dichloromethane (10/1). Phosphirene complex **4** was isolated as yellow crystals (2 g, 66%). ^{31}P NMR (81 MHz, CDCl$_3$): $\delta = -201.0$ (^1J(P,W) = 294.9 Hz); ^{13}C NMR (50 MHz, CDCl$_3$): $\delta = 89.3$ (d, ^1J(C,P) = 18.8 Hz; P-*C*≡C), 93.9 (d, ^2J(C,P) = 5.8 Hz; P-C≡*C*), 120.9 (s; C≡C-*Ph*, C$_{ipso}$), 195.9 (d, ^2J(C,P) = 9.1 Hz; *cis*-CO), 198.4 (d, ^2J(C,P) = 34.8 Hz; *trans*-CO); MS: *m*/*z* (%): 634 (6) [M^+], 494 (100) [$M^+ - 5$CO]; elemental analysis (%) calcd for $C_{27}H_{15}O_5PW$: C 51.10, H 2.36; found: C 51.48, H, 2.42.

6: Biphosphirene **6** was isolated as light yellow crystals by chromatography with hexane/dichloromethane (4/1). ^{13}C NMR (50 MHz, CDCl$_3$): $\delta = 137.4$ (d, ^1J(C,P) = 5.4 Hz; C$_{ipso}$, Ph-P), 143.2 (pseudo t, ^1J(C,P) ~ ^2J(C,P) = 13.6 Hz; Ph-*C*(P)=C-P); MS: highest mass 785 [$M^+ - 10$CO − H].

7: Biphosphirene **7** was isolated as a yellow powder by chromatography with hexane/dichloromethane (10/1). ^{13}C NMR (50 MHz, CDCl$_3$): $\delta = 89.5$ (d, ^1J(C,P) = 23.6 Hz; P-*C*≡C), 95.2 (d, ^2J(C,P) = 6.5 Hz, P-C≡*C*), 120.5 (s; C≡C-*Ph*, C$_{ipso}$), 144.6 (pseudo t, ^1J(C,P) ~ ^2J(C,P) = 15 Hz; Ph*C*(P)=C-P); MS: highest mass 632; elemental analysis (%) calcd for $C_{40}H_{20}O_{10}P_2W_2$: C 44.07, H 1.85; found: C 44.03, H 1.75.

8: Triphosphirene **8** was isolated as light yellow crystals by chromatography with hexane/dichloromethane (4/1); elemental analysis calcd (%) calcd for $C_{51}H_{25}O_{15}P_3W_3$: C 40.24, H 1.66; found: C 39.98, H 1.56.

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[8] X-ray structure data for **8a**: crystal dimensions 0.40 × 0.16 × 0.16, monoclinic, $P2_1/n$, $a = 12.467(5)$, b = 18.565(5), $c = 22.346(5)$ Å, $\beta = 91.220(5)°$, $V = 5171(3)$ Å^3, $Z = 4$, $\rho_{calcd} = 1.955$ g cm^{-3}, $\mu = 6.817$ cm^{-1}, $F(000) = 2872$, $\theta_{max} = 30.03°$, *hkl* ranges: −13–17; −21–26; −31–25, 33543 data collected, 15050 unique data ($R_{int} = 0.0699$), 12185 data with $I > 2\sigma(I)$, 649 parameters refined, GOF(F^2) = 1.008, final R indices: $R1 = 0.0414$, $wR2 = 0.1095$; max/min residual electron density 2.082(.251)/−3.219(.251) e Å^{-3}. Data collection was with a Nonius Kappa CCD diffractometer, ϕ and ω scans, Mo$_{K\alpha}$ radiation ($\lambda = 0.71073$ Å), graphite monochromator, $T = 150$ K, structure solution with SIR97,refinement against F2 (SHELXL97) with anisotropic thermal parameters for all non-hydrogen atoms, calculated hydrogen positions with riding isotropic thermal parameters. Crystallographic data (excluding structure factors) for the structures reported in this paper have been deposited with the Cambridge Crystallographic Data Centre as supplementary publication no. CCDC-151986. Copies of the data can be obtained free of charge on application to CCDC, 12 Union Road, Cambridge CB21EZ, UK (fax: (+44) 1223-336-033; e-mail: deposit@ccdc.cam.ac.uk).

