

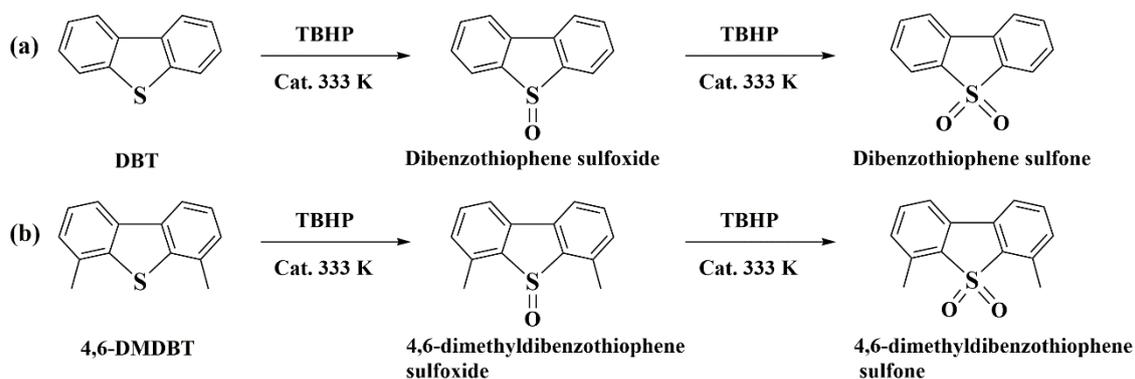
# Supplementary Materials: Synthesis of Hierarchical Titanium Silicalite-1 Using a Carbon-Silica-Titania Composite from Xerogel Mild Carbonization

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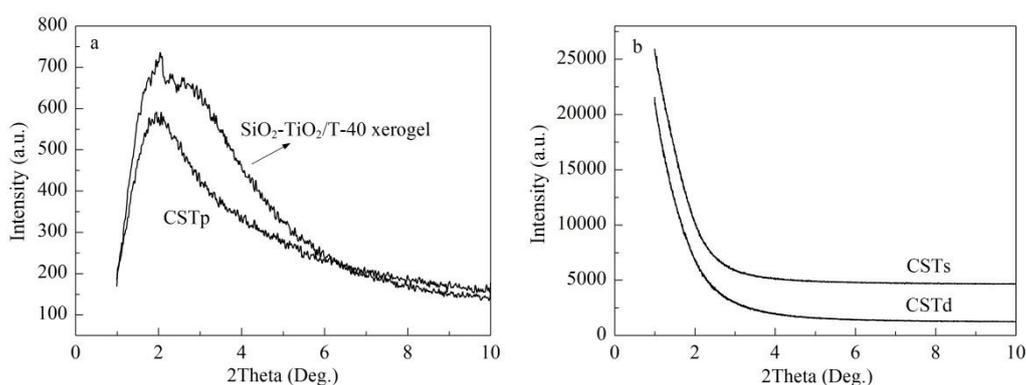
**Table S1.** Textural properties of carbon materials from different parent materials.

| Sample <sup>a</sup>          | S <sub>BET</sub><br>(m <sup>2</sup> g <sup>-1</sup> ) | S <sub>micro</sub><br>(m <sup>2</sup> g <sup>-1</sup> ) | S <sub>ext</sub><br>(m <sup>2</sup> g <sup>-1</sup> ) | V <sub>micro</sub><br>(cm <sup>3</sup> g <sup>-1</sup> ) | V <sub>meso</sub><br>(cm <sup>3</sup> g <sup>-1</sup> ) |
|------------------------------|---|---|---|--|---|
| C from CSTd                  | 535.8   | 202.8   | 333.0   | 0.10   | 0.13  |
| C from as-synthesized HTS-1d | 508.1   | 141.9   | 366.2   | 0.07   | 0.42  |
| C from CSTp                  | 279.4   | 80.6  | 198.8   | 0.04   | 0.24  |
| C from CSTs                  | 976.2   | 143.9   | 832.3   | 0.07   | 1.80  |
| C from as-synthesized HTS-1s | 905.7   | 75.1  | 830.6   | 0.03   | 1.84  |

