Electronic Supplementary Information

Wettability of Graphene Oxide Functionalized with N-Alkylamines: A Molecular Dynamics Study

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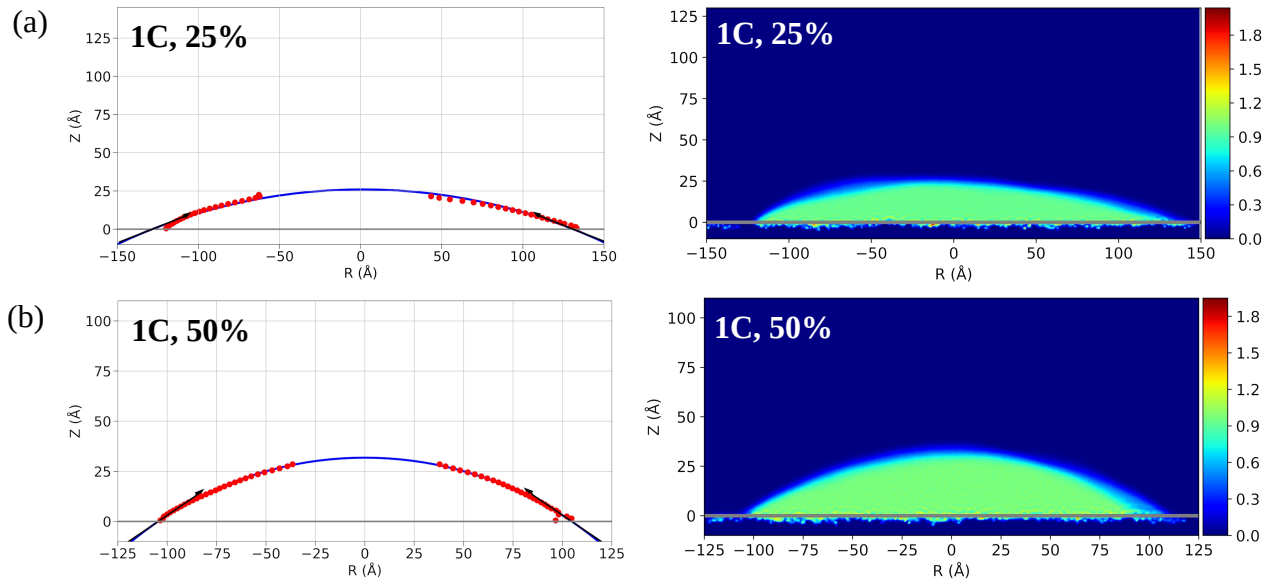


Figure S1. Water liquid boundary (red points), fitted circle (blue line), surface plane (gray line) and tangent line to the points of contact of the three phases (black line) for graphene oxide functionalized with N-methylamine (1C) for (a) 25% and (b) 50% of substitution degree (left). Selected surface planes are shown on the mass density contour maps .(right)

