Influence of flow on chemical reaction rates[a]

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(Received 22 November 1978; accepted 20 November 1980)

The influence of convective flow on diffusion-controlled steady-state reaction rates in chemically reactive mixtures is studied for a number of different model systems. Two different flow types are considered: flow in a frictional medium and constant flow in a medium containing many reactive centers. The significance of the results for the combustion of fuel sprays is discussed.

I. INTRODUCTION

In many chemical systems that are of considerable scientific and practical interest, the rate of chemical reactions is governed by hydrodynamic factors, such as molecular diffusion and convection. Examples of such systems are many industrial flow-type chemical reactors, fluidized beds, and combustion of fuel sprays.

Theoretical work on this subject goes back to the classical work of Smoluchowski.[1] The simplest prototype problem is the steady-state diffusion of pointlike particles to a macroscopic sink. The steady-state diffusion equation describing the concentration distribution $c(\mathbf{r})$ of the diffusing particles is

$$D\nabla^2 c = 0 , \tag{I.1}$$

where D is the diffusion coefficient, supplemented by boundary conditions, e.g.,

$$c \to c_0 , \quad \text{as} \quad r \to \infty \tag{I.2}$$

and

$$c = 0 , \quad \text{at the surface of the sink} . \tag{I.3}$$

The latter condition states that the absorption of the diffusing particles at the surface of the sink is *complete*, i.e., the probability that a particle which arrives at the boundary of the sink is absorbed, is unity. Obviously, in real chemical systems this is not the case, and a "radiative" type boundary condition gives a better description which takes account of the possibility of nonreactive encounters.[2] For a spherical sink of radius a, the solution of Eq. (I.1) is well known, e.g., the integrated flux is given by

$$I \equiv \frac{D}{c_0}\, a^2 \int d\Omega \left(\frac{\partial c}{\partial r}\right)_{r=a} = 4\pi D a . \tag{I.4}$$

Equation (I.1) is not appropriate for flowing media. If the fluid in which the diffusing particles are dissolved streams past the sink with a known velocity distribution $\mathbf{u}(\mathbf{r})$, then the equation governing the convective diffusion of the point particles to the sink is

$$(\mathbf{u}\cdot\nabla - D\nabla^2)\, c = 0 . \tag{I.5}$$

It is much more difficult to solve Eq. (I.5) than Eq. (I.1), and, not surprisingly, the work on this subject has focused on several simplifying assumptions,[3–9] e.g., expansions valid at either large or small values of the Peclet number $u_0 a/D$, where u_0 is the asymptotic flow velocity.

Another aspect of diffusion-controlled chemical reaction theory that has received considerable attention in recent years is the "multisink" problem.[10–14] If there are many sinks to which the point particles can diffuse, there will be a change in the rate of absorption per sink due to intersink competitive effects. Most of the work on this subject has been involved with *stagnant media* in which $u_0 = 0$. Relatively little attention has been given to the effect of flow on reaction rates in multiparticle systems, the work of Happel and Pfeffer[15–17] being a notable exception. In the present paper some novel analytical results are given on this subject.

Part of the motivation for this work derives from a recent investigation[14] in which it appeared that the dependence of the calculated burning rate of a fuel spray on the number density of fuel droplets is much stronger than what is observed experimentally.[18–20] It is partly in order to explain this observation, at least qualitatively, that the present study was undertaken.

The model studied in Sec. II consists of one reactive sphere and $N-1$ unreactive spheres, which do not absorb the diffusing species, but do influence the flow by means of frictional drag. The flow pattern is obtained by solving the so-called Debye–Bueche (DB) equation[21–24] which involves a continuum model for the frictional medium. This flow turns out to be simply related to the Stokes flow close to the surface of the reactive sphere. Consequently, the reactive flux is simply related to the one obtained by Levich and others in the case of high Peclet numbers.[3] In Sec. III the full N-reactive-spheres problem is addressed. Again the flow pattern is obtained from the DB equation. Since the resulting convective diffusion equation is prohibitively difficult to solve in all generality, we replace the full DB field $\mathbf{u}(\mathbf{r})$ by an effective, constant field $\mathbf{u}_{av}$, which is essentially the average of the DB field over the volume of the reactive mixture. The resulting reactive flux is studied as a function of the asymptotic velocity u_0 and the number density ρ of absorbers (see Fig. 3). In Sec. IV the experimental significance of the results in Sec. III is discussed.

