CONF-891044--10

ON THE PRODUCTION OF A BEAM OF THE LONG-LIVED 16^+ ISOMER IN HAFNIUM-178

W. Kutschera, I. Ahmad, W. J. Childs, R. V. F. Janssens and R. C. Pardo
Argonne National Laboratory, Argonne, IL 60349
L. F. Mausner
Brookhaven National Laboratory, Upton, NY 11973

CONF-891044--10

DE90 009720

ABSTRACT

The long-lived high-spin isomer of ^{178}Hf, with $T_{1/2}$ = 31 y and $I^{\pi} = 16^+$, presents a unique opportunity for a projectile and/or target in nuclear and atomic physics. Various aspects in using ^{178m}Hf for this purpose are discussed in this paper. One possible production mode, the irradiation of Ta with protons, was investigated. Cross sections of ~40 and ~200 μb were found for proton energies of 46 and 92.5 MeV, respectively. First ideas for a possible isomeric enrichment procedure by selective laser deflection are presented.

1. INTRODUCTION

Most radioisotopes considered for radioactive beams are so short-lived that they require an on-line production and acceleration. There are, however, a few radioisotopes with properties of special interest which live long enough to separate production and use completely. This has the advantage that production, purification and acceleration can be performed at facilities which are best suited for the respective purpose. In addition, such radioisotopes can be used to produce both a beam and a target. Well-known examples for such radioisotopes are ^{3}H ($T_{1/2}$ = 12.3 yr) and ^{14}C ($T_{1/2}$ = 5730 yr). They have been used as beams at accelerators for a number of years[1]. The level of radioactivity connected with making beams of these isotopes can be handled at many accelerator facilities.

In the present paper, we should like to discuss the prospects of producing a beam of the 16^+ isomer in ^{178}Hf. A long half-life of 31 years and the very high spin make ^{178m}Hf a unique nuclear projectile, which would allow one to study high-spin phenomena in a new way.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

MASTER

The submitted manuscript has been authored by a contractor of the U. S. Government under contract No. W-31-109-ENG-38. Accordingly, the U. S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U. S. Government purposes.

Considering both the beam and the target consisting of ^{178m}Hf, one would have the exceptional situation of an intrinsic system spin of 32 $\hbar$. Once produced, the level of activity does not present an insurmountable problem for a heavy-ion accelerator environment. For example, a ^{178m}Hf activity of 100 μCi corresponds to 5.2×10^{15} atoms of ^{178m}Hf. Assuming a concentration of 1 μCi ^{178m}Hf per milligram of a suitable matrix source material, a typical consumption rate of 1 mg/h, an ionization efficiency of 10^{-3}, and a transmission of 10%, one could deliver a ^{178m}Hf beam intensity of 1.5×10^{6} atoms/s for about four days. With a ^{178m}Hf concentration of 10 μCi/mg and a hundred times higher activity (10 mCi), which would still not pose a big radiation problem, one could probably deliver a beam of 10^{7} atoms/s for about a month of running time.

In section 2, we briefly discuss the nuclear properties of ^{178m}Hf and in section 3 we review the previous production methods. In section 4 our current results on a ^{178m}Hf production are presented. In section 5 we discuss the problem of separating the isomer from its ground state and present some first ideas how this might be achieved with the help of laser techniques. Finally, we discuss some future prospects of a ^{178m}Hf beam in section 6.

2. NUCLEAR PROPERTIES OF ^{178m}Hf

Figure 1 shows the decay scheme of the ^{178m}Hf isomer, as it was established through a series of measurements in the past[2-6]. Other properties of ^{178}Hf can be found in updated compilations on A=178 nuclei[7,8]. The exceptionally long half-life of the isomer is explained as a five times K-forbidden ($\Delta K - L = 5$) E3 transition from a $K^{\pi} = 16^{+}$, $I^{\pi} = 16^{+}$ isomeric state to a $K^{\pi} = 8^{-}$, $I^{\pi} = 13^{-}$ state. The population of the isomer is difficult since it is easily bypassed by transitions from higher lying states of lower or equal spin. For example, the 14^{+} and 16^{+} members of the ground state band[9] are at an excitation energy of 2777.6 and 3436.2 keV, respectively. Similarly, states with spins of 11^{+}, 13^{-}, 14^{+} and 14^{-} are located a few hundred keV above the isomer[5].

