

## Decay Properties of $^{265}\text{Sg}$ and $^{266}\text{Sg}$

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Using the On-Line Gas Chromatography Apparatus OLGA, the presently known most neutron-rich Sg isotopes,  $^{265}\text{Sg}$  and  $^{266}\text{Sg}$ , produced in the fusion reaction  $^{22}\text{Ne} + ^{248}\text{Cm}$ , were chemically isolated [1] and their nuclear decay properties measured. Final samples were assayed by  $\alpha$ -particle and spontaneous fission (SF) spectrometry using the ROTating wheel Multidetector Analyzer ROMA. Preliminary results of these experiments were already reported in previous annual reports [2,3]. In order to evaluate the statistical significance of the observed decay chains, a careful analysis of the data was performed. The analysis revealed a number of additional decay chains that could be attributed to the decay of  $^{265}\text{Sg}$ .

The nuclides  $^{265}\text{Sg}$  and  $^{266}\text{Sg}$  were identified by observing time correlated  $\alpha$ - $\alpha$ ( $\alpha$ ) and  $\alpha$ -SF decay chains. With a total beam dose of  $7.5 \times 10^{17}$   $^{22}\text{Ne}$  beam particles, 13 correlated decay chains of  $^{265}\text{Sg}$  (with an estimated number of 2.8 random correlations) and 3 decay chains of  $^{266}\text{Sg}$  (0.6 random correlations) were identified. Deduced decay properties were  $T_{1/2} = 7.4^{+2.3}_{-2.7}$  s and  $E_{\alpha} = 8.69$  MeV (8%), 8.76 MeV (23%), 8.84 MeV (46%) and 8.94 MeV (23%) for  $^{265}\text{Sg}$ ; and  $T_{1/2} = 21^{+30}_{-12}$  s and  $E_{\alpha} = 8.52$  MeV (33%), 8.77 MeV (66%) for  $^{266}\text{Sg}$ . The half-life value of  $^{265}\text{Sg}$  is in good agreement with the half-life determined from two  $^{265}\text{Sg}$  decays observed in the decay chain of  $^{277}\text{112}$  [4]. The  $\alpha$ -lines are in good agreement with  $\alpha$ -particle energies measured in the discovery experiment by Lazarev et al. [5], and with the  $\alpha$ -particle energy measured in one  $^{277}\text{112}$  decay chain [4]. The half-life of  $^{266}\text{Sg}$  has been measured for the first time and is in good agreement with theoretical predictions [6]. Similar  $\alpha$ -particle energies were observed in the discovery experiment [4]. Upper limits for SF of  $\leq 35\%$  and  $\leq 82\%$  were established for  $^{265}\text{Sg}$  and  $^{266}\text{Sg}$ , respectively. The resulting partial SF half-lives are  $T_{1/2}^{\text{SF}}(^{265}\text{Sg}) \geq 13$  s and  $T_{1/2}^{\text{SF}}(^{266}\text{Sg}) \geq 11$  s. All upper or lower limits are given with a 16% error probability. While the lower limit for the partial SF half-life of  $^{266}\text{Sg}$  is not significantly longer than the value estimated in the discovery experiment [5], or in Ref. [7], a lower limit for the partial SF half-life of  $^{265}\text{Sg}$  has been established for the first time in this work. Using the upper and lower error limits of the half-lives of  $^{265}\text{Sg}$  and  $^{266}\text{Sg}$ , the partial  $\alpha$ -decay half-lives are between  $T_{1/2}^{\alpha}(^{265}\text{Sg}) = 4.7 - 16.5$  s (68% c.i.) and  $T_{1/2}^{\alpha}(^{266}\text{Sg}) = 9 - 228$  s (68% c.i.). An accurate value of the partial SF half-life of  $^{266}\text{Sg}$  is of special interest for the test of theoretical models. Smolanczuk et al., [8] predicted an enhanced nuclear stability near the deformed shells  $N=162$  and  $Z=108$  against SF. In Fig. 1 the experimentally determined partial half-lives for  $\alpha$ - and SF-decay for  $^{260}\text{Sg}$  [9,10] and  $^{266}\text{Sg}$  (this work) are compared with theoretical calculations [6,8]. Our data clearly confirms the predicted enhanced nuclear stability.

