Supporting Information for: Relationship between the Relaxation of Ionic Liquid Structural Motifs and That of the Shear Viscosity

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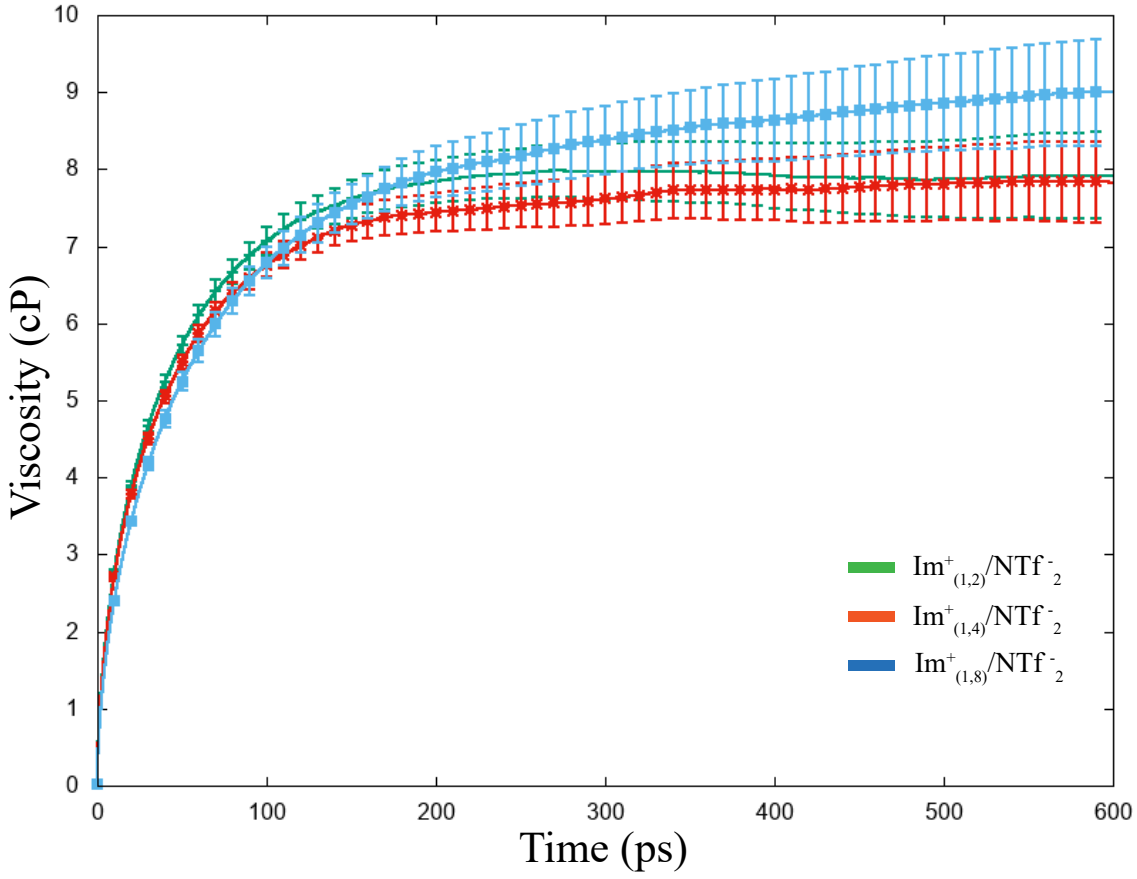

Figure S.1: The Green-Kubo expression $\frac{1}{k_B TV}\int_0^t \langle \sigma^{zx}(0)\sigma^{zx}(t')\rangle dt'$ and error bars at the 95 percent confidence level for $Im^+_{1,2}/NTf_2^-$, $Im^+_{1,4}/NTf_2^-$, and $Im^+_{1,8}/NTf_2^-$. The overlap in the error bars becomes even larger close to the corresponding t_∞ (data not show for clarity).

