Supplementary Material

On the effects of induced polarizability at the water-graphene interface via classical charge-on-spring models

Yerko Escalona,[1] Nicolas Espinoza,[2,3] Mateo Barria-Urenda,[2,3] Chris Oostenbrink,[1] and Jose Antonio Garate[2,3,a)]

[1)] *Institute for Molecular Modeling and Simulation, University of Natural Resources and Life Sciences, 1190 Vienna, Austria*

[2)] *Centro Interdisciplinario de Neurociencia de Valparaíso (CINV), Universidad de Valparaíso, Valparaíso, Chile*

[3)] *Millennium Nucleus in NanoBioPhysics (NNBP), Universidad de Valparaíso, Pasaje Harrington 287, Valparaíso, Chile.*

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[a)] Author to whom correspondence should be addressed.

email: jose.garate@uv.cl

TABLE S1. Polarizability and charge properties of the models employed

	δ	$\alpha\,(10^{-3}\mathrm{nm}^3)$	$\mathrm{Q_{pol}}$
$\mathrm{O_{COS/G2\text{-}D2}}$	-1.0530	1.1	$\pm$ 8
$\mathrm{H_{COS/G2\text{-}D2}}$	0.5265	0	0
$\mathrm{O_{SPC}}$	-0.8200	0	0
$\mathrm{H_{SPC}}$	0.4100	0	0
$\mathrm{C_{CCCP}}$	0	1.1	$\pm$ 8
$\mathrm{C_{CCC}}$	0	0	0

TABLE S2. Lennard-Jones potential parameters

	$\varepsilon\,(kJ/mol)$	$\sigma\,(nm)$
$\mathrm{C\text{-}O_{SPC}}$	0.425	0.337
$\mathrm{C\text{-}O_{COS/G2}}$	0.460	0.338
$\mathrm{C\text{-}O_{COS/D2}}$	0.474	0.337

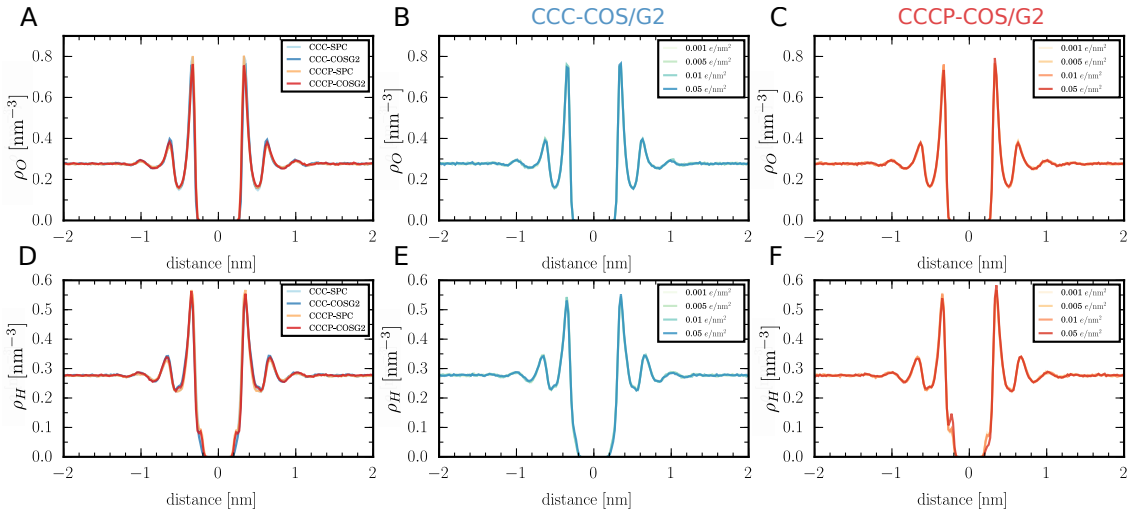

FIG. S1. Water oxygen (ρ_O) and hydrogen (ρ_H) density for hybrid systems. In A and D, ρ_O and ρ_H profiles, respectively for the CCC-SPC, CCC-COSG2, CCCP-SPC and CCCP-COSG2 systems. B and E present ρ_O and ρ_H under the effects of external electric fields for the CCC-COS/G2. B and E present ρ_O and ρ_H under the effects of external electric fields for the CCCP-COS/G2.