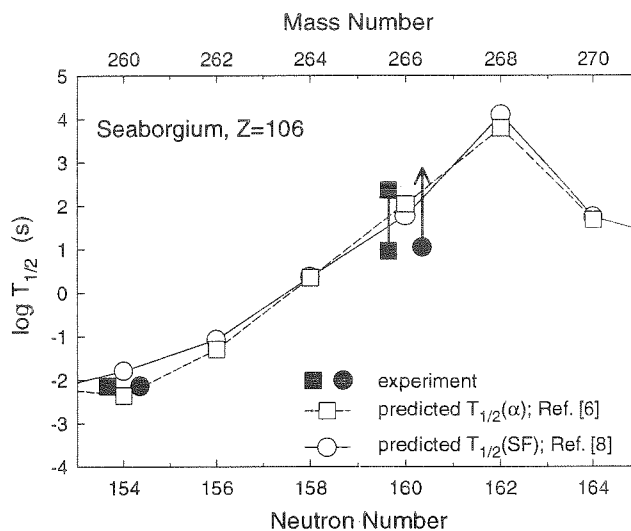


Fig.1 Predicted partial  $\alpha$ - and SF half-lives in comparison with experimental data [9,10] and this work.

In addition, also production cross sections were evaluated from our data. On the basis of yields that were typically observed in test experiments with W-isotopes, production cross sections of  $240^{+140}_{-70}$  pb and  $25^{+31}_{-9}$  pb for the production of  $\alpha$ -decaying  $^{265}\text{Sg}$  and  $^{266}\text{Sg}$  were deduced, respectively, at a  $^{22}\text{Ne}$  projectile energy between 120 and 124 MeV. The error limits account only for statistical uncertainties. In a direct catch experiment at a lower beam energy of  $119 \pm 1$  MeV, the products were transported with the gas-jet directly to a tape detection system. In this experiment a single  $\alpha$ -SF decay chain attributed to the decay of  $^{266}\text{Sg}$  was observed. A cross section of about 60 pb was calculated for this single event. All cross sections are in good agreement with values given by Lazarev et al. [5].

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## Extraction of Chloride Complexes of the Elements Nb, Ta, Pa, and 105 into an Aliphatic Amine

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Previous studies of the halide complex formation of element 105 and its extraction into triisooctyl amine (TIOA) were performed in HCl/HF mixtures [1,2]. Element 105 was shown to be extracted into the amine from 12 M HCl/0.02 M HF together with its lighter homologues Nb, Ta, and the pseudo-homologue Pa. In elutions with 10 M HCl/0.025 M HF, 4 M HCl/0.02 M HF, and 0.5 M HCl/0.01 M HF, the extraction sequence Ta > Nb > 105 > Pa was observed and element 105 behaved very differently from its closest homologue Ta. Theoretical considerations of the extraction of chloride complexes from pure HCl solutions led to the prediction of a reversed sequence of extraction [3]. To verify this experimentally, and in order to perform a systematic study of halide complexation of the group-5 elements, new batch extraction experiments for Nb, Ta, and Pa were performed with the quaternary ammonium salt Aliquat 336 in pure HF, HCl, and HBr solutions [4]. Based on these results, new chromatographic column separations were elaborated to study separately the fluoride and chloride complexation of element 105 with the automated rapid chemistry apparatus ARCA II. The separation in the pure HCl system is shown in Fig. 1. After feeding of the activities onto a 1.6x8 mm column containing Aliquat 336(Cl) on an inert support in 10 M HCl, a Ta fraction is eluted in 6 M HCl followed by a Nb fraction in 4 M HCl and a Pa fraction in 0.5 M HCl.

For the study of element 105, a 685  $\mu\text{g}/\text{cm}^2$  <sup>249</sup>Bk target was bombarded with 99 MeV <sup>18</sup>O ions at the Lawrence Berkeley National Laboratory 88-Inch Cyclotron to produce 34-s <sup>262</sup>105. The collection time of the He(KCl)-gas jet in ARCA II was 50 s, the cycle time 60 s. After feeding the activities onto the column in 167  $\mu\text{l}$  10 M HCl, a 183  $\mu\text{l}$  6 M HCl Ta fraction was eluted followed by the stripping of a Nb/Pa fraction from the column in 6 M HNO<sub>3</sub>/0.015 M HF. In 930 separations, 6  $\alpha$  singles and 1  $\alpha\alpha$ -mother-daughter correlation attributable to the decay of <sup>262</sup>105 and <sup>258</sup>Lr were registered in the Ta fraction and 12  $\alpha$  singles and 2  $\alpha\alpha$ -mother-daughter correlations in the strip fraction. From the ratio of events, a distribution coefficient  $K_d = 508^{+692}_{-237}$  in 6 M HCl is deduced. This value is shown in Fig. 2 together with the  $K_d$  values for Pa, Nb, and Ta. Thus, the extraction sequence is Pa > Nb  $\approx$  105 > Ta.

This sequence, with 105 intermediate between Nb and Ta, is theoretically predicted [5], based on

calculations of the electronic structure of hydrolyzed species of Nb, Ta, Pa, and element 105 and of their various chloride complexes formed in HCl solutions. This sequence is essentially determined by predominant changes in the electrostatic metal-ligand interaction when OH groups are replaced by Cl<sup>-</sup> ligands.

