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Procedures for Db chemical characterization in off-line experiments

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Abstract

A new procedure to isolate carrier-free isotopes of Nb and Ta after a 12-MeV proton bombardment of a natural Hf foil has been considered. The cross section of the corresponding $^{nat}Hf(p,x)^{179}Ta$ reaction has been estimated for the first time as 5.4 ± 0.5 mb. The isotopes that were produced were subsequently used for development of procedures for the chemical characterization of element 105 (Db). HF/HNO_3 and HNO_3/H_2O_2 media have been chosen and the parameters of the experiments have been optimized. Based on the results obtained in this work, off-line experimental procedures have been proposed for the evaluation of the chemical behavior of Db.

Introduction

The discovery and investigation of superheavy elements (SHE) are difficult due to the very low production cross sections of these new nuclides, at the level of several picobarns. This results in the detection of about 1 atom per day or even more infrequently. The particular type of nuclear reaction used to produce these nuclides determines the magnitude of the cross section, which is a function of the combination of target and projectile nuclides. So called "warm" fusion reactions have played a significant role in the discovery and study of elements beyond 112 (Cn). In this reaction doubly magic ^{48}Ca ions are used to bombard actinide targets. This approach was successfully implemented to synthesize SHE 113 – 118 and to characterize the parent nuclei and their descendant nuclides. It should be noted that decay chains of odd-Z elements in the vicinity of the Island of Stability [1] consist

of several sequential alpha-decays terminated by spontaneous fission (SF), mostly of Db isotopes. The only exception that has been observed so far is the decay of ^{293}117, which leads to the SF of ^{281}Rg [2]. ^{268}Db was reported as part of the ^{288}115 decay chain with a half-life of about 1 day [3, 4]. This nuclide is sufficiently long-lived that the dubnium can be chemically characterized. The ^{243}Am + ^{48}Ca reaction is therefore a method for both the synthesis of elements 115 and 113 and for studying the chemical behavior of Db.

Two experiments devoted to the identification of elements 115 and 113 via registration of Db SF fragments have been reported in the literature [5, 6]. The procedures were based on isolation of Group V elements from an initial matrix using $La(OH)_3$ precipitation followed by ion exchange or reversed-phase chromatography [7, 8]. The precipitation is required to remove the bulk of the copper catcher material, to remove the Group VI – XIV elements, and to concentrate the elements of interest in the precipitate. Cation exchange was subsequently used to isolate the Group V elements from the trivalent actinides produced both from the recoil of target material and as transfer-reaction products. During this experiment 15 SF events were detected that could be attributed to decay products of ^{288}115 based on long-term spectrometric analysis of a final sample [9]. Within a second experiment designed to isolate Db, a LaF_3 precipitation followed by either anion or extraction chromatography was used instead of cation exchange. In that experiment the isolation of the Group V elements was followed by the separation of Nb and Ta into separate fractions in order to characterize Db chemical properties. An additional 5 more SF events were detected, 2 in the Group V fractions and 3 in the Ta fractions, and the observed fission events might be considered as decay of element 105 [10]. Since two different chemical solutions were used to elute dubnium homologues it cannot be absolutely determined whether or not Db exhibited Ta-like behavior. The only

conclusion that might be drawn is that the Db distribution coefficient under the given experimental conditions was higher than that of Nb. Pa-like behavior of Db could also be dismissed because Pa would have co-precipitated with the LaF_3. The radiochemical yields of Groups III – V and VII elements in the experiments mentioned above are given in Table 1. It can be seen that the average yields of the Group V elements are about 80 % and that the scandium activity in the second experiment is two orders of magnitude lower than in the first one.

Table 1

Average yields of radionuclides, in %, for the indicated experiments (see text)

Isotope	**Experiment I**	**Experiment II**				
	IV/V-fraction	**LaF_3**	**IV-fraction**	**V-fraction**	**Nb-fraction**	**Ta-fraction**
Nb-92m	80 ± 2	1.0 ± 0.3[a]	0.26 ± 0.04[a]	86 ± 6	84 ± 2	0.11 ± 0.04
Ta-177	57 ± 19	–	–	93 ± 7	–	83 ± 5
Hf-175	64 ± 2	–	56 ± 4	1.70 ± 0.06	–	0.08 ± 0.01
Zr-89	53 ± 3	68 ± 4	52 ± 2	0.48 ± 0.10	0.41 ± 0.06	0.07 ± 0.02
Sc-48	43.7 ± 1.2	75 ± 2	0.08 ± 0.02[b]	0.14 ± 0.02	0.07 ± 0.02	–
Y-87	0.74 ± 0.07	75 ± 2	0.38 ± 0.02	0.47 ± 0.03	–	0.06 ± 0.01
Rh-102m	5.2 ± 0.7[c]	4.4 ± 0.3[c]	0.10 ± 0.01	0.06 ± 0.01	0.22 ± 0.01	0.04 ± 0.01

a – data for Nb-90 b – data for Sc-46 c – data for Rh-101m

There were some disadvantages with these two procedures. In the first case, by-products such as isotopes of Zr, Sc, and Rh did not allow for immediate α-measurements of the prepared samples because of high β-activity. Moreover, the Group V elements were eluted along with the Group IV elements; therefore, spontaneous fission events might be attributed to ^{268}Rf produced via the ^{243}Am(^{48}Ca,3n)^{288}115 $\xrightarrow{5\alpha}$ ^{268}Db $\xrightarrow{\text{E.C.}}$ ^{268}Rf reaction, which has to be taken into account when confirming the original synthesis of elements 115 and 113. Additionally, the probability of the ^{243}Am(^{48}Ca,pxn)$^{290\text{-}x}$114 reaction occurring is very low [11], which likely excludes direct production of element 114 and its daughters. The observed results were in good agreement with expectations from the previously reported decay chains of element 115 produced at the Dubna Gas-

Filled Recoil Separator (DGFRS) [3]. In order to check an assumption that dubnium undergoes electron capture prior to the SF, separation of Group IV and V elements is required, which was the motivation for performing the second experiment. In terms of purification from actinides, LaF_3 precipitation might not be as efficient as the cation-exchange step. Moreover, an extra chemical step is needed to extract the Group IV elements from the lanthanum fluoride precipitate.

