interface. Furthermore, in both the drop and the cavity experiment, the experimental data follow the same trend as the theory, which is very encouraging. It might be noted that in both the analysis and the experiment the effect of the dynamic properties on the frequencies was insignificant. Addition of surfactants lowered the frequency, but this was entirely commensurate with the lower interfacial tension.

The main conclusion drawn from this analysis and experiment is that working with different systems (gas-liquid, liquid-liquid, liquid-gas) in different size ranges, all the interfacial properties can presumably be backed out for a given surfactant or even a pure fluid. The significance (or the lack) of these properties can therefore be estimated. A large number of such similar experiments needs to be done before final conclusions can be made.

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# Diffusion of Hydrocarbons in 13X Zeolite

The kinetics of sorption of four representative hydrocarbons in 13X zeolite crystals have been investigated at temperatures within the range 409 to 513°K and pressures 0.1 to 100 torr. Extensive diffusivity data are presented showing the dependence on sorbate concentration and temperature. The form of the concentration dependence of the diffusivity for these systems is very similar to that of the small monatomic and diatomic molecules in 5A zeolite. This similarity is understandable, since the key factor which determines the diffusion behavior is the relative size of diffusing molecule and sieve window. For these hydrocarbons in the 13X sieve, as for the monatomic and diatomic gases in 5A, the critical diameters of the diffusing molecules are all appreciably smaller than the free diameter of the windows.

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## SCOPE

Despite the widespread application of the X and Y zeolites as cracking catalysts, available information on the diffusion of hydrocarbons in these materials is sparse. This is because the crystal size of the commercially available X and Y zeolites is small (~ 1$\mu$m), and the diffusional time constants are therefore too small to be determined by conventional techniques. In the present study, a specially prepared sample of 13X zeolite with crystal size ~16.5$\mu$m was used in order to achieve sorption rates which were slow enough to measure gravimetrically. Diffusivities were determined for four representative hydrocarbons ($nC_7H_{16}$, $C_6H_{12}$, $C_6H_6$, and $C_6H_5 \cdot CH_3$) over a range of temperatures from 409° to 513°K and pressures 0.1 to 100 torr. The concentration and temperature dependence of the diffusivity have been studied in detail, and comparative data for the diffusion of $nC_7H_{16}$ in 5A zeolite are presented for a similar range of pressure and temperature.

## CONCLUSIONS AND SIGNIFICANCE

Diffusivities for the four hydrocarbons in 13X zeolite are strongly concentration dependent, and the pattern of concentration dependence is similar to that observed previously for the diffusion of small monatomic and diatomic molecules in 5A zeolite (Ruthven and Derrah, 1975) and distinctly different from the behavior of the light hydrocarbons in the A zeolite (Ruthven, Loughlin, and Derrah, 1973). Over the range of the experimental measurements, the diffusivities for the $C_6$ and $C_7$ species in 13X sieve are of order $10^{-8} - 10^{-9}$ $cm^2 \cdot s^{-1}$, which is more than an order of magnitude greater than the diffusivities for *n*-heptane in 5A sieve under comparable conditions. The diffusional activation energies for the $C_6$ and $C_7$ hydrocarbons in 13X sieve range from 5.0 to 6.2 kcal, and there is no correlation with critical molecular diameter. By contrast, activation energies for the light hydrocarbons in the 5A sieve show a clear correlation with molecular diameter and a different pattern of concentration dependence of diffusivity. The key factor which determines the diffusional behavior is the relative size of the diffusing molecule and the sieve window. On this basis, the similarities of the present systems with the behavior of small monatomic and diatomic molecules in the 5A sieve and the pronounced differences from the behavior of the hydrocarbons in 5A sieve are clearly understandable.

