Electronic Supplementary Material (ESI) for New Journal of Chemistry.

SUPPORTING INFORMATION FOR

Tuning the stability of liquids by controlling the formation of interfacial surfactants

Xiaoying Yang[a], Zhaocan Zhong[a], Shiyuan Zhou[a], Peiyang Gu[a], Qingfeng Xu[a]*,

Jianmei Lu[a]*

[a]College of Chemistry, Chemical Engineering and Materials Science of Soochow University

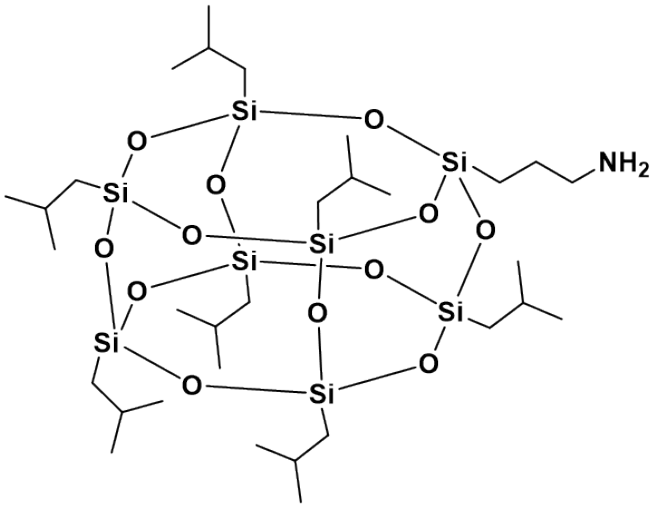

Figure S1. The chemical structure of POSS-NH_2.

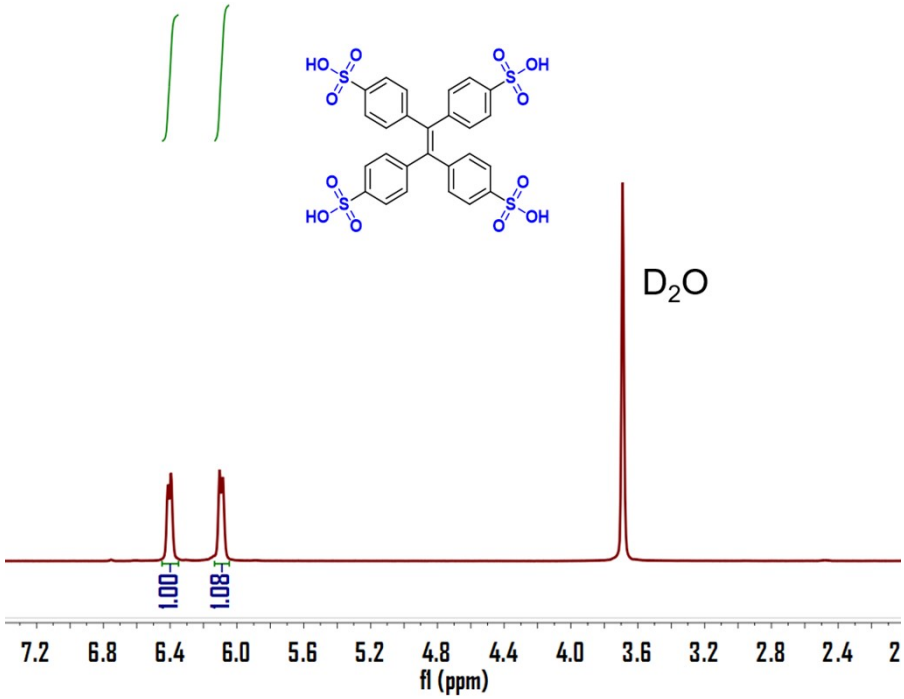

Figure S2. ^{1}H NMR (300 MHz) of TPE-SO_3H in D_2O.

