

FT-ICR MS studies of ion-molecule reactions of Ru⁺ and Os⁺ with oxygen

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Abstract

The reactions of stored ruthenium and osmium cations with oxygen have been studied in a Fourier transform ion cyclotron resonance (FT-ICR) mass spectrometer. In case of osmium the reaction products OsO⁺ and OsO₂⁺ have been observed and corresponding reaction-rate constants have been determined. In addition, there is an unreactive fraction of Os⁺ ions due to the presence of a slightly endothermic reacting ground state. Only the excited states react with oxygen. For ruthenium no spontaneous reaction with oxygen has been observed unless the cyclotron motion of Ru⁺ was excited. The results are discussed with respect to a similar investigation in a Penning trap-TOF mass spectrometer [U. Rieth, A. Herlert, J.V. Kratz, L. Schweikhard, M. Vogel, C. Walther, *Radiochim. Acta* 90 (2002) 337] and with respect to possible future studies of ion-molecule reactions of the homologous super-heavy element 108 (hassium).

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1. Introduction

The discovery of the heaviest elements [1] immediately raised the question of their properties. In particular, there is a strong interest in the chemical characterization with respect to their position in the periodic table of elements [2]. As the nuclear charge increases, strong relativistic effects on the electron configuration are expected [3,4], similar and in excess of those already observed, e.g., in the case of gas-phase ion chemistry of gold [5]. Experimental data and energy levels from Hartree-Fock calculations have been compared for the trans-actinides and their homologous elements in order to study the change in chemical behavior [6]. The results from gas-chromatographic experiments for the elements 104 (rutherfordium) and 105 (dubnium) showed deviations due to relativistic effects [7,8] whereas for the elements 106

(seaborgium) and 107 (bohrium) the expected position in the periodic table was confirmed [9,10]. Fully relativistic density functional calculations on the volatile group 8 tetroxides RuO₄, OsO₄ and HsO₄ predict a similarity of the element 108 (hassium) to its lighter homologues ruthenium and osmium [11]. Recently, the chemical separation and characterization of hassium has been performed with a new gas-chromatographic separation system [12], and the oxidation of seven Hs atoms to HsO₄ could be found and compared to the production of OsO₄ confirming the predicted chemical behavior of element 108 [13].

In addition to the aqueous and gas-chromatographic experiments, the use of the ion trapping device SHIPTRAP [14] is planned for the investigation of superheavy elements. This device is a Penning-trap apparatus located at SHIP, a kinematic separator for reaction recoil particles from thin targets. For the production of superheavy elements, the targets are irradiated by beams of heavy ions from the linear accelerator UNILAC at GSI [15]. After their flight through SHIP the recoil ions are stopped in a “gas cell”, a chamber filled with

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nobel gas, bunched and thermalized in a radiofrequency trapping system and finally transferred into a Penning trap where they are captured in flight. While the ions are stored their reaction kinematics with respect to given gases may be studied. The variable storage duration will allow the investigation of short- as well as long-lived trans-actinides, including those with a half-life $T < 100$ ms which are not accessible to gas-chromatographic methods. As a first test of the future gas-phase ion-molecule reactions of the heaviest elements at SHIPTRAP, the oxidation of ruthenium and osmium has been investigated recently [16] at the ClusterTrap, a Penning trap-TOF mass spectrometer [17]. The reaction of osmium up to the dioxide OsO_2^+ was observed and the osmium and osmiumoxide reaction rates were determined [16], although earlier bond-energy values infer a slightly endothermic reaction with oxygen [18]. In addition, the observation of a spontaneous reaction of ruthenium was observed in contradiction to a previously reported endothermic behavior [19] and thus left the results in a status which asked for further investigations. In the following, an independent experiment on the ion-molecule reactions of Ru^+ and Os^+ with oxygen is presented, where FT-ICR mass spectrometry was applied.

