Supporting Information:

Mechanistic modeling of spontaneous penetration of carbon nanocone into membrane vesicle

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S1. Membrane wrapping of carbon nanotube and graphene nanosheet

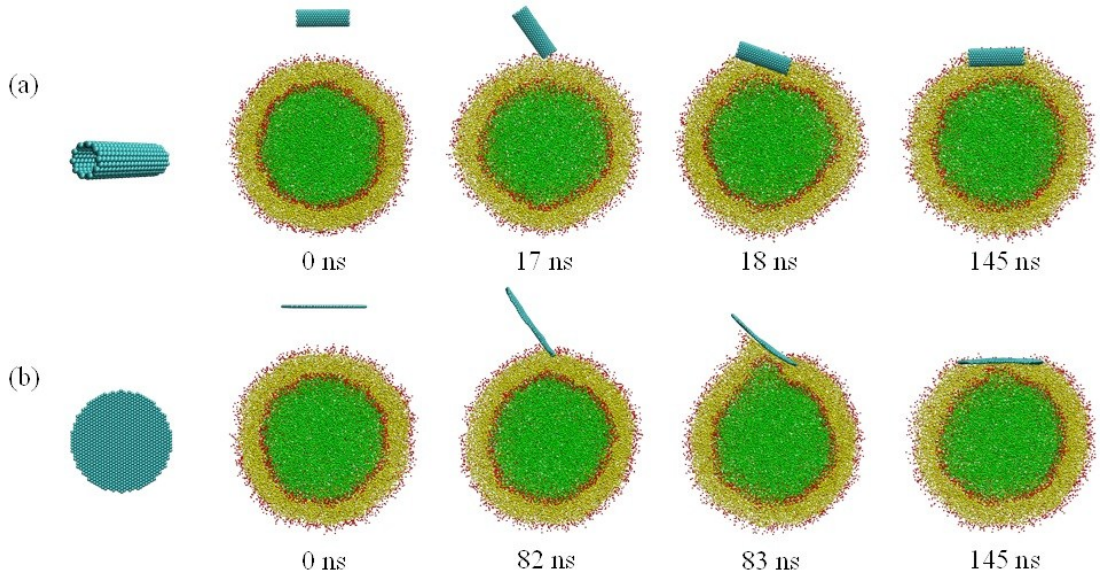

Fig. S1. Representative simulation snapshots showing the penetration process of (***a***) carbon nanotube and (***b***) graphene nanosheet.

S2. Theoretical model of carbon nanocone entering lipid membrane

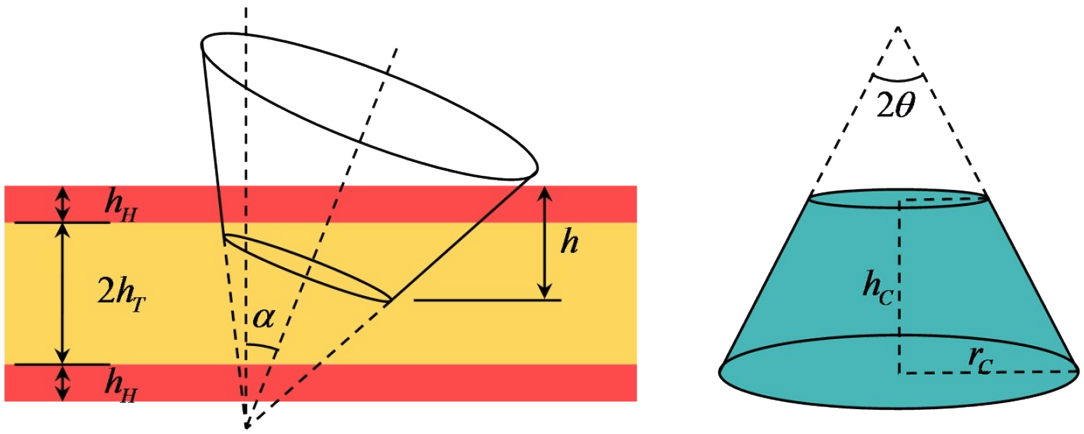

Fig. S2. Schematic representation of the theoretical model of carbon nanocone entering lipid membrane.

S3. Free energy analysis

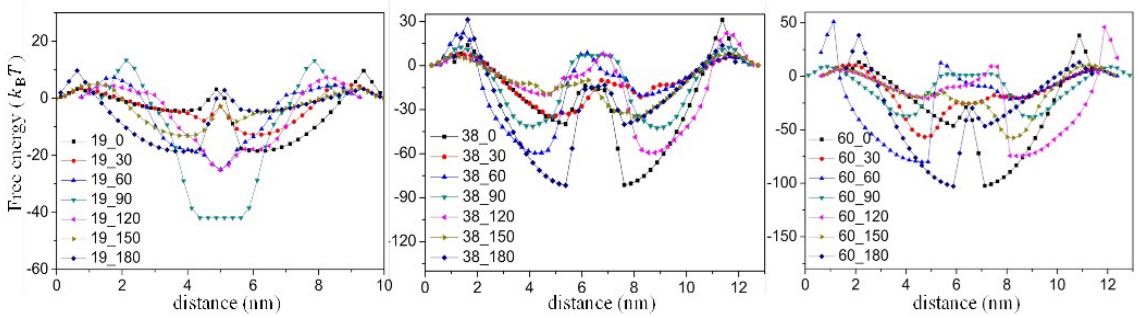

Fig. S3. Free energy evolutions of nanocones with apex angles of 19°, 38° and 60° and different orientations.

