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Diffusion-Limited Reaction in Supported Liquid-Phase Catalysis

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A method for determining the overall effective diffusivity in supported liquid-phase catalysts is proposed. Use of the method requires isobaric diffusion measurements in nonreacting systems with variable liquid loadings in the porous support. This information along with a model can be used to predict the liquid loading required to give a maximum reaction rate in diffusion-limited reacting systems. The study presented here was limited to the molecular mass transport regime.

NOMENCLATURE

A	Component A.
B	Component B.
C	Molar concentration in the gas phase (g-moles/cm^3).
C_1	Constant dependent only upon structure of porous support and giving relative Knudsen flow permeability (cm).
C_2	Constant dependent only upon structure of the porous support and giving ratio of molecular diffusivity within the porous support to the free gas diffusivity at $q = 0$, dimensionless.
$C_2{}^G$	Parameter dependent upon the liquid-loading parameter q and giving ratio of molecular diffusivity within the liquid-loaded support to the free gas diffusivity, $D_{eff}{}^G/D_{AB}$, dimensionless.
C_{A_0}	Molar concentration of component A in the gas phase at $Z = 0$ (g-moles/cm^3).
D'	Liquid-gas diffusivity-ratio parameter defined by Eq. (12), dimensionless.
D_A^L	Molecular diffusivity of species A in the liquid phase (cm^2/sec).
D_{AB}	Free gas mutual diffusivity in a binary mixture of components A and B (cm^2/sec).
$D_{AB}{}^0$	Pressure independent mutual diffusivity defined as $D_{AB}{}^0 = D_{AB}P$ (dyn/sec).
D_{eff}	Overall effective diffusivity in a liquid-loaded support (cm^2/sec).
$D_{eff}{}^G$	Gas-phase contribution to the overall effective diffusivity in a liquid-loaded support (cm^2/sec).
$D_{eff}{}^L$	Liquid-phase contribution to the overall effective diffusivity in a liquid-loaded support (cm^2/sec).
E	Effectiveness factor defined by Eq. (8), dimensionless.
G	Grouping of terms defined by Eq. (23), dimensionless.
H	Henry's constant, representing the ratio of the molar concentration of component A in the liquid phase to its molar concentration in the gas phase (cm^3 gas/cm^3 liquid).
k''	First-order reaction rate constant in supported liquid-phase catalysts (sec^{-1}).
K_A	Effective Knudsen diffusivity defined as $K_A = C_1(R_gT/M_A)^{1/2}$ (cm^2/sec).
K_m	Mixture effective diffusivity defined as $K_m = y_AK_B + y_BK_A$ (cm^2/sec).
L	Either the length or the half thick-

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	ness of an infinite slab of the pelleted support (cm).
M	Molecular weight (g/g-mole).
N	Molar flux (g-mole/cm^2 sec).
P	Total pressure (dyn/cm^2).
q	Liquid-loading parameter, representing the ratio between the actual amount of liquid dispersed to the maximum amount needed to fill the porous support at atmospheric pressure, dimensionless.
$\bar{q}$	The value of q at which r'_A reaches a maximum while γ'_M and D' are held constant, dimensionless.
r	Reaction rate per unit surface area, defined by Eq. (14) (g-mole/sec cm^2).
r'	Reaction rate defined by Eq. (16), dimensionless.
R_g	Gas constant, 8.31×10^7 (ergs/g-mole-°K).
T	Absolute temperature (°K).
y	Mole fraction.
y_{A_L}	Mole fraction of component A at $Z = L$ in the pelleted support.
y_{A_0}	Mole fraction of component A at $Z = 0$ in the pelleted support.
Z	Distance in direction of mass transfer (cm).
ϵ	Porosity of the solid support, representing the volume of liquid needed to fill the porous support per unit volume of the porous support, dimensionless.
ϕ	The Thiele modulus in supported liquid-phase catalysts, defined by Eq. (9), dimensionless.
$\bar{\phi}$	The Thiele modulus ϕ evaluated at $q = \bar{q}$ as given by Eq. (18), dimensionless.
γ'_M	The molecular Thiele modulus in supported liquid-phase catalysts, defined by Eq. (11), dimensionless.