II. SINGLE SPHERICAL ABSORBER IN A FRICTIONAL MEDIUM

In this section we consider a simple model for the diffusion of point like solute particles to an absorbing

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 0021-9606/81/052904-06$01.00

sphere of radius a which is immersed in a frictional medium. The frictional medium can be thought of as a suspension of many spherical centers, which do not absorb the diffusing particles, but do affect the flow field through frictional attenuation. A continuum description of the latter effect is given by the Debye–Bueche equation[21–24]

$$\eta\nabla^2\mathbf{u} - \zeta\rho\mathbf{u} - \nabla p = 0 , \quad (II.1)$$

where ζ is the friction coefficient of a single droplet, $\rho = \rho(\mathbf{r})$ is the number density of frictional centers, η is the viscosity of the fluid, and p is the hydrostatic pressure. For $\rho(\mathbf{r})$, a cavity model is adopted: The spherical absorber is placed at the center of a spherical cavity of radius R, outside of which there is a constant number density ρ of frictional centers, and inside of which there are no such centers, i.e., $\rho(\mathbf{r}) = 0$ for $r < R$ and $\rho(\mathbf{r}) = \rho$ for $r > R$.

In addition to Eq. (II.1) we impose the equation of incompressibility

$$\nabla \cdot \mathbf{u} = 0 , \quad (II.2)$$

the boundary conditions

$$\mathbf{u} \to \mathbf{u}_0 , \quad \text{as } r \to \infty , \quad (II.3)$$

$$\mathbf{u} = 0 , \quad \text{at } r = a , \quad (II.4)$$

and the continuity conditions

$$\mathbf{u} \text{ continuous at } r = R , \quad (II.5)$$

$$\mathbf{r} \cdot \Sigma \text{ continuous at } r = R , \quad (II.6)$$

where Σ is the stress tensor[25]

$$\Sigma = -p\mathbf{I} + 2\eta(\nabla \cdot \mathbf{u})^0 . \quad (II.7)$$

Here $\mathbf{I}$ is the unit tensor and $(\mathbf{x})^0$ denotes the symmetrized part of $\mathbf{x}$.

The program to be carried out is (i) to solve the boundary-value problem constituted by Eqs. (II.1)–(II.6); and (ii) to substitute this flow field into the convective diffusion equation (I.5) and compute the resulting reactive flux.

With the following ansatz[26]:

$$\mathbf{u} = \nabla \times (\nabla \times f\mathbf{u}_0) \qquad \text{for } a < r < R , \quad (II.8)$$

$$\mathbf{U} = \mathbf{u}_0 + \nabla \times (\nabla \times F\mathbf{u}_0) , \quad \text{for } r > R , \quad (II.9)$$

f and F depend only on radial distance r, and a set of ordinary vectorial differential equations for f and F can be found. After much tedious algebra, the following solutions result:

$$\mathbf{u} = \tfrac{1}{15} f_1 r^2 (\mathbf{rr}\cdot\mathbf{u}_0 - 2\mathbf{u}_0) - \tfrac{1}{2} f_2 (1/r)(\mathbf{rr}\cdot\mathbf{u}_0 + \mathbf{u}_0) - \tfrac{2}{3} f_3 \mathbf{u}_0 + f_4 (1/r^3)(3\mathbf{rr}\cdot\mathbf{u}_0 - \mathbf{u}_0) \quad (a < r < R) , \quad (II.10)$$

