



## Original article

## Fractional radioactive decay law and Bateman equations

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## ABSTRACT

The aim of this work is to develop the fractional Bateman equations, which can model memory effects in successive isotopes transformations. Such memory effects have been previously reported in the alpha decay, which exhibits a non-Markovian behavior. Since there are radioactive decay series with consecutive alpha decays, it is convenient to include the mentioned memory effects, developing the fractional Bateman Equations, which can reproduce the standard ones when the fractional order is equal to one. The proposed fractional model preserves the mathematical shape and the symmetry of the standard equations, being the only difference the presence of the Mittag-Leffler function, instead of the exponential one. This last is a very important result, because allows the implementation of the proposed fractional model in burnup and activation codes in a straightforward way. Numerical experiments show that the proposed equations predict high decay rates for small time values, in comparison with the standard equations, which have high decay rates for large times. This work represents a novelty approach to the theory of successive transformations, and opens the possibility to study properties of the Bateman equation from a fractional approach.

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## 1. Introduction

In several nuclear engineering phenomena, certain isotopes transform into other ones, which in turn undergo similar transformations in a successive way, which can be represented as:

$$X_1 \xrightarrow{\lambda_1} X_2 \xrightarrow{\lambda_2} \dots \xrightarrow{\lambda_{n-1}} X_n \quad (1)$$

where  $X_i$  and  $\lambda_i$ , with  $1 \leq i \leq n$ , denote the concentration and the decay constant of the isotope  $i$ , respectively. The mass balance equation for an element in (1) can be set as:

$$\frac{dX_i(t)}{dt} = \lambda_{i-1}X_{i-1}(t) - \lambda_iX_i(t) \quad (2)$$

For  $i = 1$ , there is no contribution from other isotopes and Eq. (2) is reduced to the radioactive decay law [1]:

$$\frac{dX_i}{dt} = -\lambda_iX_i(t) \quad (3)$$

Bateman [2] found a solution for Eq. (2) using the Laplace transform and considering the following initial conditions:

$$\begin{cases} X_1(0) \neq 0 \\ X_i(0) = 0, 2 \leq i \leq n \\ \lambda_i \neq \lambda_j \forall i, j \text{ with } 1 \leq i, j \leq n \text{ and } i \neq j \end{cases} \quad (4)$$

Such solution can be written as:

$$X_n(t) = X_1(0) \prod_{k=1}^{n-1} \lambda_k \sum_{i=1}^n \prod_{\substack{j=1 \\ j \neq i}}^n \frac{\exp(-\lambda_i t)}{\lambda_j - \lambda_i} \quad (5)$$

As it can be observed, the radioactive decay law plays an important role in the process of building the Bateman equations, because it allows to write the gain and the loss functions in the mass balance equation given in (2). It has been concluded that such law does not exhibit memory effects [3], and therefore the Bateman equations also lack of them. Nevertheless, some authors [4] suggested the opposite for the alpha decay process, considering that there are memory effects in such phenomenon, which can be simulate through fractional calculus and reformulating Eq. (3) as:

$$D_C^\alpha X_i(t) = -\lambda_i^\alpha X_i(t) \quad (6)$$

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where  $D_C^\alpha$  is the Caputo fractional derivative of order  $\alpha$ , with  $0 < \alpha < 1$ . This last equation is known as *fractional radioactive decay law*. Since there are decay chains who contain multiple and consecutive alpha decays, it is convenient to build a more general model that includes memory effects in such schemes. Following the Calik's et al. procedure [4], such effects can be introduced in the mass balance equations through fractional derivatives, leading to the formulation of the fractional Bateman equations (FBE), which are developed in the present paper.

Therefore, the aim of this work is extending the mentioned findings about the memory effects in alpha decay, to the case of successive transformations, focusing in providing a mathematical framework to build the Bateman equations from a fractional approach. Since there is a particular interest in developing analytic solutions for this fractional model, in the present work we use the Caputo's derivative definition, because it allows using initial conditions of the problems in the standard way [5].

