Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information

Enhancing the Seawater Desalination Performance of Multilayer Reduced Graphene Oxide Membranes by Introducing in-Plane Nanopores: A Molecular Dynamics Simulation Study

Table S1. Lennard-Jones parameters and partial atomic charges of atoms

atom	σ (Å)	ε (kcal/mol)	charge (q)
O (H_2O)	3.5365	0.1521	-0.834
H (H_2O)	0.0000	0.0460	0.417
Na	2.5860	0.1046	+1
Cl	4.4020	0.1046	−1
CA (sp^2 carbon)	3.5500	0.0700	0
CT (C in epoxy)	3.5000	0.0659	0.14
OS (O in epoxy)	2.9000	0.1400	-0.28
CF (C bonded with phenolic hydroxyl)	3.5500	0.0700	0.15
OH (O in phenolic hydroxyl)	3.0700	0.1699	-0.585
HO (H in phenolic hydroxyl)	0.0000	0.0000	0.435
C (C in carboxyl)	3.7500	0.1049	0.52
OH2 (O of -OH in carboxyl)	3.0000	0.1699	-0.53
O_3 (=O in carboxyl)	2.9600	0.2100	-0.44
HO2 (H of -OH in carboxyl)	0.0000	0.0000	0.45

Figure S1. Initial configuration of the rGO, r1NPGO, r2NPGO, r4NPGO and r1NPGO-3nm monolayers (a), and side-view of the resulting three-layer membranes (b). The orange, red, and white spheres represent carbon atoms, oxygen atoms and hydrogen atoms, respectively.

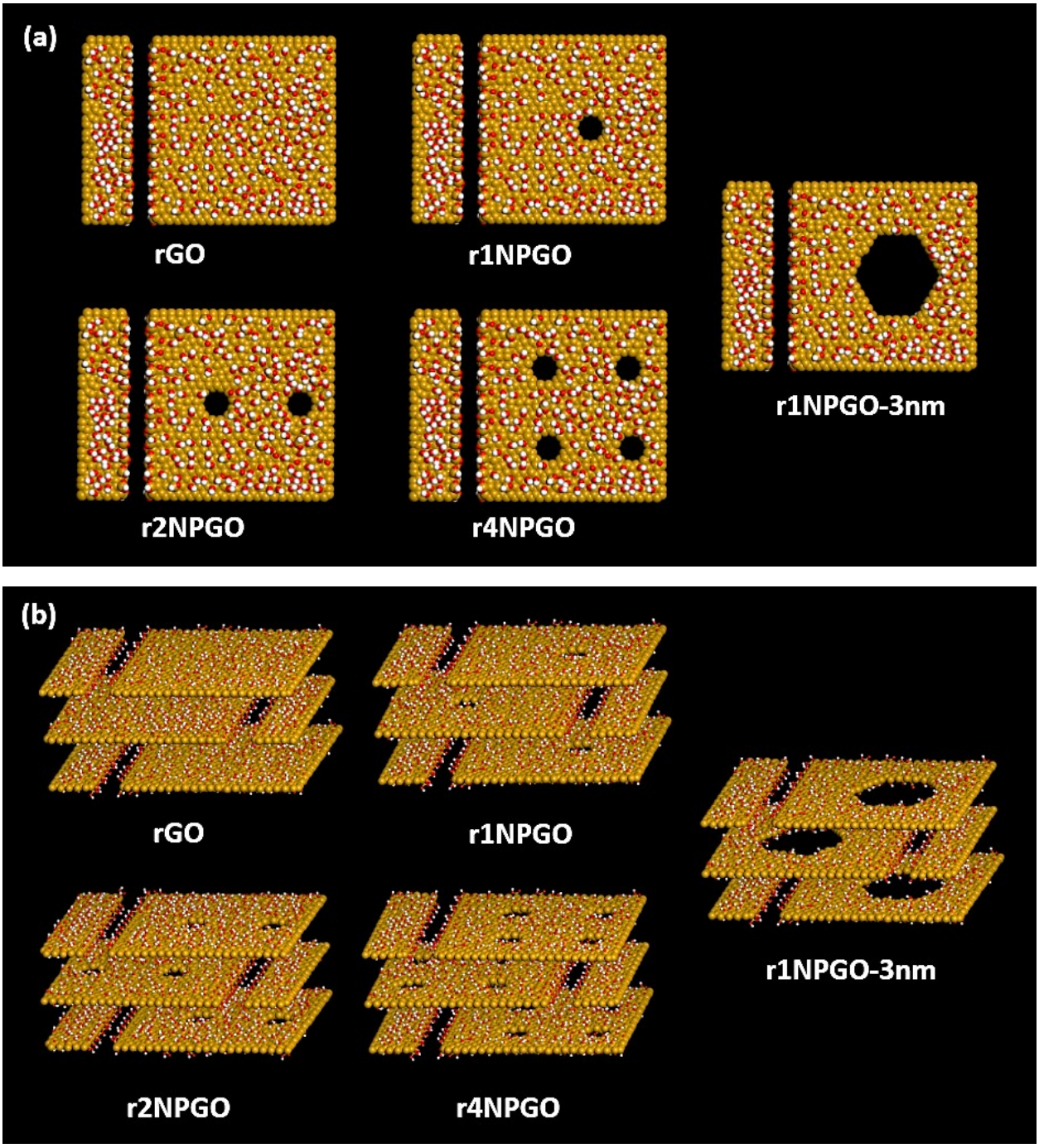


Figure S2. Number of water molecules in pure water box that are transferred from the feed side to permeate side as a function of time at 200 MPa for rGO, r1NPGO, r2NPGO, r4NPGO and r1NPGO-3nm membranes.

