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Quantitative Raman Investigation of Uranyl Chloride Complexation under Hydrothermal Conditions

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Solvent-subtracted reduced isotropic Raman spectra of aqueous uranyl chloride species have been measured in quartz capillary high-pressure optical cells at 25 to 300 °C and 20 MPa, and at LiCl concentrations from 0.3 to 19 m, using methods reported by Alcorn et al. [1]. Using carefully optimized experimental conditions, the major species were able to be isolated spectroscopically, and were determined to be UO_2^{2+}, UO_2Cl^+, $UO_2Cl_2^0$, $UO_2Cl_3^-$, and $UO_2Cl_4^{2-}$.

Temperature-independent Raman scattering coefficients were then calculated relative to the triflate internal standard, and the results were used to calculate the equilibrium concentrations of the major uranyl chloride complexes for each solution at every temperature, with the corresponding stepwise formation quotients determined. Density functional theory (Gaussian 09) was employed to predict the structures, relative stability, and scattering coefficients of the uranyl chloride species. These were found to be consistent with the experimental results.

[1] Alcorn *et al.*, *J. Phys. Chem. B* **123**, 7385-7409 (2019)