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Variable Transformations for Mathematical Model of Drug Release from a Swelling Hydrogel

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ABSTRACT

The aim of this study is to develop two mathematical models with nonhomogeneous moving boundary condition. The main concept behind the solution is to separate the mathematical model based on the region where a particular release mechanism takes place. The first region represents the swelling-controlled process; while the second represents the diffusion-controlled process. Variable transformation has had a great impact on this study. The advection term in the advection-diffusion equation was removed using Landau transformation in the swelling-controlled model. In the diffusion-controlled model, the moving boundary condition was transformed to a fixed boundary condition. Finally, the nonhomogeneous moving boundary condition was reduced to the homogeneous boundary condition using a steady-state solution in both models.

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INTRODUCTION

The development of an analytical method of solution of diffusion and advection-diffusion equation processes is critical for the analysis of phenomena occurring in numerous fields, including bio-medical science, material science, engineering and pharmaceutical problems. The term advection-diffusion equation refers to the solute transport due to the combined effect of diffusion and convection within a medium (Kumar, A., 2009). The main motivation for this study is the swelling and diffusion of hydrogel in a finite volume liquid (Bierbrauer, F., 2005; Wang, S., X. Lou, 2009).

However, the moving boundary problem was taken into consideration in this study. This is a particular kind of boundary value problem for partial differential equations (PDEs), and is adapted to the case in which a phase boundary can move with time. It occurs in numerous physical applications involving diffusion, including: heat transfer, where a phase transition occurs; moisture transport such as swelling grains or polymers; and deformable porous media problems, where the solid displacement is governed by diffusion (Barry, S.I., J. Counce, 2008). Moving boundary value problems have also been considered in the body of literature on polymer swelling, with particular relevance to drug delivery systems. In these systems, the boundary may move due to a

swelling and dissolution process. For an overall summary of this, refer to (Kanjickal, D.G., S.T. Lopina, 2004; Siepmann, J., A. Gopferich, 2001).

We present a mathematical model for the changing of the hydrogel in the finite volume liquid, which takes into account, not only a moving boundary for the swelling controlled, but also for diffusion controlled, which is the second moving boundary. The motion of the former boundary is governed by a swelling controlled process; while the motion of the latter boundary is governed by a diffusion controlled process.

Mathematical models for diffusion-controlled drug release have been developed in prior studies (Doumenc, F., B. Guerrier, 2001; Colin, R., 1998; Lu, S., 1998). Fick's law has commonly been used as the base equation for models with suitable initial and boundary conditions. The analytical solution for the diffusion-controlled drug release models for regular geometries (MohdMahali, S., 2011; Wang, S., 2009; Wang, S., X. Lou, 2009) has been derived in some ideal conditions. The diffusion-controlled model with a simplified geometry that has been analytically solved was proven to be comparable with the numerical solution of the same model, with 3D geometry. Despite the availability of the analytical solution for diffusion-controlled drug delivery in various conditions, the analytical solution for drug delivery involving a swelling effect remains

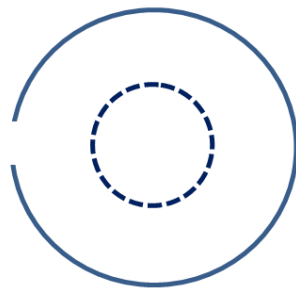
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restricted. Various mathematical theories have been proposed in developing models for drug deliveries involving swelling devices.

Mathematical Model:

Drug delivery scientists encounter challenges in developing safe and effective oral delivery methods for therapeutic proteins. Hydrogels were investigated to overcome this problem for their potential use as an oral delivery system for protein. Hydrogels are 3D polymeric networks that imbibe a large volume of water, while remaining insoluble, due to the physical or chemical cross-linking of individual polymer chains [5,9]. Stimuli-responsive hydrogels undergo dramatic changes in swelling and network structure in response to environment stimuli such as pH, temperature, ionic strength, enzymes and light.

We consider a device of cylindrical geometry with height h_d loaded with a certain amount of drug. This device is placed in a cylindrical container with height h_c filled with unstirred liquid (Note that $h_d = h_c$). An assumption is made that the expected diffusion is in an axial direction. The cross-section of the set-up is shown in Figs. 1 and 2.



