Supplementary Materials

Surface Inhomogeneity of Graphene Oxide Influences Dissociation of $A\beta_{16-21}$ Peptide Assembly

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Context:

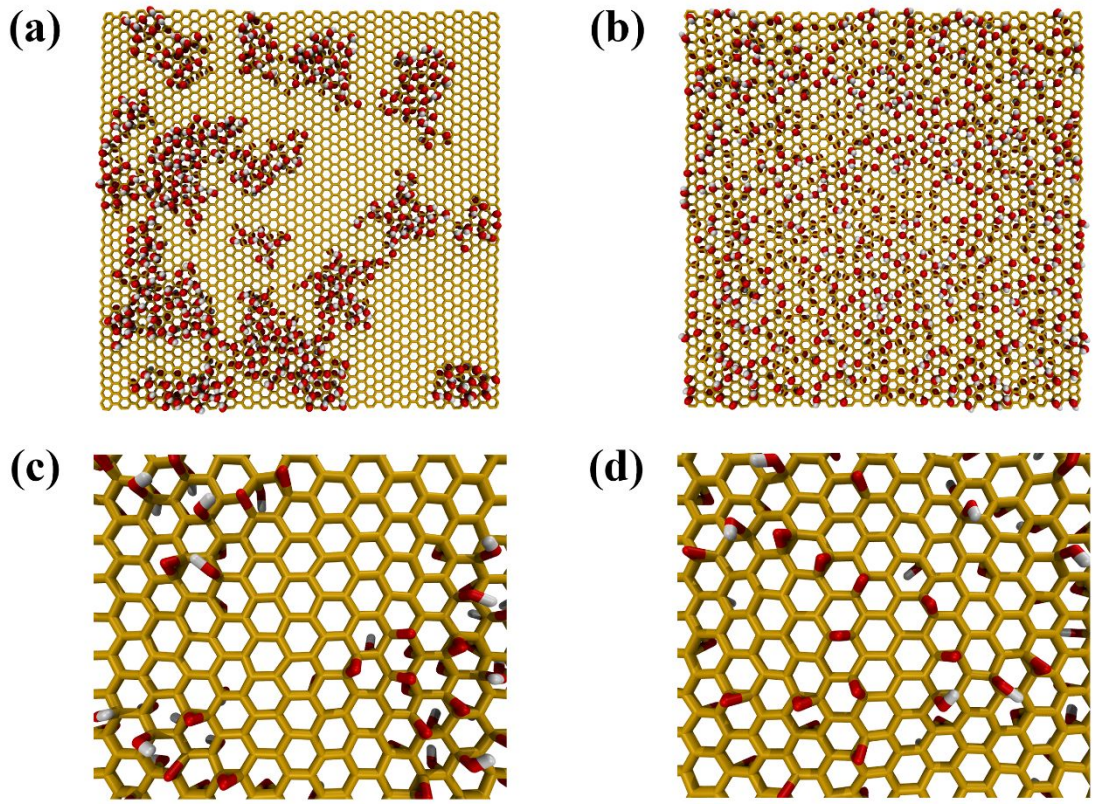

Fig. S1 A nonuniform GO sheet with concentrated distributions of epoxy and hydroxyl groups ((a), and (c) for the zoomed-in view); and a uniform GO sheet with random distribution of epoxy and hydroxyl groups ((b), and (d) for the zoomed-in view). The carbon, oxygen, hydrogen atoms in GO are colored in yellow, red and white, respectively. Both GO sheets have a C/O ratio of 10/3.

Table S1. Partial charges (in units of the elementary charge), σ (nm) and ε (kJ mol^{-1}) for atoms in epoxy and hydroxyl groups of GO.

Functional group	Atom	Charge (*e*)	σ (*nm*)	ε (*kJ* mol^{-1})
Epoxy	C	0.18	0.340	0.360
	O	-0.36	0.300	0.711
Hydroxyl	C	0.18	0.340	0.360
	O	-0.57	0.307	0.880
	H	0.39	0.000	0.000

S2. Discussion of temperature effect in simulations.

In the main text, we raise the temperature of solvents to 500 K in our simulations, in order to accommodate photothermal effect. The local high temperature is expected to accelerate the dissociation of Aβ fibrils. Here, we also performed simulations at normal temperature (300 K). The initial configurations are same as simulations in the main text (**Fig. 1(a, c)**). As shown in **Fig. S2 (a, b)**, the structure of $A\beta_{16\text{-}21}$ assembly is disturbed at some level on GO-nonuniform, whereas the Aβ structure remains largely intact on GO-uniform at t = 400 ns. **Fig. S2 (c)** demonstrates β-sheet contents of Aβ peptides on GO-nonuniform and GO-uniform after 400-ns simulations at 300 K versus 500 K. On GO-uniform, the β-sheet content (51.9%) at 500 K is slightly lower than that (54.4%) at 300 K. On the other hand, on GO-nonuniform, the β-sheet content is strongly reduced to 29.2% at 500K, which is much lower than 43.3% at 300 K.

