CHEMICAL REACTION WITH HEAT LIBERATION ON THE SURFACE OF A THERMALLY CONDUCTIVE PARTICLE MOVING IN A GAS

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A determination is made of the temperatures and concentration fields resulting from occurence of a multicomponent chemical reaction on the surface of a sphere moving in a gas at low Reynolds and Peclet numbers. It is assumed that the particle is thermally conductive, and that the rate of surface chemical reaction depends in an arbitrary manner on the temperature and concentration distribution near the particle. The problem is solved by the method of combining asymptotic expansions in small Peclet numbers. Approximate analytical formulas are obtained for the mean Sherwood and Nusselt numbers.

The concentration distribution produced by a reacting particle moving in a gas in the diffusion reaction regime was obtained previously [1-4] for the isothermal case, and also for surface reactions of first [5-7], second [5], and arbitrary orders [8]. In [9] the temperature field without and within the thermally-conductive particle was determined for the case of complete reagent absorption on the surface.

1. Formulation of the Problem

We will consider convective diffusion to a thermally conductive reacting sphere, with a translational Stokes flow passing over it, and a nonisothermal multicomponent chemical reaction occurring on the surface, the reaction rate being dependent on temperature and concentration in an arbitrary manner. It is assumed that the presence of the surface reaction does not affect flow and particle parameters.

The dimensionless equations of convective diffusion and thermal conductivity, together with the boundary conditions expressing homogeneity of temperature and concentrations far from the particle, continuity of temperature and heat balance on the surface, the "reaction law," and the finiteness of temperature at the center of the particle have the form [9, 10]

$$\begin{array}{ll} \Delta c_{m} = \operatorname{Pe}_{m} r^{-2} J(\psi, c_{m}) & (1 < r < \infty), & m = 1, \dots, M; \\ \Delta T = \operatorname{Pe}_{0} r^{-2} J(\psi, T) & (1 < r < \infty); \\ \Delta t = 0 & (0 \leq r < 1); \\ r \to \infty, c_{m} \to 0, T \to 0; \\ r = 1, T = t; \\ r = 1, \partial c_{m} / \partial r = f_{m}(c_{1}, \dots, c_{M}, T); \end{array}$$

$$(1.1)$$

$$(1.2)$$

$$(1.2)$$

$$(1.2)$$

$$(1.3)$$

$$(1.4)$$

$$(1.6)$$

$$r = 1, \quad \frac{\partial T}{\partial r} - \delta \frac{\partial t}{\partial r} = \sum_{m=1}^{M} h_{m} (c_{1}, \ldots, c_{M}, T); \qquad (1.7)$$

$$r = 0, |t| < \infty; \tag{1.8}$$

$$\psi = \frac{1}{2} \left(r^2 - \frac{3}{2} r + \frac{1}{2r} \right) (1 - \mu^2), \quad J(\psi, x) = \frac{\partial(\psi, x)}{\partial(r, \mu)}, \tag{1.9}$$

$$\begin{aligned} c_m^* &= c_{m\infty} (1 - c_m), \ T^* &= T_\infty (1 - T), \ t^* &= T_\infty (1 - t), \\ \mathrm{Pe}_m &= a U D_m^{-1}, \ \mathrm{Pe}_0 &= a U \chi^{-1}, \ \delta &= \lambda_1 \lambda^{-1}, \ h_m &= c_{m\infty} D_m H_m (\lambda T_\infty)^{-1}, \\ f_m (c_1, \ldots, c_M, T) &= -a (c_{m\infty} D_m)^{-1} F_m (c_1^*, \ldots, c_M^*, T^*), \ \mu &= \cos \theta, \end{aligned}$$

where c_m^* are the reagent concentrations; T* and t* are the temperatures in the gas flow and inside the particle; $e_{m\infty}$ and T_{∞} are the concentrations and temperature at infinity; Peo and Pem are the thermal and diffusion Peclet numbers; α is the particle radius; U is the flow velocity far from the particle; D_m are the diffusion coefficients; χ is the thermal diffusivity; ψ is the flow flunction; H_m is the heat of the m-th reaction; F_m is the rate of the surface reaction; λ_1 and λ are the thermal conductivity coefficients of particle and gas); M is the number of reagents participating in the reaction); r, θ is a spherical coordinate

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29

system fixed to the particle; $\partial(\psi, x)/\partial(r, y)$ is the Jacobian of the functions ψ and x.

