FIGURE S1

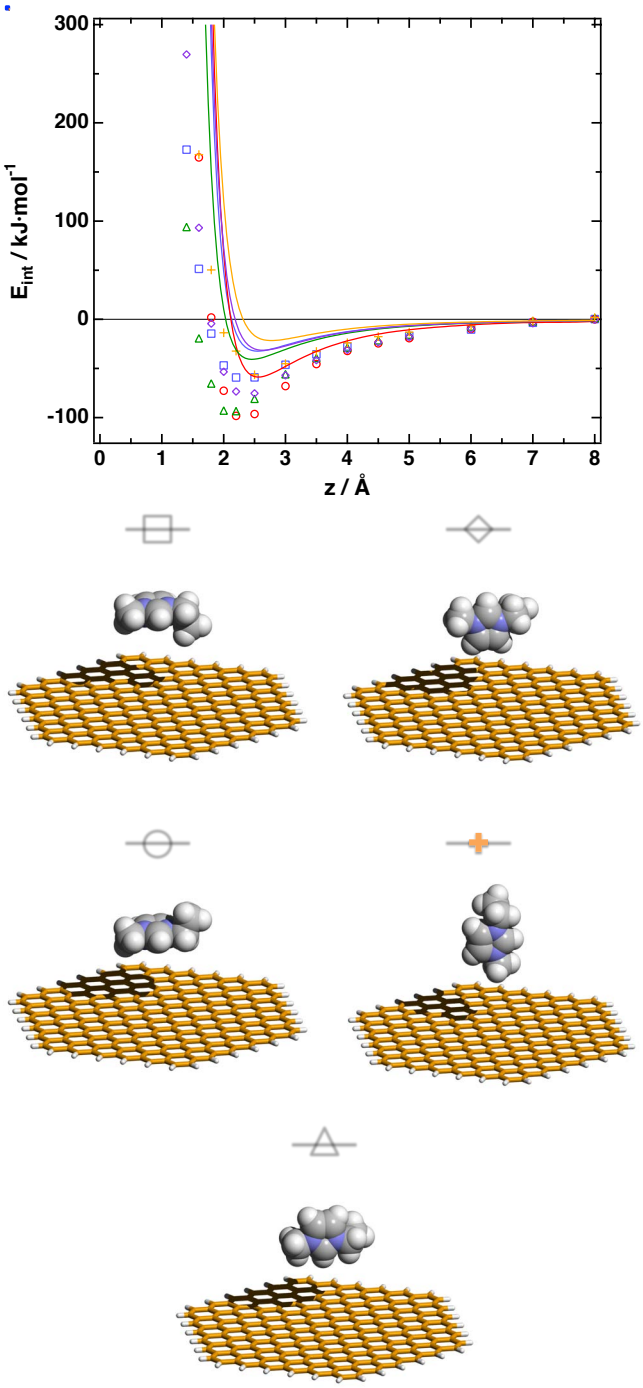

Figure S1. Discrepancies of the interaction energies between the $[C_2C_1im]^+$ cation and the graphene cluster calculated by BLYP-D[1-3]/TZVP[4-5] (symbols) and those calculated using a Lennard–Jones potential[6-10] with σ = 3.55 Å and ε = 0.2929 kJ·mol^{-1}. It illustrates the importance of developing a specific model for the interactions between ionic liquids and metal surfaces.

