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The Interplay of Methyl-Group Distribution and Hydration Pattern of Isomeric Amphiphilic Osmolytes

Agieienko V., Hölzl C., Horinek D., Buchner R.
Kazan Federal University, 420008, Kremlevskaya 18, Kazan, Russia

Abstract

© 2018 American Chemical Society. The intermolecular interactions and dynamics of aqueous 1,1-dimethyurea (1,1-DMU) solutions were studied by examining the concentration dependence of the solvent and solute relaxations detected by dielectric spectroscopy. Molecular dynamics simulations were carried out to facilitate interpretation of the dielectric data and to get a deeper insight into the behavior of the system components at the microscopic level. In particular, the simulations allowed for explaining the main differences between the dielectric spectra of aqueous solutions of 1,1-DMU and of its structural isomer 1,3-DMU. Similar to the previously studied compounds urea and 1,3-DMU, 1,1-DMU forms rather stable hydrates. This is evidenced by an effective solute dipole moment that significantly exceeds the value of a neat 1,1-DMU molecule, indicating pronounced parallel alignment of the solute dipole with two to three H2O moments. The MD simulations revealed that the involved water molecules form strong hydrogen bonds with the carbonyl group. However, in contrast to 1,3-DMU, it was not possible to resolve a "slow-water" mode in the dielectric spectra, suggesting rather different hydration-shell dynamics for 1,1-DMU as confirmed by the simulations. In contrast to aqueous urea and 1,3-DMU, addition of 1,1-DMU to water leads to a weak decrease of the static permittivity. This is explained by the emergence of antiparallel dipole-dipole correlations among 1,1-DMU hydrates with rising concentration.

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