Subscripts

A	Component A.
B	Component B.
L	Value at $Z = L$.
m	Mixture.
M	Molecular.
0	Value at $Z = 0$.

Superscripts

G	Gas phase.
L	Liquid phase.

INTRODUCTION

A theoretical treatment, followed by experimental verification, of diffusion-limited reactions in supported liquid-phase catalysts has been given by Rony (*1*, *2*). His pioneering work prompted us to initiate the work presented here.

Use of supported liquid-phase catalysis is relatively new. One of its advantages is the creation of high gas–liquid interfacial areas and short liquid-phase diffusion paths.

Although the theoretical model proposed by Rony (*1*) has not been fully verified experimentally, it is in agreement with his experimental data on a hydroformylation reaction (*2*). A disadvantage of Rony's model, however, is the fact that it contains two adjustable constants which can only be determined from experiments with reaction occurring in the supported liquid-phase catalyst. Since these adjustable constants have no known physical meaning, it is probable that they will vary from one reaction system to another. This currently prevents Rony's model from being predictive, although the basic concept is novel.

The approach presented here is somewhat different from Rony's and, from our point of view, appears to have several advantages. The model proposed relies on experimental data obtained from several isobaric diffusion experiments in nonreacting systems. This information, in conjunction with our model, can then predict diffusion limitations on reacting systems in supported liquid-phase catalysts. The only constant in the proposed model does have physical meaning. It characterizes the porous support and is independent of reacting species.

Another major advantage of our model is its ability to adequately treat systems in which the liquid has been dispersed in a nonideal porous support. This is in contrast with Rony's approach, which theoretically treats the porous support as an ideal bundle of pores and then forces the model to include nonideal porous supports. A theoretical extension of his model to nonideal supports might require a third adjustable constant.

Theory

In supported liquid-phase catalysis, the liquid is used as a carrier of the catalyst and often is dispersed in an inert porous solid. A typical gaseous reactant, A, dissolves in the liquid and then reacts homogeneously in the presence of the dissolved catalyst to produce a gaseous product, B.

The dispersion of the liquid reduces the diffusion resistances in that phase significantly. However, by only partially filling the porous matrix with liquid, the diffusional transport of gaseous reactant, A, still occurs mainly in the gas phase.

The system under consideration here is an infinite slab of porous support which is partially or fully loaded with liquid catalyst. A first-order, homogeneous reaction, A → B, is assumed to occur in the liquid bulk. In this system, the gas phase can be pictured as interconnected channels of free space. On the other hand, the liquid phase can be considered as a large number of liquid domains randomly distributed in the solid matrix. A theory describing the behavior of the above system is based on the following assumptions:

1. Regardless of whether or not the porous support is highly nonideal, it is assumed that the diffusion fluxes in both the gas and liquid phases are one-dimensional.
2. At any cross-section in the porous structure, the liquid phase is in physical equilibrium with the gas phase.
3. The solid matrix is inert relative to reaction and adsorption of species A and B. Consequently, surface diffusion and heterogeneous reaction are not included in the model.

Since the transport in the gas phase can occur by either molecular diffusion or Knudsen diffusion or both, the treatment here is limited to transport by molecular diffusion only. The flux of reactant A by molecular diffusion in the gas and liquid phases can be described by Eq. (1):

$$\mathbf{N}_{\mathrm{A}} = -D_{\mathrm{eff}} \frac{dC_{\mathrm{A}}}{dZ}, \tag{1}$$

where D_{eff} is the overall effective diffusivity in the system described above. To theoretically predict D_{eff} would be extremely difficult, simply because of the geometric complexities of the system under consideration. Therefore, a semi-empirical approach for relating the effective diffusivity to liquid loading is adopted in this study.