2. Experimental setup and procedure

The experiments were performed with a commercial FT-ICR mass spectrometer (EXTREL FT/MS 2001-DX), which is equipped with a 2.7-T superconducting magnet and a “dual-cell” trap configuration, i.e., two cubic ICR cells, each of two-inch size. Fig. 1(a) shows a schematic overview of the exper-

imental setup. For the present experiments only the source cell has been used. A scheme of the experimental sequence is shown in Fig. 1(b): Before the production and capturing of the ions the cell was emptied by pulsing the trapping plate potential to -10 V. The osmium and ruthenium ions were produced by laser ablation from a corresponding target with a CO_2 laser at a wavelength of $10.6 \mu\text{m}$. These targets consisted of copper plates where osmium and ruthenium had been electrolytically deposited on [16]. They were mounted on the automatic solids-probe of the spectrometer. The target was set to about 5 mm distance from the trapping plate of the cell and rotated to bring new osmium or ruthenium material to the position of the laser pulse focus on the probe. Both trapping plates may be set to a variable potential of 1 to 10 V (with respect to the excite/detect plates which were on ground potential). In general, a trapping potential of $U_0 = 1$ V has been applied. For capturing, the endcap plate potential is set to 0 V to allow the ions to drift into the source cell. $80 \mu\text{s}$ after the laser pulse, i.e., just before the ions can leave the cell, the trapping plate potential is reset to 1 V. In addition to the laser ablation from the target, ions can be produced by ionization of gas particles in the cell volume by 70-eV electrons from an electron gun. The electron beam is applied for about 200 ms and since the ions are produced inside the ion trap the potentials need not to be changed during the electron bombardment.

The stored ruthenium or osmium ions were subjected to the oxygen gas, which is introduced to the cell through a leak valve at a pressure range of 4×10^{-8} mbar to 2×10^{-6} mbar (the pressure in the cell volume was determined as described in the Appendix). In addition to the reaction gas, a colli-

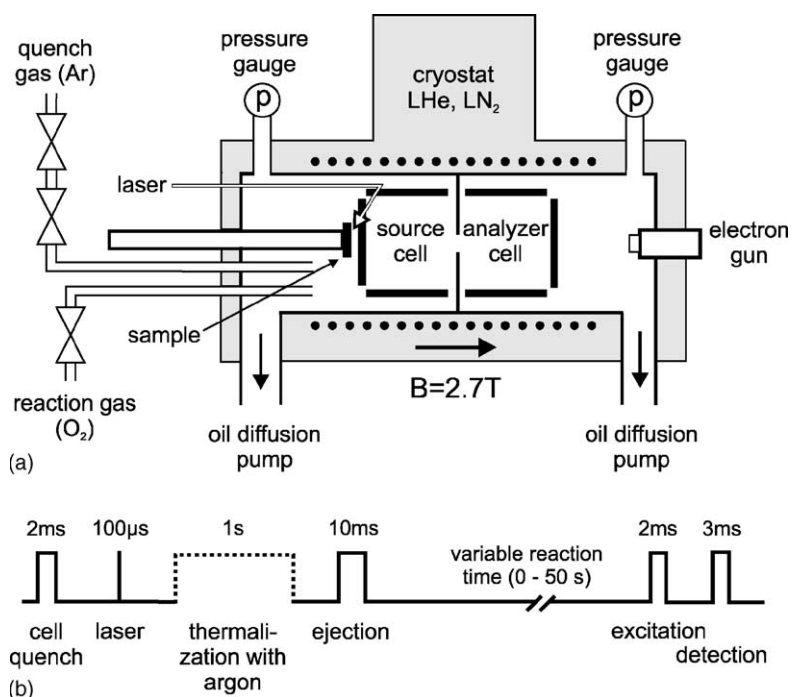


Fig. 1. (a) Overview of the experimental setup. (b) Experimental sequence. The thermalization with argon (dashed line) was not applied in all measurements.

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