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Grey and black-box modelling based on neural networks and artificial immune systems applied to solid dissolution by rotating disc method

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ABSTRACT

The dissolution rates of urea, sodium bicarbonate, and sodium carbonate in water and aqueous solutions were determined using the rotating disc technique. The experiments showed that the dissolution rate increases with increasing disc surface area, temperature, and rotating speed, while it decreases with the solute concentration increase in the dissolution medium. The comparison between experimental values for the dissolution rate and those calculated from Levich equation evidenced a satisfactory agreement in the case of the urea dissolution and poor compliance for the sodium bicarbonate and sodium carbonate dissolution. This poor results and the lack of a good model for making predictions in different situations determined the generation of empirical and semiempirical models (black and grey box approaches) which include neural networks developed with Clonal Selection algorithm (belonging to the Artificial Immune System class) and combination between neural network and phenomenological model. Satisfactory results were obtained with neural networks (black box models) and hybrid models (grey box models).

1. Introduction

Dissolution of solid in liquid phase is an important step in many processes, such as chemical engineering, nuclear engineering, mineral processing, agriculture, pharmaceuticals, electrochemical processes, environmental protection, and so on [1–5]. It is important to know the influence of intrinsic and extrinsic factors on the dissolution mechanism and kinetics, occurring in the development process and sizing equipment.

Dissolution of solids in liquids is complex and is influenced by a number of factors that characterize the properties of solvent and solute and working conditions. The process takes place in several successive stages, two of which being the most important: passage of solute molecules from the solid and transfer of separated molecules towards the bulk liquid phase [6,7].

A number of researchers have used various techniques to study the dissolution of solids in liquids and, based on the relations determined, they proposed models of mass transfer in the dissolution process. Since the surface mass transfer is constant and the hydrodynamic parameters can be easily controlled, one of the most commonly used technique for solid dissolution is represented by the rotating disc technique [8]. This technique has some advantages over other methods: the dissolution takes place only through the disc bottom plane surface, the mass transfer surface is constant, the hydrodynamic parameters can be easily controlled, end effect are negligible, and the heat and mass transfer coefficients are constants [9].

The rotating disc technique has been used in a number of carbonate minerals dissolution studies due to the importance of this family of minerals in a variety of natural and industrial processes: calcite and dolomite dissolution in various aqueous systems [10], scaling and fouling of metal and other surface [11], acidization of petroleum wells [12]. Studies have been conducted to determine the step limiting of the dissolution (mass transfer or reaction at the solid–liquid interface) [10] or the effects of various parameters (temperature, pressure, rotating speed, composition, properties of the dissolution medium, solubility gradient, surface area of the disc) on the rate/flux of dissolution [4]. On the other hand, the rotating disc method is convenient, simple technique to determine kinetics and mechanism of drug dissolution [13], to classify the drugs by the value of the dissolution flux [14]. The dissolution flux plays an important role to a routine quality control test, for evaluating drug release from conventional and novel dosage forms [5].

The theory of the dissolution by rotating disc method was developed by Levich [15]. Levich solved the Navier Stockes and continuity equations to approximate the convective transport of species from

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a bulk solution to the surface of a rotating disc under laminar flow ($Re < 10^4$). The model assumes an infinitely large vessel (concentration is less than 10% of the concentration at the surface of the disc), a large disc diameter so as to ignore edge effects, a constant diffusion layer thickness over the entire surface, the equilibrium established at the solid–liquid interface, and the disc surface constant during the experiment. The model result is accurate for large values of the Schmidt number ($Sc = (\nu/D) \gg 1$). Newman [16] obtained an analytical solution for $Sc > 100$.

In 1986 Tudose and Petrescu [9] have proposed a model in which the mass transfer occurs from the disc surface to the bulk liquid and have solved the convective diffusion equation considering the radial component of the fluid velocity.

Rotating disc technique was used by Ozmetin [8] for studying silver dissolution in nitric acid. Based on the Levich model and the experimental data, a semi-empirical model was derived as a function of rotating disc speed, reaction temperature, nitric acid concentration, and rotating disc surface area.

The complexity of real world processes such as those encountered in the chemical engineering field is often translated into great difficulties when trying the capture their dynamic and behaviour into a model. In the majority of cases, the tools used to create these models are represented by the mathematical relations describing the physical and chemical laws that govern the systems. However, these laws are complex and difficult to apply requiring specialized software (as in case of [17]), are not always fully known and understood, fact that leads to a simplified model (that does not capture the entire dynamic of the system and/or provides results far from the experimental ones). Therefore, different approaches were proposed for modelling, control and optimization of engineering systems [18,19].

