Solution of inverse anomalous mass transfer problems using a hyperbolic space-fractional model and differential evolution

Solução de problemas de transferência de massa anômala inversa usando um modelo hiperbólico espaço-fracionário e evolução diferencial

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ABSTRACT
The study of anomalous diffusion phenomenon characterizes an important field of science due to limitations of traditional laws considered to represent the conduction term found in mass and heat transfer models. Mathematically, this phenomenon can be represented by empirical and phenomenological models with different levels of complexity. For the latter, differential models with fractional order have been considered. In addition, based on hyperbolic diffusion theory, this fractional differential model can be represented by a second-order derivative on time. This fractional hyperbolic differential model presents two new parameters (fractional order and time relaxation factor) that should be estimated. For this purpose, an inverse problem considering experimental data needs to be formulated and solved. In this context, the present contribution aims to formulate and solve two
inverse anomalous diffusion problems considering a fractional hyperbolic advection-dispersion model to obtain the fractional order and the time relaxation tensor using real experimental data sets. To solve the direct problem the Finite Difference Method is extended for fractional context by using Grünwald-Letnikov Derivative. To solve each inverse problem, the Differential Evolution algorithm is considered as an optimization tool. The obtained results are compared with those found considering the simplification of the hyperbolic space-fractional model. In all analyzed cases, the DE algorithm was able to find good estimates for both parameters and it was demonstrated that the objective function considering the fractional hyperbolic advection-dispersion resulted in lower residual values.

**Keywords:** fick law, inverse anomalous problem, real experimental data, fractional hyperbolic advection-dispersion model, differential evolution.

**RESUMO**

O estudo do fenômeno de difusão anômala caracteriza um importante campo da ciência devido às limitações das leis tradicionais consideradas como representando o termo de condução encontrado em modelos de transferência de massa e calor. Matematicamente, este fenômeno pode ser representado por modelos empíricos e fenomenológicos com diferentes níveis de complexidade. Para este último, modelos diferenciais com ordem de fracionamento foram considerados. Além disso, com base na teoria da difusão hiperbólica, este modelo diferencial fracionário pode ser representado por uma derivada de segunda ordem no tempo. Este modelo diferencial hiperbólico fracionário apresenta dois novos parâmetros (ordem fracionária e fator de relaxamento temporal) que devem ser estimados novamente. Para este efeito, é necessário formular e resolver um problema inverso, tendo em conta os dados experimentais. Neste contexto, a presente contribuição visa formular e resolver dois problemas de difusão anômalos inversos, considerando um modelo de dispersão de advecção hiperbólica fracionada para obter a ordem fracionada e o tensor de relaxamento do tempo usando conjuntos de dados experimentais reais. Para resolver o problema direto, o Método de Diferença Finita é estendido para contexto fracional usando o Derivado de Grünwald-Letnikov. Para resolver cada problema inverso, o algoritmo de Evolução Diferencial é considerado como uma ferramenta de otimização. Os resultados obtidos são comparados com aqueles encontrados considerando a simplificação do modelo espaço-fracionário hiperbólico. Em todos os casos analisados, o algoritmo DE foi capaz de encontrar boas estimativas para ambos os parâmetros e demonstrou-se que a função objetiva considerando a dispersão de advecção hiperbólica fracionada resultou em valores residuais mais baixos.

**Palavras-chave:** lei fictícia, problema anômalo inverso, dados experimentais reais, modelo de dispersão hiperbólica de advecção e fracionária, evolução diferencial.
1 INTRODUCTION

The traditional Fourier and Fick laws are widely used in different fields of science and engineering to represent the diffusive term in phenomenological models. However, under certain conditions, for example when there are: i) very small scales of spatial and temporal variables, the temperature and concentration gradients are considerably high, ii) the induction of some chemical reactions in adsorption processes and the phenomena of time delay, and iii) particle retention and acceleration of diffusive processes, these laws cannot produce good results when compared to experimental data sets (Brandani et al., 2000; Bevilacqua et al., 2011ab; Silva, 2016). According to Gomez et al. (2010), the linear parabolic diffusion theories for these laws considers an infinite speed of propagation. It is true that although the linear parabolic theory propagates disturbances at an infinite speed, their amplitudes decay exponentially. For cases in which linear parabolic diffusion theories cannot be considered, it is necessary to characterize the so-called anomalous phenomena.

Experimentally, both classical and anomalous diffusion phenomena can be observed considering the Boltzmann transformation (Pel et al., 1996). In this case, for a one-dimensional and non-stationary problem, the x-axis is represented as \( xt^{-\gamma} \), where \( x \) and \( t \) are the spatial and temporal variables, respectively, and \( \gamma \) is the scale factor. Thus, if \( \gamma \) is equal to 0.5, the classical diffusion is identified, i.e., the mean squared displacement is a nonlinear function of time. Otherwise, the anomalous diffusion is identified (Pelet et al., 1996).

