Supporting Information:

Heterogeneous dynamics and its length scale in simple ionic liquid models: A computational study

Soree Kim, Sang-Won Park, and YounJoon Jung*

Department of Chemistry, Seoul National University, Seoul 08826, Korea

^{*} Electronic mail:yjjung@snu.ac.kr



FIG. S1. Displacement distributions of particles. (a)ACM-cation (b)ACM-anion (c)UCM-cation (d)UCM-anion



FIG. S2. Time dependence of the displacement distributions. (a)ACM-cation; T=1.75 (b)ACM-anion; T=1.75 (c)UCM-cation; T=0.35 (d)UCM-anion; T=0.35



FIG. S3. Time dependence of the Self-van Hove functions. (a) ACM-cation; T=1.75 (b) ACM-anion; T=1.75 (c) UCM-cation; T=0.35 (d) UCM-anion; T=0.35



FIG. S4. Probability distributions of the exchange time (dashed line) and the persistence time (solid line). (a)ACM-cation (b)ACM-anion (c)UCM-cation (d)UCM-anion



FIG. S5. The ratio of the mean persistence time and the mean exchange time. (a)ACM (b)UCM



FIG. S6. Power law relationship between the mean persistence time and the mean exchange time. (a)ACM (b)UCM



FIG. S7. Length scale d dependence on the mean persistence time and the mean exchange time. The data are obtained for the cation in SCM. Power law exponents are shown in the graph. (a) Power law relation, $\tau_{\rm e} \sim \tau_{\rm p}^{\nu}$, between the mean persistence time and the mean exchange time is shown. As d is increased, the exponent is getting closer to 1. (b) Power law relations between the mean persistence time and the structural relaxation time are shown. As d is increased, the exponent is increasing. Even at d = 1.4 which is similar value of $2\pi/q$ where q = 4.8 is the value of the first peak position of the structure factor, the exponent is still far from 1. (c) Power law relations between the mean exchange time and the diffusion constant are shown. Because of the fragile nature of ionic liquids system, there are sublinear relations between two values, irrespective of d.



FIG. S8. Dynamic susceptibility, $\chi_4(t)$, of anions in each system calculated using Eq.5 at various temperatures: from left to right, T = 6.24, 4.16, 3.33, 2.49, 2.08, 1.87, 1.66, 1.54, 1.46, 1.33, 1.25, 1.16 and 1.12 (SCM), T = 6.24, 4.16, 3.33, 2.83, 2.49, 2.25, 2.08, 1.98, 1.87, 1.79 and 1.75 (ACM), T = 1.56, 1.14, 0.94, 0.77, 0.67, 0.58, 0.50, 0.44, 0.40, 0.37, 0.35, 0.33 0.31 and 0.29 (UCM). Loglog plots are also shown (inset). The time value that makes $\chi_4(t)$ maximum is defined as the characteristic time scale of the dynamic heterogeneity, t_4^* (black dots).



FIG. S9. The dynamic susceptibility calculated using Eq.7. Wavevector k is set by $k = 2\pi/\lambda_{\text{max}}$, where $\lambda_{\text{max}} = 0.92, 0.94, 1.0$ are the shortest peak position of radial distribution function between cation and anion for SCM, ACM and UCM, respectively. Temperatures are same with FIG.S8. Log-log plot is illustrated in the inset and the maximum points are shown with black dots. (a)ACM-cation (b)ACM-anion (c)UCM-cation (d)UCM-anion



FIG. S10. Time dependence of the dynamic correlation length, $\xi_4(t)$, of the anion at various temperatures (a)SCM, (b)ACM and (c)UCM: from left to right, T = 6.24, 2.49, 1.87, 1.54, 1.33, and 1.16 (SCM), T = 3.33, 2.49, 2.08, 1.87, 1.79, and 1.75 (ACM), T = 1.56, 0.94, 0.67, 0.50, 0.40, 0.37, and 0.35 (UCM). The error bars are shown only for the lowest temperature.



FIG. S11. Collapse of dynamic structure factor. (a)SCM-cation (b)ACM-cation (c)UCM-cation (d)SCM-anion (e)ACM-anion (f)UCM-anion. Every data shows good agreement with the function $f(x) = 1/(1 + x^2)$ (solid line).



FIG. S12. Relation between τ_{α} and t_4^* . Two time scales are almost equal for every model systems. Dashed line shows $\tau_{\alpha} = t_4^*$ line.