

## Supplementary Material

# Mesoporous TiO<sub>2</sub>-SiO<sub>2</sub> adsorbent: ultra-deep desulfurization of organic-S at room temperature and atmospheric pressure

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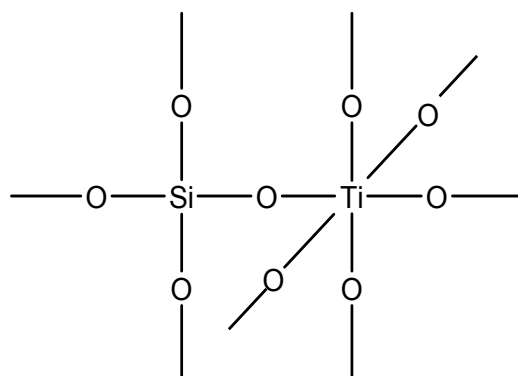
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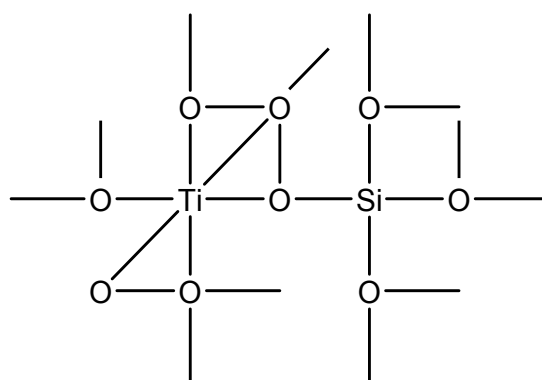
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**Section S1:** Tanabe's hypothesis



(a) charge difference =  $(+4/6-2/2) * 6 = -2$



(b) charge difference =  $(+4/4-2/3) * 4 = +4/3$

**Fig. S1.** Model structures of  $\text{TiO}_2\text{-SiO}_2$  and  $\text{SiO}_2\text{-TiO}_2$  pictured according to Tanabe's hypothesis.

(a) with  $\text{SiO}_2$  as the major oxide; (b) with  $\text{TiO}_2$  as the major oxide.

The  $\text{SiO}_2$  is major component oxide in the structure of  $\text{TiO}_2\text{-SiO}_2$ , which is shown in Figure.S1 (a). According to Tanabe's hypothesis the charge difference for each bond is  $(+4/6-2/2) = -1/3$ , and for all the bonds the valence unit is  $(+4/6-2/2) * 6 = -2$ . In this case, the Brønsted acidity is assumed to appear in the presence of an excess of the negative charge, because three protons are considered to be associated with six oxygens to keep the electric neutrality. Although the  $\text{TiO}_2\text{-SiO}_2$  adsorbent exhibits big specific surface area and showed good performance, the  $\text{TiO}_2\text{-SiO}_2$  binary oxide itself exhibits the Brønsted acidity or Lewis base which is not beneficial for adsorbing sulfur at low temperatures. However, if the binary oxide mode was changed into  $\text{TiO}_2\text{-SiO}_2$  with  $\text{TiO}_2$  being the major component, the result will be contrary for

effect. The structure of  $\text{TiO}_2\text{-SiO}_2$  with  $\text{TiO}_2$  being the major component oxide is shown in Figure.S1 (b). According to Tanabe's hypothesis, the charge difference for each bond is  $(+4/4-2/3)=+1/3$ , and for all the bonds the valence unit of  $(+4/4-2/3)*4=+4/3$  is excess. In this case, the Lewis acidity is assumed to appear in the presence of an excess of the positive charge. Thereby the adsorption capacity of thiophene sulfurs and their derivatives would be significantly enhanced at low temperatures owing to high specific surface and rich Lewis acidity of the mesoporous  $\text{TiO}_2\text{-SiO}_2$  binary oxide.

### Section S2: The effect of preparation method on desulfurization performance

Figure.S2 give a clear comparison of the desulfurization efficiencies of Ti-Si-40 adsorbent by different preparation methods in the same conditions. As is can be seen, the titania and silica powder adsorbent cannot meet the deep desulphurization requirement by simple extrusion, confirmed that  $\text{TiO}_2\text{-SiO}_2$  complex oxides can generated adsorption sites by sol-gel.

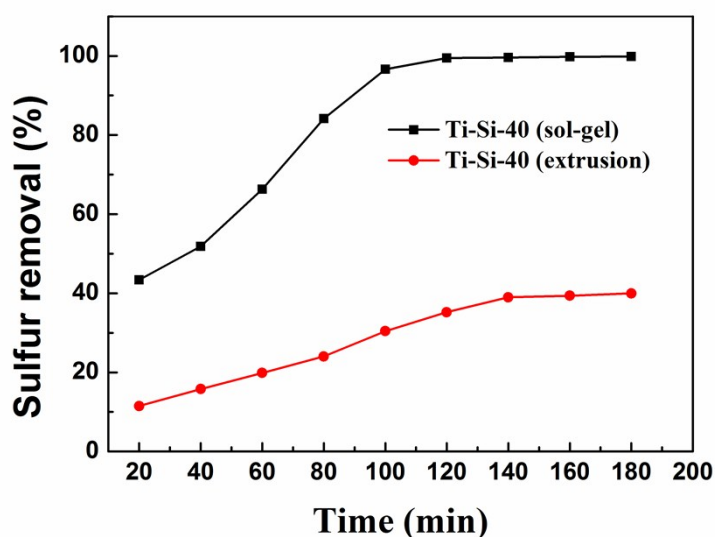


Fig. S2. Adsorption efficiency of Ti-Si-40 adsorbent by different preparation methods