The goal of this work is to develop a relatively simple and fast procedure in order to characterize dubnium as a decay chain product of the decays of elements 115 and 113. As described above, a separation between Group IV and V elements is required.

Experimental

Carrier-free tracers of Nb and Ta radionuclides were produced by bombarding a metal ^{nat}Hf foil of 1.15 g and 99% purity at the 1 MV Tandem accelerator at the Center for Accelerator Mass Spectrometry (CAMS) at Lawrence Livermore National Laboratory (LLNL). The foil was irradiated for 10.5 h with 12 MeV protons on target at a current of 100 – 400 nA. After irradiation the foil was allowed to cool for 4.5 days and was then dissolved in conc. HNO_3 with several drops of conc. HF. The resulting solution was evaporated to dryness and reconstituted in a 6 M HF/4 M HCl matrix. The solution was loaded onto a 4x80 mm column of Dowex 1x4, -400 mesh resin that had been pretreated with the same acid mixture. The Hf was eluted from the column with 3 mL of 6 M HF/4 M HCl while the Group V elements were retained on the resin. Nb and Ta were then eluted from the column with 20 mL of 6 M HF/4 M HCl and 10 mL of 6 M HF/4 M HNO_3 respectively. The fractions were evaporated to dryness and reconstituted in 1 M HF prior to the model study.

Hf-175 was produced by irradiating a ^{nat}Lu foil for 12 hours with 12-14 MeV protons on target at CAMS. The Lu foil was dissolved in aqua regia. The solution was taken to dryness, then re-dissolved in conc. HCl and loaded onto a 4x40 mm column of AG 1X8, 100-200 mesh that had been pretreated with conc. HCl. About 30 mL of conc. HCl was passed through the column to remove Lu. The Hf was then eluted with 15 mL of 6 M HCl. The Hf was taken to dryness, and the procedure repeated.

The anion exchangers used for the separation studies were all in the Cl^--form and included AG 1x8, 100-200 mesh, Dowex 1x8, 200-400 mesh and Dowex 1x4, -400 mesh. The constant flow rate of eluents was chosen to be 0.25 mL min^{-1}. A standard High Purity Ge-detector with associated Maestro software was used to determine an activity level of each isotope in the samples.

Results and Discussions

Production cross sections for proton-induced reactions in natural Hf are given

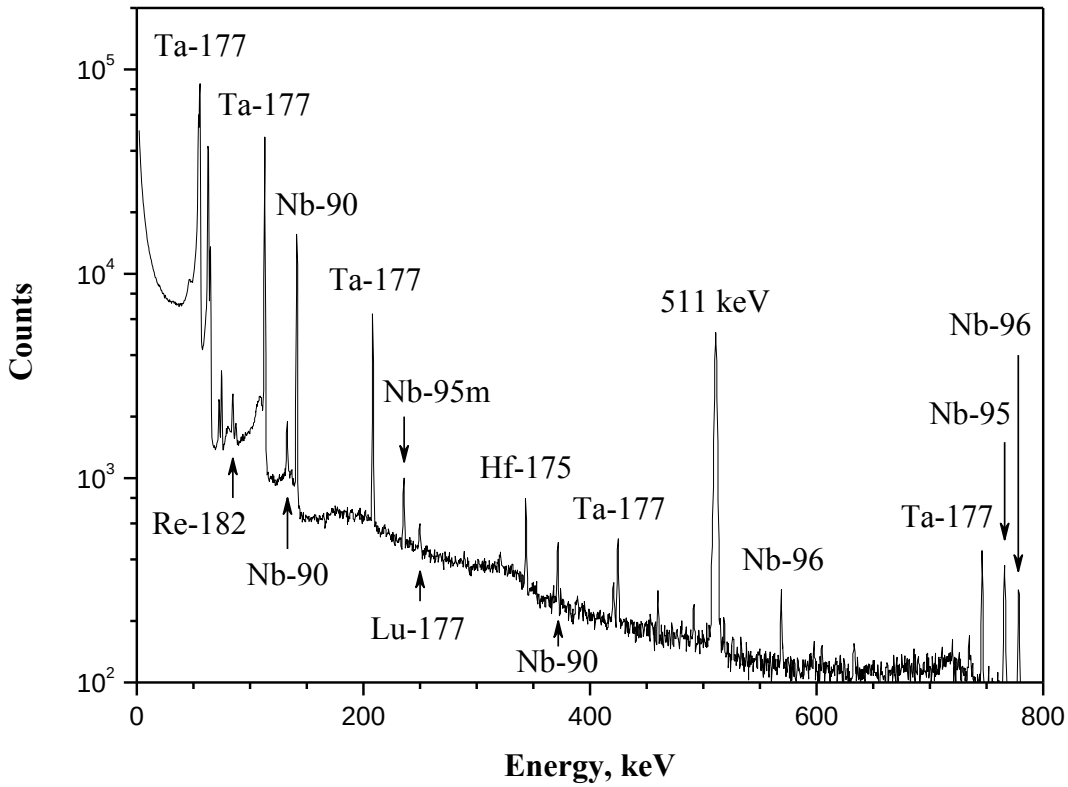

Fig. 1. Gamma-spectrum of an aliquot of dissolved Hf foil post-irradiation (Nb-92m is also present but isn't shown)

in Ref. [12]. The largest reported cross section is that of ^{177}Ta, which is the longest-lived Ta isotope considered in the paper. Under our experimental conditions we expected a cross section of 155 ± 13 mb for the ^{nat}Hf(p,x)^{177}Ta reaction. This value is in good agreement with the reported calculated value of 135 mb for the ^{177}Hf(p,n)^{177}Ta reaction [13], which is considered to be the primary reaction channel.

