Mathematical model for mass transfer coefficient determination in dissolution process

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Abstract: - Mathematical models play an essential role in simulation of dynamic processes as they provide insights into the description and behaviour of real systems. Models of chemical processes are derived from laws of conservation, thermodynamic, and control design. In the present paper, we study the modelling of dissolution process determining the quasi steady state material balance for velocity profile, the profile of compound concentration through the space dimension and time. We discuss the assumptions implied in the analysis and deduce a method of estimating the mass transfer coefficient of a solid in terms of the variation of solid weight, rate of dissolution and total flow rate. The use of mathematical modeling aids in the evaluation of simplified equations that account satisfactorily for determining the dissolution rate of solids in a fluid system.

Key-Words: - dissolution, mathematical model, diffusion, mass transfer coefficient, mass transfer rate, boundary conditions

1 Introduction

Mathematical modelling finds a huge number of successful applications in a vast amount of science, social and engineering fields, including biology, chemistry, physics, computer sciences, artificial intelligence, bioengineering, finance, economy and others. Accordingly, the mathematical description that represents the interactions and dynamics of a system contains properties, inner parameters and information about inputs and outputs represented by algebraic equations or by a system of ordinary or partial differential equations. In almost all cases, solution is achieved using algorithms and numerical methods with application of boundary conditions. A process can be studied in steady state if the state variables (v) remain constant as time changes, that is $dv/dt = 0$, for all t, or in dynamic state, in which variation with time occurs.

Process modelling is generally achieved by following the three next steps. Firstly, it is necessary to delimit the research problem in function of the relevant parameters and laws that define the process. In this step a deep knowledge, experience and intuition of the system is required. It is important to consider simplifications and assumptions to propose the idealized model. This allow us to represent the system in a scheme or block diagram with the variables (controlled, manipulated, and disturbance) Following, we need to apply fundamental laws to express in mathematical terms the relationship between different parameters describing phenomena that occur in the system. Here, we require manipulating and simplifying the equations in order to obtain a system of equations (ordinary or partial). After, the equations are solved by means of numerical methods, algorithms and digital computing; simulations can be carried out to model the process, as well as model fitting, interpolation and data prediction. Thus, the model can be validated and verified by comparison with experimental or published data. This allows representing graphically the effect and influence between the main parameters that describe the system under study [1]–[3].

The use of mathematical models in engineering and science fields can be represented in optimum control, estimation of parameters, design of distributed systems, optimization and simulation process. Several challenges can be faced during process design, such as the solution of non linear algebraic equations, numerical integration of ordinary differential equations and partial derivatives by means of discretization using finite difference methods [4]. When the system of differential equations obtained does not have analytical solutions, the design of algorithms using numerical methods is the most common alternative to find the solution. Numerical methods have the advantage of low cost, relative easy implementation

and availability through a diversity of commercial software. Additionally, we can currently find libraries containing subroutines of calculus that use a diversity of sophisticated numerical methods which can realize complex mathematical process, such as numerical analysis, statistical applications or probability [5].

Several efforts have been performed to develop new models capable of determining thermodynamic properties, reactor modelling, distillation column, kinetic parameters, environmental and safety data. Models can be obtained based on predictive methods or in equations and correlation where parameters are adjusted to experimental data. Dissolution is a process in which a solute forms a solution in a solvent. In the study of dissolution we normally come across with several factors that affects this process, namely surface area, agitation, temperature, enthalpy and entropy which affects dissolvability of salts as well as the nature of solvent and solute.[6], [7]. In aqueous dissolution, water is the main component in which the solute is dissolved. The compositional variation that describes the concentration of the components in the solution can be expressed by taking into account the number of moles of each element that is dissolved with respect to the total number of moles, expressed as molar fraction (X) $X_i = n_i / \sum n$, where X_i represents the molar fraction of component i, n_i refers to the moles of component i and $\sum n = (n_a + n_b + \cdots + n_j)$. The sum of molar fractions in a solution has to be equal to 1.

