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Superheavy Element Flerovium (Element 114) is a Volatile Metal

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ABSTRACT: The electron shell structure of superheavy elements, i.e., elements with atomic number \( Z \geq 104 \), is influenced by strong relativistic effects caused by the high \( Z \). Early atomic calculations on element 112 (copernicium, Cn) and element 114 (flerovium, Fl) having closed and quasi-closed electron shell configurations of \( 6d^{10}7s^2 \) and \( 6d^{10}7s^27p_{1/2} \), respectively, predicted them to be noble gas-like due to very strong relativistic effects on the \( 7s \) and \( 7p_{1/2} \) valence orbitals. Recent fully relativistic calculations studying Cn and Fl in different environments suggest them to be less reactive compared to their lighter homologs in the groups, but still exhibiting a metallic character. Experimental gas-solid chromatography studies on Cn and Fl in different environments suggest them to be less reactive compared to their lighter homologs in the groups, but still exhibiting a metallic character. Experimental gas-solid chromatography studies on Cn have, indeed, revealed a metal-metal bond formation with Au. In contrast to this, for Fl, the formation of a weak bond upon physisorption on a Au surface was inferred from first experiments. Here, we report on a gas-solid chromatography study of the adsorption of Fl on a Au surface. Fl was produced in the nuclear fusion reaction \( ^{244}\text{Pu}(^{48}\text{Ca}, 3\text{-}4\text{n})^{288,289}\text{Fl} \) and was isolated in-flight from the primary \( ^{48}\text{Ca} \) beam in a physical recoil separator. The adsorption behavior of Fl, its nuclear \( \alpha \)-decay product Cn, their lighter homologs in groups 14 and 12, i.e., Pb and Hg, and the noble gas Rn were studied simultaneously by isothermal gas chromatography and thermochromatography. Two Fl atoms were detected. They adsorbed on a Au surface at room temperature in the first, isothermal part, but not as readily as Pb and Hg. The observed adsorption behavior of Fl points to a higher inertness compared to its nearest homolog in the group, Pb. However, the measured lower limit for the adsorption enthalpy of Fl on a Au surface points to the formation of a metal-metal bond of Fl with Au. Fl is the least reactive element in the group, but still a metal.

1. Introduction

Superheavy elements (SHE) are unique in two respects. Their nuclei exist only due to nuclear shell effects, and their electron structure is influenced by increasingly important relativistic effects. Syntheses of SHE with proton number \( Z \) up to 118 have been reported. Elements with \( Z=104-112 \) are members of the \( 6d \) series in the Periodic Table of the Elements. The \( 7p \) valence shell is expected to be filled in the elements with \( Z=113-118 \). The discovery of elements with \( Z=114 \) and \( Z=116 \) was recently officially accepted and they were named flerovium (Fl) and livermorium (Lv), respectively. Lighter transactinides with \( Z=104-108 \) were experimentally shown to be members of groups 4 through 8 of the
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superheavy element chemistry and needs to be solved experimentally in a more sensitive and detailed study.

Here, we report on chemical studies of Fl after preseparation with TASCA, which led to the observation of two Fl atoms.

2. Experimental

To isolate Fl we exploited a combination of the TransActinide Separator and Chemistry Apparatus (TASCA) and the Cryo Online Multidetector for Physics And Chemistry of Transactinides (COMPACT). TASCA served for suppression of the primary beam and of the background from Rn isotopes and their decay products. This allowed the observation of the characteristic radioactive decay of Fl and its (grand)-daughters under significantly improved background reduction compared to previous studies performed without preseparation.

The gas chromatography and detection system comprised two COMPACT detector arrays which were placed behind TASCA. The experimental set-up is schematically shown in Figure 1.

![Figure 1](image)

**Figure 1.** Schematic drawing of the TASCA-COMPACT arrangement used for the gas chromatographic investigation of the volatility of Fl and its reactivity towards a Au surface. The ⁴⁸Ca beam (1) passed through the rotating ²⁴⁴Pu-target (2) assembly. The separator TASCA consists of one dipole (D), where unwanted nuclear reaction products (3) were deflected, and two quadrupole (Q) magnets. At the exit of TASCA a vacuum window (4) separated the low-pressure required in TASCA from the high-pressure applied in the recoil transfer chamber (RTC) (5). After passing the vacuum window, Fl was thermalized in the gas inside the RTC and was transported in its elemental state with the carrier gas through a 2-cm long PTFE tube (6) into a series of two COMPACT detector arrays (7) connected by a 30-cm long polytetrafluoroethylene (PTFE) capillary (8) (2 mm i.d.). A negative temperature gradient was applied along COMPACT II using a liquid nitrogen cryostat (9) at the exit.