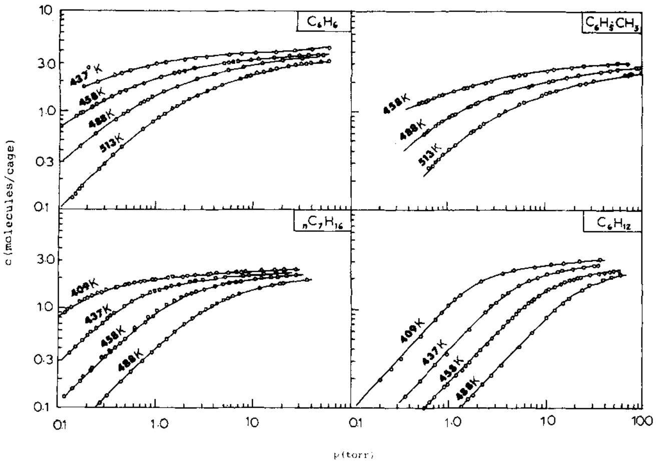

**Fig. 1. Equilibrium isotherms for sorption of $nC_7H_{16}$, $C_6H_{12}$, $C_6H_6$ and $C_6H_5 \cdot CH_3$ in 13X zeolite crystals.**

Because of the industrial importance of molecular sieve processes, diffusion in zeolites has been extensively studied. Available information has been summarized in several recent reviews (Barrer, 1971; Ruthven, 1975). Previous studies have, however, been concerned mainly with diffusion in the small port zeolites such as zeolite A, chabazite, and erionite. For the diffusion of hydrocarbons in the more open structures of the X and Y zeolites, only very limited data are available, although such information is of considerable practical importance in view of the widespread use of these zeolites as cracking catalysts. In this laboratory we have studied the diffusion of a series of hydrocarbons (Ruthven, Loughlin, and Derrah, 1973) and other small molecules such as the inert gases, oxygen and nitrogen in the 4A and 5A sieves (Ruthven and Derrah, 1975), and in natural erionite (Ruthven and Derrah, 1975). These studies revealed a complex pattern of behavior which indicated that the diffusion mechanism depends upon the relative sizes of the diffusing molecule and the zeolite micropore. The present investigation of the diffusion of four representative hydrocarbons (*n*-heptane, cyclohexane, benzene, and toluene) in 13X zeolite crystals was therefore undertaken as a logical continuation of this work.

## EXPERIMENTAL

Transient sorption curves were determined experimentally by using a Cahn vacuum microbalance system to follow the response of a small sample of zeolite crystals (~40 mg) when subjected to a small differential step change in sorbate pressure. The volume of the system was sufficiently large to ensure that, after the initial step, the pressure remained essentially constant throughout the course of an experiment. Pressures were measured on a Barocel manometer.

The crystal size of the commercially available X and Y sieves is small (~1 μ), and since the zeolitic diffusion coefficients are relatively large owing to the open pore structure, the diffusional time constants ($r^2/D$) are small and difficult to measure. Preliminary experiments, carried out in the vacuum microbalance system with Linde 13X crystals (Union Carbide), showed a rapid initial adsorption when the half time was less than the response time of the system (~5 s) followed by a much slower approach to equilibrium. Estimates of heat transfer rates showed that the slower process which was observed in the final 20% of the uptake curve could be due to thermal equilibration. In order to determine intracrystalline diffusivities, it was necessary to use specially prepared larger zeolite crystals. The sample used in the present study was kindly provided by Professor H. Pfeifer and Dr. J. Kärger of the Karl-Marx University, Leipzig. Mean crystal diameter as determined by optical photomicrography was 16.5 μ, and the size distribution range was quite narrow.