In order to obtain generalizations of Fourier's Law, the Cattaneo (1958), Vernotte (1958) and Tzou (1995) equations have been considered to expand this law by including delay terms in the temperature gradient and heat flow. This extension can be used in rapid solidification processes and cryotherapy for treating tumors and fusion of nanoparticles (Schwarzwälder et al., 2020). Another way to generalize this law is to apply models that present fractional order derivatives. Using these models, very interesting and promising results have been obtained, such as in anomalous problems related to heating thin metal films by laser and in applications involving multiple layers in fabrics (Mozafarifard et al., 2020).
In a similar way, new approaches have also been proposed to generalize Fick’s Law. In this case, fractional differential equations also can be used to generalize the Fourier’s Law (Gerasimov, 2021). The application of this type of model can be observed in anomalous mass transfer problems that involve fractured geological media, movement of proteins and amorphous solids (Li et al., 2020). Models that present superior derivatives (order greater than two) have also been considered (Paradisi et al., 2001; Bevilacqua et al., 2011a,b). Furthermore, models where the diffusion coefficient is a time function or concentration dependent can also be found (Lima et al., 2019; Zhokh, 2019).

In a fractional context, various mathematical models used to represent the anomalous phenomenon have been proposed. Neuman and Tartakovsky (2009) studied different models (space-time-nonlocal, time-nonlocal and space-fractional) to represent non-Fickian advective–dispersive transport of nonreactive tracers through heterogeneous porous and/or fractured continua. Kumar et al. (2015) proposed a hyperbolic space-fractional bio-heat transfer model based on a single-phase-lagging constitutive equation. Yan et al. (2018) evaluated three time-nonlocal transport models (multi-rate mass transfer model, tempered time fractional advection–dispersion equation model and the continuous time random walk framework) by combining theoretical analyses and applications for laboratory sand column transport experiments and field tracer tests. Sun et al. (2019) studied the sodium chloride transport in a single fracture and captured non-Fickian transport by using a fractional advection-dispersion model. Similarly, Qiao et al. (2020) investigated chloride ion transport in the single vertical fracture under different rough-walled conditions also using a fractional advection-dispersion equation. It is important to mention that the interest for this kind of model is due to an increase in the degree of freedom in the model, accurate explanations of physical phenomena and fractional derivatives are non-local in nature compared to local behavior of integer derivatives (Kumar et al., 2015).

Although the generalization of traditional laws is extremely interesting, the complexity of the models arising from incorporating these new ideas makes them much more difficult to solve (analytically or numerically) (Kheybari et al., 2020). Thus, in general, it can be said that the numerical resolution of problems that characterize anomalous
phenomena poses a great challenge as they require the development or improvement of existing strategies to reduce the associated computational cost (Greer et al., 2006).

Many studies have been carried out to propose and solve inverse problems in an anomalous context to determine the parameters that characterize this phenomenon. This problem consists of determining the parameter vector that minimizes the sum of errors observed by using a particular optimization strategy. Examples can be observed in studies that evaluate heat transfer in fabrics and porous materials (Ghazizadeh et al., 2012; Brociek et al., 2019). The parameter estimation has also been performed in an anomalous context in terms of mass transfer in porous media and water absorption in different materials (Fan et al., 2016; Lima et al., 2019). To solve these inverse problems, different methods can be used, among which we can mention the Differential Evolution and Ant Colony algorithm in heuristic context (Brociek et al., 2019; Lima et al., 2019).

In this contribution, two inverse anomalous problems considered to identify the parameters in a fractional hyperbolic advection-dispersion model by using the Differential Evolution (DE) algorithm are formulated and solved. In order to evaluate the objective function in each inverse problem, the direct problem needs to be solved. For this purpose, the Finite Difference Method (FDM) is extended for fractional context by using Grünwald-Letnikov Derivative (Podlubny, 1999). In order to evaluate the quality of the solutions found, parabolic space-fractional and parabolic space-integer models also are solved. This paper is structured as follows. The next section presents the mathematical formulation of the anomalous diffusion phenomenon. The proposed methodology considered to solve the direct and inverse problems is described in Sections 3 and 4, respectively. The obtained results are shown in Section 5. Finally, the conclusions of this work are drawn in the last section.

2 MATHEMATICAL FORMULATION OF ANOMALOUS DIFFUSION PHENOMENON

The classical theory for pure-diffusive processes is defined by equations (Gomez et al., 2010):
\[ \frac{\partial C}{\partial t} + \nabla J = S \]  \hspace{1cm} (1)

\[ J = -D \nabla C \]  \hspace{1cm} (2)

where \( C \) is the concentration, \( J \) is the flux, \( S \) is a source term and \( D \) is the diffusivity which is assumed positive definite and independent of \( C \). Equation (1) represents the mass conservation and Eq. (2) represents the constitutive relation that defines the flux. Replacing Eq. (2) in Eq. (1), we obtain:

\[ \frac{\partial C}{\partial t} - \nabla (D \nabla J) = S \]  \hspace{1cm} (3)

In order to generalize the classical diffusion equation, Cattaneo (1958) and Vernotte (1958) proposed a more general expression to represent Fick's law. This is given by:

\[ J + \tau \frac{\partial J}{\partial t} = -D \nabla C \]  \hspace{1cm} (4)

where \( \tau \) is the time relaxation factor.

