Transactinides

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Thermochemical and Physical Properties of Element 112**

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The high nuclear charges of the heaviest elements influence their electronic structure and hence their chemical properties.[1–5] The experimental results so far obtained for the heaviest chemically investigated elements reveal that seaborgium,[6,7] bohrium,[8] and hassium[9] behave as typical representatives of the corresponding Group 6, 7, and 8 of the periodic table. Apparently, relativistic effects do not introduce a perceptible destabilization of the highest oxidation states of these elements in the chemical environments they have been studied in. An even stronger increase of relativistic effects is predicted for the transactinides of Groups 12–18.[1–3] Element 112, a representative of Group 12 of the periodic table, has a predicted closed-shell electronic ground-state configuration of $[Rn]5f^{14}6d^{10}7s^2$, rendering this element one of the key elements regarding relativistic effects in the electronic structure.[3,5,10] Recently, our gas chromatography experiments with only two observed atoms of element 112 revealed evidence for a metallic adsorption interaction with the stationary gold surface.[11] Here we present new experimental results with an increased number of observed atoms that confirm these observations and improve their statistical significance. From the complete data set, thermochemical and physical data are deduced for element 112 and compared to the corresponding properties of its homologues in Group 12: Zn, Cd, and Hg. The increased stabilization of the atomic state of element 112 reveals a further enhancement of the relativistic effects with increasing atomic number Z in Group 12.

For element 112 a noble-metallic character was predicted from empirical extrapolations.[12,13] Relativistic atomic calculations revealed a contraction of the spherical 7s orbitals, leading to the prediction of an enhanced stability of the elemental atomic state for element 112. Accordingly, a noble-gas-like behavior was postulated.[14] Modern calculation methods confirmed a stronger binding of the 7s orbitals.[3] However, the spin–orbit coupling of the 6d orbitals is predicted to lead to an electronic ground state configuration with a $6d^{5/2}$ orbital that is similar energetically and spatially to the 7s orbital, indicating that element 112 could be a noble transition metal[5,15] or even a semiconductor.[16] Based on these strongly differing predictions, it is decisive for experimentalists to be able to distinguish in chemical experiments whether element 112 behaves more as a noble metal or as a noble gas. Therefore, investigation of gas adsorption properties of element 112 on metal surfaces has been suggested.[17] In such studies the energy content of the adsorption bond between element 112 and the metallic stationary phase, the standard adsorption enthalpy at zero surface coverage (ΔH_{ads}^{Au}), is determined. This quantity differentiates between metal-bond formation and weak van der Waals physisorption interaction. The semiempirical macroscopic adsorption model based on the Miedema approach[18] was improved[13] and predicts a weaker metallic interaction of element 112 with gold than for Hg. Relativistic calculations employing the interaction of atomic element 112 with gold clusters also point to a metallic interaction.[19] An adhesion model for noble gases on metals was extended to estimate the physisorption interaction of a noble-gas-like element 112 with gold[20] (Table 1, ΔH_{ads}^{Au} calcd).

In the first experiments performed in 2001–2004[21–24] an unambiguous understanding of the chemical properties of element 112 was not achieved. In 2006 a series of experiments at FLNR Dubna made possible for the first time the unambiguous detection of element 112 after its chemical separation.[11] These chemical experiments with element 112

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Table 1: Experimental and calculated thermochemical data for Group 12 elements and radon.

	$-\Delta H_{\mathrm{ads}}{}^{\mathrm{Au}}$ [kJ mol^{-1}]		ΔH_{subl} [kJ mol^{-1}]		B.p. [K]
	Exptl	Calcd	Exptl	Calcd	
Rn	27 ± 3[20]	27[20]	20[26]		211[25]
Zn			130[25]		1180[25]
Cd			111[25]		1040[25]
Hg	98 ± 3[27]		64[25]		630[25]
112	52^{+4}_{-3}[a] 52^{+46}_{-7}[11]	12[11] 30 ± 5[20] 67[19] 78[20] 84 ± 10[13]	38^{+10}_{-12}[a]	22[12] 39 ± 3[13] 110 ± 3[16]	357^{+112}_{-108}[a]

[a] This work.