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Steam-Stable MSU-S Aluminosilicate Mesostructures Assembled from Zeolite ZSM-5 and Zeolite Beta Seeds[**]

Yu Liu, Wenzhong Zhang, and Thomas J. Pinnavaia[*]

The structural integrity of Al-MCM-41 and related mesoporous aluminosilicate molecular sieves has been significantly improved in recent years through direct assembly and post-synthesis treatment methods.[1] Nevertheless, the hydrothermal instability and mild acidity remain inferior to conventional zeolites and limit potential applications in petroleum refining and fine chemicals synthesis.[2] One might expect to improve both the stability and acidity of these materials if zeolite-like order could be introduced into the pore walls. One approach, first introduced by van Bekkum and co-workers,[3] is to transform the preassembled walls of Al-MCM-41 into zeolitic structures by post-assembly treatment with a microporous zeolite structure director, such as tetrapropylammonium cations. More recent studies have shown that the walls of the mesostructure can indeed be converted to a zeolitic product, but the microporous zeolite phase (ZSM-5) is segregated from the mesostructure, giving rise to ZSM-5/MCM-41 composites.[4] These composites exhibited an enhancement in acidity for hydrocarbon cracking in comparison to mechanical mixtures of ZSM-5 and MCM-41 and an improvement in steam stability for purely siliceous composites.[4c]

We recently reported an alternative approach to more acidic and hydrothermally stable mesostructures based on the direct assembly of nanoclustered aluminosilicate precursors that normally nucleate zeolite type Y crystallization.[5] These protozeolitic species, known as "zeolite seeds", promote zeolite nucleation by adopting AlO$_4$ and SiO$_4$ connectivities that resemble the secondary building units of a crystalline zeolite.[6] The assembly of the Na$^+$-nucleated zeolite type Y (faujasitic) seeds under hydrothermal conditions in the presence of cetyltrimethylammonium ions afforded hexagonal MSU-S mesostructures with Si/Al ratios in the range 1.6:1 to 10:1. The replacement of Na$^+$ by NH$_4^+$ ions in the as-made mesostructure, followed by calcination in the presence of the surfactant, afforded exceptionally acidic and steam-stable mesostructures. However, the steam stability was enhanced by structure-stabilizing occlusions of carbon that formed during the calcination process. That is, the steam stability at 800 °C was in part a consequence of the exceptional acidity of a framework that formed structure-stabilizing carbon, and not entirely a result of an intrinsically stable framework.

Here we also make use of protozeolitic nanoclusters to form exceptionally acidic and steam-stable aluminosilicate

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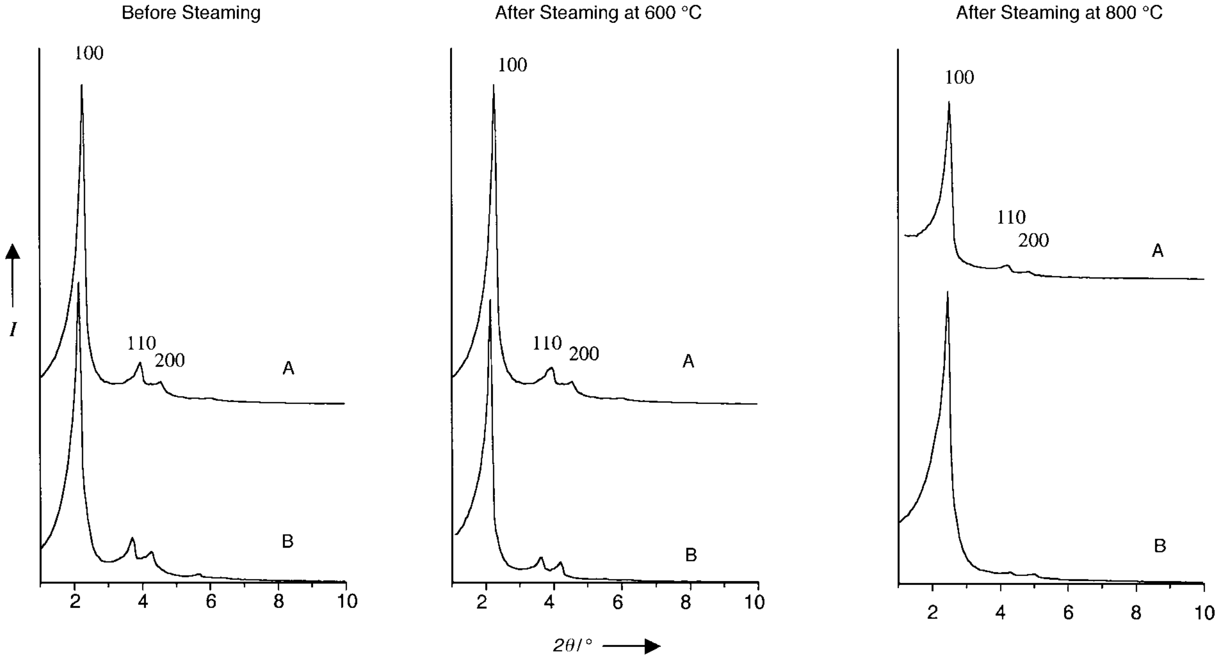
mesostructures, but the zeolitic seeds are formed from *organic* structure directors. Moreover, the Si/Al ratios are much *higher* (>10.0:1) than the ratios needed to template faujasitic (FAU) zeolite seeds (~1.6:1–10.0:1). The resulting mesostuctures, unlike those formed from FAU seeds, are remarkably steam stable even in the absence of occluded coke. Since many organic cations are known to function as zeolite-nucleating agents,[7] zeolite seeds templated by organic cations promise to be much more versatile for the supramolecular assembly of aluminosilicate mesostructures that are intrinsically steam stable.