From our steady-state diffusion experiments in liquid-loaded pellets with diffusion taking place only in the gas phase, the relationship between the ratio of the gas-phase effective diffusivity to molecular diffusivity and liquid loading can best be described by Eq. (2):

$$\frac{D_{\mathrm{eff}}{}^{\mathrm{G}}}{D_{\mathrm{AB}}} = C_2(1 - q)^2, \tag{2}$$

where $D_{\mathrm{eff}}{}^{\mathrm{G}}$ is the gas-phase effective diffusivity in a liquid-loaded pellet; D_{AB} is the binary molecular diffusivity of A or B in free space; C_2 is one of the dusty-gas constants (*3*) which depends only on the structure of the solid support; and finally, q is the liquid-loading parameter representing the ratio between the actual amount of liquid dispersed to the maximum amount needed to fill the porous pellet at atmospheric pressure. The data justifying Eq. (2) are reported in this paper.

At this point, an additional assumption in the model relates liquid loading to the ratio of the liquid-phase effective diffusivity to the molecular diffusivity in a manner similar to that used for the gas phase. This is given by Eq. (3):

$$\frac{D_{\mathrm{eff}}{}^{\mathrm{L}}}{D_{\mathrm{A}}{}^{\mathrm{L}} H} = C_2 q^2, \tag{3}$$

where $D_{\mathrm{eff}}{}^{\mathrm{L}}$ is the contribution, to the overall effective diffusivity, resulting from diffusion fluxes in the liquid phase. It is of interest to note that Eq. (3) is in full agreement with the findings of Satterfield, Ma and Sherwood (*4*) concerning effective diffusivities in liquid-filled porous pellets where $q = 1$. The inclusion of Henry's constant H in Eq. (3) is required to obtain the fluxes in the liquid phase based on gas-phase concentration gradients.

It is further assumed that the overall effective diffusivity equals the sum of the contributions from the gas and liquid phases. This is described by Eq. (4):

$$D_{\text{eff}} = D_{\text{eff}}{}^{\text{G}} + D_{\text{eff}}{}^{\text{L}} = C_2 D_{\text{AB}}(1-q)^2 + C_2 D_{\text{A}}{}^{\text{L}} H q^2. \quad (4)$$

Having established the overall effective diffusivity, the component balance equation for an infinite slab of supported liquid-phase catalyst is given by Eq. (5):

$$D_{\text{eff}} \frac{d^2 C_{\text{A}}}{dZ^2} - k'' H \epsilon q C_{\text{A}} = 0, \quad (5)$$

which is subject to the following boundary conditions:

$$\text{at } Z = 0, \qquad C_{\text{A}} = C_{\text{A}_0}, \quad (6)$$

$$\text{at } Z = L, \qquad \frac{dC_{\text{A}}}{dZ} = 0. \quad (7)$$

The inclusion of Henry's constant in Eq. (5) is required since reaction occurs in the liquid phase whereas the concentration of the reactant is defined for the gas phase.

The solution to Eq. (5) with the boundary conditions gives the effectiveness factor, as shown in Eq. (8):

$$E = \frac{\tanh \phi}{\phi}, \quad (8)$$

where ϕ is a Thiele modulus defined by Eq. (9) as,

$$\phi = L\left(\frac{k'' H \epsilon q}{D_{\text{eff}}}\right)^{1/2}. \quad (9)$$

Substituting Eq. (4) into Eq. (9) gives,

$$\phi = L\left(\frac{k'' H \epsilon}{C_2 D_{\text{AB}}}\right)^{1/2} \times \left(\frac{q}{(1-q)^2 + (D_{\text{A}}{}^{\text{L}} H / D_{\text{AB}}) q^2}\right)^{1/2}. \quad (10)$$

We can define the molecular Thiele modulus as,

$$\gamma'_M = L\left(\frac{k'' H \epsilon}{C_2 D_{\text{AB}}}\right)^{1/2}, \quad (11)$$

and also define a liquid-gas diffusivity-ratio parameter as,

$$D' = \frac{D_{\text{A}}{}^{\text{L}} H \cdot}{D_{\text{AB}}} \quad (12)$$

Substitution of Eq. (11) and (12) into Eq. (10) gives,

$$\phi = \gamma'_M \left(\frac{q}{1 - 2q + q^2(1 + D')}\right)^{1/2}. \quad (13)$$

