

Electronic Supplementary Information (ESI)
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**Mixed Template Effect Adjusted by Amine Concentration in
Ionothermal Synthesis of Molecular Sieves**

Renyan Pei,^{ab} Ying Wei,^{ab} Keda Li,^{ab} Renshun Xu,^{ab} Yunpeng Xu,^a Lei Wang,^a Huaijun Ma,^a Bingchun Wang,^a Zhijian Tian^{a*} Weiping Zhang^a and Liwu Lin^a

^a *State Key Laboratory of Catalysis, Dalian Institute of Chemical Physics, Chinese Academy of*

Sciences, 457 Zhongshan Road, Dalian 116023, China

^b *Graduate School of Chinese Academy of Sciences, Beijing 100049, China*

E-mail: tianz@dicp.ac.cn

Tel: +86-411-8437-9151; Fax: +86-411-8437-9151;

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S1. Experimental section

Ionic liquid was prepared by neutralization of methyl imidazoles and butyl bromide, The reagents were purified by redistillation in laboratory, including N-methyl imidazole (99.9%, Kaile Chemical Factory, zhejiang China) and Butyl bromide (AR, Sinopharm Chemical Reagent Co., Ltd)

Other commercially obtained reagents were used as received in ionothermal synthesis of molecular sieves according to the gel composition listed in Table 1 in the text. The following are detailed information of reagents.

Phosphoric acid (85% in water, AR, Sinopharm Chemical Reagent Co., Ltd.)

Hydrofluoric acid (AR, Shenyang chemical reagent factory, 40% in water)

Aluminium iso-propoxides (AR, Sinopharm Chemical Reagent Co., Ltd.)

n-DPA (n-dipropyl amine AR, Sinopharm Chemical Reagent Co., Ltd.)

DEA (diethyl amine AR, Sinopharm Chemical Reagent Co., Ltd.)

TriBA (tributylamine AR, Sinopharm Chemical Reagent Co., Ltd.)

n-BA (n-butylamine AR, Sinopharm Chemical Reagent Co., Ltd.)

CHA (Cyclohexanamine AR, Sinopharm Chemical Reagent Co., Ltd.)

HMTA (Hexamethylenetetramine AR, Sinopharm Chemical Reagent Co., Ltd.)

Imidazole (AR, Sinopharm Chemical Reagent Co., Ltd.)

Pyridine (AR, Sinopharm Chemical Reagent Co., Ltd.)

S2. Characterizations of materials

Powder X-ray diffraction (PXRD) analyses of the as-synthesized samples are performed on a PANalytical X'Pert PRO diffractometer fitted with Cu K α radiation ($\lambda = 1.5406 \text{ \AA}$) operating at 40mA and 40 kV. Scanning electron microscopy (SEM) was carried out on Hitachi S4800 field-emission scanning electron microscopes. The FTIR spectra were recorded on a Perkin Elmer spectrum GX spectrometer by 10 mm potassium bromine pellet with a resolution of 4 cm^{-1} and OPD Velocity of $0.2 \text{ cm per second}$. The number of scan is 64 and the interval is 1 cm^{-1} . ^{13}C , ^{27}Al and ^{31}P CP/MAS NMR measurements are performed on a Varian Infinityplus-400 spectrometer. The thermal behavior was investigated with Perkin Elmer Pyris 1 TGA instrument. The atmosphere was dynamic dry air, with a flow rate of 20 ml min^{-1} . The selected heating rate was $20^\circ\text{C min}^{-1}$ and the temperature range is 30 to 850°C . The weight of samples used in thermal analysis experiments is rigidly at a range of 2.4 to 2.6 mg.

S3. Powder XRD patterns of samples

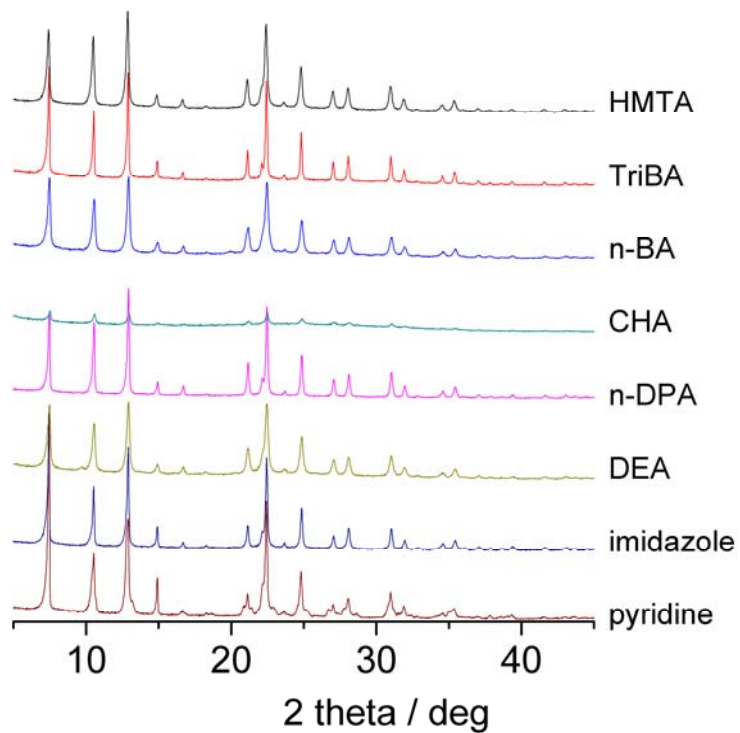


Fig. 1 XRD patterns of products with LTA topology in ionothermal synthesis with addition of various organic amines