A ⁴⁸Ca beam of typically 2·10¹² particles·s⁻¹ was accelerated by the UNILac Versal Linear ACcelerator (UNILAC) at the GSI, Darmstadt, Germany, to an energy of 259.4 MeV. In total, a projectile dose of 4·10¹⁴ particles was collected. The projectiles first passed through (2.50±0.05)-µm thick Ti targets and were deflected in the narrow channel. Depending on their incidence angle, they pass through different effective thicknesses of detector dead layer and gas, thus undergoing energy loss to a different degree, before entering the active detector area.

Volatile species including Fl, which was transported in its elemental state, were flushed with this carrier gas mixture through a 2-cm long PTFE tube (3 mm inner diameter, i.d.) into the first of the two COMPACT detector arrays (Figure 1). Each array consisted of 32 pairs (gap: 0.6 mm) of (1 x 1) cm²-large positive-intrinsic-negative (PIN) epitaxial silicon photodiodes with an active area of (9.7 x 9.8) mm² and an effective thickness of 150 µm. The calculated geometrical efficiencies for detecting an α particle or spontaneous fission from atoms present inside a detector array were 93% and ~100%, respectively. All detectors were covered by a (35-50)-μm thick Au layer deposited by evaporation. COMPACT I was operated as an isothermal chromatography (IC) detector array at room temperature (21 °C). It retained metallic elements that form a strong chemical bond with Au at room temperature, such as Pb or Hg. Chemical species that did not adsorb in COMPACT I exited, passed through a 30-cm long PTFE capillary (2 mm i.d.) and entered COMPACT II. COMPACT II was added five days after the start of the 29-day long experiment. A negative temperature gradient from +20 to −162 °C was applied along COMPACT II (thermochromatography detector array, TC) using a liquid nitrogen cryostat at the exit. For the first three days of operation, the lowest temperature in COMPACT II was −86 °C due to a weak thermal contact between the detector array and the cryostat. Volatile and inert elements pass COMPACT I at characteristic low temperatures. The energy resolution of the COMPACT detectors was ≈ 120 keV (FWHM). A higher resolution could not be reached because α particles are emitted isotropically at various angles in the narrow channel. Depending on their incidence angle, they pass through different effective thicknesses of detector dead layer and gas, thus undergoing energy loss to a different degree, before entering the active detector area.

excitation energy, E*, of 40-45 MeV were formed. After deexitation by evaporation of three or four neutrons, the resulting ²⁸⁸,²⁹⁰Fl nuclei recoiled from the target into the separator TASCA operated in the “Small Image Mode”. The primary beam and unwanted nuclear reaction products were deflected inside the dipole magnet to a beam stop, while Fl was guided to the focal plane (Figure 1). Magnets were set to focus ions with a magnetic rigidity, B·p, of 2.27 T·m into a ~(3 x 5) cm² area in TASCA’s focal plane. Monte Carlo simulations indicate that 35% of the Fl ions reached this area. There, they penetrated a (40x30) mm²-large MYLAR™ window of (3.3±0.1) μm thickness mounted on a 1-mm thick supporting grid of 80% geometrical transparency and entered the Recoil Transfer Chamber (RTC). The window separated the low-pressure region in TASCA (0.5 mbar) from the high-pressure one in the RTC (~900 mbar). In the 29-cm large RTC, made from polytetrafluoroethylene (PTFE), the Fl ions were thermalized in a dried (measured dew point below −60°C) gas mixture (He:Ar=70:30; gas purities: 99.9999% (He) and 99.9999% (Ar)), which flushed the RTC at a total flow rate of 1300-1800 mL·min⁻¹. The Ar admixture (30%) in the carrier gas served to increase gas slowing power inside the RTC, which allowed minimizing the RTC volume. Short-lived Hg and Pb isotopes, chemical homologues of Cn and Fl, were produced using ¹⁴²Nd and ¹⁴⁴Sm targets, respectively. By producing pulses of ¹⁸²Hg recoils (0.4 s beam on and 50 s beam off) and measuring the time-delay until their decay in COMPACT, the most probable transport time to COMPACT I was determined to be (0.81±0.06) s at a gas flow rate of 1300 mL·min⁻¹.

