Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Tetrahedral Crosslinking of Dia-type Nets into Zeolitic GIS-type Framework for Optimizing Stability and Sorption

Yan-Xi Tan,[a] Yanan Si,[a,b] Wenjing Wang[a] and Daqiang Yuan*,[a]

a. State Key Laboratory of Structural Chemistry, Fujian Institute of Research on the Structure of Matter, Chinese Academy of Sciences, Fuzhou, 350002, Fujian, China. E-mail: ydq@fjirsm.ac.cn

b. University of the Chinese Academy of Sciences, Beijing, 100049, China.

1. Materials and general methods

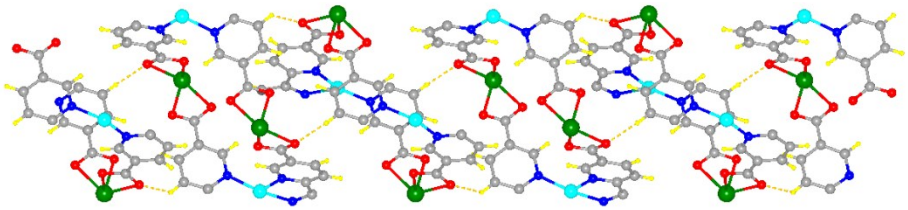

Fig. S1. Hydrogen bonds between adjacent diamondoid cages.

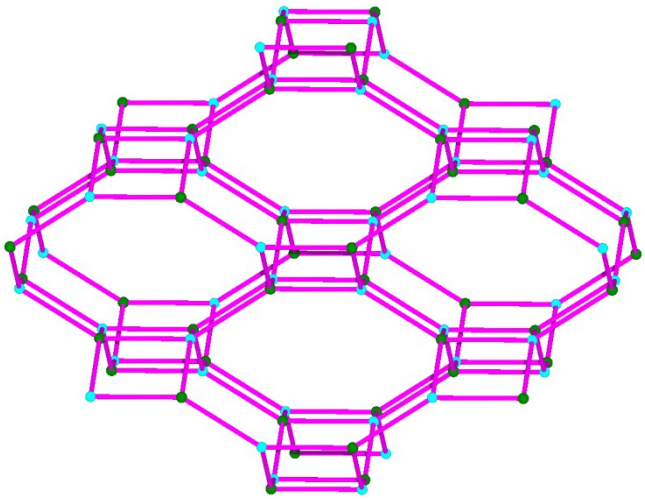

Fig. S2. GIS topology of **FJI-Y3**.

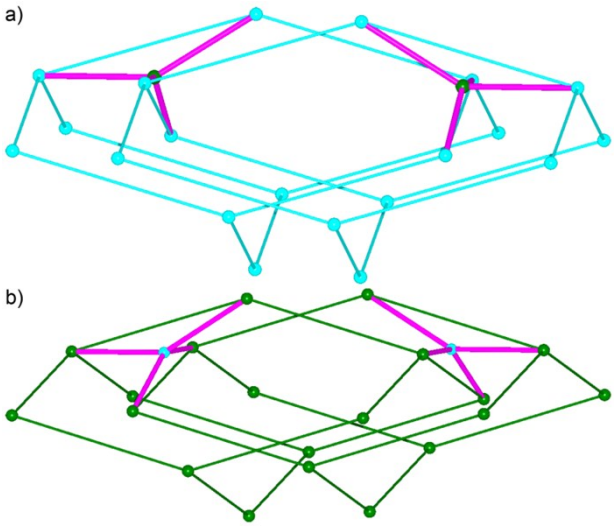

Fig. S3. Tetrahedral crosslinking of 4-fold interpenetrating *dia*-type nets.

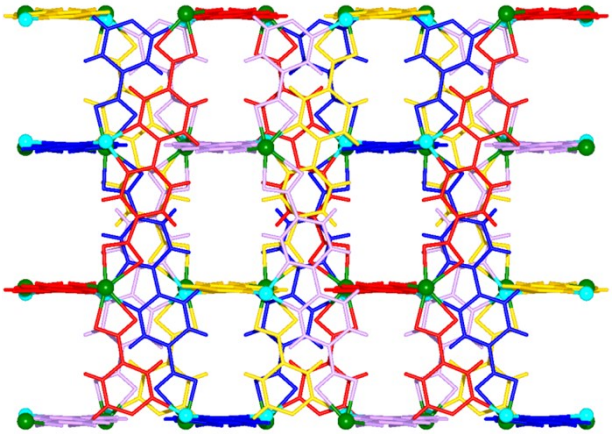

Fig. S4. The 3D interpenetrating framework of **FJI-Y3** viewing along *a* or *b* axis.

