

# Sorption of Subgroup IV, V and VI Elements on Ion Exchangers from HCl/HF Solutions. Model Experiments for Chemical Studies of the Elements 105 and 106 in Aqueous Solution

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## Abstract

The distribution coefficients of Zr, Hf, Nb, Ta, Mo, W, and Pa on cation and anion exchange resins from mixed HCl/HF solutions were determined in batch experiments by radiometric measurements. From solutions of 0.05 to 0.1 M HCl with  $1.0 \times 10^{-3}$  M HF a separation of subgroup V and VI from subgroup IV elements is possible on DOWEX 50X8. This system is proposed for the fast on-line separation of the elements 105 and 106. In column experiments with cation exchangers W can be separated from Mo. This opens the possibility to determine the chemical behaviour of element 106 in comparison to the lighter homologs. Differences in the sorption behaviour of Ta, Nb and Pa on cation exchange resins from 0.1 M HCl/2.0  $\times 10^{-4}$  M HF solution should allow a statement as to which of these elements, element 105 has the highest similarity.

## 1. Introduction

Chemical studies of the elements 105 and 106 require an effective separation from the actinides and element 104 produced either directly in nuclear reactions or as a decay product. For the investigation of element 106, the separation from 105 is of minor importance because of its low production rate in the nuclear reaction aimed to produce element 106.

Due to relativistic effects, simple extrapolations within a subgroup often fail. Chemical experiments performed with 34-s  $^{262}\text{105}$  and 27-s  $^{263}\text{105}$  in the gas phase [1] as well as in solution showed that the chemi-

cal properties of element 105 are more similar to those of the lighter homolog Nb and the pseudo-homolog Pa [2–4] than to the direct homolog Ta [5–7].

Chemical investigations on element 106 have not yet been performed. This is due to the low cross section and the short half-life of 0.9-s  $^{263}\text{106}$  [8] – until very recently the longest-lived known isotope of element 106. With the discovery of  $^{265}\text{106}$  and  $^{266}\text{106}$  with expected half-lives of 2–30 s [9] chemical studies are expected to be more feasible.

For the development of chemical systems applicable in fast on-line experiments with transactinides, usually the lighter homologs and pseudo-homologs are used in model experiments. Very often chemical systems are chosen in which (i) all elements of the same subgroup show a common chemical behaviour to separate them from the other reaction products, and (ii) which can be modified in such a way that differences in the chemical behaviour of individual elements in a subgroup can be determined.

One of the methods used for fast separation of heavy elements in aqueous solution is ion exchange on cation and anion resins with various acids or complexing agents as eluents. Reviews on the sorption behaviour and complex formation of subgroup V and VI elements fluorides and chlorides as well as mixed complexes mainly deal with strong acid solutions (0.5–12 M HCl, 0.01–10 M HF) [10–13]. Mixed HCl/HF solutions in weak concentrations were proposed for ion exchange experiments with the elements 104 and 105 [14, 15]. In a previous study [16], it was

shown that Ta can be separated from Hf on the strongly acid cation exchange resin DOWEX 50WX8 using  $1.0 \times 10^{-3}$  M HF/0.05–0.1 M HCl solution. From this solution, Hf is sorbed on the resin, whereas Ta passes through. At HF concentrations higher than  $1.0 \times 10^{-3}$  M, Hf forms neutral and anionic fluoride complexes and the sorption decreases. Lanthanides as model elements for the trivalent actinides are known to be sorbed completely on the cation exchanger under these conditions [17].

From theoretical calculations on the electronic structure and redox potentials it is predicted that  $(106\text{O}_4)^{2-}$  will be the most stable ion in aqueous solution within subgroup VI [18]. Because W and Mo are known to form anionic complexes with chloride and fluoride [19, 20], it is expected that ion exchange from dilute HCl/HF solution should be applicable for the separation of element 106, too.