In the present work, the dissolution rate of solids with different solubility in distilled water and in aqueous solution of the solute was studied, along with the influence of disc surface area, dissolution medium temperature, and rotating speed of the disc on the dissolution rate. It should be noted that, compared with other known studies, this work was carried out using solutions with high concentrations. A comparison between experimental and calculated dissolution rate, based on the only available model of Levich [15], lead to unsatisfactory results which determined the development of other types of models.

Empirical models represent acceptable alternatives to phenomenological ones, the open literature containing a multitude of variants from which biological inspired algorithms represent promising tools. The current work employs an approach based on two bio-inspired algorithms: (i) Clonal Selection algorithm (CS) belonging to the Artificial Immune System (AIS) and (ii) Feed Forward Neural Network that is a particular case of an Artificial Neural Network (ANN).

AIS is a group of computational paradigms represented by highly abstract models of the biological immune systems [20]. The main motivation of using immune systems as a source of inspiration for computational systems resides in its capabilities related to the self-evolution, self-organization, and self-sustainability [21]. In addition, unlike other biological systems such as nervous systems, the immune system is not centrally controlled and, therefore, detection and response can be locally executed [22].

ANNs are computational structures inspired by neuro-science, having specific characteristics such as parallel processing, learning, and fault tolerance. They are excellent mathematical tools that can be used for dealing with complex, non-linear problems. Also, they are able to extract system features from experimental data using no prior knowledge about the process [23]. This capability is also employed in the current work, the ANN acting as a model for the considered process, its determination being performed using information taken from experimental data. This approach is extensively used in chemical engineering, where various processes are modelled by different types of neural networks.

Although AIS algorithms are used for solving a great range of problems from different domains, in the chemical engineering field the applications are scarce [24,25]. On the other hand, related to the combination 'AIS-ANN-chemical engineering processes', to the authors' knowledge, only a few studies can be found [26]. For instance, different variations CS-ANN were applied for the removal of heavy metals from residual water [27] and optimization of CO_2 absorption in pneumatic contractors [28]. In the current work, a modified version of the methodology used in [27] is employed based on the necessity to obtain a good model for the process.

Neural network designed in an optimal form with CS algorithm and hybrid models represented by combinations between ANNs and phenomenological Levich model were the models developed in this work. The main contribution of this paper is related to the designing of new good (accurate) models, easy to develop and use, which can provide supplementary information about the process through predictions for inexperienced conditions, in terms of saving materials, time, and energies. Another benefit of the developed models derives from the possibility to include them into optimal control procedures. Compared to the existing (enumerated above) phenomenological models, our models provide accurate results and work on a large condition domain because they are based on input–output data.

2. Experimental technique

2.1. Materials

Three solids, urea, sodium carbonate, and sodium bicarbonate, were chosen as the solute in this study, based on their practical importance in agriculture, chemical, pharmaceutical, and food industries. The solubility in water of the three substances is different and decreases from urea to sodium bicarbonate. The dissolution medium was either distilled water or aqueous solution of urea (30 g urea/100 g water and 40 g urea/100 g water), 15% aqueous solution of sodium carbonate, and 5% aqueous solution of sodium bicarbonate. In this way, the influence of solvent composition on the dissolution of the solute can be determined.

The solute and dissolution medium properties were determined from the literature [29–31]. The solubility data of urea, sodium carbonate, and sodium bicarbonate in water are reported in the literature and are presented in Table 1 [31–33].

2.2. Methods

A rotating disc/stationary fluid system is simple to construct and provides reproducible data via a design equation based on fluid mechanics and forced convection principles. The latter characteristic is important since it links the flux or dissolution rate with transport quantities (diffusion coefficient and viscosity), with solubility, surface area, and stirring rate.

The dissolution method is based on the mass transfer from the disc of solute (urea, sodium carbonate, or sodium bicarbonate) to dissolution medium (distilled water or aqueous solution of urea, sodium carbonate, or sodium bicarbonate).

Both the dissolution rate and dissolution flux describe how fast the solute is released from solid surface into dissolution medium:

$$v_D = \frac{\Delta m}{\Delta t} \quad (1)$$

$$J = \frac{\Delta m}{A \cdot \Delta t} \quad (2)$$

where v_D is the dissolution rate ($kg\,s^{-1}$), Δm is the amount of dissolved substance (kg), Δt is the dissolution time (s), J is the