Thus, the so-called hyperbolic diffusion theory is derived substituting Eq.(4) in Eq.(2):

\[ \frac{\partial C}{\partial t} + \nabla J = S \]  \hspace{1cm} (5)

\[ J + \tau \frac{\partial J}{\partial t} = -D \nabla C \]  \hspace{1cm} (6)
It is important to mention that when $\tau$ is equal to zero, Eq.(6) is reduced to Eq.(2) (parabolic theory). In addition, at the steady state, both theories are equivalent even for $\tau$ different from zero (Gomez et al., 2010).

After replacing Eq.(6) in Eq.(5), a hyperbolic partial differential equation is obtained. Traditionally, various authors have studied fractional-order diffusion models (Liu et al., 2014, Afshari et al., 2015; Jia and Wang, 2016; Jia and Wang, 2019; Yazdani et al., 2020). In this case, a fractional derivative in diffusion terms is commonly analyzed.

In order to characterize the model considered in this contribution, a one-dimensional hyperbolic partial differential equation with integer order in time (in terms of first and second derivatives), and with two spatial derivatives, being a first derivative in space with order integer and a second derivative with fractional order in space is proposed. Mathematically, the fractional hyperbolic advection-dispersion model considered in this work is given by:

$$Q_1 \frac{\partial^2 u(x,t)}{\partial t^2} + Q_2 \frac{\partial u(x,t)}{\partial t} + Q_3 \frac{\partial^{\alpha} u(x,t)}{\partial x^\alpha} + Q_4 \frac{\partial u(x,t)}{\partial x} + Q_5 u(x,t) + Q_6 = 0,$$

where $x$ and $t$ are the independent variables, $u$ is the dependent variable, $\alpha \ (1 < \alpha \leq 2)$ is the fractional order and $Q_i \ (i=1, \ldots, 6)$ represents the coefficients that weigh the terms of the model presented. In this model, the following initial and boundary conditions are considered:

$$u(x,t) = f(x), \quad t = 0, \quad 0 < x \leq x_t,$$

$$\frac{\partial u(x,t)}{\partial t} = g(x), \quad t = 0, \quad 0 < x \leq x_t,$$

$$\beta_1 u(x,t) + \beta_2 \frac{\partial u(x,t)}{\partial x} = \beta_3, \quad x = 0, \quad 0 < t \leq t_t.$$
\[ \beta_4 u(x,t) + \beta_5 \frac{\partial u(x,t)}{\partial x} = \beta_6, \quad x = x_i, \quad 0 < t \leq t_f \tag{11} \]

where \( f(x) \) and \( g(x) \) are functions that define the initial conditions, \( x_f \) is the superior limit for spatial variable, \( t_f \) is the final time, and \( \beta_i (i=1, \ldots, 6) \) are constants that define the kind of boundary condition considered.

It is important to emphasize that this model presents one fractional derivative in relation to the diffusive spatial term. In this case, fractional derivatives in more terms (for example, in temporal terms) could be considered (Wang et al., 2021). However, when considering a model with numerous fractional contributions, the real physical effect in relation to the presence of these derivatives in obtained profiles must be evaluated. In this case, only a fractional derivative in second-order in space is considered in this contribution. Finally, in this model, the presence of a term with a second-order derivative in relation to time can be mentioned. This term indicates that the disturbance of system under analysis propagates as a wave that has a finite speed, contrary to what Fick’s Law states (Ordóñez-Miranda and Alvarado-Gil, 2009).

3 SOLUTION OF THE DIRECT PROBLEM

To solve the problem defined by Eqs.(7)-(11), the classical Finite Difference Method (FDM) is extended by the fractional context. For this purpose, \( t_k = k\Delta t, \quad k = 0, 1, 2, \ldots, n_t \) and \( x_i = i\Delta x, \quad i = 0, 1, \ldots, n_x \), where \( \Delta t = t_f/n_t \) (grid step in time) and \( \Delta x = x_f/n_x \) (grid step in space). Let \( u_i^k \) be the numerical estimate of the value exact solution \( u(x,t) \) at the mesh point \((x_i,t_k)\) and the following expressions to approximate both partial derivatives with integer and fractional orders (Podlubny, 1999):

\[
\left. \frac{\partial^2 u_i^k}{\partial t^2} \right|_i = \frac{u_i^{k+1} - 2u_i^k + u_i^{k-1}}{(\Delta t)^2} + \sum_{j=1}^6 \beta_j \frac{u_i^{k+1} - u_i^{k-1}}{2(\Delta x)^2} \tag{12}
\]
where Eq. (12)-(14) represent the classical approximation for first and second derivatives with integer order and Eq.(15) is the approximation of Grünwald for fractional second-order derivative. In this equation, $w_j$ corresponds to the function defined as:

$$w_j = \begin{cases} 1 & \text{if } j = 0 \\ 1 - \frac{\alpha + 1}{j} w_{j-1} & \text{otherwise} \end{cases}$$

