MODELLING FOR ENGINEERING & HUMAN BEHAVIOUR 2022

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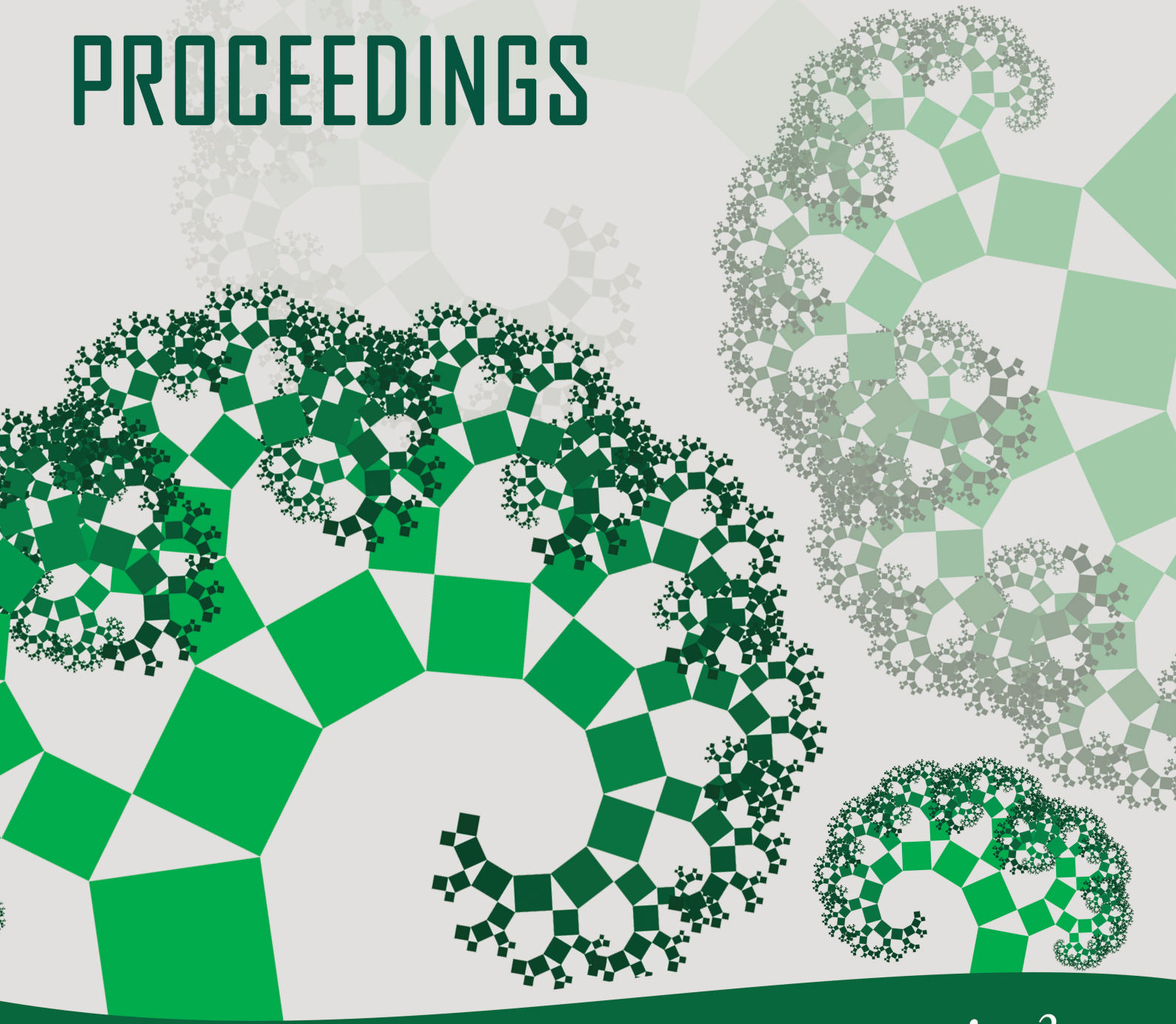

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Probabilistic analysis of the pseudo-n order adsorption kinetic model

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1 Introduction

One of the major problems of recent decades is environmental pollution. Among the various causes of contamination, human activity is one of the main ones. Intense industrial activity has brought with it multiple ecological damages, including the generation of polluting wastes and residues that have a negative impact on the environment. At present, one of the serious consequences of environmental problems is water pollution, especially the discharge of toxic industrial effluents such as lead, copper or organic dye waste into wastewater [2–5,5]. The effects of pollutants cause detrimental impacts on both the environment and human health [2, 3]. These can be avoided by treating contaminated wastewater. In recent years, numerous techniques such as reverse osmosis [2, 4], ultrafiltration [6], photocatalysis [6], electrochemical degradation [6], coagulation [2,4] and adsorption [2, 3, 6], among others, have been studied and applied to remove pollutants from wastewater. Adsorption has been recognized as superior to other techniques due to its high efficiency, simple design, easy operation and maintenance, and availability of different adsorbents at relatively low cost [2, 4–6, 6, 8, 9].