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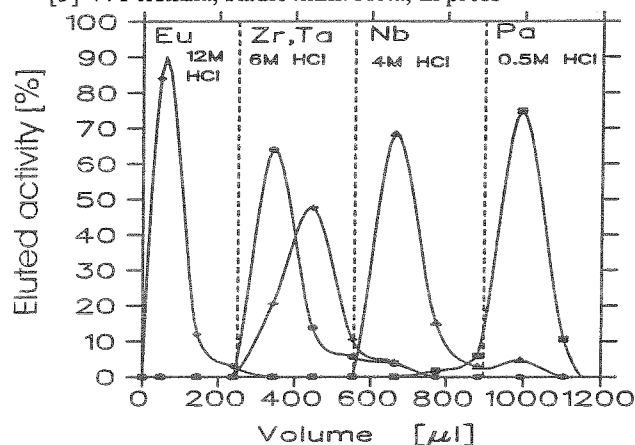


Fig. 1: Separation of Eu, Ta, Nb, and Pa on a 1.6x8 mm Aliquat 336(Cl) column. Eluents: Eu in 10 M HCl, Ta in 6 M HCl, Nb in 4 M HCl, and Pa in 0.5 M HCl.

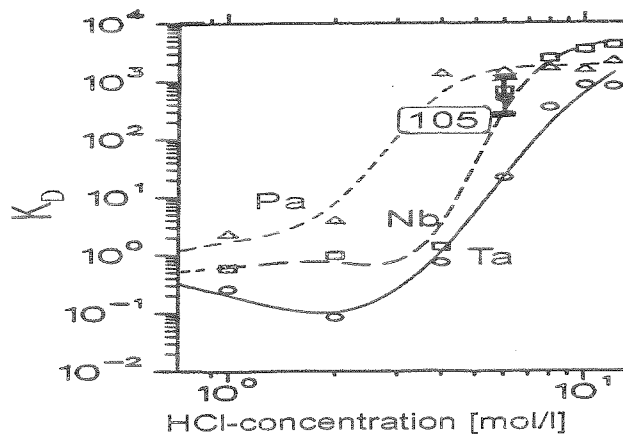


Fig. 2: Partition coefficients  $K_d$  in the system Aliquat 336(Cl)/HCl. The vertical bar represents the behaviour of element 105

## Aqueous Chemistry of Seaborgium

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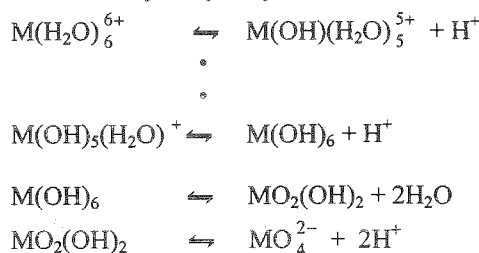
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We have previously reported on the first study of chemical properties of element 106, seaborgium, in aqueous solutions [1-4]. Seaborgium was rapidly eluted from a cation exchange column together with its homologues molybdenum and tungsten in 0.1 M HNO<sub>3</sub>/5·10<sup>-4</sup> M HF. Its chemical form was presumably a neutral or anionic oxygen containing fluoride. However, species containing no fluoride could not be excluded. In order to verify that fluoride complexing played a role in our previous study of seaborgium chemistry, we performed another series of cation exchange separations in which 0.1 M HNO<sub>3</sub> without HF was used as eluent. A target of 691 µg/cm<sup>2</sup> <sup>248</sup>Cm containing 22 µg/cm<sup>2</sup> <sup>152</sup>Gd was bombarded with <sup>22</sup>Ne<sup>5+</sup> ions at 123 MeV at an average intensity of 0.4 particle µA. The <sup>152</sup>Gd in the target produced <sup>169</sup>W on-line which was used to monitor the chemical yield of tungsten by γ-ray spectroscopy. The reaction products were transported with a He(KCl)-gas jet to the automated chemistry apparatus ARCA. The collection and cycle time was 45 s. The activity was dissolved in 0.1 M HNO<sub>3</sub> and eluted from 1.6x8 mm cation exchange columns (Aminex A6) within 8 s. For an improved decontamination from Bi/Po activities, the effluent passed a 0.6x5 mm column filled with finely grained Pd/H<sub>2</sub> where Bi/Po were electrochemically deposited. The effluent was sprayed onto 400-500 µg/cm<sup>2</sup> Ti foils mounted on Al frames and evaporated to dryness by IR-light. The Ti foils were inserted between pairs of PIPS detectors and assayed for α-particles. The energy resolution was 60 keV on the average and 550 keV for α-particles penetrating the Ti foil. The detection efficiency was thus doubled as compared to the previous experiments. For correlated pairs of mother-daughter decays this increases the detection efficiency by a factor of 4.