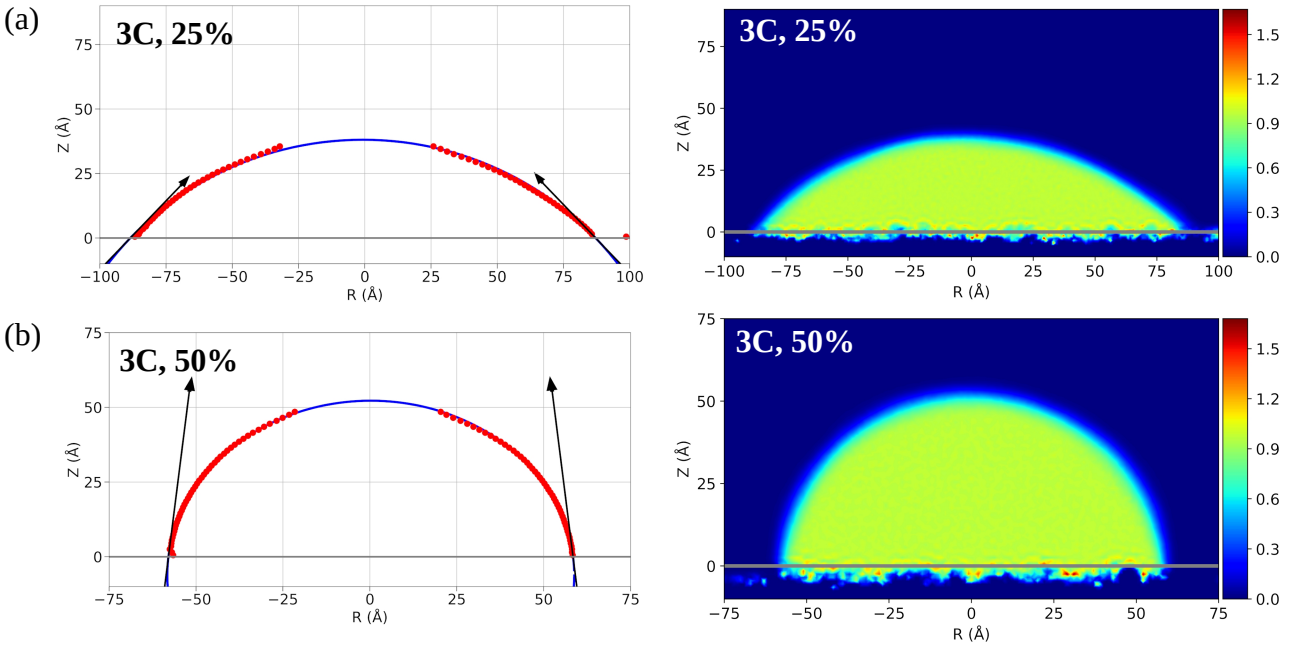


Figure S2. Water liquid boundary (red points), fitted circle (blue line), surface plane (gray line) and tangent line to the points of contact of the three phases (black line) for graphene oxide functionalized with N-propylamine (3C) for (a) 25% and (b) 50% of substitution degree (left). Selected surface planes are shown on the mass density contour maps (right).

