

Lifetime of a soluble solid particle in a stagnant medium: approximate analytical modelling involving fractional (half-time) derivatives

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Approximate analytical solutions concerning lifetime of soluble solid particles in an unbounded stagnant medium have been developed by simple application of fractional half-time derivative in the Riemann-Liouville sense to express the relationship between the net surface mass flux and the concentration at the interface. The solutions start with the initial formulation of Rice and Do on the time-depletion of the radius of a spherical particle expressed through terms including the solubility parameter as the only key parameter controlling the process of dissolution. The two approximate developed solutions use different scaling and dimensionless variables: The 1st solution is developed by an introduction of a similarity variable $\eta = R/\sqrt{Dt}$ while the 2nd solution applies the classical scaling using the initial sphere radius as a length scale that leads to dimensionless radius $r = R/R_0$ and time $\tau = Dt/R_0^2$. Both solutions provide approximate relationships close to that of Rice and Do.

Keywords: particle dissolution, analytical model, fractional half-time derivative.

INTRODUCTION

A Particle dissolution is an old problem¹ related to drug dissolution^{2, 3}, mineral leaching⁴, chemical processing^{5, 6, 7}, controlled drug release^{8, 9, 10}, etc. It is still under intense investigation^{3, 11, 12, 13, 14, 15}. The solution to diffusion-dependent and moving boundary problems¹⁶ is not an easy task and both numerical¹⁷ and analytical approaches have been employed¹¹. The analytical modeling of solid dissolution^{11, 18} is an attractive area because the final relationships of the solutions are in closed forms allowing easily estimating relationships between the key parameters of the process. The modeling approaches employ perturbation solutions^{19, 20}, the integral balance method²¹, heat transfer analogy²² etc.

The present report refers to the calculation of the lifetime of solid particles in stagnant unbounded media, a problem forever young since the time of Noyes and Whitney¹. The lifetime²³ is important to many clinical investigations of drugs and industrial processes of dissolving species or minerals. In most cases the concentration field in the surrounding fluid is not of primary interest but only the time of complete dissolution should be estimated. Rice and Do²⁴ developed an elegant analytical solution for a single sphere expressed through the term of solids solubility factor. This solution was the starting point of the present approximate solutions using initial formulation of Rice and Do²⁴ and expressing the particle lifetime in terms involving the solubility factor as a process parameter.

PROBLEM STATEMENT

Governing equations

Consider a spherical particle of radius R changing its radius due to dissolution in a stagnant liquid by simple diffusion alone

$$\rho \left(\pi \frac{4}{3} \right) \frac{d(R^3)}{dt} = -4\pi R^2(t) N_{r=R} \quad R(t=0) = R_0 \quad (1a)$$

The net flux of mass to the surrounding medium is

$$N_{r=R} = - \frac{D}{1-C^*/\rho_0} \frac{\partial C}{\partial r} \Big|_{r=R} \quad (1b)$$

while the concentration field in the stagnant liquid is described by

$$D \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial C}{\partial r} \right) = \frac{\partial C}{\partial t}, \quad R \leq r \leq \infty \quad (2a)$$

$$C = C^*, \quad r = R \quad (2b)$$

$$C \rightarrow C_0, \quad r \rightarrow \infty \quad (2c)$$

$$C = C_0, \quad t = 0, \quad r > R \quad (2d)$$

This problem has been solved by Rice and Do²⁴ by developing the solution of the model (2) and then expressing $\partial C/\partial r$ at $r = R$ in order to define $N_{r=R}$. The present work refers to the potential of the fractional calculus^{25, 26, 27} to avoid the entire domain solutions in diffusion problems because the relationship between the function value and its derivative can be established at the interface of the body and the surrounding medium by the Riemann-Liouville fractional derivative. The approach has been successfully applied for estimating heat resistances of contacting bodies²⁴ by means of semi-derivatives and semi-integrals.

