

Ion-molecule reactions of Ru^+ and Os^+ with oxygen in a Penning trap

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Summary. Gas-phase ion chemistry in a Penning trap has been explored for future experiments on the chemical properties of the heaviest elements. The Mainz Cluster Trap, a Penning trap experiment devoted to metal cluster research, has served as a model apparatus for SHIPTRAP which is being installed behind the velocity filter SHIP (Separator for Heavy Ion reaction Products) at the Gesellschaft für Schwerionenforschung (GSI) at Darmstadt. The reactions of stored Ru^+ and Os^+ with oxygen have been studied and the reaction products MO^+ and MO_2^+ ($M = \text{Ru}, \text{Os}$) have been observed. The corresponding rate constants have been measured and the results are discussed with respect to future studies of similar ion-molecule reactions of the element 108 (hassium) in SHIPTRAP.

1. Introduction

In the past decade, considerable progress has been made in the field of heavy element research [1]. This includes studies of the chemical properties of rutherfordium (element 104), dubnium (element 105), seaborgium (element 106) [2], and bohrium (element 107) [3] in the liquid phase and in the gas phase, respectively. These results were mostly obtained by the application of highly automated separation methods coupled to a He-cluster jet transportation system [4]. Alternatively, gaseous compounds have been formed *in situ* by reactions of recoil atoms with reactive gases directly behind the target [2]. For the heavier elements hassium (element 108), meitnerium (element 109) and further [1], low production rates and interfering activities produced in the nuclear reaction make it more difficult to use the techniques mentioned above. Thus, a device that combines a fast selection and the possibility of gas-phase chemistry is needed, preferably installed behind an electromagnetic separator such as SHIP [5], where the separated heavy-ion fusion products are available as ions. Therefore, ion-molecule reactions (see [6, 7] and references therein) may be considered as a new way to study their chemical properties.

Ion trapping devices have proven to be a versatile tool in mass spectrometry and for the investigation of ion-molecule reactions in the gas phase [6, 8, 9]. An Ion Cyclotron Resonance (ICR or Penning) trap is currently being installed behind the velocity filter SHIP [1, 5] at GSI in Darmstadt, Germany. This new apparatus, SHIPTRAP [10], will be suitable for future experiments on the heaviest elements. The reaction recoils, which are produced by irradiation of thin target foils by use of the heavy-ion linear accelerator UNILAC, are pre-selected with SHIP. All passing recoils are stopped in a buffer-gas chamber [11], cooled and accumulated in a radiofrequency quadrupole (RFQ) buncher [12] and finally injected into the Penning trap setup. Here, the ion bunch is purified and the elements of interest can be exposed to reactive gases. The reaction products will be characterized by their decay properties and by mass spectrometry.

It has been demonstrated by ISOLTRAP [13] at ISOLDE/CERN [14], that nuclides with lifetimes of less than one second (*e.g.* ^{33}Ar with $T_{1/2} = 174$ ms [15]) are accessible for experiments with Penning trap mass spectrometers [16, 17]. Therefore, it seems possible to use SHIPTRAP for the investigation of, *e.g.*, hassium, which has a lifetime of about 10 s [18].

The present work explores the merits of ion-chemical investigations of superheavy elements in a Penning trap. In order to test the possibilities of chemical studies of hassium ions, experiments have been performed with its lighter homologous elements ruthenium and osmium. Oxygen was chosen as a reactive gas, because it is well known that neutral ruthenium and osmium form very stable compounds with oxygen. In addition, relativistic density functional calculations for OsO_4 , RuO_4 , and HsO_4 have shown element 108 (hassium) to have properties similar to osmium and ruthenium [19]. Reactions of the ruthenium ion, Ru^+ , have already been studied by guided ion beam mass spectrometry. The ion's reaction with molecular oxygen was found to be endothermic [20]. Thus, Ru^+ , at low temperature and as long as in the electronic ground state, is not expected to react with oxygen. To our knowledge, similar experiments with Os^+ have not been reported in the literature.