2. TGA curves

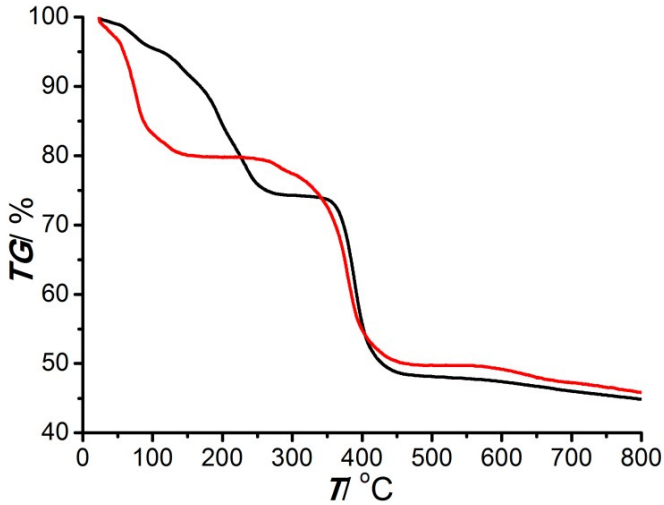

Fig. S5. TGA curves of as- synthesized **FJI-Y3** (black line) and MeOH-exchanged **FJI-Y3** (red line) samples.

3. Sorption isotherms

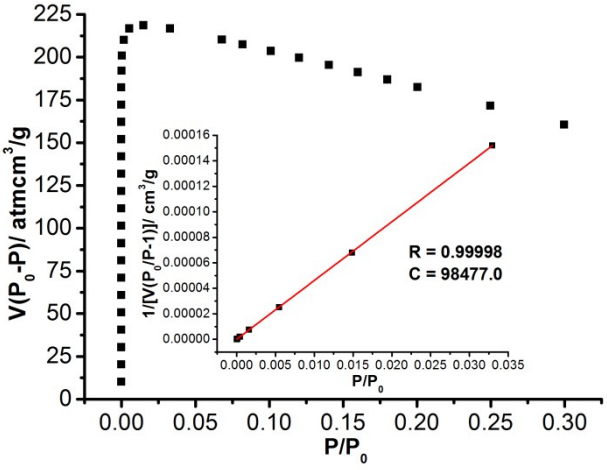

Fig. S6. BET surface area plot for **FJI-Y3-ht**.

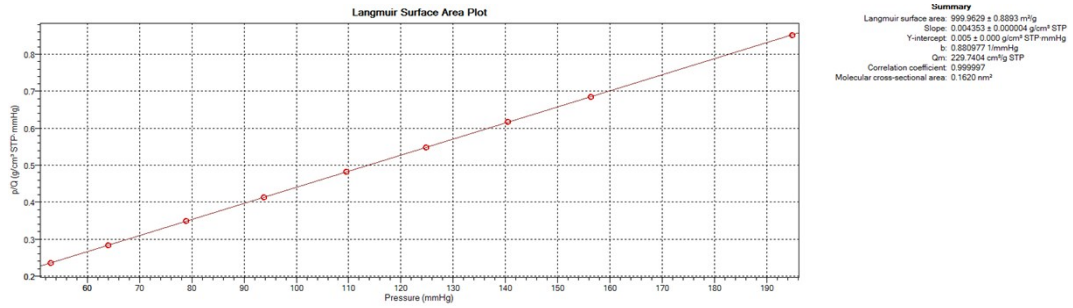

Fig. S7. Langmuir surface area plot for **FJI-Y3-ht**.

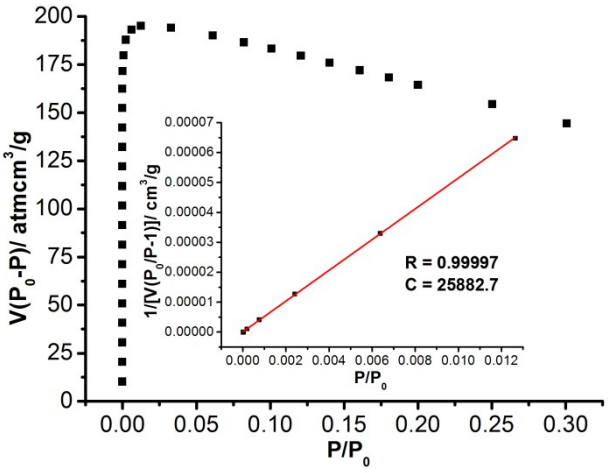

Fig. S8. BET surface area plot for **FJI-Y3-ht´**.