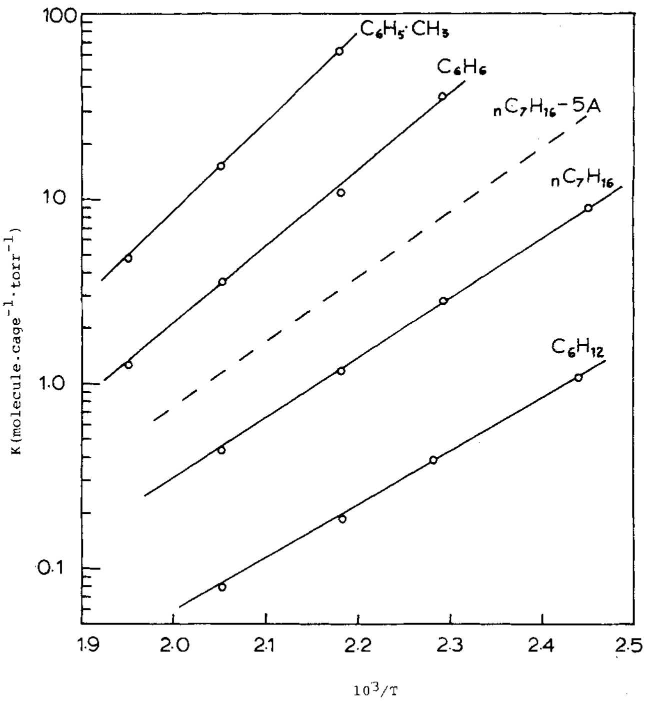

**Fig. 2. vant Hoff plot showing temperature dependence of Henry constants for sorption in 13X zeolite.**

Diffusional time constants and hence the diffusivities were calculated from the solution of the Fickian diffusion equation for a system of uniform spherical particles, subjected to a step change in surface concentration at time zero (Crank, 1956):

$$\frac{m_t}{m_\infty} = 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp(-n^2\pi^2 Dt/r^2) \qquad (1)$$

In view of the octahedral crystal habit and the relatively narrow size distribution, this is a reasonable approximation. Prior to a run, the zeolite sample was degassed at 400°C, $10^{-5}$ torr for at least 12 hr. Sorbate concentrations have been expressed as molecules per cage by using the conversion factor 1 mmole/g dehydrated zeolite = 1.68 molecules/cage, which is based on the known density of the crystal and the dimensions of the unit cell.

The temperature range for the experimental study (400° to 500°K) was selected in order to cover the entire range of concentrations from the Henry's law region to saturation over a convenient pressure range (0.1 to 100 torr).

## RESULTS AND DISCUSSION

### Equilibrium Data

Equilibrium isotherms for the four hydrocarbons at several temperatures are shown in Figure 1. The curves show the expected approach to saturation at high pressures and to Henry's law in the low concentration region. The data are consistent with previously reported isotherms (Barrer and Sutherland, 1956; Barrer, Bultitude, and Sutherland, 1957; Bartholomeuf and Ha, 1974), but since the present isotherms extend to lower concentrations, the Henry's law constants may be obtained with greater confidence. At the higher temperatures, the Henry constants (defined by $c = Kp$) were obtained directly from the limiting slopes of the isotherms, while at the lower temperatures the values were obtained from the intercepts of plots of $\ln(p/c)$ vs. $c$, extrapolated to zero concentration (Barrer and Davies, 1970, 1971). The vant Hoff plot showing the temperature dependence of the Henry constants is shown in Figure 2, while the values of $K_0$ and $q_0$, calculated according to Equation (2), are given in Table 1:

TABLE 1. KINETIC AND EQUILIBRIUM CONSTANTS FOR SORPTION IN 13X ZEOLITE

| Sorbate | $\sigma$ (Å) | $D'_* \times 10^9$ ($cm^2 \cdot s^{-1} \cdot$ molecule$^{-1}$ cage) | $E$ (kcal · mole$^{-1}$) | $K_o \times 10^7$ (molecule · cage$^{-1}$ · torr$^{-1}$) | $q_o$ (kcal · mole$^{-1}$) |
|---|---|---|---|---|---|
| $nC_7H_{16}$ | 5.1 | 22.0 | 6.20 | 0.71(0.35) | 15.2(16.7) |
| $C_6H_{12}$ | 6.6 | 10.2 | 4.96 | 0.78 | 13.4 |
| $C_6H_6$ | 7.26 | 4.9 | 5.0 | 0.18 | 18.5 |
| $C_6H_5 \cdot CH_3$ | 7.26 | 6.6 | 5.6 | 0.06 | 21.0 |

Values of $K_o$ and $q_o$ in parenthesis are for sorption of *n*-heptane in 5A zeolite over the same temperature range. The critical diameter $\sigma$ is defined as the diameter of the smallest cylinder which can circumscribe the molecule in its most favorable equilibrium conformation.

