## **Support Information**



**Figure S1**: (a) End-to-end distance distribution of lipid molecules in the collision process between hydrophobic nanoparticle and the vesicle at  $V_0 = 0.813$  m/s. (b) A representative snapshot of the cross-section of the vesicle before the collision ( $t < 0.012 \ \mu$ s), (c) in the middle of the collision ( $0.2 < t < 0.7 \ \mu$ s), (d) in the latestage interface embedded state ( $t > 4.2 \ \mu$ s).



**Figure S2**: (a) A number density profile in the radial direction for the initial configuration (at t = 0). (b) A number profile of beads in the z-direction for the initial configuration (at t = 0). In both (a) and (b), the zero in the horizontal axis refers to the center of mass of the vesicle. Color code: Head groups in blue, tail groups in red, and water beads in cyan.

## **Supporting Movies:**

All supporting movies are cross-section view. Color code: Head groups and hydrophilic section of the nanoparticle in blue, tail groups and hydrophobic section of the nanoparticle in red, water beads originally inside the vesicle in cyan, water beads once outside the vesicle in yellow. Water beads are not displayed in Movies S1-S10 and S12.

**Movie S1**: A *rebound* pathway for the homogeneous hydrophilic nanoparticle at  $V_0 = 0.26$  m/s and 0 < t < 0.47 µs.

**Movie S2**: An *expulsion* pathway for the homogeneous hydrophilic nanoparticle at  $V_0 = 0.51$  m/s and 0 < t < 2.35 µs.

**Movie S3**: A *penetration* pathway for the homogeneous hydrophilic nanoparticle at  $V_0 = 1.21$  m/s and 0 < t < 0.45 µs.

**Movie S4**: A *drag-in* pathway for the homogeneous hydrophobic nanoparticle at  $V_0 = 0.36$  m/s and 0 < t < 1.17 µs.

**Movie S5**: A *retraction* pathway for the homogeneous hydrophobic nanoparticle at  $V_0 = 1.43$  m/s and 0 < t < 1.05 µs.

**Movie S6**: A *reverse* pathway for the Janus nanoparticle with initial orientation of Fig. 1c1 at  $V_0 = 0.51$  m/s and 0 < t < 1.17 µs.

**Movie S7**: A *normal* pathway for the Janus nanoparticle with initial orientation of Fig. 1c1 at  $V_0 = 1.15$  m/s and 0 < t < 0.56 µs.

**Movie S8**: A *capsulization* pathway for the homogeneous hydrophobic nanoparticle at  $V_0 = 0.81$  m/s and 7.03 < t < 9.38 µs.

**Movie S9**: A *capsulization* pathway for the homogeneous hydrophilic nanoparticle at  $V_0 = 0.81$  m/s and 0.23 < t < 2.11 µs.

**Movie S10**: A *capsulization* pathway for the Janus nanoparticle with initial orientation of Fig. 1c2 at  $V_0 = 0.81$  m/s and 3.51 < t < 5.86 µs.

**Movie S11**: Self-rotation of the Janus nanoparticle in water with initial orientation of Fig. 1c3 at  $V_0 = 0.57$  m/s and 0 < t < 0.31 µs.

**Movie S12**: A *penetration* pathway for the Janus nanoparticle with initial orientation of Fig. 1c2 at  $V_0 = 1.41$  m/s and 0 < t < 0.52 µs.

**Movie S13**: Water discharge in the *expulsion* pathway with the homogeneous hydrophilic nanoparticle at  $V_0 = 0.51$  m/s and 0 < t < 2.7 µs.

**Movie S14**: Water discharge in the *capsulization* pathway with the homogeneous hydrophilic nanoparticle at  $V_0 = 0.81$  m/s and 0 < t < 2.7 µs.

**Movie S15**: Water discharge in the *reverse* pathway with initial orientation of Fig. 1c1 at  $V_0 = 0.47$  m/s and 0 < t < 1.41 µs.

**Movie S16**: Water discharge in the *penetration* pathway with initial orientation of Fig. 1c1 at  $V_0 = 1.44$  m/s and 0 < t < 1.64 µs.

**Movie S17**: Water discharge in the *penetration* pathway with initial orientation of Fig. 1c2 at  $V_0 = 1.44$  m/s and 0 < t < 2.34 µs.

**Movie S18**: Water discharge in the *penetration* pathway with initial orientation of Fig. 1c3 at  $V_0 = 1.44$  m/s and 0 < t < 2.81 µs.