

ABSTRACT FORM

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SPECTROPHOTOMETRIC STUDY OF THE STABILITY OF Pu(IV) AND Np(IV) LIMITING CARBONATE COMPLEX.

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Contradictory stoichiometry of transuranian(IV) aqueous complexes in carbonate solutions is published. Still, the chemical composition of the U(IV) limiting complex is well established : it is $U(CO_3)_5^{6-}$. It clearly dissociates into $U(CO_3)_4^{4-}$ when ionic strength or carbonate concentration is decreased. Other species like hydroxocarbonate complexes are also certainly formed in some chemical conditions, but the penta and tetracarbonate complexes are often enough to interpret most of the experimental results on the f elements(IV).

Based on this information and on the analogy between all the actinides, the aim of this work is to compare the stability of the U, Np and Pu(IV) carbonate limiting complex. For this, the dissociation of the limiting complex of Pu(IV) is investigated by spectrophotometry. The initial sodium carbonate solution is transformed into sodium bicarbonate by CO_2 bubbling. The presence of an isobestic point seems to prove, that only two species are in equilibrium in the solution under study. Curve fitting clearly shows the exchange of only one CO_3^{2-} in a pH range from 7 to 11 and in solutions of ionic strength between 0.3 and 4.5M. The influence of the ionic strength is taken into account by using the Specific Interaction Theory (S.I.T.), and the standard constant of the equilibrium between the penta and tetracarbonate complex is deduced. The same study performed on Np(IV) leads to similar conclusions. The standard stability constant measured for Np and Pu are close to the uranium one : these experimental data indeed confirm the similar behaviour of the actinide(IV) in carbonate solutions.

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