Hence, the present note refers to a new approach in analytical solution of problems relevant to dissolution of solids in unbounded and understated medium. With this approach, the determination of the concentration field outside the particle boundary is unnecessary and the surface concentration gradient should only be known.

Solution strategy

The classical solution of the problem addresses two consequent steps:

1. Determination of the surface mass flux (1b) through the solution of the problem (2a–2d), which is a classical semi-infinite diffusion problem^{11, 29}.

2. Determination of the radius of the sphere undergoing dissolution as a function of the time through (1a). The first step needs determination of the gradient $(\partial C/\partial r)_{r=R}$ in the solid-liquid interface through the solution of the problem (2a–2d) for the liquid phase. Then, the ordi-

nary differential equation (1a) concerning $R(t)$ has to be solved as a second step.

The present approach avoids development of the concentration distribution in the bulk phase (step 2) as it was done by Rice and Do²⁴. It directly links the surface concentration and the gradient $(\partial C/\partial r)_{r=R}$ through the Riemann-Liouville fractional derivative. Moreover, the solution concerning the sphere radius (1a) defining the time of complete dissolution is solved in terms of a similarity variable $\eta = R/\sqrt{Dt}$ because the semi-infinite diffusion problem in the liquid phase (2a-d) has no definite length scale^{11, 29}.

PROBLEM SOLUTION

Surface flux: a solution by fractional calculus

Classically, by the transforms, $\theta = C - C^*$ and $u = \theta r$, we get

$$\frac{\partial u}{\partial t} = D \frac{\partial u^2}{\partial r} \tag{3a}$$

The transformation to the variable θ leads to zero initial conditions that allows to apply the Riemann-Liouville fractional semi-derivatives after splitting (3a) as²⁵

$$\frac{\partial^{1/2} u}{\partial t^{1/2}} = -\sqrt{D} \frac{\partial u}{\partial r} \tag{3b}$$

Here

$$\frac{\partial^{1/2} u}{\partial t^{1/2}} = \frac{1}{\Gamma(1/2)} \frac{d}{dt} \int_0^t \frac{u_{r=R}}{\sqrt{u-\tau}} dt \tag{3c}$$

is a half-time fractional derivative in the Riemann-Liouville sense [25], and $\Gamma(\bullet)$ is the Gamma function, where $\Gamma(1/2) = \sqrt{\pi}$.

At $r = R$ we have from (3c):

$$-\frac{\partial u}{\partial r} \Big|_{r=R} = \frac{1}{\sqrt{D}} D_t^{1/2} u \Big|_{r=R} - \frac{u_0}{\sqrt{\pi t}} \tag{4a}$$

Since, $u \Big|_{r=R} = R\theta^* = R(C - C^*) \Big|_{C=C^*} = 0$, we get from (4a):

$$-\frac{\partial u}{\partial r} \Big|_{r=R} = \frac{1}{\sqrt{D\sqrt{\pi t}}} R(\theta_0 - \theta^*) = \frac{1}{\sqrt{D\sqrt{\pi t}}} R(C_0 - C^*), \theta^* = 0 \tag{4b}$$

With $u^* = \theta^* R$, $u_0 = \theta_0 R$ the boundary flux $(\partial C/\partial r)_{r=R} = N_{r=R}$ is given by

$$-(\partial u/\partial r)_{r=R} = \frac{1}{\sqrt{D}} \frac{\partial^{1/2} u}{\partial t^{1/2}} - \frac{u_0}{\sqrt{\pi t}} = \frac{1}{\sqrt{D}} \frac{(u^* - u_0)}{\sqrt{\pi t}} \tag{4c}$$

On the other hand, we have $\theta = C - C^*$ and $u = r\theta$, as well as

$$\frac{\partial u}{\partial r} = \theta + r \frac{\partial \theta}{\partial r} \tag{5a}$$

$$\frac{\partial u}{\partial r} \Big|_{r=R} = R \frac{\partial C}{\partial r} \Big|_{r=R} - (C_0 - C^*) \tag{5b}$$

