Supporting Information

Mechanistic Insights into the Inhibition and Size effects of Graphene Oxide Nanosheets on the Aggregation of an Amyloid-β Peptide Fragment

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This material contains the description of how to set up partial charge of atoms in oxidation groups of GO nanoparticles, five tables and eight figures.

Partial charges setup for atoms in oxidation groups of GO nanoparticles. The partial charges of atoms in oxidation groups of GO (Fig. S1) were taken from similar groups of three residues (TYR, GLU and ASP) and the base of DNA with minor modification. Oxygen atom in epoxy group (-C-O-C-) have a partial charge of -0.4e, which is similar to the charge of O4' atom in deoxyribonucleic acid (-0.37e, more details can be seen in Table S1). Both of the two carbon atoms in epoxy group have a partial charges of 0.2e and the epoxy group is electric neutral. Based on the hydroxyl group of TYR side chain (Table S2), the charges of oxygen and hydrogen atoms in hydroxyl group (-O-H) were set to be -0.55e and 0.35e, respectively. The carbon atom connected with oxygen atom has a partial charges of 0.2e to ensure the hydroxyl group electroneutral. For carboxyl group ($-COO^-$), ASP (Table S3) and GLU (Table S4) were introduced as references. The carbon atom has 0.7e and two oxygen atoms have

equal charges of -0.8e. The carbon atom which connects to the carbon atoms of carboxyl groups was set to be -0.1e and the total charge of carboxyl group is -1e to simulate the deprotonated state. Other carbon atoms didn't mentioned above in the GO are uncharged.

Table S1-S5:

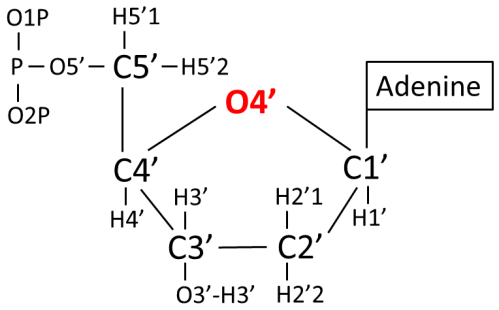
dAMP					
Atom	Charge	Atom	Charge	Atom	Charge
P	1.16590	H1'	0.18380	C2	0.57160
O1P	-0.77610	N9	-0.02680	H2	0.05980
O2P	-0.77610	C8	0.16070	N3	-0.74170
O5'	-0.49540	H8	0.18770	C4	0.38000
C5'	-0.00690	N7	-0.61750	C3'	0.07130
H5'1	0.07540	C5	0.07250	H3'	0.09850
H5'2	0.07540	C6	0.68970	C2'	-0.08540
C4'	0.16290	N6	-0.91230	H2'1	0.07180
H4'	0.11760	H61	0.41670	H2'2	0.07180
O4'	**-0.36910**	H62	0.41670	O3'	-0.52320
C1'	0.04310	N1	-0.76240		

Table S1. The partial charges of atoms in 2'-Deoxyadenosine 5'-mono-phosphate (dAMP) under the AMBER99SB-ILDN force field.

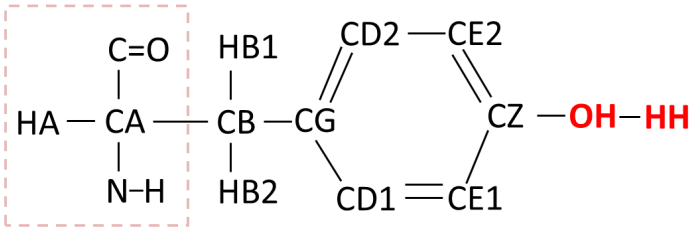
TYR					
Atom	Charge	Atom	Charge	Atom	Charge
N	-0.41570	CG	-0.00110	**HH**	**0.39920**
H	0.27190	CD1	-0.19060	CE2	-0.23410
CA	-0.00140	HD1	0.16990	HE2	0.16560
HA	0.08760	CE1	-0.23410	CD2	-0.19060
CB	-0.01520	HE1	0.16560	HD2	0.16990
HB1	0.02950	CZ	0.32260	C	0.59730
HB2	0.02950	**OH**	**-0.55790**	O	-0.56790

Table S2. The partial charges of atoms in Tyrosine (TYR) under the AMBER99SB-ILDN force field.