4575 experiments were performed. Only one correlated pair (8.26 MeV-8.18 MeV) of α-particles was registered. A Monte Carlo simulation yields 0.49 random correlations which means that the observed correlation has a 30% probability to be random. From the number of beam particles, the target thickness, the jet transportation yield, the detection efficiency and the chemical yield of tungsten (thereby assuming that the chemical yield of seaborgium is identical to the chemical yield of tungsten, i.e. 59% on the average), 4.7<sup>+3.7</sup><sub>-2.5</sub> (68 % conf. level) correlations were expected. The probability distribution around this expectation value leaves a probability of 14 % for the experimental observation to be compatible with the

expected number of correlations. From the lack of seaborgium events we conclude that seaborgium in the absence of HF sorbs on the cation exchange resin.

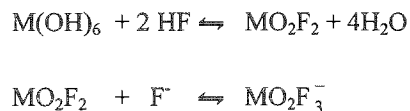
This non-tungsten like behaviour of seaborgium under the given conditions may be attributed to its weaker tendency to hydrolyze:



For Mo and W, the sequence of subsequent hydrolysis reactions in diluted HNO<sub>3</sub> reaches the neutral species MO<sub>2</sub>(OH)<sub>2</sub> [5]. A weaker tendency to hydrolyze for seaborgium would stop this sequence earlier, e.g. with M(OH)<sub>5</sub>(H<sub>2</sub>O)<sup>+</sup>, which sorbs on a cation exchange resin.

A decreasing tendency to hydrolyze (Nb>Ta>105>Pa) is reported [6] to determine the extraction of the group-5 chlorides into aliphatic amines, thus, a similar behaviour in the neighbouring group-6 is conceivable, but needs to be verified.

In the presence of fluoride ions having a strong tendency to replace OH-ligands, the formation of neutral or anionic fluoride species is favoured:



Thus, in the previous experiments with seaborgium in the presence of fluoride ions, neutral or anionic fluoride complexes, e.g. MO<sub>2</sub>F<sub>2</sub> or MO<sub>2</sub>F<sub>3</sub><sup>-</sup>, were likely to be formed and were eluted from the cation exchange columns [1-4].

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## Calculations of Dissociation Energies for RfCl<sub>4</sub>

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The DS-DV code of Rosen and Ellis has been recently further improved<sup>1</sup>, so that it became possible to calculate directly dissociation energies of molecules containing heavy atoms and a large number of ligands with sufficient accuracy.

The single-particle equations for the wave functions  $\phi_i$  and energy eigenvalues  $\epsilon_i$  are

$$\left[ t + V^n + V^c + V^{ex} \right] \phi_i = \epsilon_i \phi_i \quad i=1, \dots, N \quad (1)$$

The direct Coulomb potential  $V^c$  has been approximated by a potential  $\tilde{V}^c$  resulting from a model density

$$\rho(r) \equiv \tilde{\rho}(r) = \sum_k d_k f_k(r) \quad (2)$$

which is given as a linear combination of spherical, symmetric functions  $f_k(r)$  centered at the atoms of the molecule. The expansion coefficients are determined from an additional least-squares fit in which the error

$$\Delta = \iint d^3r d^3r' \frac{[\rho(r) - \tilde{\rho}(r)][\rho(r') - \tilde{\rho}(r')]}{|r-r'|} \quad (3)$$

in the total energy is minimized. The total energy is then

$$E = \sum_i \tilde{\epsilon}_i - \frac{1}{2} \int \tilde{\rho} \tilde{V}^c d^3r - \frac{1}{4} \int \rho V^{ex} d^3r + \sum_{p>q} \frac{Z_p Z_q}{|R_p - R_q|} + \Delta \quad (4)$$

where  $\tilde{\epsilon}_i$  is obtained via replacing  $V^c$  by  $\tilde{V}^c$  in Eq. (1). If  $\tilde{\rho}$  is determined such that Eq. (3) is minimized, one can show that replacing  $V^c$  by  $\tilde{V}^c$  in Eq. (1) yields a variationally consistent total energy from Eq. (4).

Another improvement is the calculation of the Fock and overlap matrix elements,  $H_{ij}$  and  $S_{ij}$ , respectively, with the integration scheme of Boerrigter et al.<sup>2</sup> In addition, a special transformation has been introduced to treat the singularity of the relativistic wave function at the origin; it is now possible to achieve high accuracy with a reasonable number of points (e.g.,  $10^{-8}$  with about 1400 integration points).

