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Abstract

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A PROBLEM OF DIFFUSION WITH A MOVING BOUNDARY

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Let, at the boundary of a solid particle of radius ρ, a chemical interaction occur between the substance of the solid particle and the liquid solution so rapidly that the subsequent dissolution of the particle is determined only by the rate of diffusion of the newly formed particles into the depth of the liquid solution. We shall further assume that diffusion of the newly formed particles from each particle occurs in the volume bounded by a sphere of radius R. Then, at the boundary of the sphere,

$$\left.\frac{\partial C}{\partial r}\right|_{r=R} = 0. \tag{1}$$

From each particle of radius ρ, diffusion of the newly formed particles occurs according to Fick' s second law

$$\frac{\partial C}{\partial t} = D\Delta C. \tag{2}$$

For spherically symmetric diffusion we shall have the equation

$$\frac{\partial C}{\partial t} = D\left[\frac{\partial^2 C}{\partial r^2} + \frac{2}{r}\frac{\partial C}{\partial r}\right] \tag{3}$$

under the following initial and boundary conditions:

$$\begin{gathered} C(r,0) = 0, \qquad r > \rho_0; \\ C(\rho,t) = C_{\text{p}}; \\ \left.\frac{\partial C}{\partial r}\right|_{r=R} = 0, \end{gathered} \tag{4}$$

where C_{p} is the limiting concentration of newly formed particles. Introducing the new function

$$u = (C - C_{\text{p}})r \tag{5}$$

the conditions (4) are transformed into

$$\begin{gathered} u(r,0) = -C_{\text{p}} r, \qquad r > \rho_0; \\ u(\rho, t) = 0; \\ \left.\frac{\partial u}{\partial r}\right|_{r=R} = \frac{u(R,t)}{R}, \end{gathered} \tag{6}$$

and equation (3) into the simpler form:

$$\frac{\partial u}{\partial t} = D\frac{\partial^2 u}{\partial r^2}.$$

According to Fick' s first law, the amount of newly formed substance passing in time dt through the transverse section $4\pi\rho^2$ is equal to

$$dm = 4\pi\rho^2 D \left.\frac{\partial C}{\partial r}\right|_{r=\rho} dt. \tag{8}$$

But since the total mass of the unreacted substance is

$$m = \frac{4}{3}\pi\rho^3 C_0, \tag{9}$$

where C_0 is the concentration of the substance inside the particle, then, differentiating (9) with respect to time and substituting the resulting expression into (8), we find

$$\frac{d\rho}{dt} = \frac{D}{C_0}\left.\frac{\partial C}{\partial r}\right|_{r=\rho}. \tag{10}$$

The value C_0 is found from the condition of saturation of a sphere of radius R to the concentration C_{p}. Upon complete dissolution of the particles,

$$C_0 = \left(\frac{R}{\rho_0}\right)^3 C_{\text{p}}, \tag{11}$$

where ρ_0 is the initial radius of the particle.

Substituting $C(r,t)$ from (5) into equation (10), we obtain

$$\frac{d\rho}{dt} = \frac{D}{C_0\rho}\frac{\partial u(\rho,t)}{\partial r}. \tag{12}$$

The solution of equation (7) under the initial and boundary conditions (6) and the coupled equation (12) is somewhat analogous to the solution of the Stefan problem ([1]). In ([2]) a quite rigorous method for solving the Stefan problem was proposed. However, the first solution of the Stefan problem carried through to numerical values was given in ([3]).

As in ([3]), set

$$u(r,t) = u(R,t)\frac{r-\rho}{R-\rho} + \frac{2}{R-\rho}\sum_{n=1}^{\infty} B_n(t)\sin\frac{n\pi(r-\rho)}{R-\rho}. \tag{13}$$

Equation (13) satisfies the condition at the moving boundary. We now subject this equation to the condition at the boundary of the sphere of radius R, and from this find $u(R,t)$:

$$u(R,t) = -\frac{2\pi R}{\rho(R-\rho)}\sum_{n=1}^{\infty} B_n(t)n(-1)^n. \tag{14}$$

