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PRODUCTION, CHEMICAL AND ISOTOPIC SEPARATION OF THE LONG-LIVED ISOMER ^{178m_2}Hf ($T_{1/2}$ = 31 YEARS)

Yu.Ts.Oganessian, S.A.Karamian, Y.P.Gangrski, B.Gorski, B.N.Markov, Z.Scheglowski, Ch.Briançon[1], D.Ledu[1], R.Meunier[1], M.Hussonnois[2], O.Constantinescu[2] *, M.I.Subbotin[3]

The ^{178}Hf, with its long-lived ($T_{1/2}$ =31 y) high-spin isomeric state $I^{\pi} = 16^{+}$ is a challenge for new and exotic nuclear physics studies. The first experiments are described, performed in order to produce a reasonable microweight quantity of this hafnium isomer with an isomeric to ground-state ratio as high as possible (here 5%). The reaction ^{176}Yb (^{4}He, 2n) using enriched target has been studied by measuring the excitation functions and the isomeric to ground-state ratio. About $3 \cdot 10^{14}$ isomeric atoms have been produced up to now in irradiations with high-intensity beams (~100 μA) at the U-200 cyclotron in Dubna. Chemical separation methods could be checked over using about 10^{13} atoms of the isomer. Isotopic separation experiments have been performed in Orsay and preliminary results are given for the separation efficiency.

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

Получение, химическое и изотопное выделение долгоживущего изомера ^{178m_2}Hf ($T_{1/2}$ = 31 год)

Ю.Ц.Оганесян и др.

^{178}Hf и его долгоживущий изомер ($T_{1/2}$ = 31 год) с высоким спином $I^{\pi} = 16^{+}$ могут быть предметом исследования новых и экзотических ядерных состояний. В данной статье описаны первые эксперименты по получению микровесовых количеств данного изомера при достаточно высоком изомерном отношении (около 5%). Реакция ^{176}Yb (^{4}He, 2n) с использованием обогащенной

[1] CSNSM-Orsay, IN2P3-CNRS, Bat.104-108, 91405 Orsay, France

[2] IPN-Orsay, Bat.100, 91406 Orsay, France

[3] Inst. of Nuclear Physics, Akad.Nauk.Kaz.SSR, Alma-Ata, USSR

*Permanent adress: Institute of Atomic Physics, Bucharest, Romania

мишени изучена посредством измерения функций возбуждения и изомерного отношения. Около $3 \cdot 10^{14}$ ядер изомера получено на сегодня в длительных облучениях высокоинтенсивным (~100 мкА) пучком на циклотроне У-200 в Дубне. Методы химического выделения опробованы при использовании около 10^{13} атомов изомера. Эксперименты по изотопной сепарации выполнены в Орсэ и получены предварительные результаты по эффективности сепарации.

Работа выполнена в Лаборатории ядерных реакций ОИЯИ.

1. Introduction

Some years ago the idea was brought out to study the nuclear structure of a high-K isomer ^{178m_2}Hf as well as the new physics which could be undertaken in using this isomer as a target. This isomer is indeed a unique case of an yrast trap with a high-spin $I^{\pi} = 16^+$ at the relatively low excitation energy of 2.447 MeV and especially with its long lifetime: $T_{1/2}$ = 31 years. This high value of $T_{1/2}$ can be explained by the decay properties of the 16^+ state to the 13^- and 12^- states of the $K^{\pi} = 8^-$ band, the $\Delta K = 8$ hindrance retarding even more the already slow E3 and M4 transitions.

A collaboration between JINR-Dubna (Laboratory of Nuclear Reactions) and Orsay (Centre de Spectrometrie Nucleaire et de Spectrometrie de Masse (CSNSM) and Institut de Physique Nucleaire (IPN)) has been established in order to produce this isomer, to perform the chemical extraction of the hafnium isotopes, to achieve the isotopic separation of the different hafnium isotopes and to provide targets with microweight quantities of the isomer suitable for the planned experiments.

The following references [1-5] related to the ^{178m_2}Hf production using different reactions may be quoted. Previously the highest isomeric to ground-state ratio of about 0.007 was obtained (ref.[2]) in the case of 540 MeV protons incident on a tantalum target. Khoo and Lovhoiden (ref.[4]) have measured the prompt and short delayed γ-radiation emitted in the reaction: 26 MeV ^{4}He projectiles incident on ^{176}Yb — and have found $\approx$ 0.01 feeding of the 14^- level which is close in energy to the 16^+ state. In ref.[5], a ^{178m_2}Hf/^{178g}Hf ratio of $\approx$ 0.005 was obtained using the same reaction with E_{α} = 27.5 MeV.