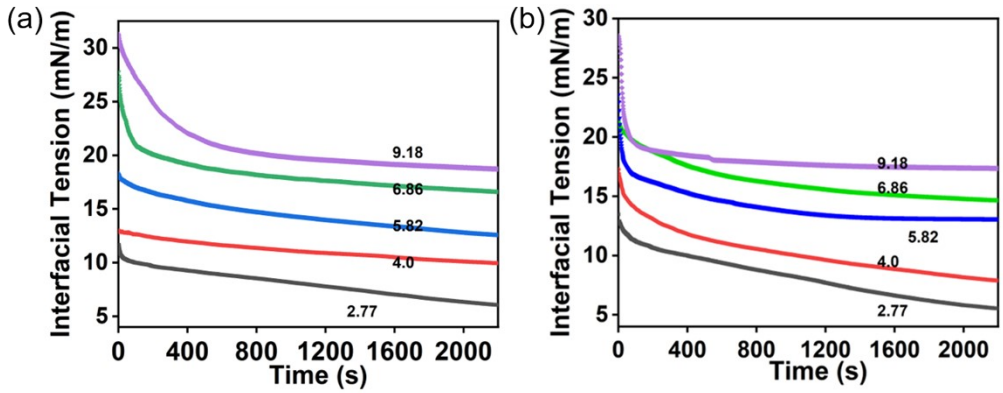

Figure S3. (a) Time evolution of the interfacial tension of Su-TPE (0.01 M) in water with different pH to POSS-NH_2 in toluene; (b) Time evolution of the interfacial tension of Su-Py (0.01 M) in water with different pH to POSS-NH_2 in toluene. $C_{POSS\text{-}NH2}$ = 1.0 g/L.

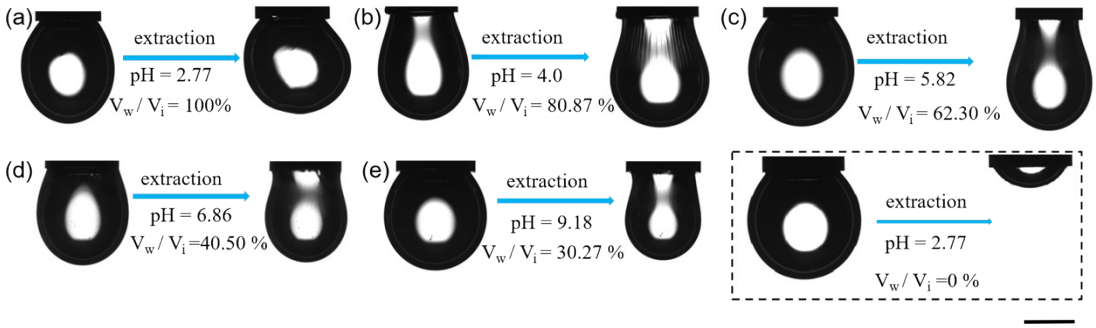

Figure S4. a-e) Changes in droplets shape of Su-Py (0.01 M) in water with different pH to POSS-NH_2 in toluene for 2600 s; f) Changes in droplets shape of Su-TPE (0.01 M, pH=2.77) in water to POSS-NH_2 in toluene for 2600 s. Scale bar: 2.0 mm.

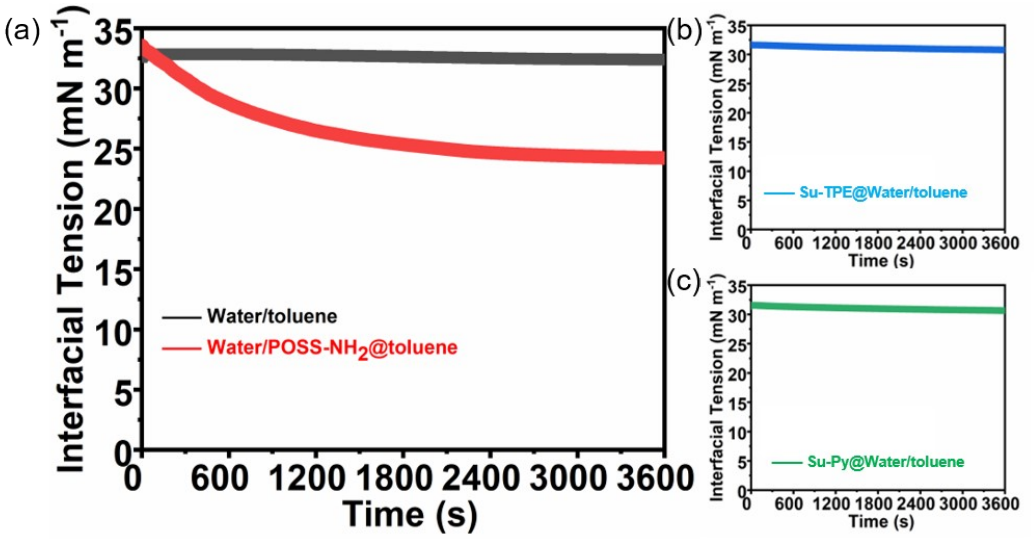

Figure S5. The time evolution of the interfacial tension of different oil-water systems. a) water/toluene and water/POSS-NH_2@toluene, b) Su-TPE@water/toluene, c) Su-Py@water/toluene ([Su-TPE] =0.01 M, [Su-Py] =0.01 M, [POSS-NH_2] =0.1 g L^{-1}).

