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X-ray spectroscopic investigations of fluids in the hydrothermal diamond anvil cell: The hydration structure of aqueous La^{3+} up to 300 °C and 1600 bars

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ABSTRACT

The first direct measurements are reported for the structure of the hydrated La^{3+} ion in an aqueous solution (containing 0.007 *m* La) over a range of temperatures from 25 to 300 °C and pressures up to 1600 bars. The radial distribution of atoms around the La^{3+} ion was measured using the X-ray absorption fine structure (XAFS) technique. La L_3-edge spectra were collected in the fluorescence mode from nitrate solutions in a modified hydrothermal diamond anvil cell using the PNC-CAT X-ray microprobe at the Advanced Photon Source, Argonne National Laboratory. Analysis of the XAFS spectra collected at all temperatures indicates that each La^{3+} ion has a hydration number of nine and that the solvating waters surround the ion in a tricapped trigonal prismatic arrangement. As temperature is increased from 25 to 300 °C, the bond distance between the equatorial-plane O atoms and the La^{3+} ion increases from 2.59 ± 0.02 to 2.79 ± 0.04 Å, whereas the bond distance between La^{3+} and the O atoms at the ends of the prism decrease to 2.48 ± 0.03 Å. This study also demonstrates the unique capability of the modified hydrothermal diamond anvil cell for in situ low energy X-ray spectroscopic analysis of elements in dilute aqueous solutions at elevated temperatures and pressures.