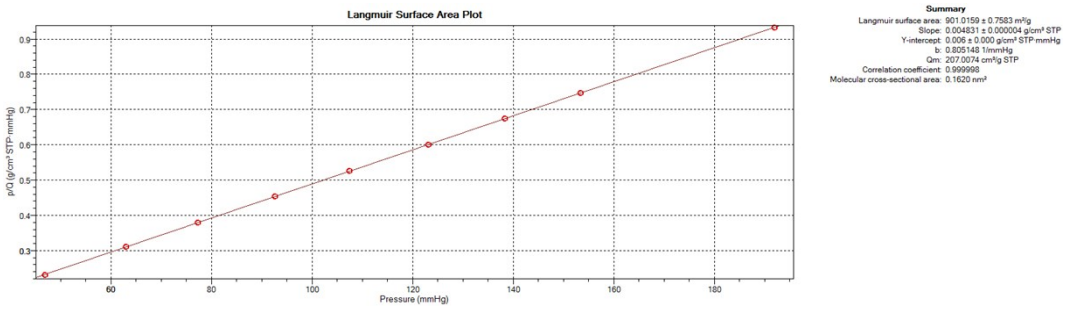

Fig. S9. Langmuir surface area plot for **FJI-Y3-ht´**.

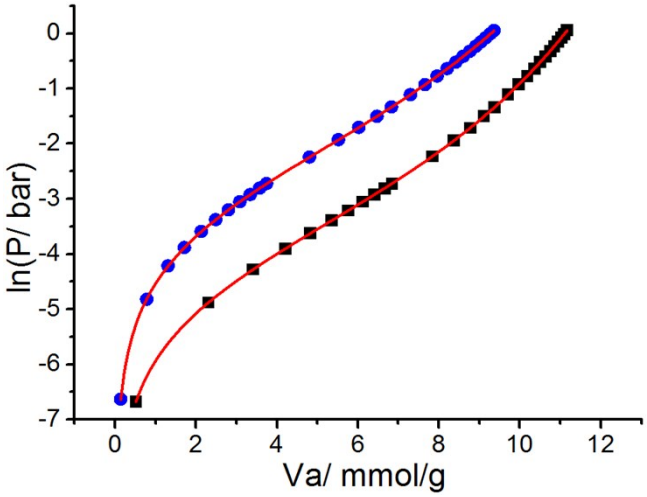

Fig. S10. D_2 adsorption isotherms under 77 (black) and 87 K (blue) for **FJI-Y3-ht** fitting by virial method.

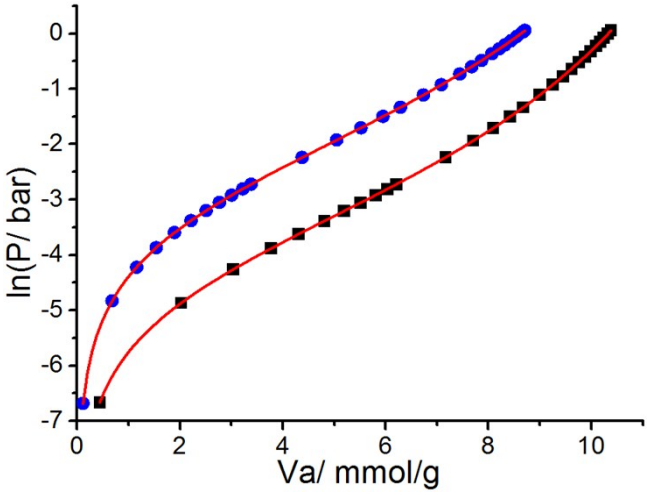

Fig. S11. H_2 adsorption isotherms under 77 (black) and 87 K (blue) for **FJI-Y3-ht** fitting by virial method.

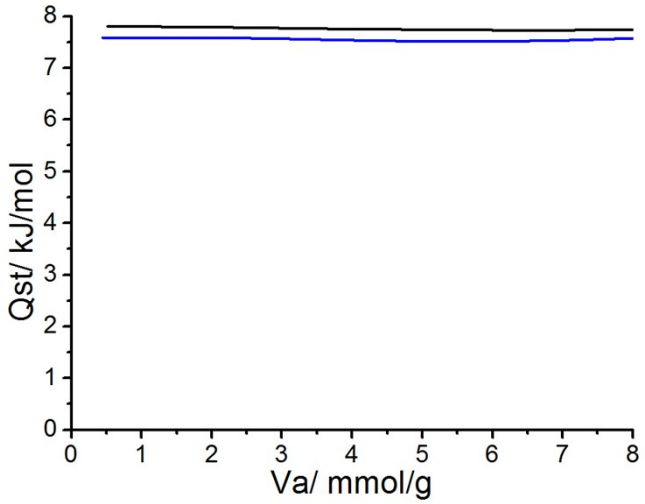

Fig. S12. The isosteric heat of D_2 (black) and H_2 (blue) adsorption for **FJI-Y3-ht** estimated by the virial equation.