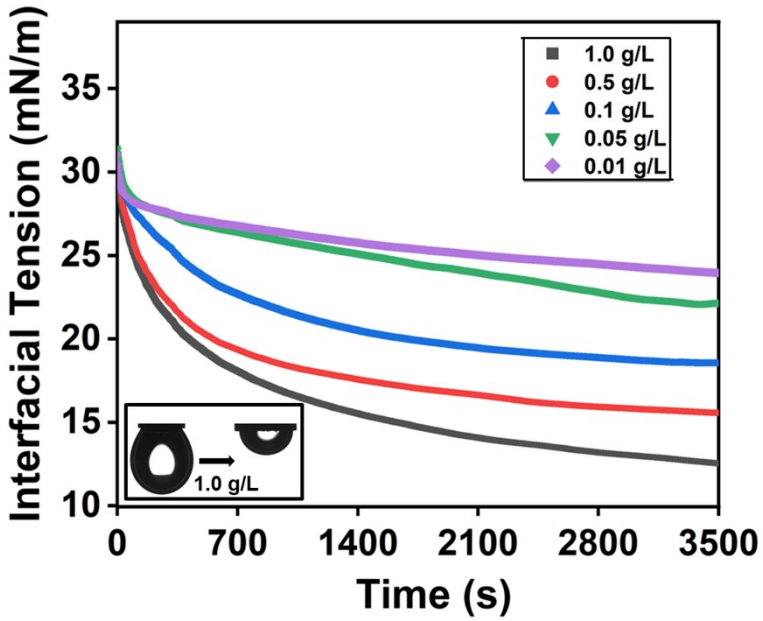

Figure S6. The time evolution of the interfacial tension of different concentrations of POSS-NH_2 in toluene to pure water (The inset shows the change in droplet shape after the assembly of pure water in 1.0 g/L POSS-NH_2 solution in toluene for 2600s).

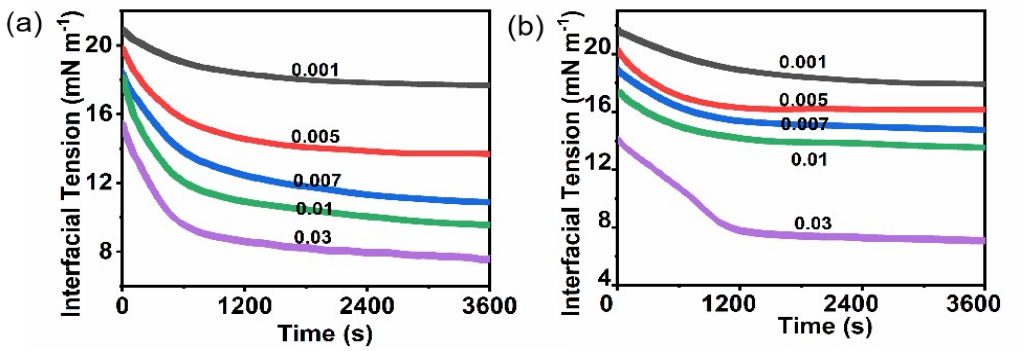

Figure S7. a) Time evolution of the toluene interfacial tension of different concentrations of Su-Py (pH = 4.0) to 0.1 g L^{-1} POSS-NH_2 in water. b) Time evolution of the toluene interfacial tension of different concentrations of Su-TPE (pH = 4.0) to 0.1 g L^{-1} POSS-NH_2 in water.