Equating the right-hand sides of (5a) and (5b) we get

$$-\frac{\partial C}{\partial r} \Big|_{r=R} = -\frac{(C^* - C_0)}{R} \left(1 + \frac{R}{\sqrt{\pi} \sqrt{Dt}} \right) \tag{6a}$$

Now with (6a) and (1b) we read

$$N_{r=R} = -\frac{D(C^* - C_0)}{R(1 - C^*/\rho_0)} \left(1 + \frac{R}{\sqrt{\pi} \sqrt{Dt}} \right) \tag{6b}$$

Expressing $(\partial C/\partial r)_{r=R}$ from (4a) and by the transforms $\theta = C - C^*$ and $u = r\theta$ we get

$$N_{r=R} = \frac{D}{R} S \left[1 + \frac{1}{\sqrt{\pi}} \frac{R}{\sqrt{Dt}} \right], S = \frac{C^* - C_0}{1 - C^*/\rho_0} \tag{7a, 7b}$$

The expression (7a) is the same as that developed by Rice and Do²⁴. However, the development of the concentration profile in the bulk phase was avoided thanks to the fractional half-time derivative (3c). Moreover, the transform $\theta = C - C^*$ zero initial conditions $u_0 = 0$ permits the Riemann-Liouville fractional derivative to be used to relate the flux and the function value at solid-liquid interface. In the next section we develop two approximate solutions starting from the result (7a, 7b) and the balance equation (1a) but with different scaling and formation of the defenseless variables.

Particle dissolution (Change of particle radius in time)

Solution - 1

Now, with the expression (7a) the mass balance equation (1a) reads as

$$R \frac{dR}{dt} = -DS \left[1 + \frac{1}{\sqrt{\pi}} \frac{R}{\sqrt{Dt}} \right] \tag{8}$$

Since in the dissolution process the particle size is time-varying and the process has no characteristic length scale we introduce a similarity variable $\eta = R/\sqrt{Dt}$ which is a crucial step differing from the approach used by Rice and Do²⁴. Now, with the new variable we get

$$\frac{d\eta^2}{dt} + \frac{\eta^2}{t} = -\frac{2S}{t} - \frac{1}{t} \frac{S}{\sqrt{\pi}} \eta \tag{9a}$$

With the substitution $\eta^2 = y$ we get

$$\frac{dy}{dt} + \frac{y}{t} = -\frac{2S}{t} - \frac{1}{t} \frac{S}{\sqrt{\pi}} \sqrt{y} \tag{9b}$$

Re-arranging (9b) with $\sqrt{y} = z$ have

$$2 \frac{d(\sqrt{y})}{dt} + \frac{\sqrt{y}}{t} = -\frac{1}{t} \frac{A}{\sqrt{y}} - \frac{B}{t}, A = 2S, B = 2S/\sqrt{\pi} \tag{10a}$$

or more conveniently as a separable equation

$$2 \frac{dz}{dt} = -\frac{1}{t} \left(z - \frac{A}{z} - B \right) \tag{10b}$$

The solution of (10b) (Performed by Maple) is

$$z + \frac{B}{2} \ln(-z^2 + Bz + A) + \frac{(2A+B^2)}{\sqrt{4A+B^2}} \arctan h \left(\frac{-2z+B}{\sqrt{4A+B^2}} \right) = -\ln t \tag{11a}$$

Expressing the time t explicitly from (11a) we get

$$t = \frac{1}{\exp \left[z + \frac{B}{2} \ln(-z^2 + Bz + A) + \frac{(2A+B^2)}{\sqrt{4A+B^2}} \arctan h \left(\frac{-2z+B}{\sqrt{4A+B^2}} \right) \right]} \tag{11b}$$