$$\mathbf{U} = \mathbf{u}_0 + (f_5/\kappa^2)\mathbf{rr}\cdot\mathbf{u}_0\left(-\frac{3}{r^5} + \frac{3x}{r^5} + \frac{3\kappa x}{r^4} + \frac{\kappa^2 x}{r^3}\right) + \frac{f_5}{\kappa^2}\mathbf{u}_0\left(\frac{1}{r^3} - \frac{x}{r^3} - \frac{\kappa x}{r^2} - \frac{\kappa^2 x}{r}\right) + f_6\,\mathbf{rr}\cdot\mathbf{u}_0 \times\left(\frac{3}{r^5} + \frac{3\kappa}{r^4} + \frac{\kappa^2}{r^3}\right) + f_6\,\mathbf{u}_0 \times\left(-\frac{1}{r^3} - \frac{\kappa}{r^2} - \frac{\kappa^2}{r}\right) \quad (r > R) , \quad (II.11)$$

$$p = p_0 - \eta\mathbf{u}_0\cdot\mathbf{n}\left(\frac{2}{3} f_1 r + \frac{f_2}{r^2}\right) \qquad (a < r < R) , \quad (II.12)$$

and

$$P = p_0 - \eta\mathbf{u}_0\cdot\mathbf{r}\left(\kappa^2 r + \frac{f_4}{r^2}\right) \qquad (r > R) , \quad (II.13)$$

where

$$\kappa^2 \equiv \zeta\rho/\eta , \quad x \equiv e^{-\kappa r} , \quad \mathbf{n} \equiv \mathbf{r}/r .$$

These solutions obey Eqs. (II.1) and (II.2) and satisfy Eq. (II.3). The six constants f_1–f_6 are determined by imposing Eqs. (II.4)–(II.6). This gives rise to six linear equations which are given in the Appendix. The $\kappa^2 r$ term in Eq. (II.13) shows that one has to apply infinite pressure to maintain steady linear flow through an infinitely extended frictional medium.

The solution of the set of equations (A1)–(A6) was found using MACSYMA, a computer language developed at the M.I.T., designed to manipulate algebraic (symbolic) expressions. Rather than giving the complete results, which are long and cumbersome, we shall give the first term in the Taylor expansion of $\mathbf{u}$ close to the surface of the absorbing sphere:

$$u_r = u_0\cos\theta\left\{\frac{3}{2}\,\phi\left(\frac{r-a}{a}\right)^2 + O\left[\left(\frac{r-a}{a}\right)^3\right]\right\} , \quad (II.14)$$

$$u_\theta = -u_0\sin\theta\left\{\frac{3}{2}\,\phi\left(\frac{r-a}{a}\right) + O\left[\left(\frac{r-a}{a}\right)^2\right]\right\} , \quad (II.15)$$

where

$$\phi = \frac{2}{3}\;\frac{270 + 270\sigma + \sigma^2(126 - 90A^2 - 45A^3 + 9A^5) + \sigma^3(36 - 90A^2 + 45A^3 + 9A^5)}{180 + \sigma(180 - 180A) + \sigma^2(24 - 45A + 30A^3 - 9A^5) + \sigma^3(4 - 9A + 10A^3 - 9A^5 + 4A^6)} ,$$

with

$$A \equiv a/R , \quad \sigma \equiv \kappa R . \quad (II.16)$$

In the limit that $\rho \to 0$, we have $\phi \to 1$ and Eqs. (II.14) and (II.15) reduce to the familiar case of Stokes flow near a spherical body.

Levich has developed an analytical treatment of the flux of solute particles to a spherical absorber which is valid if the following two conditions hold: (i) The Reynold's number is low enough so that the flow pattern is Stokesian; (ii) the Peclet number $2u_0a/D$ is so large that a thin boundary layer of diffusing solute particles is