In this work, calculations of the dissociation energy ( $D_e$ ) and equilibrium bond distances ( $R_e$ ) are presented for group 4 highest chlorides MCl<sub>4</sub>, where M = Zr, Hf and element 104, rutherfordium (Rf). They are summarized in Tables 1 and 2 along with results of other calculations.

Table 1. Binding energies,  $D_e$  (eV), for MCl<sub>4</sub> (M=Zr, Hf and Rf) obtained as a result of different calculations

| Method | ZrCl <sub>4</sub> | HfCl <sub>4</sub> | RfCl <sub>4</sub> | Ref. |
|--------|-------------------|-------------------|-------------------|------|
| DS-DV  | 19.32             | 19.53             | 17.71             | this |
| DF     | 16.32             | 14.26             | 15.55             | 4    |
| exp.   | 20.05             | 20.56             | -                 | 5    |

Table 2. Equilibrium bond distances,  $R_e$  (Å), for MCl<sub>4</sub> (M=Zr, Hf and Rf) obtained as a result of different calculations

| Method | ZrCl <sub>4</sub> | HfCl <sub>4</sub> | RfCl <sub>4</sub> | Ref. |
|--------|-------------------|-------------------|-------------------|------|
| DS-DV  | 2.35              | 2.36              | 2.44              | this |
| DF     | 2.36              | 2.32              | 2.32              | 4    |
| exp.   | 2.32              | 2.32              | -                 | 5    |

The equilibrium bond distances and binding energies have been obtained as results of a Morse fit of the corresponding potential-energy curves. One of them, as an example, is presented in Fig. 1, showing a pronounced stability of RfCl<sub>4</sub> at 2.44 Å.

The new  $D_e$  and  $R_e$  values are in very good agreement with experiment (2-5% difference). One can see that the estimates obtained earlier<sup>3</sup> are also well in line with the new results.  $D_e$  obtained by the all-electron *ab initio* Dirac-Fock (DF) method<sup>4</sup> recover about 70% of the binding energy. The discrepancy with the experiment of the DF results is accounted for the lack of the correlation.

Thus, the very good agreement of the DS-DV results with the experiment for the binding (dissociation) allows for the use of the program in the investigations of bonding in even more complicated compounds. The calculations of the electronic structure and bonding of some other compounds of Rf and element 105 (Db) monoxides are in progress.

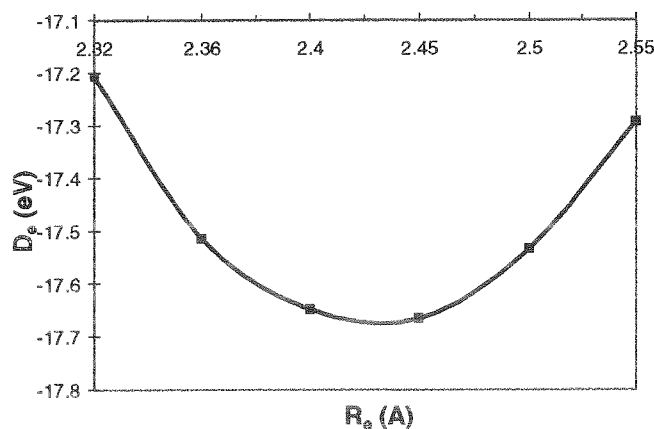


Fig. 1. Calculated potential energy curve for RfCl<sub>4</sub>.

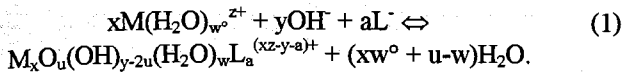
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# Complex Formation and Hydrolysis of Element 105 (Db) in HCl Solutions

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In complexing solutions, hydrolysis is competing with complex formation, which is described by the following equilibrium [1]



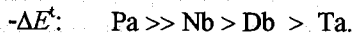
Reaction (1) defines the composition and the extent to which a hydrated metal ion is complexed in solution and finally extracted by various agents. Thus, among group 5 elements including Pa, the Pa complexes are known to be much less hydrolyzed and therefore much better extracted, while those of Ta are much more hydrolyzed and less extracted. The complex formation and extraction of Db influenced by hydrolysis has until recently been an open question.

The free energy changes of reaction (1) for different complexes of Nb, Ta, Db and Pa in HCl solutions, e.g.  $M(OH)_6^-$ ,  $M(OH)_2Cl_4^-$ ,  $MOCl_4^-$ ,  $MOCl_5^{2-}$ , and  $MCl_6^-$ , have been calculated using the model described in the previous report in this issue. The differences in the ionic and covalent parts of the interaction energy as well as in the total energy of reaction (1) for the formation of  $M(OH)_2Cl_4^-$ , as an example, are shown in Table 1.