The proposed method has two important advantages: firstly, it allows to develop analytic solutions in an analogous way in which the original Bateman equations are solved. On the other hand, except for the exponential function which is replaced by the Mittag-Leffler one, the developed solutions have an identical shape and symmetry of the integer's model. This last is very convenient from an algorithmic point of view, because it allows to implement the proposed model in burnup and activations codes in a straightforward way.

Two analytical solutions are proposed in the present work to the developed FBE: one considering that all the decay constants are different, and other allowing that some of these parameters are repeated. Numerical experiments were carried out using two different cases: a radioactive decay series who contains several consecutive alpha decays, given by  $^{238}\text{Pu} \rightarrow ^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ , and a hypothetical decay chain where there are repeated decay constants. Results show that FBE can reproduce the standard Bateman equations when the fractional order is equal to 1, and that such model exhibits high decay rates for small times, in comparison with the standard approach.

This paper is structured as follows: Section 2 contains the preliminaries of fractional calculus. The fractional Bateman equations with a single fractional order, as well as its solutions are developed in Section 3. Section 4 contains the numerical analysis, and the conclusions are given in Section 5.

## 2. Preliminaries

### 2.1. Fractional Calculus and the Caputo's derivative definition

In fractional calculus the order of the derivatives is generalized, allowing to use positive real numbers. This mathematical tool has a wide range of applications that include the modelling of biological and economical phenomena [6,7] until neurological, epidemic and non-autonomous systems process [8–10]. There are at least 15 definitions of fractional derivatives that are reported in literature [11], but in the present work the Caputo's definition will be adopted because the Laplace transform method can be extended in a straightforward way to it [12]. This last is convenient to develop analytic solutions as it will be showed in Section 3. The Caputo fractional derivative of order  $\alpha$  is defined as [13,14]:

$$D_C^{(\alpha)} \circ f(t) = \frac{1}{\Gamma(k-\alpha)} \int_0^t (t-\tau)^{k-\alpha-1} f^{(k)}(\tau) d\tau, \quad (7)$$

with  $k-1 < \alpha \leq k$  where  $\Gamma(\cdot)$  is the Gamma function. In some contexts, the  $\alpha$  number can be understood as a positive rational non-

integer number or a fraction, being this the reason that such mathematical tool takes the adjective of "fractional".

### 2.2. The Laplace transform method

Among the mathematical methods that have been used to solve differential fractional equations, one of the most important is the Laplace transform method [12], which as in the standard case, reduces the fractional differential equations to a set of algebraic equations, through the following integral transform [12]:

$$\mathcal{L}\{D_C^{(\alpha)} \circ f(t)\} = s^\alpha F(s) - \sum_{l=0}^{k-1} s^{\alpha-l-1} f^{(l)}(0) \quad (8)$$

Kexue and Jigen [15] provided the sufficient conditions to guarantee that fractional differential equations with constant coefficients can be solved through the Laplace method. Essentially, such conditions are related to the continuity of the functions as well as their property of being bounded, which will be assumed in the present work.

### 2.3. The Mittag-Leffler function

Introduced at the beginning of the last century, the Mittag-Leffler function can be considered as a generalization of the natural exponential one, and due to its importance, it has been called the "Queen of Fractional Calculus" [16,17]. Its 2-parameter version, denoted by 2PMLF, is defined as [12]:

$$E_{\alpha,\beta}(z) := \sum_{k=0}^{\infty} \frac{z^k}{\Gamma(k\alpha + \beta)}, \quad (9)$$

$\alpha, \beta, z \in \mathbb{C}$ , which is convergent always that  $Re(\alpha), Re(\beta) > 0$  [18]. When the Laplace method is used, the following relationship is crucial to solve several differential fractional systems [12]:

$$\mathcal{L}\{t^{\alpha p + \beta - 1} E_{\alpha,\beta}^{(p)}(\pm \lambda t^\alpha)\} = \frac{p! s^{\alpha - \beta}}{(s \mp \lambda)^{p+1}}, \quad (10)$$

with  $Re(s) > |\lambda|^{\frac{1}{\alpha}}$ ,  $p \in \mathbb{N}$  and where  $E_{\alpha,\beta}^{(p)}$  represents the  $p$ -integer derivative of the 2PMLF.