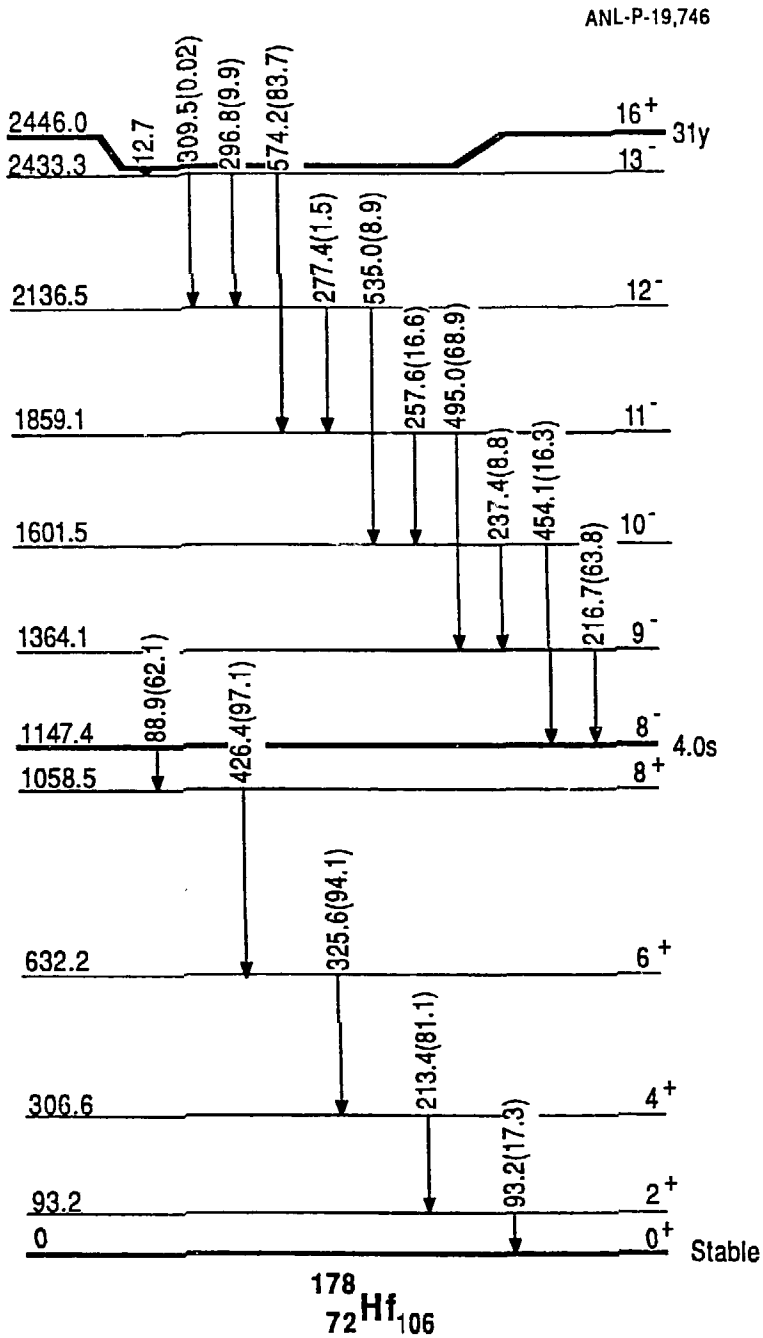

Fig. 1. Decay scheme of the 31-year isomer in ^{178}Hf. Level and transition energies are given in keV. Gamma-ray intensities per 100 decays of the 16^+-isomer are given in parenthesis, with the 4.0-sec isomer in equilibrium. The 12.7-keV E3 transition of the isomer has not been observed so far.

3. PRODUCTION OF ^{178m}Hf IN THE PAST

Several different reactions have been used in the past to produce ^{178m}Hf for spectroscopic studies[2-6]. A brief account of various methods used for production is presented below. In all cases except for the in-beam measurement[5], a chemical purification was performed prior to isomer decay measurements.