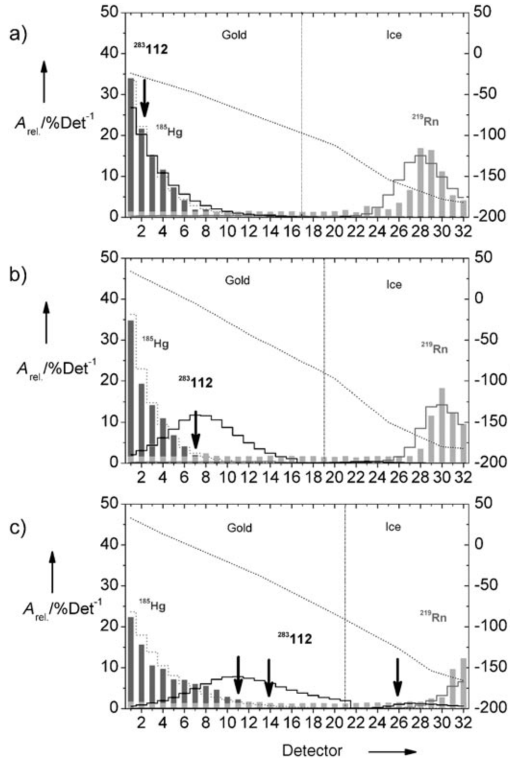

Figure 2. Thermochromatographic deposition patterns of ^{185}Hg, ^{219}Rn, and ^{283}112 in the COLD, dependent on experimental parameters. The measured relative activity per detector ($A_{\mathrm{rel.}}$, left-hand axis) of ^{185}Hg (dark gray bars, MCS: gray dashed line) and ^{219}Rn (gray bars, MCS: gray solid line) is shown. The positions of the detected ^{283}112 atoms are indicated (black arrows, MCS: black solid line). The temperature gradient is shown (black dashed line, right-hand axis). Experimental parameters: a) gas flow 860 mL min^{-1}, temperature gradient from −24 to −184 °C; b) gas flow 890 mL min^{-1}, temperature gradient from 35 to −180 °C; c) gas flow 1500 mL min^{-1}, temperature gradient from 32 to −164 °C. The beginning ice coverage of the detectors at −95 °C is indicated by the vertical line.

were focused on the investigation of the isotope ^{283}112, which was observed in the heavy ion induced nuclear fusion reaction of ^{48}Ca with ^{238}U.[28] It decays with a half-life of about 3.8 s by alpha-particle emission ($E_\alpha = 9.54$ MeV) to ^{279}Ds, which undergoes spontaneous-fission (SF) decay with a half-life of 200 ms. These decay properties of the isotope ^{283}112 were confirmed in 2007 by using the same reaction with the SHIP separator at the Gesellschaft für Schwerionenforschung (GSI) in Darmstadt, Germany.[29] Even larger production rates have been observed for ^{283}112 by using the indirect production path ^{242}Pu(^{48}Ca,3n)^{287}114 ($t_{1/2} = 0.48$ s, $E_\alpha = 10.02$ MeV)$\rightarrow^{283}$112.[28] Thus, this reaction was used in this work. The chemical setup applied in the experiments was based on the thermochromatographic in situ volatilization and online detection technique (IVO) in combination with the cryo-online detector (COLD; see references [8,11,24] for details). From the observation of only two atoms (Figure 1, chain 1 and 2) related unambiguously to the isotope ^{283}112, a first indication of a volatile metallic element 112 was obtained.[11] In 2007 we were able to increase the overall efficiency by a factor of almost three by shortening the transport time and increasing the transmission of the target grid (see Experimental Section). We observed three additional decay chains of ^{283}112 (Figure 1, chains 3–5).

The obtained thermochromatographic deposition patterns for ^{185}Hg, ^{219}Rn, and ^{283}112 at varied experimental conditions are depicted in Figure 2 and represent a characteristic example for the gas chromatographic behavior of single atoms:

- In the first experiment in 2006[11] a carrier-gas flow of 860 mL min^{-1} and a temperature gradient between −24 and −184 °C were established (Figure 2 a). A spontaneous, diffusion-controlled deposition of ^{185}Hg was observed in the first eleven detectors. ^{219}Rn deposited almost entirely on the last nine detectors. Under these conditions one decay chain (chain 1) related to ^{283}112 was observed on the second detector (−28 °C) together with ^{185}Hg, thus clearly distinct from ^{219}Rn.
- In the second part of the experiment in 2006[11] the temperature at the start of the chromatographic detector was increased, leading to a steeper temperature gradient between 35 and −180 °C (Figure 2 b). The gas flow rate was 890 mL min^{-1}. Under these conditions ^{185}Hg revealed a shorter deposition pattern up to detector eight, consistent with an increased diffusion coefficient of Hg in the carrier gas at higher temperatures. Only 70 % of the ^{219}Rn was deposited in the last six detectors owing to the slightly increased temperature in the last detectors. A decay chain (chain 2) attributed to ^{283}112 was observed on detector 7 (−5 °C), again clearly distinct from ^{219}Rn.
- In the 2007 experiment (Figure 2 c) the gas flow rate was increased to 1500 mL min^{-1} to increase the transport