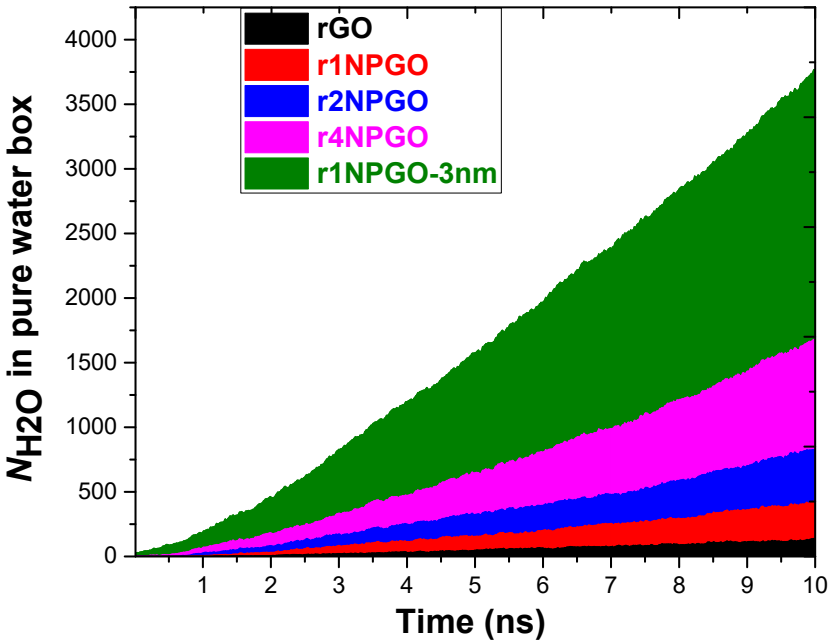


Figure S3. Two-dimensional density distribution of Na^+ ions inside (a) rGO, (b) r1NPGO, (c) r2NPGO, (d) r4NPGO, and (e) r1NPGO-3nm membranes at 10 ns. The white dashed lines show the approximate range of the three-layer membranes.

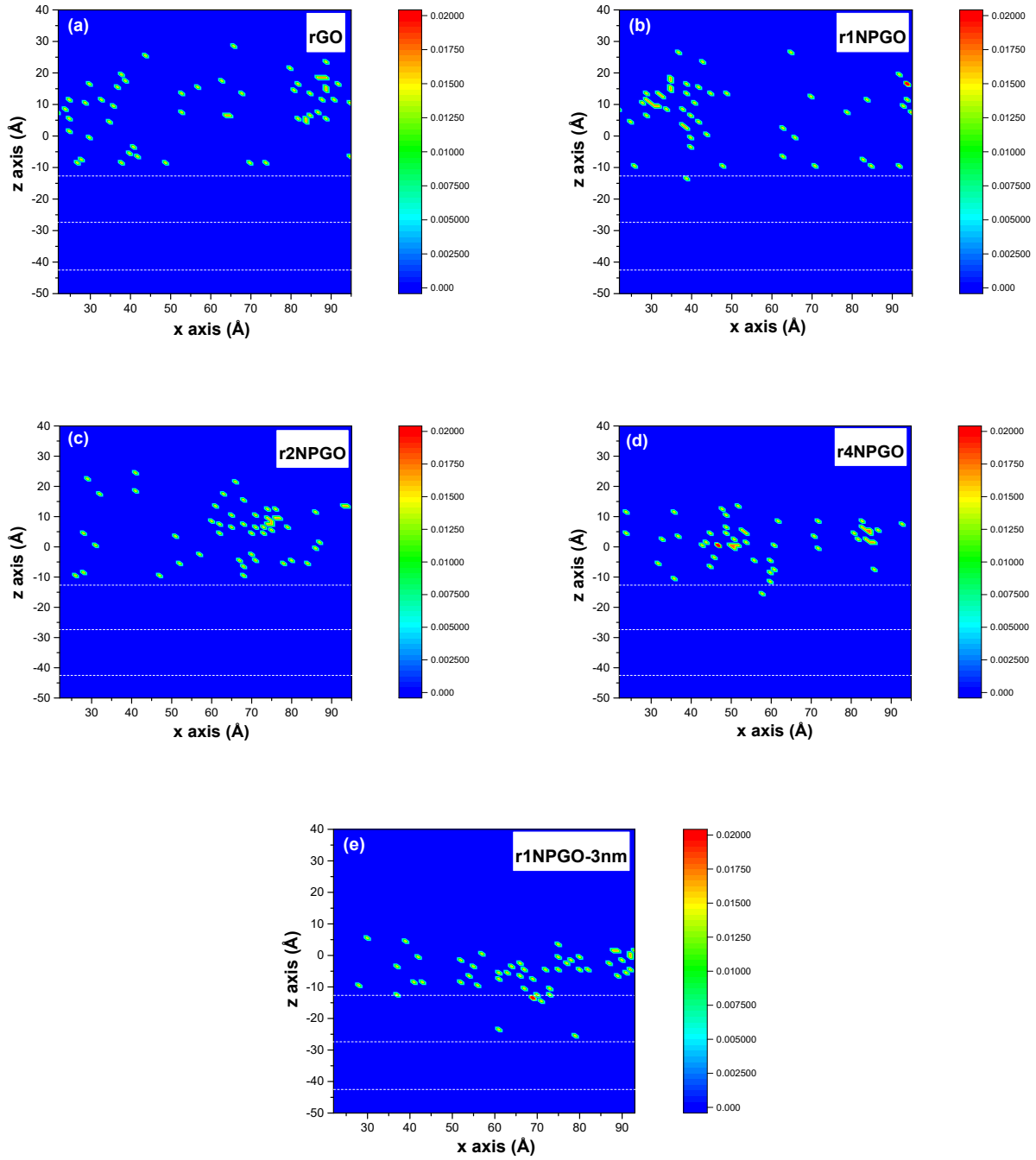


Figure S4. Two-dimensional density distribution of Cl^- ions inside (a) rGO, (b) r1NPGO, (c) r2NPGO, (d) r4NPGO, and (e) r1NPGO-3nm membranes at 10 ns. The white dashed lines show the approximate range of the three-layer membranes.