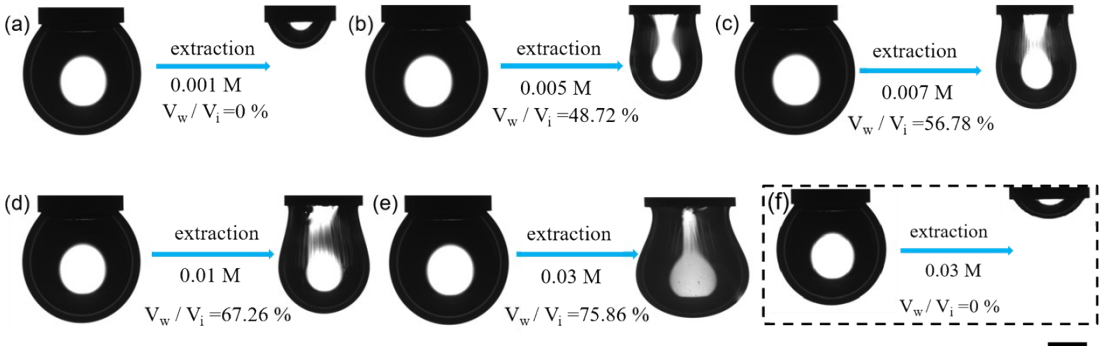

Figure S8. a-e) Changes in droplets shape of different concentrations of Su-Py assembled in the toluene phase of 0.1 g L^{-1} POSS-NH_2 for 2600 s. f) Changes in droplets shape of Su-TPE (0.03 M) assembled in the toluene phase of 0.1 g L^{-1} POSS-NH_2 for 2600 s. Scale bar: 2.0 mm.

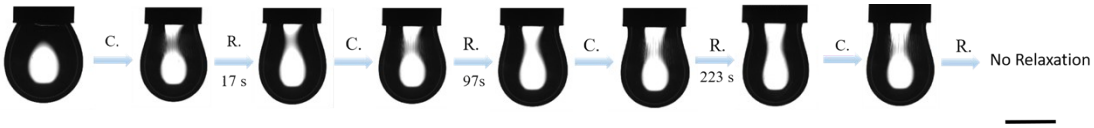

Figure S9. Snapshots of droplet's morphology evolution in an extraction- reinjection process, [Su-Py] = 0.01 M, [POSS-NH_2] =1.0 g/L, pH=4.0. Scale bar: 2.0 mm.

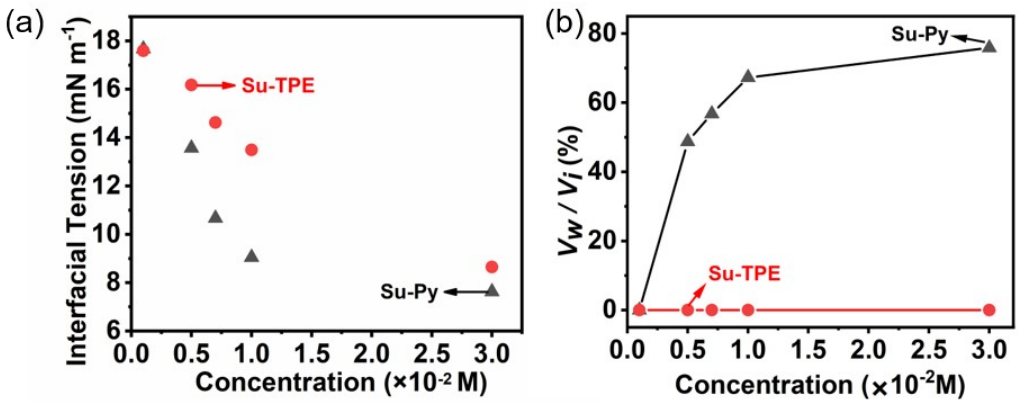

Figure S10. a) Equilibrium interfacial tension in water solution with different concentrations of Su-Py or Su-TPE ([POSS-NH_2] =1.0 g L^{-1}). b) Coverage rate of different concentrations of Su-Py or Su-TPE in water solution ([POSS-NH_2] =1.0 g L^{-1}).