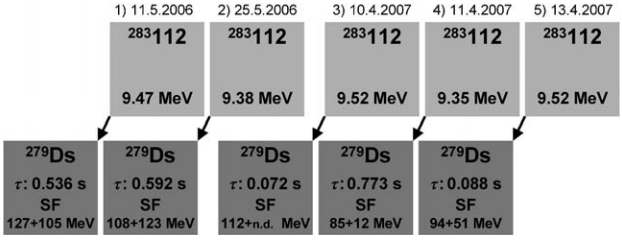

Figure 1. Detected α–SF decay chains assigned to the isotope ^{283}112; n.d. = not determined.

efficiency. The temperature gradient was held between 32 and −164 °C. Hence, the ^{185}Hg deposition region broadened drastically to 14 detectors. Only about 30 % of the ^{219}Rn deposited on the last four detectors. The faster carrier gas transported also the three observed atoms of ^{283}112 further down to detectors of lower temperature. Two of the atoms (chain 3 and 5) deposited on the detectors 11 (−21 °C) and 14 (−39 °C). The third atom (chain 4) was transported even more downstream and deposited on detector 26 (−124 °C). From results of permanent dew point measurements in the carrier gas, it has to be assumed that a thin ice layer was covering the detector surfaces held below −95 °C (Figure 2 a–c, vertical lines, –•–•). Therefore, we conclude that four events (chains 1–3 and 5) are attributed to atoms of element 112 deposited on the gold surface, and one event (chain 4) represents an atom of element 112 deposited on ice.

A microscopic kinetic model of gas chromatography based on Monte Carlo simulations (MCS)[30] was applied to evaluate the most probable standard adsorption enthalpy on gold surfaces at zero surface coverage ($-\Delta H_{ads}{}^{Au}$) from the chromatographic deposition of Hg, Rn, and element 112. The diffusion-controlled deposition pattern observed for Hg (Figure 2, gray dashed lines) allows one to determine a lower limit of $-\Delta H_{ads}{}^{Au}(\mathrm{Hg}) > 65\ \mathrm{kJ\,mol^{-1}}$, which is in agreement with literature data.[27] The ^{219}Rn deposition (Figure 2, gray solid lines) yields an adsorption enthalpy of $19 \pm 2\ \mathrm{kJ\,mol^{-1}}$, which is in good agreement with the value $-\Delta H_{ads}{}^{ice}(\mathrm{Rn}) = 20 \pm 2\ \mathrm{kJ\,mol^{-1}}$ measured for Rn on ice surfaces.[31] The statistical analysis of the deposition behavior of ^{283}112 (Figure 2, black arrows) revealed a standard adsorption enthalpy of element 112 on gold surfaces of $-\Delta H_{ads}{}^{Au}(112) = 52^{+4}{}_{-3}\ \mathrm{kJ\,mol^{-1}}$ (68 % confidence interval). Using this value the calculated deposition pattern (Figure 2 c, black lines) indicates for the highest gas flow rates a probability of about 15 % that atoms of element 112 reach detector 21 and continue the chromatographic process on ice. This explains the observation of chain 4 on detector 26. The expected number of randomly correlated decay chains of the observed type $\alpha_{9.5\pm0.2\,\mathrm{MeV}}-\mathrm{SF}_{E>50\,\mathrm{MeV}}$ during the entire experiment was calculated to be 0.05. The observed enhanced adsorption enthalpy of element 112 on gold compared to the predicted adsorption enthalpy using a physisorption model[20] indicates a metallic-bond character involved in the adsorption interaction between element 112 and gold. The calculations of the 112–Au interaction potential using density functional methods[15,19] and the macroscopic semiempirical Eichler–Miedema adsorption model[13] predict a metallic-bond character, but seem to overestimate the energetic content of this metallic adsorption bond (Table 1). However, element 112 is chemically not as inert as a noble gas, as was suggested by Pitzer in 1976.[14]

An empirical link between the single-atomic (or molecular) parameter ΔH_{ads} and the volatility of macroscopic amounts of an element (or compound), determined by the standard sublimation enthalpy (ΔH_{subl}), reveals linear relationships for various compound classes on various chromatographic materials.[32] A similar correlation was established between the $\Delta H_{ads}{}^{Au}$ values of elements and their ΔH_{subl} values [Eq. (1)].[33]

$$-\Delta H_{ads}{}^{Au} = (1.08 \pm 0.05)\Delta H_{subl} + (10.3 \pm 6.4)\ \mathrm{kJ\,mol^{-1}} \qquad (1)$$

