Electronic Supplementary Information for

Direct Measurements of the Interactions between Clathrate Hydrate Particles and Water Droplets

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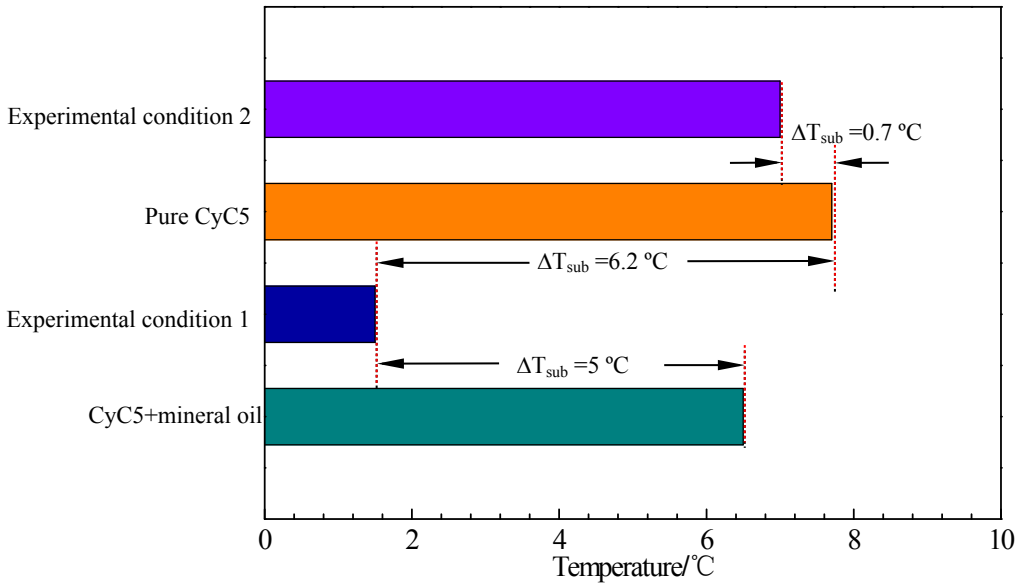
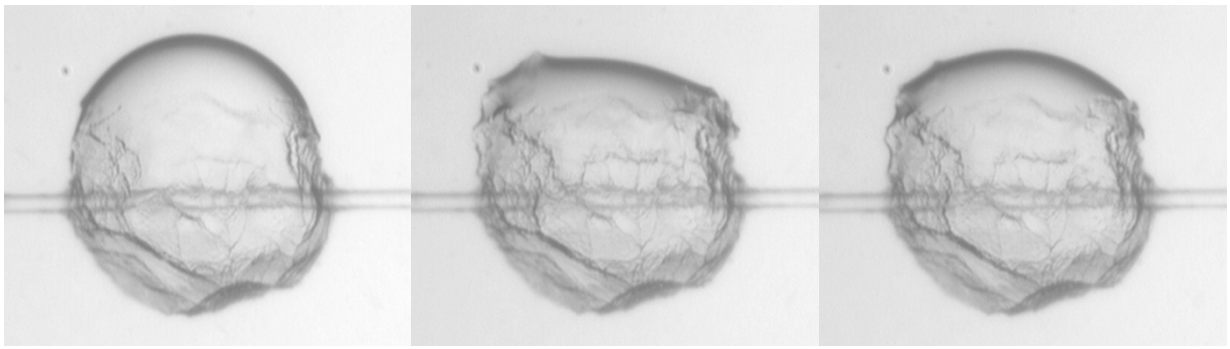
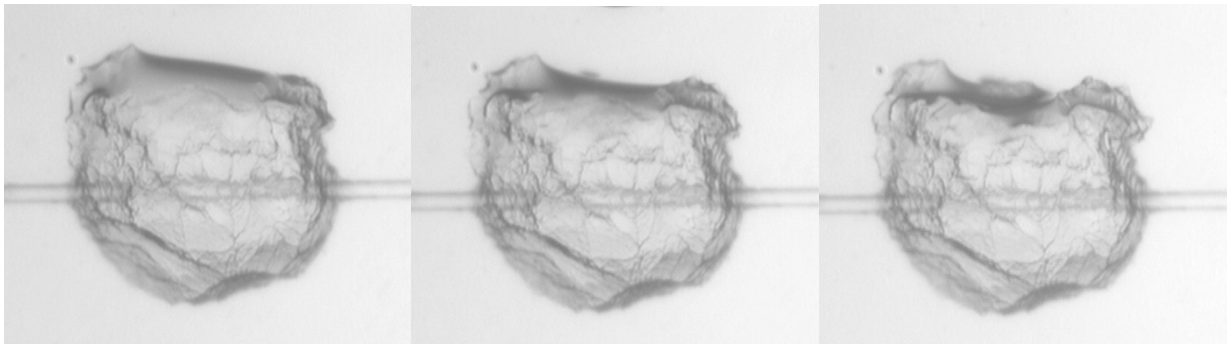