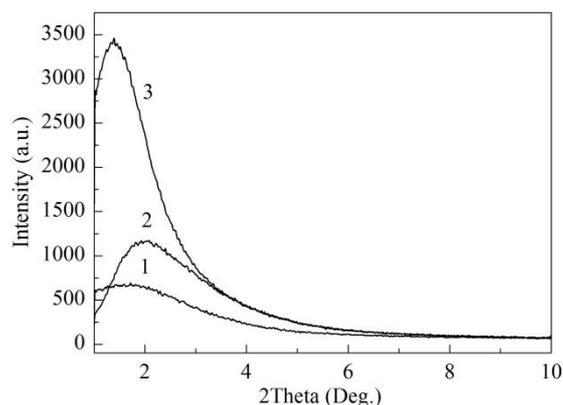
<sup>a</sup> Carbon materials obtained by removals of silica and titania from corresponding parent materials using hydrofluoric acid aqueous solution.



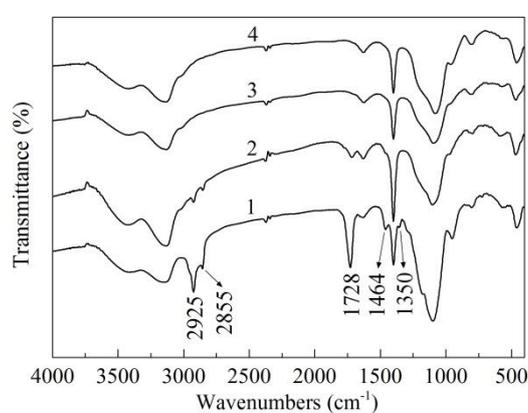
**Scheme S1.** The reaction pathway for oxidative desulfurization of DBT and 4,6-DMDBT over HTS-1s.



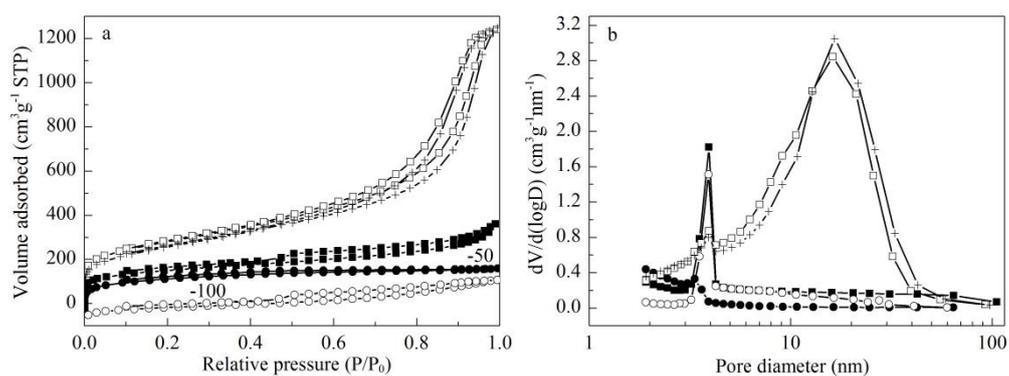
**Figure S1.** XRD patterns of samples: (a) ST/T-40 xerogel and CST composite prepared by pre-treating the ST/T-40 xerogel with dilute sulfuric acid (CSTp); (b) CST composites prepared by carbonization of ST/T-40 xerogels with (CSTs) or without (CSTd) the aid of sulfuric acid.



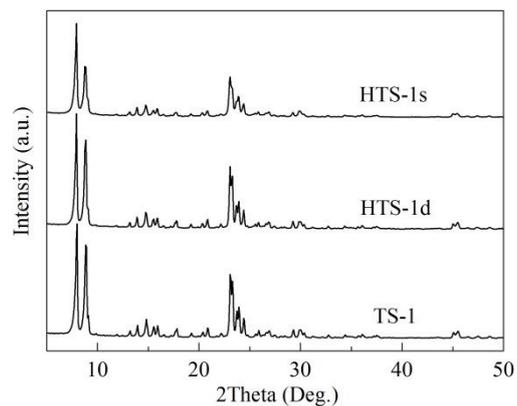
**Figure S2.** XRD patterns of SiO<sub>2</sub>-TiO<sub>2</sub> composites obtained by combustion of ST/T-40 xerogel (1), CSTd (2), and CSTs (3) in air at 823 K for 5 h.