Replacing these approximations in Eq.(7) and reorganizing the following expression, we found:

$$\left[ \frac{Q_3}{(\Delta x)^\alpha} + \frac{Q_4}{2\Delta x} \right] u_i^k + \left[ \frac{Q_1}{(\Delta t)^2} + \frac{Q_2}{\Delta t} + Q_6 \right] u_i^k - \frac{Q_4}{2\Delta x} u_i^{k-1} + \frac{Q_3}{(\Delta x)^\alpha} \sum_{j=0}^{i-1} w_j u_i^{k-1-j} =$$

$$(17)$$

This model is linear in relation to the dependent variable $u_i^k$, i.e., this model can be written in the form of $Au=b$, where $A$ is the constant coefficient matrix and $b$ is the independent term vector. It is important to mention that the $k$-th value of $u$ for any value of $i$ is computed considering the initial values in the time step, i.e., by using the values of...
$u$ at $k-1$ and $k-2$ positions. These values are known, i.e., are given by the initial conditions defined by the functions $f(x)$ (Eq.(8)) and $g(x)$ (Eq.(9)), respectively. In order to compute the values of the $u$ at $i$ equal to 0 and $n_x$ for any time $t$ ($t > 0$), the boundary conditions defined by Eqs.(10)-(11) should be applied.

In summary, if all parameters that characterize this model (final time, final length, initial and boundary conditions and vector $Q$) are known and if the number of discretization points (in both directions) also are known, the direct problem can be solved. On the other hand, if one or more pieces of information of this model are not known, but experimental data are known, an inverse problem can be formulated. This description is presented in the next section.

4 SOLUTION OF THE INVERSE PROBLEM

As mentioned earlier, in this contribution, the model parameters used to represent the anomalous diffusion phenomenon considering real experimental data presented will be determined. For this purpose, an inverse problem considering the DE algorithm needs to be formulated and solved. From a mathematical point of view, this consists of determining the parameters that characterize the model described by Eqs.(7)-(11) that minimize the difference between the experimental and calculated values. This problem can be formulated as:

$$OF = \sum_{i=1}^{M} \left( \frac{C_{i}^{\text{sim}} - C_{i}^{\text{exp}}}{\max(C_{i}^{\text{exp}})} \right)^2$$

where $OF$ is the objective function, $M$ is the number of experimental data, $C_{i}^{\text{sim}}$ and $C_{i}^{\text{exp}}$ represent the simulated and experimental values for concentration, respectively, and $\max(C_{i}^{\text{exp}})$ is the highest observed experimental value.

Figure 1 illustrates the procedure adopted in this work. Initially, the user defines the objective function (sum of difference between the prediction model and the experimental data), the design variables, the design space, the DE parameters, the fractional
order and the FFDM parameters. Then, the optimization strategy generates potential candidates considering the operators (mutation, crossover and selection). Each candidate is evaluated according to the objective function. For this purpose, the fractional model is simulated to obtain the predicted values. These values are used in set with the experimental data to define the objective function. This procedure is repeated until a stop criterion is satisfied.

Figure 1 – Flow Diagram for Proposed Solution Procedure.

Source: The authors.

4.1 DIFFERENTIAL EVOLUTION ALGORITHM

To solve the proposed inverse problem, the DE algorithm, proposed by Storn and Price (1995), is considered. This algorithm consists of the following steps:

- Initially, an initial population is randomly generated with $NP$ feasible solutions;
- An individual $(X_1)$ is randomly selected in the population to be replaced. Two other individuals $(X_2$ and $X_3$) are randomly selected in the population to perform the vector subtraction;
- The result of the subtraction operation between $X_2$ and $X_3$ is weighed by perturbation rate ($F$). This result $(F \times (X_2 - X_3))$ is added to the individual $(X_1)$. Therefore,
the new (potential) candidate \((X)\) is given by: 
\[ X = X_1 + F(X_2 - X_3) \]  
It is important to emphasize that other schemes to generate potential candidates can be used. Some of these are presented below (Storn and Price, 1995):

\[ X = X_1 + F(X_2 - X_3 + X_4 - X_5) \]  
\[ X = X_{\text{best}} + F(X_2 - X_3) \]  
\[ X = X_{\text{best}} + F(X_2 - X_3 + X_4 - X_5) \]  
\[ X = X_1 + F(X_{\text{best}} - X_2) \]  
\[ X = X_1 + F(X_{\text{best}} - X_1) + F(X_1 - X_2 + X_3 - X_4) \]  

where \(X_1, X_2, X_3, X_4\) and \(X_5\) are candidate solutions randomly chosen in the current population and \(X_{\text{best}}\) is the candidate solution associated with the best fitness value (in the current generation);

- If the resulting vector \((X)\) has a better value in terms of the objective function, it can replace the previously chosen candidate. This operation happens if a random number generated is less than the crossover probability, also defined by the user. Otherwise, the previously chosen candidate survives in the next generation. This procedure is repeated until \(NP\) completes the candidates (formed by new and current individuals).
- To finalize the algorithm, the stopping criteria is defined by the user (generally the maximum number of generations).

