Supporting information

One-pot Synthesis of Cup-like ZSM-5 Zeolite and Its Excellent Oxidative Desulfurization Performance

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1Chemicals

Sodium metaaluminate ($NaAlO_2$): chemically pure. Purity (Measured by Al_2O_3)≥41%. Tetraethoxysilane (TEOS): analytically pure. Purity≥98%. Tetrapropylammonium hydroxide (TPAOH): Purity = 25%.

2 Synthesis

2.1 The synthesis of solid ZSM-5 zeolite

17.6 mL TPAOH and 30.0 mL water was mixed, and then 0.6 g $NaAlO_2$ was added until it was dissolved. Next, 16.0 mL TEOS was added drop wise until the mixture was uniform. The mixture was put in a crystallization reactor and crystallized under 170°C for 8 hours. Then the products was filtered, washed, and dried to obtain solid ZSM-5 molecular sieves. Finally, the sample was converted into H-ZSM-5 through ion exchanges with NH_4NO_3 solution at 90^o C for three times and calcinated at 550^o C for 5h.

2.2 The synthesis of cup-like ZSM-5 zeolite

TPAOH and water was mixed, and then sodium metaaluminate was added until it was dissolved. Next, TEOS was added drop wise until the mixture was uniform.

The mixture was stirred strongly for 2h, put in a crystallization reactor and crystallized under certain temperature for 12 to 48 hours in a specially-made rotation oven. Then the products was filtered, washed, and dried to obtain small-sized hollow ZSM-5 molecular sieves. Finally, the sample was converted into H-ZSM-5 through ion exchanges with NH_4NO_3 solution at 90^o C for three times and calcinated at 550^o C for 5h.

3 Characterization

3.1 Powder XRD patterns: automated X-ray diffractometer (Rigaku Corporation of Japan, D/max-2400). Cu target Kα radiation. Tube power 12kW. Voltage 40kV. Current 100mA. Scanning rate 8° per min. Oscillation range 0.02°. Scanning range 5~50°.

3.2 IR : Bruker EQUINOX55 Fourier transform infrared spectroscopy.

3.3 SEM : scanning electron microscope (FEI, NOVA SEM 450)

3.4 TEM : transmission electron microscopy (Tecnai F30 and JEM-2100).

3.5.NH_3-TPD AutoChem II 2920

The samples were ion exchanged three times with 1M NH_4NO_3 at 90 °C for 2h, dried at 110 °C and calcined in air at 550 °C for 5h. NH_3-TPD was carried out with a Thermo TPDRO instrument equipped with a TCD detector. The sample was heated to 900 °C with 10 °C/min.

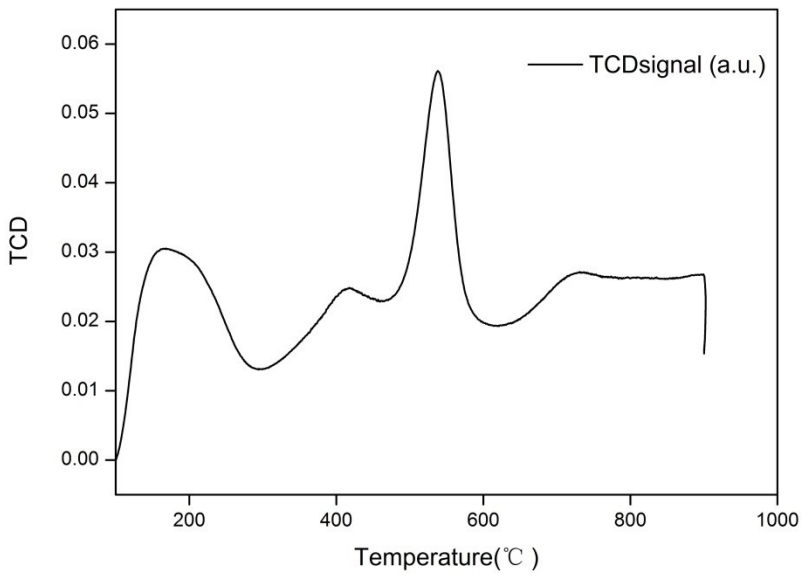

Fig. S1 NH_3-TPD of the synthesized hollow molecular sieves (crystallization temperature 130 ºC, crystallization time 48h, OH^-/SiO_2=0.3)

3.6.XPS

Elemental analysis was characterized by X-ray Photoelectron Spectroscopy (XPS; Kratos, AXIS ULtrabld).