2. Method of Solution. Heat and Mass Flux to Particle Surface.

We will study the boundary problem Eqs. (1.1)-(1.9) by the method of combinable asymptotic expansions in small Peclet numbers [1-9]. It will be assumed that

$$\varepsilon \to 0$$
, $\operatorname{Pe}_m = \varepsilon Q_m$, $Q_m = O(1)$ $(m = 0, 1, \ldots, M)$

(such a situation is typical of gases, where $0.5 \leq \operatorname{Pe}_m/\operatorname{Pe}_0 \leq 2$) and the entire flow region will be divided into two subregions: an inner $\Omega_1 = \{1 \leq r \leq O(\varepsilon^{-1})\}$ and an outer $\Omega_{\infty} = \{O(\varepsilon^{-1}) \leq r\}$ [1-9]. As usual, in the outer region a "compressed" coordinate $\rho = \varepsilon r$ is introduced, and a solution in each region is sought separately in the form of inner and outer expansions. In constructing the asymptotic solution in the inner region, boundary conditions on the particle surface, Eqs. (1.5)-(1.7), are used, while in the outer region conditions at infinity are employed, Eq. (1.4). The unknown constants appearing in the solution are determined by the combination procedure [1-9].

By analogy to [1-9] it can be shown that in the case of translational Stokes flow over the sphere, Eq. (1.9), the temperature and concentration distributions on the inner surface of the outer region Ω_{∞} (i.e., at $r \sim \varepsilon^{-1}$) can be represented in the form (m = 1, ..., M)

$$r = O(\varepsilon^{-1}), \quad T = \Psi (\operatorname{Pe}_{0^{\sharp}} \dots, \operatorname{Pe}_{M}) \left\{ r^{-1} + \frac{1}{2} \operatorname{Pe}_{0}(\mu - 1) - \frac{1}{2} \operatorname{Pe}_{0}^{2} \ln \operatorname{Pe}_{0} + O(\operatorname{Pe}_{0}^{2}) \right\},$$

$$r = O(\varepsilon^{-1}), \quad c_{m} = \Phi_{m} (\operatorname{Pe}_{0}, \dots, \operatorname{Pe}_{M}) \left\{ r^{-1} + \frac{1}{2} \operatorname{Pe}_{m}(\mu - 1) - \frac{1}{2} \operatorname{Pe}_{m}^{2} \ln \operatorname{Pe}_{m} + O(\operatorname{Pe}_{m}^{2}) \right\},$$
(2.1)

where Ψ and Φ_m are unknown functions to be determined in the course of solution.

For the inner expansion of temperature and concentrations in the region Φ_m , and also for the temperature distribution within the particle, we have

$$T = T^{(0)} + \operatorname{Pe}_{0} T^{(1)} - \operatorname{Pe}_{0}^{2} \ln \operatorname{Pe}_{0} T^{(2)} + O(\operatorname{Pe}_{0}^{2}),$$

$$t = t^{(0)} + \operatorname{Pe}_{0} t^{(1)} + \operatorname{Pe}_{0}^{2} \ln \operatorname{Pe}_{0} t^{(2)} + O(\operatorname{Pe}_{0}^{2}),$$

$$c_{m} = c_{m}^{(0)} + \operatorname{Pe}_{m} c_{m}^{(1)} + \operatorname{Pe}_{m}^{2} \ln \operatorname{Pe}_{m} c_{m}^{(2)} + O(\operatorname{Pe}_{m}^{2}),$$

(2.2)

where the functions $c_m^{(k)}$, $T^{(h)}$, and $t^{(k)}$ satisfy the equations

$$\Delta c_m^{(h)} = 0, \quad \Delta T^{(k)} = 0, \quad \Delta t^{(k)} = 0 \quad (k = 0, 2),$$

$$\Delta c_m^{(1)} = \alpha_m r^{-2} J(\psi, r^{-1}), \quad \Delta T^{(1)} = \beta r^{-2} J(\psi, r^{-1}), \quad \Delta t^{(1)} = 0.$$
(2.3)