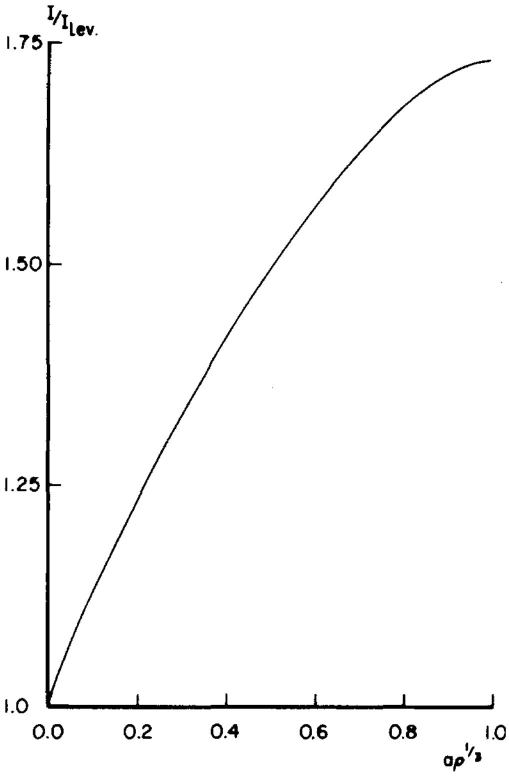

FIG. 1. The ratio of the fluxes I and I_{Lev} in a medium with and without friction, respectively, as a function of the dimensionless density parameter $a\rho^{1/3}$, according to Eq. (II.16). Here, ρ is the number density of frictional particles and a is the radius of the reactive sphere.

formed.[3] With these two provisos Levich arrived at the following expression for the particle flux:

$$I=\frac{Da^2}{c_0}\int d\Omega\left(\frac{\partial c}{\partial r}\right)_{r=a}$$
$$\approx 7.98\,(u_0 D^2 a^4)^{1/3}\equiv I_{\text{Lev}}\ . \tag{II.17}$$

For the cavity model under consideration, the flux is simply related to the single sphere result:

$$I=\phi^{1/3} I_{\text{Lev}}\ . \tag{II.18}$$

Figure 1 gives a plot of $\phi^{1/3}$ as a function of the number density ρ of droplets in the frictional medium. In obtaining this plot, the following assumptions are made: (i) The cavity radius R is equal to the average interparticle distance $\rho^{-1/3}$. (ii) The frictional constant ζ is given by the Stokes law $\zeta=6\pi a'\eta$; in a more accurate treatment the dependence of the friction coefficient on ρ would be explicitly included. (iii) The radius a' of the frictional particles is equal to that of the absorbing sphere ($a'=a$). It follows that $A=a\rho^{1/3}$ and $\sigma=(6\pi a)^{1/2}\rho^{1/6}$.

It is seen from Fig. 1 that when the density ρ of the frictional medium is increased, the flux of diffusing particles also increases. There are two factors which contribute to this effect: (i) As ρ increases, the cavity radius R decreases, so that the diffusing particles have to travel a shorter distance to reach the surface $r=a$; (ii) as ρ increases, the frictional slowing down of the flow by the suspension outside the cavity is more than offset by the concurrent increase in the applied hydrostatic pressure [see Eq. (II.13)]. Similar results have been found by Pfeffer[16] and Pfeffer and Happel.[17] The increase in the hydrostatic pressure is required to "force" the fluid through the suspension, despite the fact that fluid is incompressible, to combat the frictional force imposed on the fluid by the fixed reactive sinks.

III. MANY REACTIVE SPHERES IN DB FLOW FIELD

The problem of greatest practical interest is the study of the flux of particles to an assembly of many reactive centers. The model we consider consists of the following ingredients: We shall adopt a continuum description of the reactive cloud, in which the discrete centers are replaced by a number density field $\rho(\mathbf{r})$. The flow field is again obtained from the DB equation (II.1). In this continuum model, the depletion of solute particles through reactive encounters cannot be described by a boundary condition such as Eq. (I.3), but must be described by means of a continuous sink function $q(\mathbf{r})$ as follows:

$$(\mathbf{u}\cdot\nabla-D\nabla^2)\,c=-4\pi Dq\ . \tag{III.1}$$

In previous work[13] it was shown that in a stationary medium q and c are linked through a linear, constitutive relation

$$4\pi q=\lambda^2 c\ , \tag{III.2}$$

where

$$\lambda^2(\mathbf{r})=4\pi a\rho(\mathbf{r})\ . \tag{III.3}$$

The quantity λ has dimensions of an inverse length and is a measure of the degree of screening exerted on a sink by its neighbors. Solution of Eqs. (II.1) and (III.1) with the appropriate boundary conditions would constitute a complete solution of the problem under consideration. Unfortunately, this program is too ambitions.

