TABLES

Table S1. The dimensions (diameters and lengths) of CNTs and the number of solvent (water/urea/Gdm) molecules in 8M urea and in 6M GdmCl.

CNT	Diameter (in Å)	Length (in Å)	Water (8M urea)	Urea (8M urea)	Water (6M GdmCl)	Gdm (6M GdmCl)
(6, 6)	8.12	13.50	973	225	1031	154
(8, 5)	8.88	16.10	962	222	836	141
(9, 6)	10.22	18.22	939	221	925	137
(10, 8)	12.2	33.04	3292	777	2981	544
(17, 8)	17.29	31.55	3253	747	3264	483

Table S2. Force-field parameters used in MD simulations.

	atom	$R_{min}/2$ (Å)	ε (Kcal/mol)	q, e
CNT	C	1.992	-0.07	0.00
water	O	2.00	-0.152	-0.834
	H	0.224	-0.046	0.417
Gdm	C	2.00	-0.11	0.64
	N	1.85	-0.2	-0.8
	H	0.224	-0.046	0.46
urea	C	2.00	-0.11	0.51
	O	1.70	-0.12	-0.51
	N	1.85	-0.2	-0.62
	H	0.224	-0.046	0.31
chloride	Cl	2.27	-0.15	-1.00
Sulphate	S	2.10	-0.47	2.40
	O	1.70	-0.12	-1.10

FIGURE S1.

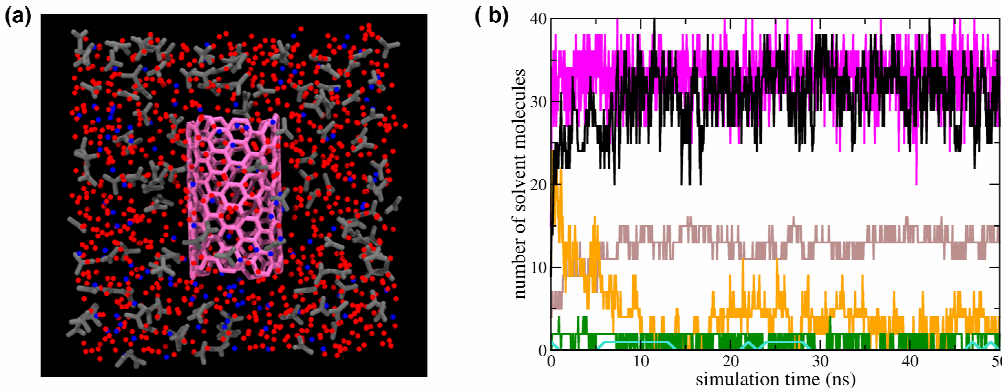

Figure S1. (a) The initial set up of a (9, 6) carbon nanotube solvated in 6M aqueous GdmCl solution. The carbon nanotube is shown in pink. The guanidinium molecules are shown in grey, water oxygens in red, chloride atoms in blue. (b) The number of cosolvent molecules in 6M GdmCl [Gdm^+: green, Cl-: cyan, water: black], in 8M urea [urea: brown, water: orange], and in pure water [water: pink] inside a 3.3 nm long (9, 6) nanotube core as a function of simulation time.