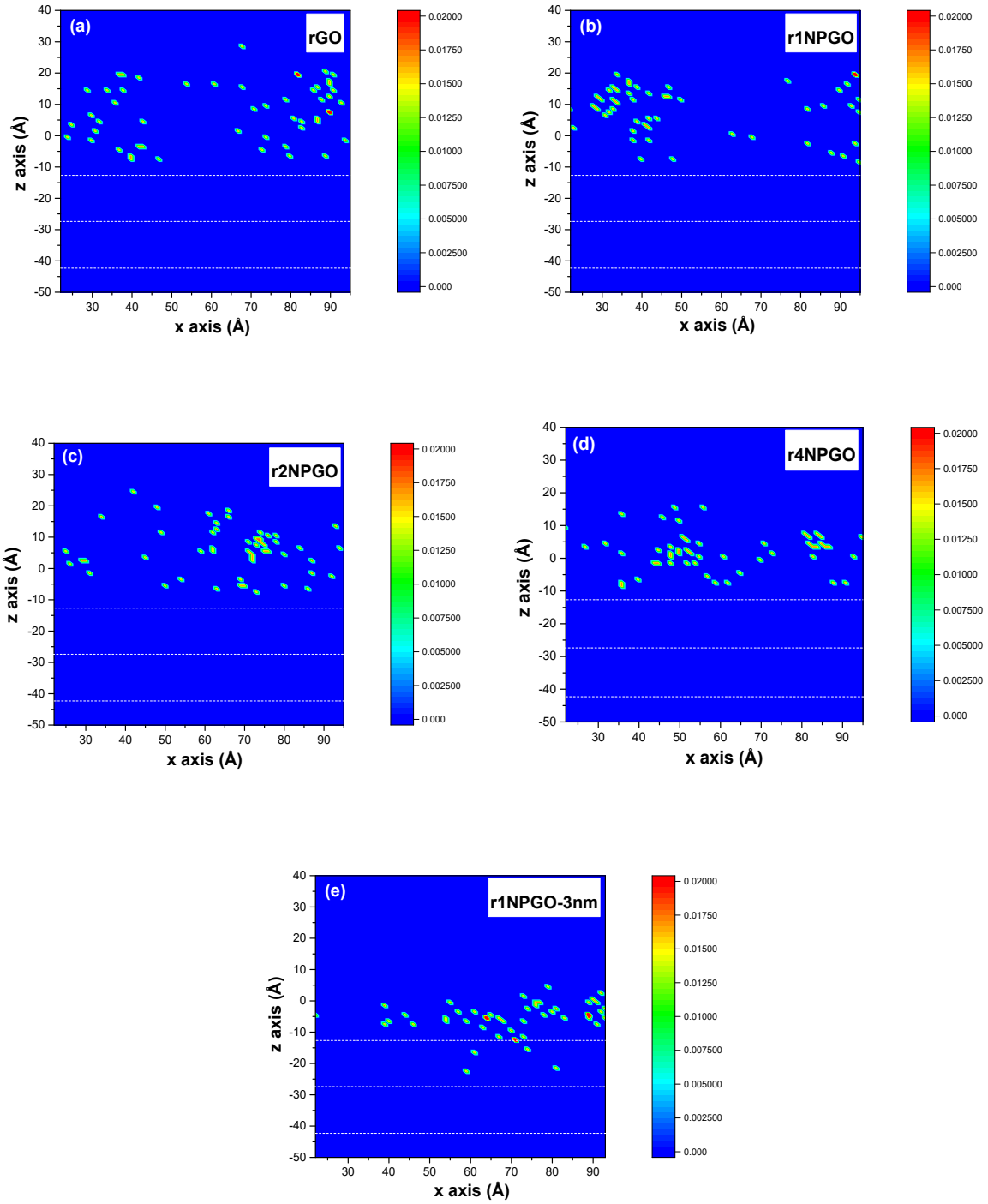
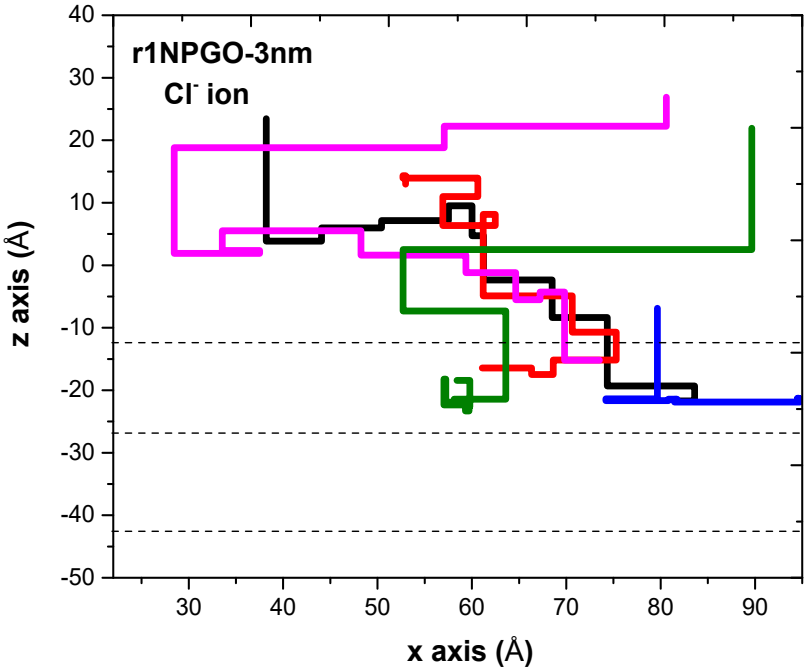


Figure S5. The trajectory of $5Cl^-$ and $3Na^+$ ions over the entire simulation duration for r1NPGO-3nm membrane.



