## **Supporting Information**

## The Intramolecular H-bonding Effects on the Growth and Stability of Schiff-base Surface Covalent Organic Frameworks

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Fig. S1 Large scale STM images of SCOF-1 on HOPG obtained by heating 120 °C for 3h without Ar protection . Imaging conditions:  $V_{bias} = 700 \text{ mV}$ ,  $I_t = 500 \text{pA}$ .

**Table S1** Structural models of conceivable networks obtained by reaction between TAPB and 2,3-DHTPA due to the asymmetrically substitution of hydroxyl functional group and the corresponding adsorption energy by Forcite using Dreiding force field.

Simplified schematic diagram				
Structural models of conceivable networks				
Adsorption energy	312.53 kcal/mol	316.93 kcal/mol	317.54 kcal/mol	318.99 kcal/mol



**Fig. S2** Large scale and high resolution STM images of SCOFs synthesized by TAPB and terephthalaldehyde in 110 °C. (a, b) without Ar protection. (c, d) in Ar atmosphere. Imaging conditions:  $V_{bias} = 693$  mV,  $I_t = 495$  pA.



**Fig. S3** The stability study of SCOF-2. (a) The pristine STM image of generated network; (b) The STM image of network after 1 mol/L HCl treatment; (c) The STM images of network after 0.01 mol/L HCl treatment; (d) The STM image of network after 0.01 mol/L KOH treatment. Imaging conditions: (a)  $V_{bias} = 630 \text{ mV}$ ,  $I_t = 550 \text{ pA}$ ; (b)  $V_{bias} = 700 \text{ mV}$ ,  $I_t = 500 \text{ pA}$ ; (c)  $V_{bias} = 414 \text{ mV}$ ,  $I_t = 546 \text{ pA}$ ; (d)  $V_{bias} = 456 \text{ mV}$ ,  $I_t = 546 \text{ pA}$ .