## 3. Fractional Bateman Equation

In certain physical systems, the properties of a given state depend not only of the present variables who described it, but also in the behavior of such variables on past states. Such property of the system is known as memory effect [19], and for the case where the states are described as functions of time  $t$ , this effect means that the system will depend on the history of the process at time, i.e., of states with  $\tau < t$ .

At first it could appear that all phenomena exhibit memory effects, but there are certain systems who cannot be described in such way. For the case of systems whose states depend on time, the absence of memory effects can be described as if they "seem not to 'age'" [20]. Therefore, in a memoryless process an item appears to 'forget' how long it has survived, which from a probability point of view can be understood as if the probability of the remained survival does not depend on past time. More generally, it is possible to conclude that, if a random variable  $X$  exhibits a memoryless behavior on a subset  $S$ , then it has the Markov property [20], which mathematically is described as:

$$P(X > a + b | X > a) = P(X > b), \quad (11)$$

where  $\forall a, b \in S$ . Eq. (11) has two important implications for the present work. Firstly, it is possible to corroborate that the radioactive decay process fulfills such property, which follows from the fact that Eq. (3) can be deduced from Eq. (11) [21]. On the other hand, applying the conditional probability formula to Eq. (11), the following equation is obtained:

$$P(X > a + b) = P(X > a)P(X > b)$$

which admits as unique solution a probability function given by  $P(X > t) = \exp(-\alpha t)$ , where  $\alpha$  is a constant [22]. Therefore, it is possible to conclude from a mathematical point of view, that the memoryless property appears in the radioactive decay law in the form of the exponential function. This last conclusion is very important, because there are some findings that suggest that there are memory effects on alpha decay [4], and therefore such phenomenon not necessarily have an exponential function in its mathematical expression.

As it is known, it is possible to compute the alpha decay half-life through theoretical models. Nevertheless, there are inconsistencies between the theoretical values and the experimental ones, being necessary to develop additional models to correct them. In one of these models, the cluster one [23], the decay constant is defined as a product of three terms: the preformation probability, the assault frequency, and the barrier penetrability. While the last two factors can be computed through theoretical methods, the first one is defined as a ratio between experimental and computed values for the half-life. In such context, a fractional alpha decay law was developed, in order to compute the decay constant without the need to use one of the factors: the preformation probability [4]. This was a very novelty and a useful procedure, which allows finding alpha decay half-lives whose values were equal to experimental ones. Therefore, it was concluded that the mentioned discrepancies between the experimental and the computed values, arise as result that the alpha decay equations, building with the exponential decay law, do not take into account the non-Markovian feature or the memory effects [4]. In such context, the present work extends that conclusion, but instead of computing decay constants, it models the isotopes concentration in successive transformations, building mass balance relationships, i.e., the Bateman equations.

### 3.1. Fractional radioactive decay law

The fractional radioactive decay law is given by [4]:

$$D_C^{(\alpha)} X(t) = -\lambda^\alpha X(t), \tag{12}$$

with  $0 < \alpha < 1$ . Applying Eq. (8) on both sides, it follows that:

$$s^\alpha \tilde{x}(s) - s^{\alpha-1} X(0) = -\lambda^\alpha \tilde{x}(s), \tag{13}$$

where  $\tilde{x} = \mathcal{L}\{X(t)\}$ . The last equation can be rewritten as:

$$\tilde{x}(s) = X(0) \frac{s^{\alpha-1}}{s^\alpha + \lambda^\alpha} \tag{14}$$

and according to Eq. (10), the solution for the last system is given by:

$$X(t) = X(0) E_{\alpha,1}(-\lambda^\alpha t^\alpha) \tag{15}$$

Starting from Eqs. (12) and (15), it is possible to develop a “fractional” mass balance equation of the problem of successive transformations given in (1), which can be solved with the Laplace transform method. Two different cases will be considered for such task:

- 1) Case 1: all decay constants are different,
- 2) Case 2: there are repeated decay constants