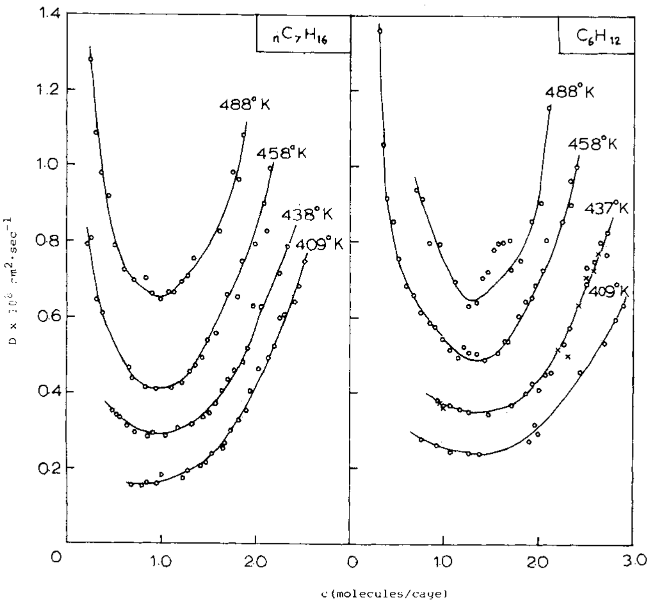

**Fig. 3. Variation of diffusivity with sorbate concentration for $nC_7H_{16}$ and $C_6H_{12}$ in 13X zeolite.**

$$K = K_o \exp(q_o/RT) \qquad (2)$$

The difference in the limiting heats of sorption of toluene and benezene (~2.5 kcal) is approximately consistent with the group contribution expected from an additional methyl group (Avgul, Bezus, and Dzhigit, 1971). Comparison with experimental data obtained over the same range of temperatures shows that the Henry constants for heptane in 13X are about one third of the values for sorption in the 5A zeolite.

**Diffusivities**

The kinetic data are shown in Figures 3 and 4 in which the Fickian diffusivities are plotted against sorbate

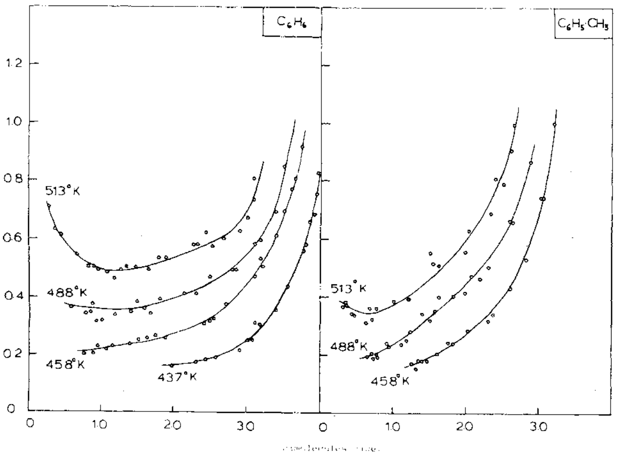

**Fig. 4. Variation of diffusivity with sorbate concentration for $C_6H_6$ and $C_6H_5 \cdot CH_3$ in 13X zeolite.**

concentration. The pronounced concentration dependence of the diffusivity is of the same form as has been reported previously for small monatomic and diatomic molecules in 5A zeolite (Ruthven and Derrah, 1975). Within the Henry's law region ($c << 1$ molecule/cage), the diffusivity decreases rapidly with increasing sorbate concentration, passing through a minimum at intermediate concentrations and increasing again as saturation is approached. Corrected diffusivities ($D_o$) were calculated from Darken's equation on the assumption that the true driving force for the zeolitic diffusion process is the gradient of chemical potential rather than the concentration gradient:

$$D = D_o(d \ln p/d \ln c) \qquad (3)$$

The factors ($d$ ln $p/d$ ln $c$) were determined directly from the empirical isotherms by differentiation.

