Supporting Information

Al-RUB-41: A Shape-Selective Zeolite Catalyst from a Layered Silicate

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Procedure for seeded-synthesis of Al-RUB-39 and its conversion to Al-RUB-41

In a typical synthesis procedure, 0.37 g of sodium aluminate [$NaAlO_2$ (Al_2O_3 = 34 ~ 39 %, Na_2O = 31 ~ 35 %)] was dissolved in 3.0 g of water and then mixed with 7.05 g of aqueous solution of the template (SDA), dimethyldipropyl ammonium hydroxide (39 wt.% DMDPAOH, Sachem Inc.) under stirring at room temperature for 1 h. In the above solution, 6.0 g of water was added and stirred for additional 5 min at room temperature. 2.25 g of fumed silica (Cab-O-Sil M7D) was then added into the resulting liquid mixture under vigorous stirring. A semi-clear gel obtained after 2 h of stirring at room temperature was mixed with 3.9 ml of 0.15 M aqueous NaOH solution under stirring at room temperature for 1 h. 0.04 g of Si-RUB-39 was then mixed with the resulting gel having a chemical composition of SiO_2 : SDA : $NaAlO_2$: NaOH : H_2O = 1 : 0.5 : 0.067 : 0.016 : 26 and transferred into a teflon-lined autoclave. The crystallization was carried out at 140^{o}C for 15 days under rotating conditions. The obtained solid, Al-RUB-39 zeolite, was then separated by filtration, washed with distilled water and dried at 70^{o}C for 24 h and calcined at 540^{o}C for 12 h.

Procedure for two-step synthesis of Al-RUB-39 and its conversion to Al-RUB-41

Step 1:

1.3 g of sodium hydroxide (NaOH) was dissolved in 35.4 g of water. This solution was then mixed with 100.9 g of aqueous solution of the template (SDA), dimethyldipropylammonium hydroxide (39 wt.% DMDPAOH, Sachem Inc.) under stirring at room temperature for 10 min. Then, 1.3 g of seed crystals of RUB-39 were added to this solution under stirring at room temperature for 20 min. Fumed silica (Aerosil) was added to this mixture in a stepwise fashion by consecutively introducing smaller portions, and a rather thick suspension was obtained. This suspension was stirred at room temperature for an additional 20 min. The resulting gel was poured into the autoclave. The molar composition of this synthesis mixture was SiO_2 : SDA : NaOH : H_2O = 1 : 0.5 : 0.0625 : 10. The first crystallization step was performed at 150 °C and the duration was 48 h. An opaque suspension with precipitation at the bottom was obtained after the first step.

Step 2:

1.47 g of sodium aluminate ($NaAlO_2$) was dissolved in 53.7 g of water under stirring for 20 min. This solution was then mixed with the precursor obtained from the first step. The mixture was stirred for 30 min. The temperature was then increased to 70 °C and 53.7 g of water was evaporated from the synthesis mixture. The resulting gel was poured into the autoclave. The molar composition of this synthesis mixture was SiO_2 : $NaAlO_2$: SDA : NaOH : H_2O = 1 : 0.0333 : 0.5 : 0.0625 : 10. The second crystallization step was performed at 140 °C and the duration was 48 h. An opaque suspension with precipitation at the bottom was obtained. The product was filtered (via centrifugation) and washed with water. The solid product was dried at 100 °C.

Conversion to Al-RUB-41

Calcination of the Al-RUB-39 product at 600°C results in the Al-RUB-41 product.

Instruments and conditions used for physicochemical characterization

Powder X-ray diffraction (XRD) patterns were collected on a Rigaku Ultima III diffractometer using a CuKα X-ray source (40 kV, 40 mA). Chemical compositions were analyzed by a Shimadzu ICPE-9000 spectrometer. Solid-state ^{27}Al MAS NMR spectra were obtained on a JEOL ECA-400 spectrometer. High-Resolution transmission electron microscopy (HR-TEM) and electron diffraction (ED) investigations were performed using a FEI Tecnai (200 kV) instrument. For characterization of morphology and crystal size, field emission scanning electron microscopy (FE-SEM) was performed using a JEOL JSM-7400F (5 kV) unit.

