

Supporting Information

**Chaotropic Effect of Ions on the Self-Aggregating Propensity of
Whitlock's Molecular Tweezer**

Madhusmita Devi and Sandip Paul*

Department of Chemistry, Indian Institute of Technology, Guwahati Assam, India-781039

(Dated: May 3, 2022)

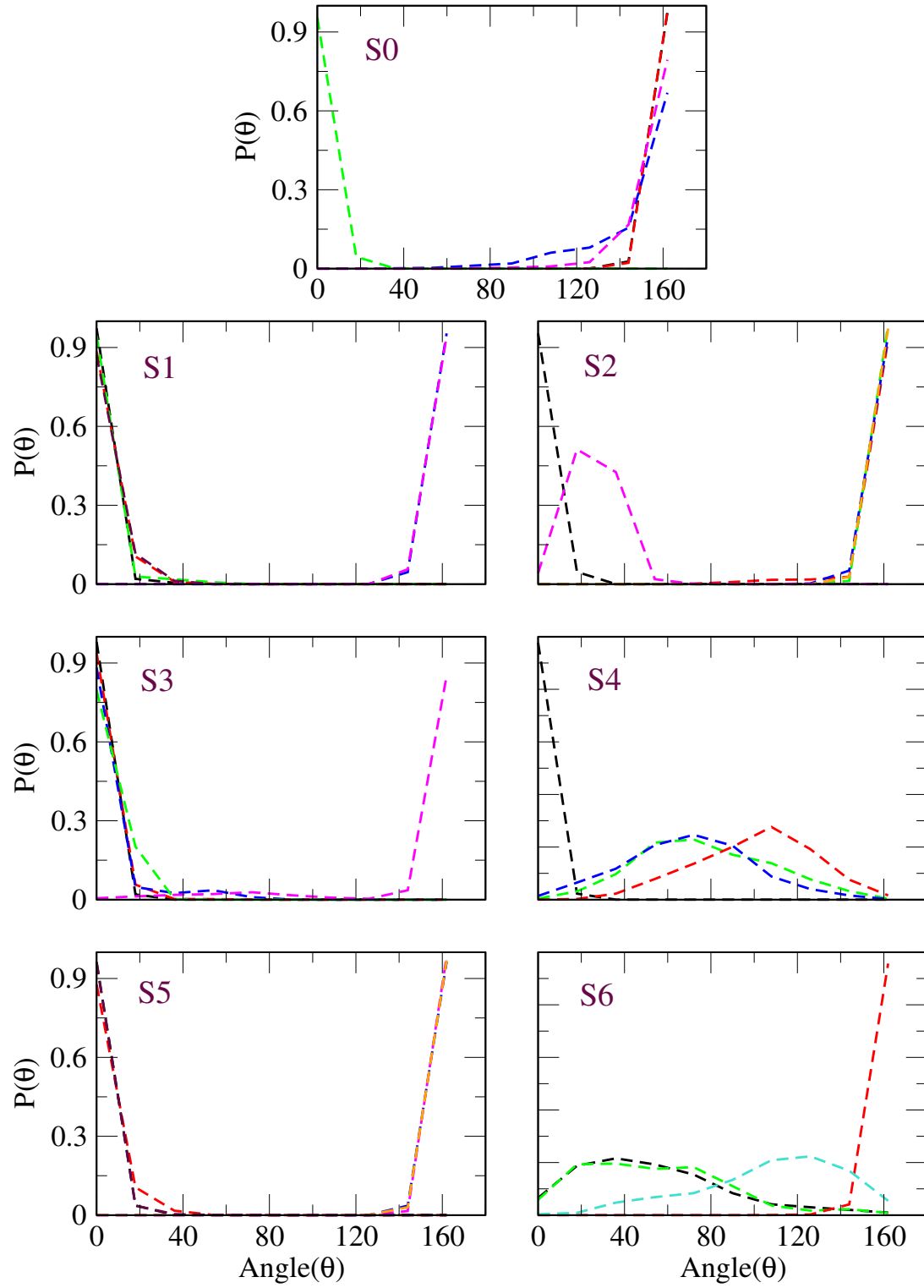


FIG. S1: Probability $[P(\theta)]$ of the angle of orientation (θ) between aromatic arms constituting two different tweezer molecules. For systems S4 and S6, angle criteria is violated for most of the tweezer pairs, indicating disruption in the π - π stacking interactions.

