the average initial particle size was in good agreement with the experimental data.

The extent of deposition during the experiment was determined and was insignificant in the calculation of the coagulation constant.

The effect of polydispersity on the coagulation constant was found to be in good agreement with the theoretical prediction.

ACKNOWLEDGMENT

This work was performed under a multi-sponsored industrial research program at IIT Research Institute. The contents of this paper are derived from the doctoral thesis of M. B. Ranade.

NOTATION

a = diameter of the particle
a_n = geometric number mean diameter, GNMD
a_m = geometric mass mean diameter, GMMD
A = constant in the Cunningham correction (0.864)
d = differential symbol
$f(a)$ = frequency of size distribution
l = mean free path of gas molecules
l_b = mean free path of aerosol particles
n = number concentration, no. particles/cm^3
n_0 = number concentration at time $t = 0$
$n(a)$ = number concentration of particles of diameter a
N = total number of particles in the aerosol cloud
r = radius of the particle
V_s = settling velocity, cm/s

Greek Letters

β = deposition loss constant, no. particles/s
η = viscosity of the gaseous medium, poise
κ = Boltzmann constant 1.38×10^{-16} ergs/°K
μ = linear dimension (microns), 10^{-4} cm
σ_g = geometric standard deviation
Φ = inverse number concentration ($1/n$), cm^3/no. particles
ψ = polydispersity factor

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Diffusion in Synthetic Faujasite Powder and Pellets

Transient sorption rates of allene and methylacetylene in synthetic faujasite (Linde 13X) powder and self-bonded pellets are studied in a constant-volume, constant-pressure system at three temperatures. The intracrystalline diffusion coefficients are found to be in the order of 10^{-11} cm^2/s and the activation energy for diffusion is 4.0 kcal/g-mole.

A simple bipore distribution model for transient diffusion in macropores and micropores is presented. It agrees well with the experimental data and quantitatively predicts the macropore diffusion coefficients. The flow in the macropores is found to be in the Knudsen flow regime.

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SCOPE

The phenomena of diffusion in porous pellets become quite complicated when zeolites are used as the pellets because they contain both a micropore structure within the crystals (intracrystalline diffusion) and a macropore structure between crystals (intercrystalline diffusion). Quantitative prediction of the individual diffusion coefficients can be of importance in interpreting the experimental data and improving engineering design in such important operations as purification of gas streams, separation processes, and catalytic conversion units.

Previous work on the study of diffusion in zeolites concentrates mostly in intracrystalline diffusion with powder materials. Recent studies on the diffusion in solids with a bipore distribution include the theoretical analysis by Furusawa and Smith (1973), both theoretical and experimental investigation on diffusion in ion exchanged resins by Ruckenstein et al. (1971), and the experimental study by Sargent and Whitford (1971) using commercial 5A molecular sieve pellets. The present work employed a constant-pressure constant-volume technique to examine quantitatively some of the effects of micropore and macropore structure on the determination of the diffusion coefficient. Diffusion coefficients were evaluated based on a mathematical model taking into account intercrystalline and intracrystalline diffusion and adsorption. Equilibrium adsorption isotherms and rates of sorption of allene and methylacetylene were measured on synthetic faujasite (13X) powder. Diffusion measurements were also made with 13X pellets formed from the material used in the powder experiments. No binder was added, thus eliminating the possible complication which it might cause.

CONCLUSION AND SIGNIFICANCE

Intracrystalline diffusion coefficients were determined by assuming that the process obeys Fick's law. Deviation from Fick's law was found at higher uptakes (fractional uptake greater than 0.5). This may be due to the concentration dependence of the diffusion coefficient. Activation energy for diffusion was found to be 4.0 kcal/g-mole. The low activation energy may be due to the relative ease of flow of gas molecules through the large pore openings of 13X zeolite.

A mathematical model was developed to describe the diffusion in a solid with a bipore distribution. Agreement between experimental measurements and theoretical predictions was good. Intercrystalline diffusion was found to be in the Knudsen regime. One distinct feature of the present work is that only one adjustable constant was used in the determination of the intercrystalline diffusion coefficient. The adsorption constant and the intracrystalline diffusion coefficient required in the calculation were determined by independent experiments. The validity of the model is, thus, enhanced.