As emphasized by Storn and Price (1995), DE presents the following advantages:

\(i\) capacity to escape from the local optimal; \(ii\) easy to implement; and \(iii\) ability to deal with problems that consider different types of variables (continuous, discrete, binary and
integer). On the other hand, the main disadvantage is the high number of objective function evaluations compared to the Classical Methods. More details about the DE can be found in Storn and Price (1995).

5 RESULTS AND DISCUSSION

To apply the proposed methodology to solve inverse problems, two real experimental data sets are considered. In this case, the following points should be highlighted.

- Processing time (PT) is estimated using an Intel Core i5-6200 Notebook computer with 8 GB of memory.
- In order to guarantee the dimensional consistence in each fractional phe-
nomenological problem, the fractional diffusive operator is represented as:

\[
\frac{\partial^2}{\partial x^2} \rightarrow \frac{1}{\sigma^{(2-\alpha)}} \frac{\partial^\alpha}{\partial x^\alpha}, \quad 1 < \alpha \leq 2
\]  

(24)

where \(\sigma\) is a parameter (with unit length) introduced to correct the dimension of the fractional term. This simple substitution guarantees that the fractional diffusive operator will always be dimensionally coherent. So that parameter \(\sigma\) does not interfere, quantitatively, in the simulated profiles, it is defined as being equal to unity.

- To represent the mass transfer in the proposed inverse problems, a Fractional Hyperbolic Advection-Dispersion Equation (FHADE) (see Eq.(7)) is considered (Gomez et al., 2010):

\[
\tau \frac{\partial^2 C}{\partial t^2} + \frac{\partial C}{\partial t} = -v \frac{\partial C}{\partial x} + \frac{1}{\sigma^{(2-\alpha)}} D \frac{\partial^\alpha C}{\partial x^\alpha}, \quad 0 < t \leq t_f, \quad 0 \leq x \leq x_f, \quad \text{and} \quad 1 < \alpha \leq 2
\]  

(25)

where \(t\) and \(x\) represent the temporal and spatial coordinates (\(x_f\) is the maximum length in \(x\)-direction), respectively, \(C\) is the concentration, \(\tau\) is the time relaxation factor, \(\alpha\) is the fractional order, \(v\) is the average velocity, \(D\) is the dispersion coefficient and \(\sigma\) is the parameter introduced to correct the dimension of the fractional term.
In order to compare the obtained results by FHADE, two simplified models are considered. The first consists of the Fractional Parabolic Advection-Dispersion Equation (FPADE), i.e., for this model, the time relaxation factor is equal to zero. The second consists of the Integer Parabolic Advection-Dispersion Equation (IPADE), i.e., for this model, the time relaxation factor is equal to zero and the fractional order is equal to 2. In this case, two additional inverse problems also are solved.

The initial and boundary conditions and real experimental data considered in each application were defined according each reference consulted.

For the simulation of each direct problem, 100 points are considered in spatial and temporal directions. These parameters were chosen from preliminary simulations.

The objective function in each case study was defined as the sum of difference between the experimental and simulated values, as shown in Eq.(18).

DE parameters: population size (25), maximum number of generations (100), crossover probability and perturbation rate equals to 0.8 and the strategy number 7 (Storn and Price, 1995). The stopping criterion adopted in this work was the maximum number of generations. Using these parameters, the number of objective function evaluations is equal to 25+25x100 times. Each inverse problem was solved 20 times considering independent runs. It is important to highlight that these parameters were chosen from preliminary simulations.

The systems of algebraic equations obtained by applying the FFDM is solved considering the LU Decomposition Method (Press et al., 2007).

5.1 CASE 1

In this first application, the experiments carried out by Zhao et al. (2016) to determine the diffusivity of CO₂ in a porous medium saturated with liquid n-tetradecane using the magnetic resonance imaging technique are considered. The container in which the sample was located consisted of a glass tube that was filled with glass spheres. The gas in the cylinder was injected into the porous medium until the equilibrium of the system
was reached, the condition in which the experiment was ended. The gas and liquid phases were kept at constant temperatures equal to 20 ºC and 30 ºC, respectively, and the pressure was varied from 2000 to 5000 kPa.