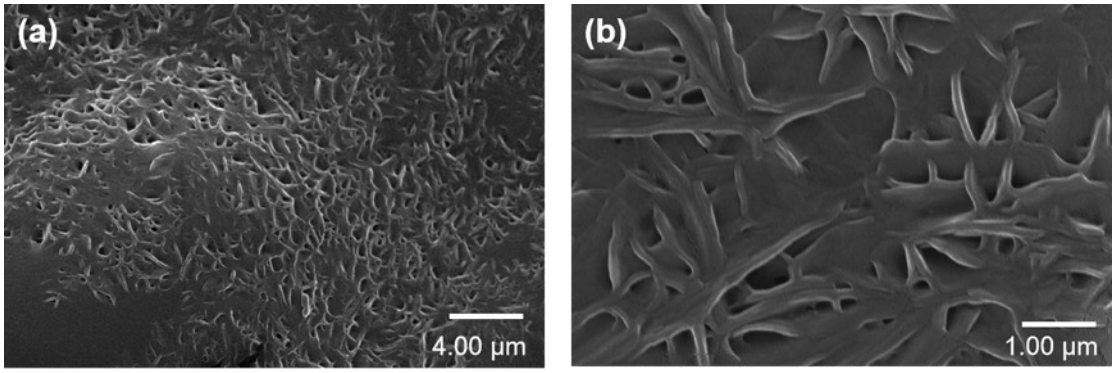

Figure S11. The SEM image of the film assembled by Su-Py (0.01 M, pH=4.0) and POSS-NH_2 (1.0 g/L). a) 4.00 μm; b) 1.00 μm.

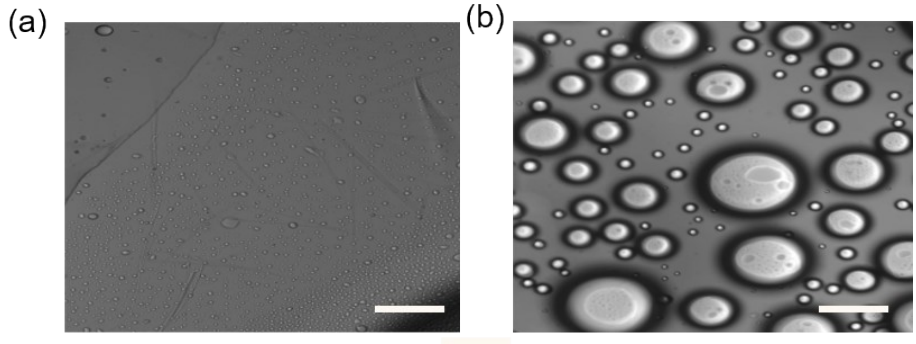

Figure S12. a) Confocal fluorescence of 0.01 M Su-Py in 1.0 g L^{-1} POSS-NH_2 mesitylene ($V_{Su\text{-}Py}$: $V_{POSS\text{-}NH2}$=1:1) in the bright field. b) Confocal fluorescence of 0.01 M Su-TPE in 1.0 g L^{-1} POSS-NH_2 mesitylene ($V_{Su\text{-}TPE}$: $V_{POSS\text{-}NH2}$=1:1) in the bright field. Scale bar: 75 μm.

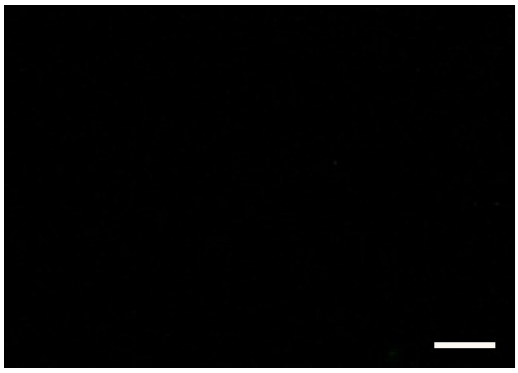

Figure S13. Confocal fluorescence of 0.01 M Su-TPE in 1.0 g L^{-1} POSS-NH_2 mesitylene ($V_{Su\text{-}TPE}$: $V_{POSS\text{-}NH2}$=1:1) in the dark field. Scale bar: 75 μm.