The necessity of using such a model to treat the experimental data obtained from measurements made on zeolite pellets was demonstrated by the fact that the calculated intracrystalline diffusion coefficient based on a simple Fick's equation would be one order of magnitude less than that obtained from the bipore distribution model. In the absence of the experimental value of the intercrystalline diffusion coefficient, it was shown that estimation of the intercrystalline diffusion coefficient by Knudsen's equation would be satisfactory for the determination of the intracrystalline diffusion coefficient.

Synthetic zeolites are becoming ever more important both as selective sorbents and as catalysts. Molecular sieves of X and Y types have found their widest commercial use in petroleum cracking processes. The structure of zeolite X, which has the basic formula $Na_2O \cdot Al_2O_3 \cdot mSiO_2 \cdot nH_2O$ (where m ranges in value from 2.4 to 3.0) was first determined by Broussard and Shoemaker (1960). Like all other aluminosilicates, zeolite X is composed of linked SiO_4 and AlO_4 tetrahedra. All corners of each tetrahedron are shared with other tetrahedra to give a three-dimensional array. The lattice of 13X may be viewed as a tightly packed arrangement of tetrahedra interlaced with void space (supercages of 12Å diameter) which are accessible to molecules of corresponding dimensions. A small pore system, formed at the interior of joined tetrahedra, also exists within the crystal structure. However, this is normally inaccessible to most molecules.

Most diffusion and sorption studies on zeolites were carried out with powder as summarized by Barrer (1971). However, zeolites in pellet form are frequently used in actual operations. Zeolite pellets normally consist of two distinct types of pore structure: micropores within the crystalline network and macropores between the crystals. The effect of the presence of the macropore on the diffusive property of the zeolite pellet has not been fully investigated. Experimental investigations on diffusion in zeolites generally involve measurement of the weight change of the sample or the pressure change of the system. A single effective diffusion coefficient is commonly used to interpret the results obtained from such dynamic measurements. However, for bidisperse pellets (for example, commercial molecular sieve pellets), this interpretation may lead to erroneous results (Furusawa and Smith, 1973).

Ruckenstein et al. (1971) have developed a model for dynamic adsorption in a bidisperse pellet. Their model takes into account diffusion and adsorption both in the micropores and the macropores and was used to quantitatively interpret some experimental sorption data. Sargent and Whitford (1971) also developed a simple model with diffusion in both micropores and macropores, but they neglected the transient effect in the macropores. They solved their equations numerically although an analytical solution would have been possible. Their model was used to determine the intracrystalline diffusion coefficient of CO_2 in 5A sieves by assuming a value of the intercrystalline diffusion coefficient.

The present study concerns the diffusion of allene and methylacetylene in Linde 13X sieve in the temperature range between 21° and 60°C. Diffusion measurements were made on both powder and self-bonded pellets pelletized from the powder used in the powder experiments. The prime objective is to investigate the mechanisms and the relative importance of intercrystalline and intracrystalline diffusion in a pellet with a bipore distribution. A mathematical model, which takes into account the diffusion in both macropores and micropores, was developed and used to obtain the diffusion coefficient in the macropores. In its determination, the diffusion coefficient was the only adjustable constant. This certainly enhances the validity of the model.

APPARATUS AND PROCEDURES

A constant-volume, constant-pressure system (the eight-liter total volume of the system was sufficiently large so that the system pressure remained essentially constant during sorption) was employed to study the rate of sorption and equilibrium capacity of Linde 13X molecular sieve. A Cahn RG automatic recording electrobalance with a sensitivity of 10^{-7} g was used to measure the weight change of the sorbent as a function of time. A schematic diagram of the system is shown in Figure 1. The whole unit was housed in an insulated cabinet whose temperature was controlled to within ±0.5°C. The sample in the balance chamber was activated under a vacuum of 5μm *Hg* at a temperature of 350°C for 12 hours. It was observed that the weight of the sample remained essentially constant after four hours of heating.