Therefore, we have studied the sorption behaviour of carrier-free Zr, Nb, Pa, W, Mo and low carrier Ta and Hf radionuclides on ion exchange resins from mixed HCl/HF solutions in detail. The aim was to develop chemical systems which are suitable for on-line separations and applicable to investigate the complex formation and chemical properties of the elements 105 and 106 produced with very low cross sections. First, studies under static conditions were carried out to get information on the general chemical behaviour under equilibrium conditions. This was followed by kinetic studies with Hf and Zr to show that their sorption on the cation exchanger is rapid enough for fast on-line experiments. Furthermore, column experiments were carried out with W and Mo.

## 2. Experimental

### Batch experiments

$^{89}\text{Zr}$  was obtained by irradiating  $\text{Y}_2\text{O}_3$  with protons at the Jülich compact cyclotron CV28 (Inst. f. Nuklearchemie, Forschungsanlage Jülich GmbH, Germany). After irradiation, the target was dissolved in 6 M HCl. From this solution,  $^{89}\text{Zr}$  was adsorbed on the anion exchanger DOWEX 1X8 and eluted with 4 M HCl.

$^{92\text{m}}\text{Nb}$  was obtained by bombarding metallic Y with 36 MeV  $\alpha$ -particles at the U-200 accelerator (Laboratory of Nuclear Reactions, JINR Dubna, Russia). Nb was separated by thermochromatography using air saturated with  $\text{SOCl}_2$  ( $T = 1000^\circ\text{C}$ ) and condensed on the surface of a quartz tube. Stock solutions were prepared by dissolving the carrier-free radionuclides in 0.1 M HCl/ $1.0 \times 10^{-3}$  M HF.

$^{233}\text{Pa}$  was produced by irradiating metallic thorium with neutrons at the IBR-2 reactor (Laboratory of Neutron Physics, JINR Dubna, Russia) for 20 days (neutron flux  $5 \times 10^{13}$   $\text{cm}^{-2}$   $\text{s}^{-1}$ ) and thorium nitrate with neutrons at the TRIGA Mainz reactor (Institut für Kernchemie, Mainz, Germany) for 6 hours (neutron flux  $7 \times 10^{11}$   $\text{n/cm}^2$   $\text{s}$ ). The metallic target was dissolved in conc.  $\text{HNO}_3$ /HF, evaporated to dryness and

fumed several times with conc.  $\text{HNO}_3$ . Boric acid was added to remove any remaining fluoride. For further purification, the activity was adsorbed on DOWEX 1X8 from 6 M  $\text{HNO}_3$  and washed with the same solution to remove Th and boric acid.  $^{233}\text{Pa}$  was then eluted with 1.5 M HF/3 M HCl and the eluate was diluted to a final concentration of  $5.0 \times 10^{-3}$  M HCl/ $1.5 \times 10^{-4}$  M HF.

The irradiated thorium nitrate was dissolved in 9 M HCl. After sorption of Pa on DOWEX 1X8 it was subsequently eluted with 4 M HCl [11, 17].

$^{99}\text{Mo}$  was obtained carrier-free by eluting a commercial  $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$  generator with 5 vol%  $\text{NH}_4\text{OH}$  solution. The eluate was evaporated to dryness and  $^{99}\text{Mo}$  was dissolved in 0.1 M HCl.

$^{182}\text{Ta}$  and  $^{181}\text{Hf}$  were produced by irradiating 1.0 mg metallic Ta and 1.7 mg  $^{180}\text{HfO}_2$ , respectively, at the IBR-2 reactor (Laboratory of Neutron Physics, JINR Dubna, Russia) for two weeks at a neutron flux of  $5 \times 10^{12}$   $\text{n/cm}^2$   $\text{s}$  yielding a specific activity of 100 MBq  $^{182}\text{Ta}$  per mg Ta. The Ta target was dissolved in a mixture of 2 M HF and 0.2 M  $\text{HNO}_3$ .  $\text{HfO}_2$  was dissolved in conc. HF, precipitated as a hydroxide and dissolved in a mixture of 1 M HF and 5 M HCl. The specific activity of  $^{181}\text{Hf}$  was 63 MBq/mg Hf. Details of the sample preparation are given in [16].

To produce  $^{178}\text{W}$ , metallic Ta was bombarded with 650 MeV protons at the phasotron "F" (Laboratory of Nuclear Problems, JINR Dubna). W was separated from the irradiated target by thermochromatography using moist air, condensed onto the surface of a thermochromatographic column [21] and subsequently dissolved in 0.5 M HCl.

