Supplementary Information

for

Balancing fracture toughness and ionic conductivity in lithium thiosilicate glassy electrolytes

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Figure S1

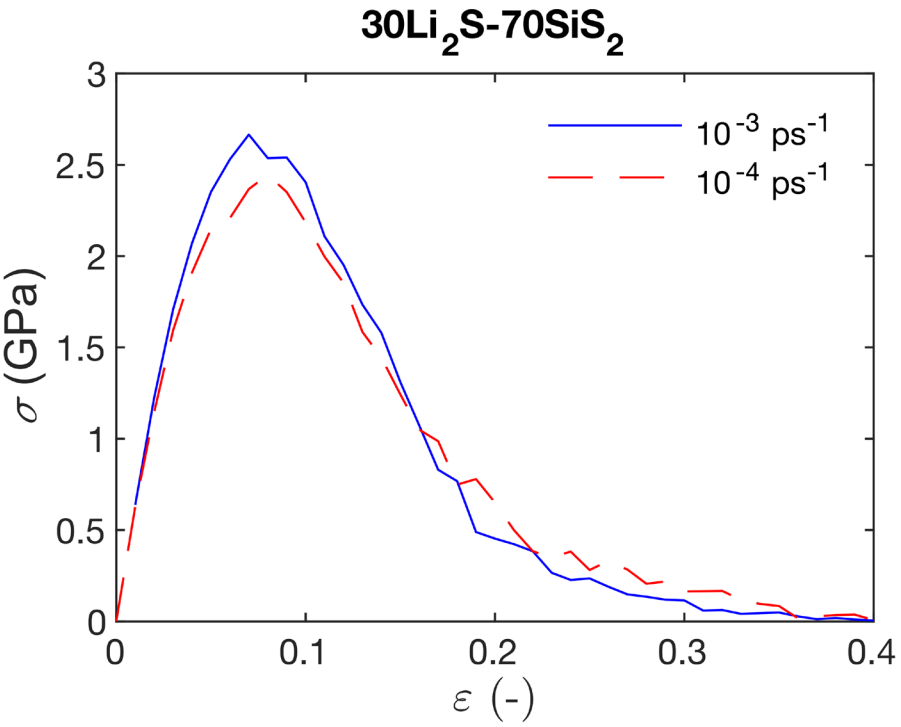


Figure S1. Comparing the effect of strain rate on the stress-strain response of the simulated $30Li_2S$-$70SiS_2$ glass.

Figure S2

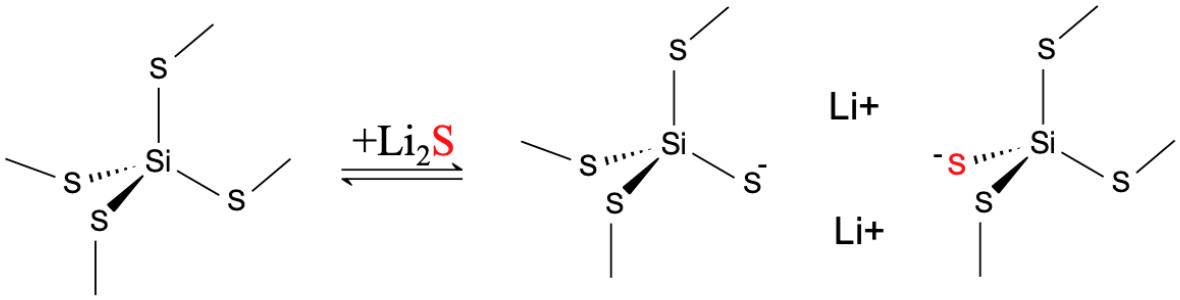
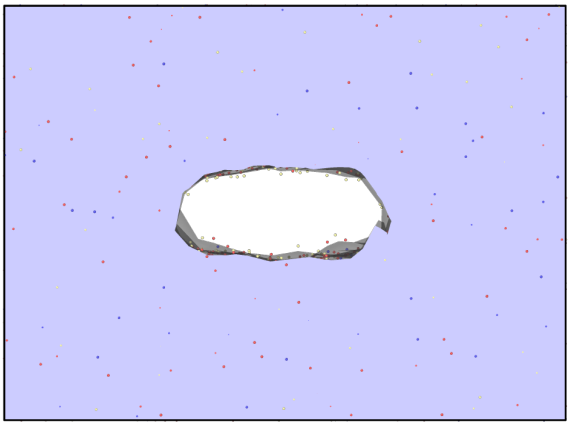