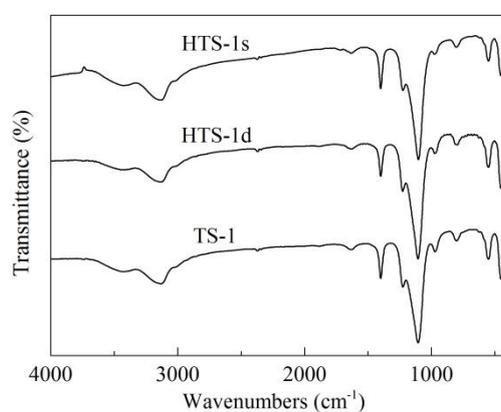
**Figure S3.** FT-IR spectra of ST/T-40 xerogel and CST composites: (1) ST/T-40 xerogel; (2) CSTp; (3) CSTs; (4) CSTd.



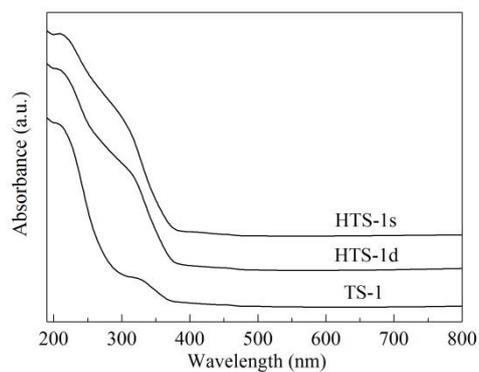
**Figure S4.** Nitrogen adsorption-desorption isotherms (a) and pore size distributions (b) of carbon materials obtained by removals of silica and titania from different parent materials with hydrofluoric acid aqueous solution: (●) carbon material from CSTd; (■) carbon material from as-synthesized HTS-1d; (○) carbon material from CSTp; (□) carbon material from CSTs; (+) carbon material from as-synthesized HTS-1s.



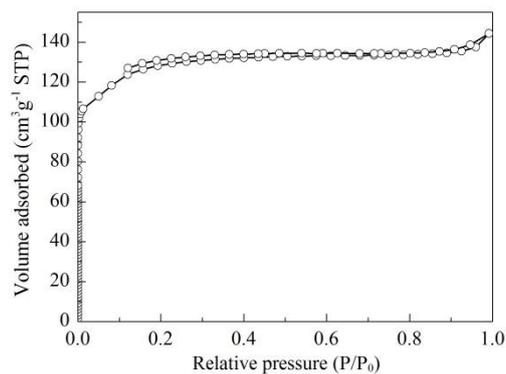
**Figure S5.** XRD patterns of TS-1, HTS-1d, and HTS-1s.