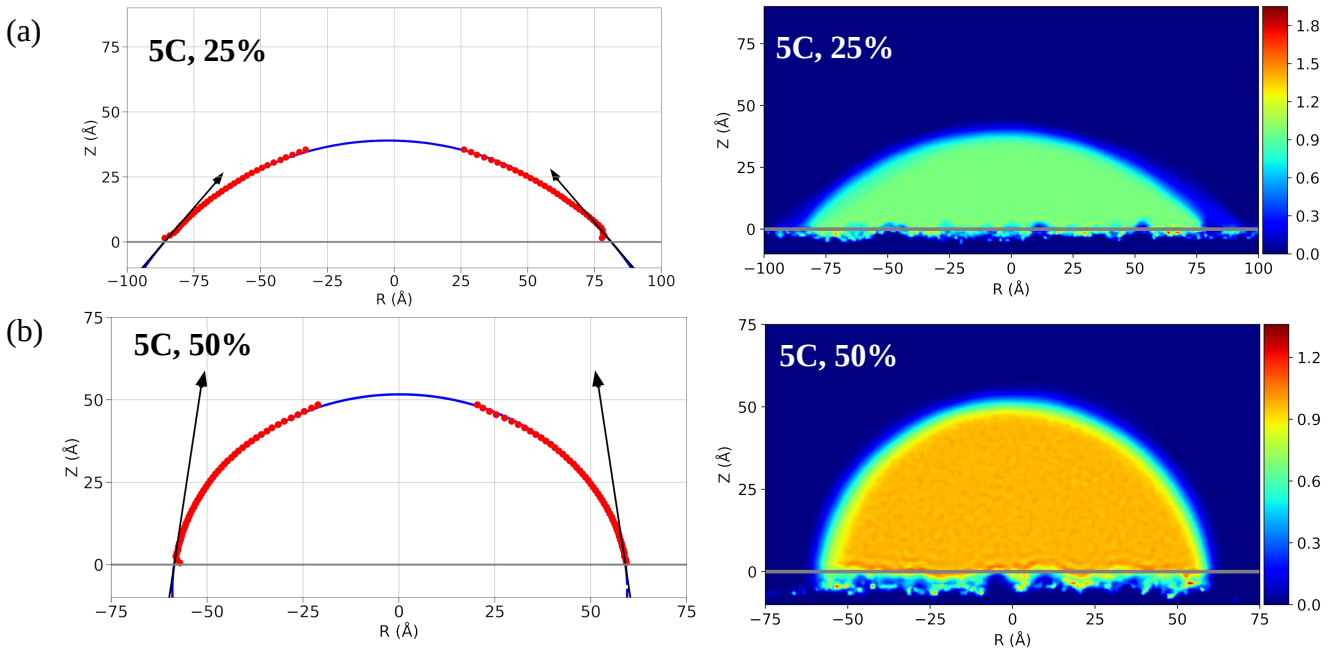


Figure S3. Water liquid boundary (red points), fitted circle (blue line), surface plane (gray line) and tangent line to the points of contact of the three phases (black line) for graphene oxide functionalized with N-pentylamine (5C) for (a) 25% and (b) 50% of substitution degree (left). Selected surface planes are shown on the mass density contour maps (right).

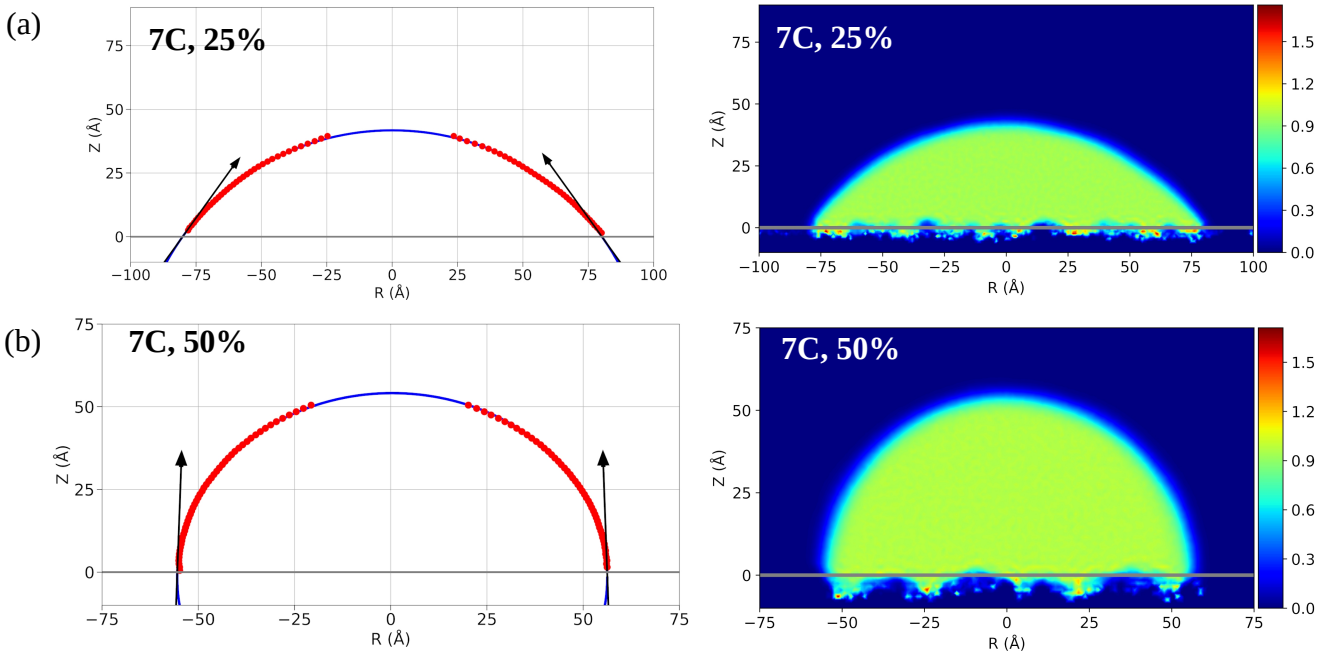


Figure S4. Water liquid boundary (red points), fitted circle (blue line), surface plane (gray line) and tangent line to the points of contact of the three phases (black line) for graphene oxide functionalized with N-heptylamine (7C) for (a) 25% and (b) 50% of substitution degree (left). Selected surface planes are shown on the mass density contour maps (right).