At the point of full particle dissolution when $R = 0$ we have from the similarity variable $\eta = R/\sqrt{Dt} = z = 0$. Hence the time of complete dissolution t_f is

$$t_{f_1} = \frac{1}{\exp \left[\frac{B}{2} \ln(A) + \frac{(2A+B^2)}{\sqrt{4A+B^2}} \arctan h \left(\frac{B}{\sqrt{4A+B^2}} \right) \right]} \tag{12a}$$

$$A = 2S, B = 2S/\sqrt{\pi} \tag{12b, 12c}$$

Therefore, the time for completing the dissolution depends only on solubility S expressed in two terms $A = 2S, B = 2S/\sqrt{\pi}$. Both of them are of equal order

of magnitude because the solubility factor is a small value with order of magnitude 10^{-3} ²⁴. In the repeating terms $2A + B^2$ and $4A + B^2$ in (12a) we may neglect B^2 because its order of magnitude is 10^{-6} . However, this approximation can yield some errors, and to avoid them, the expression (12a) can be transformed into

$$t_{f1} = \frac{1}{\exp\left[\frac{B}{2}\ln(A) + B\frac{\left(\frac{2A}{B^2}\right)}{\sqrt{4\frac{A}{B^2}+1}}\arctan h\left(\frac{1}{\sqrt{4\frac{A}{B^2}+1}}\right)\right]} \quad (13a)$$

Now, we have the order of magnitude $\frac{A}{B^2} = \frac{\pi}{2S} O(10^3)$

Now, we may derive some approximate expressions about t_{f1} through (13a) namely

$$t_f = \frac{1}{\exp\left[\frac{S}{\sqrt{\pi}}\ln(2S) + \frac{\left(8S + \frac{4S^2}{\pi}\right)}{\sqrt{8S + \frac{4S^2}{\pi}}}\arctan h\left(\frac{2S}{\sqrt{\pi}}\right)\right]} \quad (13b)$$

and assuming $\frac{4S^2}{\pi} \leq 8S$ we get

$$t_{f1} \approx \frac{1}{\exp\left[\frac{S}{\sqrt{\pi}}\ln(2S) + \sqrt{8S}\arctan h\left(\frac{\sqrt{S}}{\sqrt{2\pi}}\right)\right]} \quad (13c)$$

or

$$t_{f1} \approx \frac{1}{\exp\left[\frac{S}{\sqrt{\pi}}\ln(2S)\right]} \quad (13d)$$

Solution - 2

The Solution -1 was developed by introduction of a similarity variable $\eta = R/\sqrt{Dt}$ since we followed the classics in diffusion problems²⁹ through a semi-infinite medium. However, since the particle radius change in time is an initial value problem with $R = R_0(t = 0)$ an alternative initial scaling of eq. (8) by introducing characteristic time and length scales²⁴ can be applied, namely $r = R/R_0$ and $t_0 = R_0^2/D$, $\tau = t/t_0$ (14a, 14b, 14c)

This leads to a dimensionless form of eq. (8), namely

$$R\frac{dR}{dt} = -DS\left[1 + \frac{1}{\sqrt{\pi}}\frac{R}{\sqrt{Dt}}\right] \Rightarrow \left(\frac{R_0^2}{t_0}\right)r\frac{dr}{d\tau} = -DS\left[1 + \frac{1}{\sqrt{\pi}}\frac{R_0}{\sqrt{t_0}}\frac{r}{\sqrt{D\tau}}\right] \quad (15)$$

Using (14a, 14b, 14c) we finally read

$$\frac{dr^2}{d\tau} = -2S\left[1 + \frac{1}{\sqrt{\pi}}\frac{r}{\sqrt{\tau}}\right] \quad (16)$$

Introducing the variable $p = r^2$ we get

$$\frac{dp}{d\tau} = -2S\left[1 + \frac{1}{\sqrt{\pi}}\sqrt{\frac{p}{\tau}}\right] \quad (17)$$