Procedure for hydroconversion of decane experiments on Pt-Al-RUB-41 (0.5 wt.%)

0.5 wt.% Pt was introduced into Al-RUB-41 via ion-exchange as $Pt(NH_3)_4^{2+}$. The experiments were performed at temperatures ranging from 140°C to 300°C, 4.5 bar H_2 pressure, a H_2/HC molar ratio of 375 and a contact time of 2522 kg/s.mol. Before reaction, the Pt/H-zeolite samples were first treated with an O_2-flow of 30 ml/min at 400°C for the oxidation and then with a H_2-flow of 60 ml/min at 400°C for the reduction of platinum.

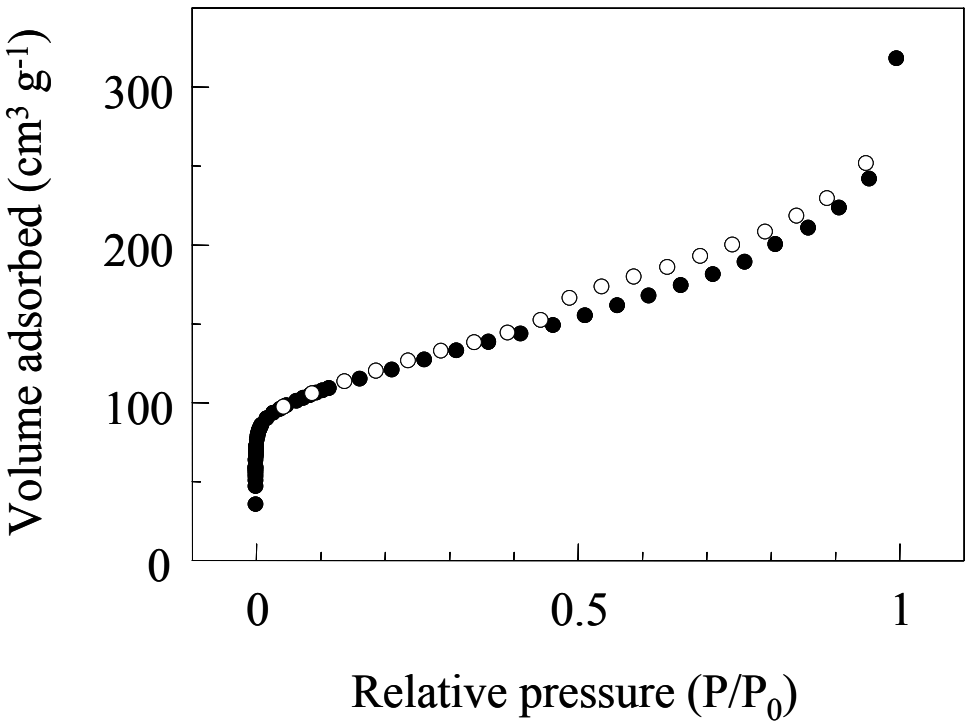

Figure S1: N_2-isotherms of Al-RUB-41. (Synthesis mixtures with SiO_2/Al_2O_3 = 200 and SiO_2/SDA = 2)

