LETTERS TO THE EDITORS

Some Comments on Ambs and Flank's Paper, "Thermal Behavior of Synthetic Faujasite"

In their recent paper, Ambs and Flank (1) (AF) examine the thermal stability of the calcination products of materials belonging to the NaY-NH₄Y system. Their results and conclusions appear to be misleading inasmuch as they failed to recognize the differences between hydrogen zeolite Y and ultrastable faujasite. The AF data show the effect of sodium content on the thermal stability of ultrastable faujasite; it does not relate to the effect of sodium content on the thermal stability of the thermal stability of the st NH₄Y may produce two quite different limiting cases depending upon the partial pressure of water and/or ammonia. Because ammonium-exchanged zeolite Y gives off large amounts of water and ammonia when heated, sample geometry and packing play an important role in determining the nature of the calcination product (3). When $P_{\rm H20}$ = 0, and/or other conditions pointed out by Kerr are met, NH₄Y \rightarrow HY \rightarrow dehydroxylated Y(Y^{*}). These reactions conventionally are represented by the equations:

normal or true hydrogen zeolite Y which Kerr (2, 3, 5) has shown to be chemically (and hence structurally) different from ultrastable zeolite Y. These differences and certain other aspects of the AF paper are discussed in greater detail below.

Kerr has shown that the calcination of

When $P_{\rm H_{20}} > 0$, Kerr has shown that NH₄Y and/or HY combine with water to produce materials from which measurable amounts of aluminum have been removed from the framework (4, 5); these materials we refer to as ultrastable faujasites. Examples are shown in Structures 3 and 4:

It seems clear to us that AF produced ultrastable faujasites (2, 3, 5) by their calcination of tightly compacted NH₄Y samples in their DTA runs (6). Samples prepared by this technique were subsequently subjected to X-ray analysis by AF. We disagree with the statement in the introduction of AF that "the ultrastable faujasite is not intrinsically different in structure from other ammonium-exchanged faujasites. . . ." In those regions of the zeolite where condensation of OH has produced structure 3 containing new Si-O-Si bonds, we believe that hexagonal prisms may have been converted to structures containing 3-rings, and adjacent site II 6-rings converted to highly distorted 8-rings.

Kerr has found that lattice collapse occurs at the indicated temperatures for the following zeolites: ultrastable zeolite Y, 1050° (3); sodium zeolite Y, 974° (4); hydrogen zeolite Y, 850° (5). From these results, it would be expected that the thermal stability of NaHY should *decrease* with decreasing sodium content; the opposite effect would be expected for the Na ultrastable faujasite. Since the system studied by AF became more stable with decreasing sodium content, it is clear that they were studying the Na ultrastable faujasite system and not the NaHY system as they supposed.

We must also point out that AF have misinterpreted the following aspects of our work: They seem to be under the impression that one of us (2) proposed that water is somehow involved in the crystal collapse of sodium zeolite Y at elevated temperatures. The studies reported in Ref. (2) concerned reactions of water with hydrogen zeolite Y—not with the sodium form. AF assumed that the sodium zeolite Y and aluminum-deficient sodium zeolite Y samples of Ref. (4) were calcined before differential thermal analyses were run. They were not. The paper clearly states that the samples were calcined prior to the sorption measurements (4).

Finally, AF implied that the results in Ref. (2) conflict with those reported in Ref. (7). Kerr (2) described the ultrastable zeolite Y which contains some hexacoordinate aluminum while Olson and Dempsey (7) described the normal hydrogen faujasite which contains only tetracoordinate aluminum. We see no conflict. As stated in Ref. (7), the final difference maps were free of any extraneous scattering matter. The background level was ≤ 0.5 e/A^3 . Hence, we can state with a relatively high degree of certainty that there were no metal cations (including Al) in nonframework positions.

References

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