Supplementary Information for: A molecular dynamics study of the structure and dynamics of screened polyelectrolyte complex materials

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Supplementary Figures

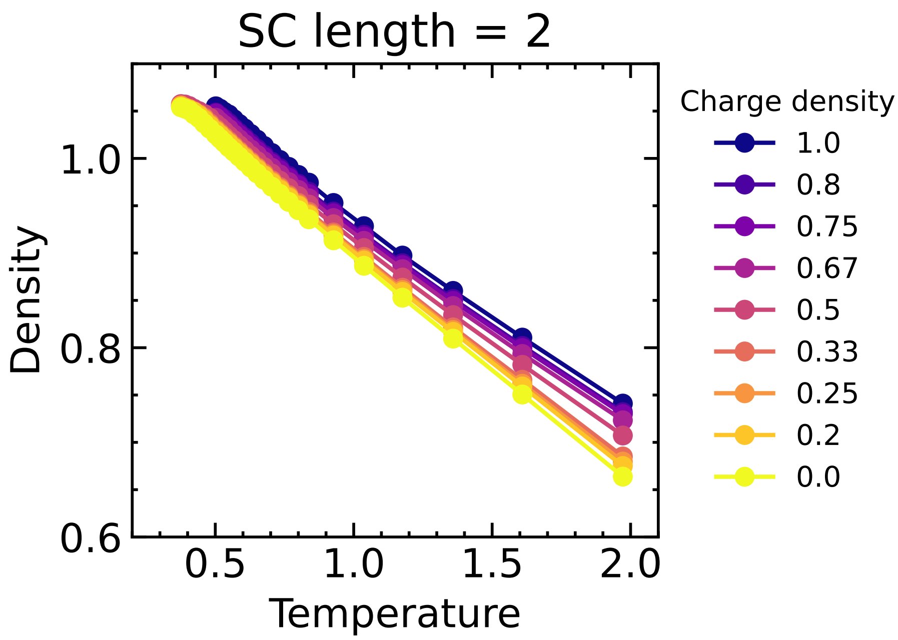

Fig. S1: The density as a function of temperature for compleximers with $SC = 2$ and varying charge density.

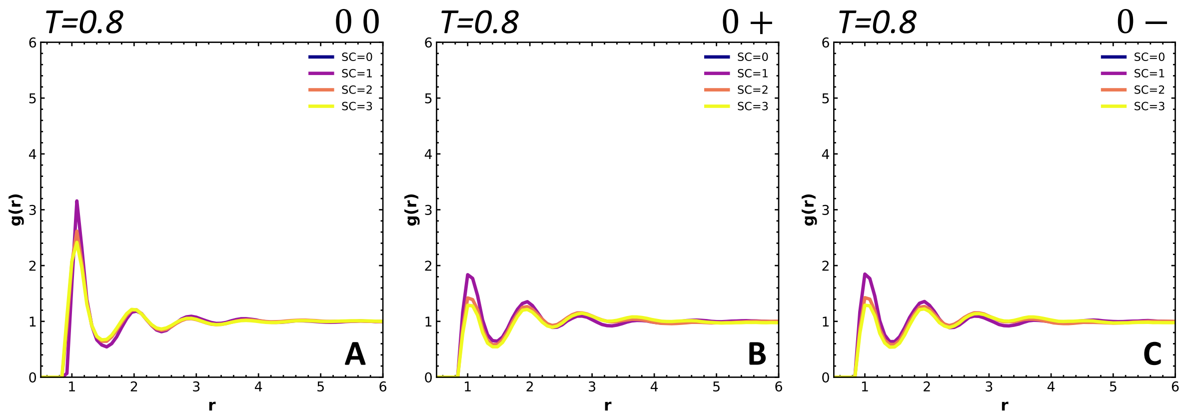

Fig. S2: The radial distribution function ($g_{ab}(r)$) of compleximers ($CD = 1$) with varying SC length at $T = 0.8$ for the pair interactions **(A)**, 00. **(B)**, 0+. **(C)**, 0−.

