Sorption Behaviour of Short-Lived W, Hf and Ta Isotopes on Ion Exchangers from HCl/HF Solutions in Fast On-line Experiments

By D. Schumann, R. Dressler, S. Fischer, St. Taut

Institute of Analytical Chemistry, University of Technology Dresden, D-01062 Dresden, Germany

R. Binder

Institute of Analytical Chemistry, University Leipzig, Germany

Z. Szełkowski, B. Kubica

H. Niewodniczanski Institut of Nuclear Physics, Krakow, Poland

L. I. Guseva, G. S. Tikhomirova

Institute of Geochemistry and Analytical Chemistry, Moscow, Russia

O. Constantinescu, V. P. Domanov, M. Constantinescu, Dinh Thi Lien, Yu. Ts. Oganessian, V. B. Brudanin

Joint Institute of Nuclear Research, Dubna, Russia

and H. Bruchertseifer

Paul-Scherrer-Institute, Villigen, Switzerland

(Received November 24, 1994; revised January 20, 1995)

Ion exchange / Short-lived tungsten, tantalum and hafnium isotopes / On-line separation / Fluoride / Chloride complexes

Abstract

In fast on-line experiments, short-lived isotopes of tungsten and tantalum can be separated from hafnium using an ion exchange system consisting of two columns (Dowex 50X8, Dowex 1X8) and 0.05 M HCl/1 · 10⁻³ M HF aqueous solution as eluent. This chemical system can be recommended for investigations of the elements 105 and 106. Moreover, determination of nuclear physical data of short-lived nuclides is possible. The half-life of ¹⁶⁵Hf was determined to be 78.6 ± 1 s.

Introduction

Until now chemical research studies of transactinide elements ended at element 105, the last member of the periodic table, which can be produced in considerable amounts and has isotopes with sufficiently long half-lives (²⁶²105: 34 s [1], ²⁶³105: 27 s [2]). The discovery of the isotopes ²⁶⁵106 and ²⁶⁶106, with expected half-lives of ~10–30 s (1993, Dubna, [3]), provides chemists the opportunity to carry out studies on the isolation and chemical properties of this interesting element. For the development of chemical separation systems the elements of subgroup IV, V and VI are usually used in model experiments as the lighter homologues of the elements 104, 105 and 106.

Fast on-line separation of transactinides from trivalent transplutonium elements (lanthanides in model

experiments) as well as from the decay products formed during the transport from the target chamber to the chemical set up is initially necessary. For the isolation and chemical investigation of short-lived transactinide elements in aqueous solution, Szełkowski *et al.* [4] developed a continuous ion exchange method consisting of three columns (Dowex 50 – Dowex 1 – Dowex 50) with acids such as HF [4], H₃PO₃ [5] or H₂C₂O₄ [6] as eluents. Sorption of transplutonium elements on the first column with high decontamination factors was possible using this equipment.

Investigations of element 106 additionally require a complete separation from element 104, which is produced in considerable amounts directly in the nuclear reaction and appears as an α -decay product of element 106. The ion exchange system described above is not applicable for such a separation, because tetravalent as well as penta- and hexavalent elements exist as anions in complex forming agents such as HF or H₂C₂O₄ under the conditions used in [4–6]. Therefore, a new approach is needed.

Mixed solutions of HCl/HF have been used for the separation of subgroup IV, V and VI elements [7–9]. Ultramicroamounts of Nb, Ta, Zr, Hf and lanthanides were investigated concerning their sorption behaviour on Dowex 50X8 from 0.1 M HCl/1 · 10⁻³ – 1 · 10⁻⁵ M HF solutions [10, 11]. Previous batch studies showed that separation of W, Mo, Ta and Pa from Zr and Hf is possible using 0.05–0.1 M HCl/1 · 10⁻⁴ – 1 · 10⁻³ M HF and the strongly acidic cation exchanger Dowex 50X8 [12]. The same behaviour is ex-

pected in the case of W and Mo. Consequently, if the elements 105 and 106 behave like their lighter homologues, then this system could be suitable for fast on-line isolation and chemical investigations of these two elements. Therefore, we studied the sorption behaviour of short-lived Hf, Ta and W radionuclides produced in heavy ion reactions on Dowex 50X8 and Dowex 1X8 from dilute HCl/HF solutions using the apparatus for the on-line separation described in [4].

Additionally, we determined the half-life of ^{165}Hf using the ion exchange method with HF solution as eluent for chemical separation. Two different values have been reported for the half-life of this isotope: 75 s [13] and 108 s [14].

