

Reply to 'An Investigation of the Postulate of 'Molecular Traffic Control' in Zeolite ZSM-5' by C. G. Pope

The concept of molecular traffic control (MTC) in zeolites has been discussed in several notes published recently in this journal (1-4). Although the proposal originated from single-component adsorption measurements near equilibrium (1), it was emphasized that the model intended to describe a dynamic situation (1, 3). It implies that at stationary state, i.e., under conditions where the concentration gradients for the reactants and products are maintained constant, reactants may "reach the catalytic sites through diffusion pathways that are less readily accessible to the products which are desorbing, . . . and vice-versa" (3).

The above requirements are clearly not met in the experiments described by Pope (4) and hence, as observed, his measurements are unlikely to lend support to the existence of a MTC effect.

It has been well appreciated that the interpretation we proposed for our adsorption data (1) could be questioned. In particular, we considered the possible role of intermolecular interactions in the ordering of the probe molecules in the pores (3), thereby rendering adsorption measurements sensitive to a variety of factors. The possible occurrence of packing effects is a valuable suggestion (4) and there is certainly a need for a more accurate description and characterization of the molecular ordering in the pores of zeolites.

We feel however that the MTC concept should be dissociated from adsorption mea-

surements although they triggered its proposal. Support should be sought from kinetic data. An investigation of the alkylation of *para*-xylene by methanol over H-ZSM-5 and H-ZSM-11 catalysts (5) has been performed in this respect.

A thorough understanding of the data reported by Pope (4) also requires clarification of several points. Firstly, the lower hydrocarbon adsorption capacities which he reports must be explained. Secondly, the desorption of the first adsorbate should be monitored during the adsorption of the second one. Indeed, it is likely to occur and if so, it will affect the meaning of the gravimetric measurements. For example, the latter effect could explain partially the poor agreement between the simple volume-filling model and the experimental data which he reports for zeolite NaY. Finally, molecules which are adsorbed at intersections in a localized mode because of their interaction with acidic sites, restrict the size of some channels (6) and decrease their accessibility to other molecules. Such an effect could occur to some extent in the adsorption of *n*-pentane following that of toluene on a protonated ZSM-5 zeolite such as that used in his study. These eventualities need further attention.

REFERENCES

1. Derouane, E. G., and Gabelica, Z., *J. Catal.* **65**, 486 (1980).
2. Lowe, B. M., Whan, D. A., and Spencer, M. S., *J. Catal.* **70**, 237 (1981).

3. Derouane, E. G., Gabelica, Z., and Jacobs, P. A., *J. Catal.* **70**, 238 (1981).
4. Pope, C. G., *J. Catal.* **72**, 174 (1981).
5. Derouane, E. G., Dejaifve, P., Gabelica, Z., and Védrine, J. C., *Faraday Discuss. Chem. Soc.* **72**, in press.
6. Gabelica, Z., Gilson, J. P., and Derouane, E. G., Proceedings of the Second European Symposium of Thermal Analysis, Aberdeen, September 1981, in press.

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