Ion exchange batch studies were carried out by shaking 5 ml of the tracer solution with 200 mg of the ion exchange resin (DOWEX 50WX8 or DOWEX 1X8, 20–50 mesh) for one hour. The  $\gamma$ -activity of the aqueous solution was measured before and after the experiment using a Ge(Li) detector. The distribution coefficient  $D$  was calculated as follows [16]:

$$D = \frac{A_i V_s}{A_s m_i}$$

with  $A_i$  activity on the ion exchanger (Bq)  
 $A_s$  activity of the solution (Bq)  
 $m_i$  amount of ion exchanger (g)  
 $V_s$  volume of the solution (ml).

### Chromatographic experiments

Carrier-free W isotopes used for column experiments were obtained by bombarding natural Hf with 49 MeV  $\alpha$ -particles at the Philips cyclotron at PSI Villigen. The main product was  $^{177}\text{W}$ . In addition  $^{93\text{m}}\text{Mo}$  was produced from Zr impurities in the target material.

The recoiling atoms were transported from the production site to the chemistry apparatus with a (He)/KCl gas jet. All products were collected on glass filters for 1–2 hours and were washed from these filters with the HCl/HF solutions used in the column ex-

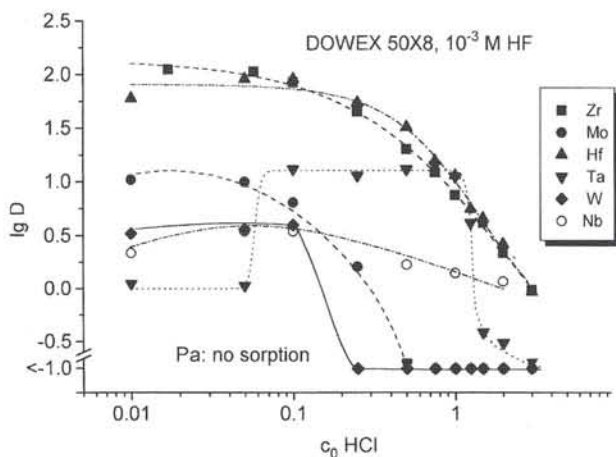


Fig. 1. Distribution coefficients of Zr, Hf, Nb, Ta, Mo, and W on DOWEX 50X8 in dependence on the HCl concentration at  $1.0 \times 10^{-3}$  M HF (data for Ta and Hf taken from [16]).

periments. The ion exchangers (column size  $3.50 \times 30$  mm, filled with 250 mg exchange resin DOWEX 50WX12, 100–200 mesh) were equilibrated with the corresponding solutions for 12–18 hours before the experiment started. The HCl/HF solution containing the W and Mo isotopes passed the column with flow rates of 0.3–4 ml/min. The  $\gamma$ -activity of the collected fractions (0.3 ml each) was measured with a HPGe detector.

### 3. Results and discussion

Sorption behaviour of Zr, Hf, Pa, Nb, Ta, W, and Mo as a function of the HCl concentration

#### *Distribution coefficients on the cation exchanger DOWEX 50WX8*

The distribution coefficients  $D$  of Zr, Hf, Nb, Ta, Mo, W, and Pa on DOWEX 50WX8 as a function of the HCl concentration in the presence of  $1.0 \times 10^{-3}$  M HF are given in Fig. 1. At concentrations lower than 0.5 M HCl, Zr and Hf are completely sorbed on the cation resin. Formation of cationic complexes of the form  $\text{MeF}^{3+}$ ,  $\text{MeF}_2^+$  or  $\text{MeF}_3^+$  can be assumed by calculating the range of existence of these species with complex stability constants known from literature [19, 20]. The respective chloride complexes of Hf and Zr are much less stable [19, 20] and can be neglected in this case, but species containing  $\text{OH}^-$  groups are also possible at HCl concentrations below 0.05 M because of the strong tendency of subgroup IV elements to hydrolyze (see below). With increasing HCl concentration the concentration of free ligand  $\text{F}^-$  decreases due to the dissociation constant of HF ( $\lg K_1 = 2.9$  [20]). Uncharged or anionic mixed chloride/fluoride complexes are formed which are not sorbed on the cation exchanger.