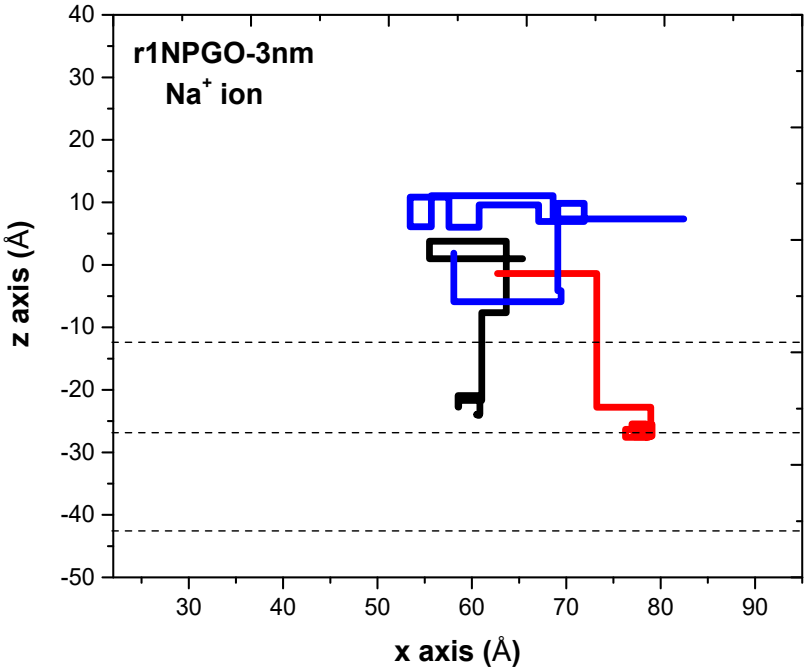
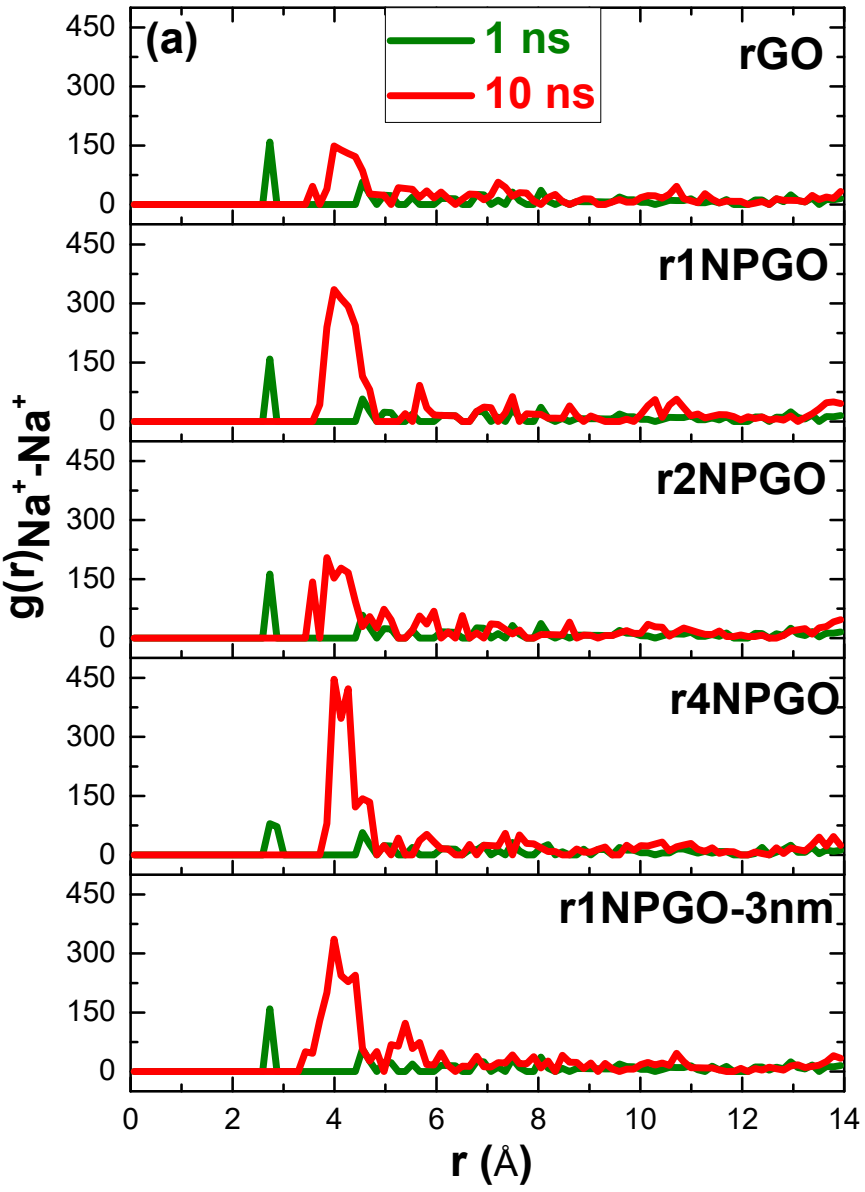
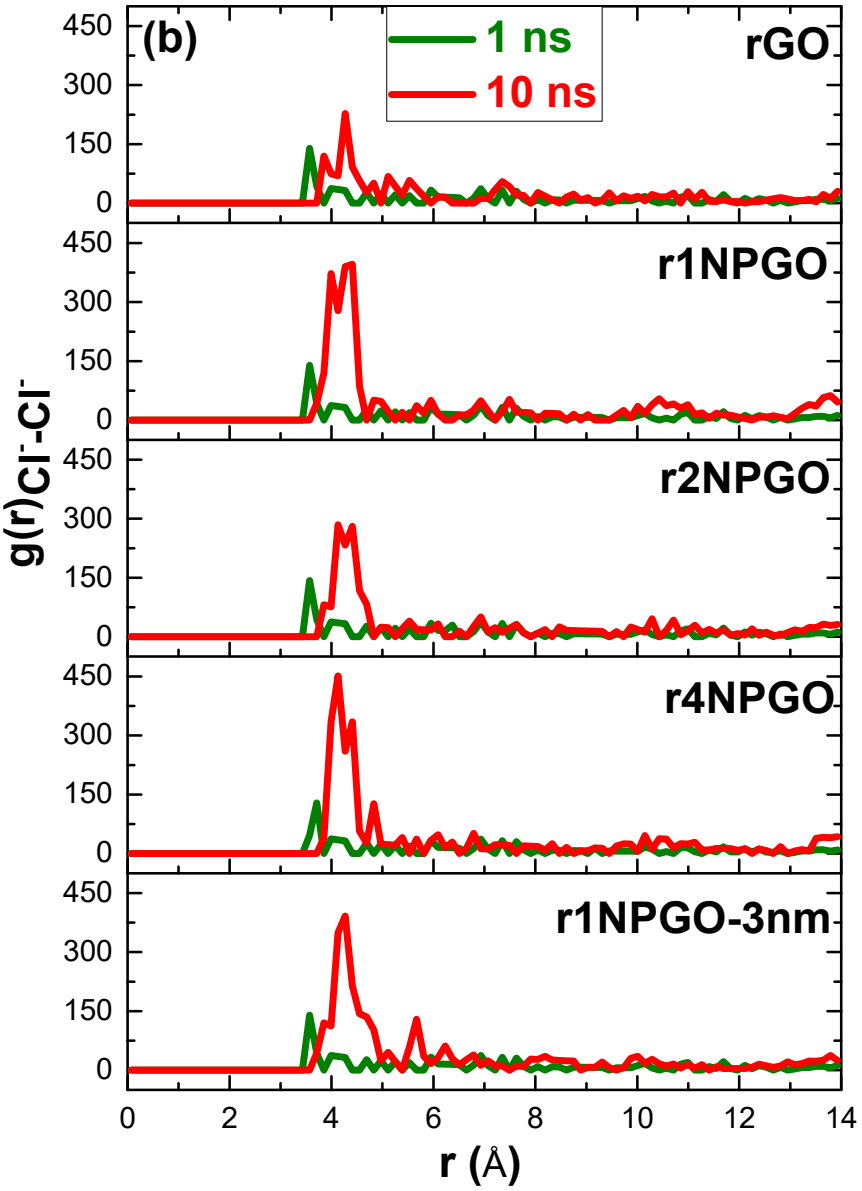