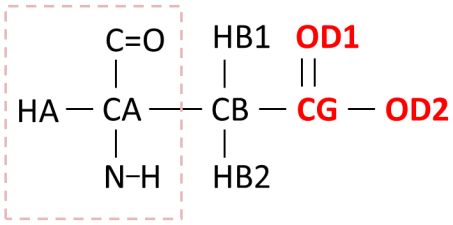
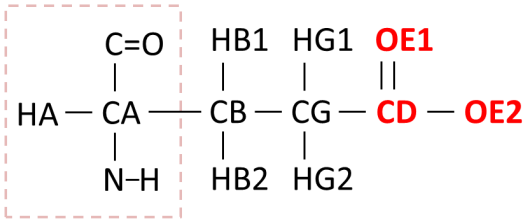
ASP					
Atom	Charge	Atom	Charge	Atom	Charge
N	-0.51630	CB	-0.03030	**OD1**	**-0.80140**
H	0.29360	HB1	-0.01220	**OD2**	**-0.80140**
CA	0.03810	HB2	-0.01220	C	0.53660
HA	0.08800	**CG**	**0.79940**	O	-0.58190

Table S3. The partial charges of atoms in Aspartate (ASP) under the AMBER99SB-ILDN force field.

GLU	

Atom	Charge	Atom	Charge	Atom	Charge
N	-0.51630	HB1	-0.01730	**CD**	**0.80540**
H	0.29360	HB2	-0.01730	**OE1**	**-0.81880**
CA	0.03970	CG	0.01360	**OE2**	**-0.81880**
HA	0.11050	HG1	-0.04250	C	0.53660
CB	0.05600	HG2	-0.04250	O	-0.58190

Table S4. The partial charges of atoms in Glutamic (GLU) under the AMBER99SB-ILDN force field.

Replicas	$A\beta_{33-42}$	$A\beta_{33-42}$ +4GO_{60}	$A\beta_{33-42}$ +2GO_{120}	Replicas	$A\beta_{33-42}$	$A\beta_{33-42}$ +4GO_{60}	$A\beta_{33-42}$ +2GO_{120}
1	307.00	307.50	307.50	25	368.57	369.97	368.11
2	309.39	309.92	309.86	26	371.34	372.79	370.82
3	311.79	312.36	312.23	27	374.13	375.62	373.55
4	314.20	314.81	314.62	28	376.93	378.47	376.30
5	316.64	317.28	317.02	29	379.75	381.33	379.07
6	319.08	319.76	319.44	30	382.59	384.22	381.83
7	321.55	322.25	321.86	31	385.45	387.12	384.64
8	324.02	324.77	324.31	32	388.26	390.03	387.45
9	326.52	327.29	326.77	33	391.15	392.97	390.28
10	329.03	329.84	329.24	34	394.06	395.93	393.13
11	331.56	332.40	331.72	35	396.99	398.91	395.99
12	334.10	334.98	334.22	36	399.94	401.91	398.88

13	336.66	337.57	336.73	37	402.91	404.92	401.78
14	339.23	340.17	339.26	38	405.88	407.96	404.69
15	341.82	342.80	341.81	39	408.88	411.01	407.62
16	344.42	345.44	344.37	40	411.91	414.08	410.57
17	347.04	348.10	346.94	41	414.95	417.17	413.53
18	349.67	350.77	349.53	42	418.01	420.29	416.51
19	352.32	353.46	352.14	43	421.08	423.42	419.52
20	354.99	356.17	354.76	44	424.18	426.57	422.54
21	357.68	358.90	357.40	45	427.31	429.74	425.58
22	360.38	361.64	360.05	46	430.44	432.93	428.64
23	363.09	364.40	362.72	47	433.62	436.14	431.71
24	365.82	367.18	365.41	48	436.79	439.37	434.81

Table S5. Temperature list of 48 replicas in each system (307-436.79 K for $A\beta_{33-42}$ system; 307.5-439.7 K for $A\beta_{33-42}$+4GO_{60} system; 307.5-450.59 K for $A\beta_{33-42}$+2GO_{120} system).

Figure S1-S8:

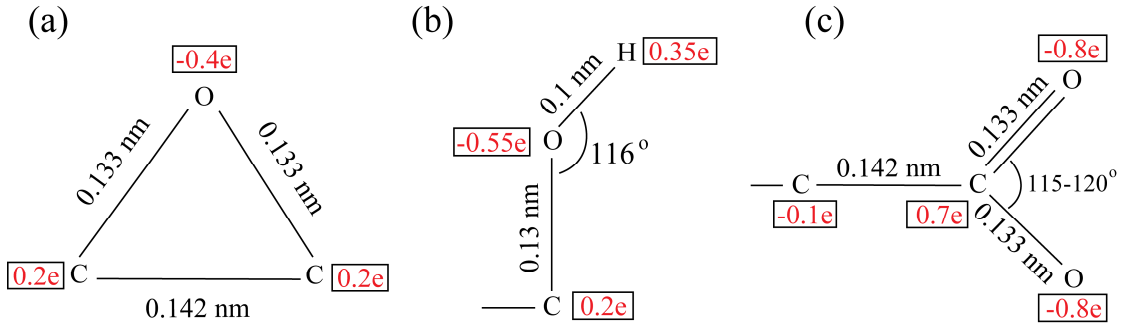


Figure S1. Illustrations of partial charges of the atoms in (a) epoxy group, (b) hydroxyl group and (c) carboxyl group