Adsorption is a surface phenomenon in which atoms, ions or molecules are trapped or deposited on the surface of a material. The efficiency of this process is studied by means of adsorption kinetics, since it provides important information on the rate and mechanism of adsorption, as well as on the time required for the adsorption process to complete [8–10]. In the literature we can find several models that have been developed to describe the adsorption kinetic process such as the pseudo-first-order model [11], the pseudo-second-order model [12], the Ritchie's equation [13] or the Elovich model [14], the first two being the most widely used. In the present work, we will introduce the pseudo-n-order (PNO) equation, whose form is as follows

$$\frac{\mathrm{d}q(t)}{\mathrm{d}t} = k_n(q_\mathrm{e} - q(t))^n, \qquad n > 1, \tag{1}$$

where $q(t)$ represents the adsorption capacity at time t (mg/g), n the reaction order, q_e the adsorption capacity at equilibrium (mg/g) and k_n the rate ratio ($(\mathrm{mg/g})^{1-n}\ \mathrm{min}^{-1}$), which, in turn, has the following expression

[1] jccortes@mat.upv.es

$$k_n = \frac{r}{q_e^{n-1}}, \tag{2}$$

where r is the adsorption rate (min^{-1}). For further details, see [10].

By integrating equation (1) yields the exact solution of the PNO model

$$q(t) = q_e \left(1 - \left(\frac{1}{1 + t(n-1)k_n q_e^{n-1}}\right)^{\frac{1}{n-1}}\right), \tag{3}$$

and substituting k_n for expression (2) the following expression is ensured

$$q(t) = q_e \left(1 - \left(\frac{1}{1 + t(n-1)r}\right)^{\frac{1}{n-1}}\right), \tag{4}$$

where the parameters q_e and r are strictly positive and $n > 1$.

In many studies these parameters are considered deterministic. However, they are calculated from experimental data, so they have measurement errors, and depend on molecular properties that are not completely know and also on random external factors such as temperature or humidity, so they contain an intrinsic uncertainty [10]. Therefore, it is more convenient to consider them as random variables. Consequently, this leads to the equation (1) becoming a random differential equation, the solution of which will now be a stochastic process of the following form

$$q(t,\omega) = q_e(\omega) \left(1 - \left(\frac{1}{1 + t(n(\omega)-1)r(\omega)}\right)^{\frac{1}{n(\omega)-1}}\right), \qquad \omega \in \Omega, \tag{5}$$

where $q_e(\omega), r(\omega)$ and $n(\omega)$ are assumed to be absolutely continuous random variables defined on a common complete probability space $(\Omega, \mathcal{F}_\Omega, \mathbb{P})$, with a known joint probability density function (PDF) $f_0(q_e, r, n)$. For simplicity, the ω-notation related to the sample dependence for random variables will be omitted.

This document is organized as follows. In section 2, we will determine the first probability density function (1-PDF), $f_1(q,t)$, of the solution (5). In section 3, the PNO random model is used to model the adsorption of cadmium on the tree fern, in order to apply the theoretical results to real case. Conclusions are drawn in the last section.

2 Computing the 1-PDF of the solution

This section is devoted to determine the 1-PDF of the solution of the PNO random model described in (5). For this purpose, we will apply the Random Variable Transformation (RVT) technique [10]. In this way, we will obtain a complete probabilistic information of the proposed model.

We set $t > 0$ and we apply the RVT technique using the following identification

$$\mathbf{v} = (q_e, r, n), \quad f_{\mathbf{v}}(\mathbf{v}) = f_0(q_e, r, n),$$
$$\mathbf{w} = (w_1, w_2, w_3) = \mathbf{r}(q_e, r, n),$$

being the mapping $\mathbf{r} : \mathbb{R}^3 \longrightarrow \mathbb{R}^3$ and its inverse $\mathbf{s} : \mathbb{R}^3 \longrightarrow \mathbb{R}^3$ given, respectively, by

$$w_1 = r_1(q_e, r, n) = q_e \quad \Rightarrow \quad q_e = s_1(w_1, w_2, w_3) = w_1,$$

$$w_2 = r_2(q_e, r, n) = q_e \left(1 - \left(\frac{1}{1 + t(n-1)r}\right)^{\frac{1}{n-1}}\right) \quad \Rightarrow \quad r = s_2(w_1, w_2, w_3) = \frac{\left(1 - \frac{w_2}{w_1}\right)^{1-w_3} - 1}{t(w_3 - 1)},$$

$$w_3 = r_3(q_e, r, n) = n \quad \Rightarrow \quad n = s_3(w_1, w_2, w_3) = w_3.$$