Thermal neutron capture: ^{177}Hf(n,γ)^{178m}Hf

Helmer and Reich[2] discovered the isomer after irradiating 100-200 mg of HfO_2 with natural isotopic abundance in a thermal-neutron flux of $\phi_n > 4\times10^{14}$ n/cm^2/s over a period of 2 years. After a cooling time of 3 years, the basic decay scheme was established. Later[3], the same group determined a neutron capture cross section to the isomer of $\sigma_m = (2 \pm 1)\times10^{-7}$ b and measured the half-life to be $T_{1/2} = 31 \pm 1$ y. We estimate that the total ^{178m}Hf production in these experiments was in the order of 10^{12} to 10^{13} atoms. From the total capture cross section of ^{177}Hf, $\sigma(n_{th}) = 390$ b, one obtains a very small ^{178m}Hf/^{178g}Hf ratio of about 10^{-9}. This makes it a rather unfavorable production mode for ^{178m}Hf.

Photo nuclear production: ^{181}Ta(γ,p2n)^{178m}Hf

De Boer et al.[4] used Ta slits from a 85-MeV electron linac which had been irradiated for about one year with unknown amounts of beam. After a cooling period of three years, Hf was chemically extracted and more details about the decay scheme of ^{178m}Hf were established. No information on the amount of ^{178m}Hf activity was reported. One may, however, expect that a photon-induced reaction would also result in a low ^{178m}Hf/^{178g}Hf ratio.

In-beam gamma-ray measurement with the ^{176}Yb(α,2nγ)^{178}Hf reaction

High-spin gamma-ray spectroscopy studies in ^{178}Hf were performed by Khoo and Lovhoiden[5] bombarding ^{176}Yb with alpha particles in the energy range from 24 to 35 MeV. In this work, a weak 126.1-keV gamma transition from a (14^-) state at 2749.1 keV was assumed to populate the 16^+ isomer. A gamma-ray intensity of 0.09% relative to 100% for the

213.4-keV, $4^+ \rightarrow 2^+$ groundstate band transition was observed. Since the $(14^-) \rightarrow 16^+$ transition would have primarily a multipolarity M2, the internal conversion would increase the decay strength by a factor of ~10. This then would lead to a $^{178m}Hf/^{178g}Hf$ production ratio of about 0.01, which would make it a relatively favorable reaction for producing the 16^+ isomer. However, the excitation energy of the 16^+ level deduced from this work is 1.4 keV higher than the one determined in the work of Van Klinken et al.[6], which seems outside the uncertainties of high-resolution gamma-ray spectroscopy. In view of the possible strong isomer population, this reaction should be studied again with emphasis on measuring the 16^+-isomer production rate.

Alpha-induced reaction: $^{181}Ta(\alpha,\alpha p2n)^{178m}Hf$

Van Klinken at al.[6] also used Ta slits from an accelerator. The slits were irradiated in a cyclotron chiefly by alpha particles with energies up to 120 MeV. Details of the irradiation history were not available. Chemical extraction from the activated parts of the slits resulted in a ^{178m}Hf source strength of 25 nCi, corresponding to 1.3×10^{12} atoms. The direct decay of the 16^+ isomer via an M4 transition to the 12^- state was found (see Fig. 1) which established the excitation energy of the isomer at 2446.01 keV. In this work, the possibility of preparing a high-spin target from the source was mentioned, pointing out the problem of substantial admixtures from ^{178}Hf in its ground state.

4. PRODUCTION OF ^{178m}Hf BY PROTON-INDUCED SPALLATION OF TANTALUM

In the present work, we have bombarded a stack of five 0.8-mm thick Ta foils with protons of 50 μA intensity and mean energies of 190, 161, 133, 92.5, and 46 MeV, respectively. With this condition, a one-hour irradiation was performed at the Brookhaven Linac Isotope Production (BLIP) facility[10]. After a relatively short cooling period of 3.5 months, gamma-ray measurements of the irradiated foils were performed. Figure 2 shows a gamma-ray spectrum from the foil irradiated with