FIGURE S2

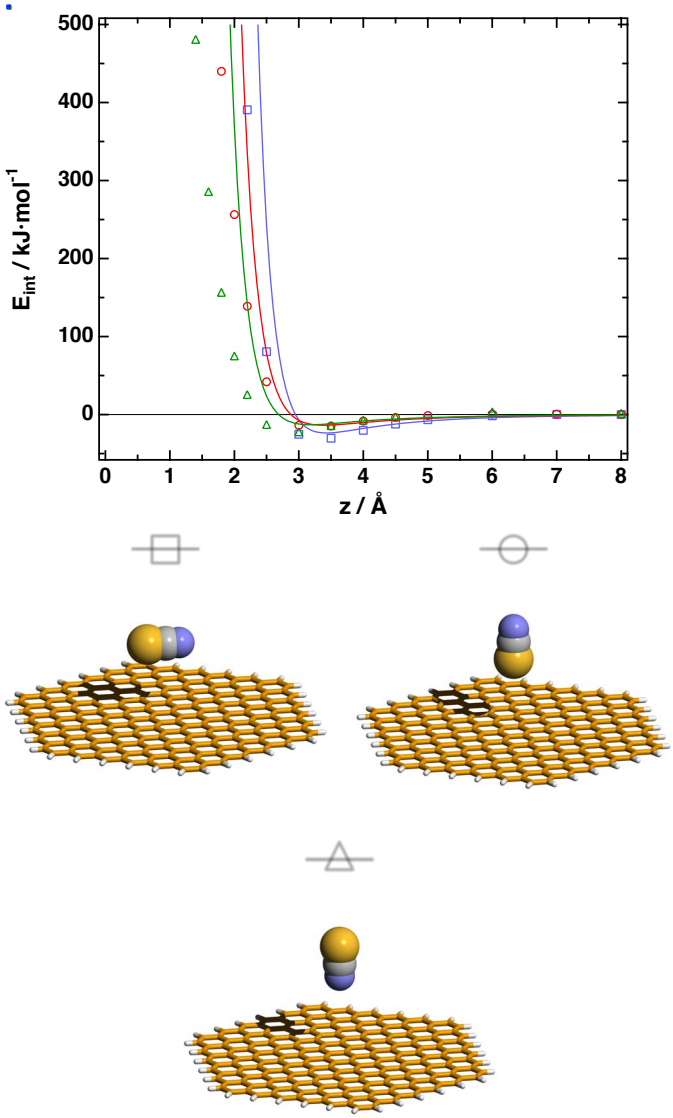

Figure S2. Discrepancies of the interaction energies between the $[SCN]^+$ anion and the graphene cluster calculated by BLYP-D[1-3]/TZVP[4-5] (symbols) and those calculated using a Lennard–Jones potential[6-10] with $\sigma = 3.55$ Å and $\varepsilon = 0.2929$ kJ·mol^{-1}.