Figure 1 shows the gamma spectrum of an aliquot of the initial solution produced through dissolution of the irradiated Hf foil. Based on the resulting gamma spectrum, it was evident that the foil also contained ^{nat}Zr and ^{nat}W impurities. Proton irradiation of these elements leads to the production of a number of Nb [14 – 16] and Re [17] isotopes. Literature data indicate that under the given experimental conditions, the cross section observed for production of ^{90}Nb might be at least twice that of ^{177}Ta; therefore, detectable carrier-free amounts of other Group V elements might also be synthesized via proton irradiation of the Hf foil.

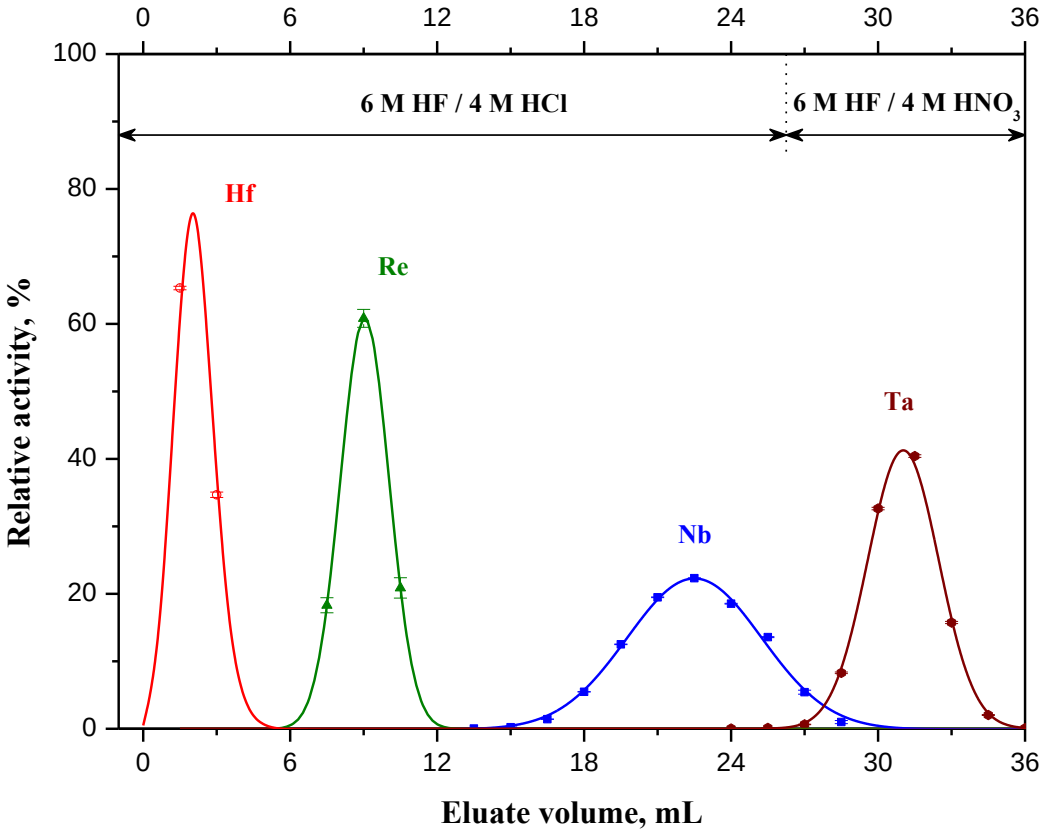

Fig. 2. Separation of Hf, Re, Nb and Ta on a 4x80 mm anion exchange column with Dowex 1x4, -400 mesh resin

The procedure used for isolation of niobium and tantalum from the irradiated Hf foil was based on the results reported in Ref. [18]. Figure 2 shows that in the 6 M HF/4 M HCl medium isocratic separation of Hf, Re, and Nb was successfully achieved and was in good agreement with the data reported in Refs. [19, 20]. The gamma spectrum of the Re fraction is shown in Figure 3.

Further analysis of the Ta fraction reveals the presence of ^{179}Ta, which was detectable only after the $^{175\text{-}178}$Ta decayed out. There are no gamma lines associated with ^{179}Ta decay, so only the Hf X-ray photons could be used for detection [21]. Through detection of the X-rays, the ^{179}Ta cross section from the ^{nat}Hf(p,x)^{179}Ta reaction was estimated to be 5.4 ± 0.5 mb. The given cross section estimate was based on the ratio of the ^{177}Ta and ^{179}Ta activities, which took the time since the end of bombardment and the known cross section of ^{177}Ta into account [12]. Figure 4 reflects excitation functions for the reactions ^{nat}Hf(p,x)$^{175\text{-}179}$Ta as a function of proton energy and includes our measurement of ^{179}Ta production at 12 MeV.