### 3.2. Case 1: different decay constants

Using a single fractional order  $\alpha$ , Eq. (2) has the following equivalent fractional formulation:

$$D_C^{(\alpha)} X_i(t) = \lambda_{i-1}^\alpha X_{i-1}(t) - \lambda_i^\alpha X_i(t), \tag{16}$$

with  $1 \leq i \leq n$ ,  $\lambda_0^\alpha X_0 = 0$ . Applying Eq. (8), such equation can be written as:

$$s^\alpha \tilde{x}_i(s) - s^{\alpha-1} X_i(0) = \lambda_{i-1}^\alpha \tilde{x}_{i-1}(s) - \lambda_i^\alpha \tilde{x}_i(s) \tag{17}$$

with  $\tilde{x}_i = \mathcal{L}\{X_i(t)\}$ . Considering the initial conditions given by  $X_i(t=0) = 0, i = 2, 3, \dots, n$ , Eq. (17) can be written in an expanded way as:

$$\begin{cases} \tilde{x}_1(s) = \frac{s^{\alpha-1} X_1(0)}{s^\alpha + \lambda_1^\alpha} \\ \tilde{x}_2(s) = \frac{\lambda_1^\alpha \tilde{x}_1}{s^\alpha + \lambda_2^\alpha} \\ \vdots \\ \tilde{x}_n(s) = \frac{\lambda_1^\alpha \lambda_2^\alpha \dots \lambda_{n-1}^\alpha \tilde{x}_{n-1}}{s^\alpha + \lambda_n^\alpha} \end{cases}$$

It is possible to observe in the last system, that the solution to the term  $i$  can be obtained through the solution of the term  $i - 1$ . Therefore, after multiple replacements, it is possible to write:

$$\tilde{x}_n(s) = X_1(0) \prod_{i=1}^{n-1} \lambda_i^\alpha s^{\alpha-1} \prod_{k=1}^n \frac{1}{(s^\alpha + \lambda_k^\alpha)}, \tag{18}$$

Using partial fraction decomposition, the right product can be expressed as a sum:

$$\prod_{k=1}^n \frac{1}{(s^\alpha + \lambda_k^\alpha)} = \sum_{k=1}^n \frac{c_k}{(s^\alpha + \lambda_k^\alpha)}, \tag{19}$$

with  $c_k = \prod_{\substack{j=1 \\ j \neq k}}^n \frac{1}{(\lambda_j^\alpha - \lambda_k^\alpha)}$

Through the last steps, the solution of the system given in (16) is reduced to find the following inverse Laplace transform:

$$X_1(0) \prod_{i=1}^{n-1} \lambda_i^\alpha \sum_{k=1}^n c_k \mathcal{L}^{-1} \left\{ \frac{s^{\alpha-1}}{s^\alpha + \lambda_k^\alpha} \right\}$$

which, according to Eqs. (10) and (19) is equal to:

$$X_n^F(t) = X_1(0) \prod_{k=1}^{n-1} \lambda_k^\alpha \sum_{i=1}^n \prod_{\substack{j=1 \\ j \neq i}}^n \frac{E_{\alpha,1}(-\lambda_i^\alpha t^\alpha)}{(\lambda_j^\alpha - \lambda_i^\alpha)} \tag{20}$$

where the upper index  $F$  is used to distinguish such fractional solution of the standard one, that was given in Eq. (5). Eq. (20) will be called the Fractional Bateman Equation (FBE), which has an identical shape that Eq. (5). In fact, their only differences are the presence of the 2PMLF, instead of the exponential function, as well as

the fractional exponent in the decay constants and on the time variable. Also, it is possible to observe that the FBE can reproduce the standard Bateman equation, in fact:

$$\begin{aligned} \lim_{\alpha \rightarrow 1} X_n^F(t) &= X_1(0) \prod_{k=1}^{n-1} \lambda_k \sum_{i=1}^n \prod_{j=1}^n \frac{E_{1,1}(-\lambda_i t)}{(\lambda_j - \lambda_i)} \\ &= X_1(0) \prod_{k=1}^{n-1} \lambda_k \sum_{i=1}^n \prod_{\substack{j=1 \\ j \neq i}}^n \frac{\exp(-\lambda_i t)}{(\lambda_j - \lambda_i)} = X_n(t) \end{aligned}$$