The actual reaction rate per unit surface area is given by Eq. (14):

$$r_{\text{A}} = 2 E L \epsilon H k'' C_{\text{A}_0} q. \quad (14)$$

By substituting Eq. (8) into Eq. (14) we obtain,

$$r_{\text{A}} = 2 L \epsilon H k'' C_{\text{A}_0} q \frac{\tanh \phi}{\phi}. \quad (15)$$

Finally, a dimensionless reaction rate can be defined as,

$$r'_{\text{A}} = \frac{r_{\text{A}}}{2 L \epsilon H k'' C_{\text{A}_0}} = \frac{q \tanh \phi}{\phi}. \quad (16)$$

One of the main purposes in formulating this model is to determine at what value of q the dimensionless rate, r'_{A}, reaches a maximum. This was also done by Rony (*2*).

Equating the derivative of r'_{A} with respect to q to zero establishes a value of $\bar{q}$ at which a maximum in r'_{A} occurs. The result of equating dr'_{A}/dq to zero is given by Eq. (17):

$$\frac{1}{4} + \bar{\phi} \frac{(1 - \tanh^2 \bar{\phi})}{4 \tanh \bar{\phi}} = \frac{\bar{q} - \bar{q}^2(1 + D')}{1 - \bar{q}^2(1 + D')}, \quad (17)$$

where $\bar{\phi}$ is given by Eq. (18):

$$\bar{\phi} = \gamma'_M \left[\frac{\bar{q}}{1 - 2\bar{q} + \bar{q}^2(1 + D')}\right]^{21/}. \quad (18)$$

It is interesting to note that the solution of Eq. (17) yields $\bar{q}$ as a function of only two parameters, namely the molecular Thiele Modulus, γ'_M, and the liquid-gas diffusivity-ratio parameter, D'. For the case in which $D' \ll 1$, and the reaction is very fast, the solution of Eq. (17) yields $\bar{q}$ as a function of only the molecular Thiele modulus. For a slow reaction, however, a very small value of D' is important in determining $\bar{q}$.

EXPERIMENTAL PROCEDURE

The measurements of effective diffusivities in liquid-loaded pellets was accomplished by using a highly modified version of the Wicke–Kallenbach diffusion cell (*5*). A detailed description of the apparatus is given by Abed (*6*). Steady-state, isobaric diffusion experiments were conducted in the cell using helium and nitrogen. The porous support consisted of ¼ × ¼, α-alumina, T-708 commercial pellets manufactured by the Girdler Catalysts Division of Chemetron Corp. The Girdler T-708 pellets are fairly macroporous; and thus, operating at pressures above 300 psig insured that the transport was dominated by molecular diffusion.

One of the problems encountered in this work was the choice of a liquid to be dispersed in the porous pellet. First of all, it is important to choose a liquid that does not dissolve the nonpolar diffusing gases, helium and nitrogen. This allows the dusty-gas model to describe isobaric diffusion in liquid-loaded pellets. Secondly, the liquid chosen must be chemically and physically inert to the diffusing gases, so that phenomena such as surface interactions between the diffusing gases and the liquid do not occur, thus eliminating surface diffusion. Thirdly, it is important to choose a liquid with low vapor pressure. This minimizes its evaporative losses when exposed to diffusing gases. A liquid which satisfactorily meets the above requirements is ethylene glycol, which is moderately polar. It was therefore chosen for dispersion in the Girdler T-708 pellets.

To achieve a uniform dispersion of the liquid in the solid support, the ethylene glycol was first dissolved in acetone, followed by immersion of the pellets in this solution for a period of at least 2 hr. The pellets were then placed in a vacuum chamber for 1 hr to remove the acetone, leaving the ethylene glycol reasonably well dispersed.

