New reactive force field allowing bond breaking/formation in classical simulation of proton transport in water, acid solutions and polymeric electrolyte membranes

D.W.M.Hofmann*, L.N.Kuleshova, B.D'Aguanno

CRS4, Center for Advanced Studies, Research and Development in Sardinia
Loc. Piscina Manna, Edificio 1, 09010 Pula (CA), Italy

We present a new effective classical force field for the simulation of the conductivity in fuel cell membranes. The force field is based on the central force model for water, i. e. the interactions between the atoms are defined by three atom pair potentials. The force field integrates the inter- and intramolecular part into a single potential. This approach has two special features: Firstly, it allows to model on the classical level bond breaking and formation, secondly, integral equation theory of liquids becomes applicable. These properties make the force field very attractive for computer simulations of proton transport and proton conductivity in water and acid solutions. It enables us to perform simulations for large systems with long simulation times, which is required for the derivation of conductivity from molecular dynamics. The potentials of the force field were optimized to reproduce the experimental radial distribution functions of water in a wide range of temperatures and pressures exactly. We show, that the force field reproduces correctly not only the RDF, but also other properties of water, e.g. density, selfdiffusion, and lifetimes of hydroniums. Finally we compare simulated properties of hydrated Nafion membranes, dissoziation degree, selfdiffusion, and conductivity with experimental data.

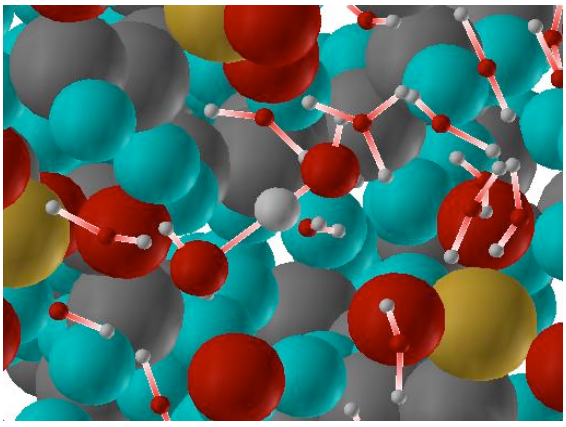

Figure 1. The transition state of hydrogen transfer between two waters in hydrated Nafion. A

[1] Detlef W.M. Hofmann, Liudmila Kuleshova, and Bruno D'Aguanno, A "reactive force field" for simulations of the structure and properties of water and its acid solution, J. Comp. Chem., under revision