where it was used that  $E_{1,1}(x) = \exp(x)$  in the last step [5]. Therefore, the FBE is a more general solution to the problem of successive transformations. As in the case of the standard Bateman equation, the FBE is also symmetrical, if it is considered as a function of the decay constants arguments. In other words, the following property is valid:

$$\begin{aligned} X_n^F(\lambda_1^\alpha, \lambda_2^\alpha, \dots, \lambda_l^\alpha, \dots, \lambda_m^\alpha, \dots, \lambda_n^\alpha) \\ = \\ X_n^F(\lambda_1^\alpha, \lambda_2^\alpha, \dots, \lambda_m^\alpha, \dots, \lambda_l^\alpha, \dots, \lambda_n^\alpha) \end{aligned} \tag{21}$$

This last equation implies that the function has the same value, independently of the order in which the isotopes that are prior to the isotope  $n$  appear (considering the same initial concentration). Such property is very important in the integer order case, to overcome the numerical difficulties related to the differences between decay constants of short-lived nuclides [24], and it is advantageous that the FBE also has such property. Finally, the similarities between the shape of the FBE with the standard Bateman Equation, imply that an algorithm that is used to solve the Bateman equation can be extended, in a straightforward way, to solve the fractional model, being only necessary to include the mentioned differences.

### 3.3. Case 2: repeated decay constants

It was assumed that all the decay constants were different when Eq. (5) was obtained. It is possible to eliminate such limitation, removing the third line in (4), which generates a more general solution to the Bateman equations. For such task it is necessary to assume that, of the total  $n$  decay constants, only  $m$  are different. Such decay constants will be denoted by  $\Lambda_j, 1 \leq j \leq m$ , and following a similar procedure to the one given in the last section, the Bateman solution that admits repeated decay constants is given by [25]:

$$\begin{aligned} X_n^R(t) &= X_1(0) \prod_{i=1}^{n-1} \lambda_i \sum_{j=1}^m \sum_{h=1}^{r_j} B_{j,r_j-(r_j-h)} t^{h-1} \frac{\exp(-\Lambda_j t)}{(h-1)!} \\ \text{where } B_{j,r_j-u} &= \frac{1}{u!} \lim_{s \rightarrow -\Lambda_j} \frac{d^u}{ds^u} \prod_{c=1}^m \frac{1}{(s + \Lambda_c)^{r_c}} \end{aligned} \tag{22}$$

In Eq. (22), the super index  $R$  is used to denote “repeated”. In the present section, a similar formula will be developed to the fractional case. Starting from Eq. (18), where no assumptions about the decay constant have been carried-out, it is possible to group the terms that have the same decay constant in the following way:

$$\tilde{x}_n(s) = X_1(0) \prod_{i=1}^{n-1} \lambda_i^\alpha \prod_{j=1}^m \frac{s^{\alpha-1}}{(s^\alpha + \Lambda_j^\alpha)^{r_j}} \tag{23}$$

where  $r_j - 1$  represents the number of times that the decay

constant  $\Lambda_j$  is repeated. Unlike the Case = 1, Eq. (23) cannot be expressed in simple-root partial fractions decomposition. In this case it is necessary to use the following identity [25]:

$$\begin{aligned} \prod_{j=1}^m \frac{1}{(s^\alpha + \Lambda_j^\alpha)^{r_j}} &= \sum_{j=1}^m \sum_{h=1}^{r_j} \frac{A_{j,r_j-(r_j-h)}}{(s^\alpha + \Lambda_j^\alpha)^h} \\ \text{where} \\ &= \frac{1}{u!} \lim_{s \rightarrow -\Lambda_j^\alpha} \frac{d^u}{ds^u} \prod_{c=1}^m \frac{1}{(s^\alpha + \Lambda_c^\alpha)^{r_c}} \end{aligned} \tag{24}$$

Therefore, the solution of (24) can be found through the following expression:

$$\mathcal{L}^{-1} \left\{ \tilde{x}_k \right\} = X_1(0) \prod_{i=1}^{n-1} \lambda_i^\alpha \sum_{j=1}^m \sum_{h=1}^{r_j} A_{j,r_j-(r_j-h)} \mathcal{L}^{-1} \left\{ \frac{s^{\alpha-1}}{(s^\alpha + \Lambda_j^\alpha)^h} \right\}$$