In the following, the experimental setup and procedure is described followed by the data evaluation. The reaction rates are discussed with respect to excited states that seem to be

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involved (as inferred from the known endothermicity of the ruthenium reaction) and in view of future studies of similar ion-molecule reactions of the element 108 at SHIPTRAP.

2. Experimental setup

For the present investigation, the experimental setup of the Mainz Cluster Trap has been used (see Fig. 1). Since it has already been described in detail [21–23], only the features relevant for the present experiment are briefly reviewed.

Positively charged ions of the element under investigation are produced in a Smalley-type cluster source [24]. Material from the surface of a metal wire is vaporized by the light of a pulsed, frequency doubled Nd:YAG laser (5–10 mJ, 10 ns) and the desorbed material is expanded adiabatically into the vacuum within a pulsed helium gas jet. During the recombination process, small neutral, anionic and cationic aggregates (clusters) are formed [25]. However, for the present experiments on Ti, Zr, Ru and Os, the atomic cations have been used, only.

The ions are guided into a Penning trap [26], which consists of a ring and two endcap electrodes of hyperbolic shape. Between these electrodes, a trapping voltage is applied and produces a static quadrupolar field for axial confinement (5 V potential depth). For radial confinement, a homogeneous magnetic field of a superconducting magnet ($B = 5$ T) is superimposed in axial direction. Ions are injected into and ejected out of the trap through holes in the center of the endcaps [27]. The ring electrode is segmented and allows the application of rf signals for the manipulation of the ions' motion [28]. By selective excitation of the cyclotron motion, unwanted ions can be radially ejected from the trap.

The ions of interest are centered by a combination of buffer gas collisions and a quadrupolar excitation [29] and confined within a region of about 2 mm diameter in the middle of the trap [30]. After a reaction period of up to several seconds, the charged products are ejected out of the trap into a time-of-flight (TOF) mass-spectrometer for mass analysis. Single ion detection is performed by use of a conversion electrode and a Micro Channel Plate (MCP) detector. Typically, the data of 200 to 500 cycles with 10 to 20 observed

ions each are added to obtain statistically significant signal intensities.

The present experiment studies the ion molecule reactions $M^+ + O_2 \rightarrow MO_n^+$ ($n = 0, 1, 2$) ($M = Ru, Os$) and their rate constants. The reactive gas (oxygen) is continuously leaked into the trap region through a needle valve and provides a constant background pressure (typically $5 \times 10^{-7} < p < 2 \times 10^{-5}$ mbar). After the metal ions are captured in flight, trapped contaminant and prebuilt oxide ions are radially ejected as described above. By the end of this selection process, only the educts of interest are left in the trap. During the reaction periods (between 10 μ s and 20 s), all charged species remain stored and, thereafter, the products are mass analyzed. Both the remaining metal ions which did not react as well as the metal oxide ions are detected. In order to minimize the influence of fluctuations in precursor ion production, a reference cycle with a reaction period of 100 ms is performed in alternation to the above described cycle and the data are normalized with respect to these reference data.

The ion source is designed for the use of a wire as target material [25]. However, no solid osmium and ruthenium wires are available commercially. Thus, a substrate wire is coated with metallic Os or Ru. For the selection of the supporting wire, several criteria have to be met. First, in order to be fed through the source, the wire (< 1 mm diameter) has to be pliable. Second, the material has to withstand the coating process performed in a sulphuric acid solution ($pH = 1.5$). And, most important, in order to separate the ion signals in the mass spectra that correspond to material from the wire and the coating, bare ions, oxide-ions as well as cluster ions must have masses that are different from osmium, ruthenium and their oxides. Several materials exhibit the pliability and chemical durability needed, but the third criterion is harder to meet as illustrated in Fig. 2. Bare copper and its oxides are much lighter than ruthenium and osmium, causing no mass spectrometrical ambiguities. However, the copper trimer has some isotomeric overlap with osmium (Fig. 2, top), proving copper to be unsuitable. Tantalum meets all three criteria (Fig. 2, bottom) and has therefore been chosen as wire material.