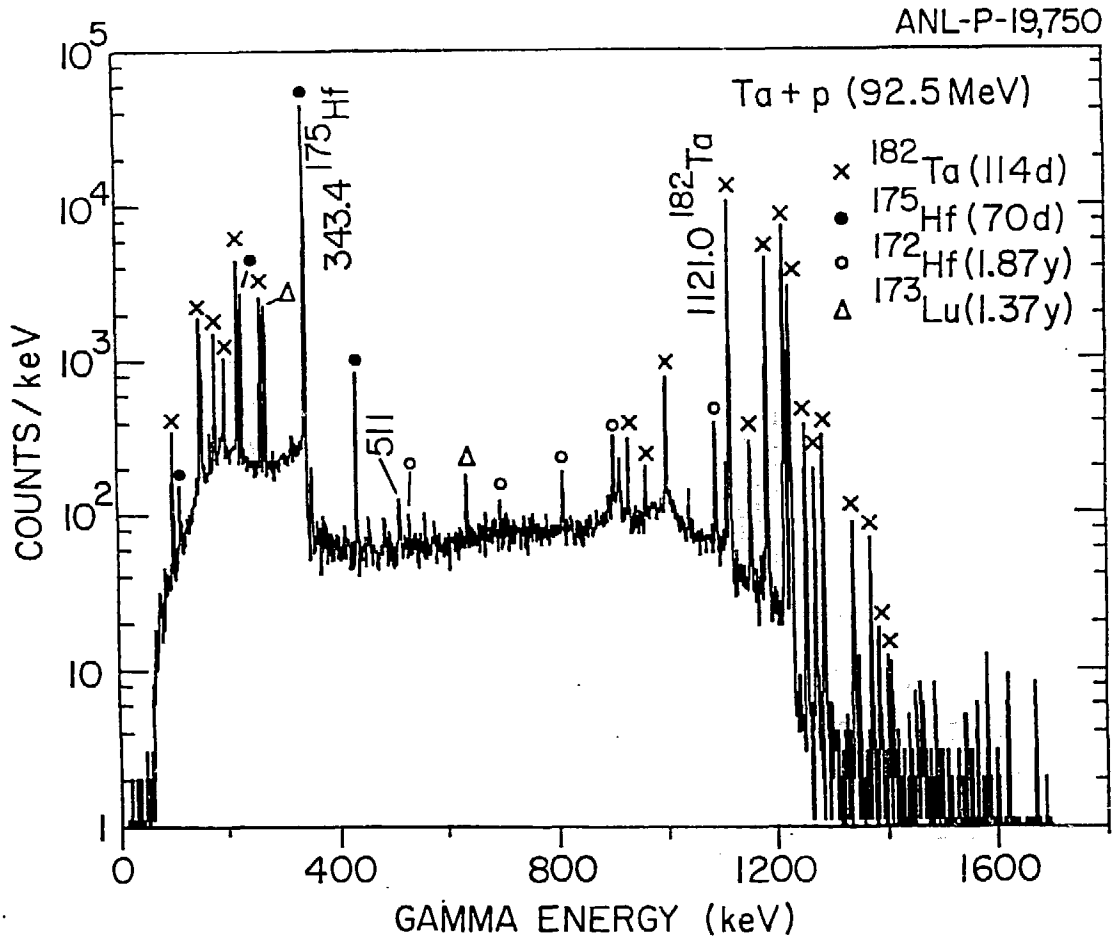

Fig. 2. Sum of γ-ray singles spectra accumulated for 25 minutes with four Ge detectors of the Argonne Notre Dame BGO gamma ray facility. Only a small central piece of the Ta foil irradiated with 92.5 MeV protons was used for this measurement.

92.5-MeV protons. It is dominated by gamma-lines from the 114-day activity of ^{182}Ta, apparently produced by secondary-neutron activation of ^{181}Ta. The strongest gamma-line is due to the decay of ^{175}Hf, produced by the cumulative yield of the reactions ^{181}Ta(p,7n)^{175}W (34 m), ^{181}Ta(p,p6n)^{175}Ta (10.5 h), and ^{181}Ta(p,α3n)^{175}Hf (70 d). Several gamma lines from the decay of long-lived ^{172}Hf (1.87 y) can also be seen. This activity is produced by evaporating three more particles and accumulating the corresponding shorter-lived isobars as in the case of ^{175}Hf. No trace of the ^{178m}Hf decay lines can be seen.

