hf and Lu high-spin isomers of interest for our study are located in the mass region 175-180. From the Tables II-IV and Fig. 3 one can notice that the mass distribution of the products in this mass range is more or less flat. Moreover, the measured and calculated values are in a good agreement. This gave us confidence to use the calculated values for the yields of stable nuclei that cannot be estimated with the activation technique. Such an assumption becomes important for the estimation of the isomer-to-ground state ratio for ^{178}Hf and ^{179}Hf since we can use the measured cross-section for the isomer and the realistic estimate for the ground state cross-sections. In Table V we report the results obtained for ^{178}Hf at irradiation with 660 MeV protons for all the cases we studied until now.

Table V. Cross-sections (in mb) for independent formation of the ^{178}Hf nuclei (σ_{indep}) and of the $^{178\text{m}2}\text{Hf}$ isomers (σ_{m}) after spallation of different targets with protons of 660 MeV. The measured σ_{m} values are combined with the LAHET results for σ_{indep} and the isomer-to-ground state ratios ($\sigma_{\text{m}}/\sigma_{\text{g}}$) are deduced.

TARGET	^{181}TA	$^{\text{NAT}}\text{W}$	^{186}W	$^{\text{NAT}}\text{RE}$
σ_{indep}	15	4.3	5.7	1.0
$\sigma_{\text{m}}/\sigma_{\text{g}}$	0.021	0.044	0.092	0.15
σ_{m}	0.31	0.18	0.48	0.13

If we compare these results with the estimates based entirely on calculations for the case of the enriched target of ^{186}W listed in Table I of the Report on the Third Item to the present Contract we can get an idea of the accuracy of our calculations.

As expected, the use of the enriched ^{186}W resulted in higher cross sections for the $^{178\text{m}2}\text{Hf}$ isomer by a factor 2÷3 than in the case of natural Tungsten. At 450 MeV the increase is even higher than at 660 MeV and also the isomer-to-ground state ratio is better (see Table VI). When compared with the cross sections measured for other target materials (Table V) it can be seen that the use of ^{186}W leads to the highest production yield of Hafnium isomers. The increase of the $^{178\text{m}2}\text{Hf}$ isomer population cross section from natural

Tantalum to the enriched Tungsten is not spectacular (being less than a factor 2) but the isomer-to-ground state ratio improves significantly (almost five times). The major gain is that the cross section for ^{172}Lu remains at the same level and this has strong consequences on the cooling time of the irradiated samples.

Table VI. Cross-sections (in mb) for independent formation of the ^{178}Hf nuclei (σ_{indep}) and of the $^{178\text{m}2}\text{Hf}$ isomers (σ_{m}) after spallation of the enriched ^{186}W targets with protons of different energies. The measured σ_{m} values are combined with the LAHET results for σ_{indep} and the isomer-to-ground state ratios ($\sigma_{\text{m}}/\sigma_{\text{g}}$) are deduced.

ENERGY(M EV)	300		450		660	
	^{186}W	natW	^{186}W	natW	^{186}W	natW
σ_{indep}	3.00	2.20	4.90	3.40	5.7	4.3
$\sigma_{\text{m}}/\sigma_{\text{g}}$	0.145	0.084	0.119	0.066	0.092	0.044
σ_{m}	0.38	0.17	0.52	0.21	0.48	0.18

When comparing different options for the isomer production we have to take in account also the background production. Contaminant radionuclides will have strong consequences on the actual production process as they determine the cooling time of the irradiated samples, the thickness and irradiation time of the samples to avoid their accumulation (the presence of undesired material in the samples increases the absorption of the triggering radiation). In Tables VII and VIII we compare the cross sections for producing the isomer with the contaminant production.

Table VII. Cross-section (in mb) for the formation of high-spin isomers and background-activity isotopes after spallation of different targets with 660 MeV protons.

Nuclide	Target			
	natTa	natW	^{186}W	natRe
$^{179\text{m}2}\text{Hf}$	0.52	0.36	0.80	0.12
$^{178\text{m}2}\text{Hf}$	0.31	0.18	0.48	0.13
$^{177\text{m}}\text{Lu}$	0.15	0.13	0.26	0.04
^{178}W	5.9	23	21.8	36
^{175}Hf	56	55	55.6	59

^{172}Hf	47	53.5	57.4	55
^{173}Lu	61	61	60	61

Table VIII. Cross-section (in mb) for the formation of high-spin isomers and background-activity isotopes after spallation of enriched ^{186}W targets at different beam energies.

Nuclide	Energy		
	300	450	660
$^{179\text{m}2}\text{Hf}$	0.72	0.87	0.80
$^{178\text{m}2}\text{Hf}$	0.38	0.52	0.48
$^{177\text{m}}\text{Lu}$	0.16	0.26	0.26
^{178}W	56.8	32.6	21.8
^{175}Hf	95.7	78.0	55.6
^{172}Hf	76.7	69.0	57.4
^{173}Lu	71.7	76.0	60.0

At 660 MeV the production of contaminant radionuclides is more or less constant for all the targets. The only exception is ^{178}W which has a much lower cross-section in the case of natural Tantalum irradiation. However, production of ^{178}W is not a problem since it can be eliminated from the target through chemical separation and one can choose to change often the targets during the irradiation process to avoid its accumulation and decay to ^{178}Hf ground state. From Table VIII we see that the contaminant cross-sections decrease at higher energy.

It results that for the irradiation of enriched ^{178}W targets higher energies are more convenient as they lead to higher cross-sections for the isomer, good isomer-to-ground state ratio and lower contaminant radionuclides production.

5. Conclusions

We may conclude that we have accomplished the goal of the present Contract. We reported on the successful irradiation of three targets of enriched ^{186}W at the synchrocyclotron of the LNP, JINR Dubna with protons of 300 MeV, 450 MeV and 660

MeV, respectively. Moreover, the particular design of the targets allowed us to irradiate samples of natural W together with the enriched material and we provided supplementary results from the analysis of these samples. In this way we could compare the yields for the isomers and contaminant radionuclides when irradiating the natural and enriched Tungsten targets under exactly the same conditions.

The high sensitivity of the analysis method developed by us allowed to make accurate measurements even with small quantities of irradiated material. The use of small samples is more convenient also from the point of view of the chemical separation. We identified about 70 radionuclides, the lowest cross-section measured with good accuracy being at the level of one microbarn.

As predicted by the LAHET Monte Carlo calculations the use of enriched material leads to an increase of the isomer cross-section by a factor 2÷3 as compared to the natural Tungsten while the contaminant background is kept at the same level. The experimental results offered a nice confirmation of the accuracy of the LAHET calculations performed by us and we could use them to make estimates of the isomer-to-ground state ratio.

The increase of the $^{178m2}\text{Hf}$ isomer cross-section by a factor almost two as compared to the $p + ^{\text{nat}}\text{Ta}$ irradiation while the production of the ^{172}Lu is kept at the same level has an important consequence on the cooling time of the samples. After the irradiation of the natural Tantalum samples [4] we reported a reduction of the cooling time from 20 years in the case of the LAMPF irradiation with 800 MeV protons [8] to about 6 years in our case. Now we report on a further reduction of the cooling down to about 4 years for the production of the same quantity of $^{178m2}\text{Hf}$ isomer.

Comparing the new results with the ones from the previous irradiation of natural Tantalum and Rhenium [4,5,6] we can conclude that the use of ^{186}W results in the highest isomer yield and best isomer-to-ground state ratio.

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