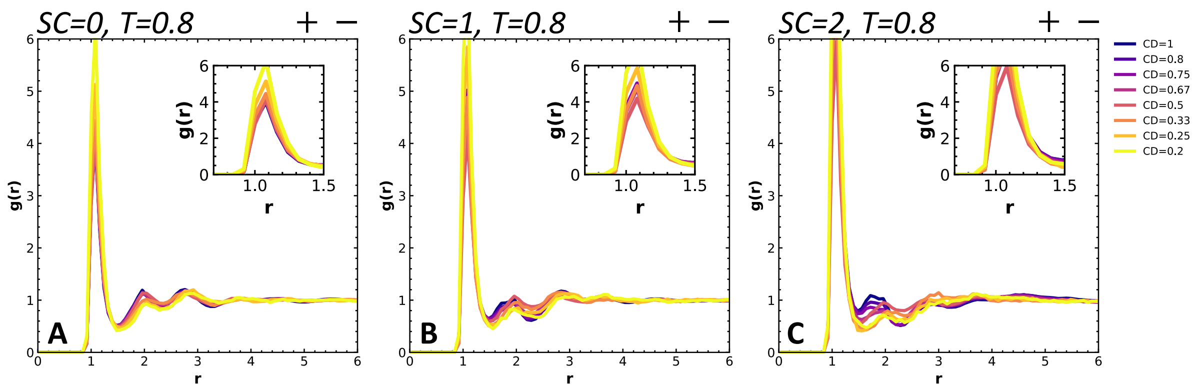

Fig. S3: The radial distribution function ($g_{+-}(r)$) of compleximers with varying CD and SC length for the +− pair interactions at $T = 0.8$, for **(A)** $SC = 0$, **(B)** $SC = 1$, and **(C)** $SC = 2$. The inset shows the broadening of the first peak.

