Experimental report

25/01/2024

Proposal: 6-03-454 **Council:** 4/2020

Title: Dynamics of a Hybrid Aqueous-Nonaqueous Electrolyte

Research area: Soft condensed matter

This proposal is a new proposal

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Samples: [LiN(CF3SO2)2][C3H6O3]_0.56[H2O]1.44
[LiN(CF3SO2)2][C3D6O3]_0.56[D2O]1.44
[LiN(CF3SO2)2][C3H6O3]_0.56[D2O]1.44

Instrument	Requested days	Allocated days	From	To
WASP	3	3	31/05/2021	03/06/2021

Abstract:

A new class of electrolytes has recently been reported, hybridizing aqueous with non-aqueous solvents (HANE), that inherits the non-flammability and non-toxicity characteristics from aqueous and better electrochemical stability from non-aqueous systems. In this proposal we plan to measure LiTFSI dissolved in 1-1 H2O + DMC and 1-1 D2O + d-DMC to verify the molecular diffusivities and ionic conductivities predicted by the Molecular Dynamics numerical simulations. These measurements will complement QENS measurements performed last year on the NEAT spectrometer at BENSC. With the longer (by about a factor 20) Fourier time proved ny WASP, we expect to resolve dynamics of the water molecules and the three-times heavier DMC molecules.

Conlusion part of submitted publication:
G. Guenther, M. Russina, B. Farago, O. Borodin, M. A. González, T. Yamada, O. Yamamuro, D. L. Price and M.-L. Saboungi, Nanosecond dynamics of a hybrid aqueous-nonaqueous electrolyte, *J. Chem. Phys*.,

In this work we have shown that, for hydrogenous systems with significant amounts of coherent scattering, combining QENS with NSES makes it possible to extract the incoherent contribution. In the present case, the limited time range of the QENS data made it necessary to extend this contribution to times of 10 ns, where the incoherent scattering functions have relaxed to near zero. The fact that extrapolations with two difference functions – stretched exponential and linear – gave generally similar results, both for the MD simulation predictions and the experimental results, provides confidence in the validity of this procedure.

17 The use of two different isotopic compositions – with H2 O-hDMC and D2 O-hDMC – and the difference of the two sets of results has made it possible to derive experimental values for both the DMC and water apparent diffusion coefficients. The values are similar for DMC and water, whereas the MD simulations are predicting significantly lower values for the heavier DMC molecules.

The comparison with of the experimental results for the structure with MD predictions presented in Ref. 7 showed the MD simulation was over-estimating the DMC-water segregation, perhaps leading to a reduced coupling between the dynamics of the two solvents. Notwithstanding this discrepancy, the overall experimental confirmation of the MD predictions for the dynamics of this system vindicates the MD simulations and the predictions of electrolyte performance presented by Wang et al.