Values of $D_o$ are plotted against sorbate concentration, on logarithmic coordinates, in Figure 5. For both *n*-heptane and cyclohexane, the resulting plots lie close to straight lines of unit negative slope, indicating that to a first approximation

$$D_o = D'_o/c \qquad (4)$$

where $D'_o$ is a temperature dependent constant. Comparative data for the diffusion of *n*-heptane in 5A zeolite over the same range of temperatures and sorbate concentrations are also shown in Figure 5 (Ruthven and Doetsch, 1974). The difference in behavior for these two sieves is striking, since for the 5A sieve, $D_o$ is almost constant and of order $10^{-11}$ – $10^{-10}$ $cm^2 \cdot s^{-1}$, whereas the values of $D_o$ for the 13X sieve are about 100 times larger and vary inversely with concentration.

The same form of inverse concentration dependence of $D_o$ is also shown by benzene and toluene in the high concentration region, but for these species the concentration dependence becomes less pronounced at low concentrations. The values of $D'_o$ calculated from the experimental data are plotted against reciprocal temperature in Figure 6. It is evident that for all four hydrocarbons the temperature dependence of this parameter may be adequately correlated by an Arrhenius equation:

$$D'_o = D'_* \exp(-E/RT) \qquad (5)$$

Values of the parameters $D'_*$ and $E$ are given in Table 1. The diffusional activation energies of all four species are similar, and there appears to be no correlation with molecular diameter.

As a result of the difficulties involved in the experimental measurement of diffusivities in the commercially available X and Y sieves only, very limited data are available for comparison with the results of the present study. An ingenious chromatographic method involving the use of different carrier gases to separate the contributions from micropore and macropore diffusional resistance was used by MacDonald and Habgood (1972) to estimate diffusivities of benzene and octane in commercial

NaX(13X) zeolite. The reported diffusivities at temperatures of 370° to 430°C were within the range 1 to 3 × $10^{-9}$ $cm^2 \cdot s^{-1}$. These values are of the same order as the present values, although the temperatures of the present study are considerably lower. A basic assumption of the chromatographic method is that the diffusivity is independent of concentration, at least within the Henry's law region. The present results indicate that this assumption is certainly not justified for these systems, and the diffusivity values derived from the chromatographic method are therefore open to question.

In a recent paper, Ma and Lee (1976) have attempted to extract zeolitic diffusivities for light hydrocarbons in 13X sieve using a two resistance diffusion model similar to that of Ruckenstein, Vaidyanathan, and Youngquist (1971) to account for the mass transfer resistances of both the macropores of the pellet and the micropores of the zeolite crystal. At temperatures of about 300°K, diffusivities for $n$-butane, iso-butane, and 1-butene were found to be of the order $10^{-14}$ – $10^{-15}$ $cm^2 \cdot s^{-1}$. These values, which are of the same order as the diffusivities for typical ionic solids at temperatures well below the melting point (for example, $Pb$ in $PbI_2$ at 450° to 500°K) (Jost, 1952), seem very low considering the nature of the intracrystalline fluid. The present values ($10^{-8}$ – $10^{-9}$ $cm^2 \cdot s^{-1}$), which are of the same order as the diffusivities for typical organic molecular solids at the melting point (Chadwick and Sherwood, 1971), are physically more reasonable. Two critical assumptions in the model used by Ma and Lee are linearity of the equilibrium isotherm over the relevant concentration range, and constancy of the zeolitic diffusivity. Comparison with the equilibrium data of Barrer and Sutherland (1956) shows that the isotherms are quite nonlinear, and the present results suggest that the approximation of a constant diffusivity is not justified. It is possible that the errors inherent in these approximations may be responsible for the large discrepancy in the diffusivities obtained by Ma and Lee.

A lower limit to the zeolitic diffusivity may be established entirely independently by considering kinetic data for catalytic cracking reactions. Miale, Chen, and Weisz (1966) showed that at 500°K in an active rare earth exchanged type X catalyst the first-order rate constant for $n$-hexane cracking is about 0.008 $s^{-1}$. Analysis of the activation energies for different catalysts showed that the reaction is not significantly affected by intracrystalline diffusion, so one may conclude that the Thiele modulus ($\phi = r\sqrt{k/D}$) must have been smaller than about 1.0. With $r \sim 10^{-4}$ cm, this gives $D > \sim 10^{-10}$ $cm^2 \cdot s^{-1}$ as a lower limit. This is consistent with the present data for diffusion of $n$-heptane at the same temperature.