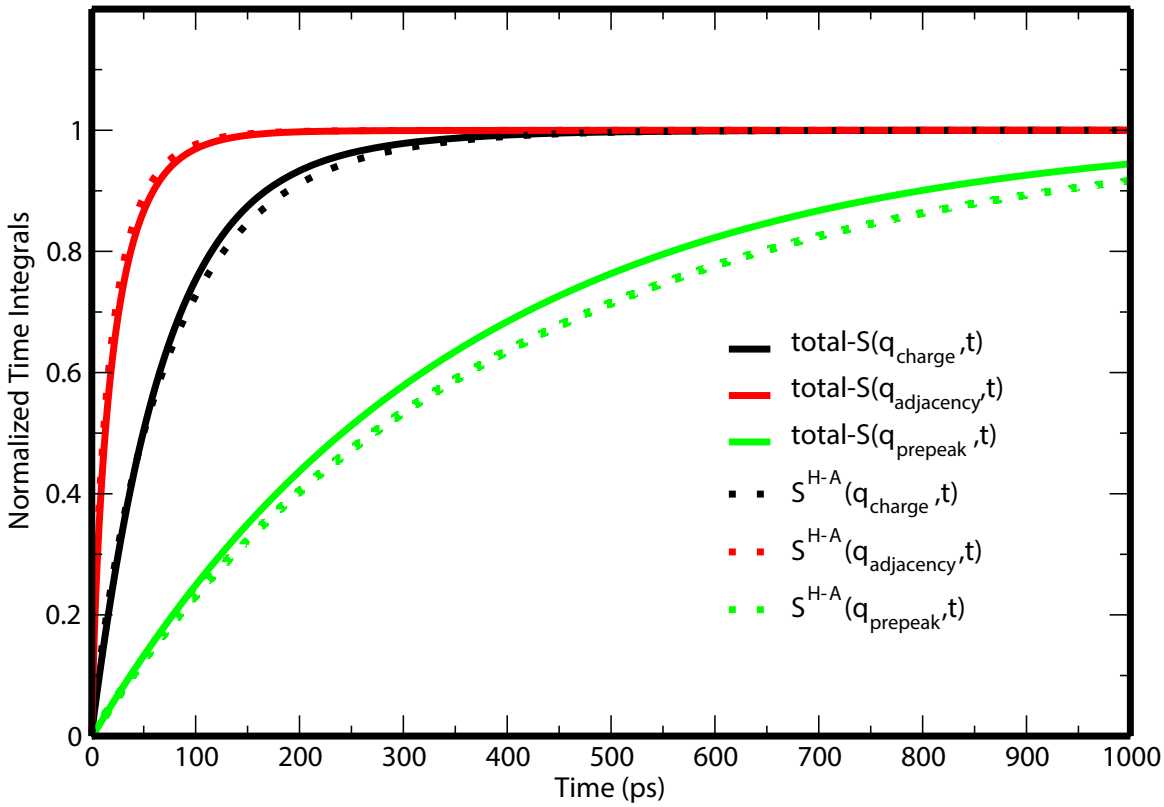

Figure S.2: For $Im^{+}_{1,8}/NTf^{-}_{2}$, a comparison between $\int_0^t S(q,t')^2\ dt'/\int_0^{t_\infty} S(q,t)^2\ dt$ and $\alpha(q,t)$, at the q-regions associated with the structural motifs.

