

Metal carbonyl complexes – a new compound class accessible for transactinides*

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Gas-phase chemical studies of transactinide elements were so far restricted to simple, thermally stable, inorganic compounds. Metal-carbonyl complexes would provide a link to metal-organic chemistry. Binary, mononuclear, volatile carbonyl complexes are known for all lighter elements of group 6 and 8 of the periodic table. Seaborgium hexacarbonyl has been predicted to be stable [1]. Its experimental study would be interesting, because relativistic effects are predicted to influence the metal-CO bond.

We explored the method of rapid in-situ synthesis of transition-metal carbonyl complexes with short-lived isotopes. First tests were performed at the TRIGA Mainz reactor, using the ²⁴⁹Cf(n,f) reaction. Recoiling fission products were thermalized either in pure N₂ or in a CO/N₂-mixture. All volatile compounds were transported in a gas stream to an activated charcoal trap, which was monitored with a γ -ray detector. Figure 1 shows a typical spectrum from the CO/N₂ measurements.

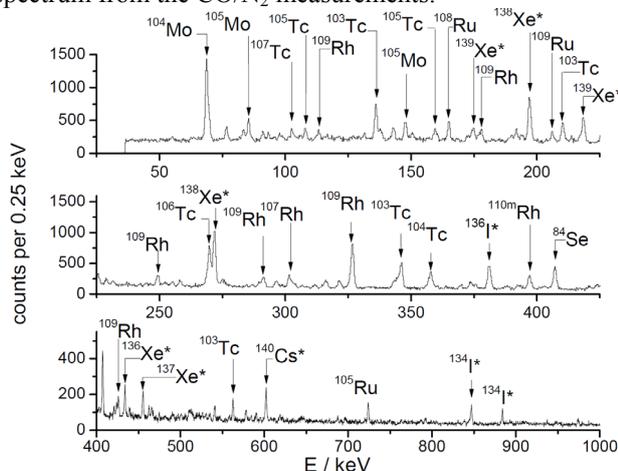


Figure 1: γ -ray spectrum of fission products transported in a CO/N₂ mixture collected for 2 min in a charcoal trap. Subsequently, the sample was measured for 2 min. γ -lines, which were also visible in spectra of pure N₂ experiments, are marked with *.

Short-lived isotopes of Se, Mo, Tc, Ru and Rh were only observed in the spectra when CO was added. These elements form volatile compound with the CO. Transport with cluster (aerosol) material can be excluded.

To test this method under experimental conditions relevant for transactinides, α -decaying ¹⁶³W, ¹⁶⁴W, ¹⁷⁰Os and ¹⁷¹Os were produced in ¹⁴⁴Sm(²⁴Mg,4-5n) and ¹⁵²Gd(²⁴Mg,4-5n) reactions at the gas-filled recoil separa-

* Work supported by the Helmholtz Institute Mainz, the Research Center Elementary Forces and Mathematical Foundations (EMG), the BMBF under contract No. 06MZ223I, and the Swiss National Science Foundation under contract No. 200020 126639 #evenj@uni-mainz.de

tor TASCA. Evaporation residues were separated from the primary beam and from unwanted transfer products within TASCA. They were thermalized in mixtures of He and CO in a Recoil Transfer Chamber (RTC) [2] at the TASCA focal plane. Volatile carbonyl complexes – most likely Os(CO)₅ and W(CO)₆ – were formed in the RTC and were transported with the gas stream to the thermochromatography detector COMPACT [3]. The COMPACT detector array is a chromatography channel consisting of SiO₂ covered PIN diodes, suitable to register α particles emitted from volatile species inside the channel. A negative temperature gradient was applied along the chromatography column. Figure 2 shows thermochromatograms of Os(CO)₅ and W(CO)₆. The measurements are compared to Monte Carlo Simulations. From the deposition patterns of W and Os, adsorption enthalpies of W(CO)₆ of (-46.5 ± 2.5) kJ/mol and $(-43^{+3.5}_{-2.5})$ kJ/mol for Os(CO)₅ were deduced. These values indicate physisorption of these carbonyl complexes on SiO₂.

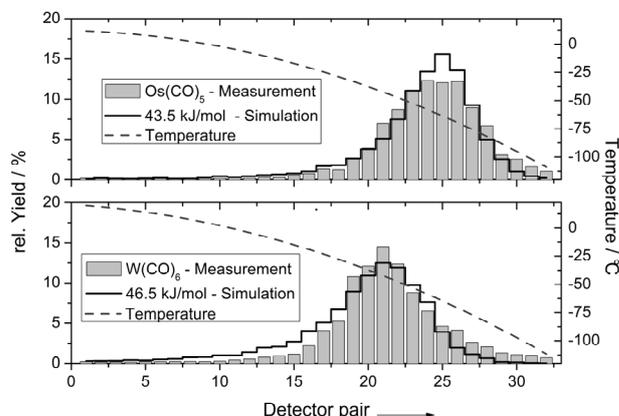


Figure 2: Upper graph: combined thermochromatogram of ¹⁷⁰Os(CO)₅ and ¹⁷¹Os(CO)₅. Lower graph: combined thermochromatogram of ¹⁶³W(CO)₆ and ¹⁶⁴W(CO)₆.

Based on the results of our experiments, Sg(CO)₆ and Hs(CO)₅ are now within reach for transactinide chemistry. These compounds are suitable for chemical characterization by thermochromatography and appear highly promising for nuclear spectroscopy under low background conditions.

References

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