In this model, the initial and boundary conditions are given by (Chang and Sun, 2018):

\[ C(0, x) = 0, \quad t = 0 \quad \text{and} \quad 0 \leq x \leq x_t \]  \hspace{1cm} (26)

\[ \frac{\partial C(0, x)}{\partial t} = 0, \quad t = 0 \quad \text{and} \quad 0 \leq x \leq x_t \]  \hspace{1cm} (27)

\[ C(t, 0) = \begin{cases} 1, & 0 < t \leq 10, \\ 0, & t > 10 \end{cases}, \quad t > 0 \quad \text{and} \quad x = 0 \]  \hspace{1cm} (28)

\[ \frac{\partial C(t, x_t)}{\partial x} = 0, \quad t > 0 \quad \text{and} \quad x = x_t \]  \hspace{1cm} (29)

It is important to mention that Eq.(27) was defined as a complementary condition to simulate the FHADE model, as \( C(0, x) \) is equal to zero. In addition, the following information on each model, based on Chang and Sun (2018), is defined:

- **IPADE (\( \alpha = 2 \) and \( \tau = 0 \)):** the aim is to determine the average velocity \( (v) \) and the dispersion coefficient \( (D) \) considering the following design space: \( 0.001 \text{ mm/min} \leq v \leq 0.006 \text{ mm/min} \) and \( 0.001 \text{ mm}^2/\text{min} \leq D \leq 0.050 \text{ mm}^2/\text{min} \).

- **FPADE (1 < \( \alpha \) \leq 2 and \( \tau = 0 \)):** the aim is to determine \( v, D \) and the order fractional \( (\alpha) \) considering the following design space: \( 0.001 \text{ mm/min} \leq v \leq 0.006 \text{ mm/min} \), \( 0.001 \text{ mm}^2/\text{min} \leq D \leq 0.050 \text{ mm}^2/\text{min} \) and \( 1.5 \leq \alpha \leq 2 \).

- **FHADE(1 < \( \alpha \) \leq 2 and \( \tau > 0 \)):** the aim is to determine \( v, D, \alpha \) and \( \tau \) considering the following design space: \( 0.001 \text{ mm/min} \leq v \leq 0.006 \text{ mm/min}, 0.001 \text{ mm}^2/\text{min} \leq D \leq 0.050 \text{ mm}^2/\text{min}, 1.5 \leq \alpha \leq 2 \) and \( 0 \leq \tau \leq 1 \text{ min} \).
• It is important to mention that the limits for each design variable were obtained after preliminary runs.
• Finally, the $OF$ is evaluated for $t$ equal to 192 min and $0 \leq x \leq x_f$ (16.5 mm) considering the experimental data obtained by Zhao et al. (2016).

Table 1 presents the results obtained by using DE considering different models.

<table>
<thead>
<tr>
<th>Model</th>
<th>$\alpha$</th>
<th>$\tau$ (min)</th>
<th>$v$ (mm/min)</th>
<th>$D$ (mm$^2$/min)</th>
<th>$OF$</th>
<th>PT (min)</th>
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</thead>
<tbody>
<tr>
<td>IPADE</td>
<td>-</td>
<td>-</td>
<td>0.0010</td>
<td>0.0337</td>
<td>0.5053</td>
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<td>0.5435$\times 10^{-9}$</td>
<td>0.2543$\times 10^{-12}$</td>
<td>18.46</td>
</tr>
<tr>
<td>FHADE</td>
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<td>0.1575</td>
<td>0.0017</td>
<td>0.0324</td>
<td>0.1850</td>
<td>18.53</td>
</tr>
</tbody>
</table>

$^a$Best value, $^b$Standard deviation and $^c$Average value required by each run of DE.

Source: The authors.

This table shows that this optimization strategy was able to estimate the values for the design variables, as observed by the average and standard deviation values (in relation to $OF$). As observed, the increase in the number of design variables reduce the standard deviation. In addition, the decrease in the objective function value when the number of design variables increases is observed, i.e., the FHDAE model presents the best value for $OF$. This result is expected, since increasing the number of design variables and, consequently, the number of freedom degrees increases the model's ability to adhere to experimental data. In relation to processing time, all models result in similar results, i.e., the increase in complexity of model does not increase the computational time.

The obtained average velocity by IPADE model is lower than that found by FHDAE. This can be explained by the preferential motion in which path particles move faster, which characterizes a super-diffusion process. The non-local spatial features of this process can be captured by the fractional order term. On the other hand, the reduction in the average velocity is considered as an alternative to adding the reduction of velocity-dispersion. This result can be justified by the presence of parameter $\tau$, which is related to sub-diffusion process due to filling of some pores in medium by gas particles. This
process slows down the particles. The ratio between the average and the dispersion calculation for the models presents low values. This demonstrates that the dominant phenomena correspond to dispersion, which allows both parameters to be considered constant.

Figure 2 presents the experimental and simulated concentration profiles for each model considering $t$ equal to 192 min.

This figure shows a good agreement between the experimental data and simulated values for each model. It can be observed that the FHADE model has a greater capacity to represent the experimental points when compared to the other models. Physically, the concentration increases until reaching the maximum value. Subsequently, the concentration decreases with the increment of value of the spatial variable. The position at which the maximum concentration value occurs as the number of model parameters increases can also be seen, as observed in Table 1.