In deriving the second group of Eqs. (2.3) (at k = 1) it is considered that the zero order terms of the inner expansion depend solely on radial coordinate r and are determined by a binomial of form $A + Br^{-1}$, where for each $c_m^{(0)} T^{(0)}$ and $t^{(\circ)}$ there are unique constants A and B, which are determined from boundary conditions (1.5)-(1.8), and the conditions for combination with the solution in the region Ω_{∞} (2.1); α_m , $\beta = \text{const.}$

Integrating Eq. (2.3) over the surface S_r of a sphere of radius r, we obtain

$$\mathbf{L} \langle c_m^{(k)} \rangle = 0, \quad \mathbf{L} \langle T^{(h)} \rangle = 0, \quad \mathbf{L} \langle t^{(h)} \rangle = 0 \qquad (k = 0, 1, 2),$$

$$\mathbf{L} \equiv \frac{1}{r^2} \frac{d}{dr} \quad r^2 \frac{d}{dr^2} \langle u \rangle \equiv \frac{1}{4\pi r^2} \int_{S_r}^{0} u ds = \frac{1}{2} \int_{-1}^{1} u d\mu.$$
(2.4)

In deriving Eq. (2.4) it was considered that $\langle J(\psi, r^{-1}) \rangle = 0$. The general solution of the equations for mean Eq. (2.4) has the form

$$\langle c_m^{(k)} \rangle = a_{m1}^{(k)} + b_{m1}^{(k)} r^{-1}, \quad \langle T^{(k)} \rangle = a_2^{(k)} + b_2^{(k)} r^{-1}, \quad \langle t^{(k)} \rangle = a_3^{(k)}$$

$$(k = 0, 1, 2, a_{m1}^{(k)}, b_{m1}^{(k)}, b_2^{(k)}, a_2^{(k)}, a_3^{(k)} = \text{const}).$$

$$(2.5)$$

In writing the last equation of (2.5) it was considered that the solution must be finite within the particle, Eq. (1.8).

From Eqs. (2.2) and (2.5) for the full means we obtain

$$\langle c_m \rangle = \Phi_m (\operatorname{Pe}_0, \ldots, \operatorname{Pe}_M) \left\{ r^{-1} - \frac{1}{2} \operatorname{Pe}_m - \frac{1}{2} \operatorname{Pe}_m^2 \ln \operatorname{Pe}_m \right\},$$



$$\langle T \rangle = \Psi \left(\operatorname{Pe}_{0^{j}} \dots, \operatorname{Pe}_{M} \right) \left\{ r^{-1} - \frac{1}{2} \operatorname{Pe}_{0} - \frac{1}{2} \operatorname{Pe}_{0}^{2} \ln \operatorname{Pe}_{0} \right\},$$
(2.6)
$$\langle t \rangle = \Psi \left(\operatorname{Pe}_{0}, \dots, \operatorname{Pe}_{M} \right) \left\{ 1 - \frac{1}{2} \operatorname{Pe}_{0} - \frac{1}{2} \operatorname{Pe}_{0}^{2} \ln \operatorname{Pe}_{0} \right\}.$$

The form of the functions $\langle c_m \rangle$ and $\langle T \rangle$ (2.6) is determined by use of the procedure of combination with the solution in the outer region equation (2.1) with consideration of the equality $\langle \mu \rangle = 0$, while the form of $\langle t \rangle$ is determined by Eq. (1.5)

Since $c_m^{(0)}$, $T^{(0)}$ and $t^{(\circ)}$ depend solely on distance from the center of the sphere, then for any (analytic) function f, to an accuracy of $O(\epsilon^2)$, the expression

$$\langle f(c_1, \ldots, c_M, T) \rangle = f(\langle c_1 \rangle, \ldots, \langle c_M \rangle, \langle T \rangle)$$
(2.7)

is valid, which can be proved by direct test with consideration of Eqs. (2.2) and (2.6) and the properties of the operation $\langle \rangle$.