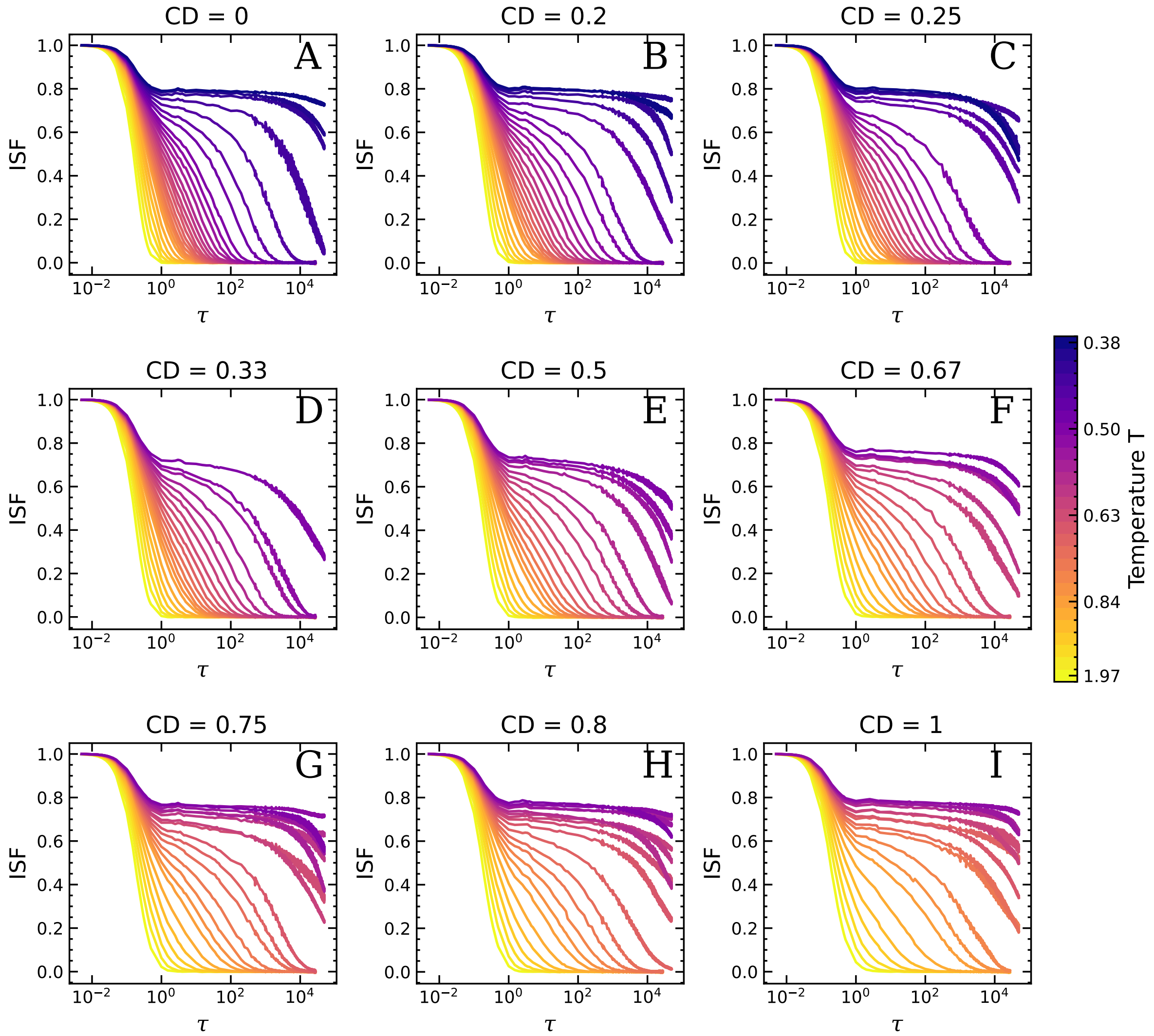

Fig. S4: The intermediate scattering function of compleximers with varying charge density CD, for $SC = 0$. **(A)**, $CD = 0$. **(B)**, $CD = 0.2$. **(C)**, $CD = 0.25$. **(D)**, $CD = 0.33$. **(E)**, $CD = 0.5$. **(F)**, $CD = 0.67$. **(G)**, $CD = 0.75$. **(H)**, $CD = 0.8$. **(I)**, $CD = 1$. The ISF of some temperatures below T_g do not decay to 0.2, these curves are omitted from the τ_α calculations.