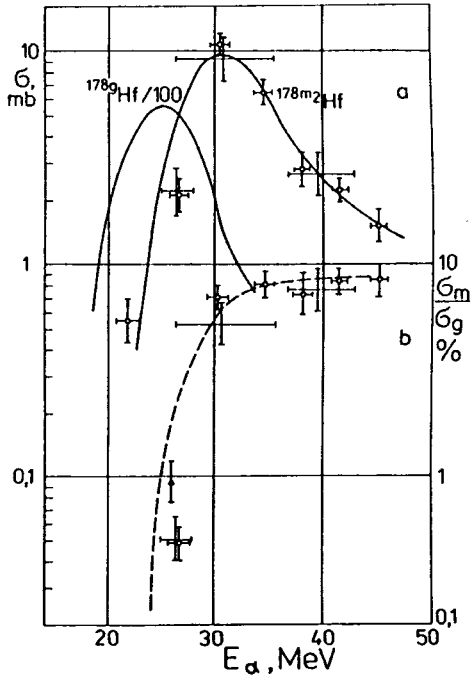
In this paper we shall describe the following experiments: the isomer production using the (^{4}He + ^{176}Yb) reaction, the chemical separation of hafnium from the other products of irradiation and finally the isotopic separation of the hafnium isotopes.

2. Production

A crucial point is to get the best isomeric ratio as well as the best absolute yield of ^{178m_2} Hf in optimizing the reaction, the energy of the chosen projectile and the thickness of the target.

As is well known from statistical theory of nuclear reactions, the yield of the high-spin isomeric state is growing with increasing angular momentum of the reaction residue. The latter is determined by the target spin, by the angular momentum I_{max} of the incident particle and by the angular momenta of the emitted particles. As is known (see, for example, ref. /6/), in compound nucleus reactions the relative yield of evaporation residues with high-spin grows drastically with the increasing projectile energy. Consequently the excitation function curve of the high-spin isomer is shifted up by some MeV to higher energies as compared to the ground-state one. As a result the enhancement of the ^{178m_2}Hf yield in the reaction ^{176}Yb(^{4}He, 2n) is expected in the region of $E_\alpha \geq 30$ MeV.

Several irradiations of thick enriched ^{176}Yb targets (96 % enrichment) were performed in Alma-Ata and Dubna in order to estimate the excitation functions and to study the optimal conditions for the production of ^{178}Hf isomeric nuclei.



Experimental data on the excitation function and the isomeric ($I^\pi = 16^+$) to ground-state ($I^\pi = 0^+$) ratio obtained using the ^{176}Yb (^{4}He, 2n) reaction are shown in fig.1. The energy intervals E_α = 43-37 MeV correspond to the irradiations in Alma-Ata, E_α = 35.5-26.5 MeV to that in Dubna,

Fig.1. a) The excitation functions for the ^{178}Hf ground state (calculated) and for the ^{178m_2}Hf (measured) as function of the incident α-beam energy. The horizontal error bars correspond to the loss of energy due to the thickness of the target. b) The isomeric to ground-state ratio as function of the incident α-beam energy.

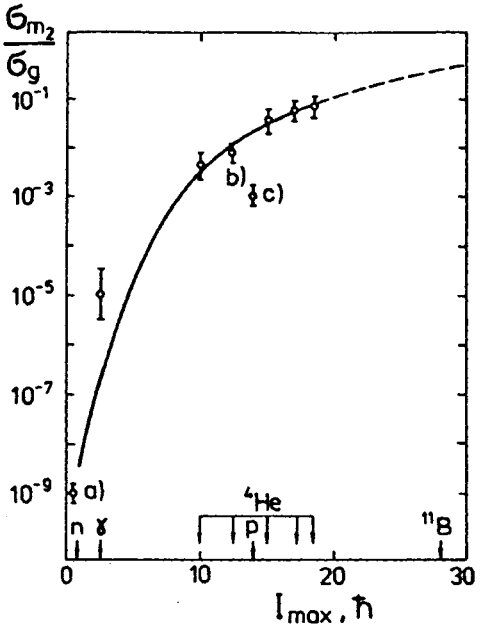


Fig.2. The $^{178m_2}Hf/^{178g}Hf$ ratio as function of the maximum spin of the incident particles (n_{th}, γ, p, α). The reaction ^{179}Hf (γ, n) $^{178m_2}Hf$ was investigated with using Dubna microtron MT-25. Points marked a), b) and c) correspond to measurements in refs./1,4 and 3/, respectively.

