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Gas Phase Chemistry of Superheavy Elements

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Long-lived isotopes of superheavy elements (SHE) beyond Rf, i.e., with atomic number Z ≥ 104 can be produced via fusion reactions between heavy actinide targets and neutron-rich projectiles at a rate of only single atoms per minute (Z=104) or per week (Z=114). Investigating these neutron-rich SHE nuclei using rapid gas-chemical separation and subsequent on-line detection provides an independent chemical characterization and an alternative separation technique to electromagnetic recoil separators. Approaching the heaviest elements, the coupling of chemistry setups to a recoil separator promises extremely high sensitivity due to strong suppression of background from unwanted species. The use of a combination of two separation techniques, physical pre-separation and gas phase chemistry opens the possibility for investigating new compound classes of superheavy elements [1,2].
Electron shells of SHE are influenced by strong relativistic effects caused by the high value of Z. The lighter transactinides with Z = 104-108 were experimentally shown to be members of groups 4 through 8 of the Periodic Table of the elements [3]. Early atomic calculations predicted copernicium (Cn, element 112) and flerovium (Fl, element 114) to be noble gas-like due to the strong relativistic stabilization of the closed-shell configuration 6d107s2 in Cn, and the very large spin-orbit splitting in 7p AOs resulting in the quasi-closed-shell configuration 7s27p1/22 in Fl [4]. Recent fully relativistic calculations studying Cn and Fl in different environments suggest those to be less reactive compared to their lighter homologues in the group, but still exhibiting metallic character [5]. The dilemma whether Cn and Fl are noble gases or rather noble metals calls for experiments. Experimental gas-chromatography studies on Cn have, indeed, revealed a metal-metal bond formation with gold [6]. In contrast to this, for Fl, the unexpected formation of a weak physisorption bond upon adsorption on gold was inferred from first experiments [7]. The recent gas chromatography study on Fl upon the adsorption on gold was performed exploiting clean Fl samples, provided after the pre-separation with the gas-filled separator TASCA [8]. Two Fl decay chains were detected under background-free conditions. The observed behavior of Fl in the chromatography column is indicative of Fl being less reactive than the nearest homolog Pb. The evaluated lower limit of the adsorption enthalpy −ΔHads(Fl) > 48 kJ/mol (95% confidence level) reveals the formation of a metal-metal bond with Au, which is at least as strong as that of Cn, and thus demonstrates the metallic character of Fl [9].
In other experiments, a first molecule of a compound class, which was previously inaccessible for superheavy elements, was recently investigated. The adsorption behavior of Sg(CO)6 was studied by gas-solid chromatography in comparison with that of its nearest homologs in the group, Mo and W [10-12].
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