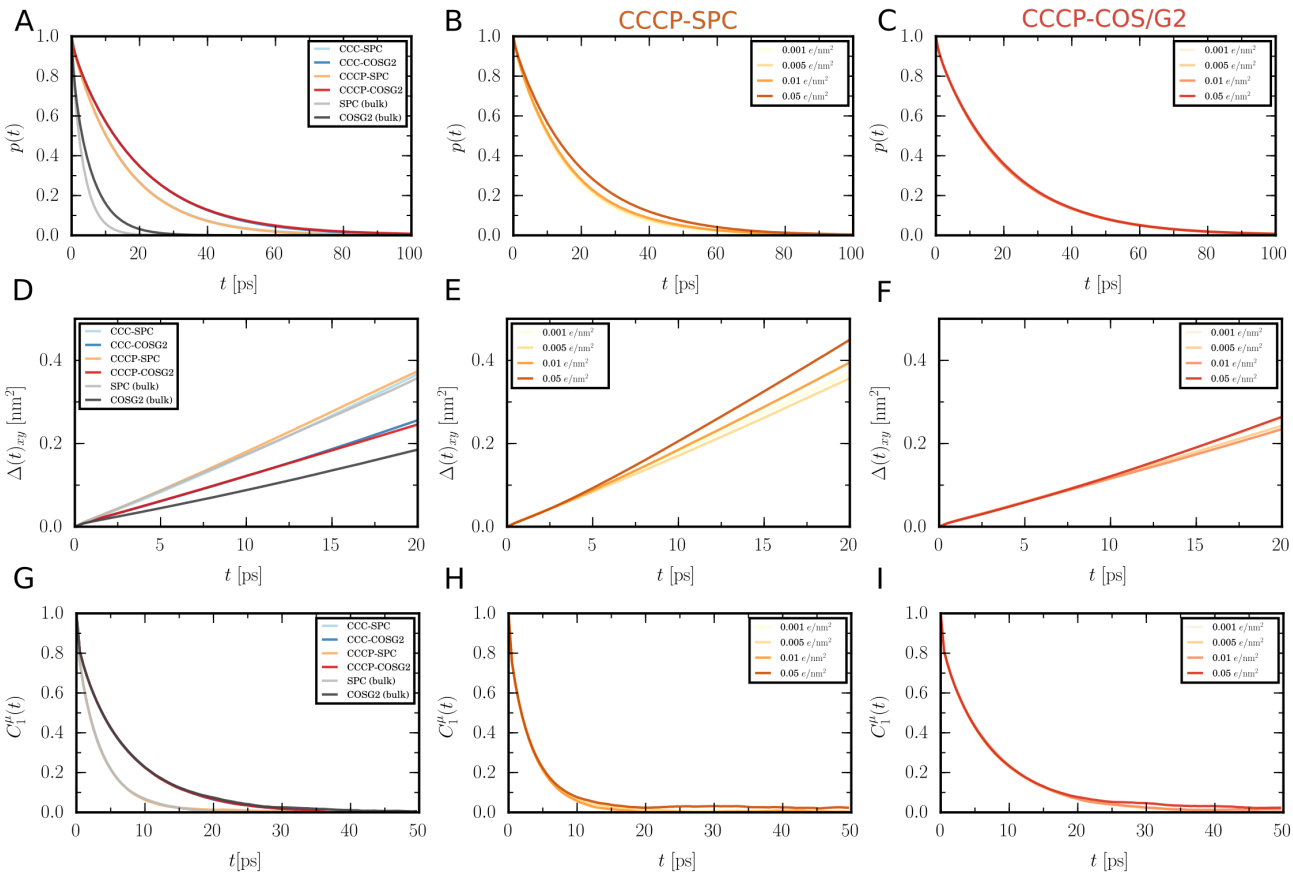

FIG. S2. Water dynamics of interfaced waters. **A** shows the survival probability of waters in the interface with graphene for the systems: CCC-SPC (light blue line), CCC-COSG2 (blue line), CCCP-SPC (light red) and CCCP-COSG2 (red line); and for the SPC (grey line) and COS/G2 (black line) waters in the bulk, which correspond to the outer layer of water with respect to the graphene. **B** and **C** correspond to the survival probability of waters in the interface with graphene at different electric field strenght condition (0.001, 0.005, 0.01 and 0.05 e/nm^2) for the systems CCCP-SPC and CCCP-COSG2, respectively. **D** shows the mean squared displacement of interfacial water in the xy axis for the different