Nb shows low sorption on the cation exchange resin over the whole range of HCl concentrations in the

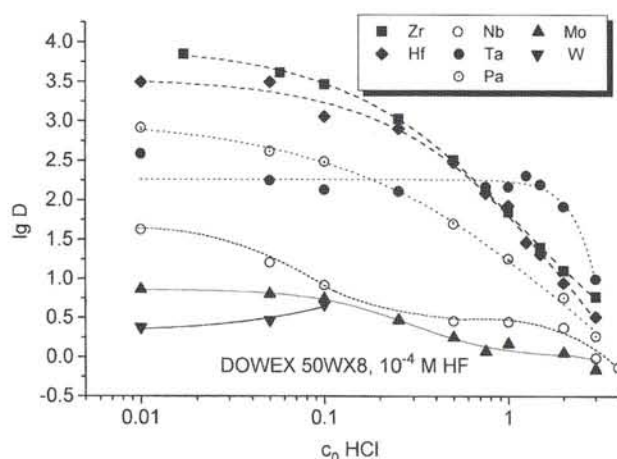


Fig. 2. Distribution coefficients for Zr, Hf, Nb, Ta, Pa, Mo and W on DOWEX 50X8 in dependence on the HCl concentration at  $1.0 \times 10^{-4}$  M HF.

presence of  $10^{-3}$  M HF ( $\lg D < 0.6$ ), Pa is not sorbed at all. This differs from the behaviour of Ta which shows a  $\lg D$  value of  $\sim 1.0$  in the interval of 0.1–2 M HCl. For Pa and Nb oxygen-containing anionic fluoride complexes are expected, while for Ta anionic species of the form  $\text{TaF}_{5+n}^-$  dominate. In the interval 0.1–1 M HCl, positively charged complexes of Ta are formed ( $\text{TaF}_4^+$ ,  $\text{TaF}_3^{2+}$  [22, 23]), which are adsorbed on DOWEX 50WX8. At HCl concentrations lower than 1 M, neutral chloride or mixed complexes are formed and the sorption on the cation exchanger decreases.

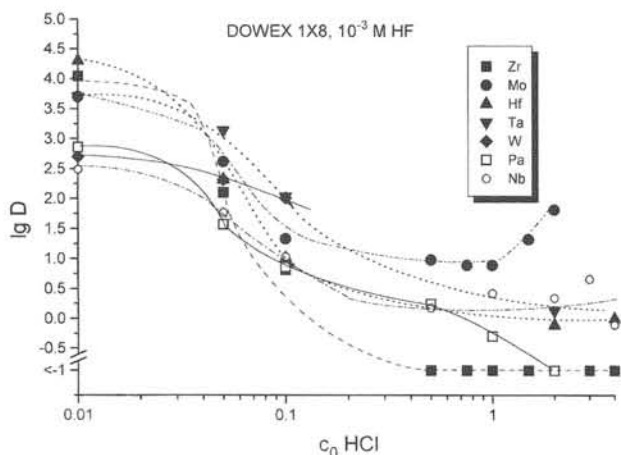
Mo and W show only slight sorption on DOWEX 50WX8. From this behaviour a separation of element 106 from 104 and actinide elements should be possible using concentrations  $< 0.5$  M HCl and  $10^{-3}$  M HF. Moreover, this chemical system should be suitable for the isolation of element 105 from trivalent actinides and element 104.

The sorption behaviour of the described elements as a function of the HCl concentration at  $1.0 \times 10^{-4}$  M HF is shown in Fig. 2. Up to a concentration of 0.1 M HCl, the tetra- and pentavalent elements remain on the cation exchanger (with exception of Nb), whereas W and Mo are not or only slightly sorbed. Consequently, separation of subgroup VI from IV and V elements should be possible on DOWEX 50WX8 (with exception of Nb) using solutions  $1.0 \times 10^{-4}$  M HF and HCl concentrations below 0.5 M.