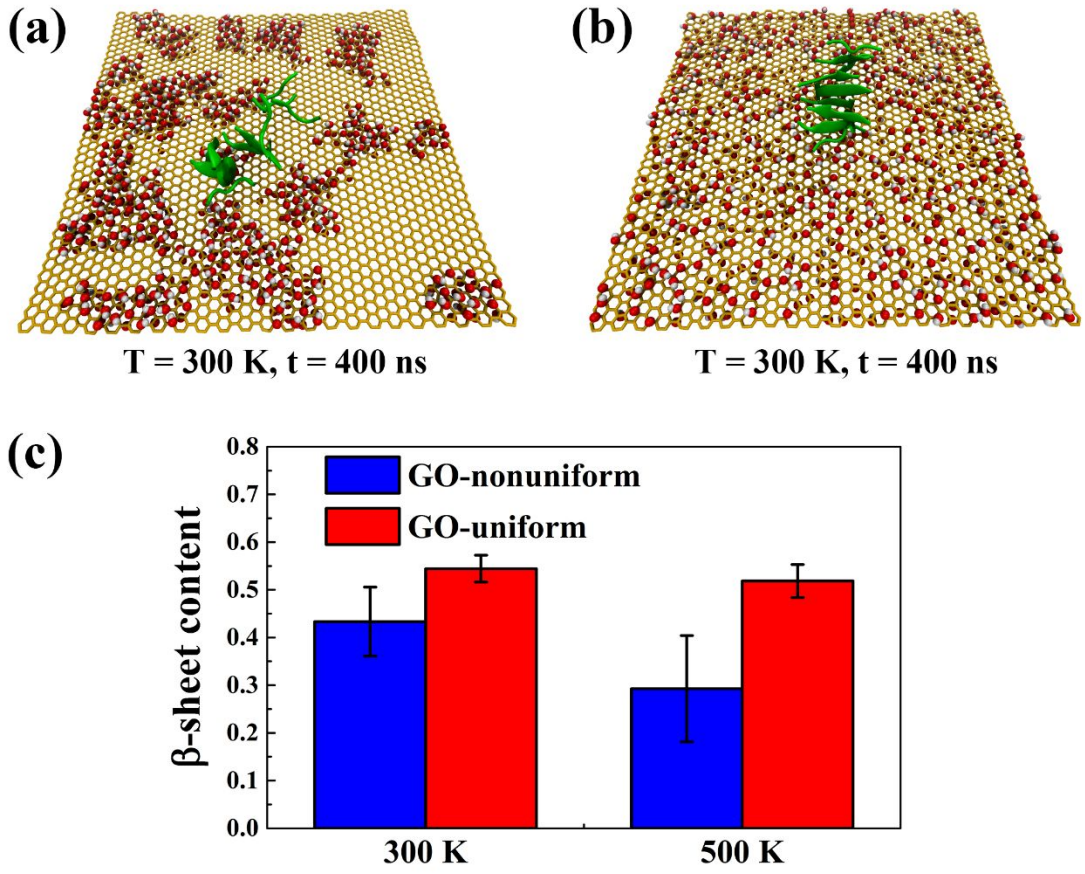

Fig. S2 Snapshots of final configurations of $A\beta_{16\text{–}21}$ peptide assemblies on GO-nonuniform (a) and GO-uniform (b) in simulations at 300 K. (c) β-sheet contents of Aβ peptides on GO-nonuniform

(blue bar) and GO-uniform (red bar) after 400-ns simulations at 300 K versus 500 K. The data was based on 20 simulations (5 independent simulations for each system).

S3. Simulation results of $A\beta_{16–21}$ peptide assemblies on other nonuniform GO sheets.

In addition, we generated another four nonuniform GO nanosheets and performed additional simulations for all of them. The initial configurations and other setups are same as the simulation in the main text (**Fig. 1(a)**). As shown in **Fig. S3(a-d)**, Aβ peptides can also be dissociated on other nonuniform GO sheets after 400-ns simulations. Moreover, **Fig. S3(e, f)** show that aromatic residues (Phe) of $A\beta_{16-21}$ prefer to bind to the sp^2 regions while the polar residues (Lys) tend to bind to the oxidized region. These additional simulations and analyses further support our conclusion in the main text.