Table 1. Differences in the ionic part,  $\Delta E^{ion}$ , and covalent part,  $\Delta E^{cov}$ , of the binding energies, and in the total energies,  $\Delta E^t$ , for the reaction  $M(OH)_6^- \rightleftharpoons M(OH)_2Cl_4^-$

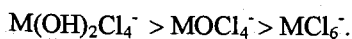
|                       | Nb    | Ta    | Db    | Pa    |
|-----------------------|-------|-------|-------|-------|
| $\Delta E^{ion}$ , eV | 12.69 | 13.44 | 13.42 | 10.85 |
| $\Delta E^{cov}$ , eV | 0.87  | 0.88  | 0.87  | 0.83  |
| $\Delta E^t$ , eV     | 13.56 | 14.32 | 14.29 | 11.68 |

The data in Table 1 show that, as in the case of hydrolysis of cations, the changes in the electrostatic part of the interaction energy (when OH groups are replaced by Cl) are larger than those of the non-electrostatic (primarily covalent) part. Thus, the complex formation of the elements considered here changes in the following order

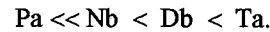


This sequence is in agreement with experiment on the complex formation for Pa, Nb and Ta.

Comparison of the  $\Delta E^t$  values for  $M(OH)_2Cl_4^-$ ,  $MOCl_4^-$  and  $MCl_6^-$  shows that, in agreement with experiment, the  $M(OH)_2Cl_4^-$  species are formed at lower HCl concentrations, while  $MCl_6^-$  at the highest concentrations, so that the complex formation changes in the order

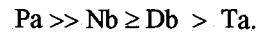


Since the complex formation considered here is a reverse process of hydrolysis of the formed species, the latter changes according to



Comparing the trends in the hydrolysis of complexes with the hydrolysis of cations (see the previous report) shows that both the cations and compounds of Pa are much less hydrolyzed than those of Nb, Ta and Db. For the other elements of interest, these processes are *not* related.

Using the theory of the solvent extraction by anion exchange which takes into account the complex formation, transfer of the ions from one phase to the other and the ion association in the organic phase, the following sequence in the distribution coefficients  $K_D$  has been predicted for the studied elements at HCl concentrations above 4 M (Fig. 1)



Recent experiments [2a] on the extraction of Nb, Ta, Pa and Db from 6M HCl have confirmed the predicted sequence.

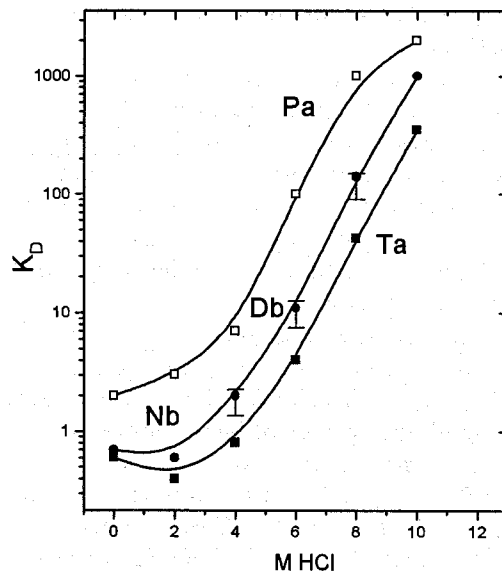


Fig. 1. Dependence of  $K_D$  on the HCl concentration. The predicted position of Db is shown by bars. The data for Nb, Ta, Pa are experiment [2a,b].

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## Reduction of W(VI) with the Automated Rapid Chemistry Apparatus ARCA II

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Tungsten (W) as a homologue of seaborgium (Sg) is used to develop a reduction experiment for Sg. Previous studies [1] with carrier free amounts of W showed the possibility to reduce W(VI) to W(III) with a solution of  $\text{Sm}^{2+}$ . While W(III) sticks to a cation exchange column in diluted HCl/HF as cation, W(VI) forms anionic oxyfluoro complexes and runs through the column. In a second step, W(III) is stripped from the column. In earlier experiments we used 4 M HCl/0.01 M HF solutions for this purpose.

Off-line experiments with reducing agents weaker than  $\text{Sm}^{2+}$  showed quite low reaction rates. Hence, although the reduction of dissolved W(VI) with  $\text{Sm}^{2+}$  seemed to be quantitative even with short reaction times, one could expect that the reaction rate could be too low for reaction times of about 1 s. On the other hand, the reduction might be assisted by the fact that on-line produced W would be dissolved directly into the reducing agent, while off-line experiments always involved the mixing of solutions. So, the main goal of the W experiments was to check whether under on-line conditions with ARCA the  $\text{Sm}^{2+}$  system would be as efficient in reducing W quantitatively as under off-line conditions.