FIGURE S2

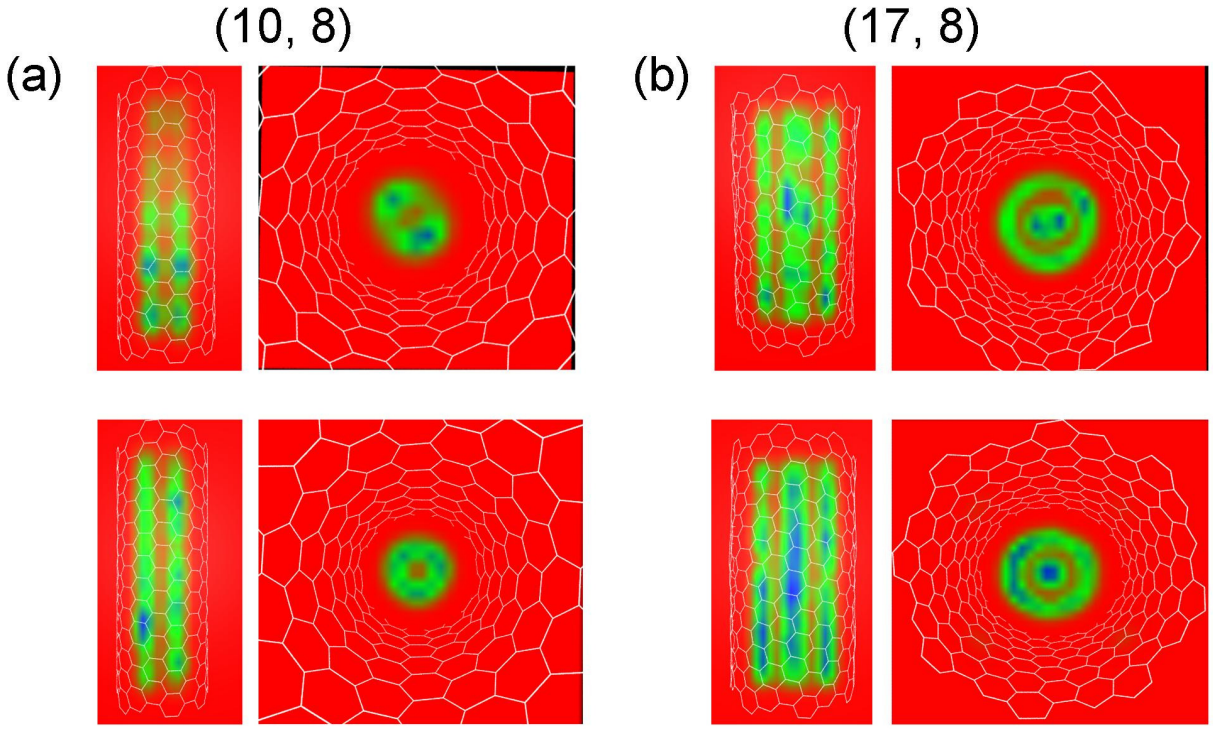

Figure S2. The axial and radial density profiles of guanidinium (top) and urea (bottom) for (10, 8) and (17, 8) CNT. The color scale for density is used such as from red (low) to green to blue (high). Existence of Gdm homo-ion dimers can be seen inside both (10, 8) and (17, 8) nanotubes.

FIGURE S3.