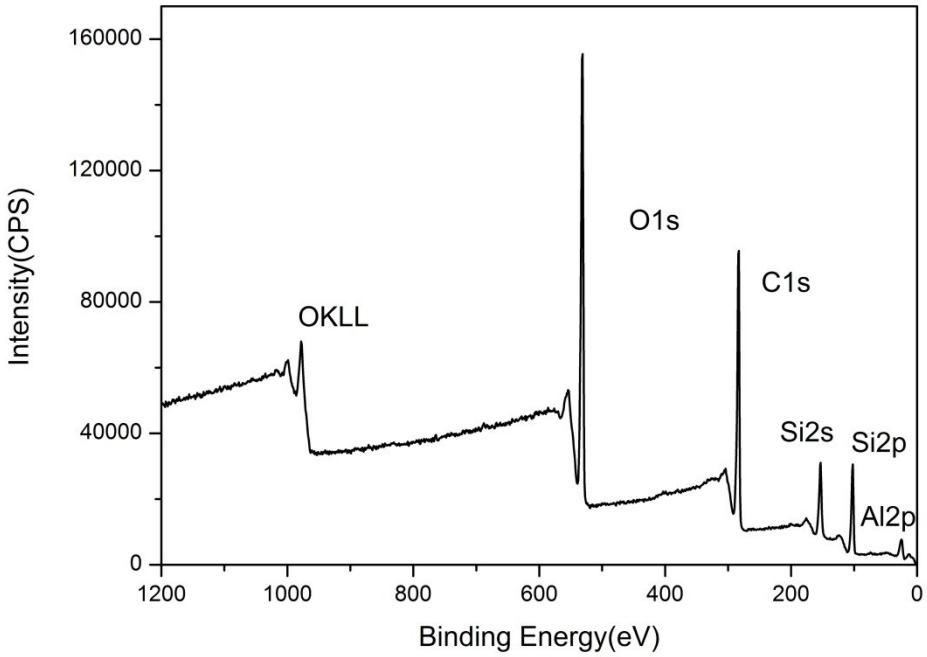

Fig. S2 Elemental analysis of synthesized hollow molecular sieves (crystallization temperature 130 ºC, crystallization time 48h, OH^-/SiO_2=0.3)

3.7.N_2 adsorption and desorption

N_2 adsorptions at 77K for the samples were performed on an apparatus of Micromeritics ASAP 2460. Before the adsorption, the samples were pretreated in vacuum at 300°C for 6 h to remove the physical adsorption of water in the holes.

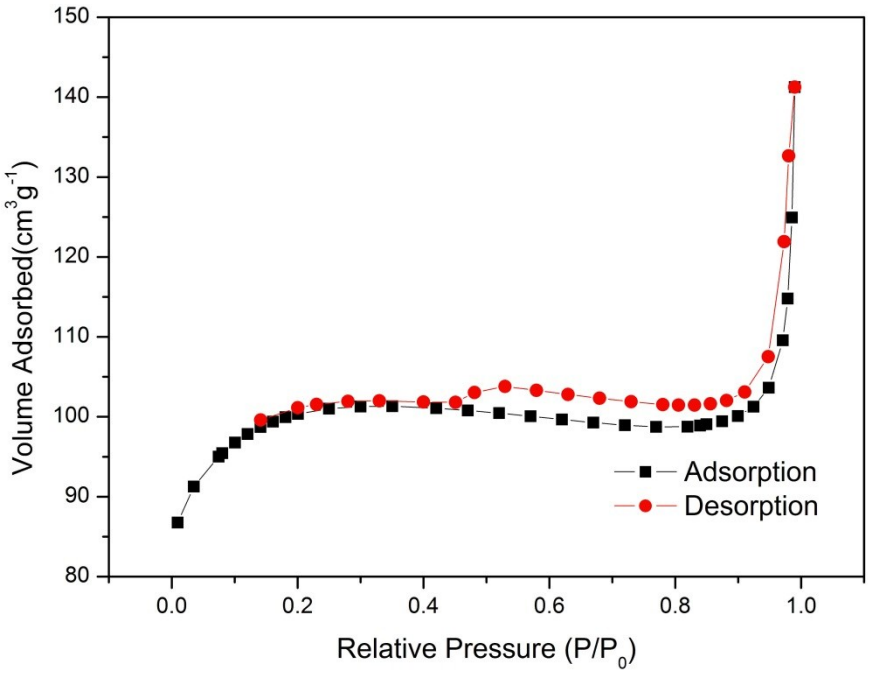

Fig. S3 N_2 desorption and adsorption of the synthesized hollow molecular sieves (crystallization temperature 130 ºC, crystallization time 48h, OH^-/SiO_2=0.3)