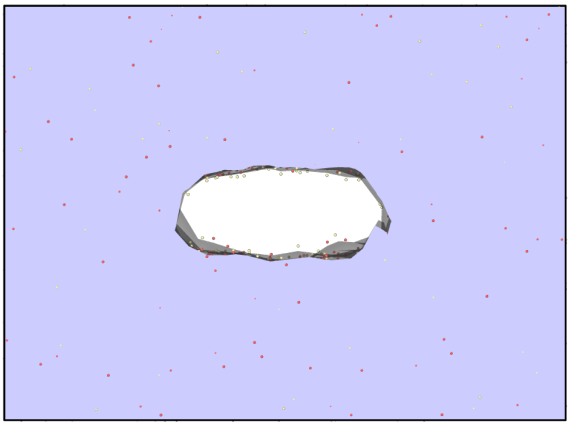
Figure S2. Schematic of how addition of Li_2S to a SiS_2-network induces Si-S bond breaking and the formation of non-bridging sulfur (S^-) in the structure.

Figure S3

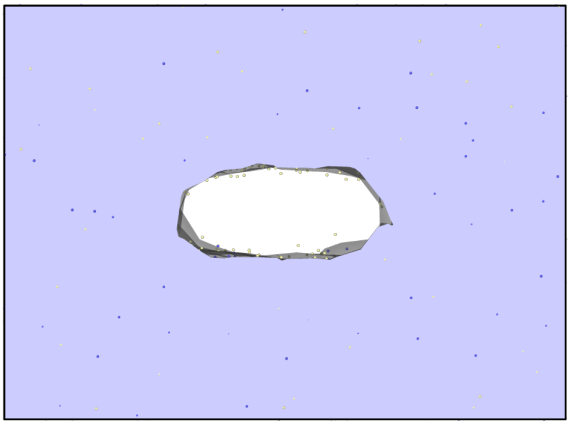
(a)
No removal
Surface area: 4900 Å^2



(b)
Si Removal
Surface area: 4900 Å^2



(c)
S removal
Surface area: 5042 Å^2



(d)
Li Removal
Surface area: 5186 Å^2

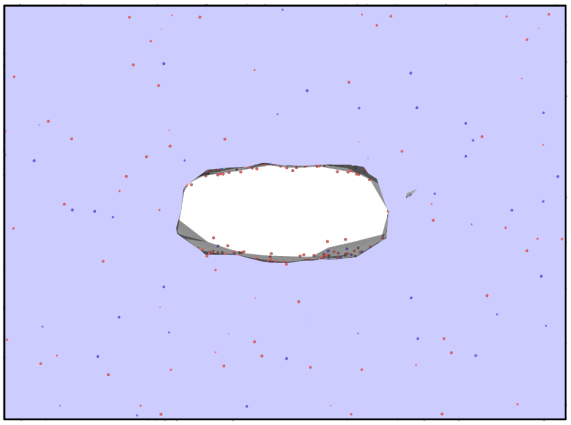


Figure S3. Calculated internal surface areas in the cases of removal of (a) none, (b) all silicon, (c) all sulfur, and (d) all lithium atoms for a $30Li_2S$-$70SiS_2$ glass (at a strain of 10%).