For definiteness we shall limit our considerations to a uniform spherical reactive cloud of radius R, i.e., $\rho(r)=\rho$, if $r<R$, and $\rho(r)=0$, if $r>R$. First, consider the DB equation (II.1), supplemented by Eqs. (II.2), (II.3), (II.5), and (II.6); Eq. (II.4) is replaced by the condition that $\mathbf{u}$ is finite at the origin. This problem has been solved by Felderhof.[24] Since we do not have a theory for solving Eq. (III.1) for an arbitrary flow field $\mathbf{u}(\mathbf{r})$, the best we can do is to replace the DB field $\mathbf{u}(\mathbf{r})$ in Eq. (III.1) by an effective field $\mathbf{u}_{\text{av}}$ which is defined as

$$\mathbf{u}_{\text{av}}=f\,\mathbf{u}_0 \tag{III.4}$$

and

$$f^2\equiv\left(\tfrac{2}{3}R^3u_0^2\right)^{-1}\int_0^R r^2\,dr\int_{-1}^{+1}d(\cos\theta)\,\mathbf{u}_{\text{DB}}\cdot\mathbf{u}_{\text{DB}} \tag{III.5}$$
$$=\frac{d^2}{\sigma^4}+\frac{2cd}{\sigma^2}\left(\text{ch}\sigma-\frac{\text{sh}\sigma}{\sigma}\right)$$
$$+c^2\left(-\frac{1}{4}-\frac{1}{4}\,\text{ch}2\sigma-\frac{\text{sh}^2\sigma}{2\sigma^2}+\frac{\text{sh}2\sigma}{2\sigma}+\frac{1}{8}\,\sigma\,\text{sh}2\sigma-\frac{\sigma^2}{4}\right),$$

where

$$\sigma\equiv\kappa R\ ,\quad \kappa^2\equiv\zeta\rho/\eta\ ,\quad d=\frac{3}{2}\ \frac{G(\sigma)}{1+\frac{3}{2}G(\sigma)/\sigma^2}\ , \tag{III.6}$$

$$c = 2d/\sigma^2 \operatorname{ch}\sigma G(\sigma) , \tag{III.7}$$

and

$$G(\sigma) = 1 - \operatorname{th}\sigma/\sigma . \tag{III.8}$$

In Fig. 2, f is plotted as a function of σ.

Consider Eq. (III.1) with $\mathbf{u}$ replaced by $\mathbf{u}_{av}$ and with the following boundary conditions: (i) c is finite at the origin, (ii) $c \to c_0$ as $r \to \infty$, and (iii) c and $\partial c/\partial r$ are continuous across $r = R$. The substitution

$$c = \exp(\boldsymbol{\alpha} \cdot \mathbf{r})\psi , \tag{III.9}$$

with $\boldsymbol{\alpha} \equiv \mathbf{u}_{av}/2D$, leads to the following solution:

$$c = \begin{cases} c_0(1 - e^{\boldsymbol{\alpha}\cdot\mathbf{r}}\psi_{out}) & (r > R) , \\ c_0 e^{\boldsymbol{\alpha}\cdot\mathbf{r}}\psi_{in} & (r < R) , \end{cases} \tag{III.10}$$

where

$$\psi_{out} = \sum_{l=0}^{\infty} d_l k_l(\alpha r) P_l(\cos\theta) , \tag{III.11}$$

$$\psi_{in} = \sum_{l=0}^{\infty} g_l i_l(\beta r) P_l(\cos\theta) , \tag{III.12}$$

$$\beta^2 = \alpha^2 + \lambda^2 , \quad g_l = G_l/\Delta_l , \quad d_l = D_l/\Delta_l , \tag{III.13}$$