A solenoid valve was installed to isolate the balance portion of the system from the reservoir. After the completion of activa-

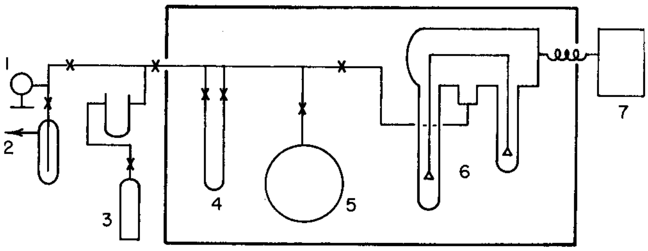

Fig. 1. Schematic diagram of the sorption apparatus. 1. McLeod gauge; 2. vacuum pump; 3. sorbate gas cylinder; 4. manometer assembly; 5. gas reservoir; 6. electrobalance assembly; 7. recorder.

tion, the sample was allowed to cool to the desired temperature. The balance portion of the system was then isolated from the rest of the system under vacuum by closing the solenoid valve. The reservoir was filled with gas to the desired pressure. The system pressure was measured by a mercury manometer which was read by a cathetometer. At the start of a run ($t=0$), the solenoid valve was opened and the sorbate gas was instantaneously admitted to the balance chamber. The change in sample weight as a function of time was monitored continuously with a Sargent recorder until equilibrium was achieved. A more detailed description of the system is given by Damle (1973).

Samples of 13X powder was obtained from Linde Division, Union Carbide Corporation (Lot No. 13945000049). The powder was used to make the pellets in this laboratory by a pelletizer under a pressure of approximately 7,000 lb./sq.in.abs. The sample was discarded after it had been used a few times. It was found that the reproducibility of the data with different samples was good. No evidence was observed for a change in sorption capacity throughout a series of runs. All data reported are based on the weight of 13X in the anhydrous form.

Both allene and methylacetylene are CP grade. One main reason for choosing these two gases is that they are being used in a mixture study to be reported in the future. The present results will be used to compare with those obtained from the mixture study in order to ascertain the mechanisms of diffusion in both single and binary sorbate systems.

RESULTS AND DISCUSSION

Diffusion in Uncompacted Powder

Sorption rates for Linde 13X powder were measured at three temperatures, 21°, 45°, and 60°C with a sorbate pressure of 1.2 mm Hg. A typical plot of the fractional uptake M_t/M_∞ for allene versus $\sqrt{t}$ is shown in Figure 2.

For the case of zeolite powder, the crystals may be assumed to be homogeneous and spherical and the intracrystalline diffusion may be assumed to follow the Fick's equation. The mathematical solution to the equation for transient diffusion into a sphere from a constant-volume, constant pressure system is given by Crank (1957)

$$\frac{M_t}{M_\infty} = 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp\left[- \left(\frac{n^2 \pi^2 D_i t}{R^2} \right) \right] \quad (1)$$

For small time, the solution becomes

$$\frac{M_t}{M_\infty} = \frac{6}{\pi^2} \left(\frac{D_i t}{R^2} \right)^{1/2} \quad (2)$$

The reduced diffusion coefficient D_i/R^2 may be obtained by plotting the theoretical dimensionless time $D_i t/R^2$ versus the experimental time t for equal values of the fractional uptake M_t/M_∞. According to Equation (2), such a plot should give a straight line through the origin with a slope equal to D_i/R^2. Experimental data plotted in this manner give reasonably straight lines. As expected, deviations from the straight line are found for large values of time possibly due to the concentration dependence of the diffusion coefficient. The diffusion coefficients calculated from the slope of the linear portion of these plots ($M_t/M_\infty < 0.5$) are shown in Table 1. Also in Table 1 are the equilibrium adsorption constants K which are obtained by taking the initial slopes of the equilibrium isotherms.

The temperature dependence of the diffusion coefficient in zeolites can generally be expressed by an Arrhenius-type equation

$$D_i = D_{i0}\, e^{-E/RT} \quad (3)$$

By assuming a constant value of E over a range of temperature, a semi-log plot of D_i versus $1/T$ for that range should yield a straight line and the activation energy E