Figure S4

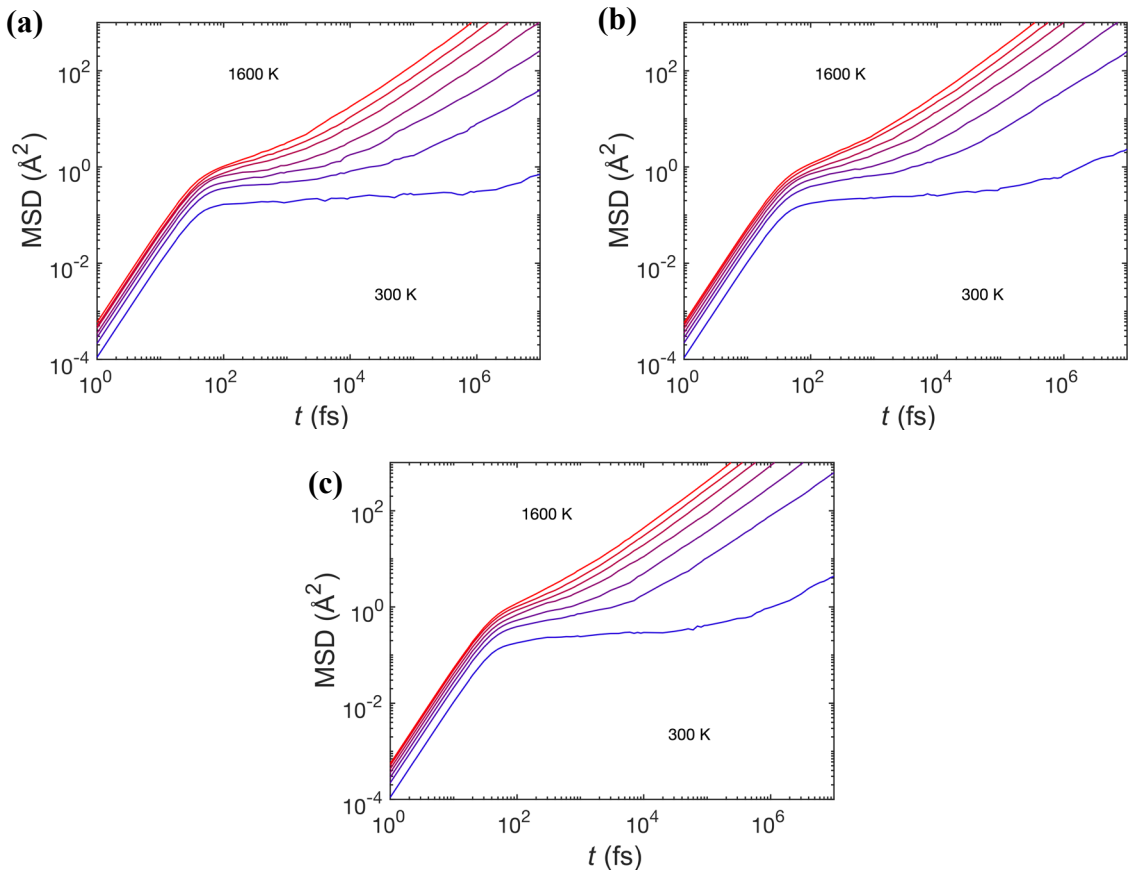


Figure S4. MSD for Li atoms in (a) $40Li_2S$-$60SiS_2$, (b) $50Li_2S$-$50SiS_2$, and (c) $60Li_2S$-$40SiS_2$ simulated glasses at temperatures of 300, 600, 800, 1000, 1200, 1400, and 1600 K.

