Radiochemical Separation of Group 5 Elements. 
Model Experiments for Investigation of Dubnium Chemical Behaviour.


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Abstract. 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 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.

Keywords: niobium, tantalum, protactinium, fluoride complexes.

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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 α-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 and 113 [2], and an option for synthesis of the longest living of known SHE isotopes 268Db.

Recently discovered at FLNR isotope 268Db 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 ($T_{1/2} < 40$ s) isotopes and were carried out only in on-line experiments.

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 group 4 elements and 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.

**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>
<thead>
<tr>
<th>Isotope</th>
<th>Synthesis reaction</th>
<th>(T_{1/2})</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{233})Pa</td>
<td>(^{233})Th ((n; \gamma)) (^{233})Th (\rightarrow) (^{233})Pa</td>
<td>27.0 d</td>
</tr>
<tr>
<td>(^{92m})Nb</td>
<td>nat(^{88})Y ((\alpha; xn)) nat(^{92m})Nb</td>
<td>10.15 d</td>
</tr>
<tr>
<td>(^{177})Ta</td>
<td>nat(^{177})Lu ((\alpha; xn)) nat(^{177})Ta</td>
<td>56.6 h</td>
</tr>
<tr>
<td>(^{88})Zr</td>
<td>nat(^{88})Sr ((\alpha; xn)) nat(^{88})Zr</td>
<td>83.4 d</td>
</tr>
<tr>
<td>(^{175})Hf</td>
<td>nat(^{87})Y ((\alpha; xn)) nat(^{175})Hf</td>
<td>70 d</td>
</tr>
<tr>
<td>(^{87m})Sr</td>
<td>nat(^{87m})Rb ((\alpha; xn)) (^{87})Y (\rightarrow) nat(^{87m})Sr</td>
<td>2.81 h</td>
</tr>
<tr>
<td>(^{173})Lu</td>
<td>nat(^{173})Ta ((p; spallation)) nat(^{173})Lu</td>
<td>1.37 y</td>
</tr>
</tbody>
</table>

These radioactive isotopes were mixed with 1\(\mu\)g of carrier of the corresponding element and 1mg of La (in form of La(NO\(_3\))\(_3\)) collected in a Teflon cup, dissolved in a few ml of conc. HNO\(_3\)/HCl and evaporated. The precipitate was dissolved in 0.5 ml of 0.2 M HCl and adsorbed on a DOWEX 50x8 column (6x30 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.
RESULTS

The chromatogram of separation of Zr, Hf and Nb is shown in figure 2. Besides the above mentioned isotopes $^{173}$Lu was added into the load solution because it is the heaviest lanthanide (actinides analog).

![Chromatogram](image)

**FIGURE 2.** Separation of Nb, Zr, Hf and Lu by cation exchange (Dowex 50x8, 100-200 mesh, 6x30 mm).

Niobium was eluted with 0.2 M HCl/5·10^{-4} M HF mixture before the group 4 elements which were eluted with 0.2 M HCl/2·10^{-3} M HF. Lutetium was desorbed with 6 M HCl. It is shown that the ion exchange separation of Zr and Hf is possible. The behaviour of separated elements confirms the results reported in work [3].

The figure 3 shows the chemical behaviour of Pa, pseudo-homologue of Db, in the dilute hydrofluoric acid media. $^{87m}$Sr was added into the load solution because it is a chemical analog of nobelium No. Nobelium can be produced as following: $\text{Db} \xrightarrow{\beta^-} \text{Rf} \xrightarrow{\alpha} \text{No}$.

Protactinium was desorbed with 0.2 M HCl/7.5·10^{-4} M HF mixture, the elements of group 4 were eluted with 0.2 M HCl/5·10^{-3} M HF. Tantalum was eluted with 1 M HF, strontium and lutetium were desorbed with 2 M HNO$_3$ and 6 M HCl respectively.

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.
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 elements: 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.

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REFERENCES