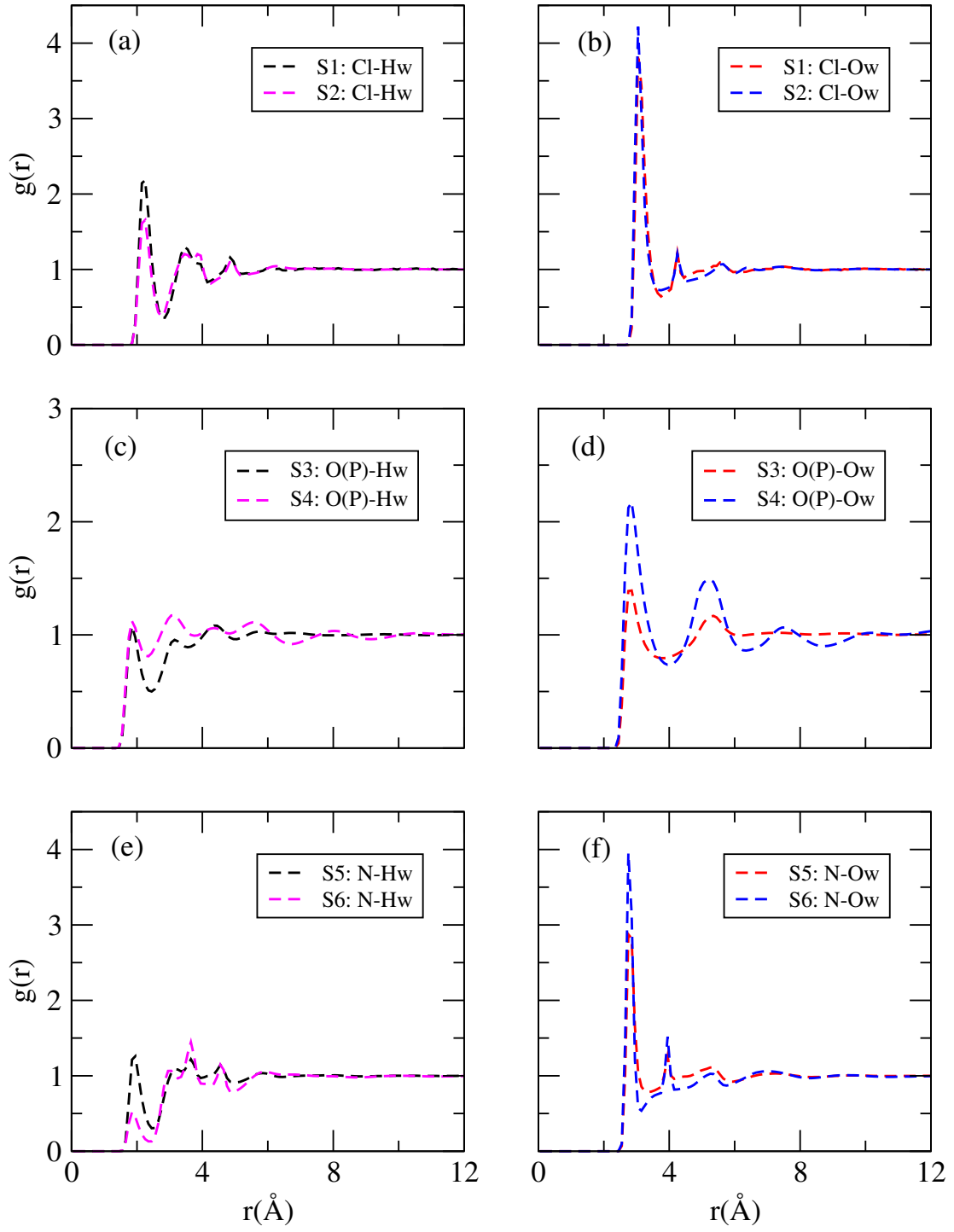


FIG. S2: Site-site radial distribution functions involving (a) Cl-H_w (b) Cl-O_w corresponding to systems, S1 and S2, (c) O(P)-H_w (d) O(P)-O_w for systems, S3 and S4, ('O(P)' indicates oxygen of perchlorate ion) and, (e) N-H_w (f) N-O_w for systems S5 and S6.