Fig. S1 Cyclopentane hydrate equilibrium conditions at atmospheric pressure. The addition of mineral oil decreases the cyclopentane hydrate equilibrium temperature. At atmospheric pressure, the hydrate equilibrium temperature of the CyC5+mineral oil mixture is about 6.5 ºC (by PVTsim®), while the hydrate equilibrium temperature of pure cyclopentane is about 7.7 ºC[1].



(i) (ii) (iii)



(iv) (v) (vi)

Fig. S2 Hydrate formation process of the remaining water on the hydrate particle at lower subcooling (0.7 ºC). i-vi: t=4467 s. The diameter of the glass fiber in the images is 38 μm.

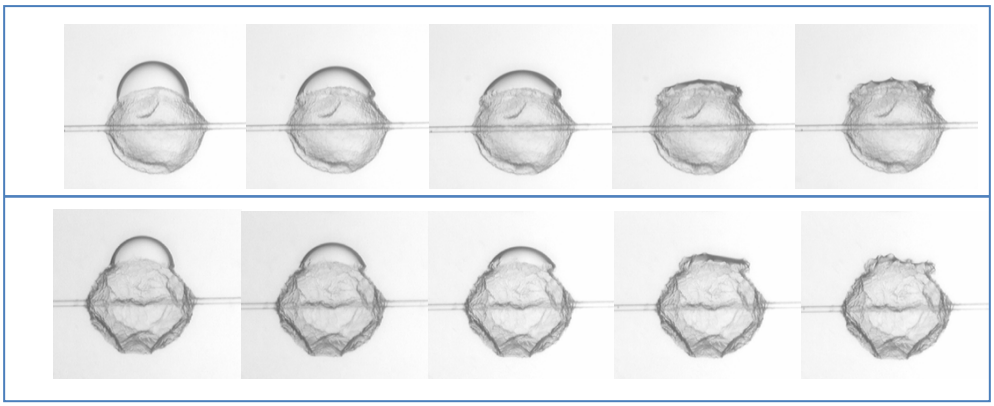


Fig. S3 Two typical repeat experiments at lower subcooling (0.7 ºC). The images also suggest the distinct hydrate morphology. The diameter of the glass fiber in the images is 38μm. Repeat 1 (top), t=3820 s; repeat 2 (bottom), t=2700 s. Time increases from left to right.