In order to detect the weak ^{178m}Hf activity, we performed coincidence measurements with the Argonne Notre Dame BGO gamma-ray facility. The facility consists of a central BGO ball with 50 hexagon elements, surrounded by 12 Compton suppressed Ge detectors. The spectrum of Figure 3 was generated by demanding a triple coincidence

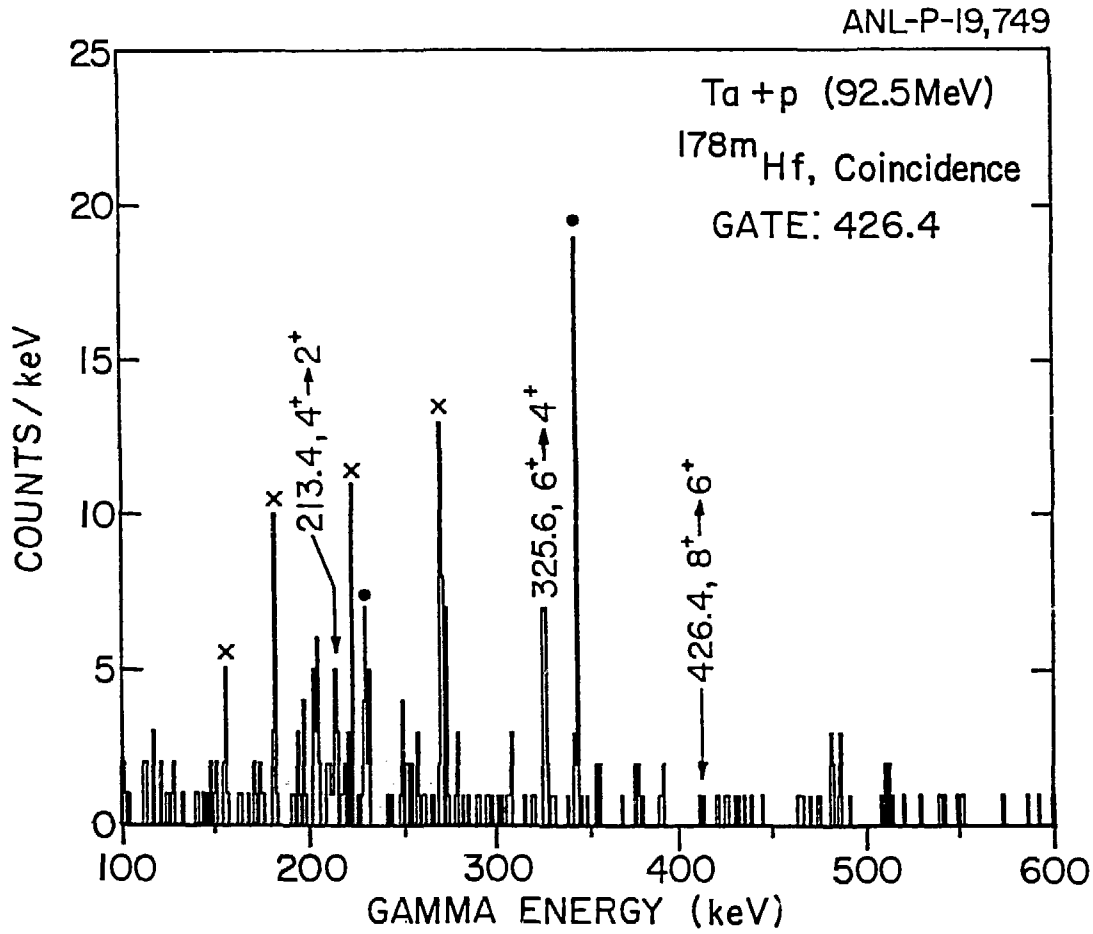

Fig. 3. Triple coincidence measurement with the full BGO array (see text) for the same sample as in Fig. 2. The accumulation time was 43 hours. A gate is set on the $8^+ \rightarrow 6^+$ transition of the ^{178m}Hf decay cascade. The background of other activities is reduced by six orders of magnitude relative to Ge singles measurements. The markers of the background lines denote the same isotopes as in Fig. 2.

between any of the elements of the BGO ball and two of the Ge detectors. With a gate on the $8^+ \rightarrow 6^+$ transition of the ^{178m}Hf decay cascade, a clear signature of the two following transitions in the decay can be

seen. Taking the coincidence detection efficiency into account, we calculate from these data an approximate production cross section of 0.2 mb for the $^{181}Ta(p,\alpha)^{178m}Hf$ reaction at 92.5 MeV.