The coating of the tantalum wire has been performed in an electrolysis cell (see Fig. 3) that has been heated in

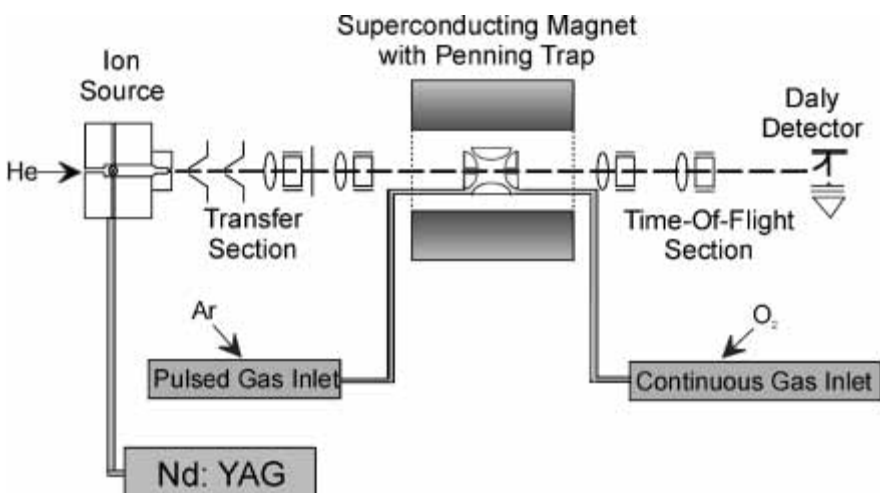


Fig. 1. Schematic overview of the experimental setup: Metal ions (M^+) are produced in an external ion source and stored in a Penning trap at a constant oxygen pressure. After formation of oxides MO_n^+ ($n = 1-2$) during a variable reaction period, all ions are ejected and analyzed by time-of-flight mass spectrometry. (For details see text.)

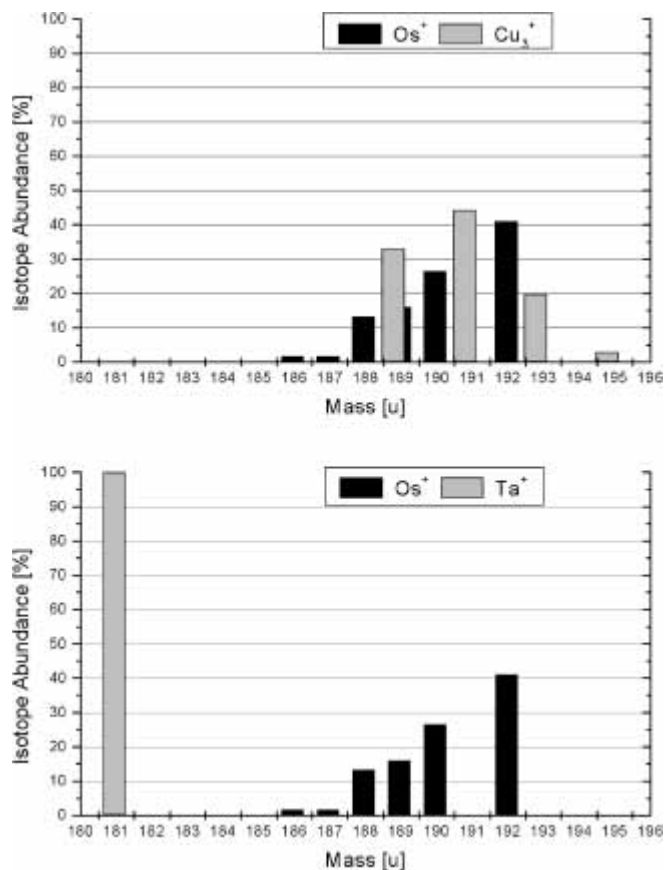


Fig. 2. Mass distribution for the core wire materials copper (top) and tantalum (bottom) compared to the distribution of the osmium isotopes.