$$G_l = (-1)^{l+1}(2l+1)\pi/(2\alpha R^2) , \tag{III.14}$$

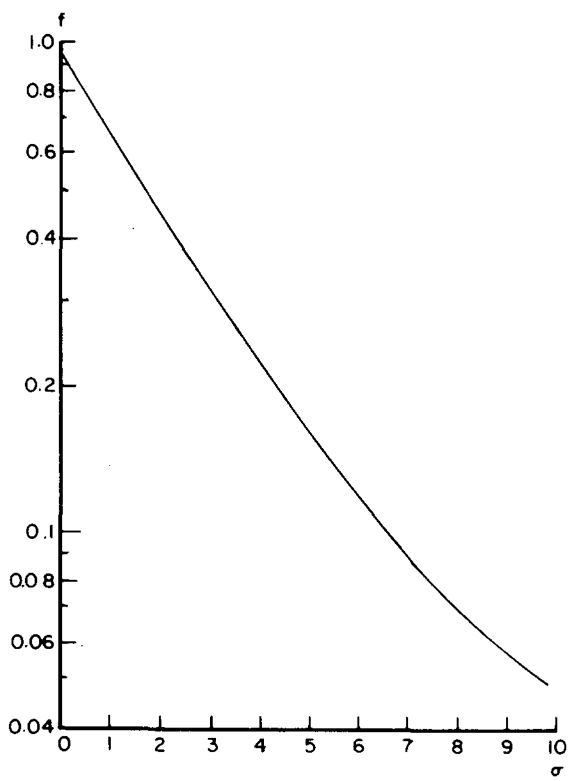

FIG. 2. The average frictional reduction f of the Debye–Bueche flow field by a uniform spherical medium as a function of the dimensionless density parameter σ, according to Eq. (III.5). $\sigma \equiv (\zeta\rho/\eta)^{1/2}R$, where ρ is the number density of frictional particles, ζ is the friction constant, η is the viscosity, and R is the radius of the sphere which contains the reactive centers.

$$D_l = (-1)^l(2l+1)[\alpha i_l(\beta R) i_l'(\alpha R) - \beta i_l(\alpha R) i_l'(\beta R)] , \tag{III.15}$$

$$\Delta_l = \alpha i_l(\beta R) k_l'(\alpha R) - \beta k_l(\alpha R) i_l'(\beta R) , \tag{III.16}$$

$$i_l(x) = (\pi/2x)^{1/2} I_{l+(1/2)}(x) , \quad k_l(x) = (\pi/2x)^{1/2} K_{l+(1/2)}(x) ,$$

$$i_l'(x) = di_l/dx , \quad k_l'(x) = dk_l/dx ,$$

where i_l and k_l are modified spherical Bessel functions of the first and third kind, respectively.[27] The volume-averaged reaction rate (in nondimensional units) is

$$q^* \equiv (\tfrac{4}{3}\pi R^3 \alpha\rho c_0)^{-1} \int d\mathbf{r}\, q(\mathbf{r}) = \frac{3}{R^3} \sum_{l=0}^{\infty} q_l h_l , \tag{III.17}$$

where g_l is defined by Eq. (III.13) and

$$h_l = (R^2/\lambda^2)[\beta i_l(\alpha R) i_{l-1}(\beta R) - \alpha i_{l-1}(\alpha R) i_l(\beta R)] . \tag{III.18}$$

We can now study several limiting cases of Eq. (III.17). In the limit that $\mathbf{u}_0 \to 0$ (no convection), only the $l = 0$ term survives and one obtains

$$q^*(\alpha, \lambda) \xrightarrow[\alpha\to 0]{} \frac{3}{\lambda^2 R^2}\left(1 - \frac{\operatorname{th}\lambda R}{\lambda R}\right) , \tag{III.19}$$

the same result as in the purely diffusive case.[10–13] In the limit that $\lambda \to 0$ (no screening), one obtains

$$q^*(\alpha, \lambda) \xrightarrow[\lambda\to 0]{} \frac{3}{2} \sum_{l=0}^{\infty} (-1)^l (2l+1) \times [i_l^2(\alpha R) - i_{l+1}(\alpha R) i_{l-1}(\alpha R)] . \tag{III.20}$$

The above equality follows from the identities

$$\sum_{l=0}^{\infty} (-1)^l (2l+1) i_l^2(x) = 1 \tag{III.21}$$

and

$$\int_0^z dx\, x^2 i_l^2(x) = \tfrac{1}{2} z^3 [i_l^2(z) - i_{l+1}(z) i_{l-1}(z)] . \tag{III.22}$$

In the limit that $\lambda \to \infty$ (an infinitely dense spray), $q^* \to 0$ as λ^{-2} and

$$I(\alpha) \equiv \lim_{\lambda\to\infty} \tfrac{1}{3}\lambda^2 R^2 q^*(\alpha, \lambda) = \frac{\pi}{2\alpha R} \sum_{l=0}^{\infty} (-1)^l (2l+1) \frac{i_l(\alpha R)}{k_l(\alpha R)} . \tag{III.23}$$

This result can be obtained by considering a single sphere of radius R in a constant flow field.