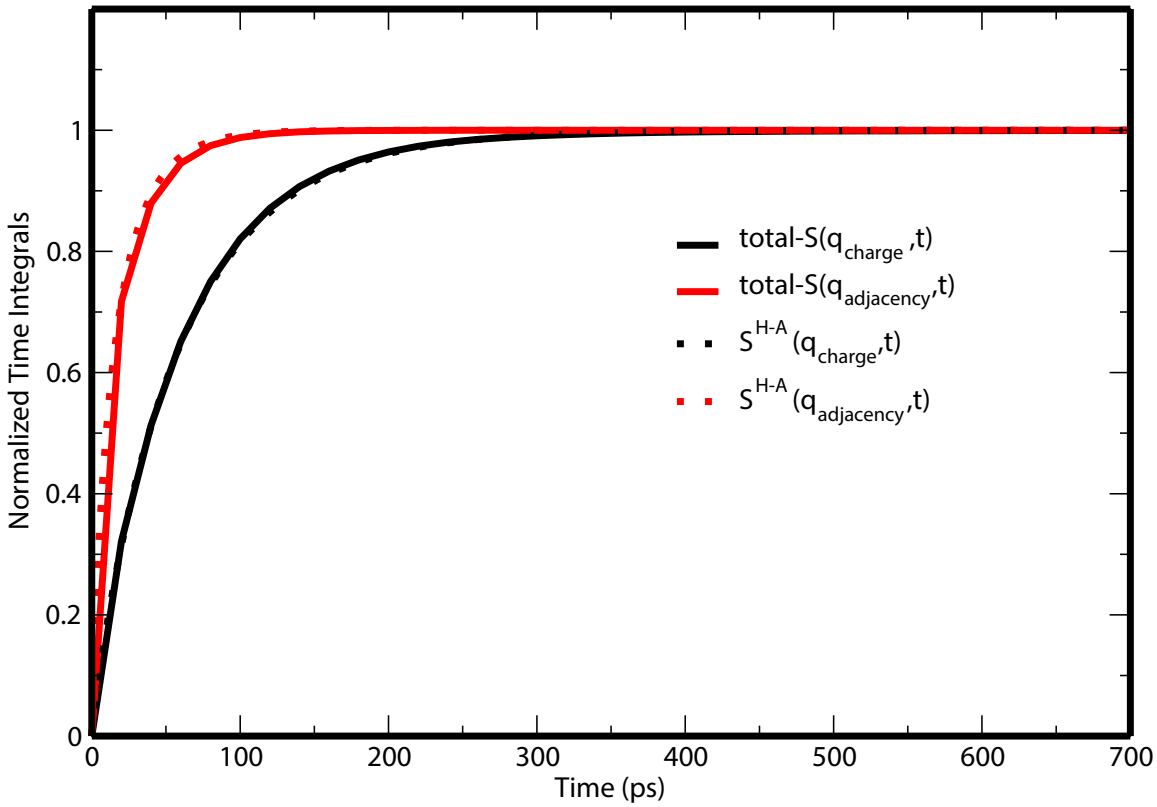

Figure S.3: For $Im^{+}_{1,4}/NTf^{-}_{2}$, a comparison between $\int_0^t S(q,t')^2\ dt'/\int_0^{t\infty} S(q,t)^2\ dt$ and $\alpha(q,t)$, at the q-regions associated with the structural motifs.

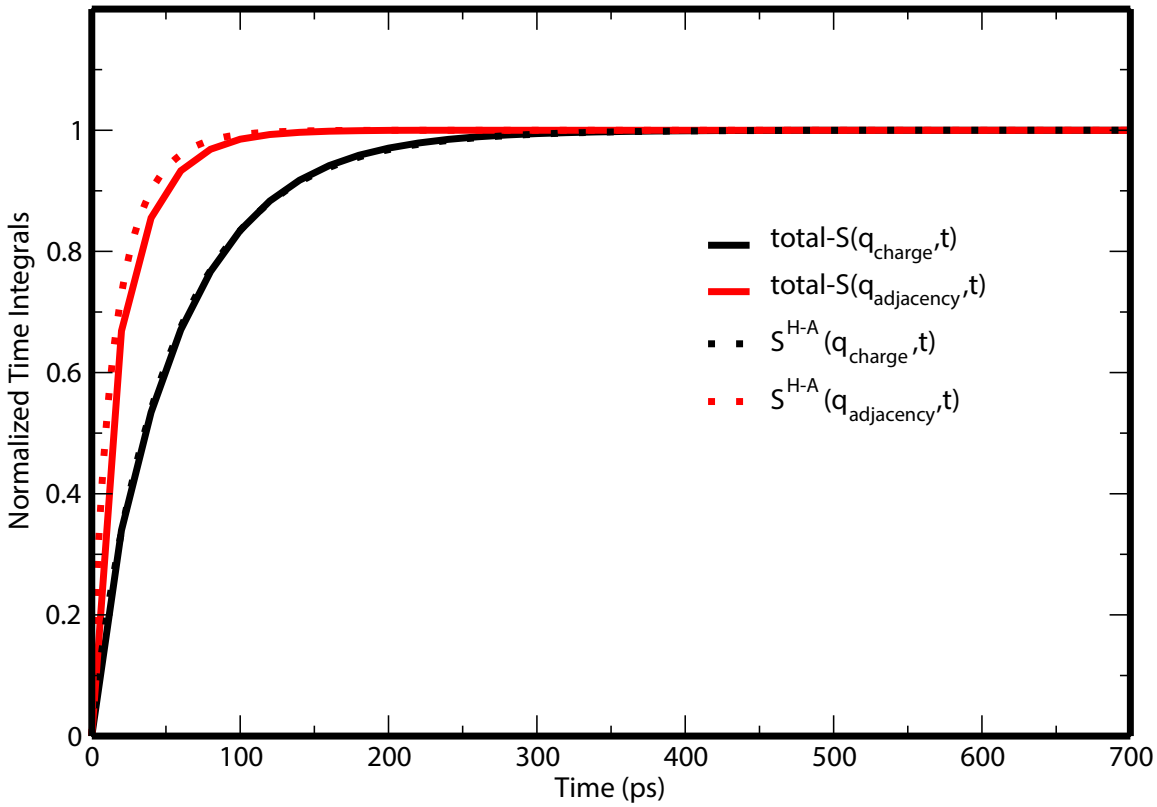

Figure S.4: For $Im^{+}_{1,2}/NTf^{-}_{2}$, a comparison between $\int_0^t S(q,t')^2\ dt'/\int_0^{t\infty} S(q,t)^2\ dt$ and $\alpha(q,t)$, at the q-regions associated with the structural motifs.