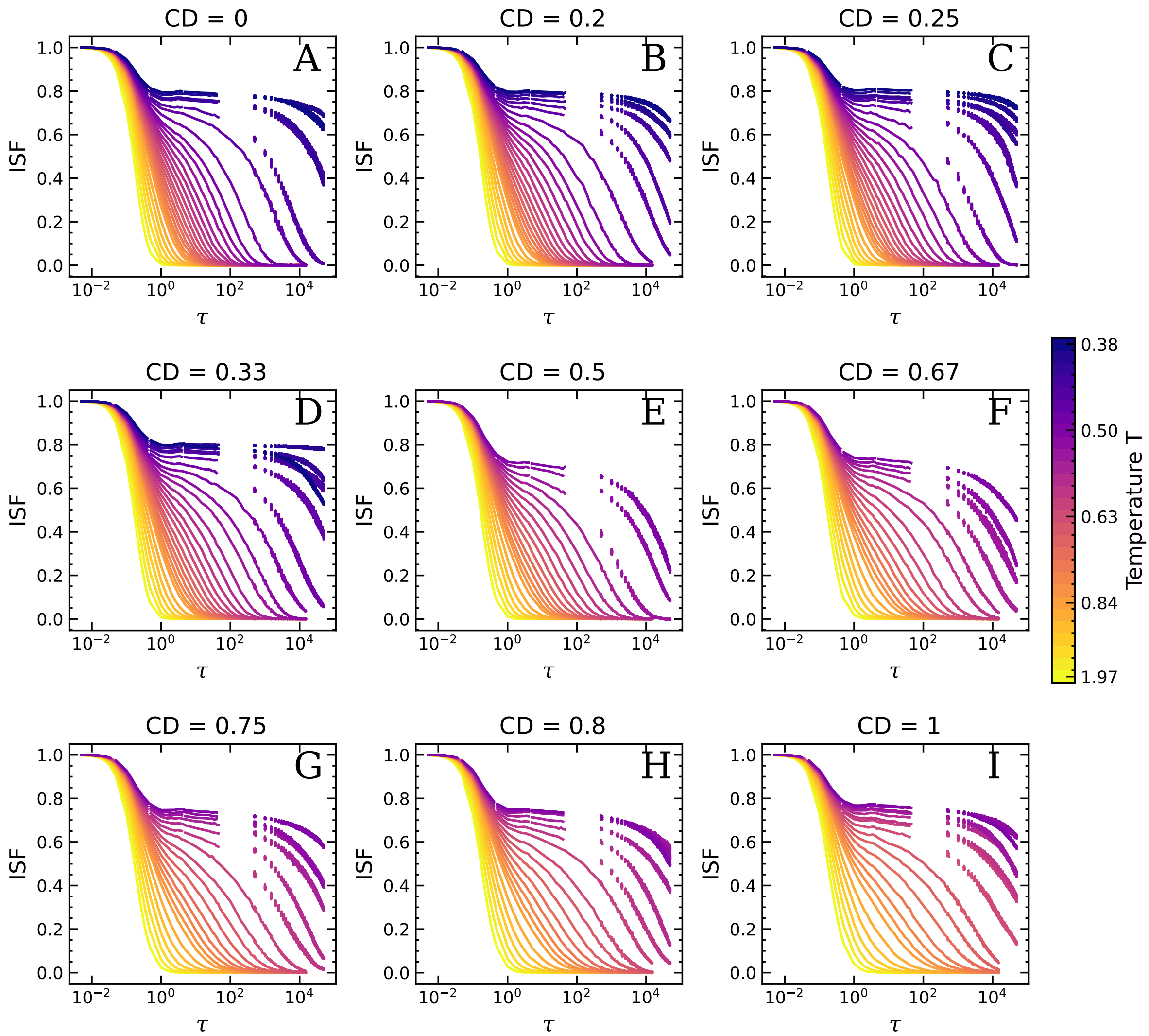

Fig. S5: The intermediate scattering function of compleximers with varying charge density CD, for $SC = 1$. **(A)**, $CD = 0$. **(B)**, $CD = 0.2$. **(C)**, $CD = 0.25$. **(D)**, $CD = 0.33$. **(E)**, $CD = 0.5$. **(F)**, $CD = 0.67$. **(G)**, $CD = 0.75$. **(H)**, $CD = 0.8$. **(I)**, $CD = 1$.

