

# Gas Chemical Investigation of Hexafluoroacetylacetonates of Zr and Hf with Preprepared Isotopes

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Gas chemical methods have proven powerful for the investigation of the transactinide elements (TAN,  $Z \geq 104$ ) which are produced in heavy-ion-induced fusion reactions. However, many chemical systems have not been accessible so far due to the plasma caused by the intense heavy-ion beam. Volatile metal complexes containing organic ligands, for example, would be immediately destroyed by this plasma. Recently, preseparation has been used as a new technique to overcome many of the current limitations in TAN chemistry [1,2]. In this technique, nuclear reaction products are separated from the heavy-ion beam and unwanted reaction products in a physical separator, extracted from this separator through a thin window and thermalized in a gas-filled Recoil Transfer Chamber (RTC) [3].

Volatile metal complexes of hafnium (Hf) and zirconium (Zr) with hexafluoroacetylacetonate (hfa) were studied and some preliminary results are reported here. The goal of this work is to later include the heaviest group 4 homolog, rutherfordium (Rf,  $Z=104$ ) and study its behavior in this chemical system.

Sort-lived ( $T_{1/2}$  between 30 s and 8 min) Zr and Hf isotopes were produced at the Lawrence Berkeley National Laboratory 88-Inch Cyclotron. The nuclear reaction products were preprepared in the Berkeley Gas-filled Separator (BGS) [4] and thermalized in the RTC, which was flushed with hfa-vapor-containing helium and kept at a pressure of  $\sim 1$  bar. The reaction products were transported with the gas flow to an oven installed directly at the exit of the RTC. There, formation of volatile complexes of the type  $M(\text{hfa})_4$  ( $M=\text{Zr}, \text{Hf}$ ) occurred. Single molecules of these complexes are volatile at room temperature and were transported with the gas flow to activated charcoal catchers through a  $\sim 5$ -m long Teflon capillary. The absolute overall yield of this process was measured for  $^{162,165,169}\text{Hf}$ . From these data, the overall "reaction plus transport" time was determined. Two settings were investigated: (i) relatively low hfa concentration and a gas-flow rate of 1.3 l/min, and (ii) relatively high hfa concentration and a gas-flow rate of 2.1 l/min. Preliminary results are shown in Fig. 1 for temperatures up to 400°C. At higher temperatures, decomposition of  $\text{hfa}/M(\text{hfa})_4$  was observed. As can be seen in Fig. 1, the minimum overall time with the current setup under the optimized current conditions is about 40 s.

In isothermal chromatography experiments, the whole 5-m long Teflon capillary was used as chromatography column. The relative yield of  $^{162}\text{Hf}$  was measured at different isothermal temperatures. The obtained breakthrough curve is shown in Fig. 2. The experiments were modeled using a

Monte Carlo Simulation (MCS) [5]. An adsorption enthalpy of  $\text{Hf}(\text{hfa})_4$  on PFA Teflon of  $-\Delta H_{\text{ads}} = (57 \pm 3)$  kJ/mol was obtained.

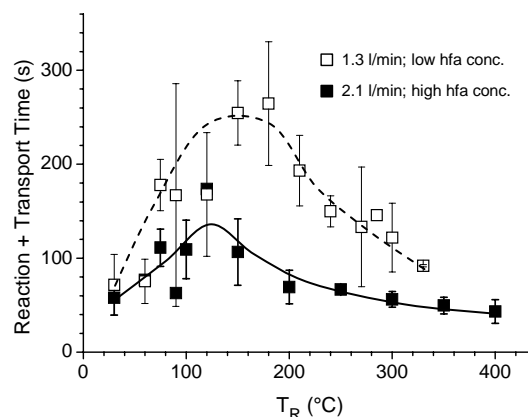


FIG. 1: Total time vs. reaction temperature  $T_R$  for  $\text{Hf}(\text{hfa})_4$ . The lines are shown to guide the eye.

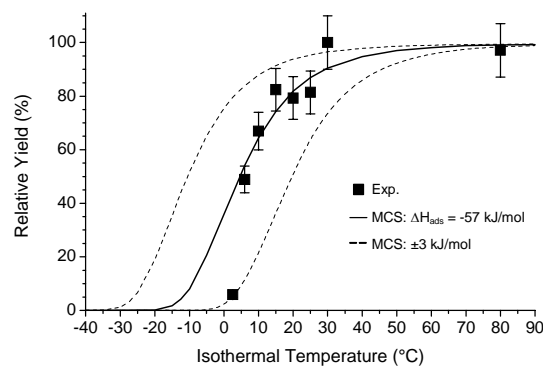


FIG. 2: Isothermal chromatogram for  $^{162}\text{Hf}(\text{hfa})_4$ .

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