The solution of (17) using Maple gives

$$\tau - \frac{1}{2}\left(\frac{\pi\ln(\pi - p(t))}{S}\right) - \frac{\sqrt{\pi}}{S}\sqrt{p(t)} + \frac{\pi\operatorname{arctanh}\left(\sqrt{\frac{p(t)}{\pi}}\right)}{S} + C_1 = 0 \quad (18)$$

Taking into account that at $t = 0$ we have $R = R_0$, that is equivalent to $\tau = 0$ and $r = 1$, as well as $p(t = 0) = 1$, we have for the constant C_1

$$C_1 = \frac{1}{S}\left[\frac{\pi\ln(\pi - 1)}{2} - \sqrt{\pi} + \pi\operatorname{arctanh}\left(\sqrt{\frac{1}{\pi}}\right)\right] \quad (19)$$

Thus, we have

$$\tau - \left(\frac{\pi\ln(\pi - p(\tau))}{2S}\right) - \frac{\sqrt{\pi}}{S}\sqrt{p(\tau)} + \frac{\pi\operatorname{arctanh}\left(\sqrt{\frac{p(\tau)}{\pi}}\right)}{S} +$$

$$+ \frac{1}{S}\left[\frac{\pi\ln(\pi - 1)}{2} - \sqrt{\pi} + \pi\operatorname{arctanh}\left(\sqrt{\frac{1}{\pi}}\right)\right] = 0$$

The final dissolution time t_{f2} at $p(\tau) = 0$ (with $\tau = t/t_0 \Rightarrow t = \tau (R_0^2/D)$) is

$$t_{f2} \approx \frac{t_0}{S}\left[\frac{\pi}{2}\ln\frac{(\pi - 1)}{\pi} - \sqrt{\pi} + \pi\operatorname{arctanh}\sqrt{\frac{1}{\pi}}\right] \Rightarrow t_{f2} \approx \frac{0.367}{S}t_0 \quad (21)$$

SOME COMMENTS

The approximate analytical solutions developed in the previous sections resulted in

$$t_{f1} \approx \frac{1}{\exp\left[\frac{S}{\sqrt{\pi}}\ln(2S) + \sqrt{8S}\arctan h\left(\frac{\sqrt{S}}{\sqrt{2\pi}}\right)\right]}$$

or

$$t_{f1} \approx \frac{1}{\exp\left[\frac{S}{\sqrt{\pi}}\ln(2S)\right]}$$

and

$$t_{f2} \approx \frac{1}{S}\left[\frac{\pi}{2}\ln\frac{(\pi - 1)}{\pi} - \sqrt{\pi} + \pi\operatorname{arctanh}\sqrt{\frac{1}{\pi}}\right] \Rightarrow t_{f2} \approx \frac{0.367}{S}t_0$$

These solutions are comparable to the solution of Rice and Do²⁴

$$\tau_f = \frac{1}{2S}\exp\left\{\left(-\frac{2S}{\sqrt{2\pi S - S^2}}\right)\left[\frac{\pi}{2} - \arctan\left(\frac{S}{\sqrt{2\pi S - S^2}}\right)\right]\right\} \quad (22)$$

For example, the approximate solution of Rice and Do [24] yields $t_{f2} \approx \frac{0.5}{S}t_0$ close to (21).

CONCLUSIONS

The present work demonstrates how the fractional half-time derivative in the Riemann-Liouville sense can be applied for direct derivation of the equation governing the time-dependent radius of dissolving solid sphere in an unbounded stagnant medium. The final outcomes are approximate relationships for the time-to-complete dissolution, i.e. the lifetime of the dissolving solids.

The two solutions developed are expressed in terms including the solubility factor starting from the formulation of Rice and Do²⁴. The 1st solution is developed by an introduction of a similarity variable $\eta = R/\sqrt{Dt}$ while the 2nd solution applies the classical scaling using

the initial sphere radius as a length scale that leads to the dimensionless radius $r = R/R_0$ and time $\tau = Dt/R_0^2$.

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