

# **RADIOCHEMICAL SEPARATION OF GROUP 5 ELEMENTS. MODEL EXPERIMENTS FOR INVESTIGATION OF DUBNIUM CHEMICAL BEHAVIOUR.**

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Chemical behaviour of group 5 elements in the aqueous hydrofluoric acid solutions was studied. The radiochemical method for the cation exchange separation of Nb (Pa) and Ta from Zr, Hf and lanthanides is presented. The opportunity for ion exchange separation of Zr and Hf is shown. The developed scheme allows excluding of the presence of SF heavy actinides in fractions of separated elements. On the basis of the data of the present work, it is possible to suggest the following order of the stability of the fluoride complexes of group 4 and 5 elements: Nb (Pa) > Zr > Hf > Ta. The order of the complex formation is in agreement with theoretical predictions. This analytical procedure can be used in future heavy nuclei synthesis experiments for the separation of dubnium (Db) from other reactions products and for its chemical identification.

## **1. Introduction**

During the last 7 years 17 isotopes of new chemical elements were synthesized in FLNR, JINR, Dubna. Due to relatively long half-lives of new isotopes it provides opportunities to study chemical properties of the heaviest known elements both in gas and liquid phases.

It is possible to synthesize superheavy elements in two ways: as a result of direct nuclear reaction or as a product of one or several consecutive  $\alpha$ -decays of a superheavy element. The second option of synthesis has given an opportunity of carrying out the chemical identification of Db as a decay product of element 115 [1]. Such way is an alternative option for the proof of synthesis of element 115 [2], and an option for synthesis of the longest living of known SHE isotopes  $^{268}\text{Db}$ .

Recently discovered at FLNR isotope  $^{268}\text{Db}$  with half-life  $T_{1/2} = 29$  h (fig. 1) provides excellent opportunity to study dubnium chemical behaviour in off-line

experiments. All previous experiments with Db dealt with short-lived ( $T_{1/2} < 30$  s) isotopes and were carried out only in on-line experiments.

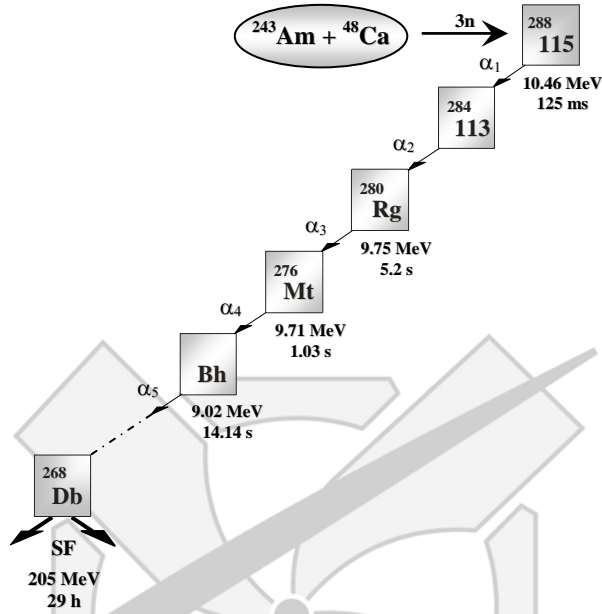


Figure 1. Decay of  $^{288}\text{115}$  observed with Dubna Gas-filled Recoil Separator in  $^{243}\text{Am} + ^{48}\text{Ca}$  reaction.

The aim of the present work was to develop a radioanalytical method for the separation of dubnium chemical homologues (Nb, Ta and Pa) and for isolation them from lanthanides (chemical homologues of actinides). The study of chemical properties of groups 4 and 5 elements was carried out by cation exchange using dilute hydrofluoric acid (HF) solutions.

## 2. Experimental

To develop the chemical separation procedures, the carrier-free tracers (see Table 1) were produced via  $(\alpha, xn)$ , spallation or  $(n, \gamma)$  reactions using the cyclotrons (U200, Synchrotron «F») and the Microtron MT-25 at JINR in Dubna.

Table 1. The brief description of radioisotopes used in work.