E_α=28-25 MeV to that quoted in ref./5/ and finally E_α=26 MeV to ref. /4/. Other points were obtained in the irradiation of the stack of enriched ^{176}Yb targets on Al foil backings at Alma-Ata cyclotron. The ground-state excitation function is calculated using statistical code version (see ref. /6/) taking also into account the phenomenological information on preequilibrium neutron emission. It is clear from fig.1 that the isomeric ratio can reach a magnitude of about 0.05-0.08. The σ_m/σ_g ratio is also shown in fig.2 for different reactions. It may be noticed that there is a regular dependence of this ratio as function of I_{max}.

The high-intensity of the beam current which is needed requires specific technics developed in Dubna of inclined or fast-rotating water-cooled targets. The target and irradiation parameters for some of the experiments carried out are given in Table 1. The inclined targets are operating successfully with an intensity of the $^4He^{++}$ beam up to 100 μA and higher. Up to now, with the two last irradiations together, about $3\cdot10^{14}$ atoms of $^{178m_2}Hf$ were produced. We may expect to get 10^{15} isomeric nuclei in 20 effective 100 μA beam days.

In the irradiation of targets with an Al backing the induced activation of Al itself leads to short-lived isotopes. Hence the radioactive Hf isotopes yield can be measured directly without chemical separation by means of γ-ray spectroscopy only after a short cooling time of about one week. This method was used to determine the isotopic distribution of Hf radioactive nuclei produced in irradiation No.3. The results are shown in Table 2 and compared with calculations including the different xn channel cross-sections for all the Yb isotopes present in the target. The yields are normalized to the one of ^{175}Hf. One may notice the fairly good agreement of the measured and calculated yields (see

Table 1. Characteristics of the irradiations of enriched ^{176}Yb targets with $^{4}He^{++}$ ions

Number of exposi-tions	Date	Target type	Target thickness (mg/cm²)	^{176}Yb enrichment (%)	Place of irradiation	Beam energy (MeV)	Integral numbers of He ions	Higher limit of the beam current (μA)	Quantity of $^{178m2}Hf$ nuclei
1.	12.89	Orthogonal Cu backing	68	93.0	Alma-Ata	43	$2.6\ 10^{19}$	20	$1.4\ 10^{13}$
2.	06.90	Orthogonal Al backing	90	96.4	Dubna	35.5	$1.5\ 10^{18}$	10	$0.4\ 10^{13}$
3.	07.90	Orthogonal Al backing	90	96.4	Dubna	35.5	$6.2\ 10^{18}$	12	$1.6\ 10^{13}$
4.	11.90	Inclined Al backing	$17/\sin\alpha=85$	96.4	Dubna	35.0	$5.0\ 10^{19}$	100	$1.2\ 10^{14}$
5.	12.90	Inclined Al backing	80	95.9	Dubna	35.0	$6.3\ 10^{19}$	100	$1.5\ 10^{14}$

Table 2. The isotopic distribution of radioactive hafnium nuclei produced in the irradiation No.3 (see Table 1). For ^{170}Hf and ^{171}Hf, only an upper limit could be calculated, following the upper limit of the amount of ^{168}Yb and ^{170}Yb given in the list of other ytterbium isotopes present in the 96% enriched ^{176}Yb

Mass number	$T_{1/2}$	Quantity	
		Experiment	Calculation
170	15.9h	$\sim 5 \cdot 10^{10}$	$< 8 \cdot 10^{11}$
171	12.2h	$1.5 \cdot 10^{11}$	$< 1.5 \cdot 10^{12}$
172	683d	$4.5 \cdot 10^{12}$	$4.4 \cdot 10^{12}$
173	24h	$0.9 \cdot 10^{13}$	$0.8 \cdot 10^{13}$
175	70d	$2.2 \cdot 10^{13}$	$2.2 \cdot 10^{13}$
$178m_2$	31y	$1.6 \cdot 10^{13}$	—
178g	stable	—	$2.9 \cdot 10^{14}$
$179m_2$	24.8d	$7.6 \cdot 10^{11}$	—

comment in the Table 2 caption). The most intensive activity is due to ^{175}Hf (70 d.) and ^{172}Lu (daughter of ^{172}Hf (683 d.)) however they are produced with a moderate yield at the used beam energy and with the enrichment up to 96% of the ^{176}Yb target.