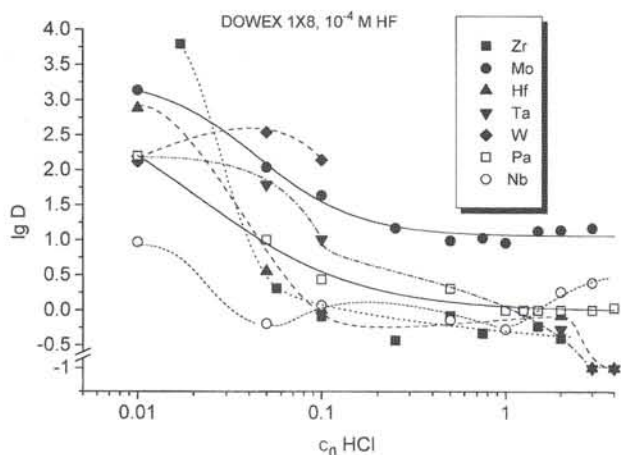
Adsorption on the vial surface, however, cannot be excluded under these conditions. Especially in lower acid solutions hydrolytic processes can take place. To distinguish between the chemical behaviour on the cation exchange resin and surface adsorption we have studied the sorption behaviour of the elements under investigation from the same solutions on an anion exchange resin.

#### *Distribution coefficients on the anion exchanger DOWEX 1X8*

Sorption experiments from HF concentrations of  $1.0 \times 10^{-3}$  M and  $1.0 \times 10^{-4}$  M on DOWEX 1X8



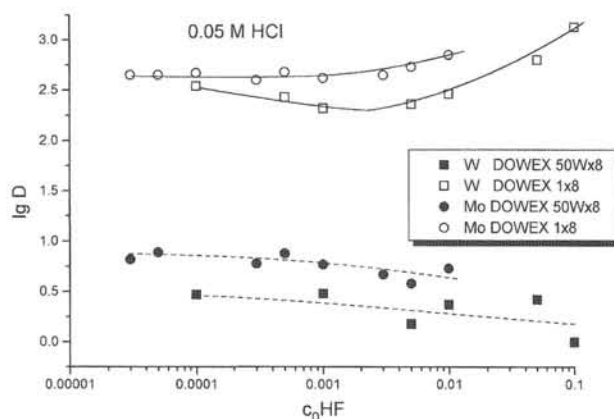
**Fig. 3.** Distribution coefficients of Zr, Hf, Nb, Ta, Mo and W on DOWEX 1X8 in dependence on the HCl concentration at  $1.0 \times 10^{-3}$  M HF.



**Fig. 4.** Distribution coefficients of Zr, Hf, Nb, Ta, Pa, Mo and W on DOWEX 1X8 in dependence on the HCl concentration at  $1.0 \times 10^{-4}$  M HF.

(Figs. 3 and 4) as a function of the HCl concentration showed that at HCl concentrations below 0.1 M W and Mo are nearly completely sorbed on the anion exchanger probably due to the formation of anionic oxo compounds. The sorption decreases at higher HCl concentration. Unfortunately, there are no reliable data on stability constants for mixed fluoride/chloride complexes of Mo and W, only data for pure chloride and fluoride complexes are published [20].

The tetra- and pentavalent elements are also strongly sorbed on the anion exchanger at HCl concentrations below 0.1 M, presumably mainly due to hydrolytic processes. Free pentavalent cationic species  $Me^{5+}$  are not stable in aqueous solution and  $Me^{4+}$  can be found only in very strong acid solutions [24]. The hydrolysis can be suppressed by strong complexing agents such as HF. If the concentration of the complexing agent decreases, then neutral hydroxo compounds are formed [24] which are easily adsorbed on surfaces. These processes are not reversible and can hardly be controlled. The strong sorption onto



**Fig. 5.** Distribution coefficients of Nb, Ta and Pa on DOWEX 50X8 and DOWEX 1X8 in dependence on the HF concentration at 0.1 M HCl.

DOWEX 1X8 at low HCl concentrations for elements which, under the same conditions, show large  $D$  values on DOWEX 50WX8 indicate the presence of hydrolytic products. Therefore, we consider only HCl concentrations of  $0.05 \text{ M} < c_{\text{HCl}} < 0.5 \text{ M}$  and HF concentrations of  $10^{-4} \text{ M} < c_{\text{HF}} < 10^{-3} \text{ M}$  as applicable for separation experiments.