Figure S5

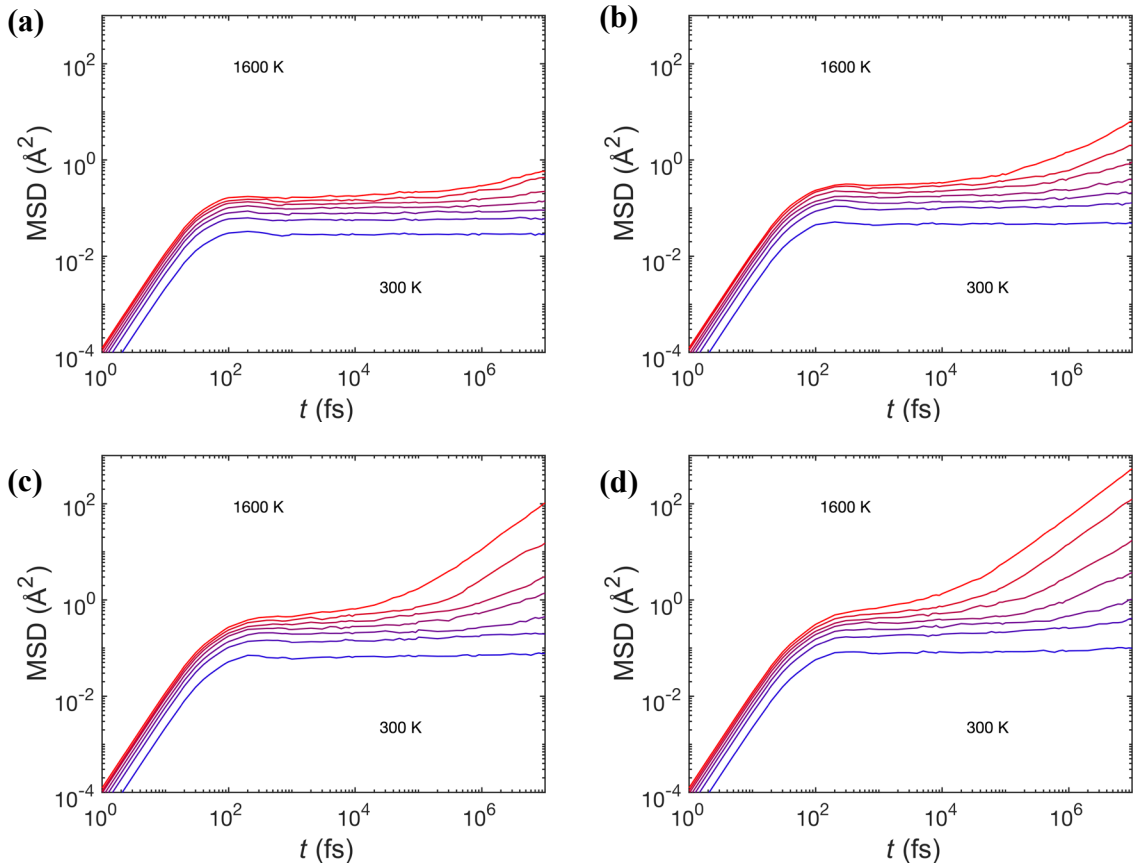


Figure S5. MSD for S atoms in (a) $30Li_2S$-$70SiS_2$, (b) $40Li_2S$-$60SiS_2$, (c) $50Li_2S$-$50SiS_2$, and (d) $60Li_2S$-$40SiS_2$ simulated glasses at temperatures of 300, 600, 800, 1000, 1200, 1400, and 1600 K.