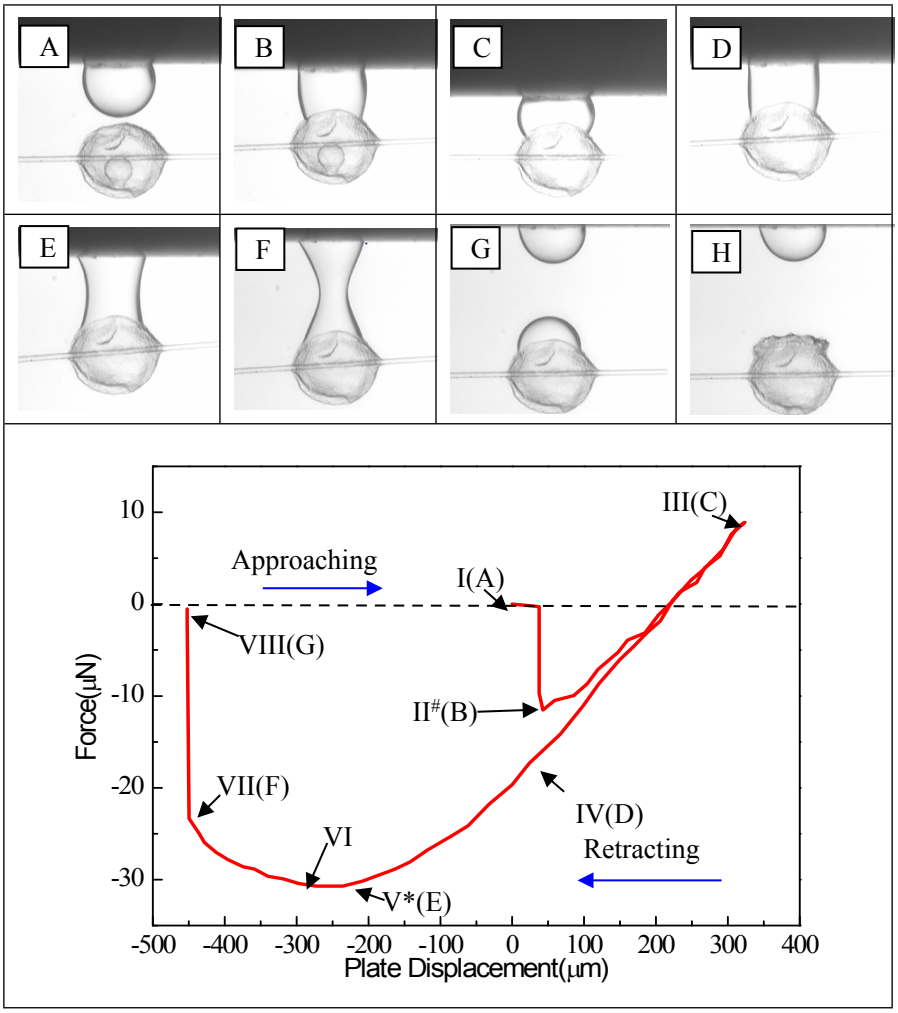


Fig. S4 Typical hydrate particle-droplet interaction force profile and corresponding microscope images of a measurement cycle in pure CyC5 at 7 ºC. II: initial interaction force; III: preload force; V: maximum interaction force; VII: rupture force. II-VII: t=257 s. The droplet and particle are allowed to rest under constant preload force for 180 s before retracting. G-H, t=3820 s.