As $q_e = w_1 > 0$, $n = w_3 > 1$ and $0 \leq \frac{q(t)}{q_e} = \frac{w_2}{w_1} < 1$, then $\left(1 - \frac{w_2}{w_1}\right)^{1-w_3} > 1$ and $r = \frac{\left(1-\frac{w_2}{w_1}\right)^{1-w_3}-1}{t(w_3-1)} > 0$, so it follows that the inverse mapping is well-defined in the conditional probability space $(\Omega, \mathcal{F}_\Omega, \mathbb{P}[\cdot|C])$, where $C = \{\omega \in \Omega : q_e(\omega) > q(t,\omega)\}$. But, due to the way the problem is defined, this condition is guaranteed so the probability of event C is 1 on the above conditional probability space.
The corresponding Jacobian is

$$J_3 = \frac{1}{tw_1(1-\frac{w_2}{w_1})^{w_3}} > 0.$$

Thus, the joint PDF of the random vector $\mathbf{w} = (w_1, w_2, w_3)$ is

$$f_{w_1,w_2,w_3}(w_1, w_2, w_3) = f_0\left(w_1, \frac{\left(1-\frac{w_2}{w_1}\right)^{1-w_3}-1}{t(w_3-1)}, w_3\right) \frac{1}{tw_1(1-\frac{w_2}{w_1})^{w_3}}.$$

Finally, considering an arbitrary t and taking into account that $q(t,\omega) = w_2$, we obtain the 1-PDF of the solution (5) as follows

$$f_1(q,t) = \int_0^\infty \int_1^\infty f_0\left(q_e, \frac{\left(1-\frac{q}{q_e}\right)^{1-n}-1}{t(n-1)}, n\right) \frac{1}{tq_e(1-\frac{q}{q_e})^n} \, \mathrm{d}n \, \mathrm{d}q_e. \tag{6}$$

From it, we can compute all the one-dimensional statistical moments of $q(t,\omega)$, such as the mean, $\mathbb{E}[q(t,\omega)] = \int_{\mathbb{R}} q f_1(q,t)\mathrm{d}q$, or variance, $\mathbb{V}[q(t,\omega)] = \int_{\mathbb{R}} q^2 f_1(q,t)\mathrm{d}q - (\mathbb{E}[q(t,\omega)])^2$, and other interesting features such as the probability that the solution lies within a particular interval of interest, $\mathbb{P}[\{\omega \in \Omega : a \leq q(t,\omega) \leq b\}] = \int_a^b f_1(q,t)\mathrm{d}q$, or the probability that the adsorbed amount exceeds a given quantity $\hat{q}$, $\mathbb{P}[\{\omega \in \Omega : q(t,\omega) > \hat{q}\}] = \int_{\hat{q}}^\infty f_1(q,t)\mathrm{d}q$.

3 An application

The aim of this section is to apply our proposed model together with the above theoretical results to a real case, particularly in a chemical environment, to study cadmium adsorption on tree ferns. In [16] they conclude that tree fern may be a suitable sorbate for sorption of metal cations due its polar and acid characters. The data are shown in Table 1.

t_i	0	4	5	10	15	20	30	45	60
q_i	0	7.172414	8.022989	9.724138	9.793103	10.551724	10.574713	11.103448	11.195402

Table 1: Adsorption capacity of cadmium ions on tree ferns, q_i, for different time instants t_i, $i \in \{1, 2, \ldots, 9\}$. Source [16].

To meet the objective of this section, it is first necessary to establish a joint PDF of the model parameters. Due to the physical significance of the variables, we can consider them independent, so that $f_0(q_e, r, n)$ in this case is the product of the marginals, $f_0(q_e, r, n) = f_{q_e}(q_e) f_r(r) f_n(n)$. Therefore, a PDF must be assigned for each of the model parameters. This is a crucial step in practice, since it is necessary to find appropriate parameter distributions that best capture the data uncertainty.

To do so, we are going to use the Random Least Mean Square (RLMS) method [17]. Considering the positivity, boundedness and the available information of the random variables in the framework of this chemical application we assume

$$q_e \sim \text{Unif}(a,b), \quad a,b>0,$$
$$r \sim \text{Ga}(\alpha,\beta), \quad \alpha,\beta>0,$$
$$n \sim \text{N}_T(\mu,\sigma^2), \quad T=[1,3].$$

Nevertheless, the generality of the results presented in the previous section allows the choice of probability distributions different from those already chosen, which is an advantage on a practical level.

