Supporting information for:

PAMAM dendrimers and graphene: Materials for removing aromatic contaminants from water

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Methods

Dendrimer OPLS-AA Parameterization

The OPLS-AA force field parameters as implemented in GROMACS were used in our simulations and are summarized in below.

Table S1: OPLS-AA parameterization for PAMAM dendrimer. Type numbers taken from GROMACS v4.5.5 implementation of OPLS-AA.

Atom Type	Description	OPLS Type
C1	CH2(N) tertiary aliphatic amines	opls_908
H1	H(C) for C bonded to N in amines	opls_911
N1	N tertiary amines	opls_902
C2	alkane CH2	opls_136
H2	alkane H	opls_140
C3	C=O in amide	opls_235
O1	O: C=O in amide	opls_236
N2	N: secondary amide	opls_238
H3	H on N: secondary amide	opls_241
C4	C on N: secondary N-CH2R amide	opls_244
C5	C in RCH2NH3+	opls_292
N3	N (RNH3+) JPC,90,2174 (1986)	opls_287
H4	H (RNH3+) JPC,90,2174 (1986)	opls_290

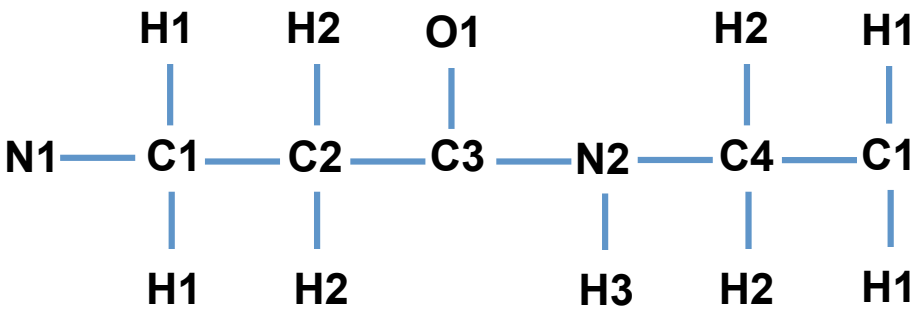

Figure S1: Single-branch parameterization of OPLS-AA for PAMAM dendrimer. Note for the terminal branches the right most C1 is replaced by a C5, and it is attached to a NH3+ group with N3 and H4 respectively. The ethyldiamine core is composed of C2, H2, and N1 types.

Graphene Oxide Partial Charges

We recalculated the partial charges on GrO since OPLS-AA partial charges were obtained for liquid hydrocarbons while GrO is a planar solid with unique electrical properties. The partial charges of the atoms on the GrO sheet were determined with density functional theory. Three different 2×2 nm^2 GrO sheets with 220 to 230 atoms and random placement of oxidized groups were generated using the technique described in the Methods section of the paper. Partial charges were then calculated using Gaussian 09[S1] with B3LYP/6-31G(d)[S2] basis set. The partial charges from the three unique sheets were averaged over 18 different atom types. Our partial charges are reported in Table S2. We considered sp^3, sp^2, and aromatic carbons as different atom types, and also created additional atom types for any atoms within 2 layers of the edge of the sheet to account for the effect of finite size of the sheet. Fourteen of the eighteen atom types have partial charges within $0.15e$ of the OPLS-AA partial charges. The largest difference between our partial charges and those of OPLS-AA was on sp^3 carbons bound to a hydrogen, where we report a charge of -0.392e and OPLS-AA reports a partial charge of 0.00.

Table S2: Partial charges determined with Gaussian 09[S1] and used for GrO systems.

