# Effect of Guest Size on the Mechanical Properties and Molecular Structure of Gas Hydrates from First-Principles

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## SI 1. Polycrystalline elastic properties

Shear modulus:

$$G_{Reuss} = \frac{5(c_{11}-c_{12})c_{44}}{4c_{44}+3(c_{11}-c_{12})} \quad \text{(S1)}$$

$$G_{Voigt} = \frac{c_{11}-c_{12}+3c_{44}}{5} \quad \text{(S2)}$$

$$G = \frac{G_{Reuss}+G_{Voigt}}{2} \quad \text{(S3)}$$

Bulk modulus:

$$B = B_{Reuss} = B_{Voigt} = \frac{c_{11}+2c_{12}}{3} \quad \text{(S4)}$$

Poisson's ratio:

$$\nu = \frac{\frac{3}{2}B-G}{G+3B} \quad \text{(S5)}$$

Young's modulus:

$$E = 2G(1+\nu) \quad \text{(S6)}$$

Compressional (longitudinal) wave velocity:

$$V_p = \sqrt{\frac{B+\frac{4}{3}G}{\rho}} \quad \text{(S7)}$$

Shear (transverse) wave velocity:

$$V_s = \sqrt{\frac{G}{\rho}} \quad \text{(S8)}$$

Zener anisotropy factor:

$$A_z = \frac{2c_{44}}{c_{11}-c_{12}} \quad \text{(S9)}$$

Every anisotropy factor:

$$A_e = \frac{c_{11}-c_{12}-2c_{44}}{c_{11}-c_{44}} \quad \text{(S10)}$$

## SI 2. Energy-volume plots

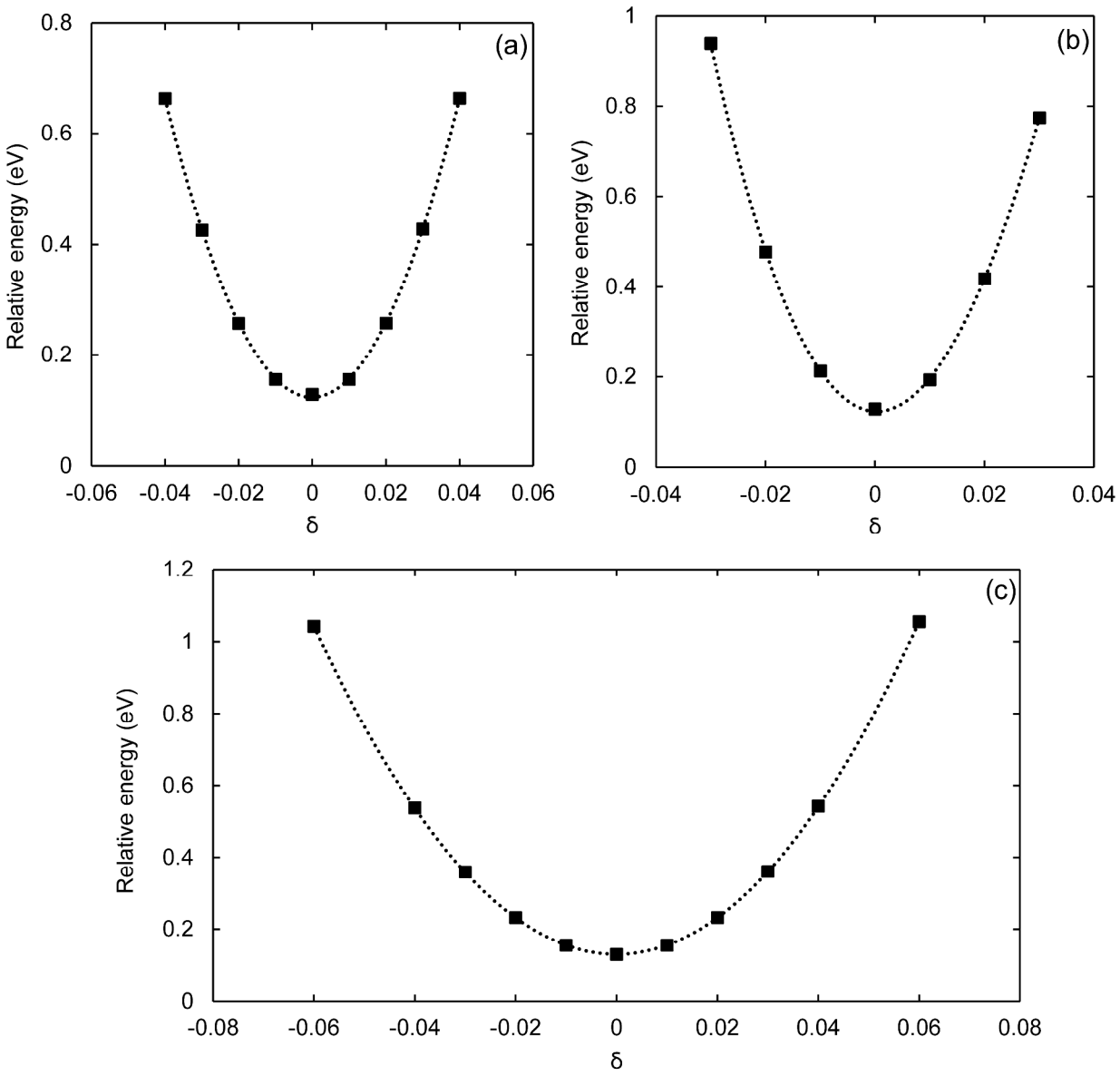


Figure S1. Change in system energy as a function of the strain $\delta$ for sII empty hydrate. a) volume-conserving tetragonal strain $\varepsilon_1$, b) 110 strain, and c) 111 shear strain.