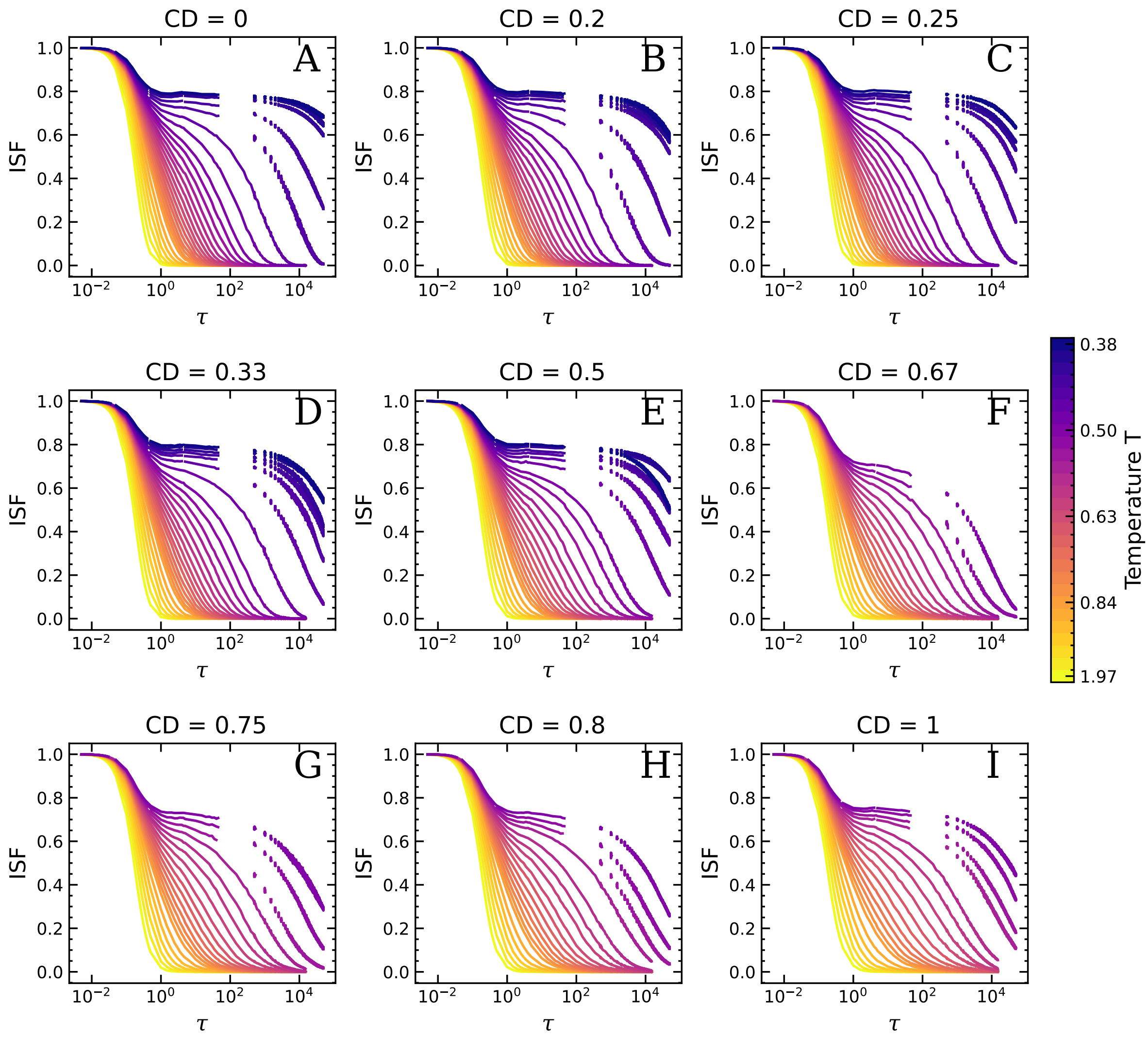

Fig. S6: The intermediate scattering function of compleximers with varying charge density CD, for $SC = 2$. **(A)**, $CD = 0$. **(B)**, $CD = 0.2$. **(C)**, $CD = 0.25$. **(D)**, $CD = 0.33$. **(E)**, $CD = 0.5$. **(F)**, $CD = 0.67$. **(G)**, $CD = 0.75$. **(H)**, $CD = 0.8$. **(I)**, $CD = 1$. The ISF of some temperatures below T_g do not decay to 0.2, these curves are omitted from the τ_α calculations.

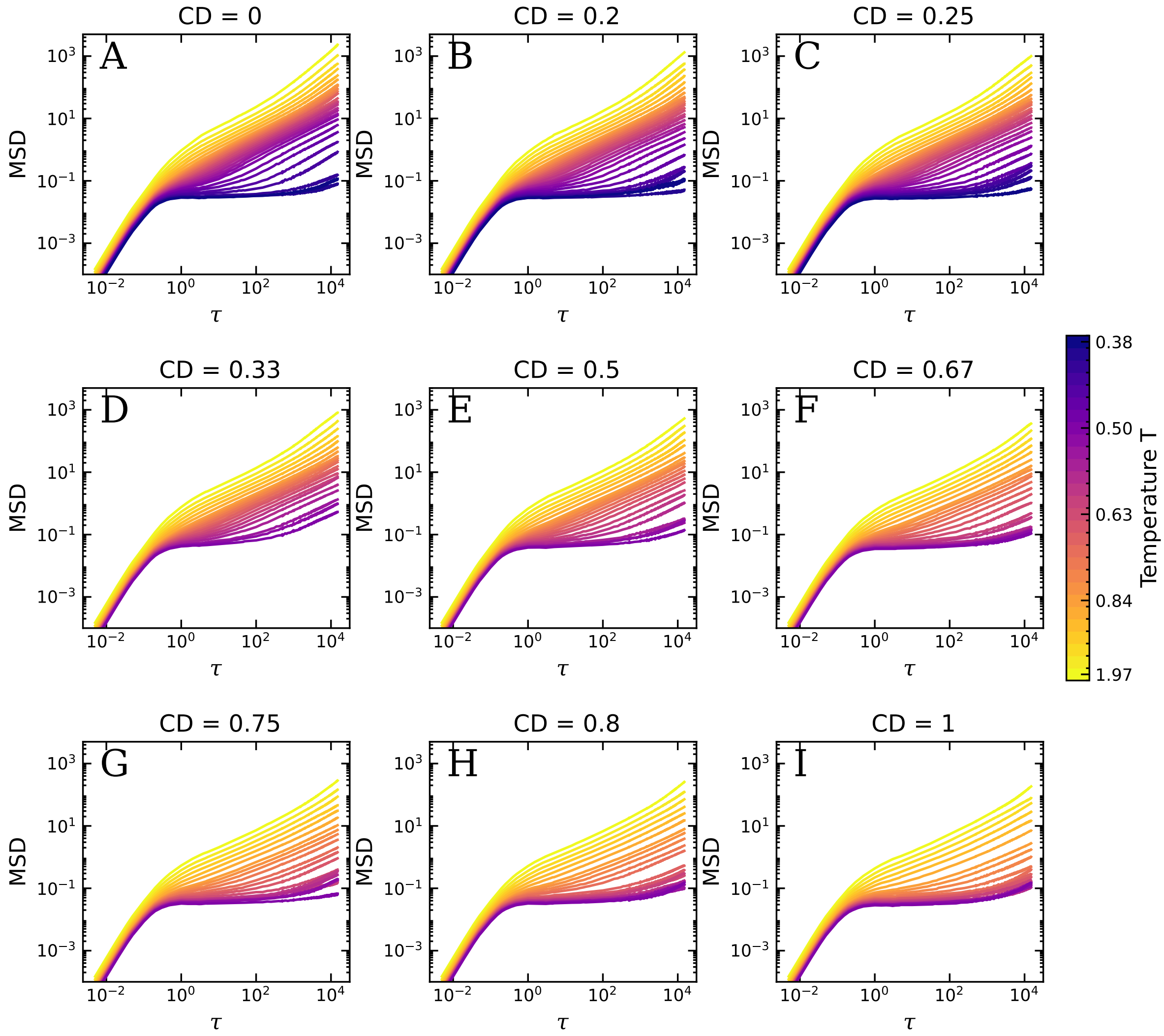

Fig. S7: The mean squared displacement (MSD) of compleximers with varying charge density (CD) at a side chain length $SC = 0$. **(A)**, $CD = 0$. **(B)**, $CD = 0.2$. **(C)**, $CD = 0.25$. **(D)**, $CD = 0.33$. **(E)**, $CD = 0.5$. **(F)**, $CD = 0.67$. **(G)**, $CD = 0.75$. **(H)**, $CD = 0.8$. **(I)**, $CD = 1$. The ISF of some temperatures below T_g do not decay to 0.2, these curves are omitted from the τ_α calculations.