With this choice of an experimental system, the flux measurements were directly usable to determine the parameter, $C_2{}^G$, from the dusty-gas model. This treatment is discussed below.

Evans, Watson and Mason (*3*) have derived the flux equation for isobaric diffusion in dry porous structures, based on the dusty-gas model. Since the liquid phase is inert in the system under consideration, the same equation can be applied to describe diffusion in liquid-loaded pellets. The dusty-gas model is given by Eq. (19):

$$\mathrm{N_A} = -\frac{C_2 D_{\mathrm{AB}}{}^{\circ} K_{\mathrm{A}} P}{(C_2 D_{\mathrm{AB}}{}^{\circ} + K_m P) R_g T}\frac{dy_{\mathrm{A}}}{dZ}. \tag{19}$$

The boundary conditions for Eq. (19) are,

$$\text{at } Z = 0, \qquad y_{\mathrm{A}} = y_{\mathrm{A}_0}, \tag{20}$$

$$\text{at } Z = L, \qquad y_{\mathrm{A}} = y_{\mathrm{A}_L}. \tag{21}$$

Substitution of Eqs. (20) and (21) into the integrated form of Eq. (19) gives,

$$\mathrm{N_A} = \frac{PC_2 D_{\mathrm{AB}}}{R_g TLG}\ln\left[\frac{1 - Gy_{\mathrm{A}_L} + C_2 D_{\mathrm{AB}}/K_{\mathrm{A}}}{1 - Gy_{\mathrm{A}_0} + C_2 D_{\mathrm{AB}}/K_{\mathrm{A}}}\right], \tag{22}$$

where G is given by,

$$G = 1 + \mathrm{N_B}/\mathrm{N_A} = 1 - (M_{\mathrm{A}}/M_{\mathrm{B}})^{1/2}. \tag{23}$$

If the transport is by molecular diffusion only, $C_2 D_{\mathrm{AB}}/K_{\mathrm{A}}$ becomes very small; and Eq. (22) reduces to,

$$\mathrm{N_A} = \frac{PC_2 D_{\mathrm{AB}}}{R_g TLG}\ln\left[\frac{1 - Gy_{\mathrm{A}_L}}{1 - Gy_{\mathrm{A}_0}}\right]. \tag{24}$$

Equation (24) can be used only if molecular diffusion is dominating. It is noteworthy that Eq. (24) can be used to determine the value of C_2 from a single, isobaric diffusion experiment. It is also important to note that C_2 will remain constant as long as the dry, porous structure remains unchanged. If the system is changed, such as by adding liquid, C_2 for the dry pellet is no longer the constant of that system and must be replaced by $C_2{}^G$. The latter is a constant in a certain liquid-loaded pellet, but it will be subject to changes if the liquid-loading parameter and/or pattern of dispersion changes. However, we do not at this early stage feel that $C_2{}^G$ is a strong function of the dispersion pattern. Relative to the liquid-loading parameter, when $q = 0$, $C_2{}^G = C_2$

which characterizes the dry matrix only. On the other hand, when $q = 1$ or when the pellet is fully loaded with liquid, C_2^G attains its minimum value of zero. Performing a series of isobaric diffusion experiments thus enables one to determine the variation of C_2^G with the liquid-loading parameter, q.

RESULTS AND DISCUSSION

Figure 1 shows the changes in the diffusion fluxes of nitrogen per unit mole fraction difference with changes in the liquid-loading parameter, q. Since the difference in mole fraction of nitrogen across the pellet was not the same for all the data points, the normalized nitrogen fluxes in Fig. 1 were obtained by dividing the measured fluxes by the mole fraction difference. As expected, the fluxes reach a maximum value at $q = 0$. Each experimental point was obtained from a single, steady-state, isobaric diffusion experiment; and all the data points in Fig. 1 were taken at pressures above 300 psig to insure transport only by molecular diffusion.

