

Synthesis and Size Tailoring of Colloidal Zeolite Particles

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A novel method is presented whereby it is possible to synthesize colloidal zeolite particles of a pre-determined size.

The synthesis of colloidal dispersions of amorphous silica and aluminosilicate particles with a narrow particle size distribution and the method of size tailoring has been described in the literature.¹⁻³

We have developed a method whereby stable colloidal

suspensions of zeolite (crystalline aluminosilicate) essentially free from unreacted amorphous material can be prepared from clear homogeneous solutions. The sols display the typical characteristics of colloidal suspensions such as Tyndall light scattering and a low rate of sedimentation. The particle size is

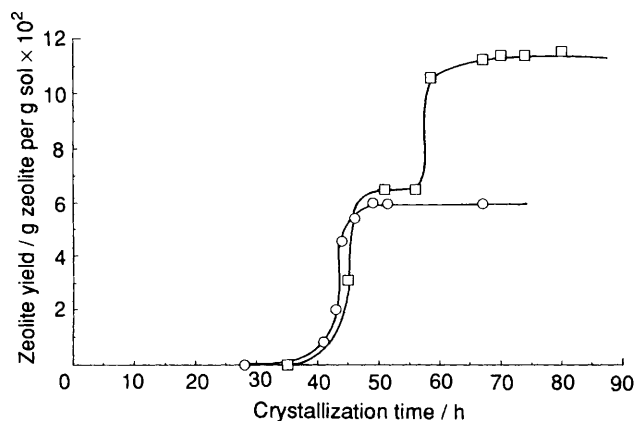


Fig. 1 The increase in zeolite yield as a function of crystallization time in run A ○ (standard run) and run B □ (mid-synthesis addition of alumina after a crystallization time of 55 h)

<100 nm and in some cases, <50 nm with a narrow particle size distribution.^{4,5}

The nature of the crystallizing system is such that the growth limiting nutrient for a given system can be identified and thereafter added at some critical time during the crystallization. The mid-synthesis addition of this nutrient thus provides a method whereby the zeolite particles can be size tailored if the initial particle size is known (as by for example light scattering).

One example of this method is the synthesis of discrete colloidal hydroxysodalite particles from a clear homogeneous solution with the molar composition 14 (TMA)₂O:0.85 Na₂O:Al₂O₃:40 SiO₂:805 H₂O (where TMA = tetramethylammonium). The solution is heat treated at 100 °C with reflux and without stirring. After a crystallization time of 55 h, a colloidal hydroxysodalite suspension, denoted run A (Fig. 1), with an ultimate particle size of 37 nm is obtained. The particle size is determined by dynamic light scattering using dilute as-synthesized sol samples. The discrete nature of the particles as well as the fact that the average particle size is less than 50 nm is confirmed by scanning electron microscopy. The ultimate product as well as intermediate products can be characterized with respect to zeolite yield by isolating the

zeolite from the soluble amorphous material with centrifugation, typically 2 h with a relative centrifugal force of 49 000 g. The mass growth curve is shown in Fig. 1. In this manner, the zeolite yield was determined as 6×10^{-2} g zeolite per g sol and a particle number of 1.1×10^{15} per g sol. A freeze-dried sample of the (by centrifugation) purified sol analysed by X-ray diffraction shows that the sample is hydroxysodalite, essentially free from amorphous material and having a unit cell constant of 8.921 Å. The specific surface area of this sample as measured by N₂ adsorption was 185 m² g⁻¹.

In a separate run, run B, (which is merely a rerun of run A with reproducible results), alumina (the growth limiting nutrient) is added after a crystallization time of 55 h in an amount corresponding to the alumina content at the start of crystallization. The mid-synthesis addition of alumina results in the growth of the particles to 48 nm. The zeolite yield in the ultimate product increases to 12×10^{-2} g zeolite per g sol and the particle number is constant. The response of the system to the addition of alumina is shown in Fig. 1, run B. The unit cell constant of the product is essentially unchanged, 8.931 Å, while the measured specific surface area shows a reduction from 185 m² g⁻¹ to 142 m² g⁻¹.

Doubling the total alumina content in the system results in a doubling of the zeolite yield with the corresponding increase in particle size, a decrease in the specific surface area as well as an unchanged particle number. These results show that the mid-synthesis addition of alumina allows for the growth upon existing particles in solution without the occurrence of a secondary nucleation event thereby allowing for the possibility to size tailor colloidal zeolite particles.

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