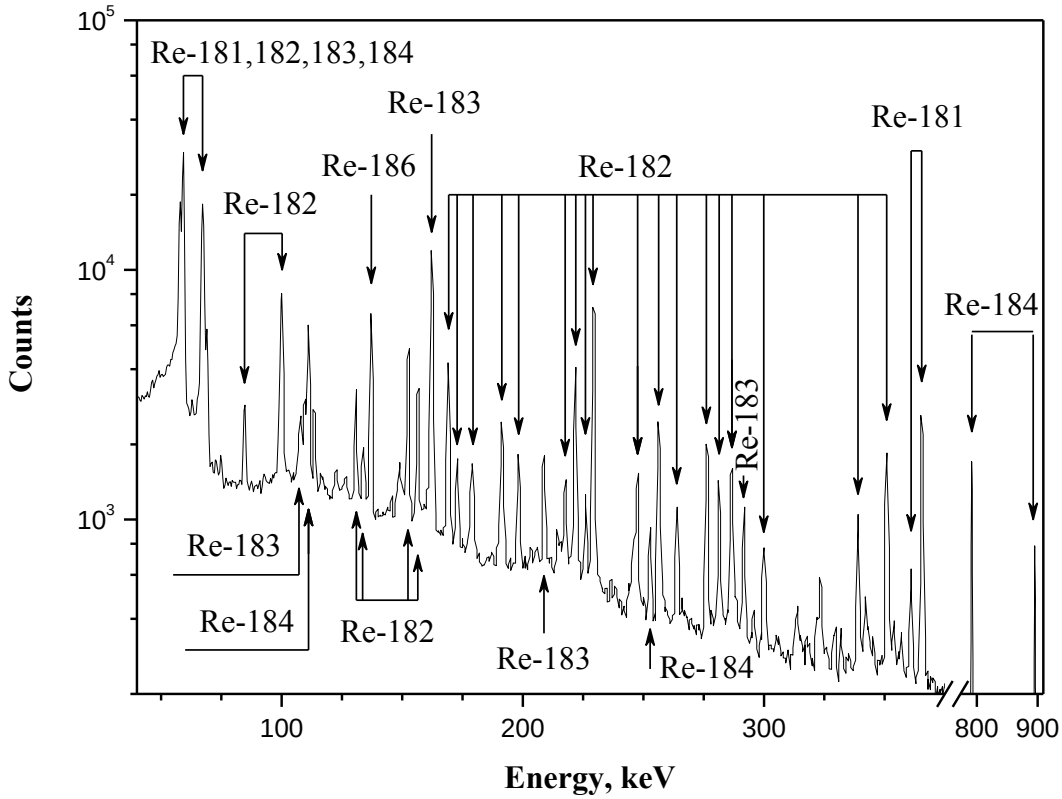

Fig. 3. Gamma-spectrum of Re-fraction

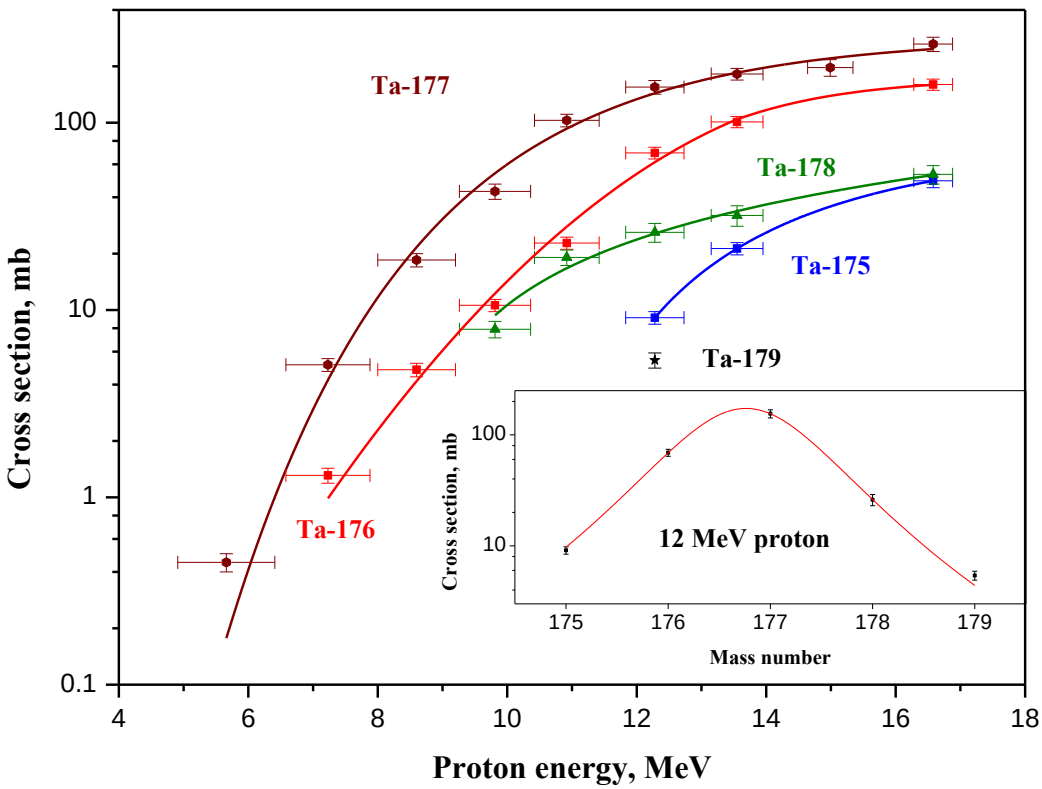

Fig. 4. Cross section of the reaction $^{nat}Hf(p,x)^{175-179}Ta$ (data for $^{175-178}Ta$ are from Ref. [12])

Chemical characterization of Db starts with the isolation of Nb and Ta together via separation from Group IV elements. In order to obtain a large total efficiency for an experiment the separation factor between Group IV and V elements must be as high as possible while minimizing the volume of each fraction in order to facilitate the fast preparation of a counting sample. Based on reported data of distribution coefficients (K_d) [18, 22] for the elements mentioned above in HF/HNO_3 media, the 11 M HF/7.7 M HNO_3 matrix was chosen to achieve the goal of separating the Group IV elements from a combined Nb and Ta fraction. Under these conditions the K_d values of Nb and Ta are relatively small and close to each other, which allows them to be eluted together in a small fractional volume. Distribution coefficients of Group IV elements are always less than those of Nb and Ta under considered conditions, so Hf and Zr will be eluted from the column in a fraction before the Group V elements.