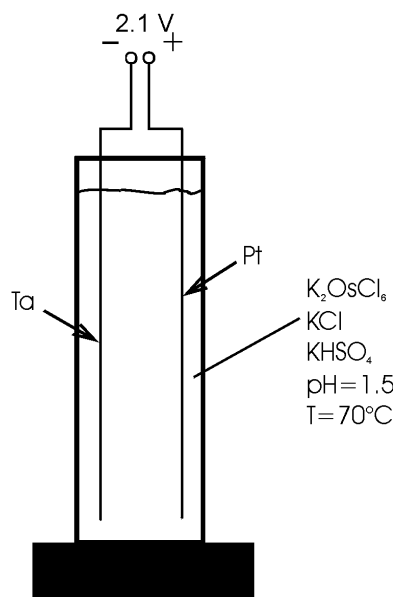


Fig. 3. Electrolysis cell for the coating of the target wire (for process details see text).

a water bath to a temperature of about 70 °C. K₂OsCl₆ served as electrolyte for the osmium coating and RuCl₃·xH₂O ($x = 1-3$) for the ruthenium coating. The potentiostatic electrolysis was performed at 2.1 V for 2.5–3 hours with an electrolyte of 1.5 g of osmium or ruthenium salt, 30 ml 0.2 M KCl solution and 30 ml 0.5 M KHSO₄ solution (adjusted to pH 1.5). A 30 cm long piece of tantalum wire

(1 mm diameter) was used as the cathode and a platinum wire as the anode [31,32]. After the electrolysis, the coated wire has been taken out of the solution and rinsed with water to remove residues of the electrolyte. It was dried in air before insertion into the ion source.

3. Results and discussion

3.1 Pressure calibration

The rate of a gas phase ion-molecule reaction depends on the collision frequency of the reactants determined by the velocity distribution and the density (pressure) of the reacting gas (or gases). The velocity is well approximated by a Maxwell distribution at room temperature. The pressure, however, cannot be directly determined in the interaction region. Hence, the reading of a gauge, which is mounted at a distance of 1.5 m from the trap, has been calibrated by use of the oxidation of titanium



as a reference reaction in the trap volume [33,34] with a known rate constant $k_{\text{Ti}^+} = (5.0 \pm 1.0) \times 10^{-10} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$ [35].

The number of Ti⁺ ions, $N_{\text{Ti}^+}(t)$, has been measured as a function of the reaction time, which shows an exponential decrease (see Fig. 4) of the form

$$N_{\text{Ti}^+}(t) = N_{\text{Ti}^+}(0) e^{-k'_{\text{Ti}^+} t}, \quad (2)$$

with the ‘pseudo’ rate constant

$$k'_{\text{Ti}^+} = k_{\text{Ti}^+} N_{\text{O}_2}, \quad (3)$$

comprising the rate constant of the reaction, k_{Ti^+} , and the number density of oxygen N_{O_2} , which is easily converted to the pressure p .

In order to confirm the linearity of the pressure reading at the TOF section as a function of the pressure at the location of the trap, the above calibration measurement has

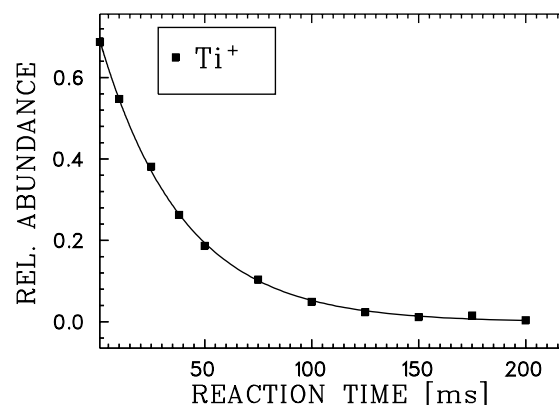


Fig. 4. Relative abundance of titanium ions as a function of the reaction time at an oxygen pressure $p = 2 \times 10^{-6}$ mbar. The line represents the fit of Eq. (2) to the data ($k'_{\text{Ti}^+} = (26.2 \pm 0.7) \text{ s}^{-1}$).