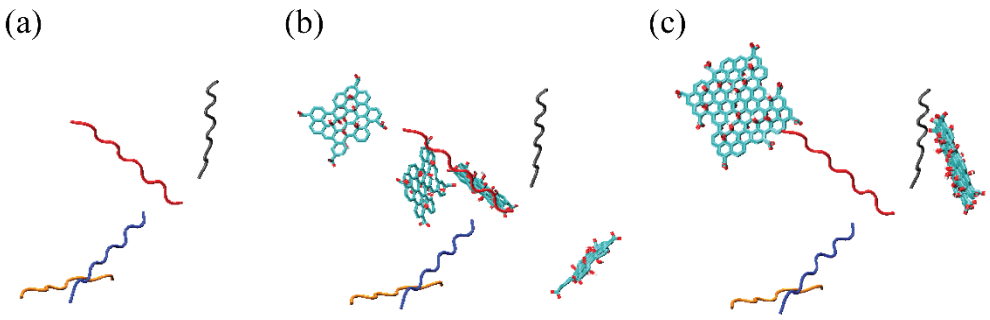


Figure S2. The initial states of (a) $A\beta_{33\text{-}42}$ system, (b) $A\beta_{33\text{-}42}$+$4GO_{60}$ system and (c) $A\beta_{33\text{-}42}$+$2GO_{120}$ system.

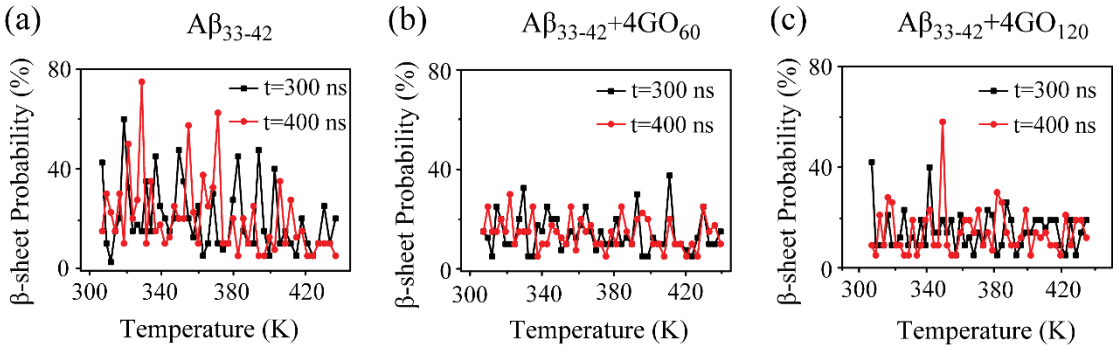


Figure S3. The probability of β-sheet among 48 temperature replicas at 300 ns and 400 ns. (a) $A\beta_{33\text{-}42}$ system, (b) $A\beta_{33\text{-}42}$+$4GO_{60}$ system, (c) $A\beta_{33\text{-}42}$+$2GO_{120}$ system.