The zeolite seeds of interest in the present work nucleate the crystallization of pentasil zeolites ZSM-5 (MFI) and Beta (BEA). Aqueous solutions of nanoclustered MFI and Beta seeds (Si/Al = 67:1) were prepared by using tetrapropylammonium (TPA^+) and tetraethylammonium (TEA^+) templates, respectively.[8] For comparison purposes, conventional aluminosilicate anions were prepared by using tetramethylammonium ions (TMA^+) in place of TPA^+ and TEA^+. TMA^+ is not known to function as a template for MFI or BEA zeolite seeds. The solids formed through the evaporation of each of the aluminosilicate solutions afforded amorphous X-ray diffraction patterns. However, the IR spectra of the solids formed from MFI and BEA zeolite seeds revealed distinct vibrations between 550–600 cm^{-1}. In contrast, no IR bands were observed in this range for the conventional aluminosilicate anions formed from TMA^+ hydroxide. A band near 550 cm^{-1} in pentasil MFI and BEA zeolites is indicative of the presence of five-membered rings.[9] Thus, the presence of an analogous band for the TPA^+ and TEA^+ aluminosilicate precursors verified the presence of pentasil zeolite subunits, even though MFI and Beta nanocrystals are absent.

Hexagonal aluminosilicate mesostructures, denoted MSU-$S_{(MFI)}$ and MSU-$S_{(BEA)}$, were assembled by reaction of the respective zeolite seed solutions with cetyltrimethylammonium bromide (CTAB) at a surfactant to silica ratio of 0.25:1 at 150 °C for two days. An equivalent procedure was used to assemble conventional TMA^+ aluminosilicate precursors into a disordered MCM-41 mesostructure. The as-made products were calcined at 550 °C to remove surfactant, treated with 0.1 M NH_4NO_3 at room temperature to displace exchangeable sodium ions, and then calcined again at 550 °C to convert NH_4^+ at exchange sites to protons. Chemical analysis indicated the absence of occluded carbon and a Si/Al ratio in agreement with the ratio contained in the initial zeolite seeds.

Figure 1 illustrates the X-ray diffraction (XRD) patterns of calcined 1.5 % Al-MSU-$S_{(MFI)}$ and 1.5 % Al-MSU-$S_{(BEA)}$ before and after exposure to 20 % (v/v) water vapor in N_2 at 600 and 800 °C for 5 h. The XRD results clearly indicate that the mesostructures retain hexagonal order upon steaming. Disordered 1.5 % Al-MCM-41 also retained some XRD order after exposure to steam at 600 °C, but at 800 °C the mesostructure was almost completely lost (XRD patterns not shown). The disorder initially associated with the 1.5 % Al-MCM-41 sample is not responsible for the collapse of the mesostructure at 800 °C. Even well-ordered Al-MCM-41 samples with four hexagonal *hkl* reflections have been shown[5] to exhibit a loss of mesostructure upon steaming at 800 °C.