FIGURE S3

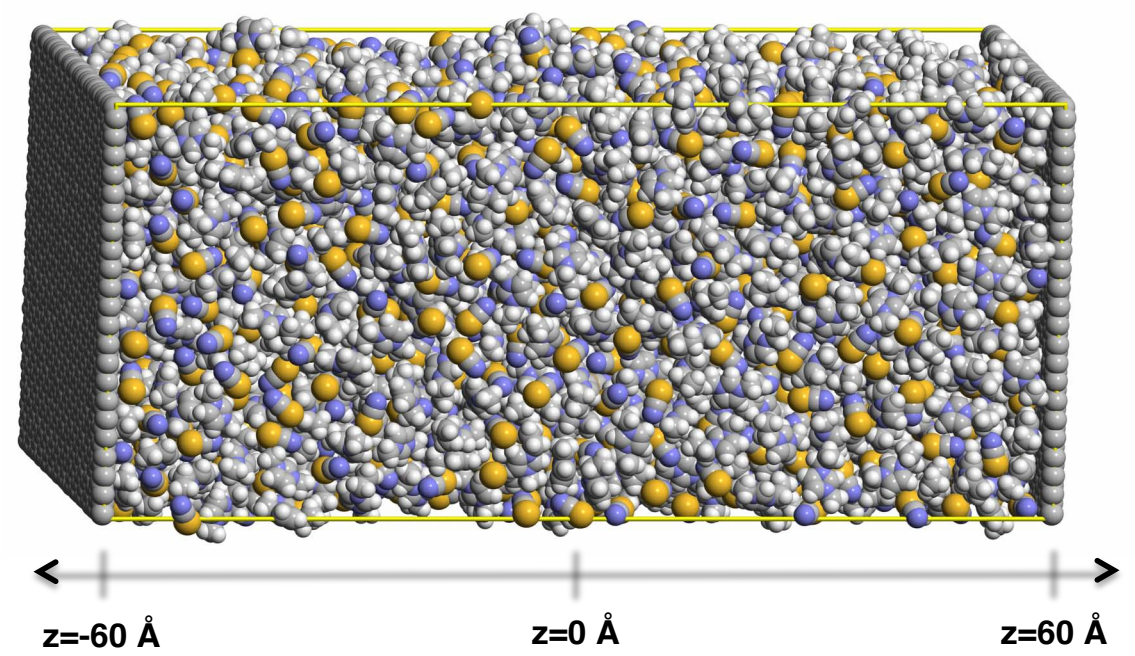

Figure S3. Snapshot of the simulation box containing the flat graphene-ionic liquid interface. The simulation box is centered in $z = 0$.