For arbitrary values of α and λ, the summation in Eq. (III.17) can be performed numerically. In Fig. 3, q^* is plotted as a function of the screening number λR, varying between 0 and 10, for different values of the Peclet number $\alpha_0 R \equiv u_0 R/2D = 0$, 5, 10, 50, and 100. Note that α differs from α_0 by a density-dependent factor f, given by Eq. (III.5). For values of αR larger than about 10, the numerical algorithm used to compute the sum in Eq. (III.17) is not reliable, due to cancellations resulting from the alternating sign of consecutive terms in Eq. (III.17). Thus, in Fig. 3, in the curve for $\alpha_0 R = 50$ when $\lambda R < 4$ and in the curve for $\alpha_0 R = 100$ when $\lambda R < 5$, the curves are smooth extensions of the large-λR sections that go through 1 at $\lambda R = 0$ as expected from Eq. (III.20).

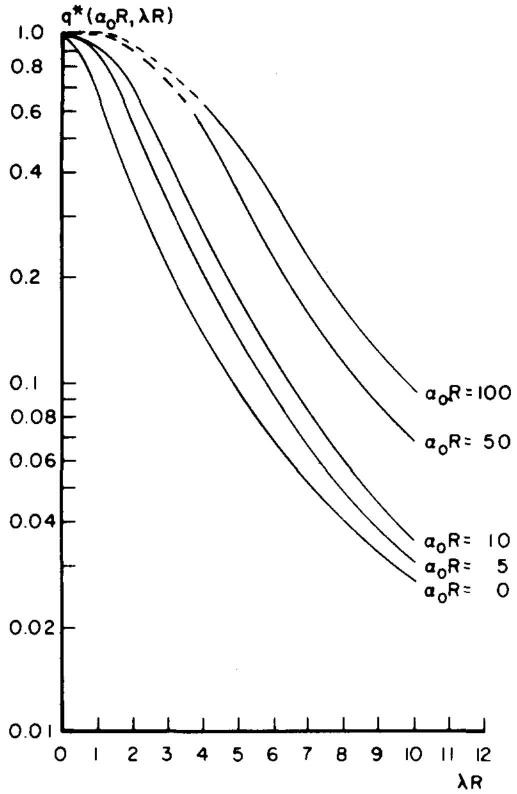

FIG. 3. The average, nondimensional reaction rate q^* of a uniform, spherical cloud of absorbers as a function of the density parameter λR for different values of the Peclet number $\alpha_0 R$, according to Eq. (III.17). Here, $q^* \equiv [(4\pi/3) R^3 c_0]^{-1} \times \int_{r<R} d\mathbf{r}\, c(\mathbf{r})$, where $c(\mathbf{r})$ is the concentration distribution of diffusing particles and c_0 is its asymptotic value and R is the radius of the reactive cloud; $\lambda \equiv (4\pi a \rho)^{1/2}$, where a is the radius of the individual reactive droplets in the cloud and ρ is the number density of droplets in the cloud; and $\alpha_0 \equiv u_0/2D$, where u_0 is the asymptotic flow velocity and D is the diffusion constant. The dotted parts of the curves labeled $\alpha_0 R = 50$ and 100 are not obtained by direct calculation, but are merely smooth extensions of the corresponding solid parts of the curves that go through 1 at $\lambda R = 0$.

It is seen that for $\lambda < \alpha_0$, the dependence of q^* on λR becomes gradually weaker as $\alpha_0 R$ is increased. For $\lambda > \alpha_0$, the curves for different values of $\alpha_0 R$ approach the curve with $\alpha_0 R = 0$ as λR increases.