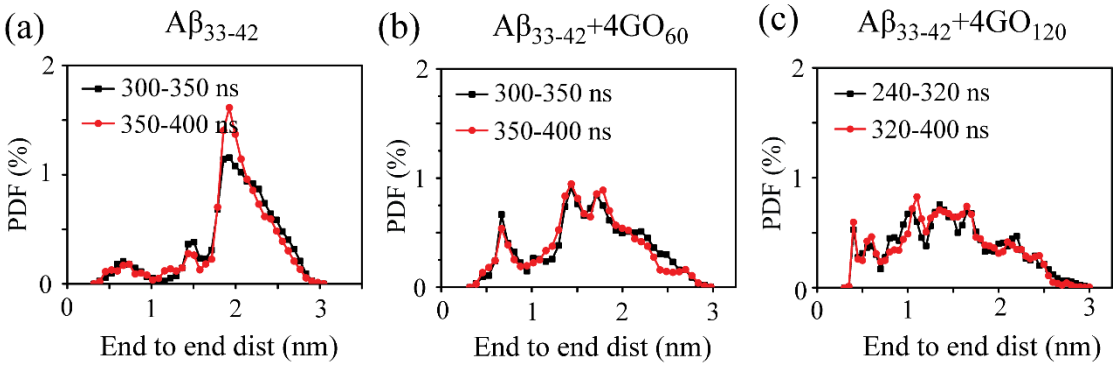


Figure S4. The probability density function (PDF) of end-to-end distances of all peptide chains within two time intervals for (a) $A\beta_{33\text{-}42}$ system, (b) $A\beta_{33\text{-}42}$+$4GO_{60}$ system, (c) $A\beta_{33\text{-}42}$+$2GO_{120}$ system.

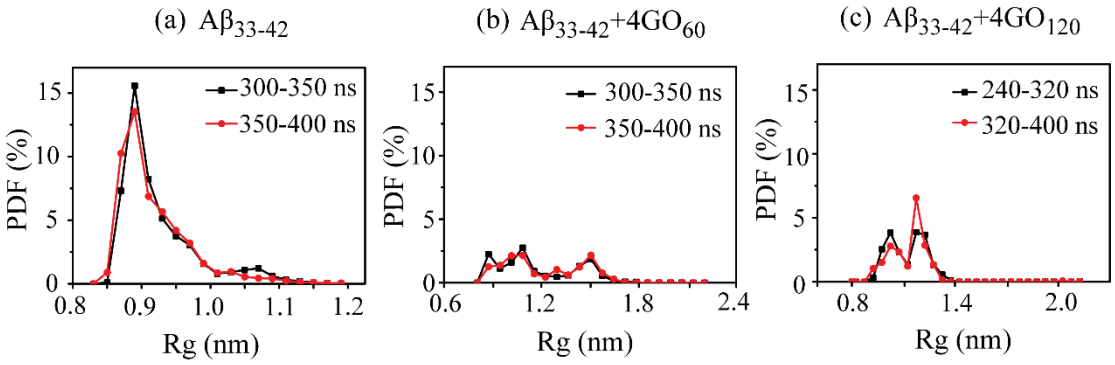


Figure S5. The PDF of the radius of gyration of the Aβ-tetramer within two time intervals in (a) $A\beta_{33\text{-}42}$ system, (b) $A\beta_{33\text{-}42}$+$4GO_{60}$ system, (c) $A\beta_{33\text{-}42}$+$2GO_{120}$ system.

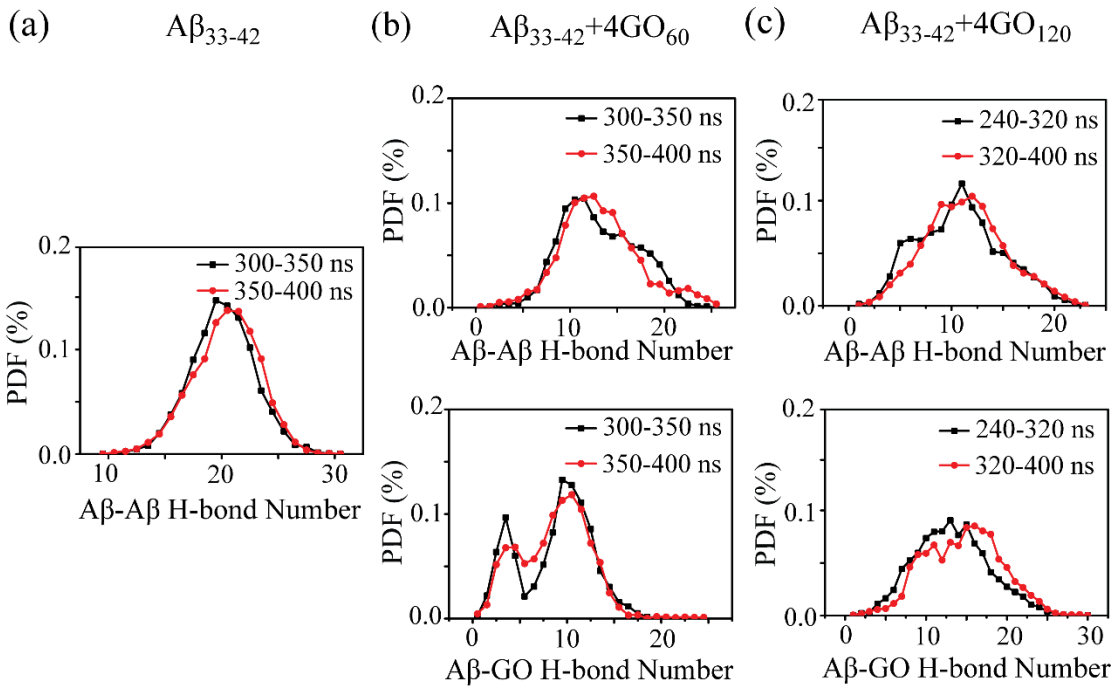


Figure S6. The PDF of the H-bond number of peptide-peptide and peptide-GO within two time intervals for (a) $A\beta_{33\text{-}42}$ system, (b) $A\beta_{33\text{-}42}$+$4GO_{60}$ system, (c) $A\beta_{33\text{-}42}$+$2GO_{120}$ system.