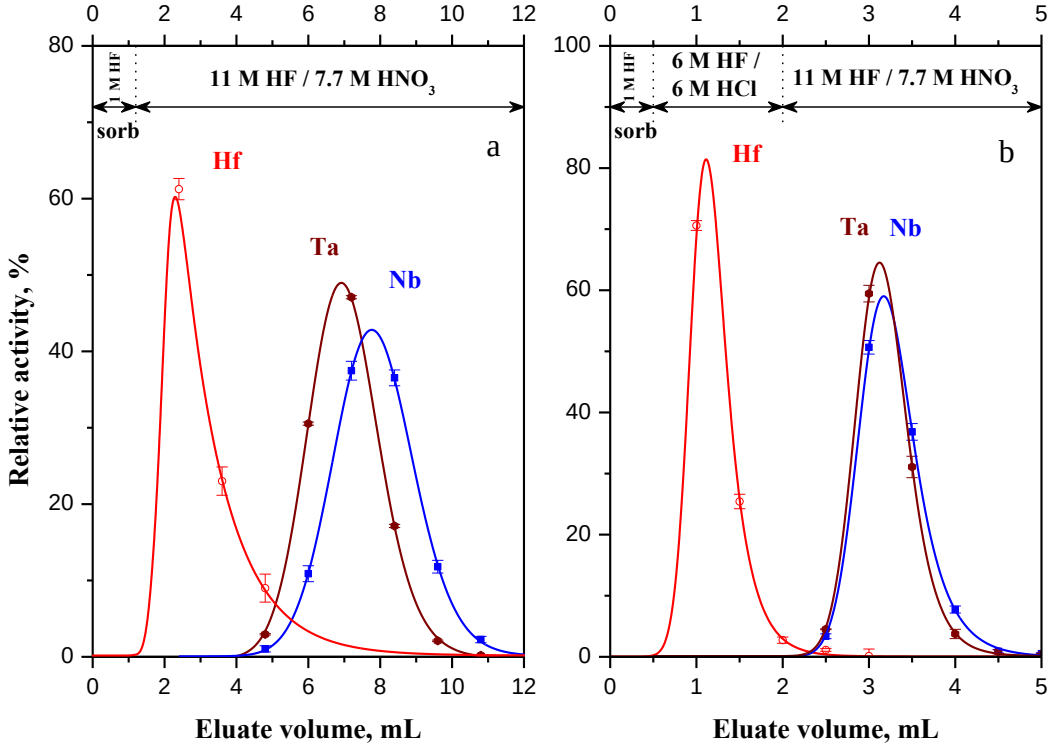

Fig. 5. Separation of Hf, Nb and Ta:

a – AG 1x8, 100-200 mesh, 4.7x57 mm, b – Dowex 1x4, -400 mesh, 4x16 mm

Figure 5 shows the resulting Hf and Nb/Ta elution curves. One can see that niobium and tantalum were eluted together and there is an isocratic separation of the Group IV and V elements (Fig. 5a). In order to reduce eluant fraction volumes, Dowex 1x4, -400 mesh resin and a smaller column size were used. Also, a stepwise elution was applied to decrease the overlap between the Group IV and V elution peaks. The Hf was eluted in a 1.5 mL fraction and the Nb/Ta fraction was eluted in about 3 mL.

Unfortunately, the 11 M HF/7.7 M HNO_3 matrix is highly aggressive and destroys the anion exchange resin almost immediately. One can see this by coloration of the eluate. While evaporating these fractions, organic residues might therefore be expected to be present on the sample surface, leading to possible deterioration of the alpha-spectra resolution and loss of fission-fragment energy. In order to optimize separation of the Group IV and V elements another HF/HNO_3

acid concentration ratio was considered. Analysis of 3D diagrams of Nb and Ta distribution coefficients [18] indicates several concentration regions where the K_d values are close to each other and relatively small. Among these the region of the least concentrated acids has been chosen. Figure 6 depicts Nb/Ta elution curves using 8 M HF/3 M HNO_3.

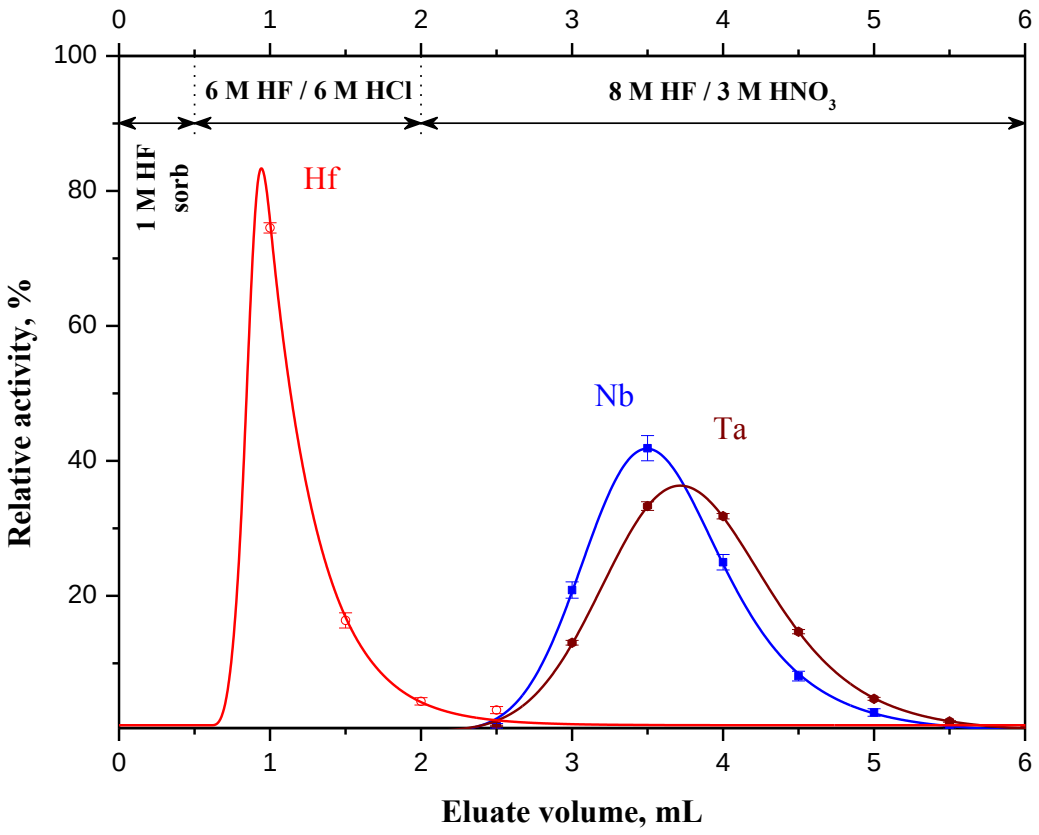

Fig. 6. Separation of Hf, Nb and Ta on 4x12 mm column with Dowex 1x4, -400 mesh resin