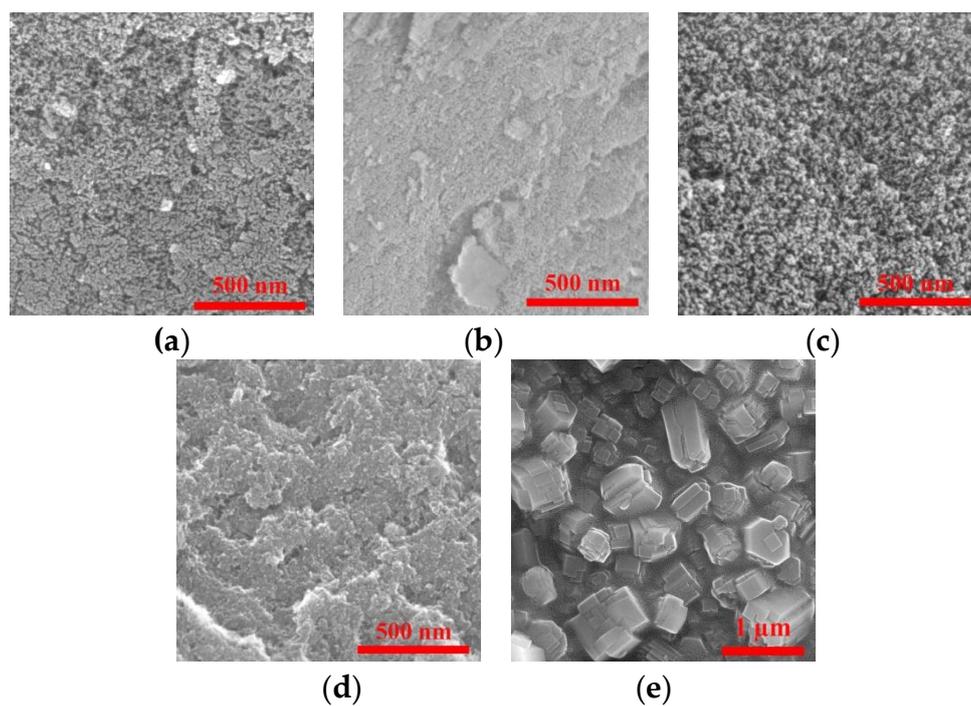
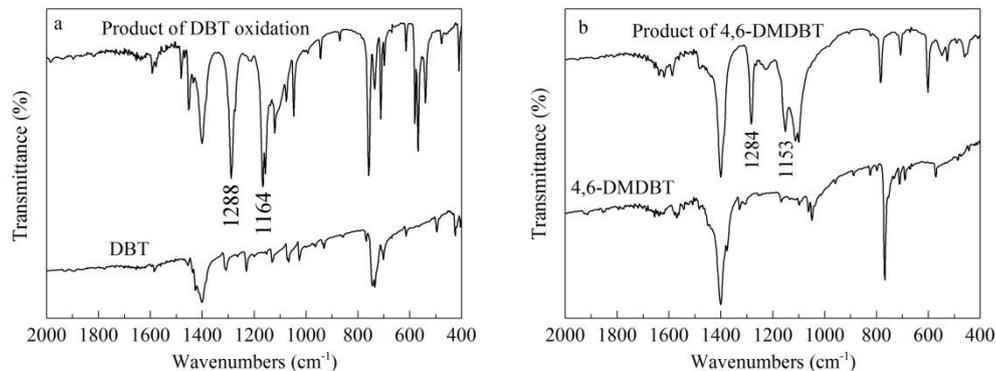


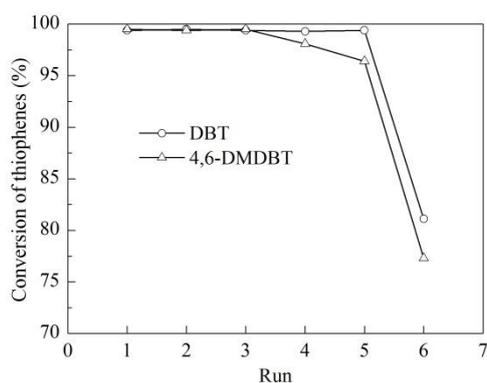
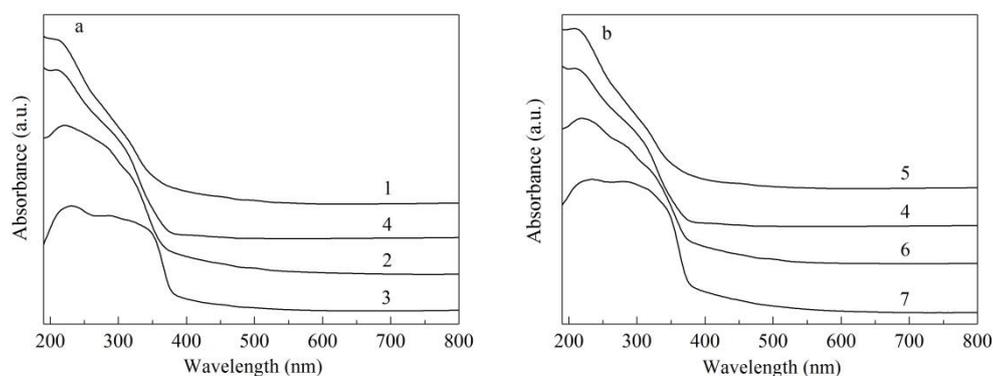
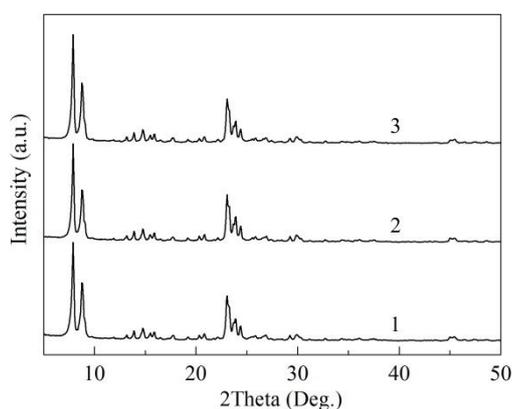
**Figure S6.** FT-IR spectra of TS-1, HTS-1d, and HTS-1s.