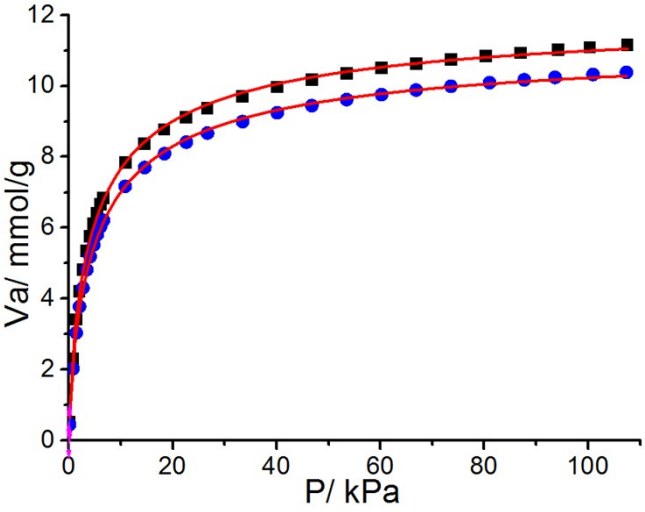

Fig. S13. Adsorption isotherms of D_2 (black) and H_2 (blue) for **FJI-Y3-ht** under 77 K. Solid lines through the experimental data are fits to the single-site L-F model.

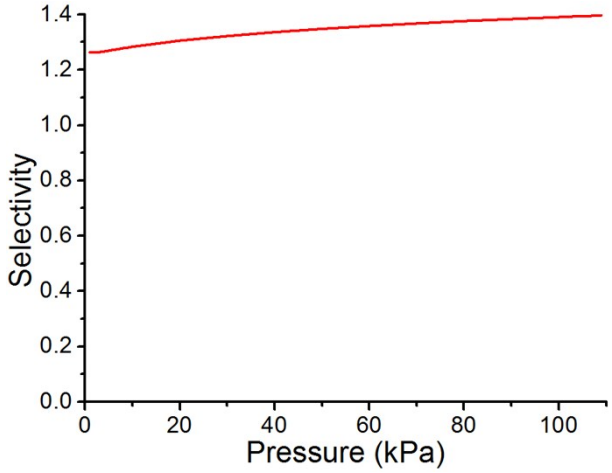

Fig. S14. Selectivity of D_2/H_2 for **FJI-Y3-ht** under 77 K.

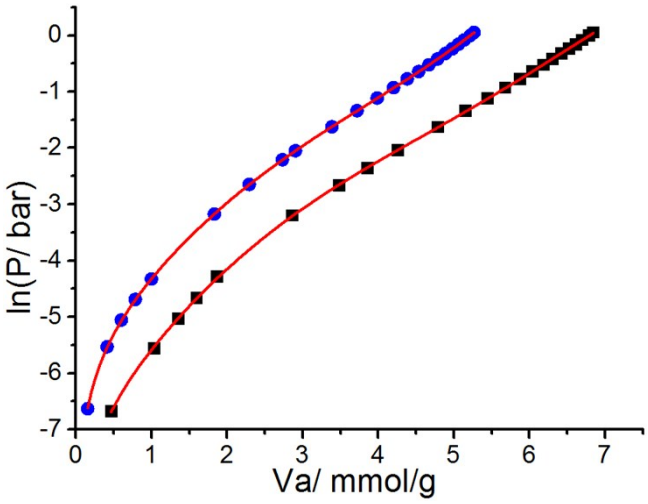

Fig. S15. C_2H_2 adsorption isotherms under 273 (black) and 298 K (blue) for **FJI-Y3-ht** fitting by virial method.