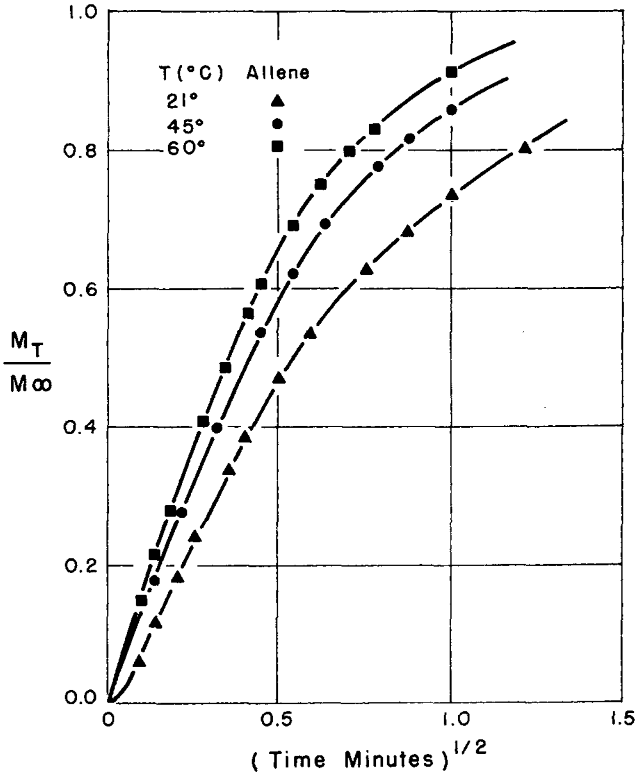

Fig. 2. Rate of sorption of allene on 13X powder.

Table 1. Results

	Allene			Methylacetylene	
Temp., °C	21	45	60	23	60
Pressure, mm Hg	1.2	1.2	1.2	1.0	1.0
K	5.45×10^4	3.28×10^4	2.48×10^4	7.5×10^4	3.3×10^4
P_0[a], cm	10^{-4}	10^{-4}	10^{-4}	10^{-4}	10^{-4}
D_c, cm²/s	1.5×10^{-11}	3.0×10^{-11}	4.1×10^{-11}	1.1×10^{-11}	2.9×10^{-11}
R[b], cm	0.145	0.145	0.145	0.145	0.145
D_p, cm²/s	0.0525	0.0550	0.0575	0.0576	0.0580
Mean square error	6.2×10^{-3}	1.6×10^{-3}	6.4×10^{-3}	1.1×10^{-2}	5.2×10^{-3}
D_K[c], cm²/s	0.289	0.300	0.308	0.290	0.308
ϵ_p	0.42	0.42	0.42	0.42	0.42
τ	5.50	5.46	5.36	5.04	5.31

[a] Estimated from photomicrograph measurements (Gulfaz, 1973).
[b] Calculated from the radius of a sphere having same volume as the pellet.
[c] Estimated from pore distribution measurements of commercial 13X pellets.

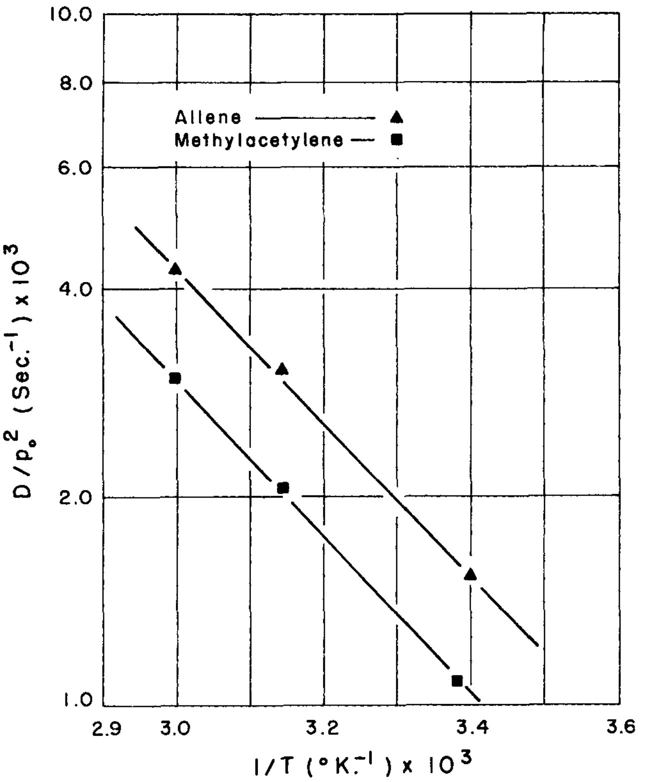

Fig. 3. Reduced diffusion coefficient as a function of temperature for 13X powder.

may be calculated from its slope. A plot of D_i/R^2 versus $1/T$ is shown in Figure 3. The calculated activation energy of diffusion for both allene and methylacetylene is 4 kcal/g-mole. The activation energies seem to be relatively low. However, activation energy in such a range has been previously reported (Satterfield and Frabetti, 1967). The low activation energy may be due to the ease of flow of sorbate molecules in the relatively large openings of the 13X zeolites.