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Accordingly, α peaks show characteristic low-energy tailing. All transported species came in contact inside and downstream of the RTC exclusively with PTFE and Au surfaces. This set-up allowed for detecting species in a wide range of volatilities, namely from the low-volatile Pb to the noble gas Rn. If Fl behaves like a metal it will adsorb in COMPACT I; if, in contrast, Fl rather behaves like a noble gas, it will deposit at a much lower temperature in COMPACT II. The overall transfer yield from TASCA to COMPACT was measured with short-lived Hg and Pb isotopes. To this end, the rate at which Hg atoms entered the RTC was determined by implanting them into a (58 x 58) mm² double-sided silicon strip detector. A subsequent experiment, in which the atoms were thermalized in the RTC and transported to COMPACT at a flow rate of 1300 mL·min⁻¹, yielded that the decay of 27% of all Hg atoms entering the RTC was observed in COMPACT. For Pb isotopes this value was lower (20%), due to additional adsorption losses of the less volatile Pb on the walls of the RTC and the connecting tube. The distributions of ¹²⁴Hg and ¹³⁸Pb isotopes in COMPACT were measured before and after the Fl measurement and found to be reproducible. Between these measurements, neither of the two COMPACT arrays mounted in the gas loop was opened. COMPACT II was warmed up every 2-3 days to remove the thin ice layer which formed on detectors held at temperatures below -75°C.

3. Results

A search for correlated decay chains starting from ²⁸⁵,²⁸⁶Fl was performed. The search conditions were the following: in case of ²⁸⁵Fl, we searched for a 9.6-10.1 MeV α particle, followed within 1 s by a >20 MeV fission fragment, which was registered in either the same or an adjacent detector pair, where the first α particle was found. For ²⁸⁶Fl, we searched for a 9.6-10.1 MeV α particle, followed by a 8.8-9.3 MeV α particle, followed by a >20 MeV fission fragment, all within 200 s. The search was extended to all detector pairs downstream of the one where the mother decay was observed, because the daughter of Fl, Cn, is known to be a volatile metal and thus can be transported by gas flow along the detector channel. This procedure revealed two correlated decay chains, which we show in Figure 2.

Both members of chain #1 were observed in detector pair #9 in COMPACT I, which was kept at 21°C. The members of chain #2 were found distributed over both COMPACT arrays: the α particle of the mother nucleus initiating the chain was detected in detector #9 “top” in COMPACT I. The last two members of the chain were detected in detector pair #52 in COMPACT II at ~32°C. Based on the good agreement of the nuclear properties of our observed chains with decay properties reported from ²⁸⁵,²⁸⁶Fl synthesis experiments, we assign chain #1 to the decay of ²⁸⁵Fl→²⁸⁵Cn and chain #2 to ²⁸⁶Fl→²⁸⁶Cn→²⁸⁶Ds, produced in the 4n and 3n evaporation channel, respectively. A search for SF decays (E > 20 MeV) revealed two additional events. In total, only these four SF events were registered - all with two coincident fission fragments. No “single” fission fragment with E_{fiss} > 20 MeV was detected. The SF events, for which no α-decay precursor was found, appeared at temperatures of ~21°C (COMPACT I, det. 17/1B, 83/83 MeV) and ~86°C (COMPACT II, det. 64T/64B, 24/85 MeV). A definite assignment of these two SF events to a certain element is not possible as SF is an unspecific decay mode.

The observed α energies in the decay chains are somewhat lower compared to the energies registered in earlier experiments in a focal plane detector, into which the ²⁸⁵,²⁸⁶Fl were implanted. This is due to energy loss in the gas layer, which the α particles penetrate in roughly half of all cases, and is in agreement with a long tail of α peaks towards the low energy side (Figure 3). Such an effect was observed in all our previous chemistry experiments, where similar cryodetectors were used. The asymmetric broadening of α peaks towards low energy can be understood as being due to the energy loss of particles penetrating the gas channel at shallow angles.