Figure 2 provides the N_2 adsorption and desorption isotherms for 1.5 % Al-MSU-$S_{(MFI)}$ and 1.5 % Al-MSU-$S_{(BEA)}$. Table 1 provides the surface areas, framework pore sizes, and pore volumes for 1.5 % Al-MSU-$S_{(MFI)}$ and 1.5 % Al-MSU-$S_{(BEA)}$ and compares them with those for 1.5 % Al-MCM-41 formed under equivalent conditions. The 1.5 % Al-MSU-S mesostructures retain more than 95 % of their surface areas and more than 87 % of their framework pore volumes with little or no pore contraction after exposure to steam at 600 °C.


Before Steaming

After Steaming at 600 °C

After Steaming at 800 °C

Figure 1. XRD patterns of calcined (550 °C, 4 h) mesoporous aluminosilicate molecular sieves before and after steaming at 600 °C and 800 °C for 5 h: A) hexagonal 1.5 % Al-MSU-$S_{(MFI)}$ prepared from zeolite ZSM-5 seeds; B) hexagonal 1.5 % Al-MSU-$S_{(BEA)}$, prepared from zeolite Beta seeds. The intensity scale is the same for the samples before and after steaming. Hexagonal unit cell parameters are provided in Table 1.

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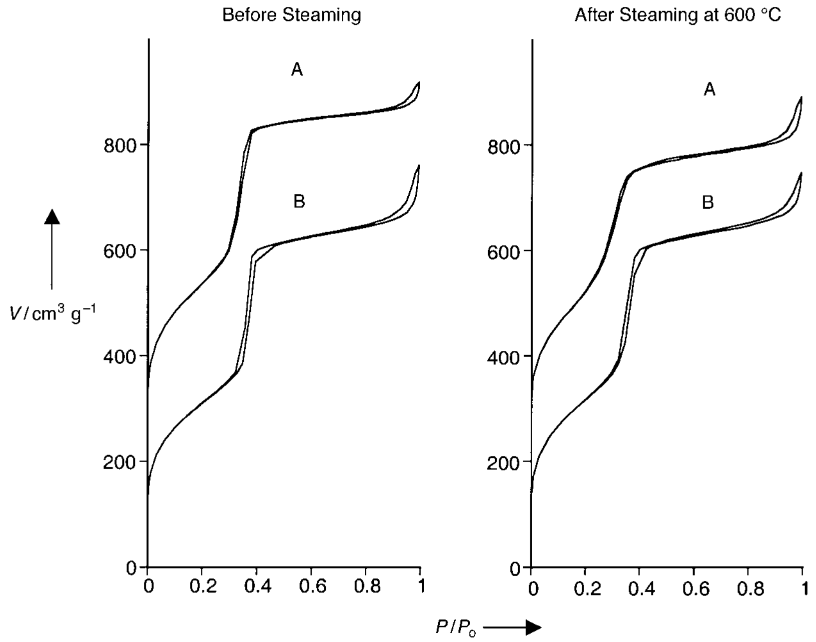

Figure 2. N_2 adsorption/desorption isotherms for calcined (550 °C, 5 h) mesoporous aluminosilicate molecular sieves before and after steaming (20 vol% H_2O in N_2, 600 °C, 5 h): A) 1.5% Al-MSU-$S_{(MFI)}$ B) 1.5% Al-MSU-$S_{(BEA)}$. The isotherms are offset by 200 cm^{3}g^{-1} for clarity.

Table 1. Textural properties and cumene conversions for mesoporous aluminosilicate sieves.