The mean Sherwood and Nusselt numbers are given by

$$\operatorname{Sh}_{m} = -\left\langle \frac{\partial c_{m}}{\partial r} \right\rangle \Big|_{r=1} = \Phi_{m}, \quad \operatorname{Nu} = -\left\langle \frac{\partial T}{\partial r} \right\rangle \Big|_{r=1} = \Psi.$$
 (2.8)

Averaging boundary conditions equations (1.6) and (1.7) and using Eq. (2.6) and the properties of Eq. (2.7), to determine the mean Sherwood and Nusselt numbers we obtain the following algebraic (transcendental) system of equations:

$$-\operatorname{Sh}_{m} = j_{m} \left(\frac{\operatorname{Sh}_{1}}{\operatorname{Sh}_{1\infty}}, \dots, \frac{\operatorname{Sh}_{M}}{\operatorname{Sh}_{M\infty}}, \frac{\operatorname{Nu}}{\operatorname{Nu}_{\infty}} \right) \qquad (m = 1, \dots, M),$$

$$\operatorname{Nu} = \sum_{m=1}^{M} h_{m} \operatorname{Sh}_{m}.$$
(2.9)

Here the last equation was obtained by substitution in boundary condition (1.7) of the expression for f_m from Eq. (1.6), and the values of $Sh_{m\infty}$ and Nu_{∞} correspond to a purely diffusion (thermal) reaction regime (which corresponds to boundary conditions on the sphere surface r = 1, $c_m = 1$, and T = 1 for Eqs. (1.1), (1.2), and (1.4)) [1]

$$Sh_{m\infty} = 1 + \frac{1}{2} Pe_m + \frac{1}{2} Pe_m^2 \ln Pe_m + O(Pe_m^2),$$

$$Nu_{\infty} = 1 + \frac{1}{2} Pe_0 + \frac{1}{2} Pe_0^2 \ln Pe_0 + O(Pe_0^2).$$
(2.10)

Thus, it has been shown that in the case of a multicomponent surface chemical reaction, accompanied by heat liberation, at small Peclet numbers to determine the integral heat and mass fluxes of reacting components to the particle it is sufficient to solve the algebraic (transcendental) system equations (2.9) and (2.10), which is significantly simpler than the original system of partial differential equations (1.1)-(1.9).

It is evident from system (2.9), (2.10) that at small Peclet numbers the ratio of the thermal conductivities of the particle and surrounding fluid does not affect the integral characteristics of the process. A change in the parameter δ leads only to a redistribution of local thermal and diffusion flows on the sphere surfaces and does not change the corresponding total fluxes.

The results of [8] can be obtained by solving the first equation of (2.9) for M = 1, $f_1 = kf$. The last equation of (2.9) gives the result of [9] for M = 1.

3. Case of Surface Chemical Reaction Following Arrhenius' Law.

We will now consider a first-order heterogeneous chemical reaction (in the presence in the flow of one reacting component M = 1), occurring in accordance with the Arrhenius law on the surface of the sphere. In this case the surface reaction rate is determined by the expression [10] $F = k_0 c^* \exp(-E/RT^*)$. Here and below the subscript will be omitted from the concentration and temperature; E is the activation energy; R is the universal gas constant; k_0 is the reaction rate constant.

System (2.9)-(2.10) reduces to the following equation for determination of the mean Sherwood number:

$$Sh = \frac{ak_0}{D} \left(1 - \frac{Sh}{Sh_{\infty}} \right) \exp\left\{ -\omega \left(1 - h \frac{Sh}{Nu_{\infty}} \right)^{-1} \right\}, \quad \omega = \frac{E}{RT_{\infty}}$$

$$(h = -h_1 > 0).$$
(3.1)

For convenience in analysis we write Eq. (3.1) in the form of a system

$$q = k \exp(-\omega/z) [1 + k \exp(-\omega/z)]^{-1}; \qquad (3.2)$$

$$q = \tan \phi(z - 1)$$
 (z = 1 + σq , $\tan \phi = \sigma^{-1}$), (3.3)

$$q = \mathrm{Sh/Sh}_{\infty}, \ k = ak_0/(D\mathrm{Sh}_{\infty}), \ \sigma = h\mathrm{Sh}_{\alpha}/\mathrm{Nu}_{\infty}.$$

Now, considering the parameter k fixed, we will study the number of roots of Eq. (3.1) as a function of variation of the parameters ω , $\sigma \in [0, +\infty)$ (the angle φ , $0 < \varphi < \pi/2$). The number of roots of Eq. (3.1) is determined by the number of intersections of straight line (3.3), passing at an angle φ to the z-axis in the plane z, q through the point (1, 0) with curve (3.2) (Fig. 1). Then, depending on the value of the parameter ω the following situations are possible; 1) at any $\sigma \in [0, +\infty)$ system (3.2)-(3.3) has a single root $q = q(\omega, \sigma)$; 2) there exists an interval (σ_1, σ_2) in which for every $\sigma_1 < \sigma < \sigma_2$ system (3.2)-(3.3) has three roots, while at the end points of the interval $\sigma = \sigma_n$ (n = 1, 2) there are two roots; in this case for every σ lying outside this interval $0 \leq \sigma < \sigma_1$ or $\sigma_2 < \sigma$, there is a single root to Eq. (3.1), $\varphi_n = \arctan \sigma_n^{-1}$.