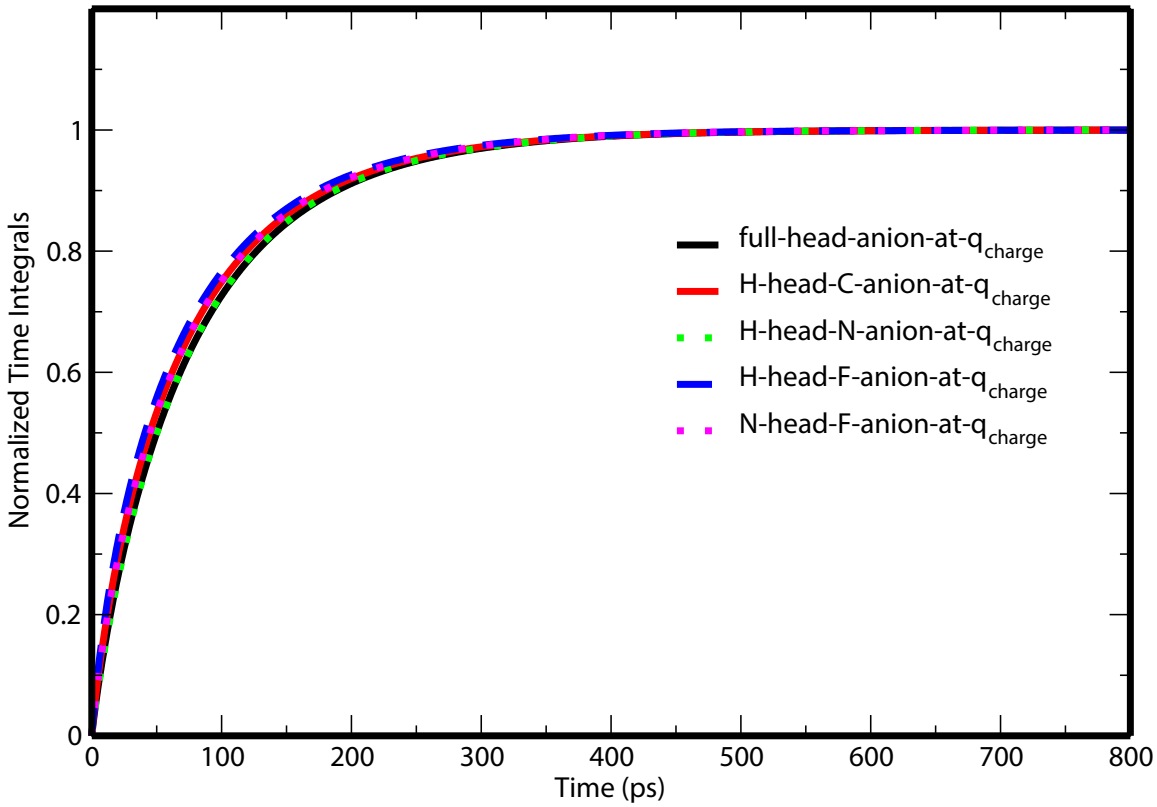

Figure S.5: For $Im^{+}_{1,8}/NTf^{-}_{2}$, α(q,t) compared with some of its partial atomic subcomponents.

Table S.1: Biexponential time constants (τ_1 and τ_2), as well as parameters A and α used to fit the relaxation of ζ(t) as defined in reference 1; computed viscosities for our three ILs. $\zeta(t) = A\alpha\tau_1(1 - e^{-t/\tau_1}) + A(1 - \alpha)\tau_2(1 - e^{-t/\tau_2})$.

IL	Viscosity (cP)	A	α	τ_1 (ps)	τ_2 (ps)
$Im^+_{1,8}/NTf^-_2$	8.8 (at 377.4 K)	0.028491	0.135804	135.514	19.5938
$Im^+_{1,4}/NTf^-_2$	7.7 (at 363.3 K)	0.055910	0.175726	64.8021	7.86063
$Im^+_{1,2}/NTf^-_2$	7.9 (at 354.4 K)	0.081545	0.196336	47.9468	3.44041

References

(1) Zhang, Y.; Otani, A.; Maginn, E. J. Reliable Viscosity Calculation from Equilibrium Molecular Dynamics Simulations: A Time Decomposition Method. *Journal of Chemical Theory and Computation* **2015**, *11*, 3537–3546.