Sample	Unit cell dimension a_0 [Å]	Surface area [m^{2}g^{-1}]	Pore volume [cm^{3}g^{-1}]	Pore diameter [Å]	Cumene conversion[a] [%]
1.5% Al-MSU-$S_{(MFI)}$					
calcined	45.3	1231	1.06	36.8	32.3
steamed 600 °C, 5 h	44.5	1192	0.93	34.7	–
steamed 800 °C, 5 h	36.6	849	0.44	24.3	–
1.5% Al-MSU-$S_{(BEA)}$					
calcined	47.3	1124	1.06	39.1	31.5
steamed 600 °C, 5 h	46.7	1065	0.94	38.0	–
steamed 800 °C, 5 h	37.0	885	0.46	26.4	–
1.5% Al-MCM-41[b]					
calcined	46.4	1013	1.08	38.7	11.7
steamed 600 °C, 5 h	35.2	639	0.39	20.1	–
steamed 800 °C, 5 h	–	55	–	–	–

[a] Reaction conditions: 6 mm i.d. fixed bed quartz reactor; 200 mg catalyst; cumene flow rate, 4.1 μmol min^{-1}; N_2 carrier gas, 20 cm^3 min^{-1}; conversions reported after 60 min on-stream at 300 °C. [b] 1.5% Al-MCM-41 was prepared by the direct assembly of conventional aluminosilicate anions formed from sodium aluminate, fumed SiO_2, and TMAOH.

In contrast, 1.5% Al-MCM-41 assembled from conventional aluminosilicate anions retains only 63% of its surface area and 36% of its framework pore volume under equivalent steaming conditions. Upon increasing the steaming temperature to 800 °C, we find that both MSU-S aluminosilicate mesostructures retain long-range hexagonal order (cf., Figure 1) and substantial mesoporosity (cf., Table 1), whereas the mesoporosity of MCM-41 is completely lost. Also, as we have shown previously[5] that "ultrastable" grafted forms of Al-MCM-41[1b] are destroyed completely under these steaming conditions.

The supramolecular assembly of steam-stable MSU-S mesostructures from zeolite MFI and Beta seeds is not limited to the 1.5% Al-MSU-S compositions illustrated here. These same nanoclusters also can be used to assemble steam-stable MSU-S mesostructures with Si/Al ratios in the range ~20:1–300:1. Also, in contrast to previously reported MSU-S derivatives assembled from faujasitic zeolite type Y seeds (denoted here as MSU-$S_{(FAU)}$), MSU-$S_{(MFI)}$ and MSU-$S_{(BEA)}$ are intrinsically stable and do not require the presence of occluded carbon for steam stability.

^{27}Al MAS NMR spectroscopy indicates that more than 90% of the aluminum centers in calcined 1.5% Al-MSU-$S_{(MFI)}$ and 1.5% Al-MSU-$S_{(BEA)}$ are in tetrahedrally coordinated sites. In accord with the tetrahedral siting of aluminum, the Brønsted acidity of these steam-stable mesostructures was verified through cumene cracking experiments. Cumene conversions for calcined forms of 1.5% Al-MSU-$S_{(MFI)}$, 1.5% Al-MSU-$S_{(BEA)}$, and 1.5% Al-MCM-41 at 300 °C are provided in Table 1. Clearly, the Al-MSU-S derivatives are far more active acid catalysts than the equivalent Al-MCM-41 prepared from conventional aluminosilicate precursors. These results, together with the steam-stability properties, suggest that MSU-$S_{(MFI)}$ and MSU-$S_{(BEA)}$ may be suitable acid catalysts for the processing of high molecular weight petroleum fractions that can not be adequately refined over microporous zeolites.

We propose that the hydrothermal stability and catalytic activity of MSU-$S_{(MFI)}$ and MSU-$S_{(BEA)}$ arise from zeolite-like subunits of AlO_4 and SiO_4 tetrahedra in the framework walls of the mesostructures. Evidence for the retention of a protozeolitic connectivity of tetrahedra was provided by IR spectroscopy. The band in the 550–600 cm^{-1} region, which is characteristic of five-membered ring subunits,[9] is well-expressed in the spectra of MSU-$S_{(MFI)}$ and MSU-$S_{(BEA)}$, but not for the MCM-41 derivative (Figure 3).

Finally, as we noted earlier, many organic cations are known to function as zeolite-nucleating agents.[7] Further advancements in the steam stability and acidity of aluminosilicate mesostructures may therefore be anticipated through the incorporation of other zeolitic subunits in the framework walls.