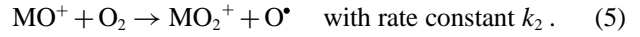
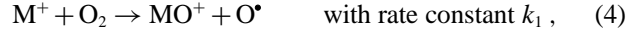
been carried out at several pressures. These measurements yield good agreement for the relevant range of $5 \times 10^{-7} < p < 2 \times 10^{-5}$ mbar.

3.2 Reactions of Os^+ and Ru^+ with oxygen

As described in Sect. 2, a sample of metal cations is prepared in the trap and exposed to oxygen at a fixed pressure for a variable reaction time ($10 \mu\text{s} < \Delta t < 20$ s). Because all charged species remain trapped, the surviving precursor ions as well as the metal-oxide ions are detected. As an example, Fig. 5a shows the relative ion intensities for the reaction of Os^+ with O_2 at $p = 7.5 \times 10^{-7}$ mbar as a function of the reaction time. The Os^+ signal shows an exponential decrease. The monoxide-ion signal (Fig. 5b) increases at some 100 ms from a finite initial value and after approximately 1 s it decreases. This decrease of the OsO^+ signal is accompa-

nied by the formation of osmiumdioxide ions (Fig. 5c). The solid lines represent fits to the measured data as discussed below.

Obviously, the reaction is composed of two sequential oxidation steps:



The solution of the differential equations of sequential reactions (or decays) is a set of exponential functions [36], which describe the intensities of the precursor and the products, in the present case:

$$N_{\text{M}}(t) = N_{\text{M}} \cdot e^{-k_1' t}, \quad (6)$$

$$N_{\text{MO}} = N_{\text{M}} \cdot \frac{k_1'}{k_2' - k_1'} \cdot (e^{-k_1' t} - e^{-k_2' t}), \quad (7)$$

and

$$N_{\text{MO}_2} = N_{\text{M}} \cdot \left(1 - \frac{k_2' \cdot e^{-k_1' t} - k_1' \cdot e^{-k_2' t}}{k_2' - k_1'} \right). \quad (8)$$

N_{M} represents the number of metal ions at $t = 0$. k_1' and k_2' are the pseudo rate constants as introduced above. The constant k_1' was obtained from a fit of Eq. (6) to the exponential decrease of the osmium-ion signal. k_2' was derived by use of Eq. (8) (where k_1' was kept fixed as determined before). For the osmium reaction at $p = 7.5 \times 10^{-7}$ mbar (see Fig. 5), the exponential equations fit the data very well. By use of Eq. (3) and the known oxygen pressure, the pressure-independent reaction rate constants k_1 and k_2 are obtained from the ‘pseudo’ rate constants k_1' and k_2' .

The osmium reaction has been investigated for seven different oxygen pressures. These measurements yield ‘pseudo’ rates that increase linearly with the pressure as expected from Eq. (3). All measurements have been performed in complete analogy for ruthenium ions. Fig. 6 shows an example of the Ru^+ and the oxidation product yields at $p = 1.6 \times 10^{-5}$ mbar. A comparison of the results with the osmium reaction indicates a much slower reaction (note that the oxygen pressure is increased by a factor of 20 as compared to the measurements shown in Fig. 5). The depletion of the precursor ions is not complete after 20 s (see Fig. 6a), and also the amount of ions that have reacted to the dioxide after 20 s (Fig. 6c) is much lower than in the case of osmium. Table 1 summarizes the results for both elements. In the case of ruthenium the mean values of the rate constants for both oxidation steps ($k_1 = (2.8 \pm 0.7) \times 10^{-12} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$, $k_2 = (6.9 \pm 3.6) \times 10^{-12} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$) are equal to or slightly smaller than the values for the osmium reaction ($k_1 = (2.6 \pm 0.3) \times 10^{-12} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$, $k_2 = (9.4 \pm 4.0) \times 10^{-12} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$).