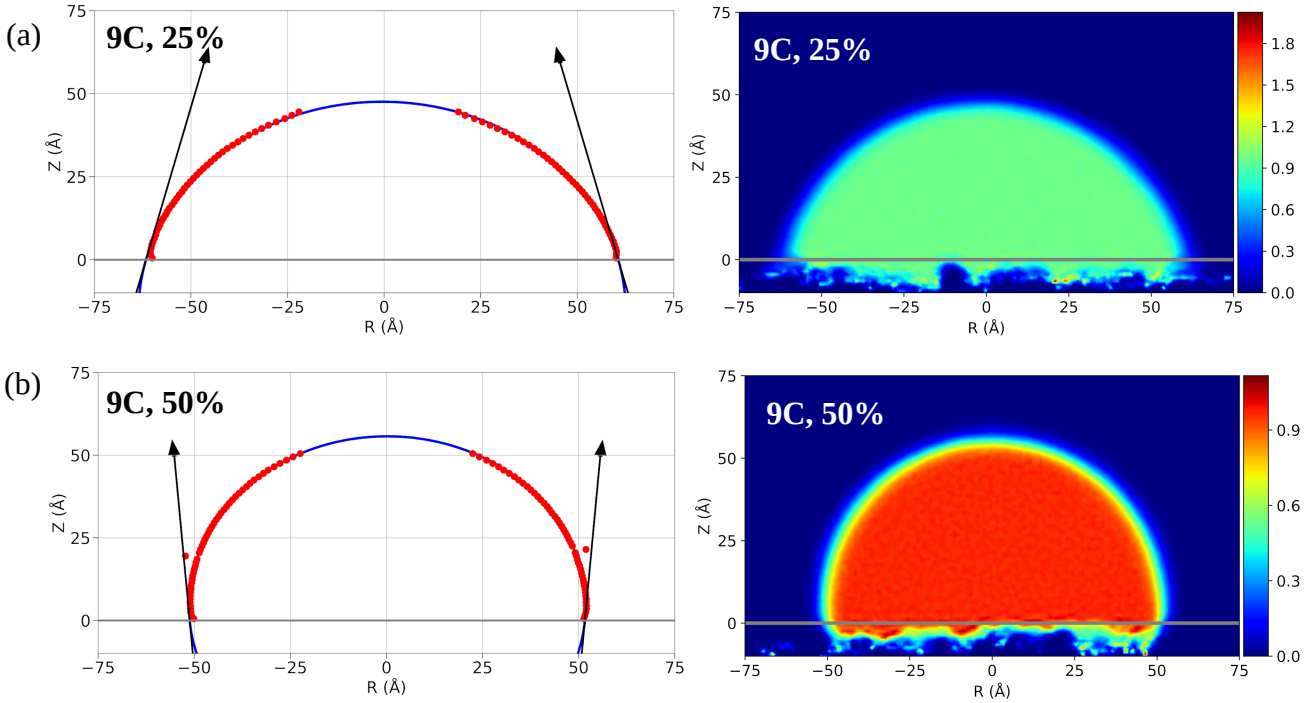


Figure S5. Water liquid boundary (red points), fitted circle (blue line), surface plane (gray line) and tangent line to the points of contact of the three phases (black line) for graphene oxide functionalized with N-nonylamine (9C) for (a) 25% and (b) 50% of substitution degree (left). Selected surface planes are shown on the mass density contour maps (right).