![Figure 2. Observed correlated decay chains assigned to Fl. Left-hand side of the boxes: α-particle energy, lifetime of the nucleus, and detector number in which the signal was measured (“T” and “B” are top and bottom detectors of a detector pair, respectively). (I)/(II) denote COMPACT I and COMPACT II, respectively.](image)

![Figure 3. Spectra measured in detector #9T of COMPACT I (left panel) and detector #52B of COMPACT II (right panel) during 22 days. α decays from ²⁸⁵Fl and ²⁸⁶Ds are shown in red.](image)
the members of chain #2, are marked, distributed over two COMPACT detectors. The only peaks visible in these spectra arise from the decay chain $^{219}\text{Rn} \rightarrow ^{215}\text{Po} \rightarrow ^{211}\text{Bi}$. Small amounts of $^{220}\text{Rn}$ were added to the carrier gas to allow for an on-line monitoring of the detection system and to provide α calibration data. No peaks are present in the spectra above the highest energy originating from the $^{219}\text{Rn}$ chain, i.e., above 7.5 MeV. This illustrates nicely the power of physical preseparation by a recoil separator in a chemistry experiment.

4. Discussion

In Figure 4 we show the temperature profile in the main part of the experiment (panel a) together with the measured distribution (solid bars) of Pb (panel b), Hg (c) and Rn (d). Also shown are the positions at which the members of the two Fl decay chains were observed (e). Monte Carlo simulations \(^{41}\) (MCS) of the migration of Pb, Hg, Rn, Cn, and Fl along the chromatography detectors were performed with 10000 atoms for each element. The distribution pattern of Rn in COMPACT was simulated using the literature value ($-\Delta H_{\text{ads}}(\text{Rn}) = 20 \text{ kJ mol}^{-1}$, ref. 28). Pb and Hg interact strongly with a Au surface ($-\Delta H_{\text{ads}}(\text{Pb}) > 295 \text{ kJ mol}^{-1}$ (ref. 42), and $-\Delta H_{\text{ads}}(\text{Hg}) = (98 \pm 3) \text{ kJ mol}^{-1}$, (ref. 20)), and their deposition temperatures on a Au surface are well above room temperature. The similarity in the observed distribution patterns for Hg and Pb, which have significantly different adsorption enthalpies on a Au surface, points at the diffusion-controlled nature of the adsorption process. The diffusion to the wall controls the process for both Pb and Hg, and they adsorb upon first contact with the surface at the beginning of the experiment (panel a) together with the measured distributions (bars) of $^{185}\text{Pb}$ (panel b), $^{182}\text{Hg}$ (panel c), and $^{219}\text{Rn}$ (panel d) together with the temperature profile in the main part of the experiment (panel a) are shown. The positions where the α particles from the members of the decay chains were observed are shown in (c): chain #1 – event in light blue in the histogram, corresponding to the light-blue bordered inserted box; chain #2 – events in red / red-bordered boxes. The fissions terminating the chains were observed in the same detector pairs as precursor α particles from Fl in chain #1 or from Cn in chain #2. The dashed lines show the results of Monte Carlo simulations for Rn and Cn using literature values (ref. 28/ ref. 19) for $-\Delta H_{\text{ads}}$.

In chain #1 originating from $^{288}\text{Fl}$, the SF decay from $^{284}\text{Cn}$ was registered, after a lifetime of 650 ms, in the same detector pair as the α decay of the mother nuclide $^{288}\text{Fl}$. A $^{284}\text{Cn}$ atom interacting with a Au surface with $-\Delta H_{\text{ads}}(\text{Cn}) = 52 \text{ kJ mol}^{-1}$ needs about 135 ms to be transported 1 cm downstream in COMPACT I by the carrier gas. Within 650 ms, a $^{284}\text{Cn}$ atom, either residing adsorbed on the surface or being immersed in the gas would be transported a few centimeters downstream the detector channel. The fact that $^{284}\text{Cn}$ remained at the same position during its entire lifetime is indicative for its implantation into the detector as a recoiling atom in the α decay of $^{288}\text{Fl}$. This is expected in about 50% of all α decays. It is due to the nuclear recoil from the α decay of the mother atom with the α particle being emitted away from the surface of the detector on which the mother atom is adsorbed. As the recoil range of α-decay products is very small, this implies that indeed the mother atom, $^{288}\text{Fl}$, was adsorbed on the detector surface when it decayed, and that the position where the decay was observed is indicative of a chemical interaction of Fl with the Au surface. In chain #2 originating from $^{289}\text{Fl}$, the last two members, starting from the daughter nucleus $^{285}\text{Cn}$, were found in detector pair #52 at a temperature of $\sim 32^\circ \text{C}$. Apparently, upon α decay of
the mother nucleus, the daughter $^{285}$Cn recoiled into the gas stream. During its lifetime of 11.6 s, it was transported along the detector channel into COMPACT II. As shown in Figure 4(e), the observed adsorption position for $^{285}$Cn agrees well with the calculated deposition pattern for this Cn isotope using the experimentally measured adsorption enthalpy.$^{19}$ This corroborates our assignment of the last two members to $^{285}$Cn $\rightarrow$ $^{281}$Ds and hence that of the mother being $^{289}$Fl.

