Catalysis Letters 25 (1994) 401–402

COMMENT

Dealumination of zeolite Y by H_4EDTA

George T. Kerr [a], Arthur W. Chester [b] and David H. Olson [c]
[a] *10 Pin Oak Drive, Lawrenceville, NJ 08648, USA*
[b] *Paulsboro Research Laboratory, Mobil Research and Development Corp.,*
Paulsboro, NJ 08066, USA
[c] *Central Research Laboratory, Mobil Research and Development Corp., PO Box 1025,*
Princeton, NJ 08540, USA

Received 12 January 1994

In this journal in a recent paper, bearing the above title, Datka, Kolodziejski, Klinowski and Sulikowski stated in their abstract, "Unlike with hydrothermal dealumination or treatment with $SiCl_4$, H_4EDTA gives defective crystals in which framework vacancies created by the expulsion of aluminum are not healed by silicon from other parts of the framework. The likely reason for this is the slow intracrystalline diffusion of $Si(OH)_4$ at low temperature" [1]. The second statement of Datka et al. is probably correct but the first statement is correct only if aluminum removal from the framework occurs at a significantly faster rate than the rate of intracrystalline diffusion of a Si-containing species which can effect healing of defects. The removal of aluminum from zeolite Y at 100°C using H_4EDTA was first reported by Kerr in 1968 and the first sentence in the abstract of that paper stated, "The slow addition of a dilute solution of ethylenediaminetetraacetic acid to sodium zeolite Y effects the removal of aluminum from the zeolite" [2]. Kerr showed that immediate addition of H_4EDTA to the zeolite resulted in a product with low crystallinity whereas addition of the acid to the zeolite over an eighteen-hour period yielded a product with good crystallinity. It is apparent that Datka et al. added H_4EDTA to the zeolite rapidly as they stated, "Dealumination was carried out by 1 h treatment..." of the zeolite with the acid.

Ten years later, in 1978, Kerr, Chester and Olson reported that when the rate of aluminum removal from zeolite Y was controlled at 10% removal per day at 100°C, up to 80% removal could be achieved with retention of most of the crystal structure. These authors stated, "Thus, if the removal of aluminum proceeds at a faster rate than the migration of silicon into defect sites, then a catastrophic defect situation arises and the crystal structure collapses. Should the exchange of hydronium ion into the zeolite (and thus removal of framework aluminum) proceed at a rate equal to or somewhat less than the rate of silicon migration, then the zeolite can "heal" itself and retain most of the crystallinity even up to quite high levels of aluminum removal" [3].

It is highly probable that solid state (MAS) NMR spectra for ^{27}Al and ^{29}Si of zeolite Y samples dealuminized as recommended by Kerr, Chester and Olson would show more complete occupancy of framework sites by silicon and aluminum.

References

[1] J. Datka, W. Kolodziejski, J. Klinowski and B. Sulikowski, Catal. Lett. 19 (1993) 159.
[2] G.T. Kerr, J. Phys. Chem. 72 (1968) 2594.
[3] G.T. Kerr, A.W. Chester and D.H. Olson, Acta Phys. Chem. 24 (1978) 169.