

recognizing the wide variation possible within each form, we respectfully suggest that other workers in this field use the terms "hydrogen zeolite," "dehydroxylated zeolite," and "ultrastable zeolite" wherever possible.

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### In Answer to Kerr, Cattanach, and Wu\*

In the preceding Letter to the Editors, Kerr *et al.* have commented that we have implied that the "decationized Y" catalyst we used was largely the dehydroxylated form. They further commented that, in our reaction mechanism, we used Lewis acid sites and surface fields to the exclusion of the protonic sites. In reply, we would like to make it clear that we did not imply the "decationized Y" catalysts in our work as the nearly fully dehydroxylated form. Neither did we neglect the important role played by the surface protonic acids in a general sense. In fact, exactly the contrary is true. For instance, we had stated in our paper that the surface protons were uniquely important for a host of chemical reactions. We cited alkylation, dealkylation, and olefin polymerization as examples. These statements implied that, with the exception of "carbon-hydrogen bond activation," which we singled out, most of the other reactions of the carbonium type were, in our view, attributable to surface protonic sites.

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Decationization, in our paper, was defined as calcining the ammonium form at high temperature. It embodied two steps—the deamination and the dehydroxylation. This definition was represented in our paper by a scheme which we took after Uytterhoven, Christner, and Hall (1). Our definition paralleled that of Rabo *et al.* (2), who defined the decationized Y as a Y zeolite in which a substantial portion of the metal cations have been replaced by protons or ammonium cations and which have been subsequently heat-treated at temperatures of between 350° and 600°C. One might, of course, disagree with our choice of the definition, but then the argument is merely a matter of semantics. The chemistry of the decationization is well documented; such as, in the works cited by Kerr *et al.* (3, 4) and by others (1, 5). Thermogravimetric work of Ward (5) and of Benesi (4) suggested that dehydroxylation started at 500°C in flowing helium. We calcined our samples at 550°C in flowing nitrogen because above 560°C the crystal structure of decationized Y would partially collapse. (6) In the samples we used, therefore, protons, Lewis acids, and elec-

tric fields all coexisted. It is noteworthy to point out that only a relatively small number of effective sites are usually needed to account for the observed chemical activity. In cumene cracking over decationized Y, a concentration of  $5.75 \times 10^{16}$  sites/g or approximately one site in every 10 000 cavities was found to be sufficient to explain the catalytic conversion (1).

We emphasized the Lewis acids and the electrical fields rather than the protonic acids not because of the relative absence of the latter sites but rather because the former two kinds explained the phenomena observed. Thus, the hexane-induced catalyst deactivation could be most straightforwardly explained by the surface reconstitution such as through oxide diffusion. The "carbon-hydrogen bond activation" was singled out from the other reactions of the carbonium ion type because there was a drastic difference in reactivities toward this reaction between the amorphous and crystalline aluminosilicates. While the conventional silica-alumina virtually does not activate the carbon-hydrogen bonds, the decationized Y could activate them with considerable facility. Because the differences was so drastic, we felt that it might reflect a difference of the reactive sites in kind (e.g., dynamic Lewis acids versus static Lewis acids) rather than in degree (e.g., stronger and more numerous Lewis acids or protonic acids). Based upon this premise, we proceeded to formulate mechanisms and found that plausible mechanisms could indeed be worked out which would satisfactorily account for the drastic difference stated. The mechanisms served to illustrate a principle—dynamic sites may behave entirely differently from the static sites. The reasoning which led us to consider the dynamic Lewis acids and electric fields as the responsible sites in the carbon-hydrogen bond activation is admittedly a casual one, and our proposed mechanisms do not preclude other alternative mechanisms. Should an alternative mechanism

(e.g., based upon protonic sites) be proposed in the future that will also account for the drastic reactivity difference and the hexane-induced deactivation, then the contest of validity among the proposals must await future experimental evidence. It should be pointed out that the experimental evidence used for such purposes must adequately take the catalyst deactivation into consideration. Initial activities, being transient (in the case of hexane on decationized Y), may not be particularly meaningful.

Finally, we wish to point out that, while the concept of the dynamic sites was derived from the needs of explaining the hexane-induced deactivation and the rather drastic difference in reactivities toward carbon-hydrogen bond activation between the amorphous and the crystalline aluminosilicates, the merits of the dynamic concept stand above the validity of the explanations. If the explanations we offered need modification in view of future experimental evidence, the concept of dynamic sites and its implications to reaction kinetics will, nevertheless, still stand.

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