Both observed Fl decays were registered in the isothermal section, in COMPACT I, while zero decays were observed in COMPACT II. Considering the low experimental statistics, a method of calculating confidence intervals for experiments with small event numbers was applied.$^{43}$ The numbers of events, which were detected in COMPACT I and COMPACT II, are $D=2$ and $N=0$, where we use the notation as in ref. 43. For the evaluation of confidence levels using Poisson statistics, $D$ and $N$ can vary from 0 to 2, with $D+N=2$. Lower ($R_{lo}$) and upper ($R_{hi}$) limits for the ratio $R = N/D$ can be calculated for different confidence intervals, as well as the most probable value of $R$, $R_{max}$ (ref. 43). Thus, the upper limit $R_{hi}$ corresponds to the maximum value of $N$, $N_{hi}$, within the selected confidence interval, and therefore, to the minimum value of $D$, $D_{lo}$. Similarly, the lower limit $R_{lo}$ corresponds to the minimum value of $N$, $N_{lo}$, and to the maximum value of $D$, $D_{hi}$, within the selected confidence interval. From the experiment we obtained the most probable value of $R$ as $R_{max} = 0$, resulting in limits $R_{lo}=0$ and $R_{hi}=1.650$ for the 95% confidence level (95% c.l.). These values correspond to $N_{hi}=1.24$ and $D_{lo}=0.76$. Thus, the experimental values $D$ and $N$ are Poisson-distributed within the intervals: $0.76<D<2$ and $0<N<1.24$ at the 95% confidence level. The minimum value for the number of events $D$, which is detected in COMPACT I at this limit, is 0.76 out of 2, i.e., 38%. To convert this into a limit for $-\Delta H_{ads}^{0}(\text{Fl})$, the deposition pattern for both observed Fl isotopes along the entire COMPACT array was simulated for various values of $-\Delta H_{ads}^{0}(\text{Fl})$ using MCS.$^{41}$ All simulations with $-\Delta H_{ads}^{0}(\text{Fl}) \geq 48 \text{ kJ/mol}$ resulted in distributions where at least 38% of all events decayed in COMPACT I. Therefore, $-\Delta H_{ads}^{0}(\text{Fl}) > 48 \text{ kJ/mol}$ was found as a lower limit for the adsorption enthalpy of Fl on a Au surface at 95% c.l. Similar calculations were performed for 90% and 68% confidence intervals, resulting in limits of 49 kJ/mol and 50 kJ/mol, respectively.

5. Conclusion

A gas phase chromatography experiment with Fl was performed. Two atoms were registered. The observed behavior of Fl in the chromatography column indicates that Fl is less reactive than Pb. The estimated minimum value of $-\Delta H_{ads}^{0}(\text{Fl}) > 48 \text{ kJ/mol}$ reveals a metallic character upon adsorption on a Au surface due to the formation of a metal-metal bond, which is at least as strong as that of Cn.$^{15,19}$ The observed behavior is in agreement with results of recent fully-relativistic calculations on the adsorption of Fl on a Au surface$^{13-15}$, but disagrees with an observation of adsorption of Fl on a Au surface merely due to physisorption.$^{27}$ To conclude, the present experimental study has established that Fl is a volatile metal, the least reactive one in group 14. It is, however, not as inert as a noble gas, as was initially assumed from atomic calculations.$^{7}$
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Author Contributions
The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

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ABBREVIATIONS
COMPACT, Cryo-Online Multidetector for Physics And Chemistry of Transactinides; IC, Isothermal Chromatography; MCS, Monte Carlo Simulation; PTFE, polytetrafluoroethylene; TASCAtm TransActinide Separator and Chemistry Apparatus; TC, Thermochromatography
The electronic structure of superheavy elements ($Z \geq 104$) and their chemical properties are dominated by relativistic effects. Recently two superheavy elements were recognized by the IUPAC and named flerovium (Fl, $Z=114$) and livermorium (Lv, $Z=116$). Fl is the heaviest element with which chemical experiments were performed, based on the observation of single atoms. Here, we report on experiments that help answering the long-standing question whether Fl behaves rather like a noble gas or like a metal.