Diffusion in Pellets

Mathematical model. In the derivation of the mathematical model for zeolite particles, it is assumed that

1. A spherical macroporous sorbent particle is an assembly of small microporous crystals which may be treated as homogeneous solids.
2. The crystals are spheres with a uniform size.
3. Diffusion in both the micropores and the macropores can be described by Ficke's equation, and the diffusion coefficients are constant.
4. The adsorption isotherm is linear and adsorption equilibrium is attained at the boundary surface of the crystals.

A similar approach has been employed by Sargent and Whitford (1971). However, they neglected the transient effects in the macropores. They used a numerical technique to solve their equations although an analytical solution of the problem was possible. Mass balances in both macropores and micropores give

$$\frac{D_p \epsilon_p}{r^2}\frac{\partial}{\partial r}\left(r^2 \frac{\partial C_p}{\partial r}\right) - \frac{3(1-\epsilon_p)}{\rho_0} D_i \left(\frac{\partial C_i}{\partial \rho}\right)_{\rho=\rho_0} = \frac{\partial C_p}{\partial t}\epsilon_p \tag{4}$$

$$D_i\left(\frac{\partial^2 C_i}{\partial \rho^2} + \frac{2}{\rho}\frac{\partial C_i}{\partial \rho}\right) = \frac{\partial C_i}{\partial t} \tag{5}$$

Equations (4) and (5) should be solved with the following initial and boundary conditions:

$$C_p(0, r) = C_{p0} \tag{6a}$$

$$C_i(0, \rho) = C_{i0} \tag{6b}$$

$$C_p(t, R) = C_{p\infty} \tag{6c}$$

$$C_i(t, \rho_0) = KC_p(t, r) \tag{6d}$$

$$\frac{\partial C_p(t, 0)}{\partial r} = \frac{\partial C_i(t, 0)}{\partial \rho} = 0 \tag{6e}$$

If one defines the following normalized variables

$$\theta_p = \frac{C_p - C_{p0}}{C_{p\infty} - C_{p0}} \qquad \theta_i = \frac{C_i - C_{i0}}{C_{p\infty} - C_{i0}}$$

$$\bar{t} = \frac{D_p t}{R^2} \qquad \delta = \frac{3(1-\epsilon_p)}{\epsilon_p} \tag{7a}$$

$$\eta = \frac{r}{R} \qquad \gamma = \frac{\rho}{\rho_0}$$

and the parameters

$$\alpha = \frac{D_i/\rho_0^2}{D_p/R^2} = \frac{D_i R^2}{D_p \rho_0^2}$$

$$\beta = \frac{3(1-\epsilon_p)}{\epsilon_p}\frac{D_i R^2}{D_p \rho_0^2} = \delta\alpha \tag{7b}$$

Equations (4) and (5) become

$$\left(\frac{\partial^2 \theta_p}{\partial \eta^2} + \frac{2}{\eta}\frac{\partial \theta_p}{\partial \eta}\right) - \beta\left(\frac{\partial \theta_i}{\partial \gamma}\right)_{\gamma=1} = \frac{\partial \theta_p}{\partial \bar{t}} \tag{8}$$

$$\left(\frac{\partial^2 \theta_i}{\partial \gamma^2} + \frac{2}{\gamma}\frac{\partial \theta_i}{\partial \gamma}\right) = \frac{1}{\alpha}\frac{\partial \theta_i}{\partial \bar{t}} \tag{9}$$

and Equations (6) become

$$\theta_p(0, \eta) = \theta_i(0, \gamma) = 0 \tag{10a}$$

$$\theta_p(\bar{t}, 1) = 1 \tag{10b}$$

$$\theta_i(\bar{t}, 1) = K\theta_p(\bar{t}, \eta) \tag{10c}$$

$$\frac{\partial \theta_p}{\partial \eta}(\bar{t}, 0) = \frac{\partial \theta_i}{\partial \gamma}(\bar{t}, 0) = 0 \tag{10d}$$