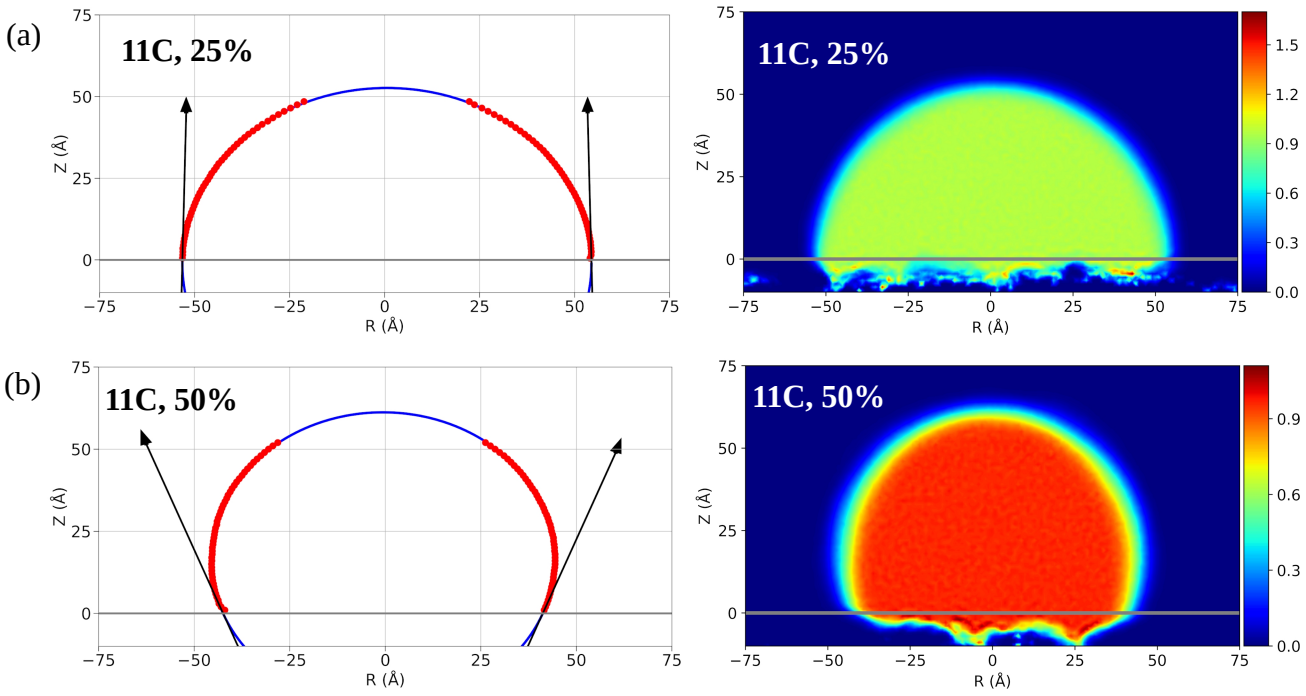


Figure S6. Water liquid boundary (red points), fitted circle (blue line), surface plane (gray line) and tangent line to the points of contact of the three phases (black line) for graphene oxide functionalized with N-undecylamine (11C) for (a) 25% and (b) 50% of substitution degree (left). Selected surface planes are shown on the mass density contour maps (right).