Using (10), it follows that:

$$X_n^{FR}(t) = X_1(0) \prod_{i=1}^{n-1} \lambda_i^\alpha \sum_{j=1}^m \sum_{h=1}^{r_j} A_{j,r_j-(r_j-h)} \times t^{\alpha(h-1)} \frac{E_{\alpha,1}^{(h-1)}(-\Lambda_j^\alpha t^\alpha)}{(h-1)!} \tag{25}$$

where the upper index,  $FR$ , is used to denote “Fractional Repeated”. It is possible to prove that Eq. (25) is reduced to Eq. (22), when the fractional order is equal to 1. For such task, it is necessary to observe the following relationships, which can be proved using the derivative chain rule:

$$\begin{aligned} \frac{d^{h-1}}{dx^{h-1}} (e^{xt}) &= t^{h-1} (e^{xt}), \\ \frac{d^{h-1}}{dx^{h-1}} (E_{\alpha,1}(xt^\alpha)) &= t^{\alpha(h-1)} E_{\alpha,1}^{(h-1)}(xt^\alpha) \end{aligned} \tag{26}$$

Using them, it is possible to conclude that  $X_n^{FR}(t)$  and  $X_n^R$ , have the same shape, as in the Case 1, being the unique difference that the exponential function was replaced by the Mittag-Leffler one. This has a very important meaning related to Eq. (11). As it was discussed before, the memoryless property of the phenomenon was contained in the exponential function. Nevertheless, if memory effects are considered, it would expect that such function will be replaced by a generalized one, which is precisely the 2PLMF. Therefore, as in the past case, this similarity allows to use an algorithm that originally was designed to solve the Bateman equations with repeated decay constants, to solve the equivalent fractional model. On the other hand, the  $p$  derivative of  $E_{\alpha,\beta}(z)$  can be computed using the following relationship [26]:

$$\frac{d^p}{dz^p} E_{\alpha,\beta}(z) = \sum_{k=0}^{\infty} \frac{(p+k)! z^k}{k! \Gamma(\alpha(p+k) + \beta)} = p! E_{\alpha,\alpha p + \beta}^{p+1}(z), \tag{27}$$

where  $E_{\alpha,\beta}^\gamma(z)$  represents the 3-Parameter Mittag-Leffler Function (3PMLF) [27,28], which is a further generalization of Eq. (9). According to Fernandez and Husain [29], such expression is locally uniformly convergent for any  $z \in \mathbb{C}$ , and  $Re(\alpha) > 0$ .

An important aspect in the developing of fractional models is related to determine a proper value of  $\alpha$ , i.e., the fractional order. At

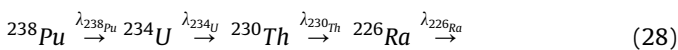
the present stage of the research, such task will not be addressed, limiting the present study to the mathematical and numerical framework of the FBE, being the experimental part a matter of a future work. It is worth mentioning that the most common decay schemes that appear in nuclear engineering and activation problems were considered in the present work, which are known as linear chains and for which there are analytic solutions. Nevertheless, there are more complicated structures known as cyclic chains, for which such analytic solutions do not exist, being necessary to approximate them using numerical methods [30]. Fortunately, there is a continuous progress in the numerical techniques to solve fractional differential equations, and particularly in the ones that are based in the Caputo's derivative [31–33]. On the other hand, there also have been several developments in the generalization of the Mittag-Leffler function [34,35], which can help to express the developed equations in a different way, reducing their complexity as it was the case of the derivatives in Eq. (27), which was reduced using the 3PMLF. Nevertheless, such task will be addressed in future stages of this research.