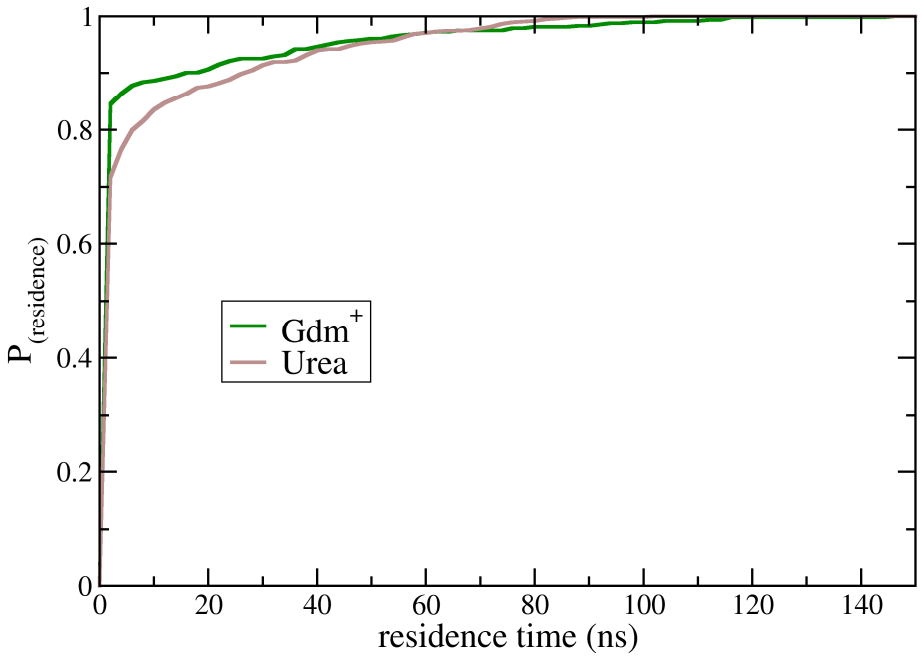

Figure S3. Cumulative distributions of the residence times $P_{(residence)}$ of Gdm^+ (green) and urea (brown) inside a (17, 8) CNT.

FIGURE S4.