IV. DISCUSSION OF RESULTS

In this section, the experimental significance of the results of the last section is briefly indicated. In two recent publications[13,14] we have formulated a simple model theory for the burning of fuel sprays, which takes screening effects between fuel droplets in the spray into account, but which ignores convective flow. In Ref. 13 we considered the same uniform spherical spray model as in Sec. III and in Ref. 14 we considered a one-dimensional spray model where the number of droplets is determined self-consistently, In both cases, the dependence of the calculated burning rate q^* on the number density ρ of droplets was found to be stronger than what is experimentally observed.[18] The following argument makes it plausible that q^* becomes progressively less dependent on ρ as the flow rate u_0 is increased. At high flow rates, a diffusion boundary layer is set up around each droplet, the thickness of which is inversely proportional to u_0. Outside the boundary layers, the concentration of diffusing particles $c(\mathbf{r})$ is more or less constant ($\approx c_0$), the concentration gradient being limited to the region inside the boundary layers of the droplets. If the average interdroplet distance is much larger than the boundary layer thickness, then there is no possibility of interdroplet competition. Hence, it follows that a high flow rates u_0, q^* is relatively insensitive to variations in ρ, as is experimentally observed. This is borne out by the results in Fig. 3.

It should be pointed out, however, that our results are only qualitatively in agreement with the experimental results of Ref. 18. In the latter work, q^* is found to be approximately 0.5 over a large range of interdroplet distances, while in our work q^* approaches 1 for $\alpha_0 \gg \lambda$. We suspect that this is not due to interparticle effects, but to the fact that the spray experiments[18] were performed under other flow conditions than the single droplet experiments[28] with respect to which the spray burning rates are expressed.

ACKNOWLEDGMENTS

One of us (R.S.) wishes to thank Dr. D. Ronis for several very helpful suggestions and stimulating discussions. The computer language MACSYMA was developed by the Mathlab group at M.I.T. supported by the Defense Advanced Research Projects Agency work order 2095, under Office of Naval Research Contract #N00014-75-C-0661.

APPENDIX

The equations which determine the six constants f_1–f_6 occurring in Eqs. (II.10)–(II.13) are summarized here. They follow from the boundary and continuity conditions (II.4)–(II.6):

$$f_1 a^2/15 + f_2/a + 2f_3/3 - 2f_4/a^3 = 0 \; , \tag{A1}$$

$$2f_1 a^2/15 + f_2/2a + 2f_3/3 + f_4/a^3 = 0 \; , \tag{A2}$$

$$-f_1/15 - f_2 - 2f_3/3 + 2f_4 = 1 + 2f_5(\kappa x + x - 1)/\kappa^2 + 2f_6 x(\kappa + 1) \; , \tag{A3}$$

$$-2f_1/15 - f_2/2 - 2f_3/3 - f_4 = 1 + f_5(-\kappa^2 x - \kappa x - x - 1)/\kappa^2 - f_6 x(\kappa^2 + \kappa + 1) \; , \tag{A4}$$

$$2f_1/5 + 3f_2 - 12f_4 = \kappa^2 + f_5(\kappa^2 - 4\kappa^2 x - 12\kappa x - 12x + 12)/\kappa^2 - 4f_6 x(\kappa^2 + 3\kappa + 3) \; , \tag{A5}$$

$$-4f_1/15 + f_2/2 + 3f_4 = f_5(\kappa^3 x + 2\kappa^2 x + 3\kappa x + 3x - 3)/\kappa^2 + f_6 x(\kappa^3 + 2\kappa^2 + 3\kappa + 3) \; . \tag{A6}$$

Here, $x = \exp(-\kappa R)$ and we have set $R = 1$.

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[26]The referee correctly points out that in a consistent treatment the shielding length λ, defined in Eq. (III.3), should depend upon the velocity u_0, reflecting the influence of the moving medium on the sink absorption rate. An indication of the influence that large u_0 might have on λ (at low ρ) is given by the single sphere result [Eq. (II.17)]. We believe that at low u_0, λ will approach the $u_0 = D$ limit displayed in Eq. (III.3). The problem of generalizing Eq. (III.5) to finite u_0 is left open.
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