0: centre of hydrogel
 $X(t)$: boundary of hydrogel
 X_c : boundary of container

Fig. 1: The 2D geometry of a disc device in a cylindrical container.

We first consider the case of 2D disc geometry. However, for simplicity, we consider a one dimensional space where $x = 0$ refers to the centre of the hydrogel; $x = X(t)$ refers to the boundary of the hydrogel moving with respect to time; and $x = X_c$ shows the boundary of the container.



Fig. 2: Geometry of disc device in the form of 1D.

Mathematical Formulation:

In order to develop a suitable mathematical model for this problem, we consider two regions: region 1 and region 2. Region 1 refers to the region inside the hydrogel device ($0 < x < X(t)$); while region 2 refers to the region between the

hydrogel surface to the boundary of the container ($X(t) < t < X_c$). In region 1, we assume the release mechanism is swelling-controlled. On the other hand, the release mechanism is assumed to be diffusion-controlled in region 2.

Swelling-controlled model:

In region 1, the drug diffuses out of the domain at the boundaries, and may swell as fluids are absorbed. The situation can be expressed in terms of an advection-diffusion equation, as follows:

$$\frac{\partial c_1}{\partial t} = D_1 \frac{\partial^2 c_1}{\partial x^2} - \frac{\partial c_1}{\partial x} u - c_1 \frac{\partial u}{\partial x} \quad 0 < x < X(t), t > 0 \quad (1)$$

$$\frac{\partial c_1}{\partial x}(0, t) = 0 \quad (2)$$

$$c_1(X(t), t) = c_2(X(t), t) \quad (3)$$

$$c_1(x, 0) = 1, \quad 0 < x < X(t) \quad (4)$$

where $c_1(x, t)$ is the concentration inside the hydrogel with D_1 as the constant diffusion coefficient, $\frac{\partial c_1}{\partial t}$ is the local rate of change of concentration over time, $\frac{\partial^2 c_1}{\partial x^2}$ represents the diffusion of the concentration, $\frac{\partial c_1}{\partial x} u$ is the dilution term, and $c_1 \frac{\partial u}{\partial x}$ is due to local volume change. For the initial condition, the concentration is uniform in the device, and zero in the liquid.

Diffusion-controlled model:

In region 2, the release mechanism is governed by the following equation:

$$\frac{\partial c_2}{\partial t} = D_2 \frac{\partial^2 c_2}{\partial x^2} \quad X(t) < x < X_c, t > 0 \quad (5)$$

$$c_2(X(t), t) = c_1(X(t), t) \quad (6)$$

$$\frac{\partial c_2}{\partial x}(X_c, t) = 0 \quad (7)$$

$$c_2(x, 0) = 0, \quad X(t) < x < X_c \quad (8)$$

where $c_2(x, t)$ is the concentration outside the hydrogel with D_2 as the constant diffusion coefficient.

Methodology:

Variable Transformation:

For the mathematical model in region 1, a Landau transformation will be used in order to remove the advection term. However, for this particular mathematical model, this transformation only takes place after the steady state solution (which will be introduced in the next section of this paper) is applied to this model. The Landau transformation is defined by:

$$\zeta = \frac{x}{X(t)}, \quad \tau = t$$

According to Bierbrauer, when the Landau transformation is applied, Equation (1) then becomes:

$$\frac{\partial c}{\partial \tau} = \frac{D}{X^2} \frac{\partial^2 c}{\partial \zeta^2} - \frac{\dot{X}}{X} c, \quad 0 < \zeta < 1, \tau > 0. \quad (9)$$

For region 2, the domain for the mathematical model will be changed by using the following transformation:

$$\bar{x} = \frac{x - X(t)}{X_c - X(t)}, \quad \tau = t.$$

The new domain is now $0 < \bar{x} < 1, \tau > 0$. By using chain rule, we have

$$\begin{aligned} \frac{\partial c_2}{\partial t} &= \frac{\partial c_2}{\partial \bar{x}} \frac{\partial \bar{x}}{\partial X} \frac{\partial X}{\partial t} + \frac{\partial c_2}{\partial \tau} \frac{\partial \tau}{\partial t} \\ &= \frac{\partial c_2}{\partial \bar{x}} \frac{\dot{X}}{(X_c - X(t))} + \frac{\partial c_2}{\partial \tau} \end{aligned}$$