It can be shown that the geometric point of tangency of straight line (3.3) with curve (3.2) (which corresponds to two roots of Eq. (3.1)) in the plane $\omega\sigma$ is given in parametric form by

$$\omega(q) = (1 - q)y^2 G^{-1}(1), \ \sigma(q) = q^{-1} G^{-1}(1),$$

$$y = \ln \{k(q^{-1} - 1)\}, \ G(n) = G(q, k, n) = (1 - nq)y - n.$$
(3.4)

The limiting curve (3.4) as $\omega \rightarrow \infty$ has two branches departing to infinity; the upper branch asymptotically approaches the curve

$$\sigma = k^{-1} \omega^{-1} \exp \omega \ (\omega \to \infty), \tag{3.5}$$

while the lower has the asymptote

$$\sigma = (q_*^{-1} - 1)\omega - 2q_*^{-1} \qquad (\omega \to \infty)_{\mathbf{s}}$$
(3.6)

where $q_* < k(k + 1)^{-1}$ is a root of the equation $G(q_*, k, 1) = 0$.

Curve (3.4) in the plane $\omega\sigma$ has a sharp-pointed singular point (ω_0 , σ_0), which is its absolute minimum, lies on the hyperbola

$$D(\omega, \sigma) \equiv \sigma \omega - 4\sigma - 4 = 0 \tag{3.7}$$

and is given by the value of the parameter $q = q_0$, where q_0 is the root of the equation $G(q_0, k, 2) = 0$.

The local behavior of curve (3.4) in the vicinity of the singular point is given by

$$\sigma - \sigma_0 = \tan \gamma (\omega - \omega_0), \quad \omega \ge \omega_0, \quad \tan \gamma = (1/2) q_0^{(-1)} - 1,$$

$$\omega_0 = \omega (q_0) = 4 (1 - q_0) (1 - 2q_0)^{-1}, \quad \sigma_0 = \sigma (q_0) = q_0^{-1} - 2.$$
(3.8)

In Fig. 2, in the plane $\omega\sigma$ the behavior of Eq. (3.4) is depicted qualitatively. In the shaded region Ω , located between the two branches of Eq. (3.4) (defined by parameter q ranges $0 < q < q_0$ and $q_0 < q < q_{\star}$, respectively) at a fixed value of $\omega > \omega_0$, the segment $l = l(\omega) = {\sigma_1(\omega) \leq \sigma \leq \sigma_2(\omega)}$ (shown by the heavy line in Fig. 2) gives the same values of σ at which Eq. (3.1) has three roots.

It can be shown that $\partial q_*/\partial k > 0$, $\partial q_0/\partial k > 0$. Therefore, upon change in the parameter k with consideration of Eqs. (3.5)-(3.8) the region $\Omega = \Omega(k)$ deforms in the following manner: The singular point, lying on the parabola equations (3.7), moves downward and to the right upon increase in k, the slope γ with respect to the ω axis then decreasing; the asymptotic curve equation (3.5), corresponding to the maximum possible values of the parameter σ , approaches the ω axis in inverse proportion to the value of k; the second asymptote, Eq. (3.6), approaches the ω axis, and its point of intersection with the ω axis remains in the right semiplane, moving to the right from zero.

In Fig. 2, dashed lines I and II correspond to asymptotic curves of Eqs. (3.5) and (3.6), the arrows on the boundary of the region Ω indicate the direction of motion of the points of curve (3.4) with increasing k, the arrows on the hyperbola $D(\omega, \sigma) = 0$ show the direction of motion of the singular point (ω_0, σ_0) with increase in k, and the arrows on the ω axis show the direction of motion of the points of intersection of asymptote (3.6) with the ω axis with increasing k, the coordinate of the intersection point always being more than two.