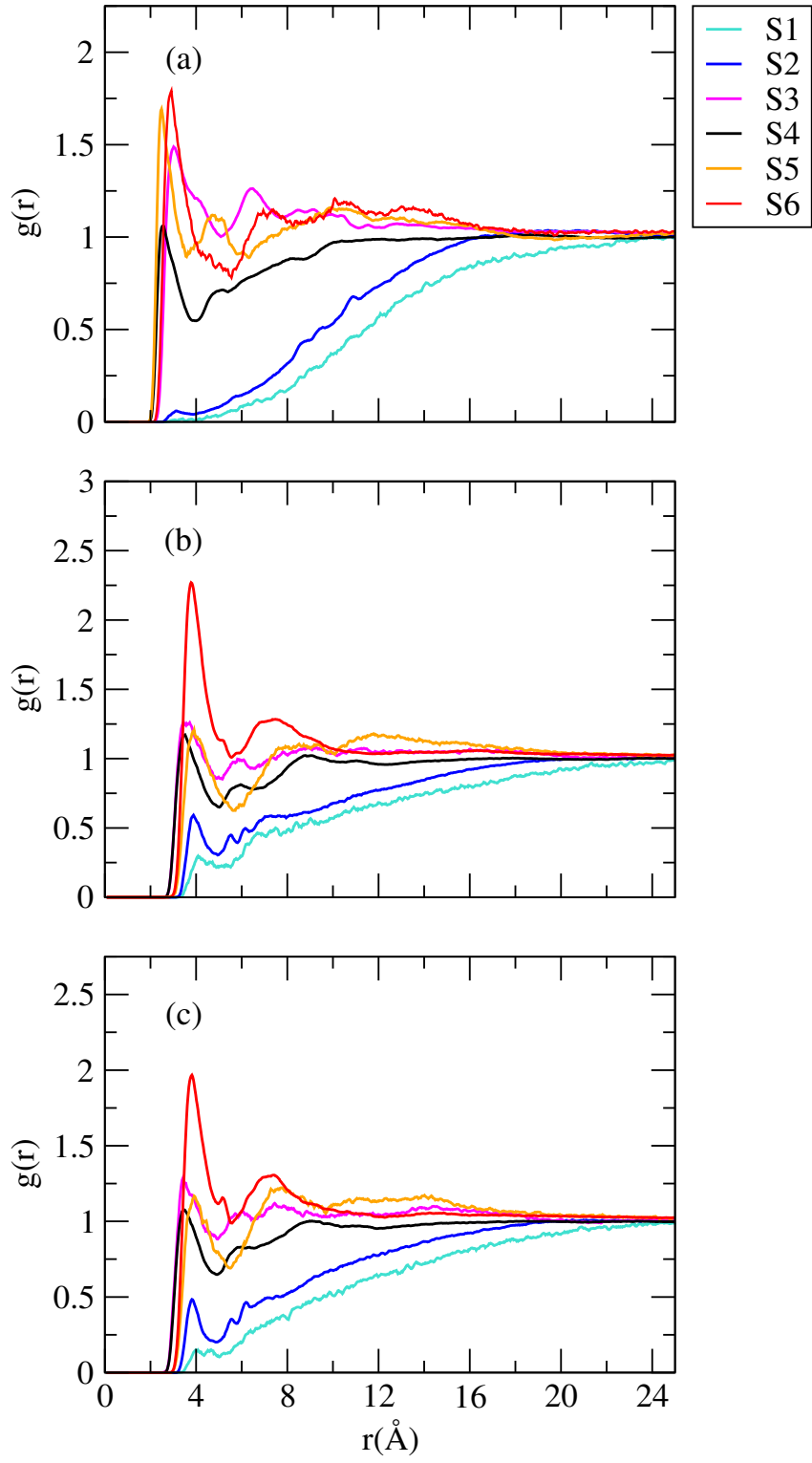


FIG. S3: Site-site radial distribution functions involving anions around selective atomic sites of the tweezer molecule. (a) H49/H50-anion, (b) C13/C14-anion, (c) C15/C16-anion.