Extensive studies of counterdiffusion in zeolites of the faujasite type have been carried out by Satterfield and Katzer (1971) and Moore and Katzer (1972). For the counter diffusion of benzene-cumene, benzene-cyclohexane, and toluene-cyclohexane in NaY zeolite at 300°K, diffusivities were found to be of order $10^{-12}$ $cm^2 \cdot s^{-1}$ which is considerably smaller than the diffusivities observed in the present work, even with allowances made for the difference in temperature. That the diffusivity in a counterdiffusion process should be significantly smaller than in a simple adsorption or desorption process is not surprising, however, in view of the possibility of interference between counterdiffusing molecules.

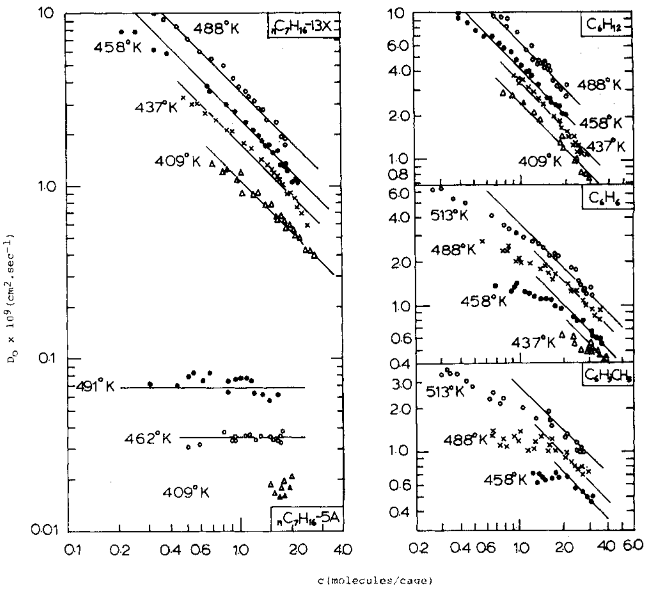

**Fig. 5. Log-log plots showing concentration dependence of corrected diffusivity ($D_o$).**

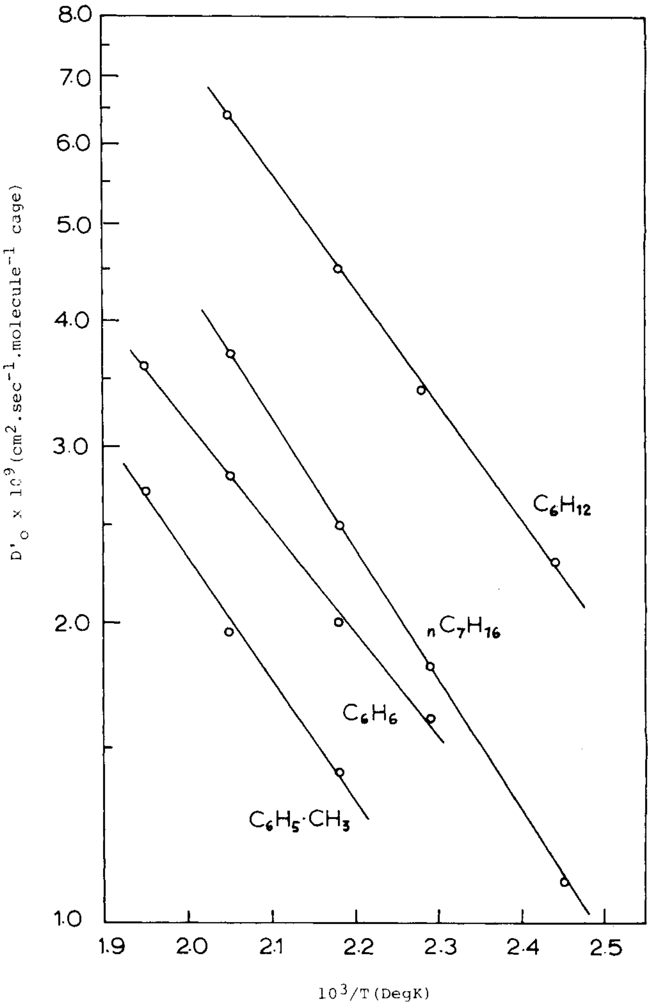

**Fig. 6. Arrhenius plot showing temperature dependence of the parameter $D'_o$ [Equations (3), (4), (5)].**

Diffusion of hydrocarbons in NaX zeolite has also been studied by N.M.R. methods. The self-diffusivities obtained from pulsed field gradient spin-echo measurements would be expected to be comparable with the corrected diffusivities ($D_o$) derived from sorption experiments. The observed values of the N.M.R. diffusivities are, however, very much larger than expected ($10^{-5}$ – $10^{-6}$ $cm^2 \cdot s^{-1}$ for $nC_7H_{16}$ and $C_6H_{12}$ at temperatures of 200° to 300°K) (Karger, Shdanov, and Walter, 1976; Karger, Lorenz, Pfeifer, and Bülow, 1976). A similar discrepancy of several orders of magnitude has been noted between the values obtained for the diffusivity of methane in 5A sieve by N.M.R. and sorption measurements (Kärger and Caro, 1975; Caro, Kärger, Pfeifer, and Schöllner, 1975). The reasons for this disagreement have not yet been established, but it is noteworthy that similarly large discrepancies have been observed between the diffusivities of molecular solids as determined by N.M.R. and by direct tracer measurements (Chadwick and Sherwood, 1975).