W (mainly  $^{170}\text{W}$ ) produced on-line at the Philips Cyclotron of the Paul Scherrer Institut was transported to ARCA II with a He/KCl gas jet system. After a collection time of 120 s, the KCl spot was dissolved in 250  $\mu\text{l}$  of a mixture of HCl, dilute HF and 0.005 M  $\text{SmCl}_2$  solution. The solution was fed onto a CIX column (Aminex A4) at a flow rate of 1 ml/min. As described above, W(VI) runs through this column and is collected as the first fraction. W(III) sticks to the column and is stripped from the column using a solution of 0.1 M HCl and 15%  $\text{H}_2\text{O}_2$  as a second fraction. By this solution, W(III) is reoxidized to W(VI) and runs through the column, while Sm sticks to the column in both of its possible oxidation states. The more simple stripping with 4 M HCl/0.01 M HF would also elute the Sm from the column. In a Sg experiment, this would disturb the  $\alpha$  measurement by producing a thick sample. The  $\gamma$ -activities of the two fractions were determined.

For the feeding solution, different concentrations of HCl and HF were used to optimize the separation of the different oxidation states of W. Further, for each concentration there was taken a reference value without reducing agent (see Table).

| solvent<br>HCl/HF | reference<br>without $\text{Sm}^{2+}$ | 'reduced'<br>fraction | reduced |
|-------------------|---------------------------------------|-----------------------|---------|
| 1M/0.001M         | 6.2%                                  | 18.3%                 | 12.1%   |
| 0.5M/0.0005M      | 13.1 %                                | 23.3%                 | 10.2%   |
| 0.05M/0.0005M     | 15.6%                                 | 26.8%                 | 11.2%   |
| 0.5M/0.001M       | 16.5%                                 | 19.0%                 | 2.5%    |
| 0.05M/0.001M      | 6.8%                                  | 16.5%                 | 9.7%    |

The percentage of reduction was not much more than 10% at most HCl concentrations. Thus, the reaction rate seems to be too slow.

Recent off-line experiments lead us to a new concept using metallic Al as reducing agent. This system has the following advantages: The amount of  $\text{Al}^{3+}$  dissolved is negligible and does not disturb the  $\alpha$  measurement. The Al column can be heated more easily than a solution; higher temperatures generally lead to higher reaction rates. Off-line experiments with reaction times of less than 0.5 s (contact time with Al) yielded about 80% of reduced species. For this experiment, an anion exchange column is used with 0.1M HF/0.1M HCl (stripping with 4M HCl/0.01M HF) where the reduced species run through the column while the W(VI) sticks to the column. This should allow for a fast access to the reduced species in a future Sg experiment.

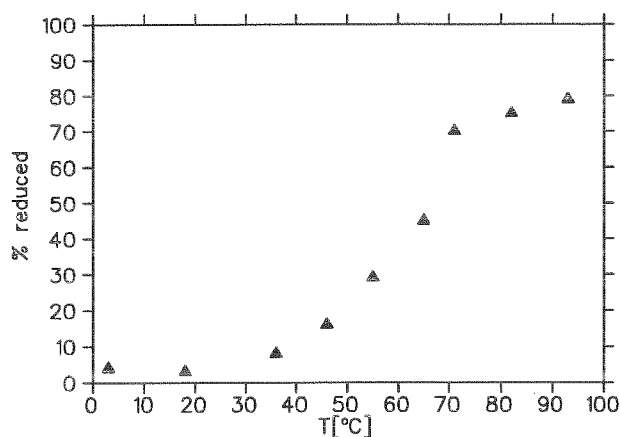


Fig.1: Temperature dependence of the reduction of W(VI) with Al. Reduced and unreduced species are separated by an anion exchange column (Dowex AG1x8)

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# ON-LINE ISOTHERMAL GAS CHROMATOGRAPHY OF SHORT-LIVED ISOTOPES OF THE SEABORGIUM HOMOLOGUES IN HUMID OXYGEN

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To characterize Sg and to look for general trends at the end of the periodic table, Sg has to be studied in comparison to its lighter homologues Mo and W. The experimental set-up has to be tested by model experiments with trace amounts of short-lived isotopes of these elements. The behavior of trace amounts of the Sg homologues in the O<sub>2</sub>-H<sub>2</sub>O(g)/SiO<sub>2(s)</sub>-system has been already studied earlier by off-line thermochromatography with long-lived nuclides [1]. It was found that all group 6 elements are transported via reaction gas chromatography MO<sub>2</sub>(OH)<sub>2(g)</sub> ↔ MO<sub>3(ads)</sub> + H<sub>2</sub>O(g) characterized by relatively slow kinetics.

In test experiments with the OLGA II apparatus total retention times of Mo species of more than one minute were measured [2], far too long for studying the nuclides <sup>265</sup>Sg and <sup>266</sup>Sg.