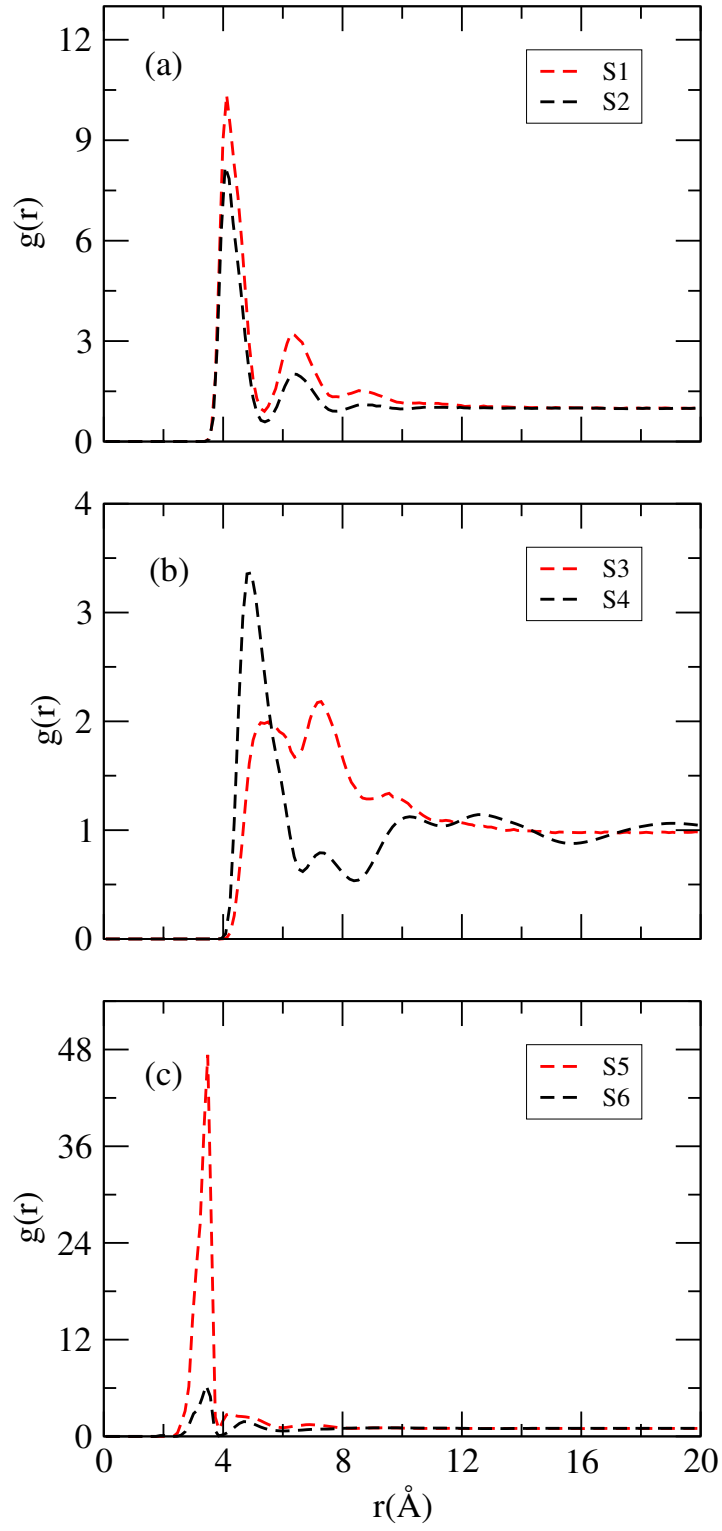


FIG. S4: Radial distribution functions involving cation-anion pairs. (a) $\text{Al}^{3+}\text{-Cl}^-$ corresponding to systems, S1 and S2 (b) $\text{Al}^{3+}\text{-ClO}_4^-$ for systems, S3 and S4 (c) $\text{Al}^{3+}\text{-SCN}^-$ for systems, S5 and S6.