It is well known that Nb forms pure fluoride complexes only at [HF] > 10 M [23]. This is the main reason why Nb elutes after Ta under these conditions. At lower HF concentrations, formation of $[NbOF_n]^{3-n}$ and $[Nb(OH)_2F_n]^{3-n}$ species should be considered and the elution order of Nb and Ta will be changed. Figure 6 shows that Nb and Ta were eluted together and their K_d values were estimated correctly. Despite the fact that the volume of the fraction containing the Group V elements was slightly larger than what is shown on Fig. 5b, this matrix might be preferred for this separation because there is no noticeable anion-exchange resin

destruction and the last mL eluted from the column contained only about 1% of the total activity of each element.

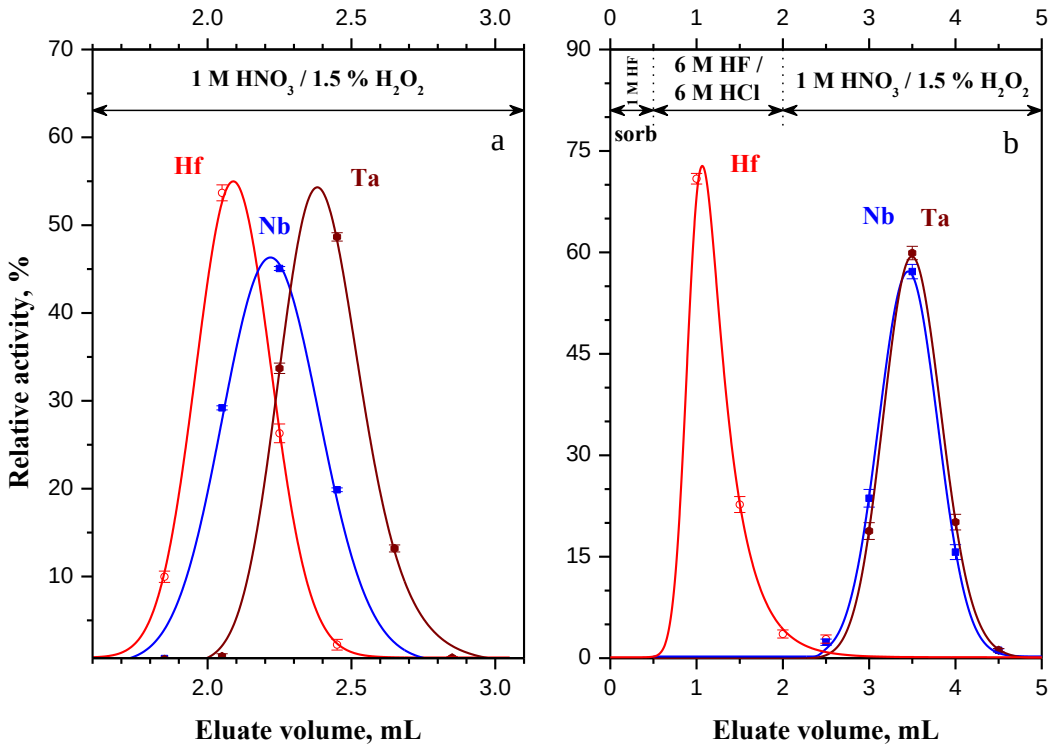

Fig. 7. Separation of Hf, Nb and Ta:

a – Dowex 1x8, 200-400 mesh, 6x10 mm, b – Dowex 1x4, -400 mesh, 4x16 mm

Distribution coefficients for Ta on anion-exchange resin in HF media are extremely high, which is why a nitric acid and hydrogen peroxide mixture is widely used to desorb it [24, 25]. Such an approach was applied in the experimental study of Db behavior [6] at the Flerov Laboratory for Nuclear Reactions (FLNR). Under these conditions Group V elements might be eluted simultaneously with the Group IV elements if preliminary chemical separations have not been implemented. Figure 7 reflects the behavior of Hf, Nb and Ta in a mixture of 1 M HNO_3/1.5% H_2O_2. Stepwise elution was again employed to separate Hf from Nb/Ta (Fig. 7b). The volume of each fraction was equal to those shown in Fig. 5b. Therefore, two independent chemical systems have been

developed for the investigation of the chemical properties of Db and its homologues.

The procedure for the entire chemical process for separation of any SHE liquid-phase study should include the following steps: collection of recoil atoms, removal of bulk amounts of the catcher material, separation and purification of the elements of interest, and final source preparation.

Two Db off-line experiments that have been discussed in the literature used Cu catchers where the recoiling reaction-product atoms were implanted into the surface layers of a Cu block. Lanthanum hydroxide precipitation was successfully employed to remove copper and Group VI – XIV elements and concentrate the Group III – V elements in the solid phase [7]. Afterwards a cation-exchange step with 1 M HF as the eluent was required to isolate trivalent lanthanides and actinides [26] from the Group IV and V elements. In Ref. [27] it was shown that Zr, Hf, Nb and Ta release from cation-exchange resin ($K_d < 2$) with eluant concentrations < 1 M HF, whereas alkaline, alkaline-earth and rare-earth elements remain on the resin. The eluant from the cation-exchange step is then loaded onto an anion-exchange column. The elements of interest in this system have sufficiently high K_d values to be adsorbed on Dowex 1x4 resin, but these values increase about 1.5-2 times for adsorption on Dowex 1x8 resin [28]. Choosing the anion-exchange procedures described in this paper to separate Group IV and V elements will result in a minimum number of chemical steps before preparation of a final counting source for measurement and an increase of yield. Moreover, it will help to reduce the time required to carry out the experiment and make it possible to automate the procedure.