#### Kinetic studies

Time dependent batch studies with Hf and Zr on DOWEX 50WX8 from 0.1 M HCl/ $1.0 \times 10^{-3}$  M HF showed that within 5 min sorption of these two elements on the cation exchanger is completed. However, in a previous study chromatographic experiments were carried out to obtain information on the kinetics for a contact time of a few seconds [25]. In these on-line experiments (U-400 cyclotron, Laboratory of Nuclear Reactions, JINR Dubna, Russia) the sorption behaviour of short-lived W, Ta, and Hf isotopes on DOWEX 50WX8 was studied using 0.05 M HCl/ $1.0 \times 10^{-3}$  M HF solution as an eluent at flow rates of 2–4 ml/min. W and Ta passed the cation exchange column while Hf and its daughter Lu remained nearly completely on DOWEX 50WX8 (decontamination factor  $2 \times 10^2$ ).

#### Sorption behaviour of W, Mo, Pa, Nb and Ta in dependence on the HF concentration

In Fig. 5, the  $\lg D$  values of Ta, Nb, and Pa on DOWEX 50WX8 and on DOWEX 1X8 are plotted as a function of the HF concentration at 0.1 M HCl. At HF concentrations below  $1.0 \times 10^{-3}$  M, cationic species of Pa are formed ( $\text{PaOF}_2^+$ ,  $\text{PaOF}_2^{2+}$  [26]) which are sorbed on DOWEX 50WX8. Nb shows only slight sorption on the cation exchange resin. With increasing HF concentration, the cationic species transform first into neutral and further into anionic fluoride complexes, the latter being sorbed on DOWEX 1X8. The strong sorption of Ta on the cation exchanger at low HF concentration is caused by hydrolysis. The increas-

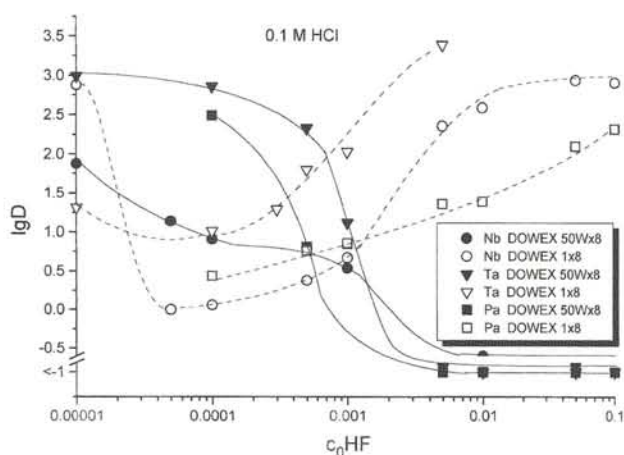


Fig. 6. Distribution coefficients of W and Mo on DOWEX 50X8 and DOWEX 1X8 in dependence on the HF concentration at 0.05 M HCl.

ing sorption on DOWEX 1X8 can again be seen as evidence for adsorption of hydrolysed species.

With 0.05 M HCl and HF concentrations higher than  $1.0 \times 10^{-3}$  M, Ta, Nb and Pa show no differences in the sorption behaviour between each other on DOWEX 50WX8 as well as on DOWEX 1X8. For all three elements anionic complexes are dominant. Hydrolysis dominates the fluoride complexing of Ta at HF concentrations lower than  $10^{-3}$  M because for both cation and anion exchange resins relatively high distribution coefficients are found.

Exploiting the differences in the sorption behaviour of subgroup V elements on DOWEX 50WX8 at concentrations of 0.1 M HCl/ $2.0 \times 10^{-4}$  M, HF should allow to find out whether the chemical properties of element 105 are more like Ta and Pa or like Nb.