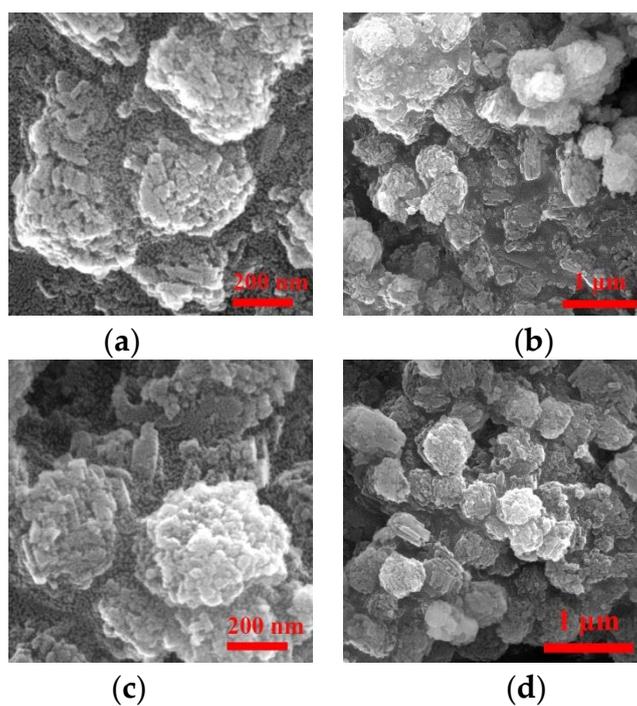


**Figure S7.** UV-Vis spectra of TS-1, HTS-1d, and HTS-1s.



**Figure S8.** Nitrogen adsorption-desorption isotherms of TS-1.**Figure S9.** SEM images of CSTs (a), CSTd (b), carbon materials obtained by removals of silica and titania from as-synthesized HTS-1s (c) and HTS-1d (d) with hydrofluoric acid aqueous solution, and TS-1 (e).**Figure S10.** FT-IR spectra of reactants and their corresponding products: (a) DBT and its oxidation product; (b) 4,6-DMDBT and its oxidation product.

**Figure S11.** Image of a typical reaction medium after the reaction was finished for 3 min.**Figure S12.** Recycle tests in the oxidation of DBT and 4,6-DMDBT over HTS-1s recovered by centrifugation and drying at 373 K.**Figure S13.** UV-Vis spectra of the fresh, used, and regenerated HTS-1s for DBT (a) and 4,6-DMDBT (b) oxidations: (1) HTS-1s regenerated by calcination after the 15th run; (2) HTS-1s recovered by centrifugation and drying after the sixth run; (3) product of DBT oxidation; (4) fresh HTS-1s; (5) HTS-1s regenerated by calcination after the 15th run; (6) HTS-1s recovered by centrifugation and drying after the sixth run; (7) product of 4,6-DMDBT oxidation.**Figure S14.** XRD patterns of fresh HTS-1s (1) and regenerated HTS-1s catalysts for DBT (2) and 4,6-DMDBT (3) oxidations.



**Figure S15.** SEM images of the regenerated HTS-1s after the 15th run: (a) and (b) for 4,6-DMDBT oxidation; (c) and (d) for DBT oxidation.