S4 Thermal gravimetric (TG) analysis

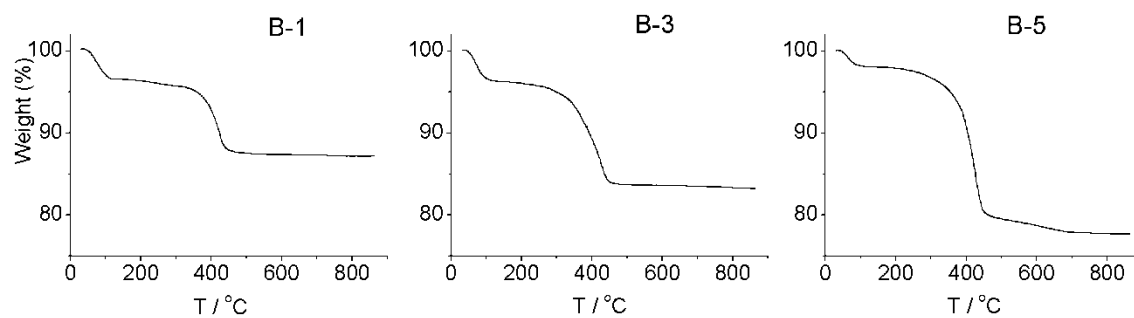


Fig. 4 Thermal gravimetric (TG) curves of sample B-1, B-3 and B-5.

S5 ^{27}Al and ^{31}P NMR spectroscopy of LTA sample B-5

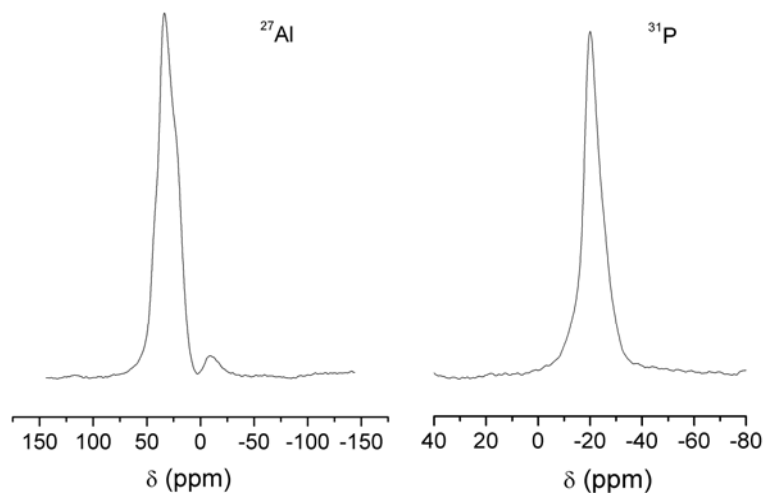


Fig. 3 ^{27}Al and ^{31}P MAS NMR spectra of B-5

^{27}Al and ^{31}P MAS NMR spectra of B-5 ($\text{AlPO}_4\text{-42}$, LTA structure) are shown in Fig. 3. The occurrence of peak at -19 ppm in ^{31}P NMR spectra results from a strong interaction of P atom with the F^- ions. This indicates that F^- ions reside inside the D4R unit. This is consistent with the result of Schreyeck et al.¹ The ^{13}C NMR spectra of several products with LTA structure could be seen in article.

S6 SEM images of LTA sample B-11 and B-12

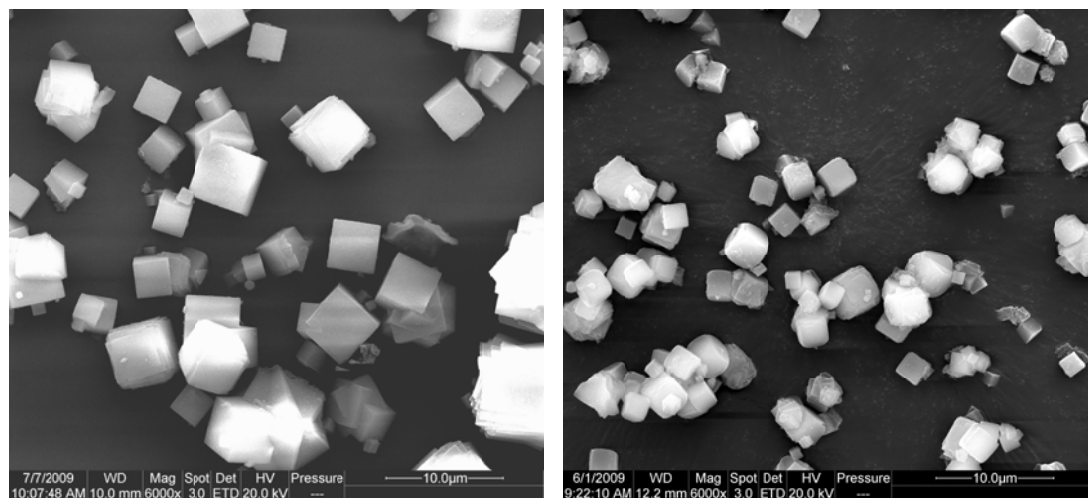


Fig. 4 SEM images of products with LTA structure synthesized in BMImBr by introducing pyridine(left) and imidazole(right)

Reference

1. L. Schreyeck, J. Stumbe, P. Caullet, J. C. Mougénel and B. Marler, *Microporous Mesoporous Mater.*, 1998, **22**, 87-106.