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The search for the Grotthuss mechanism.

Understanding of proton transport is an old and fundamental problem critical for many technologies. More than 200 years ago Grotthuss proposed the concept of correlated proton transport, when proton hopping can be collective with correlated proton jumps in the same direction along hydrogen-bonded chains of host molecules. This type of collective hopping is considered as the most efficient transport of charge. Although many computational studies have suggested the existence of Grotthuss mechanism, experimental verification has remained a challenge and only indirect indications of this mechanism have been reported.
In this study we report experimental observation of proton transfer between the molecules in pure and 85% aqueous phosphoric acid, employing quasielastic neutron scattering, dielectric spectroscopy, light scattering, and ab initio molecular dynamic simulations [1]. Detailed analysis of neutron scattering data and ab initio molecular dynamic simulations reveal that proton transfer occurs via very short 'jumps'of only ~0.5 - 0.7 Å, which is much smaller than the typical ion jump length in ionic liquids. Neutron scattering and GHz frequency dielectric studies also show that protons are the major contributor to the conductivity in these systems and that all protons are involved in proton jumps. In turn, the light scattering measurements in GHz region revealed that the timescale of structural relaxation is much slower than proton conductivity relaxation.
Our analyses also shows that protons movement is indeed correlated, however the correlations suppress the conductivity instead of enhancing as has been generally assumed for a 'Grotthuss-like'hopping mechanism. Importantly, these results suggest that collective proton transport in bulk liquids will always decrease ionic conductivity.

1. Ivan Popov, Zhenghao Zhu, Amanda R. Young-Gonzales, Robert L. Sacci, Eugene Mamontov, Catalin Gainaru, Stephen J. Paddison, Alexei P. Sokolov. Search for a Grotthuss mechanism through the observation of proton transfer. Communication Chemistry, 6, 77 (2023). DOI: https://doi.org/10.1038/s42004-023-00878-6

Topic

Energy Materials

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