

## Supporting information

# Structural Evolution in a Biphasic System: Poly(N-isopropylacrylamide) Transfer from Water to Hydrophobic Ionic Liquid†

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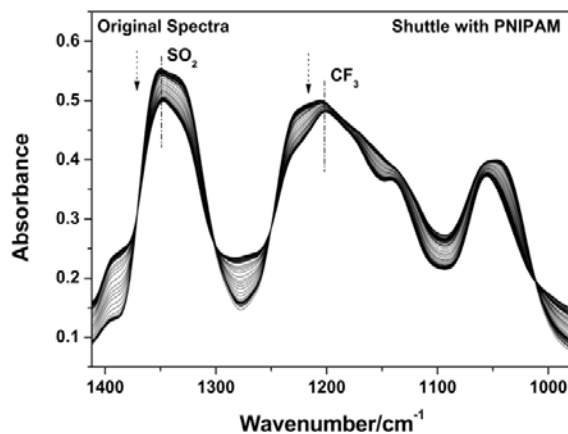


Fig. S1. Original spectra in the region (1411~985  $\text{cm}^{-1}$ ) at intervals of 6 min during PNIPAM transfer process under the homeothermic (80 °C) and quiescent condition.

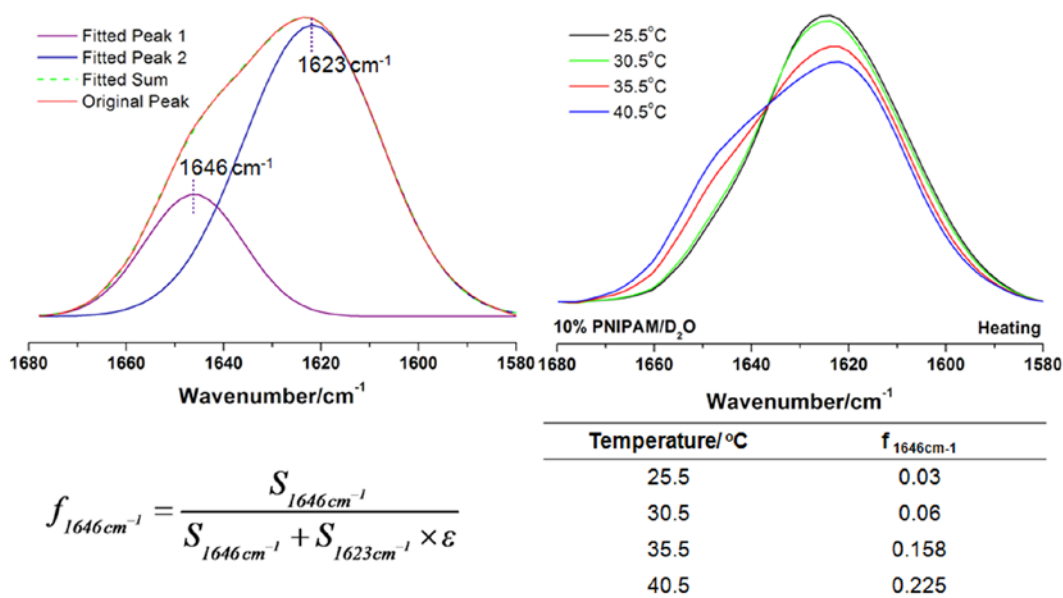


Fig. S2. FTIR Spectral variations in the C=O region of 10 wt% PNIPAM/D<sub>2</sub>O solution during heating processes. molar fractions  $f_{1646\text{cm}^{-1}}$  at different temperatures are calculated based on peak fitted curves and equation. The molar absorption coefficient  $\epsilon$  is 0.79.

Fig. S2 shows FTIR experiments on 10 %PNIPAM/D<sub>2</sub>O. It is well known that the PNIPAM changes of peak shape originate from replacement of two kinds of C=O hydrogen bonds: one is C=O···D-N(1646  $\text{cm}^{-1}$ ), the other C=O···D-O-D (1623  $\text{cm}^{-1}$ ). To clarify the quantitative changes of hydrogen bonds, peak fitting has been treated in every spectrum, and the relative content of C=O···D-N (1646 $\text{cm}^{-1}$ ) can be calculated according to the equation in Fig. S2. The results find that nearly 20% hydrated C=O groups have been changed to non-hydrated structures (C=O···D-N). It means that many C=O···D-O-D (PNIPAM) hydrogen bonds are still survived around the aggregations of PNIPAM above LCST.