Central parts of each irradiated Ta foil (~1 cm^2) were dissolved in a mixture of conc. HNO_3 and conc. HF. The solution was evaporated to dryness and redissolved in 7 M HCl/4 M HF and loaded on an anion-exchange resin column. Ta was strongly absorbed while Hf passed through[11]. Although the strong ^{182}Ta activity was efficiently removed, the remaining activities of ^{172}Hf, ^{175}Hf, ^{178}W, ^{173}Lu and others prevented the weak ^{178m}Hf gamma lines from being measured in direct gamma spectra for the the three highest proton energies. However, at Ep = 46 MeV the 574.2, 495.0 and 426.4 keV transitions from ^{178m}Hf could clearly be detected and a cross section of about 40 μb extracted by comparing their intensity with that of the 343.4 keV line in ^{175}Hf. Similarly at Ep = 92.5 MeV, a cross section of 0.2 mb consistent with that from the coincidence measurement of the unseparated material could be extracted. The results for the cross section measurements of the Hf radioisotopes is summarized in Table 1.

Table 1. Total cross sections for the production of hafnium radioisotopes by proton irradiation of Ta. Estimated errors in the cross section values are ± 30%. Blanks mean that no measurement was made.

Isotope	$T_{1/2}$	Production cross section (mb)				
		190 MeV	161 MeV	133 MeV	92.5 MeV	46 MeV
^{172}Hf[a]	683 d		43	47	8	0.4
^{175}Hf[a]	70 d	45	55	61	74	8
^{178m}Hf	31 y				~0.2	~0.04

[a] Cumulative yield.

From these data, we estimate a total ^{178m}Hf production of about $2x10^{12}$ atoms. This was accomplished in only one hour of irradiation. It is easy to calculate that with a ten times thicker Ta target and 3 weeks of irradiation one would produce about 10^{16} ^{178m}Hf atoms. However, this material needs further treatment since the $^{178m}Hf/^{178g}Hf$ ratio is likely to be in the 10^{-3} range or below.

5. ISOMER SEPARATION BY SELECTIVE LASER DEFLECTION

Ultimately, the isomeric purity of a ^{178m}Hf beam will be the limiting factor for its usefulness. To the best of our knowledge, isomeric purification has not yet been explored in a nuclear system. Here we present a method, which should work in principle. In practice, of course, many difficult details have to be solved before an efficient enrichment of the isomer can be achieved. The basic idea is to absorb many laser photons consecutively into a spontaneously re-emitting level of a very slowly moving atom. The accumulative photon momentum transfer caused by absorption from one direction and re-emission in random direction will result in a net deflection of the absorbing atoms from the initial beam direction. Such a scheme has recently been used by G. R. Janik et al.[12] to demonstrate the separation of Kr isotopes. In this experiment, metastable Kr atoms in the $4p^5 5s$ (J=2) state moving with a velocity of 440 m/s were intercepted perpendicular to the direction of motion by laser light of 811 nm, which induced transitions to the $4p^5 5p$ (J=3) state with spontaneous re-emission back to the metastable state. With about 1500 photons absorbed per atom, they observed a deflection of 19 mrad and an isotopic enrichment of 1.2×10^4. Similarly one could use the difference in hyperfine interaction between the ^{178}Hf ground and isomeric state for a selective laser deflection. However, there are several difficulties which have to be overcome. In the experiment of Janik et al.[12], only a small fraction (5×10^{-4}) of the total krypton beam was in the metastable state. The use of the metastable atoms was necessary, since transitions from the ground state get easily trapped in metastable states in the emission process. A similar condition can occur in ^{178}Hf. It was pointed out to us[13] that the originally planned excitation of the $5d^2 6s^2$, $^3F_2$ (J=2) atomic ground state with 618-nm photons to the $5d^2 6s6p$, $^3D_2$ (J=2) excited state would primarily lead to a re-emission to the metastable $^3F_3$ (J=3) state. However, excitation with 713 nm would lead to the $5d^2 6s6p$, $^3D_1$ (J=1) state which decays back to the groundstate. We have recently verified that this transition can be easily excited by measuring the isotope shifts for natural Hf with laser light of this wave length. The second challenge is the production of a very slow (cryogenically cooled) Hf

beam. Assuming that this problem can be solved, an efficient deflection of the ^{178}Hf ground state should be possible. The ground state rather than the isomer should be deflected since only one atomic transition is possible for the 0^+ ground state, whereas for the 16^+ isomeric state the hyperfine interaction will lead to 5 ground state levels with total spin F = 14 to 18 and three excited levels with F = 15 to 17. Once an isomerically enriched material is produced, an ion beam production in an ECR source[14)] would probably be the most efficient method for providing a beam for heavy-ion acceleration.