systems and the bulk. **E** and **F** shows the same but with different electric field strengths for the CCCP-SPC and CCCP-COSG2 systems, respectively. **G** shows the rotational relaxation curve of the dipole vector in the interfaced waters with graphene for the different systems and for waters in the bulk region. **H** and **I** shows the same in different electric field strength condition for the CCCP-SPC and CCCP-COSG2 systems, respectively.

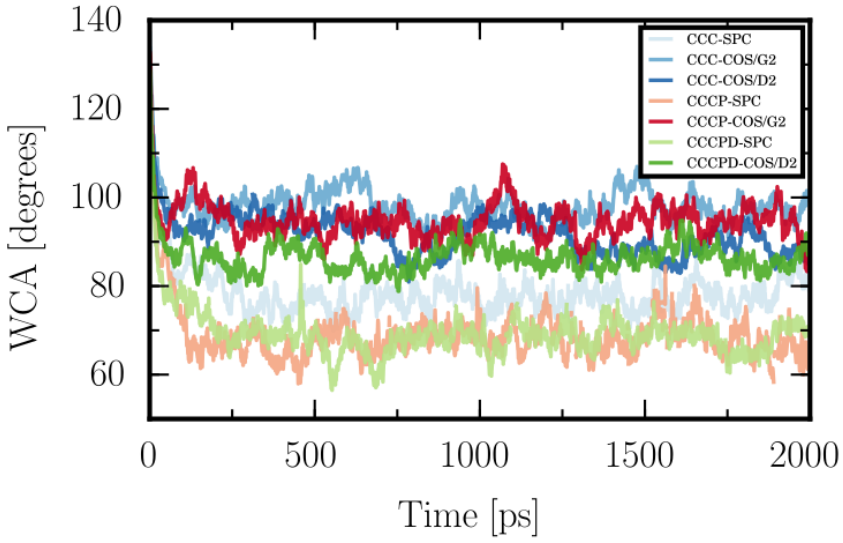

FIG. S3. Time series of Water contact angle (WCA) for combinations of polarizable and fixed charged models: CCC-SPC (light blue), CCC-COS/G2 (blue), CCC-COS/D2 (dark blue), CCCP-SPC (light red), CCCP-COS/G2 (red), CCCPD-SPC (light green) and CCCPD-COS/D2 (green)

