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## **Electronic Supplementary Information (ESI)**

## Synthesis of hierarchical TS-1 zeolites with abundant and uniform intracrystalline mesopores and their highly efficient catalytic performance for oxidation desulfurization

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**Scheme S1** Chemical structural formula of polydiallyldimethylammoniumchloride (PDADMAC)



Fig. S1 UV-vis spectra of conventional microporous TS-1 and hierarchically porous TS-1 zeolites.



Fig. S2 <sup>29</sup>Si MAS NMR spectra of the calcined conventional microporous TS-1 and hierarchically porous TS-1 zeolites.



Fig.S3  $^{13}$ C NMR spectra of aqueous PDADMAC and the liquid phase obtained by dissolving as-synthesized microporous TS-1 and hierarchically porous TS-1C zeolite with HF.



Fig. S4 GC-MS chromatograms of oxidation of DBT. The structures annotated onto the chromatograms are identified by the standard mass spectra in the NIST database. (a) The chromatographic spectrum before reaction; (b) The chromatographic spectrum after reaction.



Fig. S5 The Mass sepctrum of dibenzothiophene (DBT)



Fig. S6 The Mass sepctrum of the products (dibenzothiophene sulfone)

Table S1	Comparison of catalytic performance over some representative hierarchica
	TS-1 catalysts in oxidation desulfurization of DBT

Catalysts	S content / ppm	Oxidant / S - organic	Mass of catalyst /mg	T/ ºC	Time / min	Conversion / %	Refs.
h-TS-1	200	2	30	80	60	100	13
HTS-1	500	2	50	60	180	100	25
HTS-1	500	2	50	60	240	100	10
HTS-1	500	2	50	60	300	100	47
Ti-B-M-DA	500	6	50	60	45	100	48
Hierarchical TS-1	500	2	50	60	30	100	This work

Catalyst	Sulfur concentration	DBT Conversion/%						
	before reaction (ppm)	0min	5min	10min	15min	20min	30min	40min
TS-1	500	0	20.1	22.2	28.1	30.4	31.0	30.2
TS-1A	500	0	28.3	46.8	68.1	75.9	88.7	100.0
TS-1B	500	0	28.2	50.0	70.0	77.8	92.2	100.0
TS-1C	500	0	38.9	57.0	73.1	88.2	100.0	100.0

## Table S2 The DBT oxidation over conventional microporous TS-1 and hierarchically porous TS-1A, TS-1B and TS-1C

Reaction conditions: 10 ml 500 ppm model fuel, catalyst=50 mg, T=333 K, oxidant/sorganic = 2 (molar ratio)

Table S3 The recycle tests in the DBT oxidation over hierarchically porous TS-1A

Recycle	Sulfur concentration	DBT Conversion/%						
Times	before reaction (ppm)	0min	5min	10min	15min	20min	30min	40min
0	500	0	38.9	57.0	73.1	88.2	100.0	100.0
1	500	0	35.9	56.0	70.1	78.3	92.8	100.0
2	500	0	26.1	52.0	65.1	70.6	88.2	100.0
3	500	0	18.0	43.0	53.2	66.5	84.3	100.0

Reaction conditions: 10 ml 500 ppm model fuel, catalyst=50 mg, T=333 K, oxidant/sorganic = 2 (molar ratio)