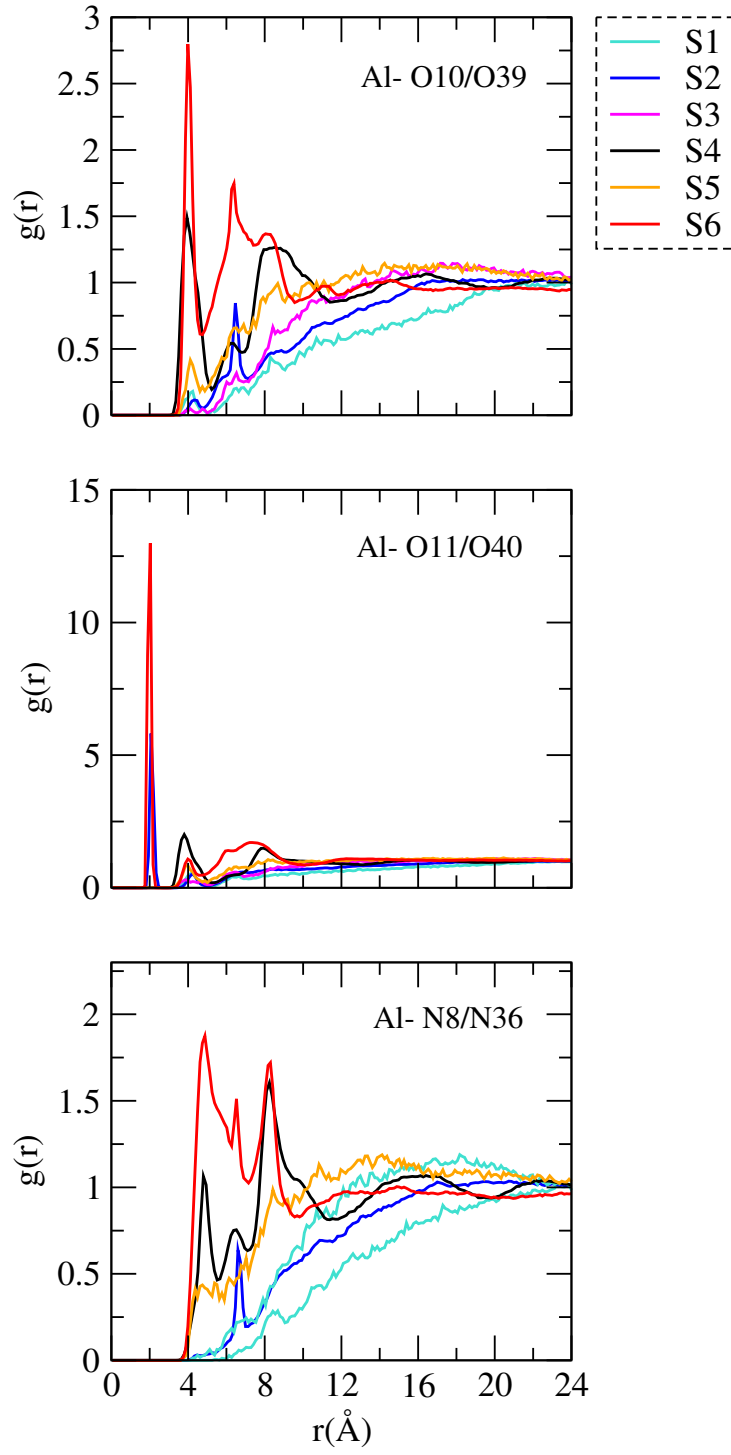


FIG. S5: Site-site radial distribution functions involving (a) Al^{3+} -O10/O39 (b) Al^{3+} -O11/O40, and (c) Al^{3+} -N8/N36.

