

ARTICLE

Supporting Information: Softening by charging: how collective modes of ionic association in concentrated redoxmer/electrolyte solutions define the structural and dynamic properties in different states of charge

Received 00th January 20xx,
Accepted 00th January 20xx

DOI: 10.1039/x0xx00000x

Hossam M. Farag,^{a,b,c} Aman Preet Kaur,^{a,d} Lily A. Robertson,^{a,e} Erik Sarnello,^{a,g} Xinyi Liu,^{a,g} Yilin Wang,^{a,c,h} Lei Cheng,^{a,f} Ilya A. Shkrob,^{a,e} Lu Zhang,^{a,e} Randy Ewoldt,^{a,c,h} Tao Li,^{a,i,g} Susan A. Odom,^{a,d} Y Z*,^{a,b,c,j}

Molarity Corrections for different %SOC solutions

As mentioned in the main text, during charging of the cell, Li⁺ ions migrate across the cell membrane, leaving behind un-occupied volume, which virtually increases the molarity of the corresponding charged solution. Such effect is very small across the majority of considered molarities, but becomes more prominent in the supersaturated regime, and in other cases where the salt cation is larger than Li⁺. For completeness we have computed the corrections that should be made for such effect. The results are shown in the following figures.

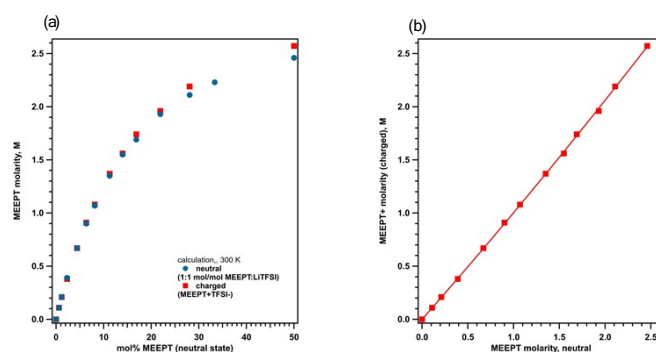


Figure 1. (a) 0% SOC and 100% SOC molarities for chosen mol% MEEPT in the neutral state. (b) Regression line for 0% SOC and 100% SOC molarities for the same chosen mol% MEEPT in the neutral state. The slope of the regression line is approximately 1.0 across all molarities.

Reduced Viscosity and Huggins Equation

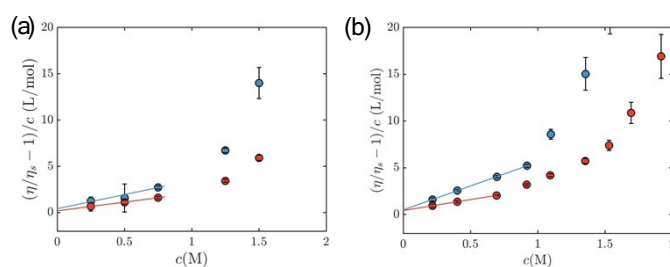


Figure 2. The reduced viscosity of neutral and charged systems as a function of concentration; (a) the measured viscosity; (b) the simulated viscosity. The solid lines represent the most credible fits to the rearranged Huggins equation.

MEEPT: Li⁺ pair distribution function (PDF) and potential of mean force (PMF)

^a Joint Center for Energy Storage Research

^b Department of Nuclear, Plasma, and Radiological Engineering, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801, United States

^c Beckman Institute for Advanced Science and Technology, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801, United States

^d Department of Chemistry, University of Kentucky, Lexington, Kentucky 40506, United States

^e Chemical Sciences and Engineering Division, Argonne National Laboratory, Lemont, Illinois 60439, United States

^f Materials Sciences Division, Argonne National Laboratory, Lemont, Illinois 60439, United States

^g Department of Chemistry and Biochemistry, Northern Illinois University, DeKalb, Illinois 60115, United States

^h Department of Mechanical Science and Engineering, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801, United States

ⁱ X-ray Science Division, Argonne National Laboratory, Lemont, Illinois 60439, United States

^j Department of Electrical and Computer Engineering, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801, United States

Electronic Supplementary Information (ESI) available: [details of any supplementary information available should be included here]. See DOI: 10.1039/x0xx00000x

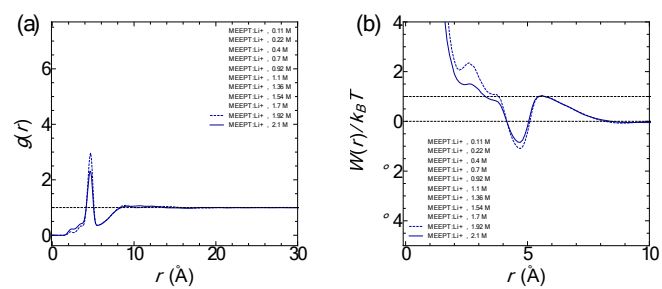


Figure 3. (a) MEEP: Li⁺ pair distribution function (PDF) $g(r)$ at different solute. (b) Corresponding solution-averaged potential of mean force (PMF). The Li⁺ interaction with two oxygens in the ether chain of MEEP becomes less prominent as salt concentration increases and is dominant at lesser concentrations.