Equilibrium relations for solid-liquid phases are giving by distribution coefficients. The analysis of mass transfer among phases is normally performed following three theoretical models, namely the film theory, the penetration theory, the boundary layer theory. In the film theory, we require a well defined film thickness and the mass transfer coefficient (k) is proportional to the molecular diffusivity D, in this theory only diffusion is present while in the penetration theory, convective and diffusive transport are present, k is proportional to $D^{1/2}$ and we require a well-defined contact time. Accordingly, when we have a flux along a surface and the interest is in the orthogonal transport from the surface to the outer region, the boundary layer theory is generally used. Different correlations are observed in the study of mass transfer of a solid in turbulent fluid, (i.e. k can be proportional to $D^{1-\alpha}$, where α can adopt values between 1/3 and 1/4) [8]. These three theories relate the mass transfer coefficient to the diffusivity. There exist, however, other theories such as the Reynolds analogy and Chilton-Colburns analogy that relates the transfer coefficient to the

heat transfer coefficient and the momentum transfer coefficient. The process of mass transfer between a fluid in motion and different shapes (plates, spheres and cylinders) has been extensively studied by several researchers [9]–[13]. Some of the most common cases are sublimation of solids into air, dissolution of solids in water and vaporization of liquids [14]. Grifoll and Giralt predicted high Schmidt number mass transfer coefficients at smooth surfaces in pipe and duct flows using a modified Van Driest damping equation [15]. In studies related to the mass transfer from a solid to a moving fluid, Linton and Sherwood investigated the dissolution of several compounds, namely benzoic acid, cinnamic acid, and β-naphthol in several fluids flowing through a tube [16]. The use of falling-film columns to study the interphase mass transfer of a solute between fluid streams has been also reported. In these multiphase systems, suspension catalysts are normally used in columns, reactors or fluidized beds[17]. Baldi and Specchia developed a model to evaluate the enhancement factor while studying solid-fluid turbulent mass transfer with chemical reaction [8].

Understanding the dissolution processes of any compound considers the study of kinetic rates, controlling factors, products of dissolution, kinetics of precipitation, chemical equilibrium, mechanism of crystallization among other factors [18], [19]. During the formation of dissolutions, homogenous materials are formed with the combination of more than one chemical species that achieve equilibrium. The components of dissolution (solute and dissolvent) form a homogenous mixture and have a regular distribution of their physicochemical properties. As a result, every portion has the same chemical and psychical characteristics but the quantity of solute and dissolvent are in proportions that varied within some limits. This can be exemplified by the capacity of solubilization that every compound has in a specific solvent. The physical properties are given by the concentration of the components, which can be separated by changes of phases such as evaporation, condensation, or fusion; however, they are not able to be separated by sedimentation or centrifugation. This is normally caused due to the size of the particles, which is normally lower than 10 Angstroms. The stability of materials can be influenced by several factors such as transport process. Additionally, the presence of elements like oxygen or other oxidants (i.e. Fe_2O_3, MnO_2, nitrogen compounds) and proper characteristics of the materials such as structure, surface, reactive area, presence of ligands, temperature affect their dissolution rate [20]–[22].

The formation of dissolution is strongly related to the attraction between the solute and the solvent. A solution is formed when attractive forces between different particles are bigger than the attractive forces between particles that are similar. Therefore, the amount of solute that is solubilised depends on the strength of attraction. The attractive intermolecular forces between particles keep the particles together; as a result, these intermolecular forces have to be broken in order to form a solution. [23], [24].

Dissolution process is important in several fields. In the pharmaceutical industry gives information about the disposition of a pharmaceutical compound within an organism or drug release (adsorption, distribution, metabolism and excretion). This is extremely important in cases where drug release should be controlled in time or gradual after specific time of administration. Several systems aimed to control drug delivery try to minimize degradation of compounds prevention of side effects and increment of drug bioavailability. Research in this field has been carried out by the application of micelles as drug carriers, soluble polymers and liposomes. Bioavailability is the fraction of a drug dose in its active form or some of the metabolites absorbed from the site of administration that reaches the systemic circulation or a particular place where it performs the main function. This property can be increased using micellar systems which have the potential to solubilise drugs with low solubility. It also depends on the properties of the pharmaceutical form and its formulation. Drugs can be administrated by various routes, such as oral, rectal, parenteral, topical, inhalational, etc.). They require being in solution to be adsorbed after crossing several semipermeable cell membranes before reaching the systemic circulation. There exists several alternatives to cross cell membranes; among the most important are passive diffusion, facilitated passive diffusion, active transport and pinocytosis. Low bioavailability can be present in cases where metabolism of the drug occurs before reaching the required plasmid concentration. This is frequent in oral administration of low hydrosoluble drugs with low adsorption [25].

2 General model of dissolution process

The general model of dissolution is a first order model, described by the differential equation given in (1)

$$\frac{dC(t)}{dt} = a(C_S - C(t)), \quad C(0) = 0 \qquad (1)$$

where $a > 0$ is constant and C_S is the limit concentration achieved after the solvent is dissolved or the solute is saturated.