# SI 3. Gas hydrate atomic structure analysis

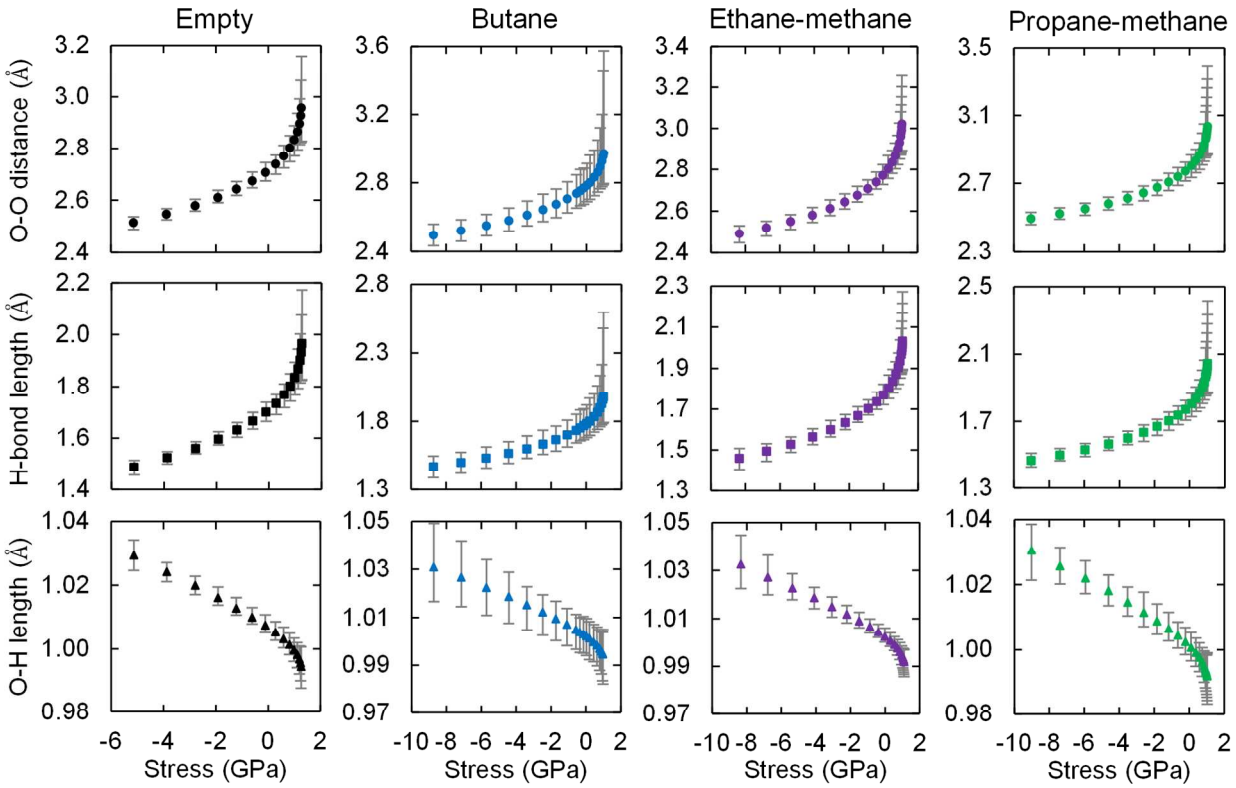


Figure S2. Bond lengths in sII gas hydrates under triaxial stress, with error bars representing the total spread of values in the unit cell lattice. The sII gas hydrates behave almost identically; under tensile stress the bond lengths reach a vertical asymptote characterized by the ideal tensile strength, while under compressive stress the H-bond length decreases and the O-H bond length increases, becoming closer in length.