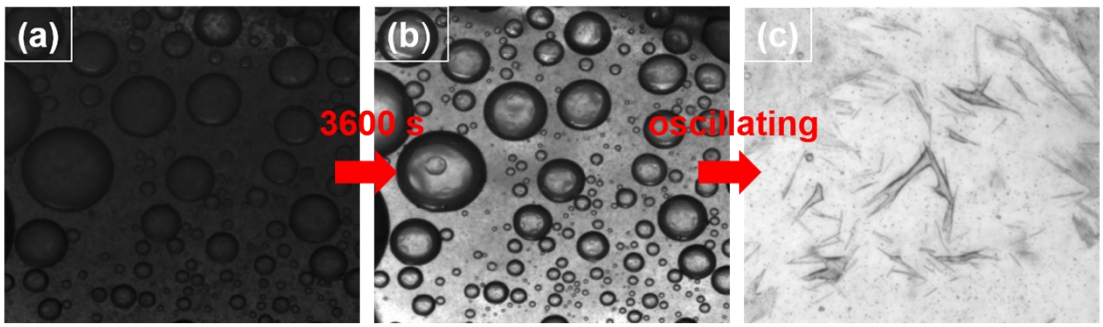

Figure S14. Optical microscopy images of Su-Py and POSS-NH_2 interface assembly. (a) At the beginning of the assembly; (b) The assembly after 3600 s; (c) Oscillation the interface. Scale bar: 0.5 mm.

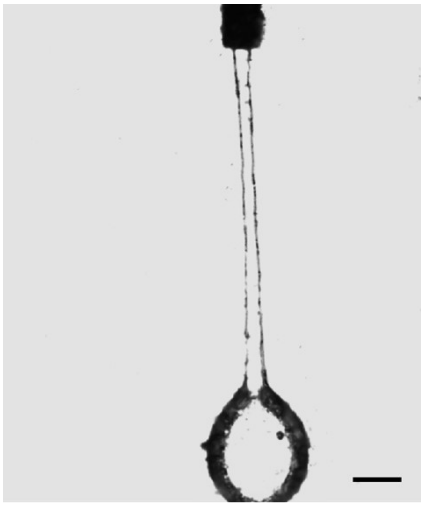

Figure S15. Manual 3D printing of Su-Py (0.01M, pH = 4.0) in 10 mg/mL POSS-NH_2 in silicone oil (10000 cst) solution. Needle Diameter: 0.5 mm, flow rate: 3 uL/s. Scale bar: 0.5 mm.

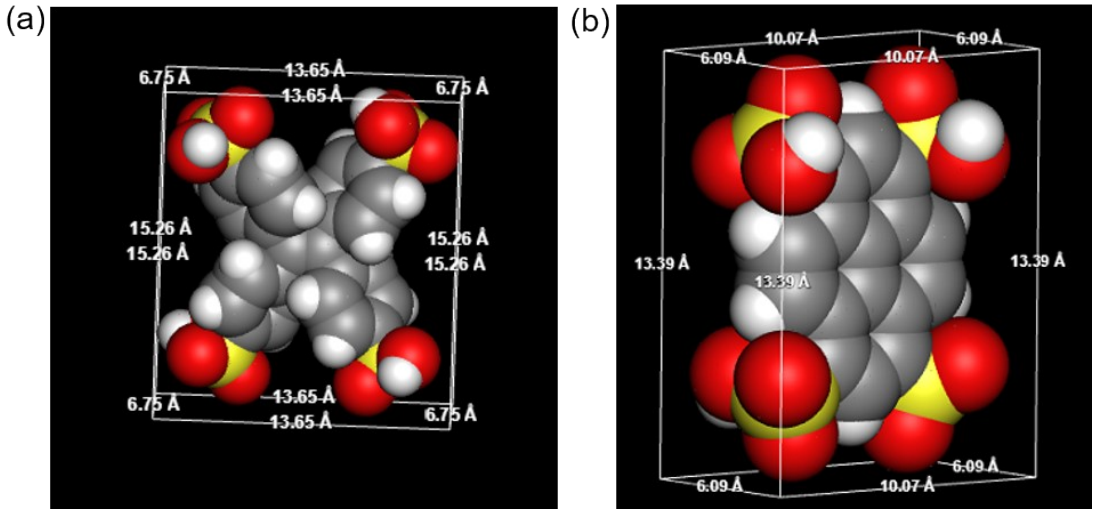

Figure S16. Molecular simulation structure of the Su-TPE (a) / Su-Py (b) showing the dimensions of the molecules.

Note: Draw the structural formula of the molecule in Chemdraw, and then determine the three-dimensional coordinates of each atom in the entire molecule. Then calculate the radius of the molecule according to the calculation method of radius summed up by Hu et al[1] in 2010.

The method of calculation refers to reference[2-3]

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(1) Hu, S, Z.; Xie Z X.; Zhou, Z, H. 70 Years of Crystallographic van der Waals Radii. *Acta Phys. Chim. Sin*, **2010**, 26(07): 1795-1800.

(2) Eigenvalue algorithm

(3) Numerical diagonalization of 3x3 matrices