## CONCLUSIONS

Previous studies of diffusion in the type A zeolites (Ruthven and Derrah, 1975) led to the tentative conclusion that the diffusional behavior is determined by the relative sizes of the diffusing molecule and the sieve windows. Values of $D_o$ for the diffusion of small monatomic and diatomic gases (argon, kryton, xenon, oxygen, nitrogen) in 4A sieve were found to be essentially independent of sorbate concentration, whereas in the 5A sieve these gases showed the same inverse concentration dependence as we now see for the $C_6$ and $C_7$ hydrocarbons in the 13X sieve. The free diameter of the twelve membered oxygen rings in the 13X sieve is about 7.4Å, which is significantly greater than the critical diameters of the hydrocarbon sorbates. The results of the present study are thus entirely consistent with previous observations and suggest that the conclusions concerning the dependence of the diffusional behavior on the relative sizes of diffusing molecule and sieve aperture are not restricted to the A type zeolites.

It was suggested that when the diffusing molecule is large relative to the sieve aperture, the dominant diffusion mechanism involves single activated jumps between cages. When the molecule is relatively small, a multiple jump mechanism becomes dominant. The data for diffusion of monatomic and diatomic gases in the A type sieves all conformed either to the constant $D_o$ pattern (single jump mechanism) or to the $D_o \propto 1/c$ pattern (multiple jump mechanism). However, if there really are two contributing transport mechanisms, one would expect to find certain systems and conditions for which the contribution of both mechanisms was comparable. At high sorbate concentrations, the jump path would be limited by intermolecular collisions, while in the low concentration limit the diffusivity would be determined simply by the jump frequency. This would give rise to an intermediate form of concentration dependence as is observed for the diffusion of benzene and toluene in 13X.

## NOTATION

$c$ = sorbate concentration (molecule/cage)
$D$ = zeolitic diffusivity
$D_o$ = corrected diffusivity, Equation (3)
$D'_o$ = parameter defined by Equation (4)
$D'_*$ = parameter defined by Equation (5)
$E$ = diffusional activation energy
$K$ = Henry's law constant (molecule·cage$^{-1}$·torr$^{-1}$)
$K_o$ = parameter defined by Equation (2)
$m_t/m_\infty$ = fraction of final equilibrium mass adsorbed or desorbed during time $t$
$p$ = pressure of sorbate
$q_o$ = limiting heat of sorption, Equation (2)
$R$ = gas constant
$r$ = radius of zeolite crystal
$t$ = time
$T$ = temperature, °K

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