Many chemical reactions can proceed in both directions, and, if the rate constants are of similar order of magnitude, an unconsidered back reaction might result in an underestimation of the actual rate constant. To check whether there is a back reaction in the first step of the proposed sequential reaction (see Eq. (4)), an additional experiment was performed: MO^+ ions formed in the source or during transfer were selected, stored in the trap and, after a variable reaction time, the products were identified. Only dioxide ions and no

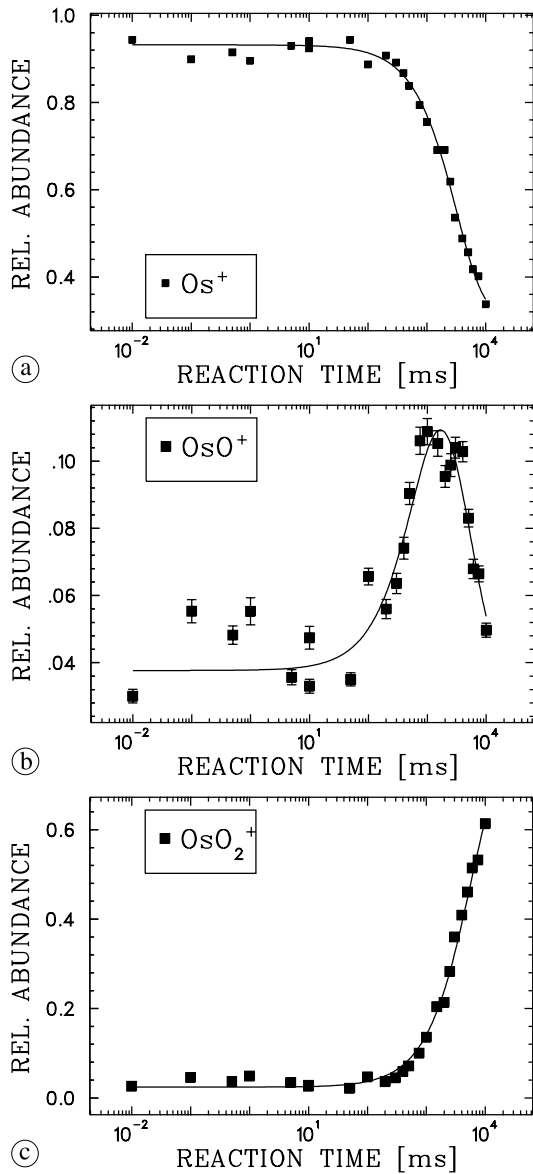


Fig. 5. Relative abundance of (a) osmium ions (b) osmium-monoxide ions and (c) osmium-dioxide ions as a function of the reaction time at $p = 7.5 \times 10^{-7}$ mbar. The solid line is a fit of Eqs. (6), (7) and (8) to the data, respectively ($k_1 = 7(1) \times 10^{-12} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$ and $k_2 = 9(2) \times 10^{-12} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$).

| Oxygen pressure p_{PT} [mbar] | Ruthenium [10^{-12} cm ³ molecule ⁻¹ s ⁻¹] | | Osmium [10^{-12} cm ³ molecule ⁻¹ s ⁻¹] | |
|---------------------------------|---|--------|--|--------|
| | k_1 | k_2 | k_1 | k_2 |
| 7.5×10^{-7} | — | — | 7(1) | 9(2) |
| 9.8×10^{-7} | 8(2) | 13(4) | 3(1) | 11(18) |
| 2.3×10^{-6} | 3(1) | 17(13) | 2.0(3) | 17(3) |
| 4.4×10^{-6} | 3(1) | 6(2) | 1.8(2) | 9(1) |
| 6.5×10^{-6} | 0.5(1) | 2(1) | 1.7(2) | 8(1) |
| 8.3×10^{-6} | 1.0(2) | 2(1) | 1.9(2) | 8(1) |
| 1.6×10^{-5} | 0.5(1) | 1.1(3) | 1.7(3) | 4(1) |

Table 1. Rate constants of the ion-molecule reactions of ruthenium and osmium with oxygen.