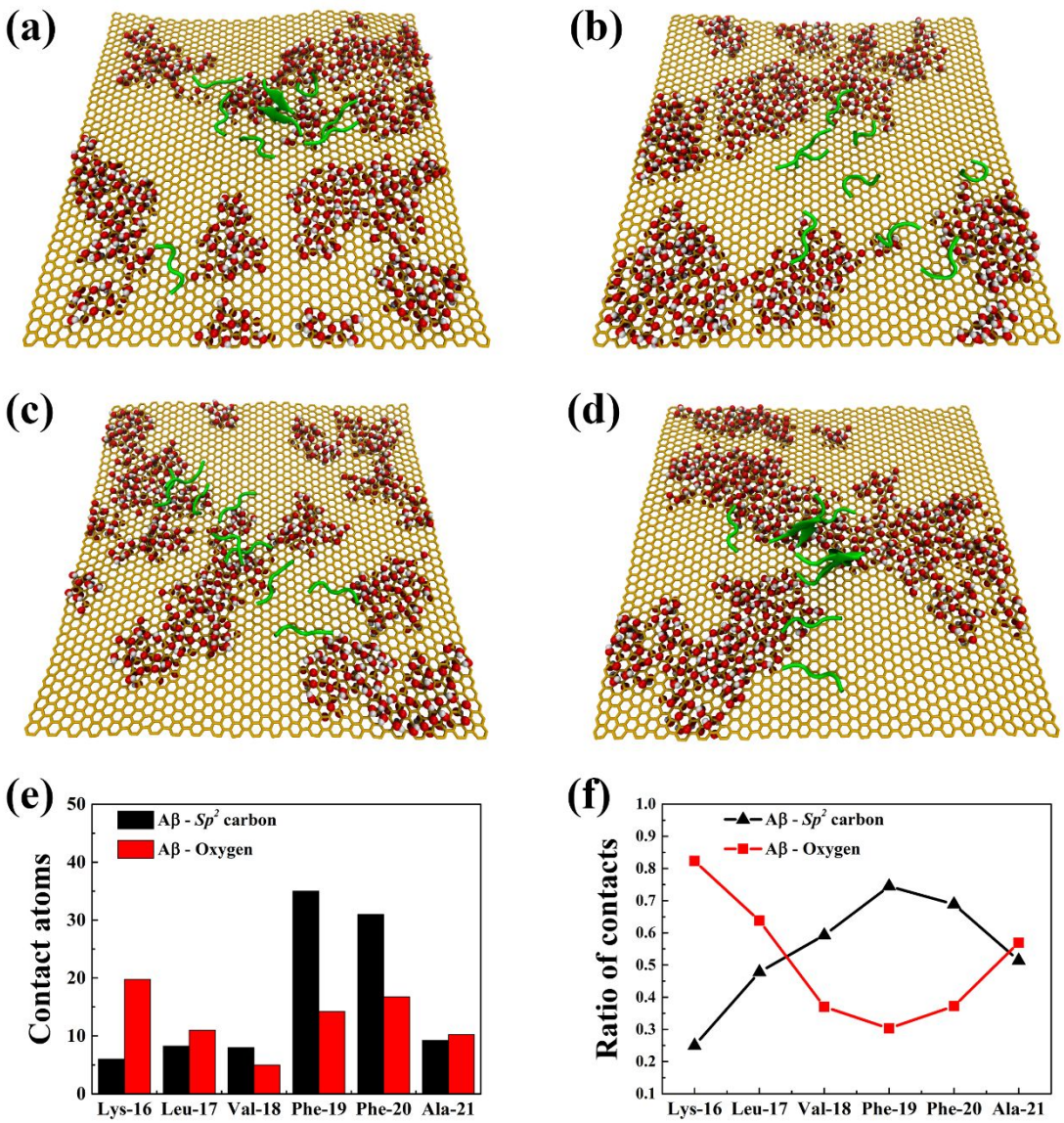

Fig. S3 (a-d) Snapshots of final configurations of $A\beta_{16–21}$ peptide assemblies on other nonuniform GO sheets with a C/O ratio of 10/3, at t = 400 ns. (e) Contact atoms of each residue on the sp^2 region (black bar) versus the oxidized region (red bar) on GO-nonuniform. (f) Ratios of contact atoms on sp^2 (black) and oxidized regions (red) to their respective total contact numbers for each residue. The data was based on the last 10-ns trajectories of above simulations.