Sorption experiments with 0.1 M HCl showed that the behaviour of W and Mo on DOWEX 50WX8 is similar and depends on slightly on the HF concentration. The sorption behaviour on DOWEX 50WX8 and DOWEX 1X8 for these two elements as a function of the HF concentration from 0.05 HCl is shown in Fig. 6. On DOWEX 1X8, W and Mo are completely sorbed between  $10^{-4}$  M and  $10^{-1}$  M HF due to the predominant formation of anionic oxo-compounds. Small amounts observed on the cation exchange resin may represent a small fraction of hydrolyzed species.

#### Chromatographic study

As can be seen from Fig. 1, Mo shows on the cation exchange resin a distribution coefficient about three times higher than that for W (Mo:  $D \approx 10$  and W:  $D \approx 3$ ) at HCl concentrations lower than 0.1 M and  $10^{-3}$  M HF. From these results differences on the cation exchanger in column experiments could be expected, but chromatographic studies with  $^{177}\text{W}$  and  $^{93\text{m}}\text{Mo}$  using 0.05 M–0.1 M HCl/ $1.0 \times 10^{-3}$  M HF and flow rates of 2–3 ml/min showed that both elements pass the cationic column (DOWEX 50X12) without sorption.

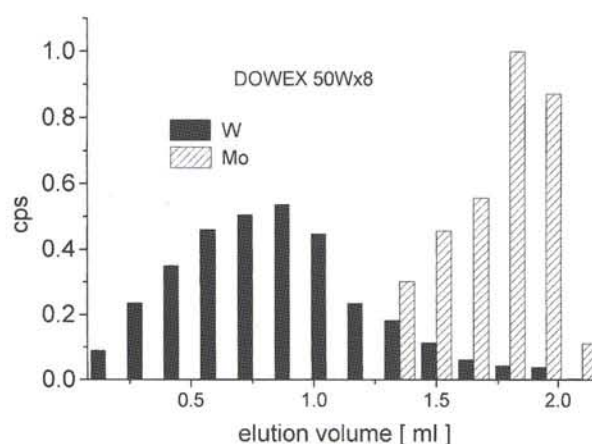


Fig. 7. Elution of W and Mo from the cationic column (DOWEX 50X12) using 0.05 M HCl/ $1.0 \times 10^{-3}$  M HF solution, flow rate 0.3 ml/min.

However, with a slow flow rate (0.3 ml/min, solution 0.05 M HCl/ $1.0 \times 10^{-3}$  M HF) a separation of W from Mo is possible, as can be seen from Fig. 7. W passes the column (DOWEX 50WX12, free column volume 0.08 ml) without sorption, Mo is found in the following fractions. These first results offer the possibility to develop chemical systems to determine the chemical behaviour of element 106 in a direct comparison with the lighter homologs Mo and W.

#### 4. Conclusions

1. A fast separation of hexavalent (Mo, W, element 106) and pentavalent elements (Nb, Ta, Pa, element 105) from tetravalent (Zr, Hf, element 104) and trivalent elements (heavy actinides) is achieved in 0.05–0.1 M HCl/ $1.0 \times 10^{-3}$  M HF on a cation exchange resin (e.g. DOWEX 50WX8). Subgroup 5 and 6 elements pass the column while tri- and tetravalent elements are sorbed.
2. A separation of subgroup 6 elements from elements of subgroup 4 and 3 is also possible from 0.1 M HCl/ $1.0 \times 10^{-4}$  M HF on a cation exchange resin (DOWEX 50WX8). An indication of an additional separation of at least some subgroup 5 elements (high  $D$  values for Ta, Pa and a lower one for Nb) provides promising aspects to further improve the purity of the subgroup 6 element fraction.
3. A solution of 0.1 M HCl/ $2.0 \times 10^{-4}$  M HF for sorption experiments on DOWEX 50WX8 may be suited to distinguish between a Nb-like or Ta/Pa-like behaviour of element 105. However, for a better interpretation of the results more information about the structure and stability constants of the lighter homologs is needed.
4. Mo and W can be separated chromatographically on a cation exchange column with 0.05 M HCl/ $1.0 \times 10^{-3}$  M HF at a flow rate of 0.3 ml/min. With further optimized parameters regarding the resin,

grain size, acid concentrations, column size etc., a separation of the individual subgroup 6 elements (Mo, W, element 106) should be possible in a time interval between 10 and 30 s.

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