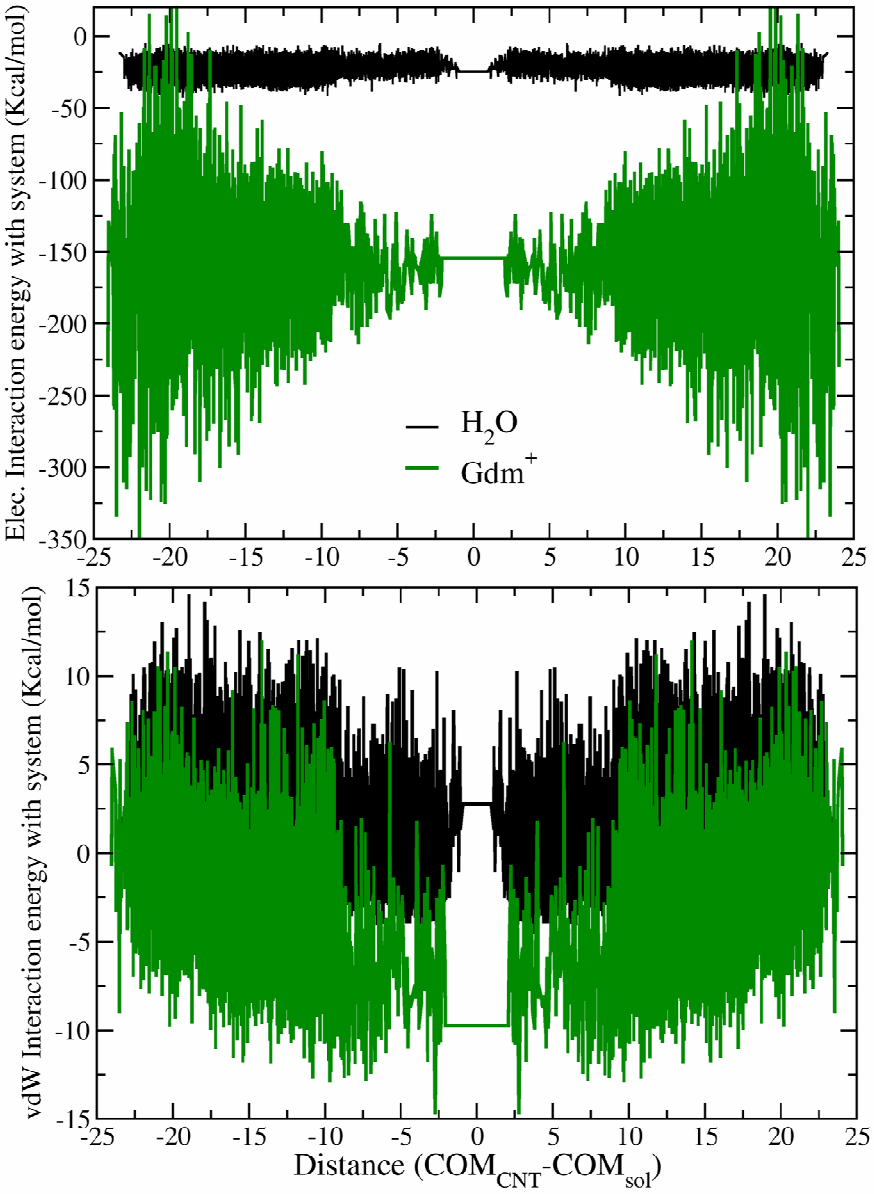

Figure S4. Interaction energies (in Kcal/mol) with rest of the system in 6M GdmCl for a water (black) or guanidinium (green) molecule as a function of its distance (in Å) from the center of mass (COM) of the (10, 8) nanotube. Top. The electrostatic interaction energy. Bottom. The van der Waals interaction energy.

FIGURE S5.

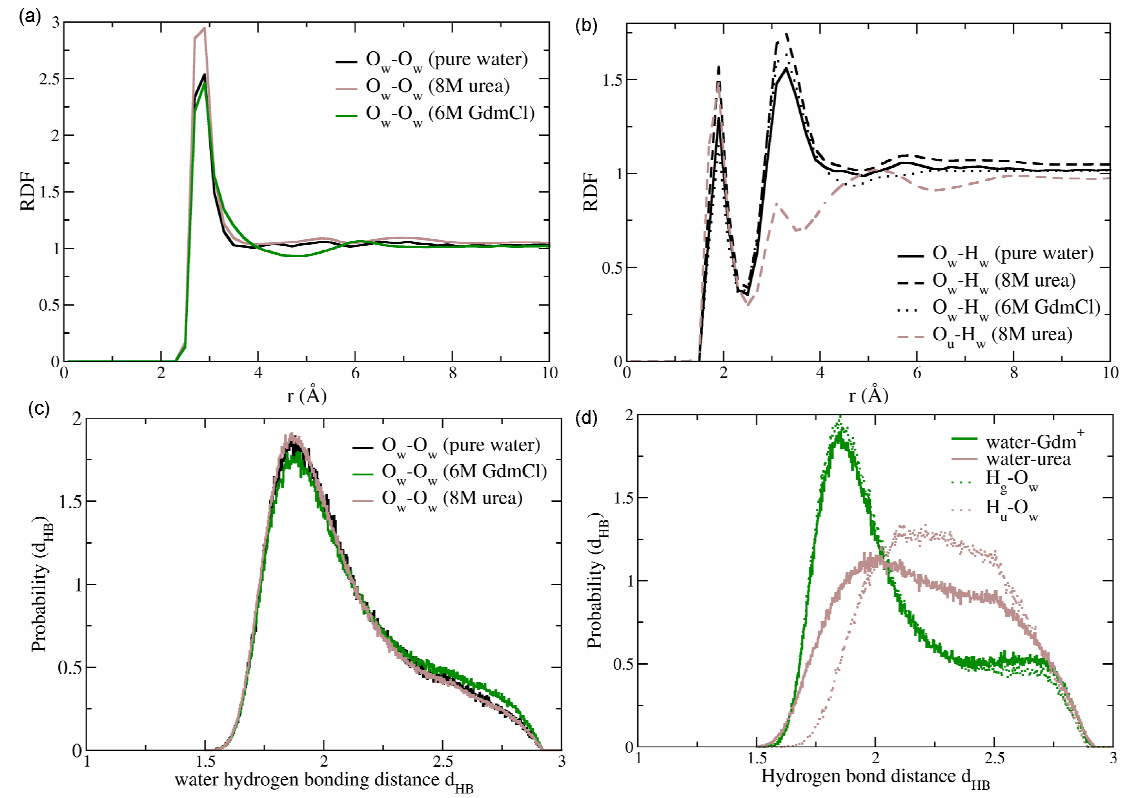

Figure S5. Comparison of water structure (a-b) and hydrogen bond distance distributions (c-d) in different cosolvent solutions and in pure water. The definition of hydrogen bonds is that the distance between donor and acceptor is no greater than 3.5 Å and the angle of donor–H–acceptor is no smaller than 120°.