r1NPGO-3nm
Na+ ion
z axis (Å)
x axis (Å)
40
30
20
10
0
-10
-20
-30
-40
-50
30
40
50
60
70
80
90

Figure S6. Radial distribution functions of Na^+-Na^+, Cl^--Cl^-, and Na^+-Cl^- for rGO, r1NPGO, r2NPGO, r4NPGO and r1NPGO-3nm membranes at 1 ns and 10 ns.




(b)
1 ns
10 ns
rGO
r1NPGO
r2NPGO
r4NPGO
r1NPGO-3nm
g(r)Cl-Cl-
r (Å)
0
150
300
450
0
2
4
6
8
10
12
14

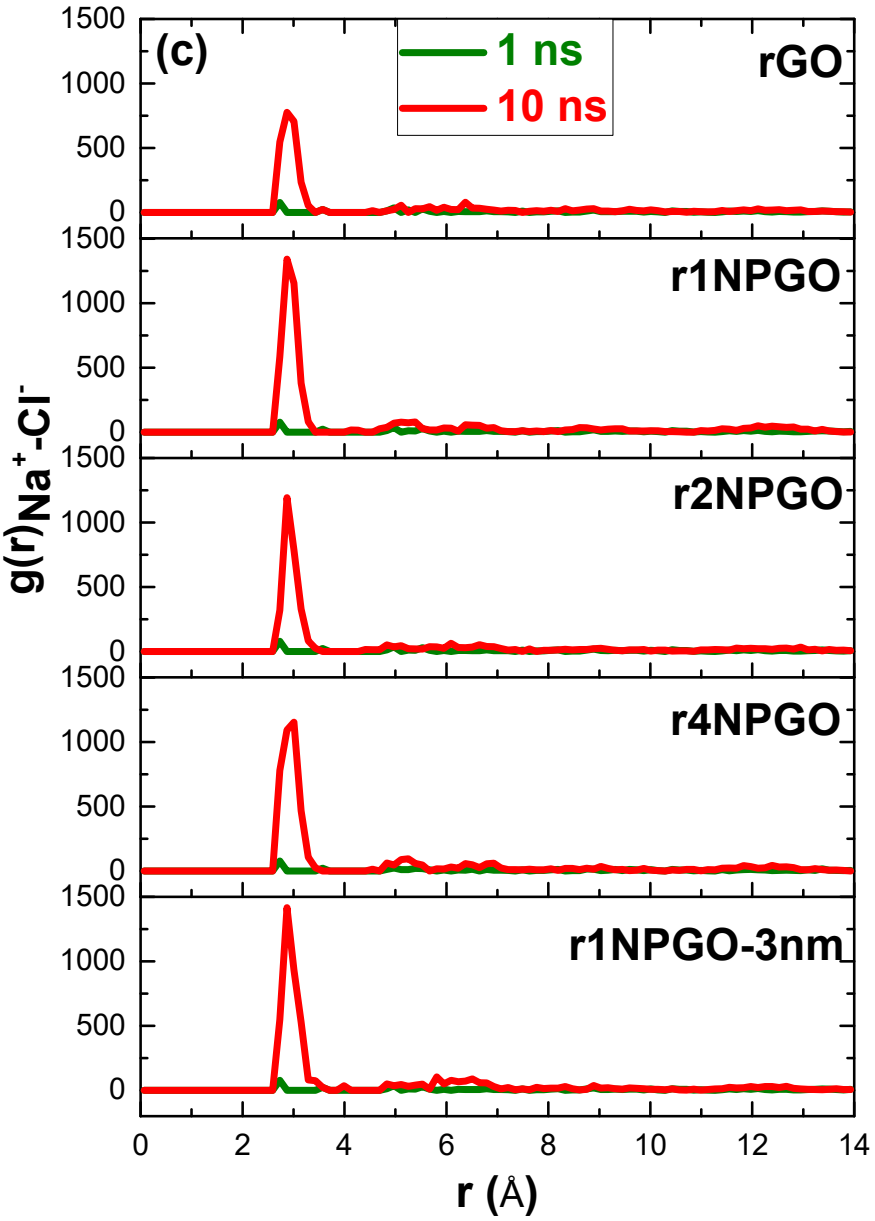

(c)
1 ns
10 ns
rGO
r1NPGO
r2NPGO
r4NPGO
r1NPGO-3nm
g(r)Na+-Cl-
r (Å)
0
500
1000
1500
0
2
4
6
8
10
12
14

Figure S7. Formation of ion clusters from Na^+ and Cl^- ions in (a) rGO, (b) r1NPGO, (c) r2NPGO, (d) r4NPGO, and (e) r1NPGO-3nm at 10 ns. The orange, red, white, purple, and lime spheres represent carbon atoms, oxygen atoms, hydrogen atoms, sodium ions, and chlorine ions, respectively.

