## **Supplementary Information for**

## Rotational Dynamics of Thiocyanate Ions in Highly Concentrated Aqueous Solutions

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In this Online Supplementary Information, we present the original FTIR spectra of the sample solutions, all the measured dispersive IR pump-probe signals, calculated hydrodynamic radius of  $S^{13}CN^{-}$  ion at different potassium thiocyanate concentrations, and previously reported viscosities of KSCN in H<sub>2</sub>O solutions.

Figure S1. The background-corrected FTIR spectra of sample solutions without intensity normalization. Figure 2 in the main text is obtained by normalizing the absorbance of the  $S^{13}CN$  band.











**Figure S3.** Hydrodynamic radius  $(R_{hyd})$  of  $S^{13}CN^{-}$  ion is calculated by using the DSE (Debye-Stokes-Einstein) equation with the rotational relaxation times obtained from the IR pump-probe signals measured at  $\omega_{pr}^{*} = 1982 \text{ cm}^{-1}$  at different potassium thiocyanate concentrations. The hydrodynamic radius of  $S^{13}CN^{-}$  ion appears to be independent of the concentration of potassium thiocyanate in D<sub>2</sub>O. For the calculation of  $R_{hyd}$ , the viscosity of KSCN in H<sub>2</sub>O reported in *J. Solution Chem.*, **21**, 1115-1129 (1992) is used.



**Figure S4**. Viscosity of KSCN in  $H_2O$  as a function of the KSCN concentration. The following graph is reconstructed by using the experimental results reported in *J. Solution Chem.*, **21**, 1115-1129 (1992).