When a liquid is flowing through the inner tube formed with crystals of a solute, a gradient of concentrations exist in the solid surface. The solute starts to dissolve slowly and gradually according to the solubility of the material in the respective solution (Fig.1). For the analysis of the system, we consider a linear velocity profile close to the solid under laminar and turbulent flow (Lévêque Approximation) [26], a thin diffusion boundary layer in comparison with the length of the solid, the dissolved material is not involved in any type of reaction, a plug flow model with perfect radial mixing, isothermal conditions and incompressible Newtonian flow with constant fluid physical properties (density, viscosity, specific heat). The concentration of the solid varies along the tube (axial coordinate) with absence of radial diffusion and resistance to mass transfer due to liquid phase contribution. The diameter of the cylindrical tube remains constant.
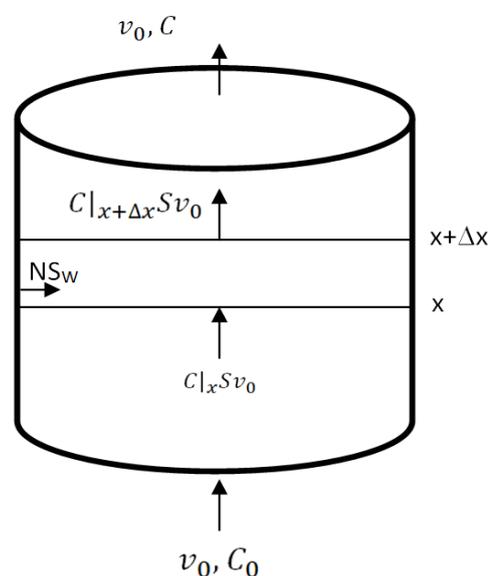
Fig.1. General representation of the system under study

To analyze the process, we proceed to formulate the problem by describing the schematic representation of the system, the governing equations, boundary conditions and initial conditions. Thus, we consider a differential element (shell) of thickness Δx, with constant cross section

area and derive the macroscopic mass balance around the elemental volume. If the process is in steady state, there is no accumulation of mass. There is no chemical reaction but there is transfer of mass between the surface of the solid and the fluid. Additionally, we assume that the system remains infinitesimally close to an equilibrium state at all times. This implies that the process is slow enough to allow the system to adjust itself internally and the properties in one part of the system do not change any faster than those at other parts [27]. As a result, the general conservation law is as expressed in (2):

$$C_x S \vartheta_0 - C_L S \vartheta_0 + \Delta R = 0 \qquad (2)$$

where C is the concentration of the solid, with a distance (x) and a thickness (L), ϑ represents the superficial fluid velocity, S the surface area of the solid, ΔR express the incremental rate expression given by the mass flux [product of the mass transfer coefficient (k), the composition linear driving force (C^*-C)] and the surface area of the tube (S_W). Equation (2) takes the form of (3)

$$C|_x \left(\frac{\pi D^2}{4}\right) \vartheta_0 - C|_L \left(\frac{\pi D^2}{4}\right) \vartheta_0 + k[C_S - C(x)]S_W = 0 \qquad (3)$$

where D is the diameter of the tube, k the mass transfer coefficient, C and C_S are the concentration of the substance and solubility respectively.
Dividing equation (3) by $\pi D \Delta x$ gives

$$C|_x \left(\frac{D}{4\Delta x}\right) \vartheta_0 - C|_L \left(\frac{D}{4\Delta x}\right) \vartheta_0 + k[C_S - C(x)] = 0 \quad (4)$$

$$\left(\frac{-D\vartheta_0}{4}\right)\frac{[C|_L - C|_x]}{\Delta x} + k[C_S - C(x)] = 0 \qquad (5)$$

Taking the limit when $\Delta x \rightarrow 0$ gives the concentration profile with distance, as expressed in equation (6)

$$\left(\frac{-D\vartheta_0}{4}\right)\frac{dC}{dx} + k[C_S - C(x)] = 0 \qquad (6)$$