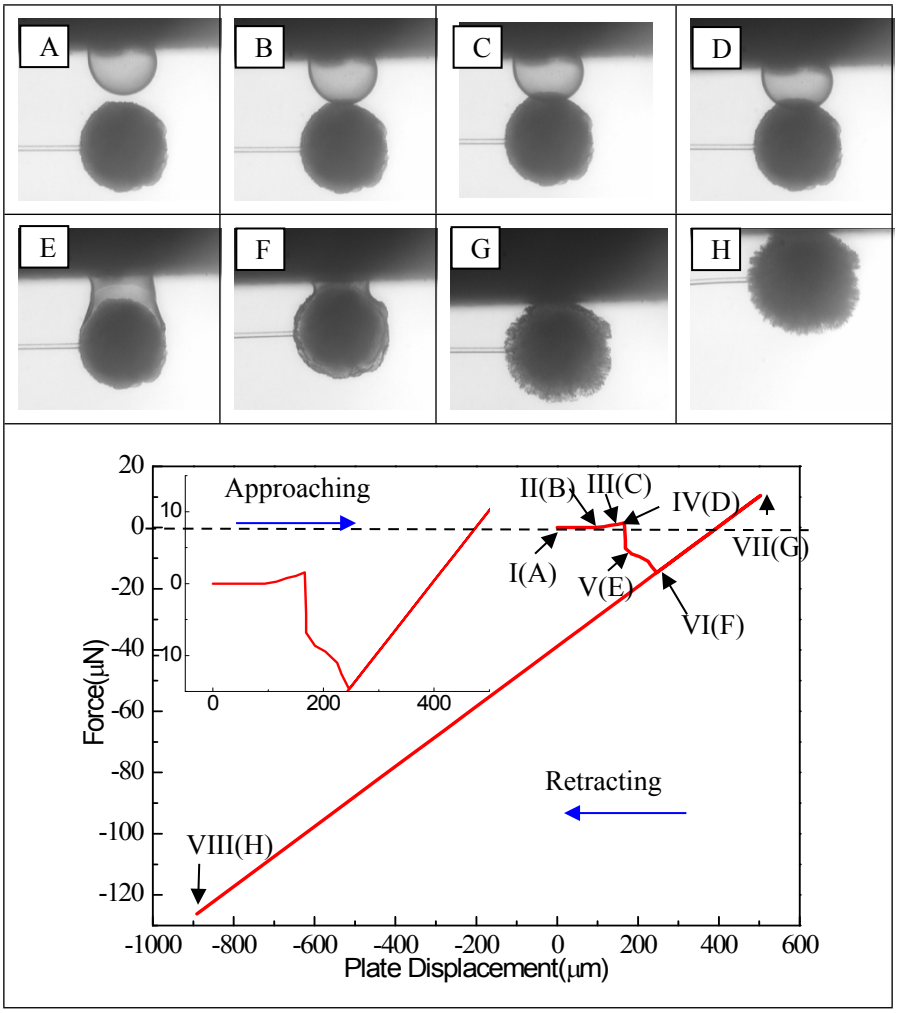


Fig. S5 Typical hydrate particle-droplet interaction force profile and corresponding microscope images of one measurement cycle in CyC5 with 1 wt.% Span80 at 1.5ºC. II: droplet-hydrate particle initial contact; IV: droplet rupture; V: initial interaction force; VIII: preload force; II-VIII: t=97 s. In this case the hydrate sinters on the surface and therefore cannot be separated.

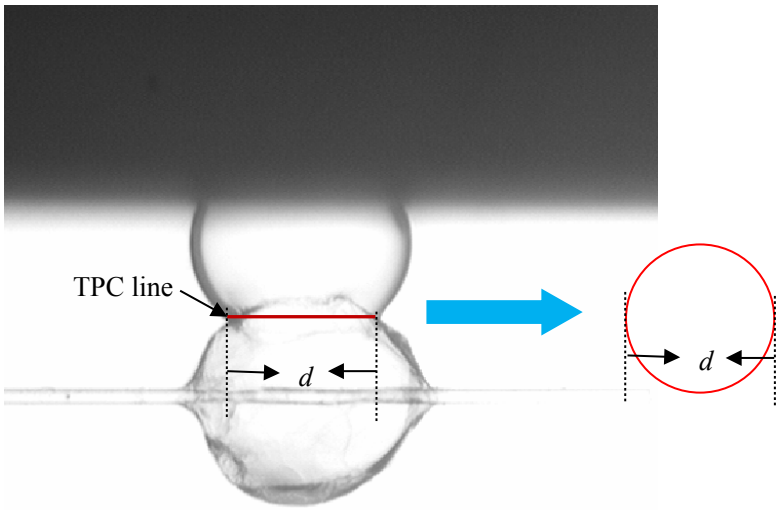


Fig. S6 Schematic of the contact area, contact area $= 0.25 \times \pi \times d^2$

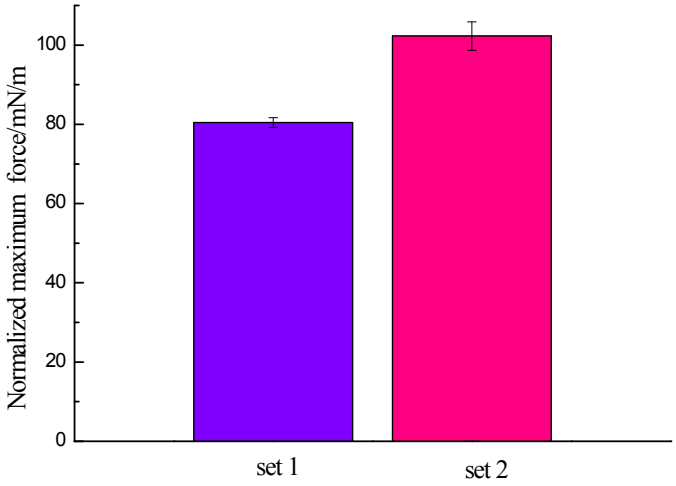


Fig. S7 Two typical normalized maximum interaction forces between CyC5 hydrate particle and droplet (no hydrate formed during the measurement). For every set, the value is based on 5 measurements. The coefficients of variation of the two sets are 1.52%, 3.59%, respectively.

Reference

1 E. D. Sloan and C. A. Koh, Clathrate Hydrates of Natural Gases. 3rd Ed., CRC Press-Taylor & Francis Group, Boca Raton, FL. 2007.