Equations (8) and (9) were solved in conjunction with Equations (10) by the Laplace transform technique to yield

$$\theta_p(\eta, \bar{t}) = 1 + \frac{4\pi}{\alpha\eta}\sum_{m=1}^{\infty}\sum_{n=1}^{\infty}\frac{m(\sin m\pi\eta)\exp(-\alpha q^2_{mn}\bar{t})}{(-1)^m q^2_{mn}\left[2 + \frac{K\beta}{\alpha}\left(csc^2 q_{mn} - \frac{\cot q_{mn}}{q_{mn}}\right)\right]} \tag{11}$$

where q_{mn} are the roots of the transcendental equation

$$\alpha q^2_{mn} + K\beta(1 - q_{mn}\cot q_{mn}) = m^2\pi^2 \qquad m = 1, 2 \ldots\ldots, \infty \tag{12}$$

It is more convenient to express the solution in terms of the ratio M_t/M_∞ where M_t and M_∞ are the net change in solute in the solid at time t and time infinity, respectively. This is done by using the following relationships:

at time t,

$$M_t = \int_0^t D_p \epsilon_p 4\pi R^2 \left(\frac{\partial C_p}{\partial r}\right)_{r=R} dt \qquad (13a)$$

and at equilibrium

$$M_\infty = \int_0^\infty D_p \epsilon_p 4\pi R^2 \left(\frac{\partial C_p}{\partial r}\right)_{r=R} dt \qquad (13b)$$

Therefore, the fractional uptake is given by

$$\frac{M_{\bar{t}}}{M_\infty} = \frac{\int_0^{\bar{t}} \left(\frac{\partial \theta_p}{\partial \eta}\right)_{\eta=1} d\bar{t}}{\int_0^{\infty} \left(\frac{\partial \theta_p}{\partial \eta}\right)_{\eta=1} d\bar{t}} = \frac{\sum_{m=1}^{\infty}\sum_{n=1}^{\infty} \frac{m^2\,[1 - \exp(-\alpha q^2_{mn}\bar{t})]}{q^4_{mn}\left[2 + \frac{K\beta}{\alpha}\,(csc^2\, q_{mn} - \cot q_{mn}/q_{mn})\right]}}{\sum_{m=1}^{\infty}\sum_{n=1}^{\infty} \frac{m^2}{q^4_{mn}\left[2 + \frac{K\beta}{\alpha}\,(csc^2\, q_{mn} - \cot q_{mn}/q_{mn})\right]}} \qquad (14)$$

It should be pointed out that Equation (14) reduces to the solution of a simple unsteady state diffusion problem for either large α or small K. The ratio α represents the relative importance of the diffusional resistances in micropores and macropores. Large α means lower resistance in the micropores and the process is controlled by the macropore diffusion. For large values of α the solution reduces to that of a simple unsteady state diffusion problem. For small values of K, this corresponds to the case where the solute concentration at the crystal surface is small and thus can easily be transported into the crystal. In the limit, as $K \to 0$, only one type of pore exists. Thus, the solution again reduces to that of a simple diffusion equation.

Some of the results calculated from Equation (12) are shown in Figures 4 and 5. Figure 4 shows M_t/M_∞ as a function of $\sqrt{\bar{t}}$ at a constant value of α with the adsorption constant K as a parameter. For small values of K, the uptake by crystals is negligible and the process may be treated as diffusion in macropores discussed in the previous paragraph and also by Ruckenstein et al. (1971). Figure 5 shows M_t/M_∞ as a function of $\sqrt{\bar{t}}$ at a constant value of K with α as a parameter. Again, as previously discussed, for large values of α, the shape of the curve approaches that of a simple unsteady state diffusion.

Determination of Macropore Diffusion Coefficient

The mathematical model was used to calculate the diffusion coefficients in the macropores. One should notice that Equation (11) has three parameters K, D_i, and D_p. However, only one adjustable constant D_p was used in fitting the experimental data to the theoretical curve. A typical comparison between the experimental data and the theoretical curves is shown in Figure 6. It can be seen that the agreement between the experiments and the model is very good. Deviations were found at higher values of uptakes ($M_t/M_\infty > 0.70$). This is to be expected as at higher uptakes, it is possible that the controlling step may be altered and the assumption of a linear isotherm also becomes doubtful. Moreover, the value of D_i determined from the powder study cannot be applied at higher uptakes. The calculated diffusion coefficient D_p is tabulated in Table 1 for three temperatures.