These results are very promising as compared to other attempts to produce ^{172m_2}Hf as compiled in ref. /3/.

3. Chemical Separation

The chemical separation of hafnium isotopes from the bulk of reaction products was carried out by selective extraction chromatography from HNO_3 solutions on columns filled in with tri-n-octyphosphine oxide (TOPO). The irradiated target (Yb_2O_3, usually 100-200 mg) was dissolved in a mixture of 9 ml 12 M HNO_3, 3 ml 11.5M HCl, 0.5 ml

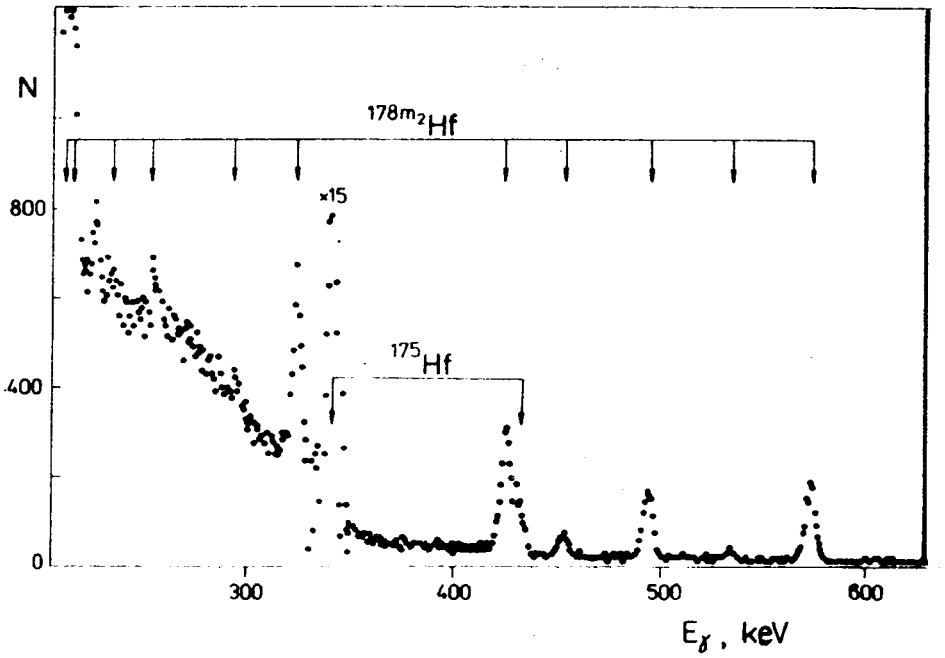


Fig.3. The γ-ray spectrum of the material chemically separated after 8 monts "cooling" time (irradiation No.2 in Table 1).

11M $HClO_4$ by heating up to 180° in an autoclave during 2 hours. This procedure is necessary because the irradiated rare-earth oxides are very hard to dissolve. The solution was dried and the residue was dissolved in 10 ml 6M HNO_3. The resulting solution was transfered to a 3 mm x 60 mm column which was filled in with 20% TOPO on the copolymer of divinylbenzene and styrene (Wofatite EP-60, 100-200 μm). After sorption, the column was washed with 30 ml 6M HNO_3 to ensure the purification of hafnium from the rare-earths and from most of the other elements produced in the irradiation process on the backings and their impurities. The hafnium elution may be performed with 2-3 ml 0.2M HF. The hafnium fraction was dried and twice evaporated with 0.5 ml 9M HCl to obtain Hf chloride.

In this separation process we could reach a high degree of purification ($\geq 10^7$) and an efficiency of ~80-90%. The quality of the separation was controlled by γ-spectroscopy. The spectrum of chemically separated material (irradiation No.2, after 8 months "cooling" time) is shown in fig.3. The high purity of the extracted matter can be observed in this spectrum: only the ^{175}Hf strongest lines are observed in addition to those of ^{178m_2}Hf. As the γ measurements have been made

just after the hafnium selective chemical extraction, the ^{172}Lu activity is negligible. Even at saturation ($T_{1/2}$ = 6.7d.), the activity due to this isotope is still one order of magnitude lower than the ^{175}Hf one.

4. Isotopic Separation

Several tests of mass separation with the PARIS-Separator (ref. /7/) of CSNSM-Orsay have been performed in order to get:

i) the mass calibration in a wide range of masses (10 units of masses),

ii) the separation yield of the hafnium isotopes.

The efficiency of the separator itself beeing high, the total yield is mainly governed by the ion-source efficiency.