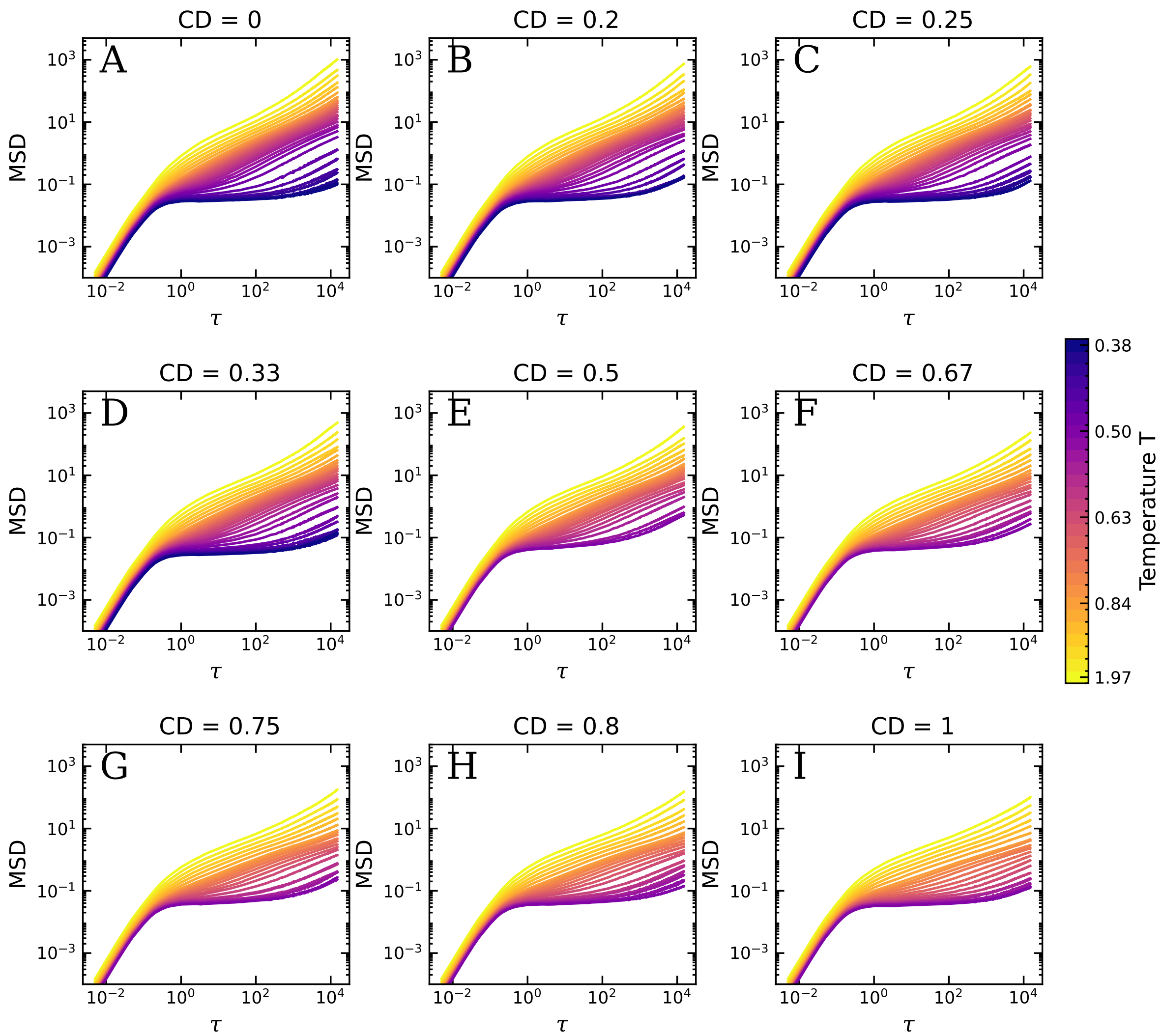

Fig. S8: The mean squared displacement (MSD) of compleximers with varying charge density (CD) at a side chain length $SC = 1$. **(A)**, $CD = 0$. **(B)**, $CD = 0.2$. **(C)**, $CD = 0.25$. **(D)**, $CD = 0.33$. **(E)**, $CD = 0.5$. **(F)**, $CD = 0.67$. **(G)**, $CD = 0.75$. **(H)**, $CD = 0.8$. **(I)**, $CD = 1$.