6. CONCLUSIONS

The production of a beam of the ^{178m}Hf isomer presents a considerable technical challenge. Methods have been discussed which, in principle, allow to reach this goal. Since the isomeric purity is the most difficult problem to solve, a reaction with the largest possible ^{178m}Hf/^{178g}Hf ratio should be used. For this the ^{176}Yb(α,2n)^{178m}Hf reaction looks most promising and should be explored. At the appropriate alpha energy between 20 and 40 MeV, this reaction also has the advantage that ^{172}Hf will not be produced. The long half-life and the correspondingly low specific activity make ^{178m}Hf a radioisotope with only a modest radiation problem. This has the great advantage that a ^{178m}Hf material can be used at almost any heavy-ion accelerator in the world capable to accelerate such heavy species. The long half-life allows to completely decouple production and purification procedures from the actual use as a nuclear beam and/or nuclear target.

Acknowledgement

We thank Gene Sprouse for fruitful discussions on laser spectroscopy of Hf isotopes, and Ralph Bane, Alice Essling, Don Graczyk and Edmund Huff for the chemical separation of Hf from Ta. This work was supported by the U. S. Department of Energy, Nuclear Physics Division, under Contract W-31-109-ENG-38.

REFERENCES

1. Kutschera, W., "Negative Ion Beams of Rare and Radioactive Isotopes", Proc. 3rd Int. Conf. on Electrostatic Accelerator Technology, Oak Ridge, IEEE CH1639-4 (1981) 222.

2. Helmer, R. G. and Reich, C. W., "Decay of an Isomeric State in ^{178}Hf with K $\geq$ 16", Nucl. Phys. A114, 649 (1968).

3. Helmer, R. G. and Reich, C. W., "Half-Life of ^{178m2}Hf and its Neutron Capture Production", Nucl. Phys. A211, 1 (1973).

4. De Boer, F. W. N., Goudsmit, P. F. A., Meijer, B. J., Kapteyn, J. C., Konijn, J., and Kamermans, R., "The Four Quasi-Particle ^{178}Hf Isomeric State, its Excitation Energy and Multipolarities of Deexciting Transitions", Nucl. Phys. A263, 397 (1976).

5. Khoo, T. L. and Lovhoiden, G., "Structural Changes in the Yrast States of ^{178}Hf", Phys. Lett. 67E, 271 (1977).

6. Van Klinken, J., Venema, W. Z., Janssens, R. V. F. and Emery, G. T., "K-Forbidden Decays in ^{178}Hf; M4 Decay of an Yrast State", Nucl. Phys. A339, 189 (1980).

7. Browne, E., "Revised A-Chain A = 178", Nuclear Data Sheets 54, 199 (1988).

8. Browne, E. and Firestone, R. B., "Table of Radioactive Isotopes", ed., Sirley, V. S., (John Wiley, New York, 1986) p. 178-2.

9. Hamilton, J. H., Ramayya, A. V., Ronningen, R. M., Sayer, R. O., Yamada, H., Maguire, C. F., Colombani, P., Ward, D., Diamond, R. M., Stephens, F. S., Lee, I. Y., Butler, P. A. and Habs, D., "Isomer Trapping Following Coulomb Excitation of High Spin States in ^{178}Hf", Phys. Lett. 112B, 327 (1982).

10. Mausner, L. F., Mirzadeh, S., Schnakenberg, H. and Scrivastaza, S. C., "The Design and Operation of the Upgraded BLIP Facility for Radionuclide Research and Production," J. Appl. Rad. and Isot., in press.

11. The chemical separation was performed in the Analytical Chemistry Laboratory at Argonne.

12. Janik, G. R., Cannon, B. D., Ogorzalek-Loo, R. and Bushaw, B. A., "Isotopically Selective Optical Deflection of a Krypton Atomic Beam" J Opt. Soc. Am. B9, 1617 (1989).

13. G. Sprouse, private communication.

14. Pardo, R., "Operating Experience of an ECR Ion Source on a High Voltage Platform", Nucl. Instr. Meth. B40/41, 1014 (1989).

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.