The tests have been performed using a mixture of ^{175}Hf obtained by α irradiation of Yb and ^{181}Hf produced by neutron irradiation of natural hafnium.

After achieving chemical separation cycles as described above, a known activity of hafnium isotopes has been evaporated as chloride on quartz wool and placed inside a carbon tube with a small hole at the end.

The ion source is schematically shown in fig.4. The separation procedure, as usual, used CCl_4 as a jet gas. The ion source was designed in order to reduce the lost of matter by undesirable condensations.

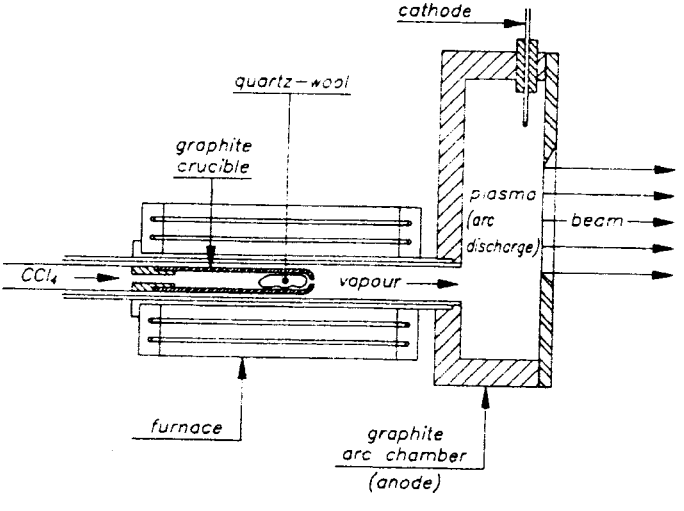


Fig.4. The scheme of the ion source of the isotopic separator PARIS in CSNSM-Orsay.

The carbon tube was placed in the furnace which was heated slowly up to 1400° C. The duration of the separation was ≅ 10 minutes.

The separated isotopes were collected on a copper foil placed in the focal plane of the separator by implanting them at 50 keV. The analysis of the remaining activity showed that no activity remained in the carbon tube. From the activity collected in the focal plane, we could deduce a separation yield of 5% for each of the hafnium isotopes. This result is quite satisfying if one takes into account the difficulties inherent to the hafnium element itself. However, this yield may be improved in two ways:

i) by a more constant distribution of the temperature in the arc chamber in order to reduce the trapping of hafnium in some regions of the ion chamber,

ii) by evaporating on the walls of the ion chamber a material non reactive to hafnium.

Anyway, following previous tests to separate hafnium isotopes in optimal conditions (ref. [8]), one should not expect a yield better than 10 %.

5. Conclusion

Experiments have been carried out to investigate the optimum conditions to produce the isomer ^{178m_2}Hf ($I^{\pi}=16^{+}$). A rather high absolute yield of $\sim 5 \cdot 10^{8}$/s of isomeric nuclei is obtained in the chosen reaction conditions, with an isomeric to ground-state ratio of 5 %. These results, together with the chemical and isotopic separations, are promising in the sense of performing new experiments with microweight quantities of this high-spin isomer used as a target.

Some experimental tests have been already performed to prepare further nuclear structure investigations as Coulomb excitation induced by heavy ions (^{208}Pb) in collaboration with GSI-Darmstadt as well as a preliminary attempt using laser spectroscopy.

This high-spin target could promote also new investigations, among others: transfer reactions with light ions, induced fission, (n, γ) and (n, n$'\gamma$) reactions, excitation of high-spin neutron resonances, etc. Such investigations could open new directions in studying the multiquasiparticle high-spin state properties.

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References

1. Helmer R.G., Reich C.W. — Nucl.Phys., 1973, A211, p.1.
2. Wilhelmy J.B. et al. — Proc. IV Intern. Conf. on Nuclei far from Stability, Geneva, 1981, p.684.
3. Kutschera W. et al. — Proc. First Intern. Conf. on Radioactive Beams, Berkeley, 1989, p.345.
4. Khoo T.L., Lövhöiden G. — Phys.Lett., 1977, 67B, p.271.
5. Kugler A., private communication.
6. Karamian S.A. — Preprint JINR P4-11339, Dubna, 1978.
7. Camplan J., Meunier R., Fatu C. — Proc. VIII EMIS Intern. Conf., Gothenburg, 1973.
8. Charvet I., Bernas R. — Nucl. Inst. and Meth., 1967, 51, p.1.

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