It is important to note that the measured flux ratio of helium and nitrogen came within ±2% of the theoretical flux ratio of 2.65 as predicted by the dusty-gas model. This is a strong indication that the dusty-gas model behaves very well in describing diffusion in liquid-loaded porous pellets. It also indicates that the ethylene glycol in our system was inert, so that no surface diffusion took place.

Equation (24) was used to evaluate C_2^G for each data point, and Fig. 2 shows the variation of C_2^G with the variable, $1 - q$. In this case, C_2^G has a value of zero at $(1 - q) = 0$ and a value of $C_2^G = C_2 = 0.35$ at $(1 - q) = 1$. The solid curve is a parabola and is given by Eq. (25):

$$C_2^G = C_2(1 - q)^2. \tag{25}$$

Comparing Eq. (25) with Eq. (2), we see that $C_2^G = \mathrm{D}_{\mathrm{eff}}^G/D_{AB}$. Regression analysis on the data points and on Eq. (25) shows excellent agreement up to $q = 0.5$. Above this value, the data points deviate slightly from the parabolic behavior indicated by Eq. (25). It is noteworthy that the parabolic dependence of C_2^G on q simplifies the treatment significantly, because C_2 is the only constant that requires determination in order to define the system of liquid-loaded pellets under consideration.

Figure 3 shows the relationship between the dimensionless rate of reaction, r'_A, and the liquid-loading parameter, q, for different values of the molecular Thiele modulus, γ'_M. In Fig. 3, the liquid–gas diffusivity-ratio parameter is constant at a value of $D' = 10^{-5}$. The lowest curve corresponds to $\gamma'_M = 6$, which is an indication that the reaction is fast and diffusion limited. For this case, the optimum value of the liquid-loading parameter occurs at $\bar{q} = 0.33$. The middle curve in Fig. 3 is

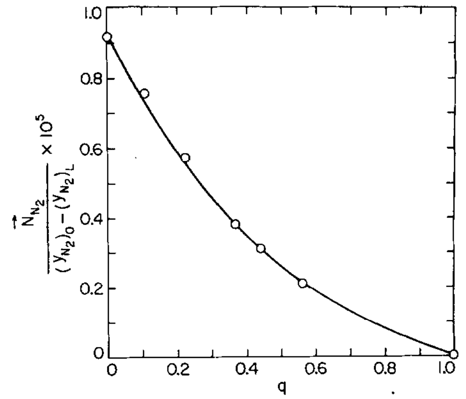

FIG. 1. The effect of liquid loading on nitrogen flux in isobaric diffusion experiments.

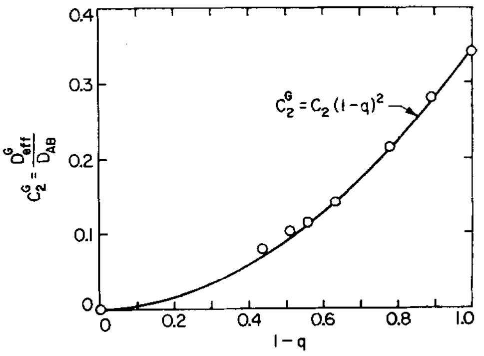

FIG. 2. The variation of the gas-phase effective diffusivity with liquid loading.

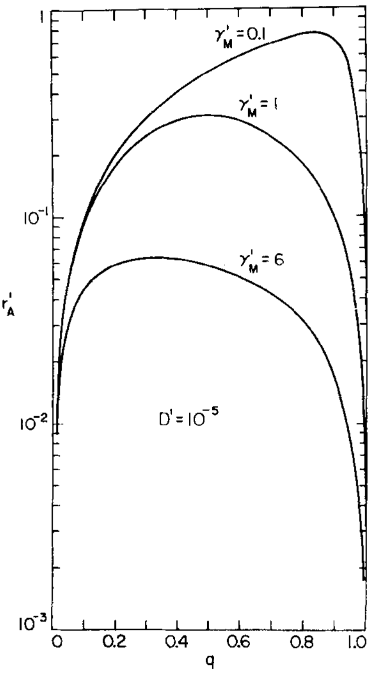

FIG. 3. The variation of the reaction rate with liquid loading showing the effect of the molecular Thiele modulus.