The system is described by the previous ordinary differential equation, which can be integrated with the following initial condition: $x = 0, C = 0$, which states that the concentration at the beginning of the tube is zero. The solution can be performed by separation of variables

$$\frac{dC}{C_S - C(x)} = \frac{4k}{D\vartheta_0} dx \qquad (7)$$

$$\int \frac{dC}{C_S - C(x)} = \frac{4k}{D\vartheta_0} \int dx \qquad (8)$$

$$-\ln|C_S - C(x)| = \frac{4k}{D\vartheta_0} x + B \qquad (9)$$

$$e^{\ln|C_S - C(x)|} = e^{-\frac{4k}{D\vartheta_0}x + B} \qquad (10)$$

$$C_S - C(x) = B' e^{-\frac{4k}{D\vartheta_0}x} \qquad (11)$$

$$C(x) = C_S - B' e^{-\frac{4k}{D\vartheta_0}x} \qquad (12)$$

At the boundary, when $x=0$, the concentration corresponds to zero ($C=0$), therefore the value of the constant B' is equal to C_S. Substituting the value of the constant gives

$$C(x) = C_S \left(1 - e^{-\frac{4k}{D\vartheta_0}x}\right) \qquad (13)$$

Equation (13) can be used to evaluate the concentration at the boundary, when $X=L$

$$C(L) = C_S \left(1 - e^{-\frac{4k}{D\vartheta_0}L}\right) \qquad (14)$$

The variation of weight (ΔW) according to time ($\Delta \tau$) can be calculated by determining the rate at which the compound is being dissolved (υ), as expressed in (15)

$$\Delta W = \upsilon \Delta \tau \qquad (15)$$

Accordingly, the rate of dissolution [g/s] is determined by multiplying the volumetric flow rate through the cylinder [m³/s], the concentration at the boundary $C(L)$[mol/m³] and the molecular weight of the substance [g/mol] as expressed in (16)

$$\upsilon = \frac{\pi D^2}{4} \vartheta_0 C(L) M_W \qquad (16)$$

Substituting (16) in (15) and using equation (14) leads to equation (17)

$$\Delta W = \frac{\pi D^2}{4} \vartheta_0 C_S \left(1 - e^{-\frac{4k}{D\vartheta_0}L}\right) M_W \Delta \tau \quad (17)$$

Equation (17) can be rearranged to have

$$\frac{4\Delta W}{\pi D^2 \vartheta_0 C_S M_W \Delta \tau} = 1 - e^{-\frac{4k}{D\vartheta_0}L} \qquad (18)$$

For any value of x, the exponential function can be represented as a power series as indicated in (19)

$$e^x = \sum_{n=0}^{\infty} \frac{x^n}{n!} = 1 + x + \frac{x^2}{2} + \cdots \qquad (19)$$

Using the approximation of equation (19) gives (20)

$$e^{-\frac{4k}{D\vartheta_0}L} \approx 1 - \frac{4k}{D\vartheta_0}L \qquad (20)$$

From (20) we can obtain (21)

$$1 - e^{-\frac{4k}{D\vartheta_0}L} \approx \frac{4k}{D\vartheta_0}L \qquad (21)$$

Substituting equation (21) into (18)

$$\frac{4\Delta W}{\pi D^2 \vartheta_0 C_S M_W \Delta\tau} \approx \frac{4k}{D\vartheta_0}L \qquad (22)$$

Finally, obtaining for k gives (23)

$$k \approx \frac{\Delta W}{\pi D C_S M_W \Delta\tau L} \qquad (23)$$

Since equation 23 is an approximation, the maximum error can be estimated according to equation (24)

$$\varepsilon_k = \left|\frac{\partial k}{\partial \Delta W}\right| \varepsilon_{\Delta W} + \left|\frac{\partial k}{\partial \Delta\tau}\right| \varepsilon_{\Delta\tau} + \left|\frac{\partial k}{\partial D}\right| \varepsilon_D \qquad (24)$$

where

$$\left|\frac{\partial k}{\partial \Delta W}\right| = \frac{1}{\pi D C_S M_W \Delta\tau L} \qquad (25)$$

$$\left|\frac{\partial k}{\partial \Delta\tau}\right| = -\frac{\Delta W}{\pi D C_S M_W \Delta\tau^2 L} \qquad (26)$$

$$\left|\frac{\partial k}{\partial D}\right| = -\frac{\Delta W}{\pi D^2 C_S M_W \Delta\tau L} \qquad (27)$$

The substitution of the values of the partial derivatives using equations (25-26), the experimental errors $\varepsilon_{\Delta W}$, $\varepsilon_{\Delta t}$, ε_D and simplifying we obtain equation (28)

$$\varepsilon_k = \frac{\Delta W}{\pi D C_S M_W \Delta\tau L}\left(\frac{\varepsilon_{\Delta W}}{\Delta W} + \frac{\varepsilon_{\Delta\tau}}{\Delta\tau} + \frac{\varepsilon_D}{D}\right) + \left|\frac{\partial k}{\partial \Delta\tau}\right| \varepsilon_{\Delta\tau} + \left|\frac{\partial k}{\partial D}\right| \varepsilon_D \qquad (28)$$