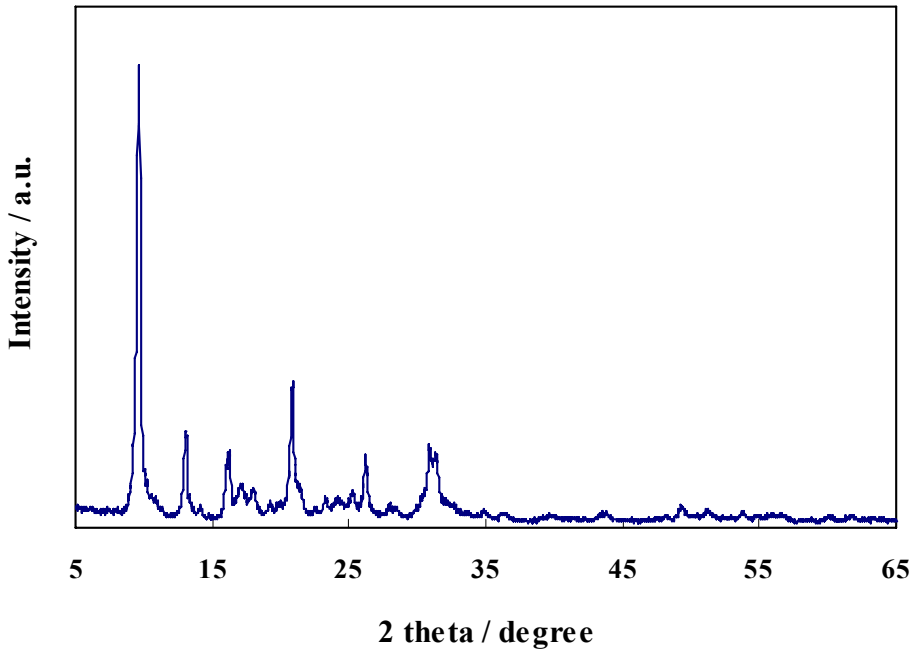

Figure S2: Powder XRD pattern of Al-RUB-41 obtained from the topotactic condesation of Al-RUB-39 prepared via the direct (one-step) crystalliaztion route. (Synthesis mixture with molar composition SiO_2 : 0.5 SDA : 0.067 $NaAlO_2$: 0.016 NaOH : 33 H_2O)

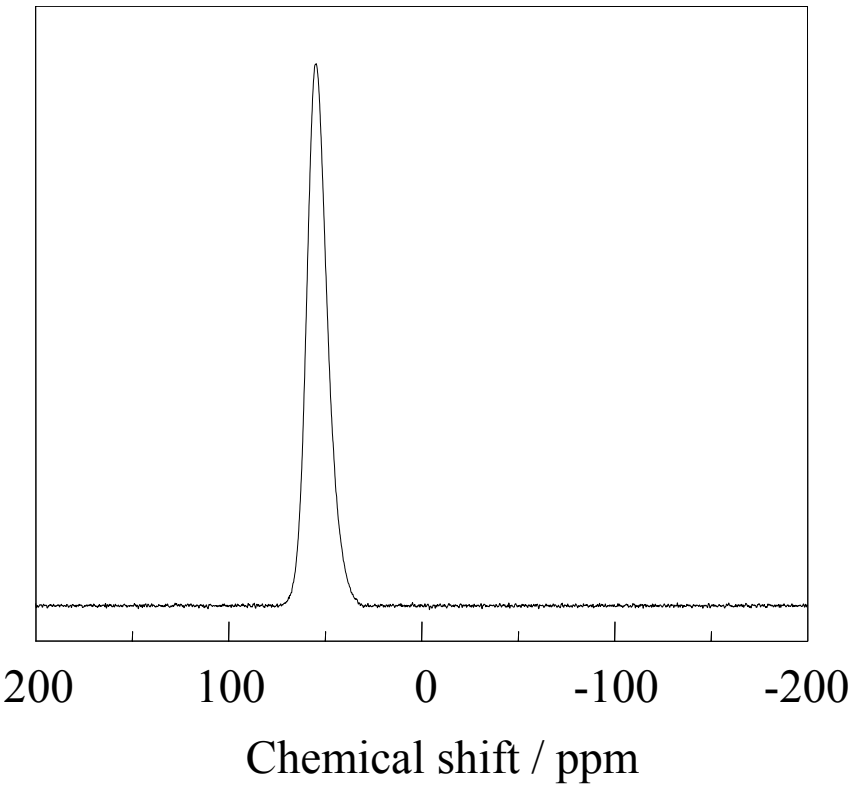

Figure S3: ^{27}Al-NMR spectrum of Al-RUB-41 obtained from the topotactic condesation of Al-RUB-39 prepared via the direct (one-step) crystalliaztion route. (Synthesis mixture with molar composition SiO_2 : 0.5 SDA : 0.067 $NaAlO_2$: 0.5 NaOH : 12 H_2O)