FIGURE S4

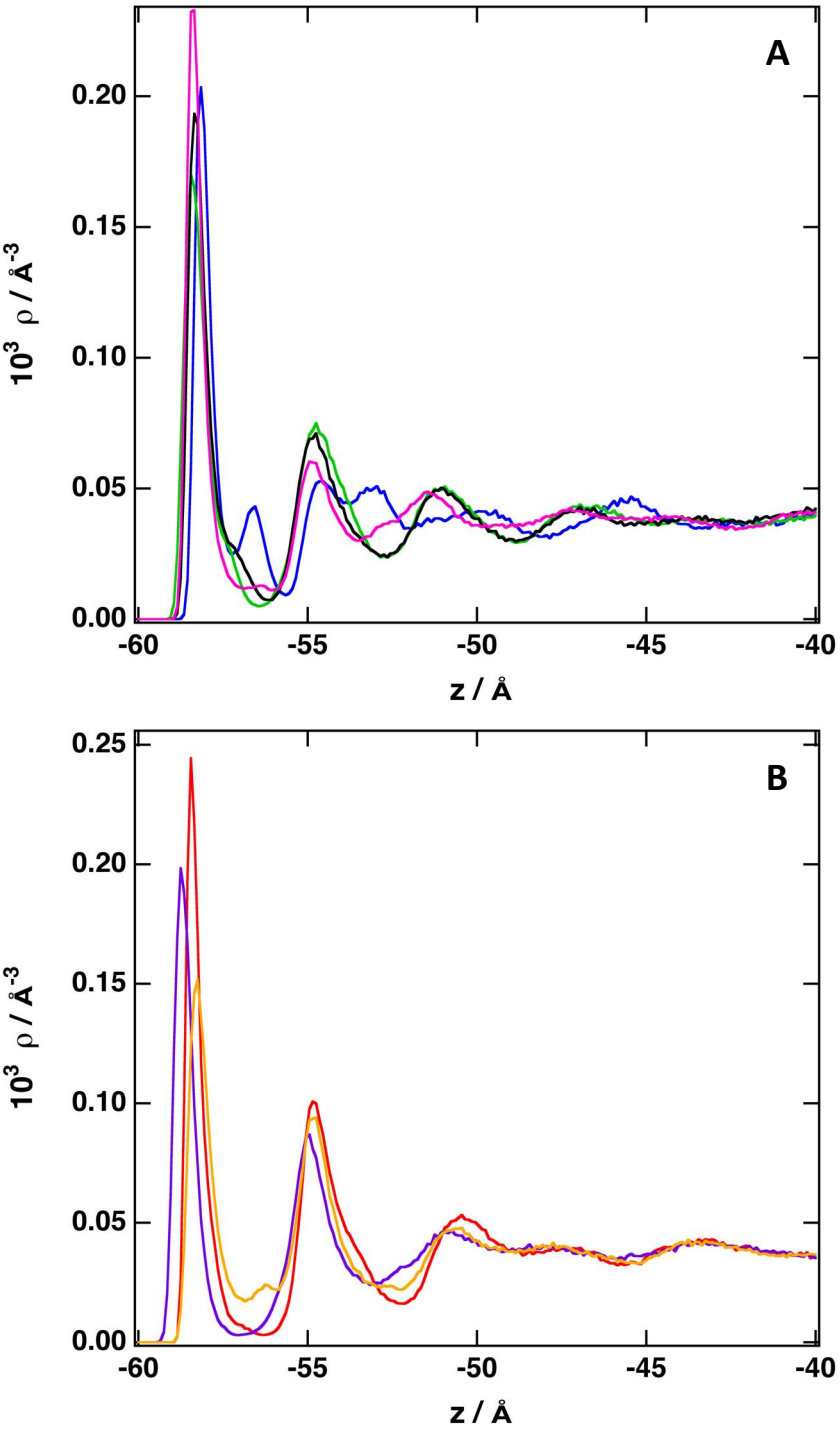

Figure S4. Number density profile of individual atoms of A) cation: C2 (blue), C4-C5 (magenta), C7 (black), C8 (green); and B) anion C (red) N (purple) and S (orange). See Figure 1 for labeling.

FIGURE S5

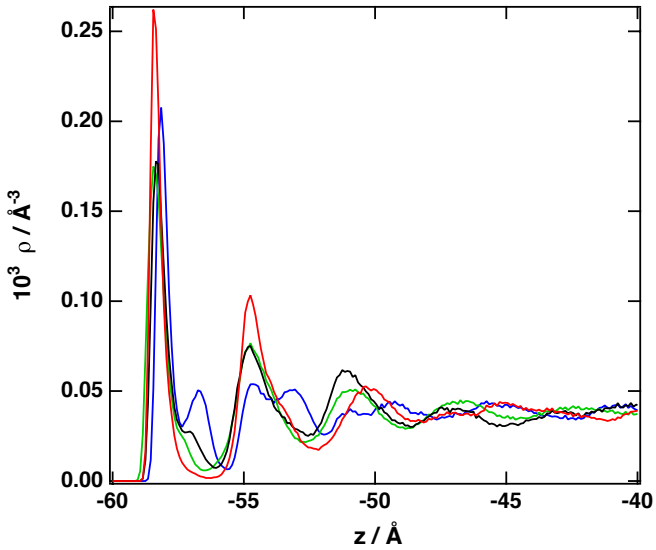

Figure S5. Number density profile of individual atoms when the polarization of the graphene surface is not explicitly considered: C2 cation (blue), C7 cation (black), C8 cation (green) and C anion (red). See Figure 1 for labeling.