where $\dot{X} = \frac{\partial X}{\partial t}$

$$\frac{\partial c_2}{\partial x} = \frac{\partial c_2}{\partial \bar{x}} \frac{\partial \bar{x}}{\partial x} = \frac{\partial c_2}{\partial \bar{x}} \frac{1}{(X_c - X(t))}$$

$$\begin{aligned} \frac{\partial^2 c_2}{\partial x^2} &= \frac{\partial}{\partial x} \left(\frac{\partial c_2}{\partial \bar{x}} \right) \\ &= \frac{\partial}{\partial \bar{x}} \left(\frac{\partial c_2}{\partial \bar{x}} \frac{\partial \bar{x}}{\partial x} \right) \\ &= \frac{1}{\partial x \partial \bar{x} (X_c - X(t))} \frac{\partial^2 c_2}{\partial \bar{x}^2} \\ &= \frac{1}{(X_c - X(t))^2} \frac{\partial^2 c_2}{\partial \bar{x}^2} \end{aligned}$$

This equation is then been substituted to Equation 5 to have

$$\frac{\partial c_2}{\partial \tau} = D_2 \frac{1}{(X_c - X(t))^2} \frac{\partial^2 c_2}{\partial \bar{x}^2} - \frac{\dot{X}}{(X_c - X(t))} \frac{\partial c_2}{\partial \bar{x}}$$

Whereas, the no flux boundary condition (7) becomes $\frac{1}{(X_c - X(t))} \frac{\partial c_2}{\partial \bar{x}}(1, \tau) = 0$. Since $\frac{1}{(X_c - X(t))} \neq 0$, we can simply put the boundary condition as $\frac{\partial c_2}{\partial \bar{x}}(1, \tau) = 0$.

Then, Equations 5-8 are changed into a new system of equations, as follows:

$$\frac{\partial c_2}{\partial \tau} = D_2 \frac{1}{(X_c - X(t))^2} \frac{\partial^2 c_2}{\partial \bar{x}^2} - \frac{\dot{X}}{X_c - X(t)} \frac{\partial c_2}{\partial \bar{x}} \quad 0 < \bar{x} < 1, \tau > 0 \quad (10)$$

$$c_2(0, \tau) = c_1(0, \tau) \quad (11)$$

$$\frac{\partial c_2}{\partial \bar{x}}(1, \tau) = 0 \quad (12)$$

$$c_2(\bar{x}, 0) = 0, \quad 0 < \bar{x} < 1. \quad (13)$$

Steady-state solution:

Most of the mathematical models related to the drug release mechanism in previous studies come with a homogeneous boundary condition. However, in the present research, we encounter the nonhomogeneous moving boundary condition in the mathematical models. The steady state solution is applied to both the swelling-controlled model and the diffusion controlled model in order to transform the nonhomogeneous moving boundary condition to a homogeneous boundary condition. In this solution, we first assume that it will be written as a combination of steady state and transient solution.

$$c(x, t) = v(x) + w(x, t)$$

where $v(x)$ is the steady state solution which is independent of t , and $w(x, t)$ is the transient solution

which varies with t . We note the fact that it is a function of x alone, yet it must satisfy the heat equation. Since $v_{xx} = v''$ and $v_t = 0$, substituting into the heat equation gives $a^2 v_{xx} = 0$. After dividing both sides by a^2 and integrating twice with respect to x , we found that $v(x) = Ax + B$.

We then rewrite the boundary condition in terms of v : $u(0, t) = v(0) = T_1$, and $u(L, t) = v(L) = T_2$. The two conditions are applied to obtain the following:

$$\begin{aligned} v(0) = T_1 &= A(0) + B = B && \rightarrow B = T_1 \\ v(L) = T_2 &= AL + B = AL + T_1 && \rightarrow A = \frac{T_2 - T_1}{L} \end{aligned}$$

Therefore, the steady state solution is $v(x) = \frac{T_2 - T_1}{L}x + T_1$.

We can then set aside the steady state solution and proceed to find the transient solution $w(x, t)$. First, we rewrite the initial-boundary value problem. In order to do so, we subtract out $v(x)$ from the initial and boundary values. Since $c(x, t) = v(x) + w(x, t)$, the results will be the conditions that the transient solution $w(x, t)$ alone must satisfy.