Conclusion

Dowex 1x4 resin has been used to isolate Nb and Ta isotopes as part of a procedure to purify an irradiated Hf foil and for procedure development for future investigations of the chemical properties of Db. The procedure to separate Group V elements from macro amounts of Hf was successfully used for the isocratic anion exchange separation of Hf, Re and Nb. Analysis of the photons emitted by the Ta-fraction allowed us to estimate the production cross section of the 12 MeV proton induced reaction $^{nat}Hf(p,x)^{179}Ta$ at a value of 5.4 ± 0.5 mb.

Several procedures have been considered for the separation of Group IV from Group V elements. HF/HNO_3 and HNO_3/H_2O_2 media have been used to achieve the final goal. 11 M HF/7.7 M HNO_3 and 1 M HNO_3/1.5% H_2O_2 solutions show almost identical elution curves of Hf, Nb and Ta. Under the first set of conditions destruction of the resin might be observed and an alternative 8 M HF/3 M HNO_3 mixture has been implemented. This leads to a slight increase of the fraction volume of the Group V elements.

A combination of consecutive steps comprising an off-line experiment to measure the chemical properties of Db has been suggested. Lanthanum hydroxide precipitation is a well known matrix to remove copper and Group VI – XIV elements after irradiation. The next step is to use a cation-exchange resin column to separate Group IV and V elements from Group I – III elements. Two parallel anion-exchange procedures have been proposed to quickly separate Group IV and V elements. Therefore, a new off-line experiment might be carried out to chemically characterize Db properties in order to both independently confirm the synthesis of elements 115 and 113 and study dubnium chemical behavior.

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References

1. Oganessian, Yu. Ts.: Synthesis of the heaviest elements in ^{48}Ca-induced reactions. Radiochim. Acta **99**, 429 (2011).
2. Oganessian, Yu. Ts., Abdullin, F. Sh., Bailey, P. D., Benker, D. E., Bennett, M. E., Dmitriev, S. N., Ezold, J. G., Hamilton, J. H., Henderson, R. A., Itkis, M. G., Lobanov, Yu. V., Mezentsev, A. N., Moody, K. J., Nelson, S. L., Polyakov, A. N., Porter, C. E., Ramayya, A. V., Riley, F. D., Roberto, J. B., Ryabinin, M. A., Rykaczewski, K. P., Sagaidak, R. N., Shaughnessy, D. A., Shirokovsky, I. V., Stoyer, M. A., Subbotin, V. G., Sudowe, R., Sukhov, A. M., Taylor, R., Tsyganov, Yu. S., Utyonkov, V. K., Voinov, A. A., Vostokin, G. K., and Wilk, P. A.: Eleven new heaviest isotopes of elements Z = 105 to Z = 117 identified among the products of ^{249}Bk+^{48}Ca reactions. Phys. Rev. C **83**, 054315 (2011).
3. Oganessian, Yu. Ts., Utyonkov, V. K., Lobanov, Yu. V., Abdullin, F. Sh., Polyakov, A. N., Shirokovsky, I. V., Tsyganov, Yu. S., Gulbekian, G. G., Bogomolov, S. L., Mezentsev, A. N., Iliev, S., Subbotin, V. G., Sukhov, A. M., Voinov, A. A., Buklanov, G. V., Subotic, K., Zagrebaev, V. I., Itkis, M. G., Patin, J. B., Moody, K. J., Wild, J. F., Stoyer, M. A., Stoyer, N. J., Shaughnessy, D. A., Kenneally, J. M., and Lougheed, R. W.: Experiments on the synthesis of element 115 in the reaction ^{243}Am(^{48}Ca, xn)$^{291-x}$115. Phys. Rev. C **69**, 021601(R) (2004).
4. Oganessian, Yu. Ts., Utyonkov, V. K., Dmitriev, S. N., Lobanov, Yu. V., Itkis, M. G., Polyakov, A. N., Tsyganov, Yu. S., Mezentsev, A. N., Yeremin, A. V., Voinov, A. A., Sokol, E. A., Gulbekian, G. G., Bogomolov, S. L., Iliev, S., Subbotin, V. G.,

Sukhov, A. M., Buklanov, G. V., Shishkin, S. V., Chepygin, V. I., Vostokin, G. K., Aksenov, N. V., Hussonnois, M., Subotic, K., Zagrebaev, V. I., Moody, K. J., Patin, J. B., Wild, J. F., Stoyer, M. A., Stoyer, N. J., Shaughnessy, D. A., Kenneally, J. M., Wilk, P. A., Lougheed, R. W., Gäggeler, H. W., Schumann, D., Bruchertseifer, H., and Eichler, R.: Synthesis of elements 115 and 113 in the reaction ^{243}Am + ^{48}Ca. Phys. Rev. C **72**, 034611 (2005).