By using this correlation the standard sublimation enthalpy can be estimated as $\Delta H_{subl}(112) = 38^{+10}{}_{-12}\ \mathrm{kJ\,mol^{-1}}$, which is in excellent agreement with predictions of volatility for element 112[13] (Table 1, ΔH_{subl} calcd). The recent quantum chemical methods seem to underestimate the elemental volatility of element 112.[16] By applying the estimated value for the sublimation entropy $\Delta S_{subl} = 106.5 \pm 2.0\ \mathrm{J\,mol^{-1}K}$,[13] element 112 can be presumed to have a boiling point of $357^{+112}{}_{-108}$ K. These values indicate that element 112 is considerably more volatile compared to its lighter homologues Zn, Cd, and Hg (Figure 3 and Table 1), thus manifesting the preserved trend of increasing stabilization of the elemental atomic state owing to relativistic effects in the electronic structure along Group 12 of the periodic table up to element 112.

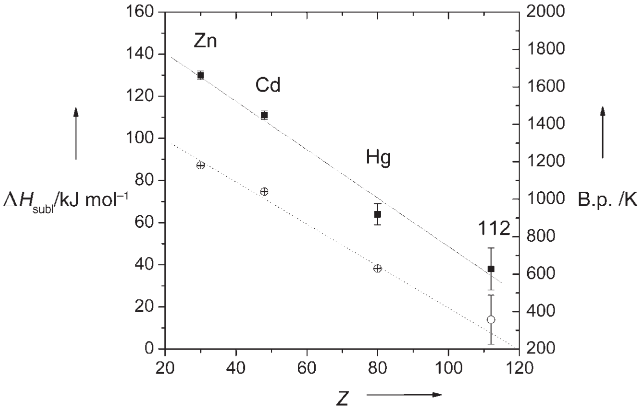

Figure 3. Trends of volatility for Group 12 elements of the periodic table. The standard sublimation enthalpy (ΔH_{subl}; black squares, left-hand axis) and the boiling points (open circles, right-hand axis) are taken as a measure for elemental volatility. The error bars for Zn and Cd are smaller than the symbols. The linear trends (dashed lines) are included to guide the eyes.

Experimental Section

In 2007 at the U-400 cyclotron at FLNR Dubna a stationary grid-supported ^{242}PuO$_2$ target (ca. 1.4 mg cm^{-2}) containing 50 µg cm^{-2} Nd in natural isotopic composition was irradiated with ^{48}Ca (278 ± 3 MeV). The projectile energy in the target material was calculated with SRIM2003[34] to be 227–242 MeV. During 16 days of experiment an overall beam dose of $3.1\cdot10^{18}$ ^{48}Ca particles were accumulated on the target. The target grid transmission was about 85 %. Neodymium was added to the target to produce the alpha-decaying ^{185}Hg ($t_{1/2} =$ 49 s) throughout the entire experiment. Additionally, several isotopes of radon, for example, ^{219}Rn ($t_{1/2} = 3.96$ s), were produced in multinucleon transfer reactions of the projectile with the target material. The recoiling nuclear reaction products were stopped in a recoil chamber covered by a quartz inlay (except for a copper beam stop). Subsequently, they were swept with the carrier gas mixture (He/Ar = 1:1, 1500 mL min^{-1}) through an oven heated to 850 °C and containing a quartz column with a quartz wool filter (for removal of aerosol particles) and Ta metal (for removal of O_2 and H_2O traces; the measured dew point of $-95\,^{\circ}\mathrm{C}$ corresponds to a water content of 0.2 ppm). Therefore, only nonreactive volatile products were transported further through the 8-m-long PFA teflon capillary to the COLD. In this thermochromatographic detector, 32 sandwiched pairs of silicon PIPS detectors ($9.7\times9.7\ \mathrm{mm}^2$ active area) were arranged in a linear array, forming a rectangular chromatographic column with an opening of $11.6\times1.5\ \mathrm{mm}^2$ with about 80% active detector area. One side was covered with a gold layer of thickness 50 nm. This detector array was mounted into a vacuum-tight channel. The temperature gradient along this channel was established by using a thermostat heating at the warm entrance of the channel and a liquid nitrogen cryostat cooling at the exit. This thermochromatography detector allowed for time-resolved alpha particle and spontaneous fission (SF) fragment spectroscopy with a temperature resolution of about $5\,^{\circ}\mathrm{C}$. The favorable detection geometry permitted online identification of single atoms deposited on the detector surface with a detection efficiency of about 100% and 86% for SF decay and α decay, respectively.

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