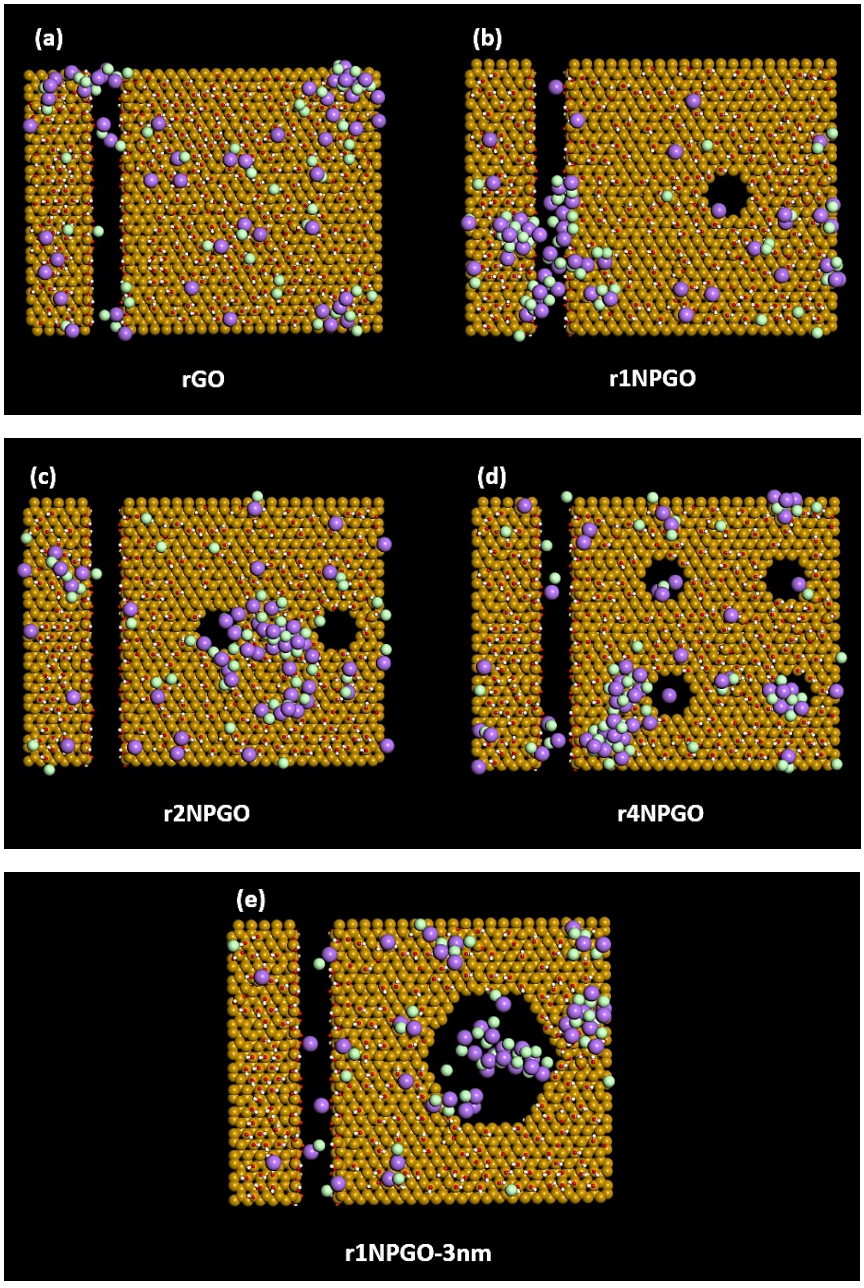
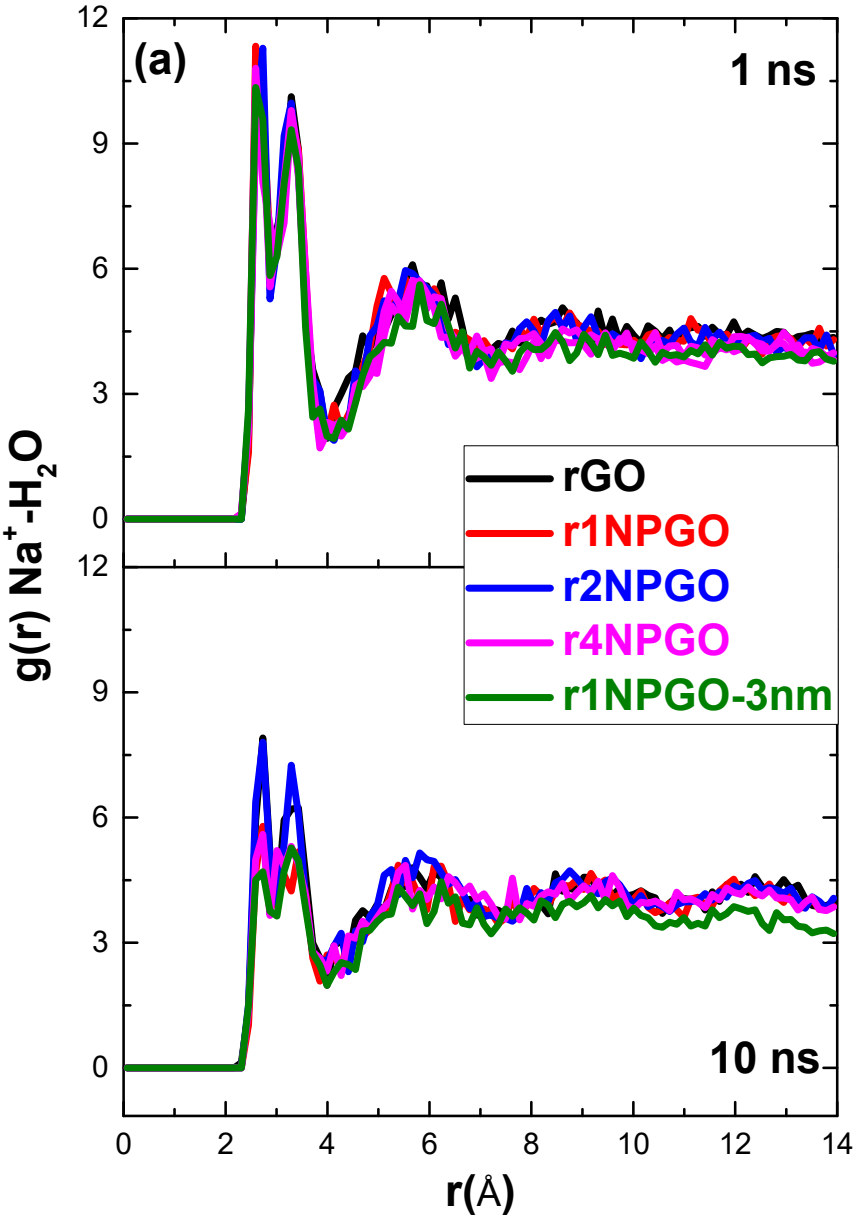


Figure S8. Radial distribution functions of Na^+-H_2O, and Cl^--H_2O for rGO, r1NPGO, r2NPGO, r4NPGO and r1NPGO-3nm membranes at 1 ns and 10 ns.