From system (3.2)-(3.3) it is evident that the following limiting relationships exist:

$$\sigma \to 0, \ q \to (1 + k^{-1} e^{\omega})^{-1}, \ \sigma \to \infty, \ q \to k(k+1)^{-1}.$$
(3.9)

With decrease in particle velocity the mean Sherwood and Nusselt numbers corresponding to a diffusion reaction regime (2.10) decrease. Then, as follows from Eq. (3.1), the point A, corresponding to a high temperature reaction regime, moves to the left along curve (3.2) to point B (see Fig. 1). After passage through B the high temperature regime can no longer exist, so that B "jumps" from the upper portion of the curve to the lower at point C, which now corresponds to a low-temperature reaction regime on the particle surface.

It is evident from Eq. (3.2) that the mean Sherwood number decreases with increase in heat of reaction h.

4. Some Remarks.

From the equations for mean Sherwood and Nusselt numbers, Eqs. (2.9) and (2.10), it is evident that these quantities do not depend on the ratio of the thermal conductivity coefficients of particle and fluid δ . We will show that the last equation of (2.9) is valid in the most general case of arbitrary particle surface form Γ and flow field at any Peclet numbers $0 \leq \text{Pe}_m < \infty$.

In fact, since the temperature field t within the particle satisfies Laplace equation (1.3), then for any closed contour γ lying within the particle, the surface integral of $\partial t/\partial n$ (where $\partial/\partial n$ is the derivative with respect to the normal to the surface) over γ is equal to zero. Now, fixing γ and Γ and considering the continuity of the corresponding integral function over γ , we find that the integral over the particle surface Γ of $\partial t/\partial n$ is also equal to zero. With consideration of this, by integrating boundary condition (1.7) over the particle surface and using Eq. (1.6), we obtain the last equation of Eq. (2.9), which expresses the law of conservation of energy. In this sense, the parameter δ , which figures in boundary condition (1.7), is of little significance.

The last equation of (2.9) permits determination of the integral thermal characteristics of the problem in those cases in which the total diffusion flux on the particle is known. It is most simple to use Eq. (2.9) when the surface reaction rate does not depend on temperature, f = f(c), M = 1. Under such conditions diffusion problem (1.1), (1.4), (1.6) can be solved independently of the thermal problem, after which the calculated mean Sherwood number and Eq. (2.9) are used to calculate the mean Nusselt number. Then in the special case of small Peclet numbers it is possible to use the results of [2, 6], obtained for the case of total absorption of matter on the reacting surface and a first-order chemical reaction f(c) =kc, occuring on the surface of a particle of arbitrary form in the presence of a translational Stokes flow. For the thermal Nusselt number, from the last equation of Eq. (2.9) we obtain

$$\mathrm{Nu} = -h\left\{\mathrm{Sh}_{\mathrm{e}} \pm \frac{\mathrm{f}}{4}\,\mathrm{Sh}_{0}^{2}\,\mathrm{Pe} \pm \frac{\mathrm{f}}{4}\,\mathrm{Sh}_{0}^{2}\,(\mathbf{F}\cdot\mathbf{i})\,\mathrm{Pe}^{2}\,\mathrm{ln}\,\mathrm{Pe}\right\} \pm O\,(\mathrm{Pe}^{2}),$$

$$\mathrm{Nu} = \frac{1}{2\pi} \int_{\Omega} \nabla T d\Gamma, \quad \mathbf{F} = \frac{F^0}{6\pi\mu a |V|}, \quad \mathbf{i} = \frac{|V|}{|V|}, \quad \mathrm{Pe} = \frac{e|V|}{D},$$

where U is the flow velocity at infinity; a is the radius of a sphere of equivalent volume; F is a dimensionless vector, equal to the ratio of the resistance force F^0 of the given particle to the value of the Stokes resistance force of a solid sphere of radius $a; \mu$ is the dynamic viscosity of the fluid; Sho is the Sherwood number corresponding to the mass exchange of a particle at rest with an immobile medium ($U \equiv 0$) in the case in which a first order chemical reaction occurs on the particle surface

The last formula of Eq. (2.9) is thus a generalization of the results of [9] to the case of reacting particles of arbitrary form and finite reaction rate.

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