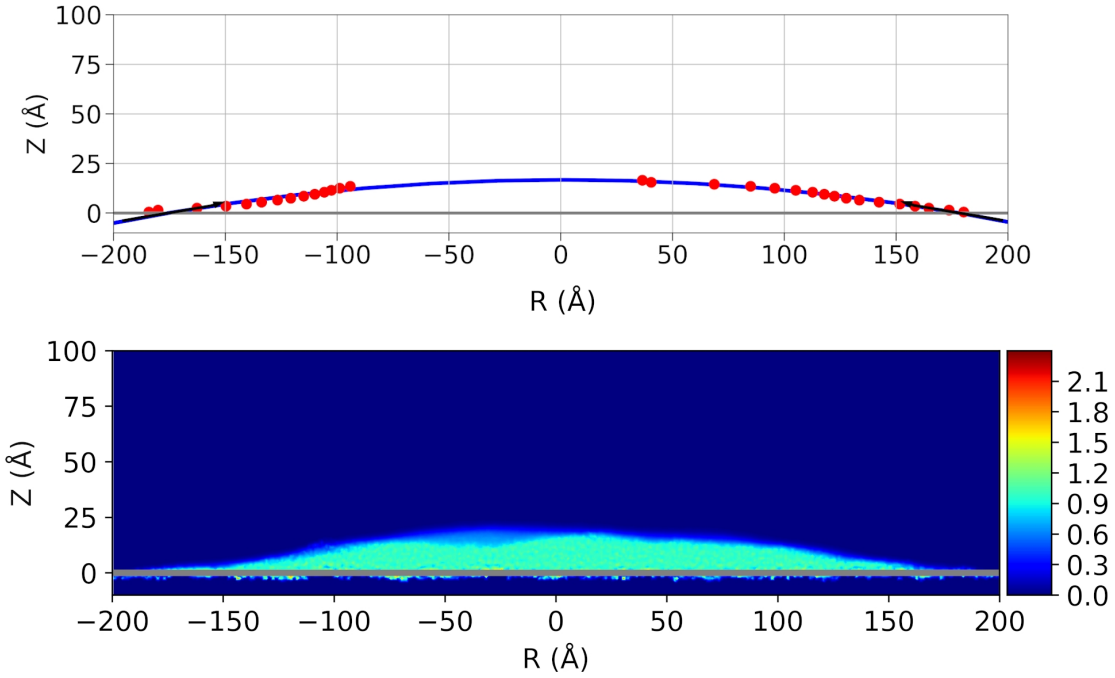


Figure S7. Water liquid boundary (red points), fitted circle (blue line), surface plane (gray line) and tangent line to the points of contact of the three phases (black line) for graphene oxide functionalized with C/O = 2 (top). Selected surface plane is shown on the mass density contour maps (bottom).

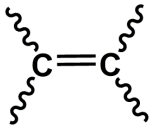
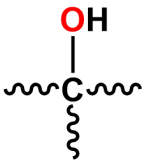
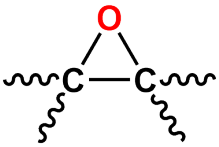
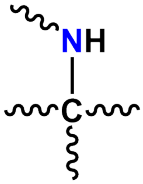
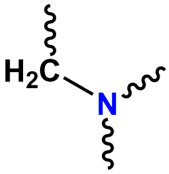
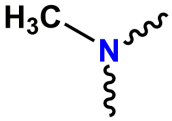
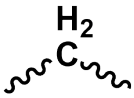
* Generation of topologies of graphene oxide functionalized with N-alkylamines.

To build the amino-modified models of GO, the nucleophilic ring-opening reaction mechanism between the N-alkylamines and epoxy functional groups was considered. Reaction between the N-alkylamines and epoxy groups yields the corresponding α-amino alcohols. Random addition of the N-alkylamine molecules on the GO model and topological file generation were performed by using a homemade python script, which is available for download at the following link "https://github.com/emejiao/Amino-modified-Graphene-Oxide". In order to use our script, it is necessary employ a LAMMPS datafile generated by make-graphitics as input.[1–4]

Table S1. OPLS-AA force field parameter for Lennard-Jones 12-6 potential.

Atom name	Type	ε (kcal/mol)	σ (Å)
C	sp^2	0.070	3.550
C	sp^3	0.066	3.500
O	-OH	0.170	3.120
O	-C-O-C-	0.140	2.900
H	-OH; -NH	0.000	0.000
H	$-CH_2-$; $-CH_3$	0.030	2.500
N	-NH	0.170	3.300

Table S2. Atomic charges of each functional group

Functional group	Atom type	Charge (e)
	C	0.000
	C	0.265
	O	-0.683
	H	0.418
	C	0.200
	O	-0.400
	C	0.183
	N	-0.778
	H	0.383
	C	0.083
	H	0.063
	C	0.023
	H	0.063
	C	-0.120
	H	0.060
	C	-0.180
	H	0.060

* **Bibliography.**

1. S. Plimpton, Fast parallel algorithms for short-range molecular dynamics, *J. Comput. Phys.*, 1995, **117**, 1–19.

2. R. C. Sinclair, J. L. Suter and P. V. Coveney, Graphene–Graphene Interactions: Friction, Superlubricity, and Exfoliation, *Adv. Mater.*, 2018, **30**, 1705791.

3. R. C. Sinclair and P. V. Coveney, Modeling Nanostructure in Graphene Oxide: Inhomogeneity and the Percolation Threshold, *J. Chem. Inf. Model.*, 2019, **59**, 2741–2745.

4. M. Vassaux, R. C. Sinclair, R. A. Richardson, J. L. Suter and P. V. Coveney, The Role of Graphene in Enhancing the Material Properties of Thermosetting Polymers, *Adv. Theory Simulations*, 2019, **2**, 1800168.