FIGURE S6

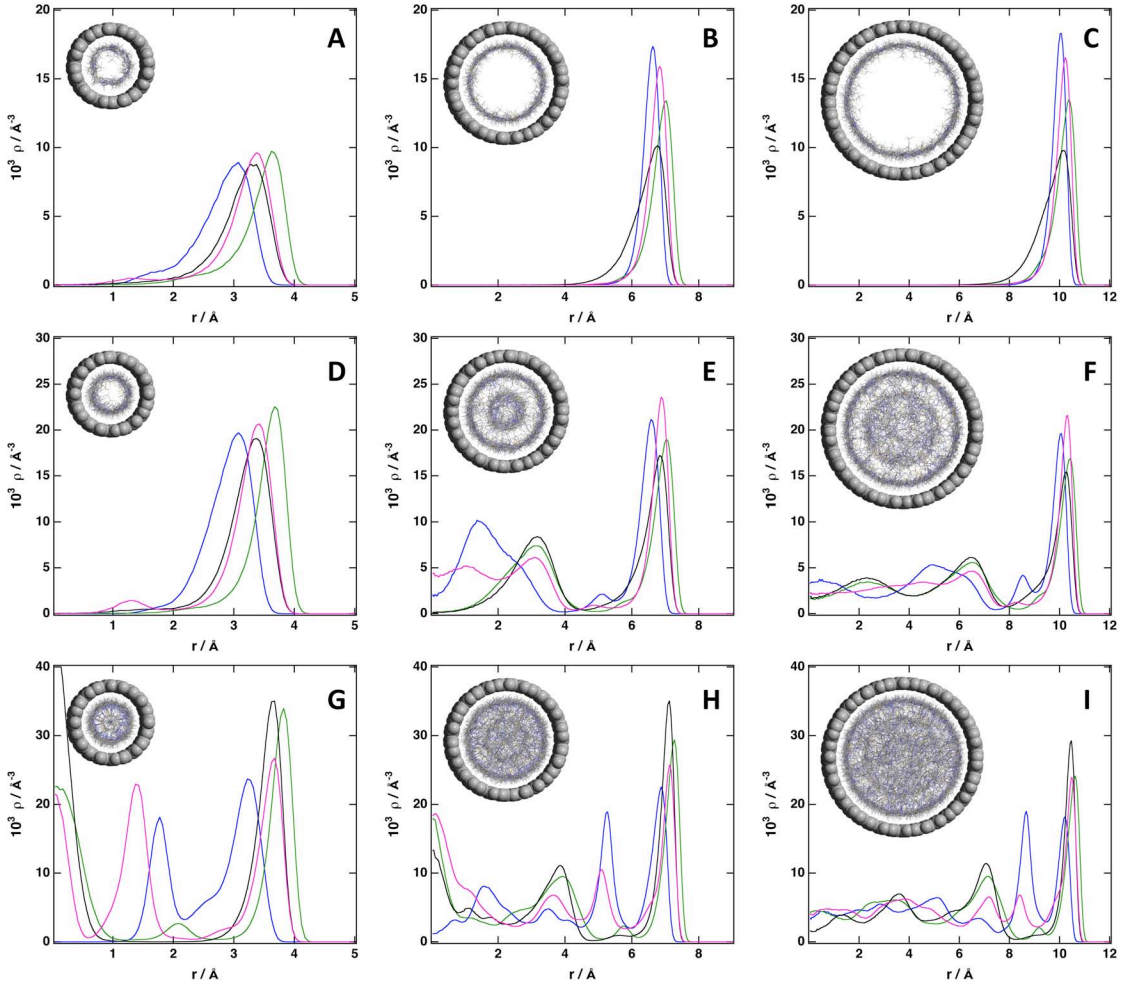

Figure S6. Number density profile of individual atoms of the cation inside of (20,0) CNT (left column), (15,0) CNT (central column) and (10,0) CNT (right column): C2 (blue), C4-C5 (magenta), C7 (black) and C8 (green). First row $\rho_{CNT} = 0.5\rho_{bulk}$, central row $\rho_{CNT} = 1.0\rho_{bulk}$, bottom row $\rho_{CNT} = 1.2\rho_{bulk}$.