Experimental

Short-lived isotopes of hafnium, tantalum and tungsten were produced via the following heavy ion reactions using a ^{20}Ne beam (10^{12} pps):

at the U-400 cyclotron (Flerov Laboratory of Nuclear Reactions, JINR Dubna, Russia). The recoiled atoms were transported from the target chamber to the chemical set up by use of a KCl/Ar gas jet (1–2 l/min) [15]. The equipment described in [4] was modified for the present experiments as follows: Instead of three columns (Dowex 50X8 – Dowex 1X8 – Dowex 50X8), we used only two (Dowex 50X8 – Dowex 1X8, 200–400 mesh, 50–100 mg). These two columns were simultaneously monitored during the experiment using a HPGe-PT and a high resolution HPGeX detector. 15 γ -spectra per 1 min were recorded from each column and summed. The spectra did not change qualitatively during the time measured. All separation experiments were performed with 0.05 M HCl/ $1 \cdot 10^{-3}$ M HF solution as eluent and flow rates of 2–4 ml/min. The decontamination factor of Hf was calculated from the ratio of the isotopes' γ -activity on Dowex 50X8:Dowex 1X8. The enriching factor of tantalum was determined by the ratio of the intensity on Dowex 1X8:Dowex 50X8.

Reaction (1) was used for the determination of the half-life of ^{165}Hf . The chemical separation system described above is not applicable in this case because separation of Hf from all other contaminants is necessary. Therefore, we used 0.2 M HF solution to adsorb the Hf completely on Dowex 1X8 [4]. The activity was collected on the anion column for three minutes, then the column was washed with 0.2 M HF and measured off-line 30 times per 15 s. The experiment was repeated 40 times and the γ -spectra were summed.

Results and discussion

Figs. 1–3 show the γ -spectra of the Dowex 50X8 and Dowex 1X8 columns eluted with 0.05 M HCl/

Fig. 1. γ -Spectra of the Dowex 50X8 and Dowex 1X8 columns; nuclear reaction: $^{147/149}\text{Sm} + ^{20}\text{Ne} \rightarrow \text{Hf}$, solution 0.05 M HCl/ $1 \cdot 10^{-3}$ M HF.

$1 \cdot 10^{-3}$ M HF for the reactions (1–3). The main products were obtained in the 3n and 4n reaction channel, although 5n and $(^{20}\text{Ne},\alpha\text{xn})$ reactions have some importance, too. For instance, the appearance of ^{166}Ta [Fig. 2, reaction (2), 5n channel] and ^{166}Hf [Fig. 3, reaction (3), $(^{20}\text{Ne},\alpha\text{xn})$] can only result from such reactions.

The energies of the γ -rays used for the detection of the elements on the columns and for calculating enriching and decontamination factors are displayed in the figures. Detailed analysis of the γ -spectra yields the following results.

1. Hafnium

The γ -spectra of the cation and anion exchange columns in the energy range of 100–300 keV are shown in Fig. 1. ^{166}Hf ($T_{1/2} = 6.77$ min) together with ^{166}Lu ($T_{1/2} = 2.65$ min/1.4 min), and ^{165}Hf ($T_{1/2} = 78.6$ s), as well as its daughter ^{165}Lu ($T_{1/2} = 2.65$ min), can be found on the cation exchange column. Only small amounts of ^{165}Hf and ^{166}Hf could be detected on the anion exchange column. From these results a decontamination factor of $3 \cdot 10^2$ was calculated, e.g. Hf and its decay product Lu are nearly completely adsorbed on the cation exchange column. This agrees very well with the results obtained in batch experiments [12].

According to literature data on stability constants of hafnium fluoride complexes [16], the species ad-

aration of tungsten and tantalum in heavy ion experiments. Using 0.05 M HCl/1 · 10⁻³ M HF solution as eluent, Hf isotopes produced directly in the nuclear reaction as well as the decay products of Ta formed during the transport to the chemical set up are completely adsorbed on the cation exchange column. Lutetium was shown to adsorb on Dowex 50X8, too. Tantalum and tungsten pass the first column and are adsorbed on the anion exchanger. Consequently, the chemical system presented should be applicable for separations of the elements 105 and 106 from heavy actinides. Separation of element 106 from 104 should be possible.

The half-life of ¹⁶⁵Hf was determined to be 78.6 ± 1 s, which supports the value reported in Ref. [13].

Acknowledgement

The authors are very grateful to Prof. I. Zvara for his permanent interest in this work. Moreover, we wish to thank Mr. G. V. Buklanov and Mr. L. N. Salamatin for preparing the targets, Mr. A. F. Novgorodov for many helpful discussions and the U-400 cyclotron staff for their service.