Chang and Sun (2018) also proposed and solved an inverse considering this experimental data set. However, the model used by authors presents fractional derivatives in relation to time and space and the model is parabolic. In this case, the authors obtained an average speed equal to 0.01 mm/min and a dispersion coefficient equivalent to 0.086 mm$^{1.85}/$min$^{0.77}$. The values of non-integer orders obtained by authors were equivalent to 0.77 and 1.85 for temporal and spatial derivatives. Qualitatively, the obtained
concentration profiles in this contribution are similar to those obtained by Chang and Sun (2018) considering a parabolic model, demonstrating the quality of the model used.

5.2 CASE 2

In this last application, three inverse problems are also formulated and solved by using the IPADE, FPADE and FHADE models considering the two materials studied by Nowamooz et al. (2013), namely sandstone and granite. These authors carried out experiments from which the concentration of an aqueous methylene blue solution was determined during its flow in sandstone and granite fractures, which were initially filled with water and later with the solution. After it was filled with the solution, each material was injected into the fractures at constant flow rates. From the processing of images recorded during the experiments and the Beer-Lambert attenuation law, the authors determined the concentration at the exit of the fractures and in different positions.

In this application, the initial and boundary conditions based on Nowamooz et al. (2013) are defined as follows:

\[
C(0, x) = 0, \quad t = 0 \quad \text{and} \quad 0 \leq x \leq x_f
\]  
(30)

\[
\frac{\partial C(0, x)}{\partial t} = 0, \quad t = 0 \quad \text{and} \quad 0 \leq x \leq x_f
\]  
(31)

\[
C(t, 0) = 1, \quad t > 0 \quad \text{and} \quad x = 0
\]  
(32)

\[
\frac{\partial C(t, x_f)}{\partial x} = 0, \quad t > 0 \quad \text{and} \quad x = x_f
\]  
(33)

As mentioned by the first inverse problem, Eq. (31) was defined as a complementary condition to simulate the FHADE model. In addition, the following information about each model is defined (Nowamooz et al., 2013):
• IPADE ($\alpha = 2$ and $\tau = 0$): the aim is to determine the average velocity ($v$) and the dispersion coefficient ($D$) considering the following design space: $0.01 \text{ mm/min} \leq v \leq 5 \text{ mm/min}$ and $0.0001 \text{ mm}^2/\text{min} \leq D \leq 5 \text{ mm}^2/\text{min}$.

• FPADE ($1 < \alpha \leq 2$ and $\tau = 0$): the aim is to determine $v$, $D$ and the order fractional ($\alpha$) considering the following design space: $0.01 \text{ mm/min} \leq v \leq 5 \text{ mm/min}$, $0.0001 \text{ mm}^2/\text{min} \leq D \leq 5 \text{ mm}^2/\text{min}$ and $1.2 \leq \alpha \leq 2$.

• FHADE($1 < \alpha \leq 2$ and $\tau > 0$): the aim is to determine $v$, $D$, $\alpha$ and $\tau$ considering the following design space: $0.01 \text{ mm/min} \leq v \leq 5 \text{ mm/min}$, $0.0001 \text{ mm}^2/\text{min} \leq D \leq 5 \text{ mm}^2/\text{min}$, $1.2 \leq \alpha \leq 2$ and $0 \leq \tau \leq 1 \text{ min}$

• All limits were defined after preliminary runs.

• The $OF$ is evaluated for each material considering different positions for each material ($[0.25 0.45 0.6 0.8] \text{ mm}$ for sandstone and $[0.2 0.4 0.6 0.8] \text{ mm}$ for granite), $x_f$ equal to 1 mm and $0 \leq \tau \leq 4 \text{ min}$. The experimental points used were extracted from Nowamooz et al. (2013).

Table 2 presents the results obtained by using DE considering different models and different materials for the second test case.

<table>
<thead>
<tr>
<th>Model</th>
<th>$\alpha$</th>
<th>$\tau$ (min)</th>
<th>$v$ (mm/min)</th>
<th>$D$ (mm$^2$/min)</th>
<th>$OF$</th>
<th>PT$^c$ (min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>IPADE</td>
<td>-</td>
<td>-</td>
<td>0.0627</td>
<td>0.9944</td>
<td>0.2018</td>
<td>18.5</td>
</tr>
<tr>
<td></td>
<td>1.9864(a)</td>
<td>-</td>
<td>0.0566</td>
<td>1.0381</td>
<td>0.1711</td>
<td>18.9</td>
</tr>
<tr>
<td>FPADE</td>
<td>1.2325×10$^{-5}$(b)</td>
<td>-</td>
<td>2.3433×10$^{-6}$</td>
<td>3.4454×10$^{-6}$</td>
<td>1.3434×10$^{-7}$</td>
<td>19.2</td>
</tr>
<tr>
<td>FHADE</td>
<td>1.9918</td>
<td>0.1014</td>
<td>1.0166</td>
<td>0.0246</td>
<td>0.1578</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Model</th>
<th>$\alpha$</th>
<th>$\tau$ (min)</th>
<th>$v$ (mm/min)</th>
<th>$D$ (mm$^2$/min)</th>
<th>$OF$</th>
<th>PT$^c$ (min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>IPADE</td>
<td>-</td>
<td>-</td>
<td>0.0714</td>
<td>0.7554</td>
<td>0.3289</td>
<td>17.8</td>
</tr>
<tr>
<td></td>
<td>1.9710(a)</td>
<td>-</td>
<td>0.0561</td>
<td>0.8504</td>
<td>0.2011</td>
<td>17.8</td>
</tr>
<tr>
<td>FPADE</td>
<td>3.5545×10$^{-5}$(b)</td>
<td>-</td>
<td>2.3433×10$^{-7}$</td>
<td>4.5544×10$^{-7}$</td>
<td>2.4343×10$^{-9}$</td>
<td></td>
</tr>
<tr>
<td>FHADE</td>
<td>1.9767</td>
<td>0.0820</td>
<td>0.8389</td>
<td>0.0195</td>
<td>0.1878</td>
<td></td>
</tr>
</tbody>
</table>