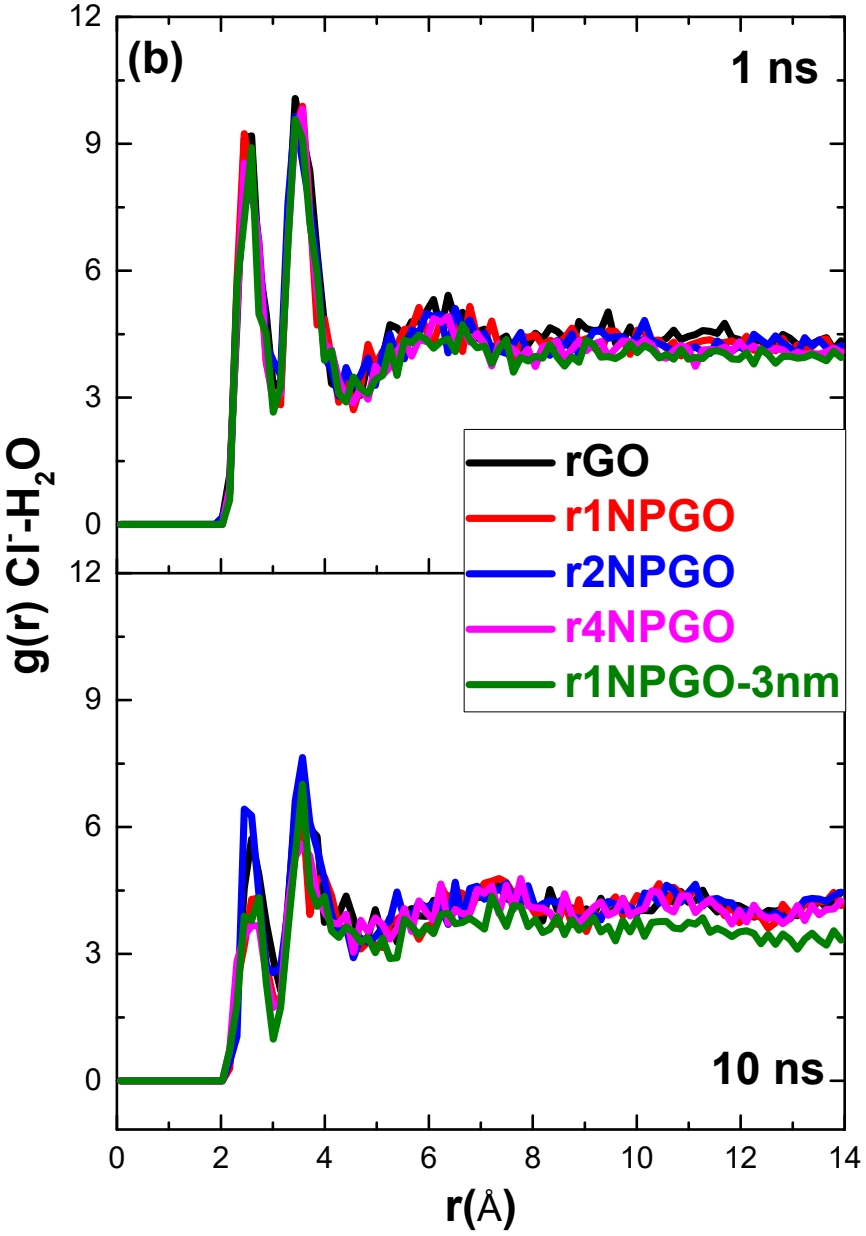
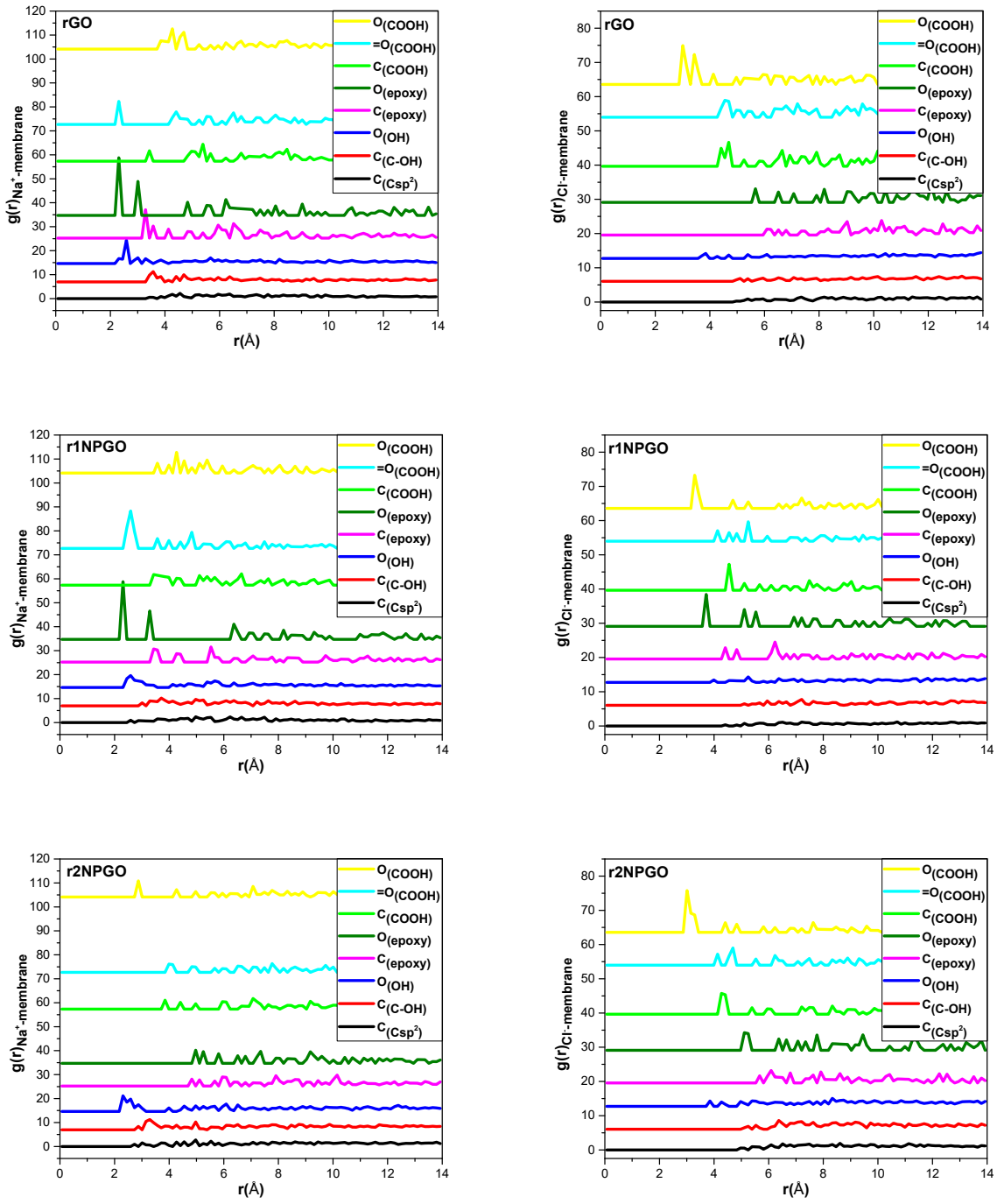
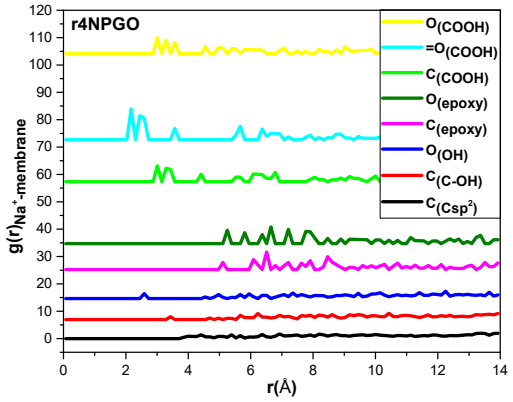
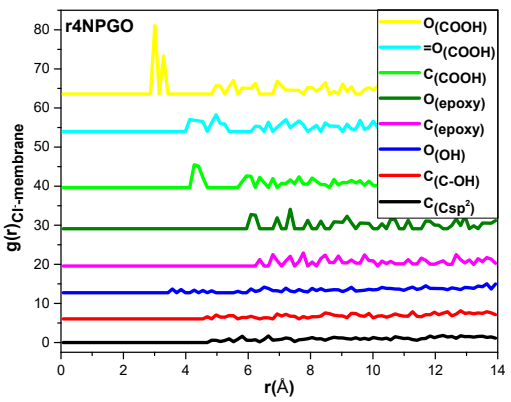
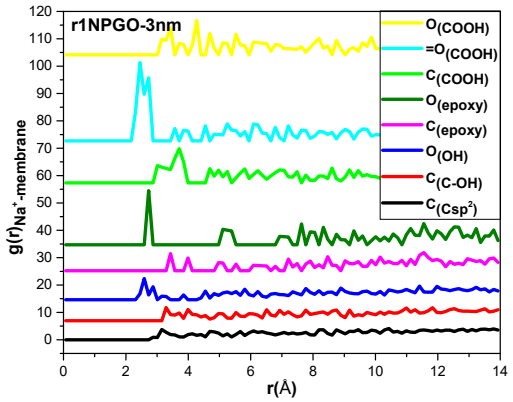