S4. DPD simulation of the self-assembly process of membrane vesicle from lipid molecules

Here, we performed DPD simulation to investigate the spontaneous formation of membrane vesicle from lipid molecules. Our results show that the lipid molecules aggregate into many small spherical micelles, which then merge to form oblate membrane. The oblate membranes can close up to form small vesicles (Fig. S4, Middle). In the next stage, the small vesicles still grow to form larger ones by including the neighboring small micelles and vesicles (Fig. S4, Right).

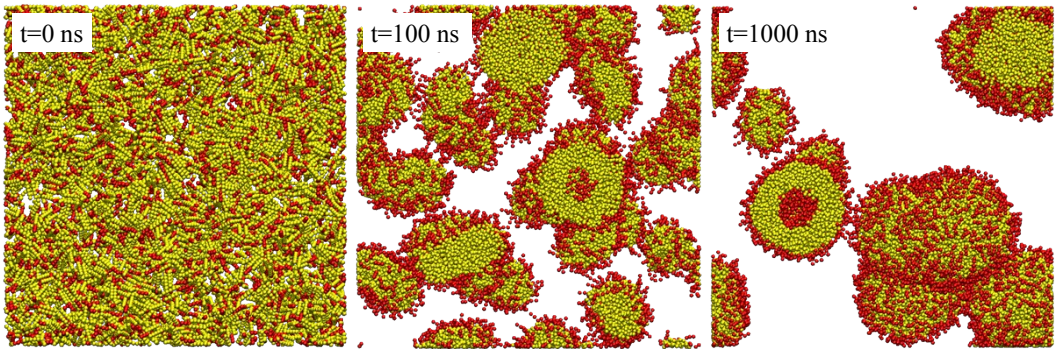

Fig. S4. The self-assembled membrane vesicle from lipid molecules. For clarity, the solvent particles are not shown.

S5. Free energy estimation via thermodynamic integration method

Besides the free energy theory, we also adopt thermodynamic integration method to calculate the free energy. We apply a harmonic potential on the CNC to constrain its motion in z direction and the CNC was forced to oscillate around a pseudo-equilibrium position. Then the derivative of the free energy was determined from the constrained interaction between the CNC and its surroundings. However, out of the flexibility of the lipid membrane, the free energy obtained from this approach could have an obvious error. Thus, we adopted the free energy theory to calculate the free energy. An example of the comparison between free energy theory and thermodynamic integration method is shown in Fig. S5.

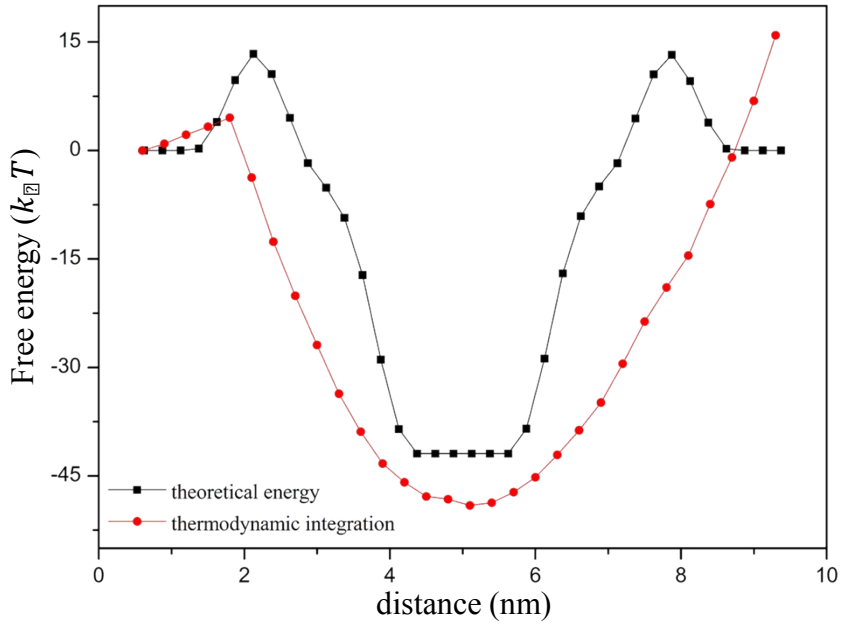

Fig. S5. Free energy of the CNC with apex angle of 19° obtained from Eq. (8) and thermodynamic integration method.