This work was supported by the Bundesministerium für Forschung und Technologie, Germany.

References

- Ghiorso, A., Nurmia, M., Eskola, K., Eskola, P.: *Phys. Rev. C* **4**, 1850 (1971).
- Schädel, M., Brüchle, W., Schimpf, E., Zimmermann, H. P., Guber, M. K., Kratz, J. V., Trautmann, N., Gäggeler, H., Jost, D., Kovacs, J., Scherer, U. W., Weber, A., Gregorich, K. E., Türler, A., Czerwinski, K. R., Hannick, N. J., Kadkhodayan, B., Lee, D. M., Nurma, M. J., Hoffmann, D. C.: *Radiochim. Acta* **57**, 85 (1992).
- Lougheed, R., *et al.* for a Dubna-Livermore collaboration, *Proc. Int. Conf. Actinides-93*, Santa Fe, Sept. 19–24, 1993, to appear in *J. of Alloys and Compounds*.
- Szeglowski, Z., Bruchertseifer, H., Domanov, V. P., Gleisberg, B., Guseva, L. I., Hussonois, M., Tikhomirova, G. S., Zvara, I., Oganessian, Yu. Ts.: *Radiochim. Acta* **51**, 71 (1990).
- Szeglowski, Z., Guseva, L. I., Dinh Thi Lien, Domanov, V. P., Constantinescu, O., Tikhomirova, G. S., Hussonois, M.: *Radiokhimiya* **35**, 59 (1993).
- Szeglowski, Z., Guseva, L. I., Dinh Thi Lien, Domanov, V. P., Constantinescu, M., Constantinescu, O., Tikhomirova, G. S., Hussonois, M.: *Radiokhimiya*, in press.
- Korkisch, J.: *Handbook of Ion Exchange Resins: Their Application to Inorganic Chemistry*, CRC Press, Inc. Boca Raton, Florida 1989.
- Gregorich, K. E., Henderson, R. A., Lee, D. M., Nurmia, M. J., Chasteler, R. M., Hall, H. L., Bennett, D. A., Gannett, C. M., Chadwick, R. B., Leyba, J. D., Herrmann, G., Hoffman, D. C.: *Radiochim. Acta* **43**, 223 (1988).
- Kratz, J. V., Zimmermann, H. P., Scherer, U. W., Schädel, M., Brüchle, W., Gregorich, K. E., Gannett, C. M., Hall, H. L., Henderson, R. A., Lee, D. M., Leyba, J. D., Nurmia, M. J., Gäggeler, H., Jost, D., Baltensperger, U., Ya Nai-Qi, Türler, A., Linert, Ch.: *Radiochim. Acta* **48**, 121 (1989).
- Bruchertseifer, H., Heller, W., Haberberger, F., Trautmann, N., Zimmermann, P., Schädel, M., Brüchle, W., Skarnemark, G., Alstad, J., Domanov, V. P., Szeglowski, Z., Vobecky, M.: *Jahresbericht der Universität Mainz*, 1991, p. 8.
- Bruchertseifer, H., Heller, W., Binder, R., Trautmann, N., Herrmann, G., Kratz, J. V., Zimmermann, H. P., Nähler, A., Schädel, M., Brüchle, W., Skarnemark, G., Alstad, J., Fischer, S., Roß, A., Dressler, R.: *ZfI-Mitteilungen Leipzig*, **166**, p. 53.
- Schumann, D., Fischer, S., Taut, St., Dressler, R., Novgorodov, A. F., Misiak, R., Trautmann, N., Schädel, M., Bruchertseifer, H.: *GSF Scientific Report 1993*, p. 241.
- Hild, T., Schmidt-Ott, W. D., Freistein, V., Meissner, F., Runte, E., Salewski, H., Michaelsen, R.: *Nucl. Phys. A* **492**, 237–252 (1989).
- Bruchertseifer, H., Eichler, B.: *Radiochem. Radioanal. Letters* **48**, 391–400 (1981).
- I. Zvara, Domanov, V. P., Shalevsky, M. R., Petrov, D. V.: *Communication of the JINR 12-80-48*, Dubna 1980.
- Höglfeld, E.: *Stability Constants of Metal-Ion-Complexes, Part A: Inorg. Ligands*, IUPAC Chemical Data Series, Nr. 21, Pergamon Press, Oxford, New York, Toronto, Sydney, Paris, Frankfurt 1982.
- Marhol, M.: *Ion Exchangers in Analytical Chemistry*, Academia Prague, 1982, p. 277.