Now substitute (13) into (7), multiply (7) by $\sin\dfrac{k\pi(r-\rho)}{R-\rho}$, and integrate with respect to r from ρ to R:

$$\begin{aligned} B'_k(t) = &- B_k(t)\left[\frac{\rho'}{2(R-\rho)} + \frac{D\pi^2k^2}{(R-\rho)^2}\right] - \sum_{n=1}^{\infty} B_n(t)n\left\{\frac{2R\rho'(-1)^n}{\rho(R-\rho)k}\right. \\ &\left. - \frac{2R\rho'(R-2\rho)(-1)^{k+n}(R-\rho)}{[\rho(R-\rho)]^2}\right\} + \sum_{n\neq k}^{\infty} \frac{n\rho'\left[(k-n)(-1)^{k+n} + (k+n)(-1)^{k-n}\right]}{(n^2-k^2)(R-\rho)} B_n(t) \\ &+ \frac{2R}{k\rho}\sum_{n=1}^{\infty} B'_n(t)n(-1)^{1+k}. \end{aligned} \tag{15}$$

Adjoin to this equation equation (12):

$$\rho' = -\frac{D}{C_0}\frac{2\pi R}{[\rho(R-\rho)]^2}\sum_{n=1}^{\infty} B_n(t)n(-1)^n + \frac{2D\pi}{C_0\rho(R-\rho)^2}\sum_{n=1}^{\infty} B_n(t)n. \tag{16}$$

The solution of the system (15), (16) is obtained from the solution of the system of $(i+1)$ equations ([3])

$$B_k^{(i)\prime} = -B_k^{(i)}\left[\frac{\rho^{(i)\prime}}{2\left(R-\rho^{(i)}\right)} + \frac{D\pi^2k^2}{\left(R-\rho^{(i)}\right)^2}\right] - \sum_{n=1}^{i} B_n^{(i)} n \left\{\frac{2R^{(i)\prime}(-1)^n}{\rho^{(i)}\left(R-\rho^{(i)}\right)k}\right.$$
$$\left. - \frac{2R\rho^{(i)\prime}\left(R-2\rho^{(i)}\right)(-1)^{n+k}\left(R-\rho^{(i)}\right)}{\left[\rho^{(i)}\left(R-\rho^{(i)}\right)\right]^2}\right\}$$
$$+ \sum_{\substack{n=1\\ n\neq k}}^{i} \frac{(k-n)(-1)^{k+n}+(k+n)(-1)^{k-n}}{(k-n)} \frac{n\rho^{(i)\prime}B_n^{(i)}}{(n+k)\left(R-\rho^{(i)}\right)} + \frac{2R}{k\rho^{(i)}}\sum_{n=1}^{i} B_n^{(i)\prime} n(-1)^{n+k}; \tag{17}$$

$$\rho^{(i)\prime} = -\frac{D}{C_0}\frac{2\pi R}{\left[\rho^{(i)}\left(R-\rho^{(i)}\right)\right]^2}\sum_{n=1}^{i} B_n^{(i)} n(-1)^n + \frac{2D\pi}{C_0\rho^{(i)}\left(R-\rho^{(i)}\right)^2}\sum_{n=1}^{i} B_n^{(i)} n. \tag{18}$$

We compute the first approximation (17) taking into account the relation

$$B_n(0) = -\frac{C_{\mathrm{p}}\left(R-\rho_0\right)\rho_0}{n\pi}, \tag{19}$$

which is obtained from the initial condition, and the relation

$$\sum_{n=1}^{i} B_n^{(i)\prime}(0)n(-1)^n = \frac{\rho_0^{(i)\prime}\left(R-2\rho_0^{(i)}\right)}{\rho_0^{(i)}\left(R-\rho_0^{(i)}\right)}\sum_{n=1}^{i} B_n^{(i)}(0)n(-1)^n, \tag{20}$$

which is obtained from $u'(R,0)=0$. This condition is valid, since $C'(R,t')=0$ for

$$t' \leqslant \frac{(R-\rho_0)^2}{2D}. \tag{21}$$

Relation (21) determines the time during which a particle participating in Brownian motion reaches the edge of the sphere of radius R (with equality).

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Note: Figure translations are in progress. See original paper for figures.