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CHEMISTRY OF VOLATILE OXYGEN-BEARING COMPOUNDS FORMED BY U, NP, PU, AM AND CM

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Tracer-scale chemistry has some distinguishing characteristics which makes it differ from common (macro) chemistry. For example, the common redox and self-redox reactions 2Pu(IV)+U(IV)—>2Pu(III)+U(VI) and 2U(V)—>U(VI)+U(IV) that do not occur in extremely diluted solutions as collisions between micro components are practically excluded. Considerable differences in the chemical behaviour micro- and macro quantities of some actinides in the gaseous phase had also been observed. The initial sample contains a tracer quantity of the actinide under study that was adsorbed on the surface of quartz powder. The experiments were carried out using open quartz thermochromatographic (TC) columns (id=3 mm); oxygen served as a reagent and helium was a carrier gas; the flow rate of the gas mixture was 20 cm^3 min^{-1}; the initial sample was heated in a gas stream at 700-750 ˚C and the final temperature was −165 ˚C; the temperature gradient was −18 ˚C cm^{-1}; the duration of each experiment was 30 min. It was found that uranium formed volatile dioxide and trioxide that were adsorbed at 450±25 ˚C and 250±25 ˚C. The values of adsorption enthalpy -ΔHao for UO_2 and UO_3 on quartz were 172±6 and 126±6 kJ mol^{-1} respectively. Two plutonium radioisotopes $^{238,239}Pu$ were used in similar experiments. Contrary to the previous results, plutonium formed three oxides. The centres of their deposition zones were registered at 450±30˚C (PuO2), 250±30 ˚C (P_uO_3) and at negative temperature −105±25 ˚C (PuO_4). The values of -ΔHao for these oxides were 175±7, 122±7 and 47±6 kJ mol^{-1} respectively. Results of the TC isolation of neptunium, americium and curium oxides are also presented. In particularly, Cm formed also three oxides: CmO_2, CmO_3 and CmO_4.

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