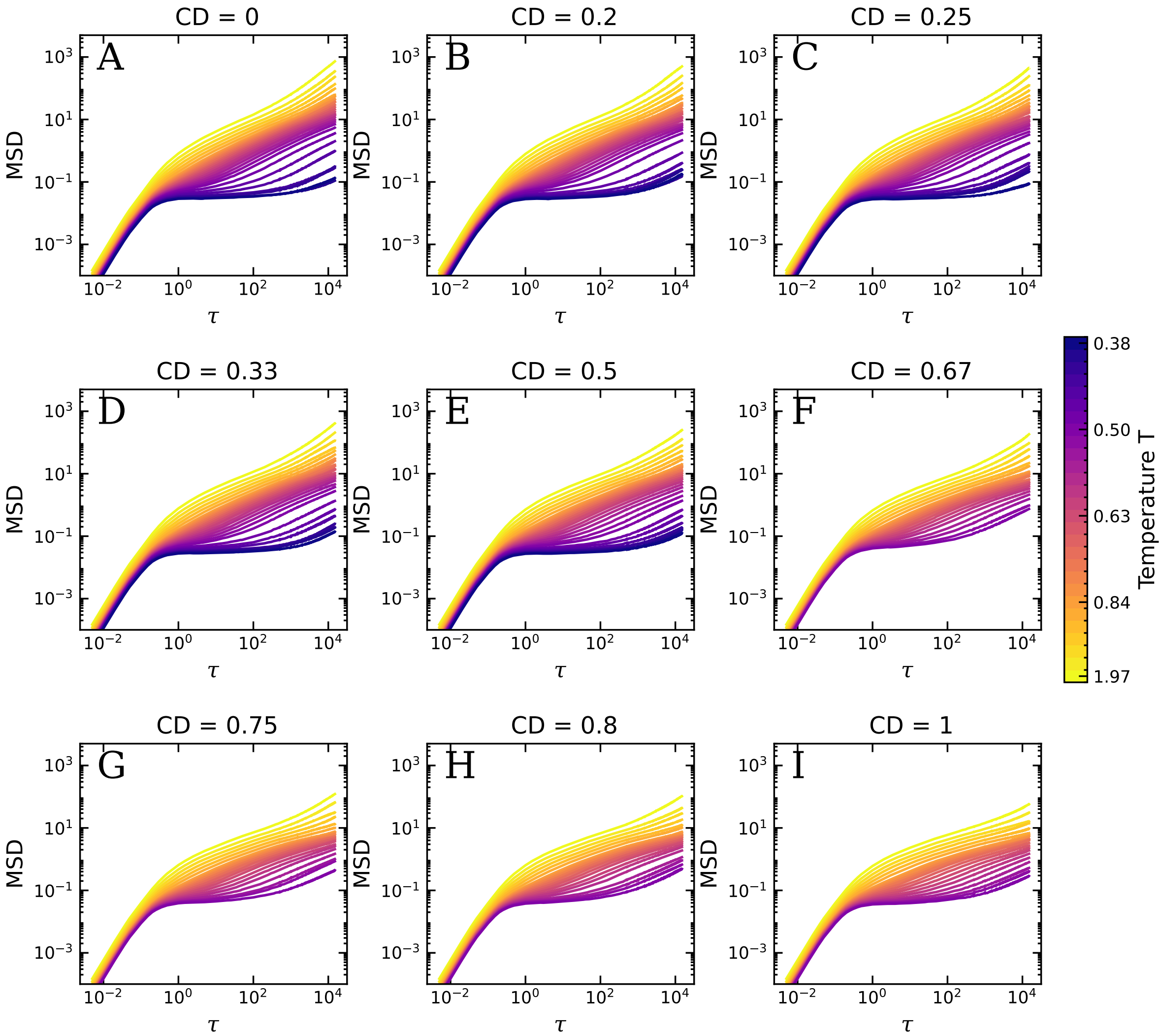

Fig. S9: The mean squared displacement (MSD) of compleximers with varying charge density (CD) at a side chain length $SC = 2$. **(A)**, $CD = 0$. **(B)**, $CD = 0.2$. **(C)**, $CD = 0.25$. **(D)**, $CD = 0.33$. **(E)**, $CD = 0.5$. **(F)**, $CD = 0.67$. **(G)**, $CD = 0.75$. **(H)**, $CD = 0.8$. **(I)**, $CD = 1$.