Changing the boundary conditions results in:

$$\begin{aligned} u(0, t) &= T_1 = v(0) + w(0, t) \\ &\rightarrow w(0, t) \\ &= T_1 - v(0) \\ &= 0 \\ u(L, t) &= T_2 = v(L) + w(L, t) \\ &\rightarrow w(L, t) \\ &= T_2 - v(L) = 0 \end{aligned}$$

Changing the initial condition results in:

$$\begin{aligned} u(x, 0) &= f(x) = v(x) + w(x, 0) \\ \rightarrow w(x, 0) &= f(x) - v(x) \end{aligned}$$

RESULTS AND DISCUSSION

We know that from steady state solution, the solution can be expressed as $v(x) = A(x) + B$, and its derivative would be $v'(x) = A$. From the boundary conditions (2) and (3), we have:

$$\begin{aligned} \frac{\partial c_1}{\partial x}(0, t) &= v'(0) = 0 \\ c_1(X(t), t) &= v(X(t)) = c_2(X(t), t) \\ v'(0) &= 0 = A \\ v(X(t)) &= A(X(t)) + B = c_2(X(t), t) \end{aligned}$$

Therefore, our steady state solution is:

$$v(x) = 0x + c_2(X(t), t).$$

Since $c(x, t) = v(x) + w(x, t)$, we may rewrite the boundary condition as follows:

$$\begin{aligned} \frac{\partial c_1}{\partial x}(0, t) &= 0 = v'(0) + w_1'(0, t) \\ c_1(X(t), t) &= c_2(X(t), t) = v(X(t)) + w_1(X(t), t) \end{aligned}$$

Then the new boundary conditions are obtained:

$$w_1'(0, t) = 0 - v'(0) = 0$$

$$w_1(X(t), t) = c_2(X(t), t) - v(X(t)) = 0$$

After that, the new initial condition is changed by subtracting the steady state solution $v(x)$ from the original value of initial condition.

$$c_1(x, 0) = 1 = v(x) + w_1(x, 0)$$

$$w_1(x, 0) = 1 - c_2(X(t), t).$$

Therefore, equations(1)-(4), are transformed to the new system of equations:

$$\frac{\partial w_1}{\partial t} = D_1 \frac{\partial^2 w_1}{\partial x^2} - \frac{\partial w_1}{\partial x} u - w_1 \frac{\partial u}{\partial x} \quad (14)$$

$$0 < x < X(t), \quad t > 0$$

$$\frac{\partial w_1}{\partial x}(0, t) = 0 \quad (15)$$

$$w_1(X(t), t) = 0 \quad (16)$$

$$w_1(x, 0) = 1 - c_2(X(t), t), \quad (17)$$

$$0 < x < X(t)$$

By using Landau transformation, the advection term is removed so that the system (14)-(17) is now,

$$\frac{\partial w_1}{\partial \tau} = D \frac{\partial^2 w_1}{\partial \zeta^2} - \frac{\dot{X}}{X} \frac{\partial w_1}{\partial \zeta}, \quad 0 < \zeta < 1, \quad t > 0 \quad (18)$$

$$\frac{\partial w_1}{\partial \zeta}(0, \tau) = 0 \quad (19)$$

$$w_1(1, \tau) = 0 \quad (20)$$

$$w_1(\zeta, 0) = 1 - c_2(1, \tau), \quad 0 < \zeta < 1 \quad (21)$$

After obtaining the homogenous boundary condition, we proceed to find the analytical solution for this problem. We may refer and compare the analytical solution to previous studies. In Bierbrauer [3], the boundary concentration is assumed to be a sink condition, to obtain the following:

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2} - c \frac{\partial u}{\partial x} - u \frac{\partial c}{\partial x} \quad 0 < x < X(t), \quad t > 0 \quad (22)$$

$$c(x, 0) = 1 \quad 0 < x < X(0) \quad (23)$$

$$X(0) = L \quad (24)$$

$$\frac{\partial c}{\partial x}(0, t) = 0 \quad t > 0 \quad (25)$$

$$c(X(t), t) = 0$$

with the final solution expressed as:

$$c(x, t) = \frac{4}{\pi} \sum_{n=0}^{\infty} \frac{(-1)^n L}{(2n+1)X(t)} \cos\left(\frac{(2n+1)\pi x}{2X(t)}\right) e^{-D\left(\frac{(2n+1)\pi}{2}\right)^2 \int_0^t X(t')^{-2} dt'}$$