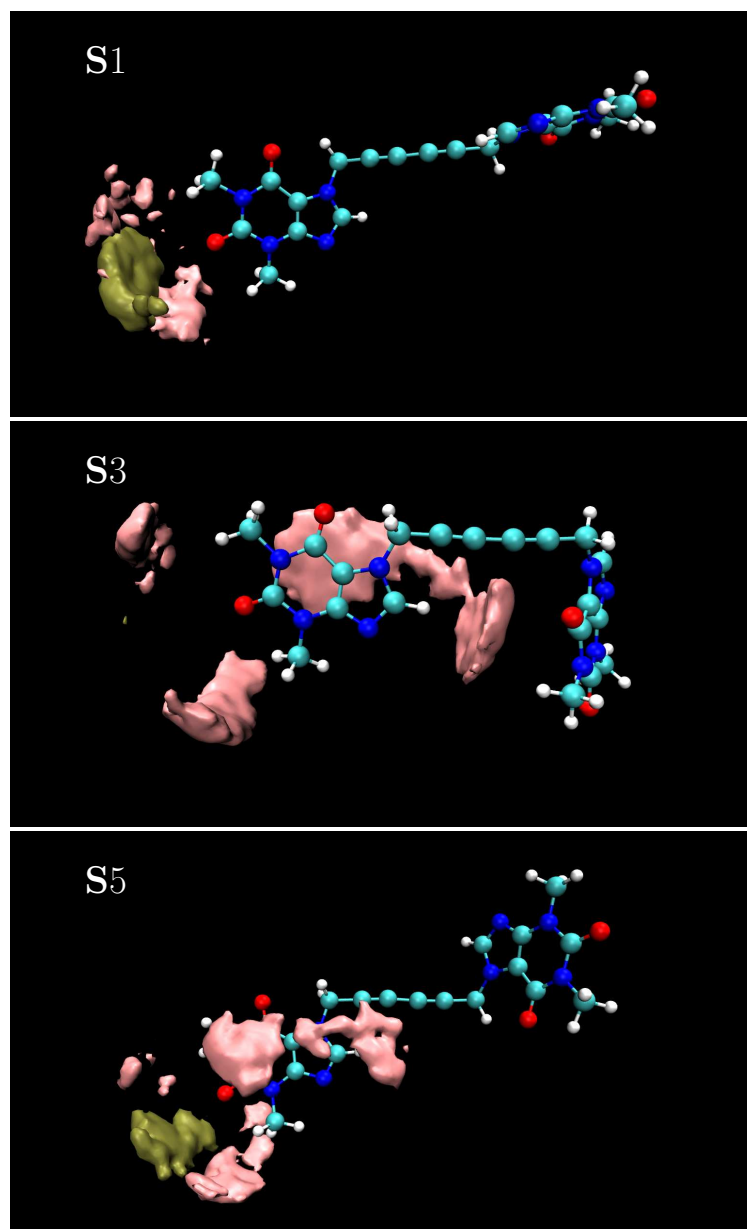


FIG. S6: Contour density plots displaying cation and anion density within 6.0 Å around tweezer arm in the lower concentrated salt systems. Tan and pink color represents cation and anion densities, respectively.

TABLE S1: Hydration number around the selected hydrophilic and hydrophobic sites of tweezer molecule are tabulated here. To account for the reduced number density of water in salt solutions, the coordination number values are normalized with respect to the box volume of the pure tweezer-water system and the same are presented in parentheses.

System	S0	S1	S2	S3	S4	S5	S6
O10/O39	1.49	1.46 (1.46)	1.56 (1.74)	1.86 (1.89)	1.91 (2.75)	1.53 (1.54)	1.75 (2.41)
O11/O40	2.18	1.98 (1.98)	2.10 (2.35)	2.38 (2.42)	2.22 (3.19)	2.09 (2.11)	2.10 (2.89)
N8/N36	1.15	0.96 (0.96)	1.35 (1.51)	1.37 (1.39)	1.81 (2.60)	1.18 (1.19)	1.62 (2.23)
C5/C31	4.46	3.26 (3.26)	3.82 (4.27)	4.76 (4.85)	4.92 (7.08)	3.94 (3.97)	4.33 (5.97)
H49/H50	2.22	1.11 (1.11)	1.70 (1.90)	2.39 (2.43)	4.11 (5.91)	1.73 (1.74)	4.05 (5.58)
C12/C45	11.49	11.23 (11.23)	10.29 (11.52)	11.67 (11.90)	10.89 (15.68)	10.54 (10.64)	9.78 (13.49)
C16/C41	10.91	10.35 (10.35)	10.45 (11.70)	11.07 (11.29)	10.68 (15.37)	10.46 (10.56)	9.72 (13.41)

* Electronic address: sandipp@iitg.ac.in