The following expression is generally used to predict the effective diffusion coefficient in a porous material

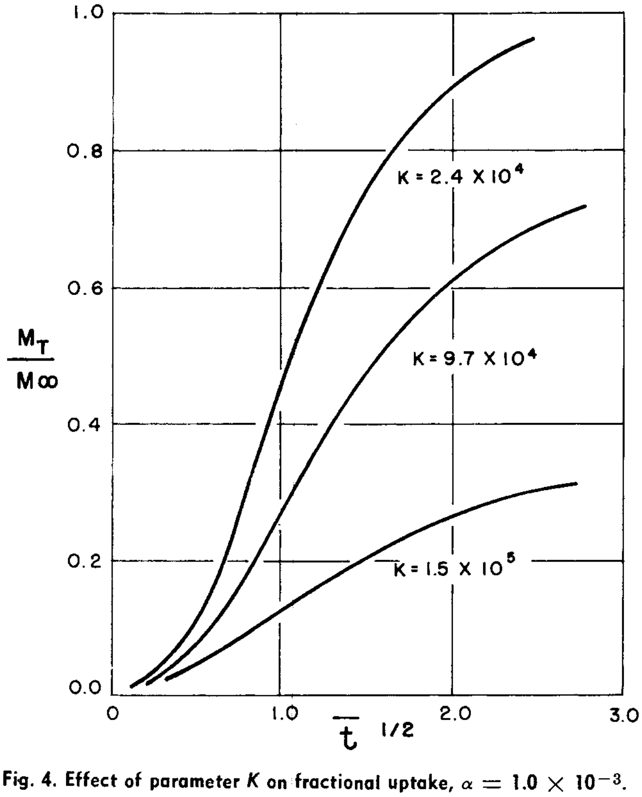

Fig. 4. Effect of parameter K on fractional uptake, $\alpha = 1.0 \times 10^{-3}$.

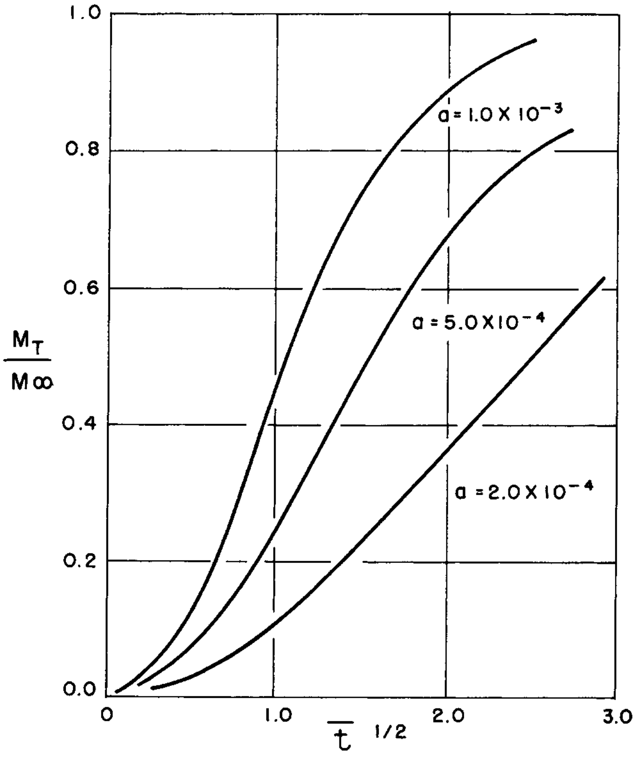

Fig. 5. Effect of parameter α on fractional uptake, $K = 1.0 \times 10^2$.