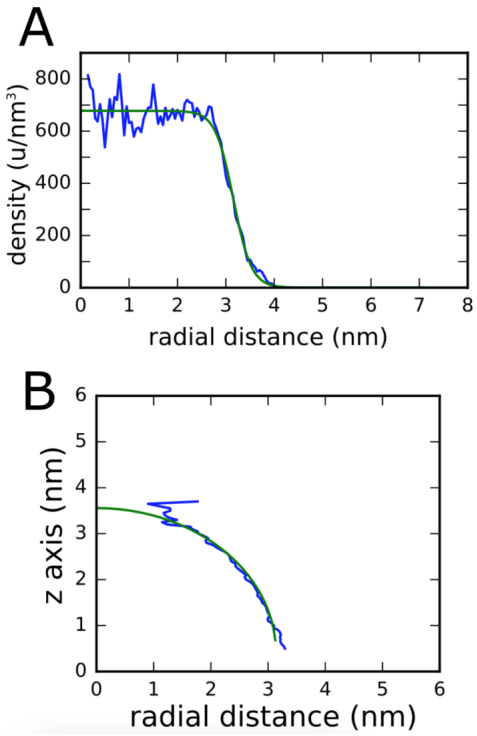

FIG. S4. Representative plots employed to compute WCA. In A, number densities as function of the radial distance along the x-y plane, for the first bin along the z axis. In B, radial distance along the x-y plane as function of the z axis. In blue and green computed data and non-lineal fits, respectively.

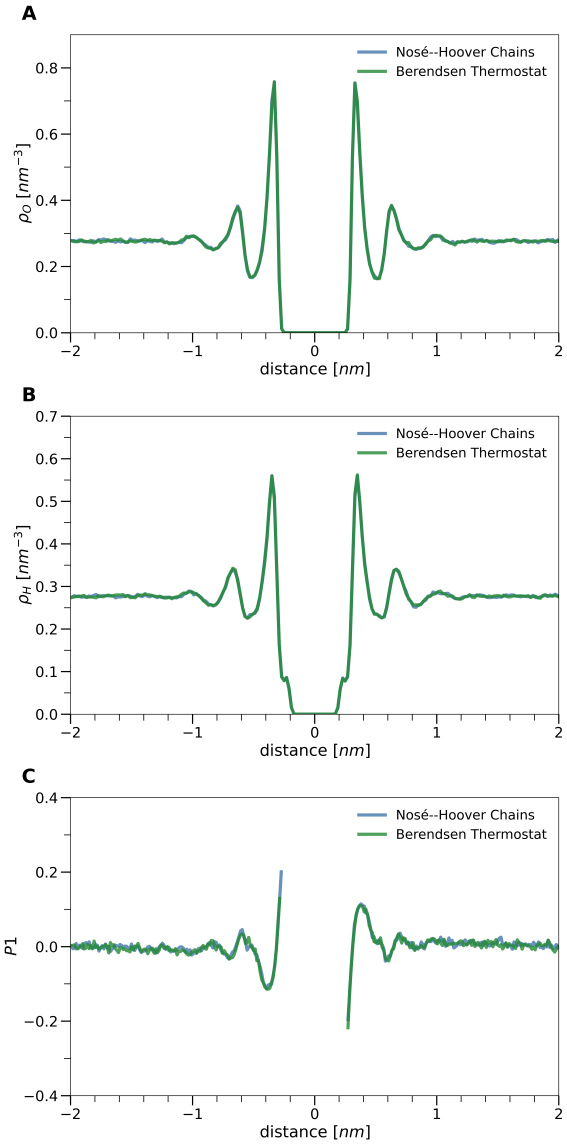

FIG. S5. Water oxygen (ρ_O) and hydrogen (ρ_H) density and dipolar orientation (P1) profiles along the graphene normal for the CCCP-COSG2 system employing different thermostats. In A, B and C, ρ_O, ρ_H and P1 profiles, respectively.