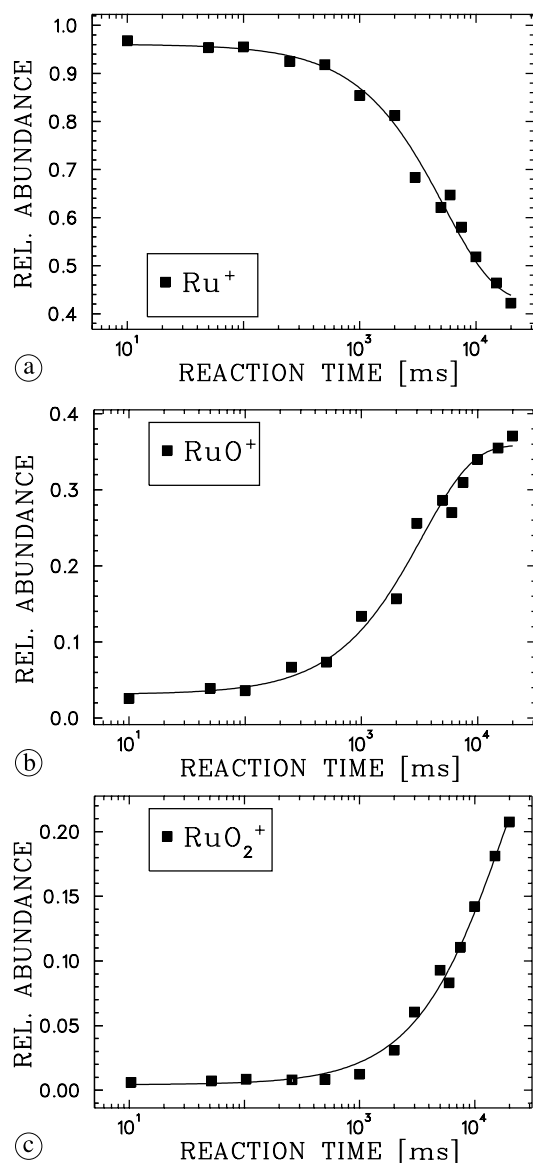


Fig. 6. Relative abundance of (a) ruthenium ions, (b) ruthenium-monoxide ions and (c) ruthenium-dioxide ions as a function of the reaction time at $p = 1.6 \times 10^{-5}$ mbar. The solid line is a fit of Eqs. (6), (7) and (8) to the data, respectively ($k_1 = (0.5(1)) \times 10^{-12}$ cm³ molecule⁻¹ s⁻¹ and $k_2 = (1.1(3)) \times 10^{-12}$ cm³ molecule⁻¹ s⁻¹).

pure metal ions were observed, contrary to the expectations in the case of a back reaction. Hence, under the present experimental conditions, no back reaction takes place and the assumption of a one-way reaction is justified.

3.3 Discussion and conclusion

As shown in the previous section, it takes more than 10 seconds to convert all osmium ions into the dioxide ion at the highest oxygen pressure applied ($p = 1.6 \times 10^{-5}$) and for ruthenium, the reaction is even slower – after 20 seconds, the reaction into the dioxide ion is far from being complete. Both reactions stop at the second oxidation stage (the dioxide ion) and no higher oxidized species, like *e.g.* MO_3^+ , have been observed.

The oxidation of ruthenium as well as osmium is well described by a simple sequential reaction. The measured rate constants k_1 for the first step $\text{M}^+ \rightarrow \text{MO}^+$ and k_2 for the second step $\text{MO}^+ \rightarrow \text{MO}_2^+$ (see Table 1) show that for both elements the reaction velocity is dominated by a slower first step that is followed by a much faster second step. To our knowledge, the ion molecule reaction of osmium with oxygen has not been studied before. However, a spontaneous reaction of ruthenium ions with oxygen is not expected. On the contrary, it is known from the literature [20] that the reaction $\text{Ru}^+ + \text{O}_2 \rightarrow \text{RuO}^+ + \text{O}^*$ is endothermic ($\Delta_f H^\circ = 5.2 \pm 1.1$ eV at $T = 0$ K, as deduced from the data in [20]). Therefore, an additional reaction has been investigated, where the rate constant is already known, *i.e.*, the reaction of zirconium ions with oxygen. Dheandhanoo *et al.* have shown that the rate constant of this reaction is equal to that of the previously used calibration reaction of titanium with oxygen [37], $k_{\text{Ti}^+} = k_{\text{Zr}^+} = (5.0 \pm 1.0) \times 10^{-10}$ cm³ molecule⁻¹ s⁻¹.