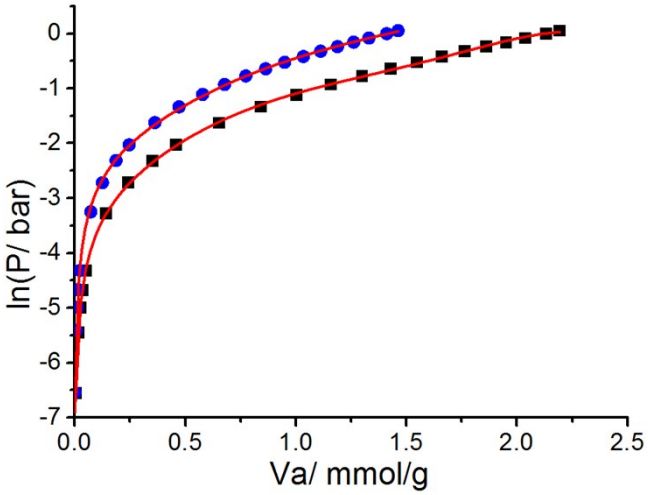

Fig. S16. CH_4 adsorption isotherms under 273 (black) and 298 K (blue) for **FJI-Y3-ht** fitting by virial method.

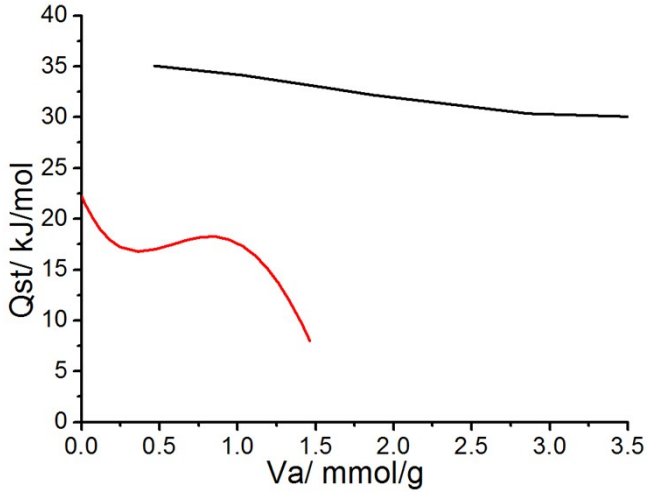

Fig. S17. The isosteric heat of C_2H_2 (black) and CH_4 (blue) adsorption for **FJI-Y3-ht** estimated by the virial equation.

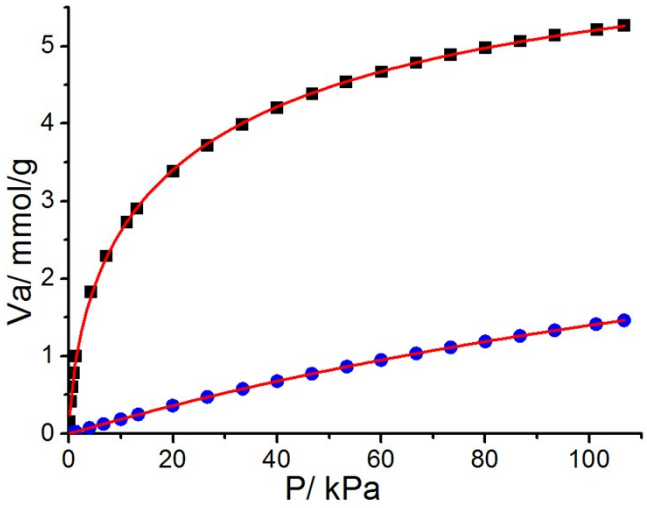

Fig. S18. Adsorption isotherms of C_2H_2 (black) and CH_4 (blue) for **FJI-Y3-ht** under 298 K. Solid lines through the experimental data are fits to the single-site L-F model.

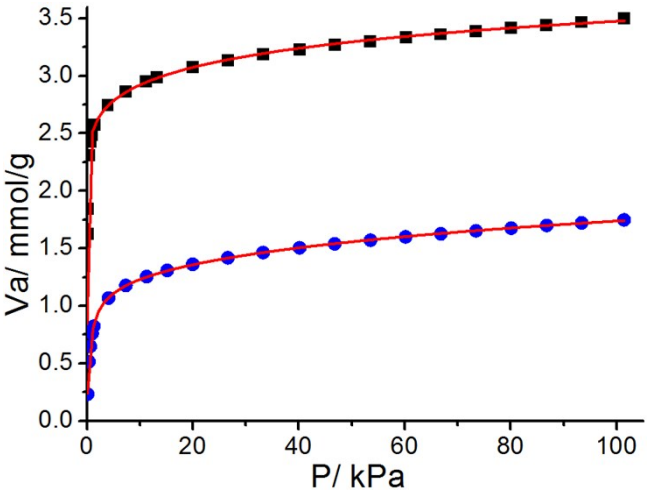

Fig. S19. Adsorption isotherms of *n*-C_4H_{10} (black) and *i*-C_4H_{10} (blue) for **FJI-Y3-ht** under 298 K. Solid lines through the experimental data are fits to the dual-site L-F model.