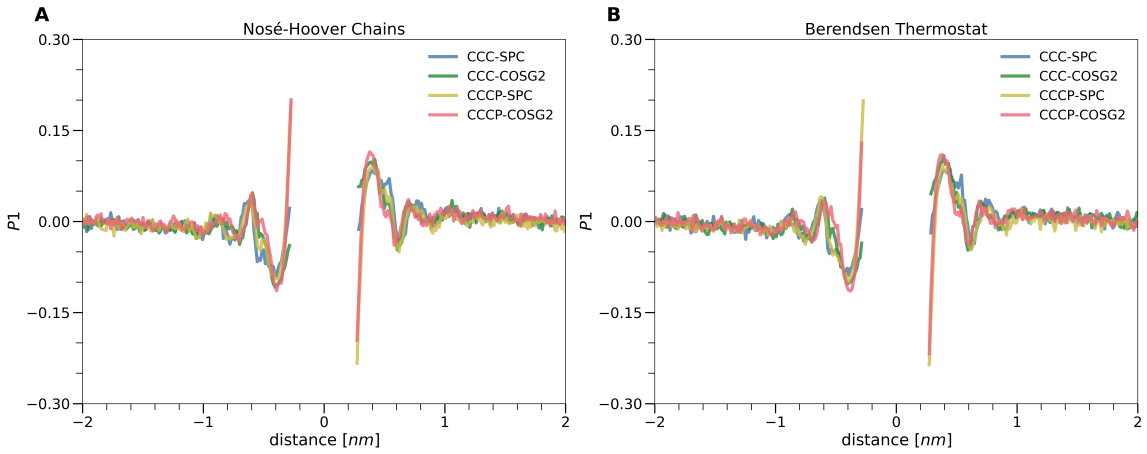

FIG. S6. Water dipolar alignment along the graphene normal for simulated systems; A, P1 values employing the Nosé-Hoover thermostat; B, P1 values employing the Berendsen thermostat.

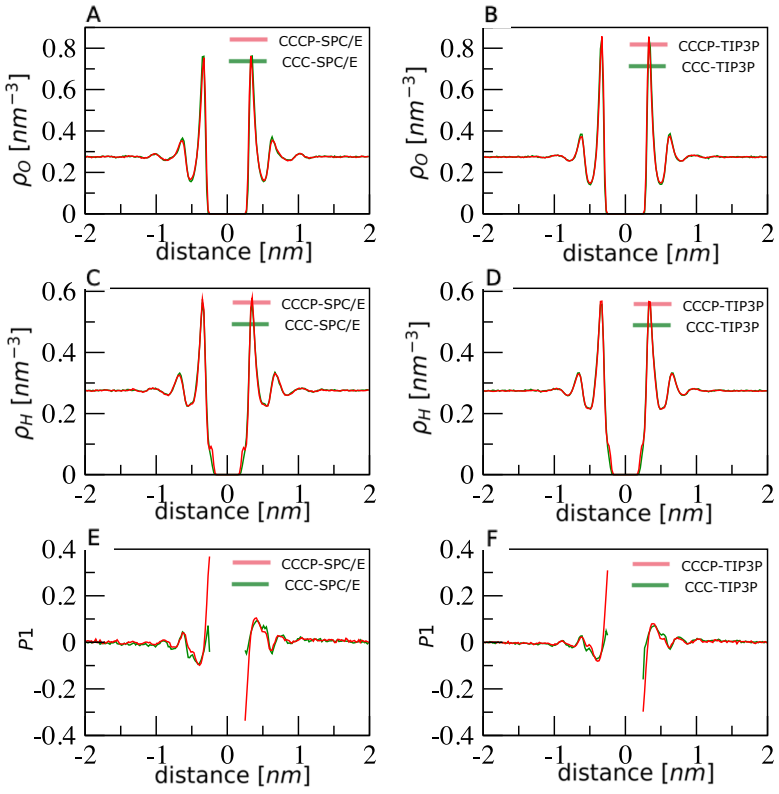

FIG. S7. Water oxygen (ρ_O) and hydrogen (ρ_H) density and dipolar orientation (P1) profiles along the graphene normal for the CCC-SPC/E, CCCP-SCP/E, CCC-TIP3P and CCCP-TIP3P systems. In A,C and E, ρ_O, ρ_H and P1 profiles for the SCP/E model, respectively. In B, D and F, ρ_O, ρ_H and P1 profiles for the TIP3P model, respectively.