Figure S6

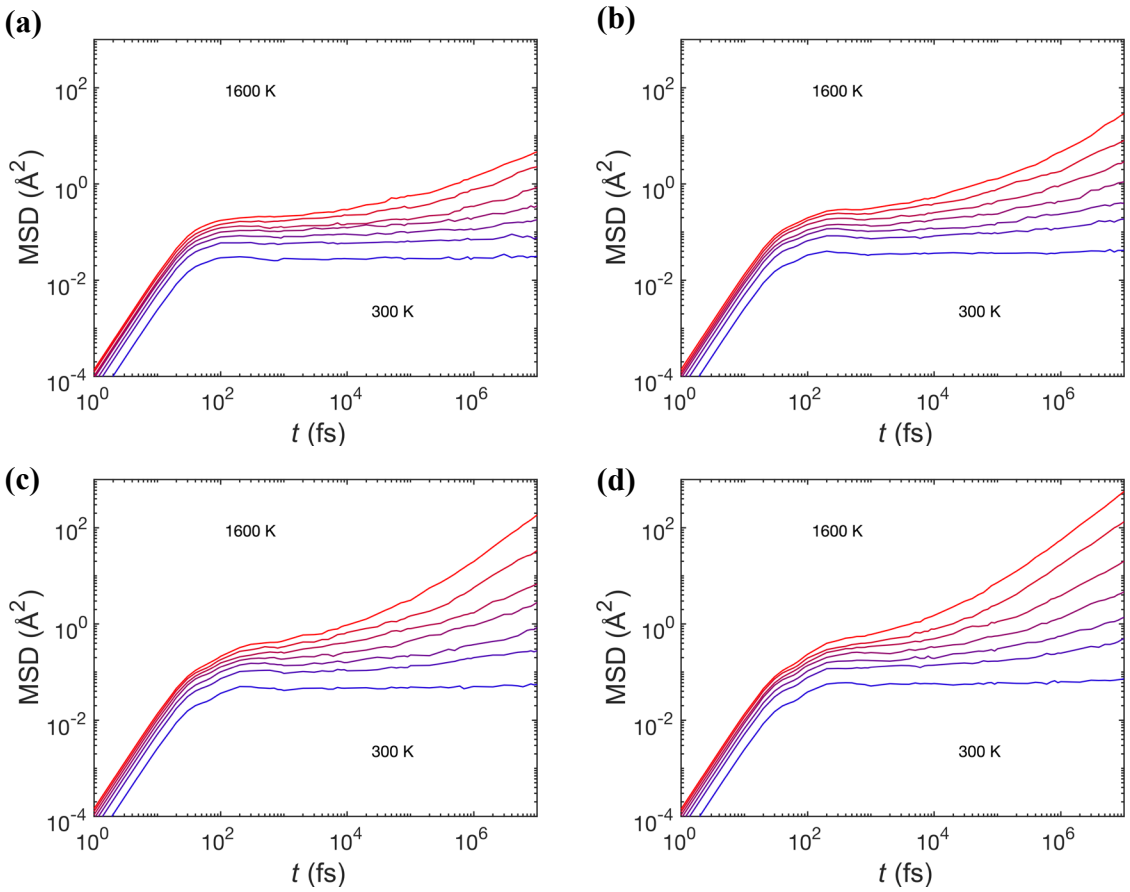


Figure S6. MSD for Si atoms in (a) $30Li_2S$-$70SiS_2$, (b) $40Li_2S$-$60SiS_2$, (c) $50Li_2S$-$50SiS_2$, and (d) $60Li_2S$-$40SiS_2$ simulated glasses at temperatures of 300, 600, 800, 1000, 1200, 1400, and 1600 K.