In this work improved experimental arrangements were tested studying short-lived isotopes of Mo and W. Mo isotopes were produced in the <sup>235</sup>U (n, f) reaction at the FZR U-120 cyclotron using neutrons from the reaction <sup>9</sup>Be (d, n) <sup>10</sup>B. To get short-lived W isotopes an enriched <sup>152</sup>Gd target was bombarded with <sup>20</sup>Ne at the PSI PHILIPS cyclotron. The He/MoO<sub>3</sub>-jet used to transport the reaction products to the chromatography apparatus was operated with a He-flow rate of 1.0 l/min. At the column entrance 0.5 l/min O<sub>2</sub> moistened with H<sub>2</sub>O at 50 °C were added to form volatile oxidehydroxides. The temperature of the reaction zone was 1350 K, the isothermal column temperature was varied between 800 and 1350 K.

In a first series the volatile Mo species leaving the chromatography column were directly deposited on thin Al foils mounted on the wheel of the GSI Rotating wheel Multidetector Analyzer ROMA to replace the time consuming recluster step between chromatography and detection of the species. The wheel was operated with a collection/detection time of 60 s. From the experimental data an enthalpy value of  $\Delta H^{\circ}_{\text{diss.ads}} = -54$  kJ/mol was determined by Monte Carlo simulation [3]. This value correspond well with the  $\Delta H^{\circ}_{\text{diss.ads}} = -50 \pm 9$  kJ/mol from former thermochromatography experiments [1]. A retention time through the chromatographic column of 7 seconds was calculated by the Monte Carlo method. About 10 seconds result from the T<sub>50%</sub>-value [4].

In a second series a further improved experimental arrangement was tested. To establish an optimum pressure for the direct deposition of about 80 mbar at a pressure below 5 mbar in the ROMA chamber and to cool the wheel a separate deposition chamber was placed between column and rotating wheel which was operated with a collection/detection time of 20 s. At a coolant temperature of 10 °C the temperature of the rotating wheel at the first detection position did not exceed 25 °C. This temperature is sufficient to avoid water condensation on the foils. The activity of the foils was measured either  $\gamma$ - or  $\alpha$ -spectrometrically. Maximum yields of 53 % for <sup>166</sup>W (t<sub>1/2</sub> = 18.8 s) and 63 % for <sup>168</sup>W (t<sub>1/2</sub> = 51 s) were measured at 1350 K relative to the activity of the gas-jet.

Fig. 1 shows relative yields of <sup>166</sup>W and <sup>168</sup>W (maximum value = 100 %) as a function of the isothermal temperature together with yield curves obtained by Monte Carlo simulation. The best fit of the experimental data was obtained with enthalpy values of  $\Delta H^{\circ}_{\text{diss.ads}} = -54$  and  $-56$  kJ/mol, respectively. These values correspond well with the value of  $\Delta H^{\circ}_{\text{diss.ads}} = -62 \pm 13$  kJ/mol derived from former thermochromatography experiments [1]. The retention time spent by the W species during their travel from the column entrance to the Al foil was determined to be about 20 s. From the nuclide yield ratio at 1350 K a retention time of 8 s was calculated according [5] which seems to be a more reliable value since a few assumptions without proven correctness were used to simulate the chromatographic process.

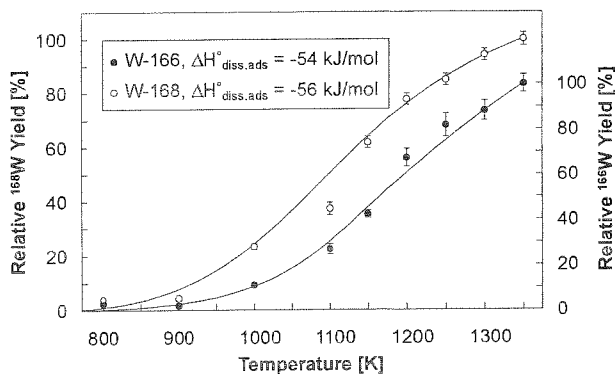


Fig. 1: Relative yields of short-lived W isotopes as a function of the isothermal temperature

The  $\alpha$ -spectrometric measurements were carried out in 2 $\pi$  geometry with a resolution of 30 - 50 keV (FWHM). Nuclides like <sup>152</sup>Ho and <sup>151</sup>Dy produced in transfer reactions were detected only in negligible quantities what shows that they were retained nearly quantitatively in the column. The results are promising with regard to future studies of Sg. However, as a prerequisite for the unequivocal detection of Sg  $\alpha$ -spectroscopy in 4 $\pi$  geometry has to be realized.

## Acknowledgements

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