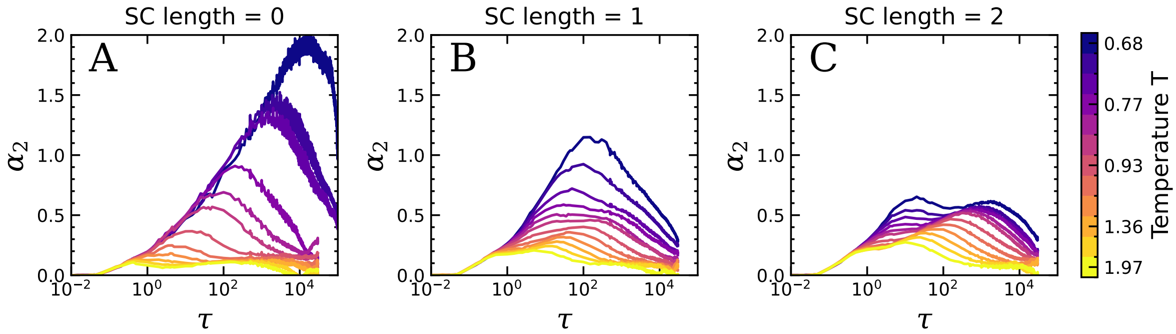

Fig. S10: The non-Gaussian parameter (α_2) of compleximers with varying side chain length (SC) at a charge density $CD = 1$. **(A)**, $SC = 0$, **(B)**, $SC = 1$, **(C)**, $SC = 2$, **(D)**, $SC = 3$.

The slowing down of the particles upon approaching the glass transition is often accompanied by dynamic heterogeneities, where the particle mobility is different in different locations of the material. To characterize these dynamic heterogeneities, we calculate the non-Gaussian Parameter (α_2), defined as:

$$\alpha_2(t) = \frac{3\langle \Delta r^4(t) \rangle}{5\langle \Delta r^2(t) \rangle^2} - 1,$$

where $\langle \Delta r^2(t) \rangle$ is the mean squared displacement (MSD) and $\langle \Delta r^4(t) \rangle$ is the fourth moment of the displacement distribution. A value of $\alpha_2(t)$ close to zero indicates a Gaussian displacement distribution, while a non-zero value reflects non-Gaussian dynamics, typically arising in systems exhibiting spatial heterogeneity of the dynamics. We find that α_2 peaks around the cage relaxation time, which is the time scale of maximum cooperative rearrangements, typically corresponding to τ_α. As the temperature decreases the peak shifts to longer times and becomes higher, indicating more pronounced dynamic heterogeneities . We find similar behavior across systems with varying SC length and varying charge density.

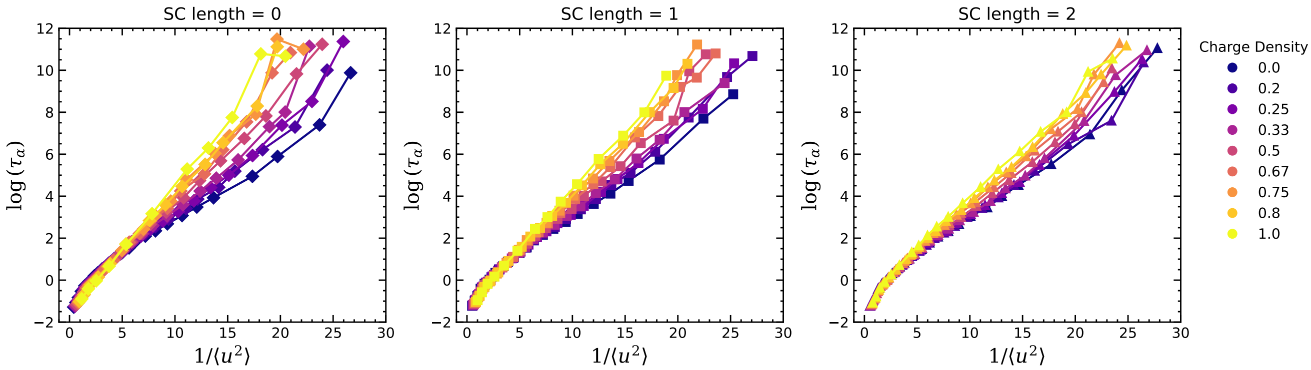

Fig. S11: $\log(\tau_\alpha)$ vs $1/\langle u^2 \rangle$ for different SC and CD.