$^a$Best value, $^b$Standard deviation and $^c$Average value required by each run of DE.

Source: The authors.
This table shows that the value of the objective function decreases when the number of design variables is increased, as expected, since the greater the number of model parameters, the greater the tendency for it to adjust to experimental data. Low values for the standard deviations in relation to parameters and objective function are verified, demonstrating that the DE algorithm always converges for the same solution. It can be seen that the OF values are a little lower when considering the inverse problems formulated for sandstone. This indicates that the models evaluated are more accurate in describing the diffusion in this material than in granite. Furthermore, it appears that the values of $\alpha$ obtained for sandstone are closer to 2 than those found for granite. This indicates that granite is more spatially heterogeneous, which was experimentally verified by Nowamooz et al. (2013). A higher value of the parameter $\tau$ is also observed for the sandstone. This indicates that in this material, the particles need a longer time interval to diffuse from one point to another than in granite.

For both materials, it can be observed that the average speeds obtained when considering the FHADE are lower than those found with the application of the IPHADE. This result can be related to the retention of particles that occurs in dead zones present in the medium, characterizing a sub-diffusion process. When using the FHADE, the obtained values for $v$ are significantly higher than those found for the two previous equations. It can be observed that the diffusion is more considerable in the sandstone, since for this material higher values of dispersion coefficient are obtained than for the granite. It can be observed that when using the two equations that do not present the parameter $\tau$, the ratio between the average speed and dispersion coefficient presents small values, which indicates that in these cases, dispersion is the predominant phenomenon. For the equation in which $\tau$ is not null, it can be seen that the value of this ratio is significantly higher. This demonstrates that in this case, the predominant phenomenon corresponds to advection.

In relation to processing time, all models resulting in similar computational time. Figure 3 presents the experimental and simulated concentration profiles for each model and each material.
In this figure, it can be observed that the predicted profiles by FHADE for all materials are closer to profiles experimentally in relation to other models. It can also be verified that the profiles predicted by considered models fit slightly better to the experimental data for the sandstone than for the granite. These results are in accordance with those presented in Table 2. Furthermore, it can also be observed that the concentration profiles are in concordance with expected physically. This was already expected as the injection of the aqueous solution into the fractures occurs at \( x=0 \). The increase in concentration with increasing time occurs until the total saturation of fractures, a condition in which the concentration is equivalent to unity. In addition, it can be observed that the complete saturation of the granite fracture occurs in times longer than the sandstone fracture. This is due to greater presence of preferential paths in granite as this is a more heterogeneous material than sandstone.

Nowamooz et al. (2013) used the traditional advection-dispersion equation, the stratified medium model and the random walk model with continuous times to represent this application. As the models studied by authors are different those considered in this contribution, the qualitative comparison is not possible. The authors demonstrated that the traditional advection-dispersion model is not able to obtain good results and that the models with more parameters present better fits in relation to the experimental data. Furthermore, the profiles simulated by Nowamooz et al. (2013) present behaviors similar to...
those observed in Fig. 3. Thus, it can be concluded that, from a qualitative point of view, the results obtained in this contribution are in accordance with those obtained by Nowamooz et al. (2013).

6 CONCLUSIONS

This work proposed and solved inverse anomalous diffusion problems considering real data, a Fractional Hyperbolic Advection-Dispersion Equation and Differential Evolution. To evaluate the direct problem, the extension of classical Finite Difference Method considering the Grünwald-Letnikov Derivative was proposed. In the inverse problem context, all the phenomenological models were able to obtain good estimates for the concentration profiles in both applications. However, the Fractional Hyperbolic Advection-Dispersion Equation resulted in better values compared to the objective function due to the greater number of design variables compared to more simplified models. In this case, we conclude that a more complete model (hyperbolic) can be used to represent the mass transfer through the advection-diffusion equation.

As suggestions for future work, we can mention the formulation and resolution of inverse problems considering the insertion of uncertainties (robustness and reliability techniques).

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