Isotope	Synthesis reaction	$T_{1/2}$
$^{233}\text{Pa}$	$^{232}\text{Th} (n; \gamma) ^{233}\text{Th} \rightarrow ^{233}\text{Pa}$	27,0 d
$^{92\text{m}}\text{Nb}$	$^{\text{nat}}\text{Y} (\alpha; xn) ^{92\text{m}}\text{Nb}$	10,15 d
$^{177\text{r}}\text{Ta}$	$^{\text{nat}}\text{Lu} (\alpha; xn) ^{177\text{r}}\text{Ta}$	56,6 h
$^{88}\text{Zr}$	$^{\text{nat}}\text{Sr} (\alpha; xn) ^{88}\text{Zr}$	83,4 d
$^{175}\text{Hf}$	$^{\text{nat}}\text{Yb} (\alpha; xn) ^{175}\text{Hf}$	70 d
$^{87\text{m}}\text{Sr}$	$^{\text{nat}}\text{Rb} (\alpha; xn) ^{87}\text{Y} \rightarrow ^{87\text{m}}\text{Sr}$	2,81 h
$^{173}\text{Lu}$	$^{\text{nat}}\text{Ta} (p; \text{spallation}) ^{173}\text{Lu}$	1,37 y

These radioactive isotopes were mixed with  $1\mu\text{g}$  of carrier of the corresponding element and  $1\text{mg}$  of La (in form of  $\text{La}(\text{NO}_3)_3$ ) collected in a Teflon cup, dissolved in a few ml of conc.  $\text{HNO}_3/\text{HCl}$  and evaporated. The precipitate was dissolved in  $0.5\text{ ml}$  of  $0.2\text{ M HCl}$  and adsorbed on a DOWEX 50x8 column ( $6\times 30\text{ mm}$ , 100-200 mesh). Precisely separated element fractions were eluted exclusively with easy evaporable strong mineral acids of different types and concentrations. Their concentrations were finally determined by the  $\gamma$ -activity measurements of the corresponding tracers with a standard spectrometer.

### 3. Results

The chromatogram of separation of groups 4 (Zr and Hf) and 5 (Nb and Ta) elements is shown in figure 2. Besides the above mentioned isotopes  $^{87\text{m}}\text{Sr}$  and  $^{173}\text{Lu}$  were added into the load solution. The former is a chemical analog of nobelium No and the latter is the heaviest lanthanide (actinides analog). Nobelium can be produced as following:  $\text{Db} \xrightarrow{\text{e.c.}} \text{Rf} \xrightarrow{\alpha} \text{No}$ .

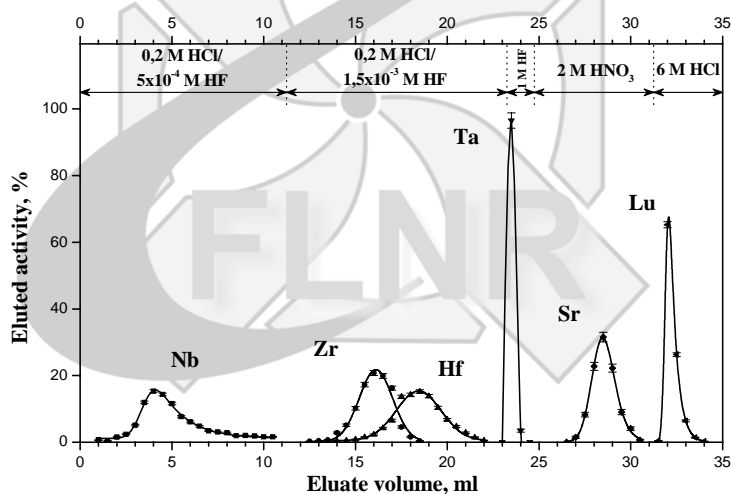


Figure 2. Separation of Nb, Zr, Hf, Ta, Sr and Lu by cation exchange (Dowex 50x8, 100-200 mesh,  $6\times 30\text{ mm}$ ).

Niobium was eluted with  $0.2\text{ M HCl}/5 \cdot 10^{-4}\text{ M HF}$  mixture before the group 4 elements which were eluted with  $0.2\text{ M HCl}/1.5 \cdot 10^{-3}\text{ M HF}$ . Tantalum was eluted with  $1\text{ M HF}$ , strontium and lutetium were desorbed with  $2\text{ M HNO}_3$  and  $6\text{ M HCl}$  respectively. The separation of Nb and Ta means that chemical conditions were optimal. Also the ion exchange separation of Zr and Hf is possible but it's necessary to change geometric parameters of a column and ion exchange

resin. The behaviour of separated elements confirms the results reported in work [3].