FIGURE S7

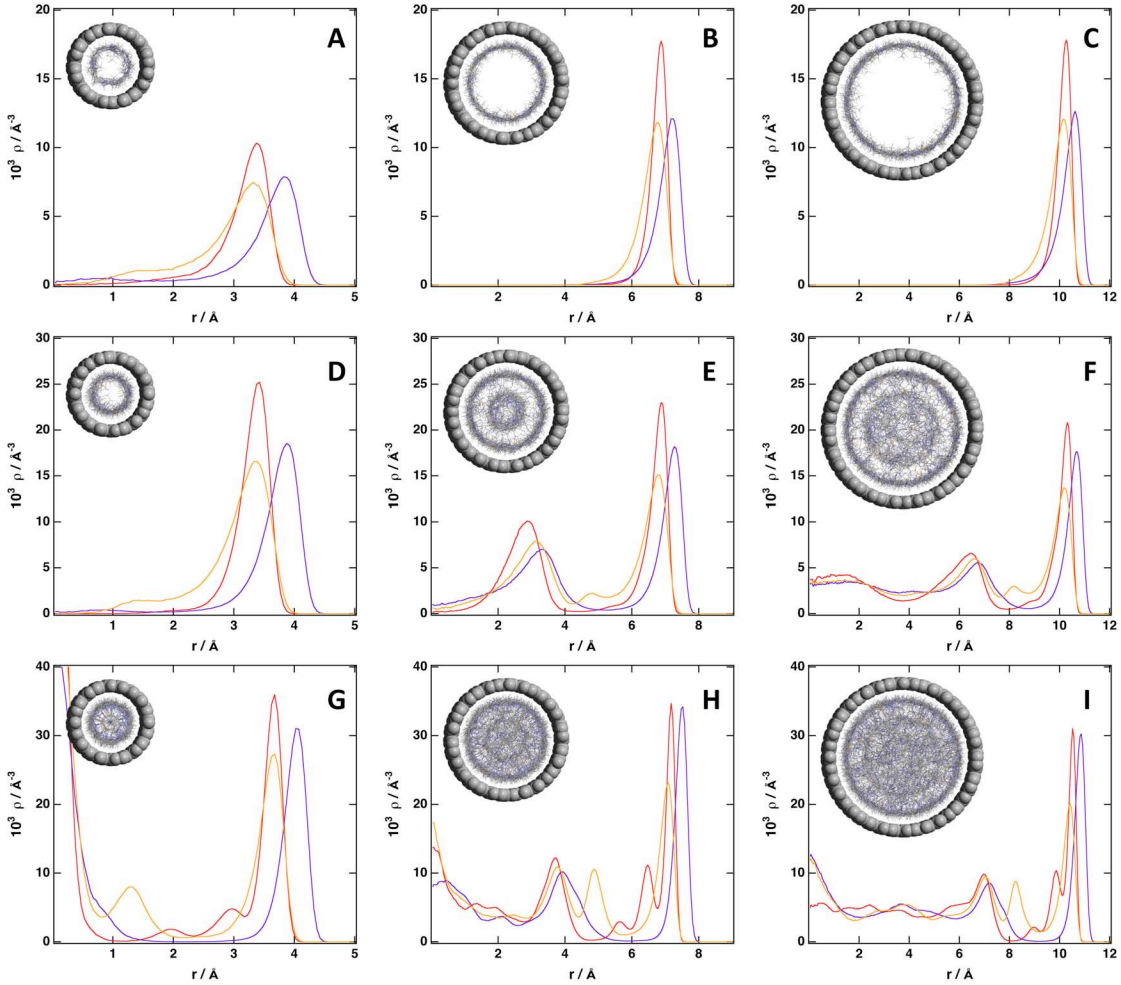

Figure S7. Number density profile of individual atoms of the anion inside of (20,0) CNT (left column), (15,0) CNT (central column) and (10,0) CNT (right column): C (red), N (purple) and S (orange). First row $\rho_{CNT} = 0.5\rho_{bulk}$, central row $\rho_{CNT} = 1.0\rho_{bulk}$, bottom row $\rho_{CNT} = 1.2\rho_{bulk}$.

Table S1. Parameters of the intermolecular potential adjusted to the BLYP-D[1-3]/TZVP[4-5] calculations. $U(r)=\frac{E_0}{(n-m)}\left[m\left(\frac{r_0}{r}\right)^n - n\left(\frac{r_0}{r}\right)^m\right]$, n=9, m=6.

Atom	E_0	r_0
C2	1.113	4.052
C4,5	1.015	3.772
C6,8	1.328	3.552
C7	0.299	2.850
C	0.828	3.754
N	0.925	3.321
S	0.800	3.835

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