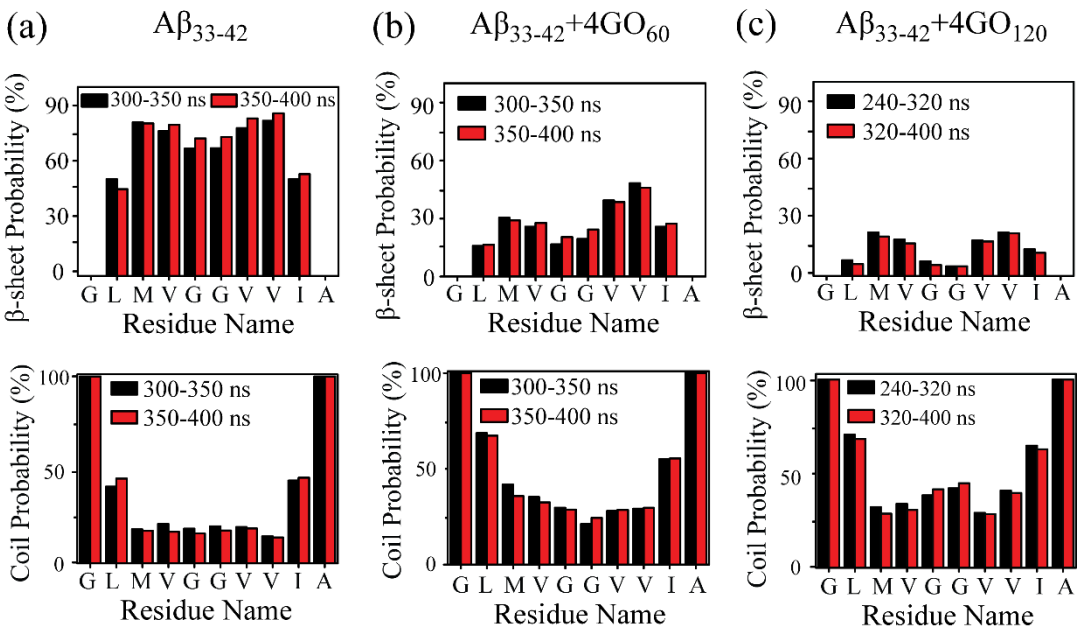


Figure S7. The calculated secondary structure (coil and β-sheet) probability of each residue in the REMD runs for Aβ-tetramer in three systems within two time intervals for (a) $A\beta_{33\text{-}42}$ system, (b) $A\beta_{33\text{-}42}$+$4GO_{60}$ system, (c) $A\beta_{33\text{-}42}$+$2GO_{120}$ system.

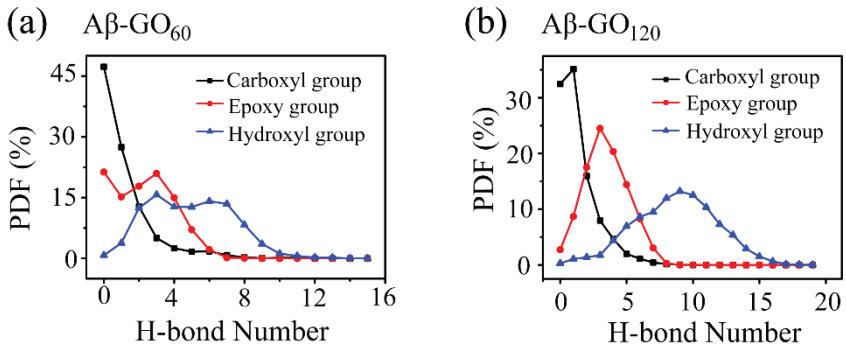


Figure S8. The PDF of H-bond number between Aβ and different oxidation groups (carboxyl group, epoxy group and hydroxyl group) of GO. (a) $A\beta_{33\text{-}42}$+$4GO_{60}$ system. (b) $A\beta_{33\text{-}42}$+$2GO_{120}$ system.