Experimental Section

MFI seeds[8a] (Si/Al = 67:1) were prepared by the reaction of 1.0 M tetrapropylammonium (TPA^+) hydroxide (6.7 mmol), sodium aluminate (0.50 mmol, Strem Chemicals, Inc), and fumed silica (33.3 mmol, Aldrich Chemicals) in water (1270 mmol) at 50 °C for 18 h. The same stoichiometric ratio of tetraethylammonium (TEA^+) hydroxide, sodium aluminate, fumed silica, and water was used to prepare a solution of BEA seeds[8b] at 100 °C. MSU-$S_{(MFI)}$ and MSU-$S_{(BEA)}$ mesostructures were assembled by reaction of the respective zeolite seeds solutions with CTAB (8.65 mmol) at 150 °C for

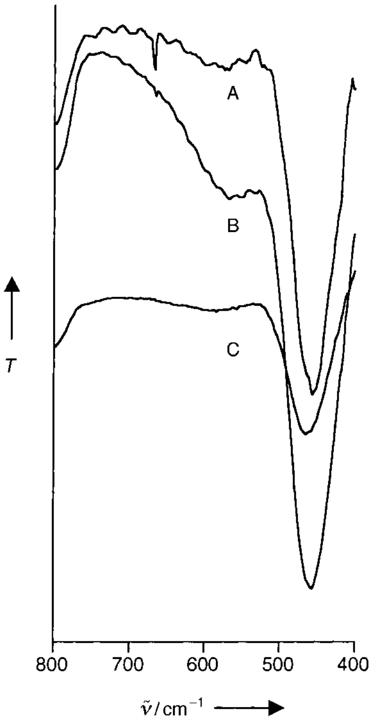

Figure 3. IR spectra of calcined mesostructures: A) 1.5 % Al-MSU-S$_{(MFI)}$ assembled from zeolite ZSM-5 seeds, B) 1.5 % Al-MSU-S$_{(BEA)}$ assembled from zeolite Beta seeds, and C) 1.5 % Al-MCM-41 formed from conventional aluminosilicate precursors.

two days. Al-MCM-41 (Si/Al = 67:1) also was assembled at 150 °C from conventional aluminosilicate precursors formed at 35 °C using tetramethylammonium (TMA$^+$) in place of TEA$^+$ and TPA$^+$.

X-ray diffraction patterns were recorded on a Rigaku Rotaflex diffractometer using Cu$_{K\alpha}$ radiation ($\lambda = 1.542$ Å). N$_2$ adsorption and desorption isotherms were determined on a Micromeritics ASAP 2000 sorptometer at -196 °C. The samples were outgassed under 10^{-5} Torr at 150 °C. ^{27}Al NMR spectra were recorded on a Varian VXR-400S spectrometer with a 7-mm zirconia rotor and a spinning frequency of 4 kHz. IR spectra of samples in the form of KBr pellets were recorded by using a Nicolet IR/42 spectrometer.

Cumene cracking reactions were performed in a 6 mm i.d. fixed bed quartz reactor with 200 mg catalyst. The cumene flow rate was 4.1 μmol min^{-1} in a 20 cm^3 min^{-1} carrier stream of N$_2$. Cumene conversions were reported under steady-state conditions after 60 min on stream at 300 °C.

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Strongly Acidic and High-Temperature Hydrothermally Stable Mesoporous Aluminosilicates with Ordered Hexagonal Structure**

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Mesoporous molecular sieves such as hexagonally ordered MCM-41 have attracted considerable attention because of their potential use as versatile catalysts and catalyst supports for the conversion of large molecules.[1–16] However, the acidity and hydrothermal stability of these mesostructured materials are relatively low compared with those of zeolites, which strongly influences their practical applications in industrial catalytic reactions of petroleum.[1] Despite several successful examples of stabilizing aluminosilicate mesostructures at low temperatures (in boiling water, 100 °C), for example by synthesizing materials with thick pore walls by using triblock copolymers,[3] by removing silanol groups by silylation,[14] by employing salt effects,[15] by using the co-templating approach,[16] and by post-treatments,[5–8] the acidity and hydrothermal stability of these mesostructures at high temperature (800 °C) are still lower than those of microporous aluminosilicate zeolites.

It is well known that microporous zeolites are very stable, commercial catalysts, and the MFI-type and Beta zeolites, which contain structure building units of 5-rings (T-O-T, T = Si and Al), exhibit extremely high acidity and hydrothermal

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