Additionally, we also analyzed the case where the relevant concentration difference for defining a mass transfer coefficient is a local logmean of the driving force at the beginning of the tube (x=0), and a logmean with the concentration driving force at the end of the tube (x=L). In this case, considering equation (14) we can obtain equation (29) and (30). Then, taking the logarithm, and rearranging by multiplying by the exit concentration $C(L)$

$$\frac{C(L)}{C_S} = 1 - e^{-\frac{4k}{D\vartheta_0}L} \qquad (29)$$

$$1 - \frac{C(L)}{C_S} = e^{-\frac{4k}{D\vartheta_0}L} \qquad (30)$$

$$-\frac{4k}{D\vartheta_0}L = \ln\left|1 - \frac{C(L)}{C_S}\right| \qquad (31)$$

$$C(L)\left(-\frac{4k}{D\vartheta_0}L\right) = C(L)\left(\ln\left|1 - \frac{C(L)}{C_S}\right|\right) \qquad (32)$$

$$\frac{(C_S - 0) - (C_S - C(L))\left(-\frac{4k}{D}L\right)}{\left(\ln\left|1 - \frac{C(L)}{C_S}\right|\right)} = C(L)\vartheta_0 \qquad (33)$$

where in the left side of equation (33) is expressed the log-mean concentration driving force, the solid area per unit volume, and the local mass transfer coefficient. By definition, $C(L)\vartheta_0$ is the total $mol//m^2$ flow out of the tube. Thus the total mass transfer flux (N) over the volume area is given by the superficial velocity multiplied by the concentration at the end of the tube multiplied by the cross sectional area of the tube (S) divided by the total area in the tube, which is the area per unit volume of the tube multiplied by the volume of the tube as expressed in (34) and substituting from equation (33), we obtain (35) and finally (36) which relates the local mass transfer coefficient multiplied by a log-mean concentration driving force. In this case, the log-mean average transfer coefficient is a local transfer coefficient. This is useful because it help us to identify a mass transfer rate in case we do not have information of the concentration profile in the bed as we just require the inlet and outlet conditions as well as the saturated concentrations.

$$N = \frac{C(L)\vartheta_0 S}{\frac{4}{D}SL} \qquad (34)$$

$$N = \frac{C(L)\vartheta_0}{\frac{4}{D}L} = \frac{(C_S - 0) - (C_S - C(L))\left(-\frac{4k}{D}L\right)}{\frac{4}{D}L\left(\ln\left|1 - \frac{C(L)}{C_S}\right|\right)} \qquad (35)$$

$$N = \frac{(C_S - 0) - (C_S - C(L))(-k)}{\left(\ln\left|1 - \frac{C(L)}{C_S}\right|\right)} \qquad (36)$$

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3 Discussion

Initially, pure water is passed through a tube constructed of a solid. Considering that the solid is slightly soluble in water ($C*$ moles solid/cm^3 solution), the inner walls of the tube will dissolve very slowly. By weighing the dried tube before and after exposure, it is possible to calculate the rate of mass transfer. As an example to this process, if a base is immersed into a solution containing a low acid concentration with phenolphthalein, there will be a change in the surface of the base, which will turn into a pink coloration. The stirring speed is an important parameter which will determine the thickness of the layer.

Furthermore, considering that the alkaline will saturate the monomolecular film of liquid close to the external solid surface, according to the Nernst diffusion layer theory of dissolution rates, there will exists a gradient of concentration between the acid and the alkali and a layer will be formed. This type of system has been discussed by other researchers but no systematic study of such cases has been made. Accordingly, if crystal of benzoic acid is immersed in an alkaline solution a similar gradient of acid and base concentrations will occur near the external solid surface.

4 Conclusion

We analyzed the dissolution process of solids in a fluid by determining the quasi steady state material balance for velocity profile, the profile of compound concentration through the space dimension and time. We discussed the assumptions implied in the analysis and deduced a method of estimating the maximum possible experimental error in calculating the rate constant.

Using mathematical modeling allows evaluating equations based on assumption that account satisfactorily for the dissolution rate of a solid in dilute solutions. In this paper, we propose a mathematical model that predicts the dissolution of a solid material immersed in a fluid. Accordingly, a log-mean average transfer coefficient was also determined, which is particularly important in cases when we have information about concentrations in the inlet and outlet. Further research is expected to be carried out to use the model as a base to develop useful models for practical applications, such as in the pharmaceutical industry to obtain information about the dissolution of medicaments in the body stream as this could play a key role in formulation of medicaments.

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