Figure S7

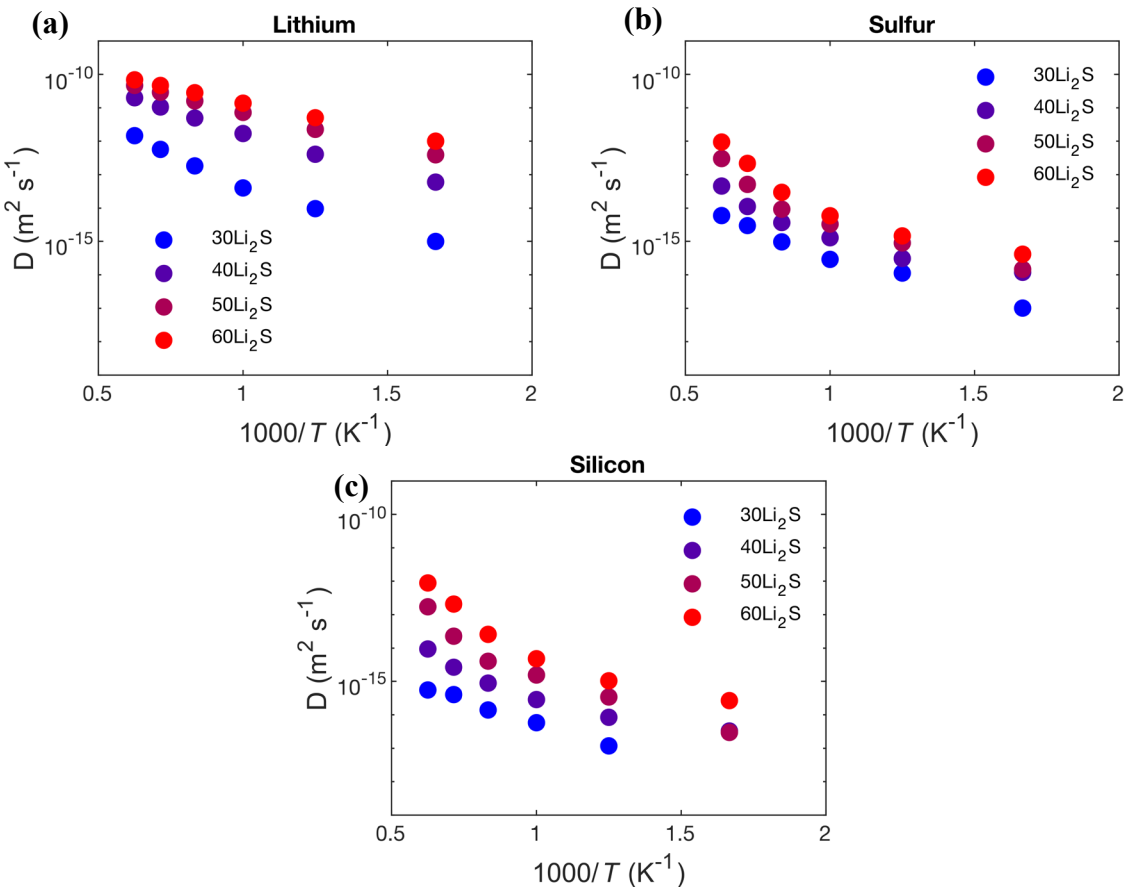


Figure S7. Diffusion coefficients for (a) Li, (b) S, and (c) Si atoms in $30Li_2S$-$70SiS_2$, $40Li_2S$-$60SiS_2$, $50Li_2S$-$50SiS_2$, and $60Li_2S$-$40SiS_2$ simulated glasses at temperatures of 600, 800, 1000, 1200, 1400, and 1600 K.