To obtain comparable data, the zirconium oxidation has been performed in the same way as the ruthenium and osmium reactions. Zirconium ions were produced by laser vaporization from a zirconium wire in the source and selection, trapping and detection were performed as described above for Os and Ru. Again, the precursor ion number was measured as a function of the reaction time and for various oxygen pressures. As expected, an exponential decrease of the precursor-ion signal was observed and with Eq. (6), the corresponding rate constants were deduced (see Table 2). From these results, a mean value of $k_{\text{Zr}^+} = (7.1 \pm 1.4) \times 10^{-10}$ cm³ molecule⁻¹ s⁻¹ for the experimental rate constant was fitted which is only slightly higher than the expected value [37].

Thus, the present results are as yet inconclusive with respect to the nature of the measured reaction rates. The titanium and zirconium measurements give consistent results while ruthenium shows a strong deviation from the expected behavior, and for osmium there are no prior reports. Possible explanations for the finite ruthenium rates are: (a) long-lived

Table 2. Rate constants of the ion-molecule reaction of zirconium ions with oxygen at various oxygen pressures.

| Oxygen pressure p_{PT} [mbar] | k_{Zr^+} (Exp) [$10^{-10} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$] |
|---|---|
| 7.5×10^{-7} | 5 (1) |
| 9.8×10^{-7} | 8 (2) |
| 2.3×10^{-6} | 7 (1) |
| 3.1×10^{-6} | 7 (1) |
| 4.4×10^{-6} | 7 (1) |
| 8.3×10^{-6} | 9 (3) |

metastable states, that may survive many collisions both in the ion source and in the trap, (b) non-Maxwellian velocity distributions of the ions in the trap, in particular due to the persistent application of quadrupolar excitation of the ions' radial motion, in order to center the ions in the middle of the trap [29].

In the first case, the observed reaction would be a superposition of the reactions of two (or more) distinctive species, Ru^+ and Ru^{2+} , where the ground state species should not react to RuO^+ . The influence of long-lived excited states is reported, *e.g.*, in the case of the reaction of Ag^+ with oxygen [20]. A thermalization of ruthenium and osmium with argon gas has been performed prior to the reaction with oxygen, but the number of collisions (about 100) was possibly not sufficient to quench all excited states [38]. The present data do not allow to decide whether there is a finite non-reacting offset of Ru^+ ions (see Fig. 6a). Under the assumption of case (b) all measured rate constants are upper bounds of the actual room temperature rates. In any case, both of these complications may apply to any of the four metals that have been used for the present study.

Nevertheless, the following important conclusion can be drawn: In principle, kinetic data on ion-molecule reactions can be obtained in a Penning trap study with single ion detection, and the stoichiometry of the reaction products can be determined unambiguously by mass spectrometry. This is a distinct advantage over conventional gas-phase or liquid-phase experiments [2] in which the stoichiometry of investigated molecules can at best be deduced indirectly on the basis of comparisons of the chemical behavior of homologous elements within a given group in the Periodic System. The low reaction rates observed, which actually may only be upper bounds, will constitute a severe problem for short-lived isotopes with lifetimes well below the inverse of the reaction rates. This problem is due to the low permissible gas pressure in the Penning trap. Therefore, the main conclusion from the present work is that ion-molecule reactions should preferably be performed already in the buffer-gas cell or the RFQ buncher of SHIPTRAP for cooling and accumulation. Thus, high gas pressures can be realized at the expense of a loss of information on kinetic data. The stoichiometry of the reaction products can still be determined by a subsequent mass measurement in the Penning trap.

Note added in proof:

In a subsequent experiment by G. Marx *et al.* (to be published) using a Fourier Transform-Ion Cyclotron Resonance

apparatus (FT-ICR) it could be shown that Ru^+ ions react with oxygen only when their radial motion is excited. Thus, apparently the persistent application of quadrupolar excitation of the ions' radial motion was responsible for the Ru^+ ions reactions in the present work.

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