5. Dmitriev, S. N., Oganessyan, Yu. Ts., Utyonkov, V. K., Shishkin, S. V., Yeremin, A. V., Lobanov, Yu. V., Tsyganov, Yu. S., Chepygin, V. I., Sokol, E. A., Vostokin, G. K., Aksenov, N. V., Hussonnois, M., Itkis, M. G., Gäggeler, H. W., Schumann, D., Bruchertseifer H., Eichler, R., Shaughnessy, D. A., Wilk, P. A., Kenneally, J. M., Stoyer, M. A. and Wild, J. F.: Chemical identification of dubnium as a decay product of element 115 produced in the reaction ^{48}Ca + ^{243}Am. Mendeleev Commun. **15**, 1 (2005).
6. Stoyer, N. J., Landrum, J. H., Wilk, P. A., Moody, K. J., Kenneally, J. M., Shaughnessy, D. A., Stoyer, M. A., Wild, J. F., Lougheed, R.W., Dmitriev, S. N., Oganessian, Yu. Ts., Shishkin, S. V., Aksenov, N. V., Tereshatov, E. E., Bozhikov, G. A., Vostokin, G. K., Utyonkov, V. K., and Yeremin, A. A.: Chemical identification of a long-lived isotope of dubnium, a descendant of element 115. Nucl. Phys. A **787**, 388c (2007).
7. Schumann, D., Bruchertseifer, H., Eichler, R., Eichler, B., Gäggeler, H. W., Dmitriev, S. N., Oganessian, Yu. Ts., Utyonkov, V. K., Shishkin, S. V., Yeremin, A. A., Lobanov, Yu. V., Tsyganov, Yu. S., Chepygin, V. I., Sokol, E. A., Vostokin, G. K., Aksenov, N. V., Hussonnois, M., and Itkis, M. G.: Chemical procedure applied for the identification of Rf/Db produced in the ^{48}Ca + ^{243}Am reaction. Radiochim. Acta **93**, 727 (2005).
8. Wilk, P. A., Landrum, J. H., Shaughnessy, D. A., Kenneally, J. M., Stoyer, N. J., Stoyer, M. A., Moody, K. J., Aksenov, N. V., Bozhikov, G. A., Tereshatov, E. E., Vostokin, G. K., Shishkin, S. V., Dmitriev, S. N.: Separation of group five elements by reversed-phase chromatography. J. Radioanal. Nucl. Chem. **275**, 651 (2008).
9. Dressler, R., Eichler, R., Schumann, D., Shishkin, S. V.: Long-term α- and spontaneous fission measurement of a Rf/Db sample chemically prepared in a ^{48}Ca on ^{243}Am experiment. Phys. Rev. C **79**, 054605 (2009).

10. Henderson, R. A., Moody, K. J.: ^{243}Am + ^{48}Ca: A second look at the Rf/Db data set. In: *LLNL internal report*, LLNL-TR-400981 (https://e-reports-ext.llnl.gov/pdf/357247.pdf) 2008.
11. Münzenberg, G.: Recent advances in the discovery of transuranium elements. Rep. Prog. Phys. **51**, 57 (1988).
12. Siiskonen, T., Huikari, J., Haavisto, T., Bergman, J., Heselius, S.-J., Lill, J.-O., Lönnroth, T., Peräjärvi, K., Vartti, V.-P.: Excitation functions for proton-induced reactions on natural hafnium: Production of ^{177}Lu for medical use. Nucl. Instr. Meth. Phys. Res. B **267**, 3500 (2009).
13. Krivan, V.: Instrumental multielement proton activation analysis of high-purity niobium using both γ-ray and X-ray spectrometry. Anal. Chem. **47**, 469 (1975).
14. Uddin, M. S., Khandaker, M. U., Kim, K. S., Lee, Y. S., Lee, M. W., Kim, G. N.: Excitation functions of the proton induced nuclear reactions on natural zirconium. Nucl. Instr. Meth. Phys. Res. B **266**, 13 (2008).
15. Khandaker, M. U., Kim, K., Lee, M. W., Kim, K. S., Kim, G. N., Cho, Y. S., Lee, Y. O.: Experimental determination of proton-induced cross-sections on natural zirconium. Appl. Rad. Is. **67**, 1341 (2009).
16. Maiti, M., Lahiri S.: Separation of no-carrier-added ^{90}Nb from proton induced natural zirconium target. J. Radioanal. Nucl. Chem. **283**, 637 (2010).
17. Tárkányi, F., Takács, S., Szelecsényi, F., Ditrói, F., Hermanne, A., Sonck, M.: Excitation functions of proton induced nuclear reactions on natural tungsten up to 34 MeV. Nucl. Instr. Meth. Phys. Res. B **252**, 160 (2006).
18. Tereshatov, E. E., Bruchertseifer, H., Voronyuk, M. G., Starodub, G. Ya., Petrushkin, O. V., Dmitriev, S. N.: Isocratic anion exchange separations of Group V elements. J. Radioanal. Nucl. Chem. **286**, 9 (2010).
19. Kim, J. I., Lagally, H., Born, H.-J.: Ion exchange in aqueous and aqueous-organic solvents. Part I. Anion-exchange behaviour of Zr, Nb, Ta and Pa in aqueous HCl-HF and in HCl-HF-organic solvent. Anal. Chim. Acta **64**, 29 (1973).
20. Caletka, R., Krivan, V.: Anion-exchange behaviour of some elements in HF-HCl medium. J. Radioanal. Nucl. Chem. **142**, 373 (1990).
21. Firestone, R. B.: *Table of Isotopes*, 8th ed., Wiley, New York 1996, p. 225.

22. Faix, W. G., Caletka, R., Krivan, V.: Element distribution coefficients for hydrofluoric acid/nitric acid solutions and the anion exchange resin Dowex 1x8. Anal. Chem. **53**, 1719 (1981).
23. Caletka, R., Krivan, V.: Behaviour of 18 elements in HF and HF-NH_4F media on anion exchanger in various ionic forms. J. Radioanal. Nucl. Chem. **142**, 359 (1990).
24. Caletka, R., Hausbeck, R., Krivan, V.: Distribution of niobium and tantalum on Dowex-1 and polyurethane foam in HF-H_2SO_4 and HF-HCl medium. J. Radioanal. Nucl. Chem. **131**, 343 (1989).
25. Bruchertseifer, H., Gäggeler, H. W., Tereshatov, E. E., Aksenov, N. V., Shishkin, S. V., Bozhikov, G. A., Vostokin, G. K., Starodub, G. Y., Dmitriev, S. N.: Anion exchange separation of Pa, Nb and Ta in aqueous mineralic acid solutions. In: *PSI annual report 2006*. PSI, Villigen 2007, p. 8.
26. Guseva, L. I., Tikhomirova, G. S.: Ion exchange behavior and separation of actinides in different oxidation state on cation exchangers in dilute solutions of hydrofluoric acid. Radiochemistry **2**, 103 (1993).
27. Caletka, R., Krivan, V.: Cation-exchange of 43 elements from hydrofluoric acid solution. Talanta **30**, 543 (1983).
28. Marhol, M.: *Ion exchangers in analytical chemistry*. Part 1. Academia, Prague 1982, p. 264.