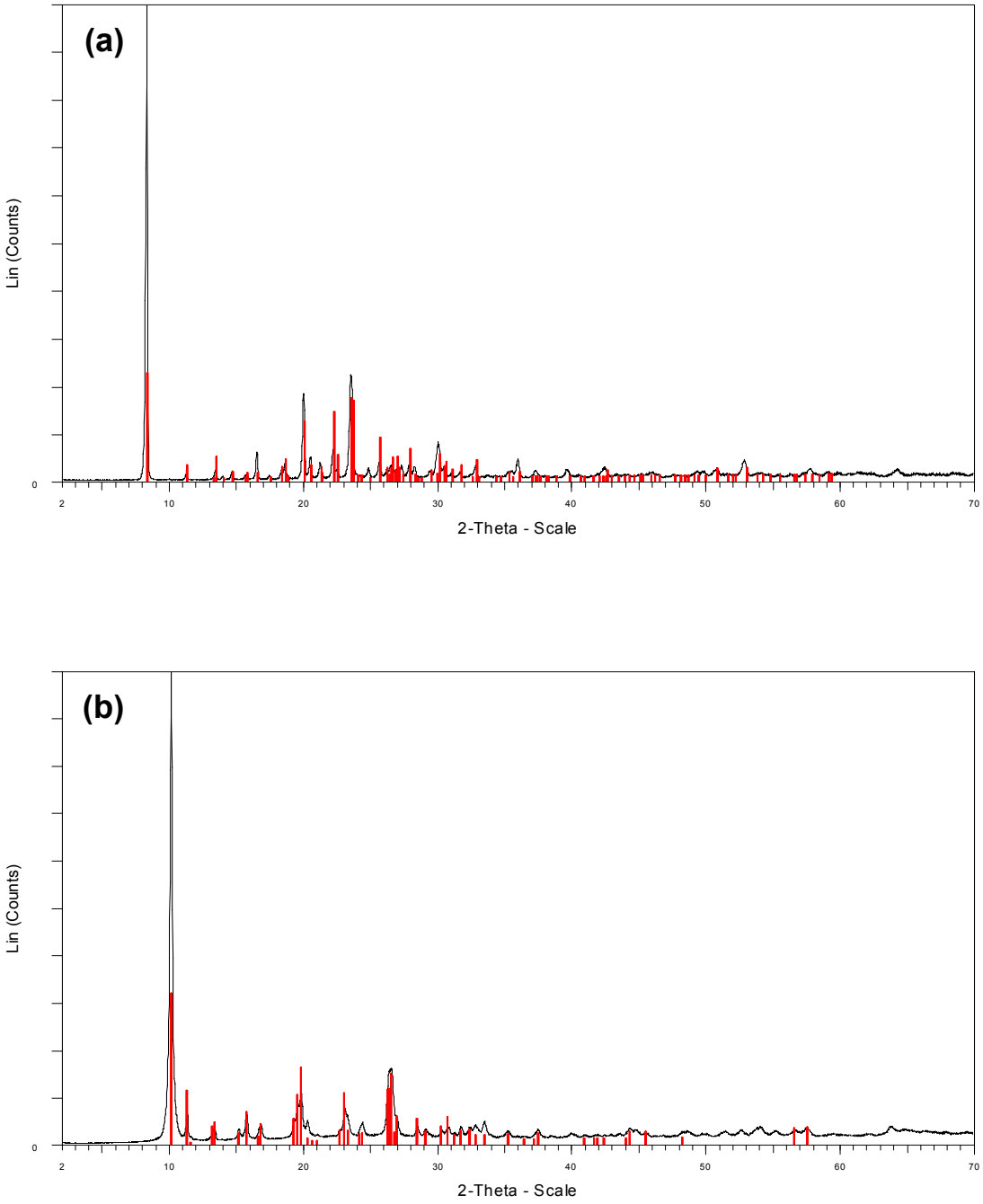

Figure S4: Powder XRD patterns of (a) Al-RUB-39 and (b) Al-RUB-41 prepared via the two-step crystalliaztion route (Synthesis mixture with molar composition SiO_2 : 0.5 SDA : 0.0333 $NaAlO_2$: 0.0625 NaOH : 10 H_2O). Red lines represent standard peak positions of all-silica (a) RUB-39 and (b) RUB-41 for reference.