To know the 1-PDF of the solution, it is required to determine the parameters of the proposed parametric distributions, since it depends on these. To this end, we will minimize the mean squared error between the observed data, q_i, and the expectation of the solution $q(t_i,\omega;a,b,\alpha,\beta,\mu,\sigma)$ evaluated at the time instants t_i. This leads to the next optimization program

$$\min_{a,b,\alpha,\beta,\mu,\sigma,>0} \quad \sum_{i=1}^{9} \left(q_i - \mathbb{E}\left[q(t_i,\omega;a,b,\alpha,\beta,\mu,\sigma)\right]\right)^2, \tag{7}$$

where the expectation of the solution is calculated as indicated in section 2, with $f_1(q,t)$ being the expression given in (6).

The output of the minimization problem (7) are the following local optimal values obtained using the Nelder-Mead's algorithm implemented in Mathematica© software

$$a=10.9934,\ b=12.1161,\ \alpha=42.0172,\ \beta=0.009594,\ \ \mu=1.8858,\ \sigma=0.3710.$$

Once the distributions of the model parameters have been estimated using the RLMS method, we obtain the 1-PDF of the solution of the PNO random model, the graph of which is shown in Figure 1.

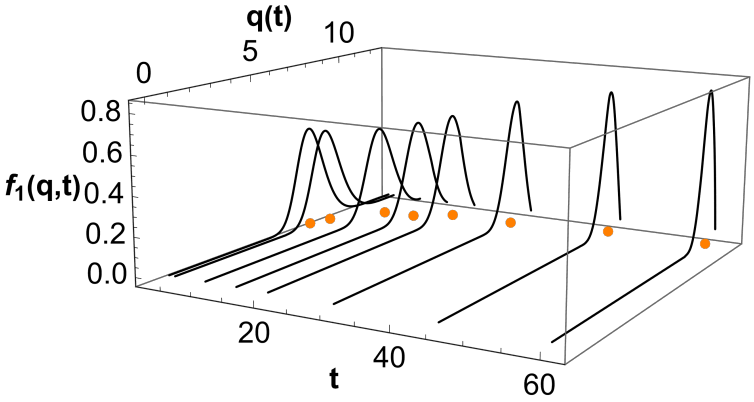

Figure 1: Visual display of the 1-PDF of the solution of the PNO random model, $f_1(q,t)$, given in equation (6), at time instants t_i for $i=2,...,9$, collected in Table 1.

It is observed that for each time instant, the PDF is concentrated approximately around the adsorbed quantity registered, following a morphology quite similar to that of a Gaussian. It is also observed that the density mass shifts with time towards quantities closer and closer to equilibrium levels, with some leptokurtic tendency and some negative asymmetry.

As mentioned above, from the 1-PDF we can get a broader analysis of the solution, thus obtaining its expectation together with the 95% confidence interval, whose representation is shown in Figure 2.

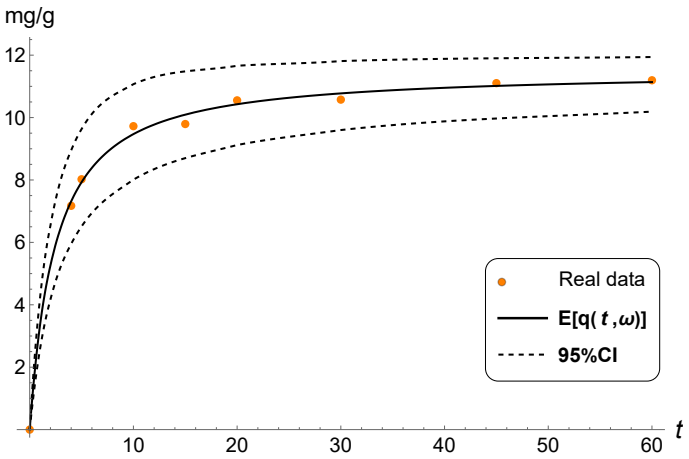

Figure 2: Visual display of the expectation (solid line) together with the 95% confidence interval (dashed lines) of the solution of the PNO random model, which represents the probabilistic fit of the data (points) indicated in Table 1.

Note that the expected value fits the data series well at all time points and that its evolution is increasing over time and stabilizes around equilibrium. As for the 95% confidence interval, we note that it captures the uncertainty coming from our data.

4 Conclusions

In this work we have proposed a randomization of a kinetic model formulated by differential equations to describe the chemical adsorption process. For this purpose, we have treated all the model parameters as random variables with arbitrary distributions. Then, we have solved it probabilistically by determining its respective first probability density function, thus obtaining a full probabilistic description of the solution at each time instant. Given real data, we have shown an uncertainty quantification technique for assigning appropriate distributions to all model parameters when applying the PNO model to real-world data.

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