(b)
1 ns
10 ns
rGO
r1NPGO
r2NPGO
r4NPGO
r1NPGO-3nm
g(r) Cl-H2O
r(Å)
0
3
6
9
12
0
2
4
6
8
10
12
14

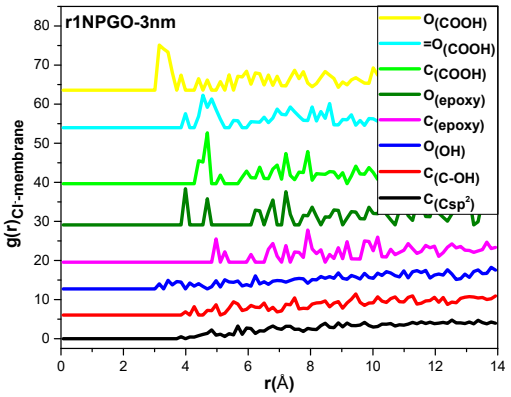
Figure S9. Radial distribution functions between Na^+ and Cl^- ions and membrane atoms for rGO, r1NPGO, r2NPGO, r4NPGO and r1NPGO-3nm membranes at 10 ns.




r4NPGO
O(COOH)
=O(COOH)
C(COOH)
O(epoxy)
C(epoxy)
O(OH)
C(C-OH)
C(Csp2)
g(r)Na+-membrane
r(Å)


r4NPGO
O(COOH)
=O(COOH)
C(COOH)
O(epoxy)
C(epoxy)
O(OH)
C(C-OH)
C(Csp2)
g(r)Cl-membrane
r(Å)


r1NPGO-3nm
O(COOH)
=O(COOH)
C(COOH)
O(epoxy)
C(epoxy)
O(OH)
C(C-OH)
C(Csp2)
g(r)Na+-membrane
r(Å)


r1NPGO-3nm
O(COOH)
=O(COOH)
C(COOH)
O(epoxy)
C(epoxy)
O(OH)
C(C-OH)
C(Csp2)
g(r)Cl-membrane
r(Å)