Atom Type	Description	Mass	Charge	OPLS Type	OPLS Charge
C1	sp^3 C bonded to epoxide	12.0107	0.1366	opls_139	0.00
C11	sp^3 C bonded to epoxide [edge]	12.0107	0.0999	opls_139	0.00
C2	sp^3 C bonded to alcohol	12.0107	0.0082	opls_139	0.00
C12	sp^3 bonded to alcohol [edge]	12.0107	-0.0414	opls_139	0.00
C3	sp^3 C bonded to hydrogen	12.0107	-0.3918	opls_139	0.00
C13	sp^3 C bonded to hydrogen [edge]	12.0107	-0.3191	opls_139	0.00
C4	sp^2 C (double bonded)	12.0107	0.1231	opls_142	-0.115
C14	sp^2 C (double bonded) [edge]	12.0107	-0.0692	opls_142	-0.115
C5	Aromatic C	12.0107	0.0461	opls_147	0.00
C15	Aromatic C [edge]	12.0107	-0.0358	opls_147	0.00
O1	Epoxide O	15.9994	-0.4625	opls_180	-0.40
O11	Expoxide O [edge]	15.9994	-0.4660	opls_180	-0.40
O2	Alcohol O	15.9994	-0.7112	opls_154	-0.683
O12	Alcohol O [edge]	15.9994	-0.7167	opls_154	-0.683
H1	H bonded to C	1.0079	0.2810	opls_140	0.06
H11	H bonded to C [edge]	1.0079	0.1595	opls_140	0.06
H2	H bonded to O	1.0079	0.4227	opls_155	0.418
H12	H bonded to O [edge]	1.0079	0.4123	opls_155	0.418

Results

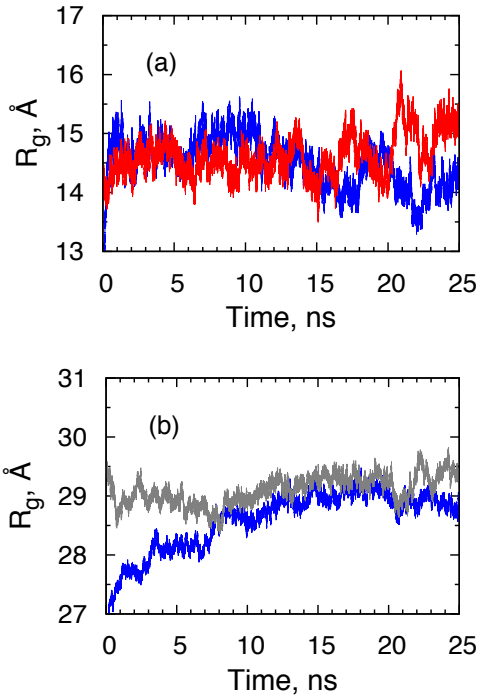

Figure S2: Time evolution of the radius of gyration (R_g) of (a) G3 (red) and (b) G6 (gray) dendrimer in water-dendrimer-NPH systems. The R_g values for G3 and G6 dendrimer in bulk water (blue) are also shown for reference.

Table S3: R_g of PAMAM dendrimers in bulk water, averaged over the last 10 ns for the dendimer-water and over the last 50 ns of the simulation for the dendrimer-water-NPH system. Results reported from other simulation studies are for the dendrimer structure under neutral pH conditions. Error on the R_g values was calculated using the block averaging technique[S3] implemented in GROMACS.

Source	**Force Field**	**R_g (nm)**			
		G3	G4	G5	G6
This work (water)	OPLS	1.42 ± 0.01	1.95 ± 0.04	2.29 ± 0.01	2.90 ± 0.02
Maiti *et al.* 2005[S4]	Dreiding	-	1.70 ± 0.01	2.22 ± 0.01	2.73 ± 0.04
Maingi *et al.* 2012[S5]	GAFF	1.58 ± 0.03	2.06 ± 0.02	2.53 ± 0.02	3.05 ± 0.01
Prosa *et al.* 1997[S6]	SAXS	-	$1.71 \pm$ n/a	$2.41 \pm$ n/a	$2.63 \pm$ n/a
Rathgeber *et al.* 2002[S7]	SAXS	1.51 ± 0.005	1.86 ± 0.007	2.31 ± 0.003	2.75 ± 0.003
Porcar *et al.* 2008[S8]	SANS	1.67 ± 0.12	2.14 ± 0.04	2.68 ± 0.04	3.30 ± 0.04
This work (solution)	OPLS	1.47 ± 0.02	1.93 ± 0.01	2.29 ± 0.05	2.91 ± 0.02

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