A Novel Method for the Preparation of Zeolite ZSM-5

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The zeolite ZSM-5 has been synthesized from amorphous aluminosilicate gels in a vapour of ethylenediamine, triethylamine, and water and characterized by XRD and chemical analysis.

The current technique for preparing zeolites is the hydrothermal method.[1] Many types of high-silica zeolites, *e.g.*, ZSM-5,[1] can be synthesized using this method and employing organic materials as templates. Recently, however, Bibby and Dale have synthesized silica- and alumino-sodalite from ethylene glycol and propanol solvents.[2] Following this high-silica zeolites, *e.g.*, ZSM-5,[3] ZSM-35,[3] silicalite-1,[4] ZSM-39,[4] and ZSM-48,[4] have been synthesized from the organic solvents. Both methods have an identical step of contacting the amorphous gels with the solvent phase. The zeolite products are separated from the solvent phase by filtration or centrifugation. As the liquid-phase is separated, a portion of the organic material is wasted which adheres onto the zeolite surface and exists in the liquid-phase. Here we report a new method for synthesizing zeolites, the vapour-phase method, which could decrease the consumption of organic materials. Using this method zeolite ZSM-5 has been prepared.

During the preparation solutions of aluminium sulphate, sodium silicate, and sodium hydroxide are mixed and stirred for 15 min. This mixture is filtered and washed. Then, the amorphous gels are put into the container, the bottom of which possesses sieves. Ethylenediamine (EDA), Et_3N, and H_2O are put into a special reactant autoclave. Then, the container is placed on the supporter of the autoclave (see Figure 1). The autoclave is sealed and left to stand in a dry oven. The reaction is carried out at 453—473 K for 5—7 days. During heating, a mixed vapour of EDA, Et_3N, and H_2O forms in the autoclave. The gels are closed in with the mixed vapour, but the gels do not come into contact with the mixed solution of the solvents directly. When the reaction is complete, the solid samples are washed with distilled water and dried at 378 ± 2 K. *X*-Ray powder diffraction and chemical analysis characterize the synthesized samples. Typical results are shown in Table 1.

Table 1. Typical experimental results.

Run	Reactant composition/molar ratio		Crystallization conditions		Products	
	Amorphous Gels	Liquid-phase	*T*/K	*t*/days	SiO_2/Al_2O_3	Phase
1	$1.4Na_2O : Al_2O_3 : 44.8SiO_2 : 228H_2O$	$1.0EDA : 4.3Et_3N : 2.8H_2O$	473	5	44.8	ZSM-5
2	$1.5Na_2O : Al_2O_3 : 86.4SiO_2 : 325H_2O$	$1.0EDA : 7.7Et_3N : 10H_2O$	453	5	86.4	ZSM-5
3[a]	$1.5Na_2O : Al_2O_3 : 86.4SiO_2$	$1.0EDA : 7.7Et_3N : 10H_2O$	453	7	86.4	ZSM-5

[a] The amorphous gels were dried at 823 K.

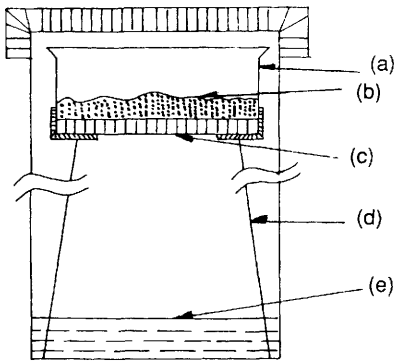

Figure 1. Reactant autoclave: (a) container; (b) amorphous gels; (c) porous sieve plate; (d) stainless steel supporter; (e) solution phase.

The reactant products taken from the autoclave appear damp. After washing and drying, the samples are identified by *X*-ray powder diffraction. The powder diffraction patterns (location and intensities of peaks) of the prepared samples are similar to those of the zeolite ZSM-5.[1] In runs 1 and 2 (Table 1) the molar ratios of SiO_2 to Al_2O_3 of the products are 44.8 and 86.4, respectively. These are constant before and after the reaction (see Table 1). Even when gels are dry, zeolite ZSM-5 can still be synthesized (run 3). These results lead us to believe that the crystallization process of ZSM-5 belongs to the solid-phase transformation process.

In summary, we report a new technique for preparing zeolites, the vapour method. The process eliminates the waste liquids which normally occur during the synthesis of zeolites. The mixed solvents in this experiment could be circulated and used several times. The method therefore decreases the consumption of amines and so synthesizes zeolites more cheaply. The method also could raise zeolite productivity.

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