For a diffusion-controlled model, the same steady state method is applied to equation (10)-(13) to reduce the nonhomogeneous moving boundary condition to homogeneous boundary condition. Since $v(x) = A(x) + B$ and $v'(x) = A$, we would have the following:

$$c_2(0, \tau) = v(0) = c_1(0, \tau)$$

$$\frac{\partial c_2}{\partial \bar{x}}(1, \tau) = v'(1) = 0$$

The following equation gives us the value of A and B.

$$v(0) = c_1(0, \tau) = A(0) + B$$

$$v'(1) = 0 = A$$

where $A = 0$ and $B = c_1(0, \tau)$.

Hence, the steady state solution would be:

$$v(x) = 0x + c_1(0, \tau).$$

Before we proceed to find the transient part $w(x, t)$, we first need to rewrite the boundary condition.

$$c_2(0, \tau) = c_1(0, \tau) = v(0) + w_2(0, \tau)$$

$$\frac{\partial c_2}{\partial \bar{x}}(1, \tau) = 0 = v'(1) + w_2'(1, \tau)$$

The new boundary condition are obtained after $v(x)$ is subtracted.

$$w_2(0, \tau) = c_1(0, \tau) - v(0) = 0$$

$$w_2'(1, \tau) = 0 - v'(1) = 0.$$

The new initial condition is obtained by applying the same step. $v(x)$ is subtracted from the original value of initial condition.

$$c_2(\bar{x}, 0) = 0 = v(x) + w_2(\bar{x}, 0)$$

$$w_2(\bar{x}, 0) = 0 - c_1(0, \tau).$$

Hence, the new system of equation for diffusion-controlled model would be as the following:

$$\frac{\partial w_2}{\partial \tau} = D_2 \frac{1}{(X_c - X(t))^2} \frac{\partial^2 w_2}{\partial \bar{x}^2} - \frac{\dot{X}}{X_c - X(t)} \frac{\partial w_2}{\partial \bar{x}} \quad (26)$$

$$0 < \bar{x} < 1, \quad \tau > 0$$

$$w_2(0, \tau) = 0 \quad (27)$$

$$\frac{\partial w_2}{\partial \bar{x}}(1, \tau) = 0 \quad (28)$$

$$w_2(\bar{x}, 0) = 0 - c_1(0, \tau) \quad (29)$$

$$0 < \bar{x} < 1.$$

As for comparison for a further analytical solution for this model, we refer to the analytical solution to diffusion-controlled drug release found in Wang and Lou. The model is presented in polar coordinates, with a homogeneous boundary condition.

$$\frac{\partial c(r, t)}{\partial t} = D \left(\frac{\partial^2 c(r, t)}{\partial r^2} + \frac{1}{r} \frac{\partial c(r, t)}{\partial r} \right),$$

$$0 < r < r_2, \quad t > 0$$

$$\frac{\partial c(r^2, t)}{\partial r} = 0, \quad t > 0$$

$$C(r, 0) = \begin{cases} M^0/V_d, & 0 < r < r_1 \\ 0, & r_1 < r < r_2 \end{cases}$$

which has the following solution:

$$C(r, t) = \frac{M^0 \sigma^2}{V_d} + \frac{2M^0 \sigma}{V_d} \sum_{n=0}^{\infty} \frac{J_1(\sigma a_n)}{a_n J_0^2} J_0\left(\frac{a_n r}{r_2}\right) e^{-D a_n^2 t / r_2^2},$$

where M^0 is the initial loading and V_d is the volume of the device.

Conclusion

The swelling-controlled and diffusion-controlled processes have been represented by the advection-diffusion equation and the diffusion equation, respectively, with a nonhomogeneous moving boundary condition. The analytical solutions from the proposed mathematical models will be further developed in the next phase of this study. This will be suitable for the swelling hydrogel problem. In other words, it can be used to estimate the effective

diffusion coefficient of a drug from a delivery device with a 2D disc geometry, to an external finite volume, as well as for similar cases.

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