Figure S8

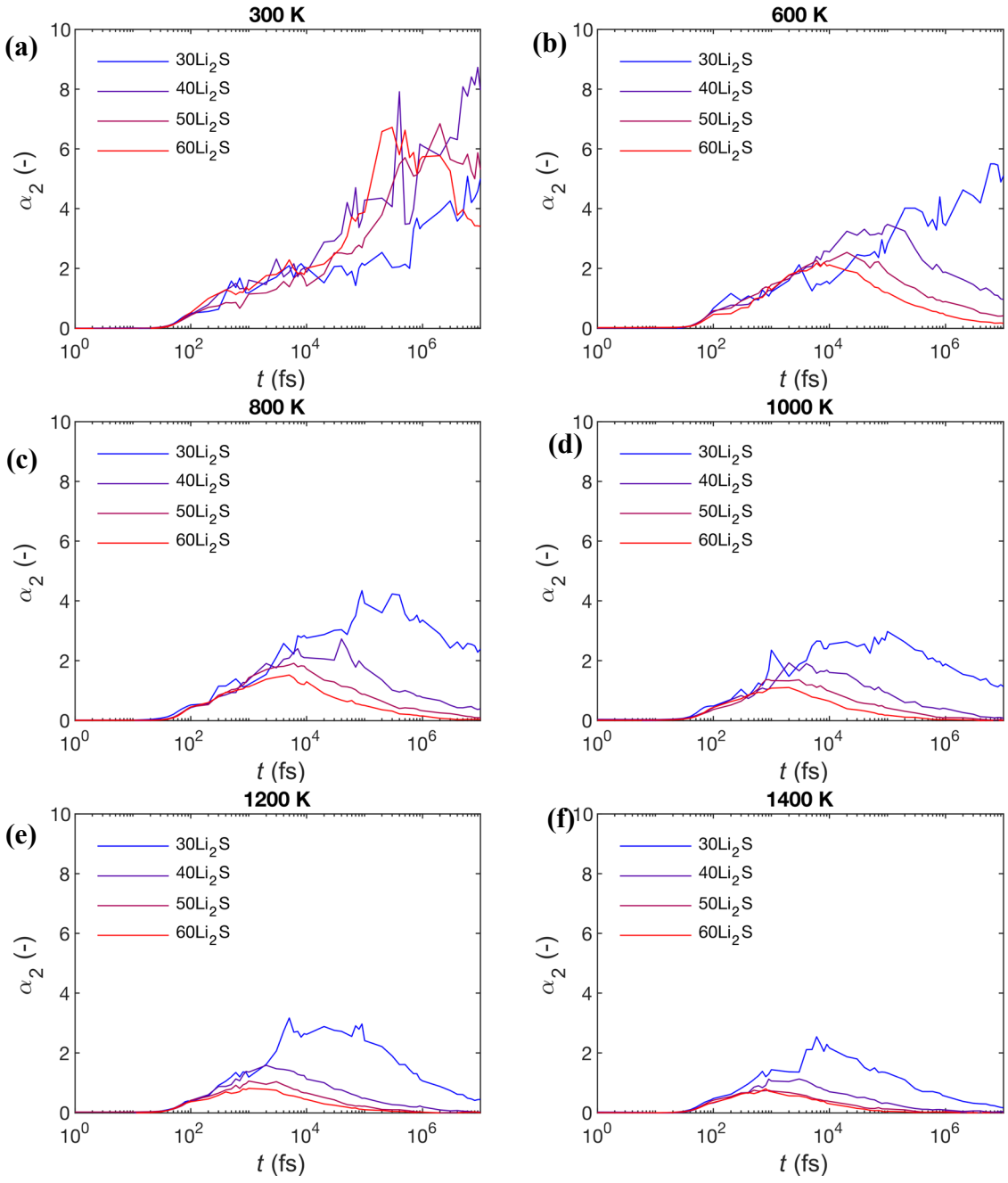


Figure S8. Non-gaussian parameter for Li atoms at (a) 300 K, (b) 600 K, (c) 800 K, (d) 1000 K, (e) 1200 K, and (f) 1400 K for the $30Li_2S$-$70SiS_2$, $40Li_2S$-$60SiS_2$, $50Li_2S$-$50SiS_2$, and $60Li_2S$-$40SiS_2$ simulated glasses. Note the difference in the scale of the vertical axis compared to that in Figure 6b in the main text.

Figure S9

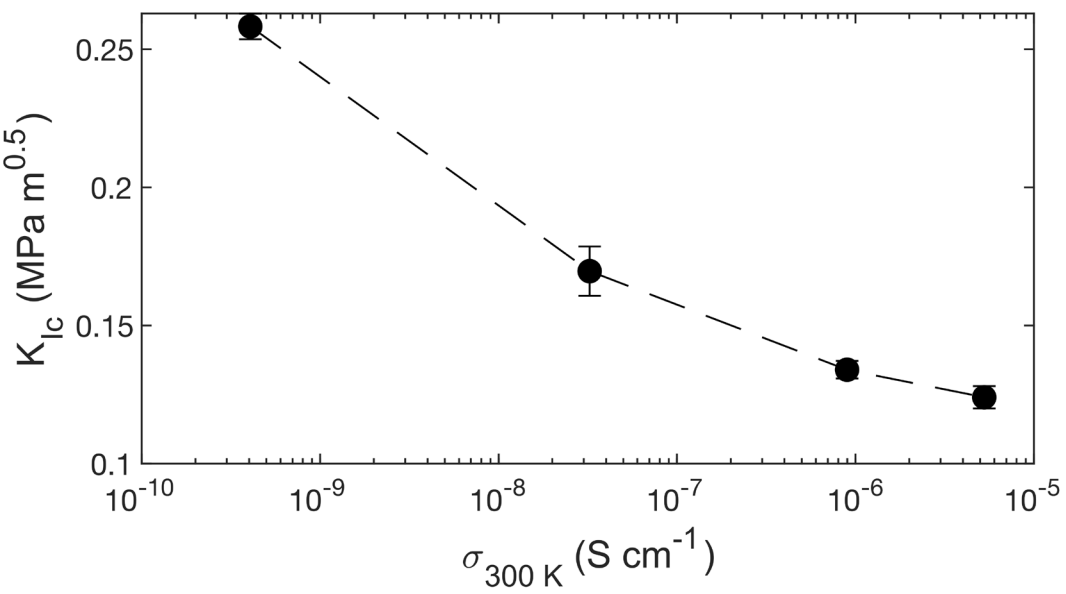


Figure S9. Fracture toughness (K_{Ic}) as a function of extracted ionic conductivity at 300 K (σ_{300K}) for the $30Li_2S$-$70SiS_2$, $40Li_2S$-$60SiS_2$, $50Li_2S$-$50SiS_2$, and $60Li_2S$-$40SiS_2$ simulated glasses. Note the log-scale on the horizontal axis.