It is known, that No is eluted from Dowex-50 ion exchange resin before all lanthanides [4]. Nevertheless, it is shown that it is possible to carry out full separation of groups 4 and 5 elements from Sr (No analogue) and Lu (heaviest actinides analogue). Thus, the presence of heavy actinides in the previous fractions is excluded.

The figure 3 shows the chemical behaviour of Pa, pseudo-homologue of Db, in the dilute hydrofluoric acid media. Protactinium was desorbed with 0.2 M HCl/ $7.5 \cdot 10^{-4}$  M HF mixture, the elements of group 4 were eluted with 0.2 M HCl/ $5 \cdot 10^{-3}$  M HF and the following concentrations of solutions were as mentioned above.

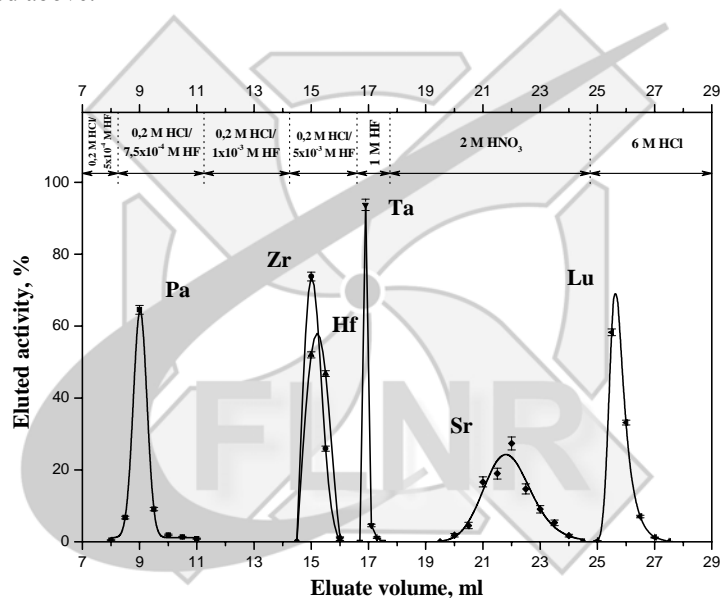


Figure 3. Separation of Pa, Zr, Hf, Ta, Sr and Lu by cation exchange (Dowex 50x8, 100-200 mesh, 6x30 mm).

#### 4. Conclusion

In the present work the opportunity for the cation exchange separation of group 5 elements and isolation them from the group 4 elements and lanthanides is shown. Radioanalytical procedure allows achieving separation of pairs of the elements Nb/Ta and Pa/Ta.

On the basis of the data of the present work, it is possible to suggest the following order of the stability of the fluoride complexes of groups 4 and 5 ele-

ments: Nb (Pa) > Zr > Hf > Ta. The behaviour of the elements is in agreement with earlier investigations and theoretical predictions [5, 6].

The usage of the proposed analytical method allows to selectively separate group 5 elements from Rf analogues and heavy actinides as well as to perform nuclear physical analysis of several fractions obtained during the study of Db chemical properties.

### Acknowledgement

The authors express their gratitude to Shishkina T.V. for supporting the radiochemical separations and Piperov Ts.N. for assistance with theoretical calculations. This work was supported by the Russian Foundation for Basic Research (grant no. 04-03-32047).

### References

1. Dmitriev S.N., Oganessian Yu.Ts., Utyonkov V.K., Shishkin S.V., Yereimin 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., Wild J.F. *Mendeleev Communications*, vol. 15, p. 1 (2005).
2. Oganessian Yu.Ts., Utyonkov V.K., Dmitriev S.N., Lobanov Yu.V., Itkis M.G., Polyakov A.N., Tsyganov Yu.S., Mezentsev A.N., Yereimin 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., Loughheed R.W., Gäggeler H.W., Schumann D., Bruchertseifer H., Eichler R. *Phys. Rev. C* 72, 034611 (2005).
3. Bruchertseifer H., Heller W., Haberberger F. et al. *GSI Scientific Report* 1990, GSI 91-1, 265 (1991).
4. Kratz J.J., Seaborg G.T. and Morse L.R. (Eds.), *The Chemistry Of The Actinide Elements*, 2nd ed., Chapman and Hall, London 1986 (2 volumes).
5. Pershina V. *Radiochim. Acta* 80, 75 (1998).
6. Pershina V., Bastug T. *Radiochim. Acta* 84, 79 (1999).