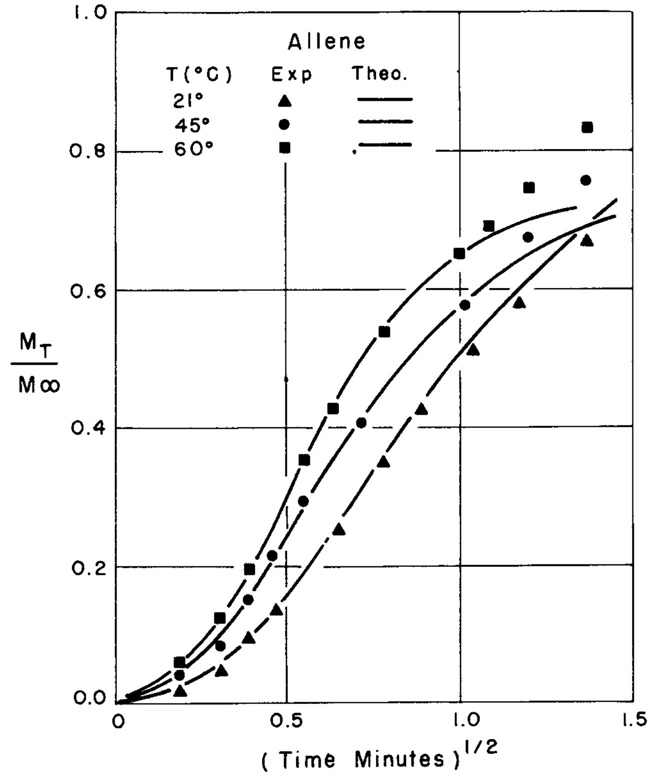

Fig. 6. Comparison of experimental and theoretical sorption curves for Linde 13X selfbonded pellet.

$$D_p = \frac{D_K}{\tau} \tag{15}$$

The diffusion coefficient D_K for Knudsen flow can be estimated by the Knudsen Equation. The values of D_K calculated from the Knudsen Equation for both allene and methylacetylene at various temperatures are also shown in Table 1. As D_p is determined from experimental data, Equation (15) can then be used to calculate the tortuosity factor τ which is also included in the table. The tortuosity factor is found to be approximately 5 for all cases studied here. This agreement in the value of the tortuosity factor is an additional verification of the validity of the model.

The fact that only one adjustable constant was used to fit the data with theoretical curves deserves some additional comments. Normally, both D_i and D_p are varied to obtain the best fit of the data with the theoretical curve. Thus, two adjustable parameters are involved. However, it is well recognized that one should minimize the number of adjustable constants when it is necessary to fit the data with mathematical models. The present model based on only a single adjustable constant is well confirmed by the good agreement with experiments.

The necessity of using such a bipore distribution model may be demonstrated as follows. One may simply want to neglect the effects of the presence of the bipore distribution and use the solution of the simple unsteady state diffusion equation to calculate the intracrystalline diffusion coefficient. Using the present data for pellets, the reduced diffusion coefficient calculated in this manner is 10^{-4} sec^{-1} which is about one order of magnitude smaller than the true value. For measurements of sorption rate made with pellets, the present model should be used to calculate the intracrystalline diffusion coefficient. The Knudsen equation could be used to estimate the macropore diffusion coefficient.

ACKNOWLEDGMENT

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NOTATION

C = the concentration of sorbate, moles cm^{-3}
D = diffusion coefficient, cm^2 s^{-1}
D_K = Knudsen diffusion coefficient, cm^2 s^{-1}
E = activation energy, kcal mole^{-1}
K = equilibrium constant
M = the net change in solute in the solid, moles g^{-1}
r = radial distance in macropores, cm
R = radius of the particle, cm
ρ = radial distance in micropores, cm
ρ_0 = radius of the crystal, cm
t = time, s
$\bar{t}$ = dimensionless time, defined in Equation (7a)
T = temperature, °K

Subscripts

0 = initial value
∞ = equilibrium (infinite time)
i = micropore
p = macropore
t = value at time t

Greek Letters

α = dimensionless rate parameter, defined in Equation (7b)
β = defined in Equation (7b)
γ = dimensionless microsphere radial position defined in Equation (7a)
δ = defined in Equation (7a)
ϵ_p = macropore void fraction
η = dimensionless macrosphere radial position defined in Equation (7a)
θ_i = dimensionless micropore concentration defined in Equation (7a)
θ_p = dimensionless macropore concentration defined in Equation (7a)
τ = tortuosity factor

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