Figure S10

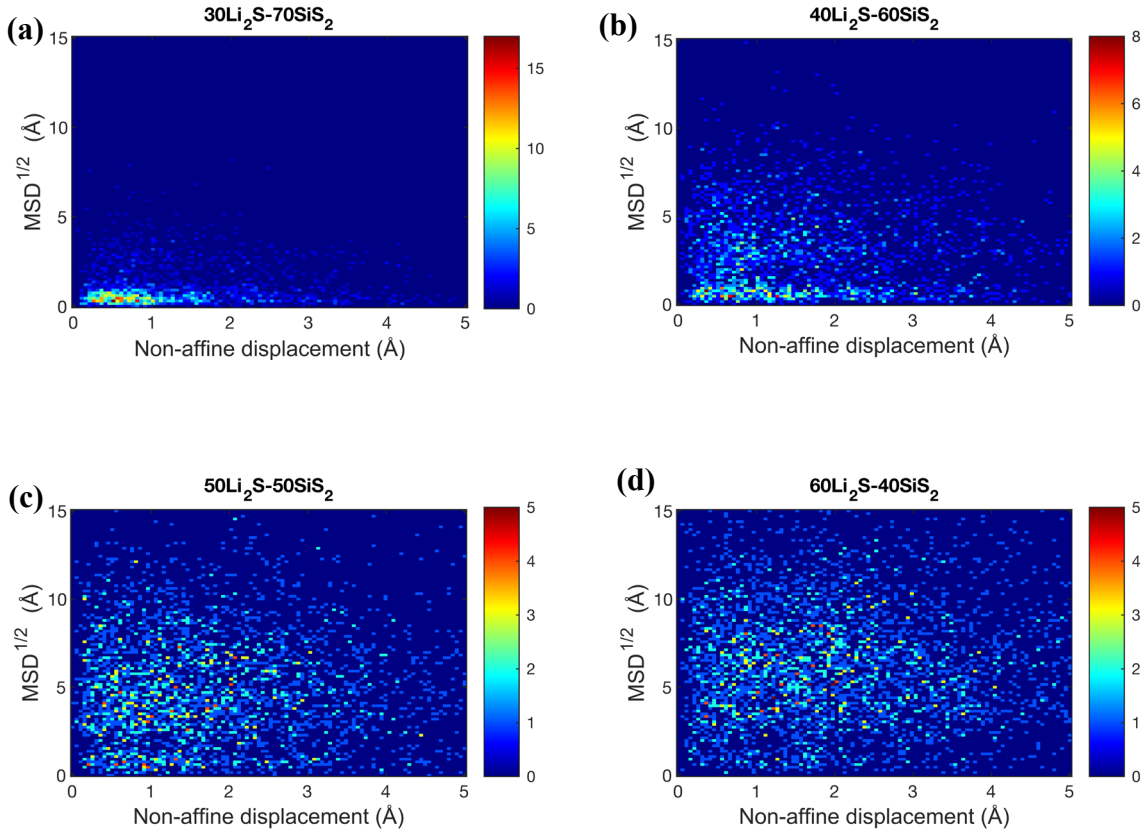


Figure S10. Square root of mean square displacement of Li atoms (after 1 ns of simulation at 1200 K) as a function of non-affine displacement of Li atoms in a uniaxial stretching simulation (values of non-affine displacement taken after a strain of ε=10%). Results are shown for (a) $30Li_2S$-$70SiS_2$, (b) $40Li_2S$-$60SiS_2$, (c) $50Li_2S$-$50SiS_2$, and (d) $60Li_2S$-$40SiS_2$ simulated glasses. Color bar represents the number of atoms. Note that panel (b) is shown in the main text as Figure 8 but included here for easy comparison.