based on $\gamma'_M = 1$ and shows that the optimum liquid loading occurs at $\bar{q} = 0.5$. In this case, the rate of reaction is considered mild, and diffusion limitations still exist. Finally, the upper curve in Fig. 3 corresponds to $\gamma'_M = 0.1$, which is an indication of a very slow reaction. Consequently, the optimum liquid loading has a high value of $\bar{q} = 0.96$. In this case, diffusion limitations are small. It is noteworthy that, when the reaction becomes very slow with no diffusion limitations, the effectiveness factor becomes unity; and, according to Eq. (16), the reaction rate, r'_A, equals q.

An important observation from Fig. 3 is the fact that for a fixed value of q, the rate of reaction, r'_A, increases with decreasing γ'_M. This is explained by the fact that for large γ'_M the reaction occurs in the outer portion of the pellet; and for decreasing γ'_M, it occurs deeper in the pellet, thus using larger liquid volume.

Figure 4 shows a plot of the optimum liquid loading, $\bar{q}$, for a first-order reaction versus the Thiele modulus, γ'_M, at different values of the liquid–gas diffusivity-ratio parameter, D'. At very small values of γ'_M, the reaction is very slow so that diffusion limitations are small and hence $\bar{q}$ is large, approaching unity. As the molecular Thiele modulus becomes larger, the reaction becomes faster; the diffusion limitation is more significant; and $\bar{q}$ becomes smaller. An interesting result of this analysis is the indication that, for a molecular Thiele modulus larger than 5, $\bar{q}$ has a constant value of 0.33.

Figure 4 also shows the effect of changing the liquid–gas diffusivity-ratio parameter, D'. It appears that, for values less than 0.001, it has no effect on the function relating $\bar{q}$ with γ'_M for $\gamma'_M > 0.1$. The upper line of the two diverging curves corresponds to $D' = 0.01$. It is expected that, for $D' > 0.01$, the influence of D' on $\bar{q}$ vs γ'_M would become much more noticeable.

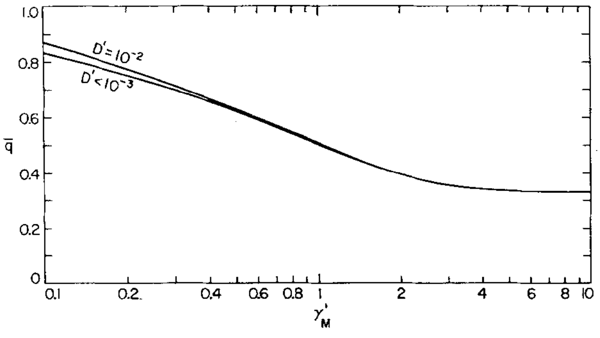

FIG. 4. The variation of the optimum liquid loading with the molecular Thiele modulus showing the effect of the liquid–gas diffusivity-ratio parameter.

CONCLUSIONS

In supported liquid-phase catalysts, as pointed out by Rony (*1*), the dispersion of liquids in porous supports adds additional complications to the transport of materials within each system. Our work suggests that effective diffusivities in liquid-loaded pellets should be determined from experimental information on diffusion fluxes in conjunction with the dusty-gas model, rather than from predictive models which rely on additional measurements such as pore-size distribution in liquid-loaded pellets. These latter measurements would be nearly impossible.

The correlation, which is given by Eq. (2), characterizes the support that was under investigation. Generalizing its usefulness for other porous supports is currently beyond the scope of our work. However, this correlation can easily be obtained for any other porous support which is to be considered.

The important contribution of this approach is the fact that, once the effective diffusivity in liquid-loaded pellets is correlated with the liquid-loading parameter, $\bar{q}$, the model will predict the behavior of a supported liquid-phase catalyst in the presence of chemical reactions.

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