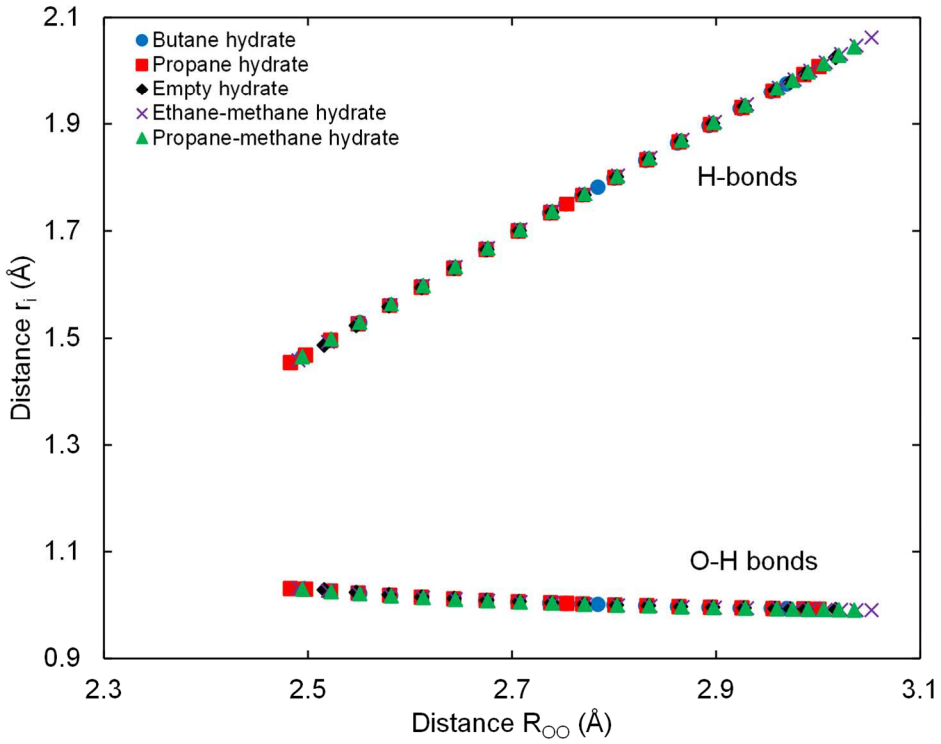


Figure S3. The dependence of the H-bond (top line) and O-H bond (bottom line) lengths on the O-O distance in all sII gas hydrates under investigation. There is no observable difference between any of the hydrates.

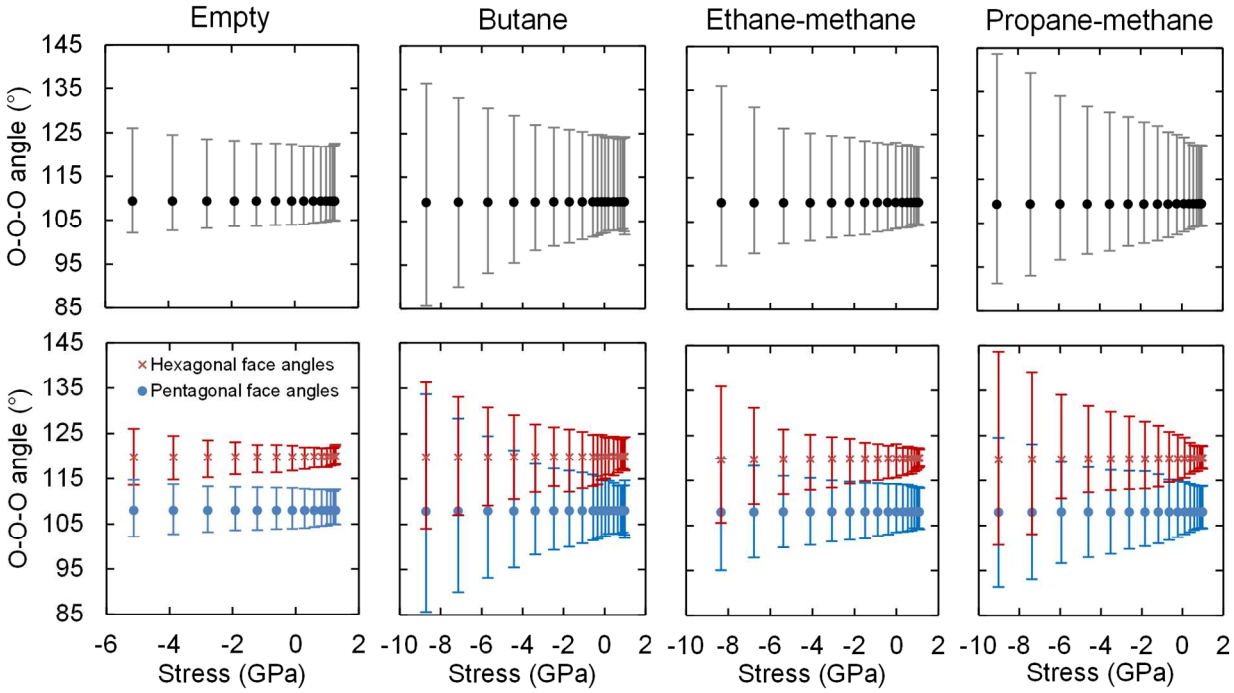

Figure S4. O-O-O angles in sII gas hydrates under triaxial stress, with error bars representing the total spread of values in the unit cell lattice. The top row shows the average values for all angles, while the bottom row shows the average values per category of angle (hexagonal, or pentagonal). While the average bond angles do not change regardless of guest size or stress magnitude, the total spread of the values increases with guest size and with increasing applied stress.