#### 4. Numerical experiments

The calculations were performed on a 3.5 GHz Intel i7 processor, 6700 HQ, under a 64-bit Windows operating system, using the Python 3.8 programming language. The Decimal and the SciPy libraries were used, the first one to guarantee a precision of 20 digits and the second one for computing the Gamma function. For the case of the Mittag-Leffler function and its derivatives, a direct programming of Eqs. (9) and (27) was carried-out, as it is showed in the Algorithm 1 in Appendix A, where the infinite in the upper limit was replaced by a value of 1000. Even when such number represents a huge computational-time, it is adequate for the moment because it provides a proper precision with the purpose to make adequate comparisons.

##### 4.1. Case 1: different decay constants

Since the mentioned findings about the memory effects are related only to alpha decay, it is adequate to consider a chain where only this type of decay appears. For this case, a range of time from 1 to 70,000 days was used, because this is a typical range in nuclear chronometry [36]. Therefore, for Case 1, the following scheme is proposed:



The corresponding values of the decay constants are listed in Table 1. For comparative purposes, a normalized unitless initial concentration equal to 1 will be considered for  ${}^{238}\text{Pu}$ . The results for the four isotopes are given in Fig. 1. For  ${}^{238}\text{Pu}$ , the Bateman equation is reduced to

$$X_1(t) = X_{238\text{Pu}}(0) \exp(-\lambda_{238\text{Pu}} t) = \exp(-\lambda_{238\text{Pu}} t)$$

and for the fractional model:

$$X_1^F(t) = X_{238\text{Pu}}(0) E_{\alpha,1}(-\lambda_{238\text{Pu}}^\alpha t^\alpha) = E_{\alpha,1}(-\lambda_{238\text{Pu}}^\alpha t^\alpha)$$

Then, the first comparison is reduced to compare the exponential function with the Mittag-Leffler one. As it can be observed in Fig. 1, in the fractional model the reduction in the concentration is faster than the standard case, at least for the first 3000 days. Nevertheless, from a certain point the tendency is reverted, and the reduction is slower than, which is the reason that there is a crossing

**Table 1**  
Decay constants of the isotopes belonging to (28).

$\lambda$	(seconds) <sup>-1</sup>
$\lambda_{238\text{Pu}}$	$2.5062204 \times 10^{-10}$
$\lambda_{234\text{U}}$	$8.9529747 \times 10^{-14}$
$\lambda_{230\text{Th}}$	$2.9158334 \times 10^{-13}$
$\lambda_{226\text{Ra}}$	$1.3737220 \times 10^{-11}$

between the graphs. On the other hand, the growth in the concentration for  ${}^{234}\text{U}$  is faster than in the standard case, at least for the first 3000 days. This coincides with the behavior that was observed for the preceding isotope,  ${}^{238}\text{Pu}$ , whose decay was faster. Since the decay of this last isotope led to the creation of  ${}^{234}\text{U}$ , it is expected a greater concentration.

Starting from 3000 days, the tendency is reverted, which as it was discussed before, is perfectly coherent with the behavior of the Mittag-Leffler function. For the graphs of  ${}^{230}\text{Th}$  and  ${}^{226}\text{Ra}$ , it is possible to observe significative differences between the fractional model and the standard Bateman one. Such differences can be explained in terms of subtractions and products between decay constants, which are more dominant that in the past isotopes. In order to exemplified such behavior, the values of  $X_{230\text{Th}}(t_{1000}) = 1000$  days) and  $X_{230\text{Th}}^F(1000$  days) will be compared. Firstly, it will be considered the following expression that appears in the standard Bateman equations given in (5):

$$\prod_{j=1}^3 \frac{\exp(-\lambda_j t_{1000})}{(\lambda_j - \lambda_i)} \approx \begin{cases} 1.5603364615 \times 10^{19} & i = 1 \\ 1.97544990700 \times 10^{22} & i = 2 \text{ (seconds)}^2 \\ -1.9770098728 \times 10^{22} & i = 3 \end{cases} \quad (29)$$

where  $\lambda_1, \lambda_2$  and  $\lambda_3$  correspond to the first three isotopes shown in (28). In Eq. (5), the expression (29) is multiplied by the factor  $\lambda_1 \cdot \lambda_2 = 2.2438142 \times 10^{-23} \text{ seconds}^2$ , after which it follows that  $X_{230\text{Th}}(t_{1000}) \approx 8.314769492319 \times 10^{-8}$ . In the fractional case, all the last terms depend on the fractional order, but it is more notorious such dependence in the factor  $\lambda_1^\alpha \cdot \lambda_2^\alpha$ . In order to show that, the following ratios will be defined:

$$R(\alpha) = \sum_{i=1}^3 \prod_{j=1}^3 \frac{E_{\alpha,1}(-\lambda_j^\alpha t_{1000}^\alpha)}{(\lambda_j^\alpha - \lambda_i^\alpha)} \div \sum_{i=1}^3 \prod_{j=1}^3 \frac{\exp(-\lambda_j t_{1000})}{(\lambda_j - \lambda_i)},$$

$$P(\alpha) = \frac{\lambda_{238\text{Pu}}^\alpha \cdot \lambda_{234\text{U}}^\alpha}{\lambda_{238\text{Pu}} \cdot \lambda_{234\text{U}}},$$

$$L(\alpha) = \frac{X_{234\text{U}}^F(t_{1000})}{X_{234\text{U}}(t_{1000})} \quad (30)$$

It follows that  $R(\alpha) \cdot P(\alpha) = L(\alpha)$ . Fig. 2 shows the behavior of such ratios as a function of the fractional order. As it can be observed, the ratio  $P(\alpha)$  decreases at a higher rate than  $R(\alpha)$ , which implies that for small fractional orders, the fractional solution will exceed the standard solution in a greater order of magnitude. Therefore, unlike the past two cases, where the Mittag-Leffler function has an important effect on the solution, leading to “crosses” in the graph, in this case this effect is overshadowed and surpassed by the effect of  $R(\alpha)$  and  $P(\alpha)$ .



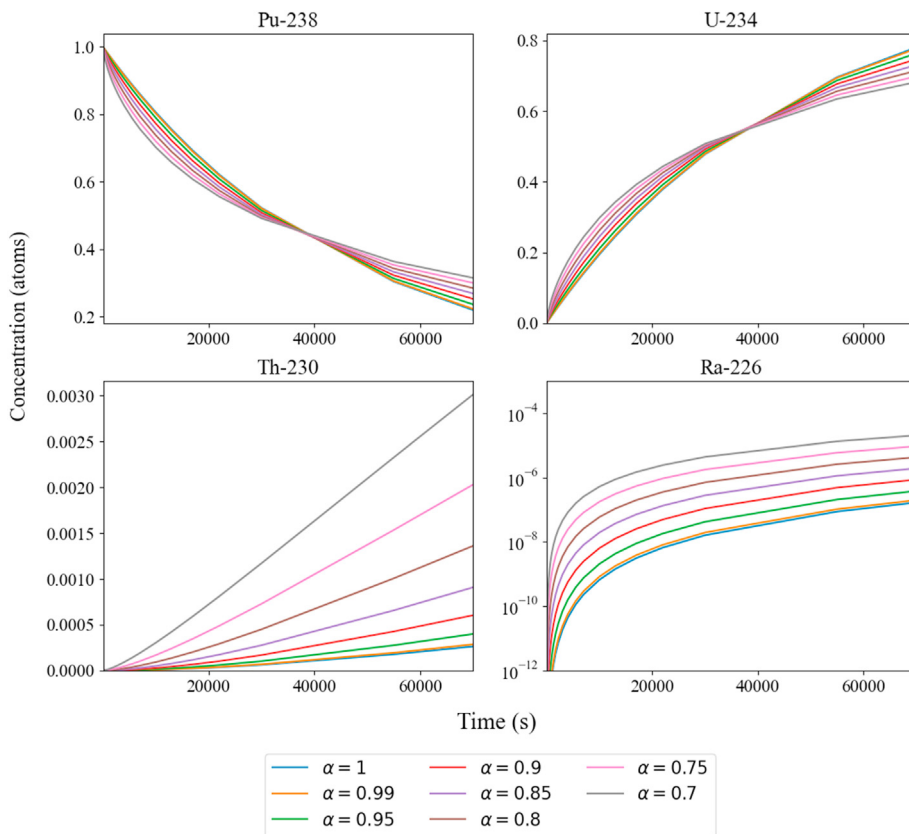
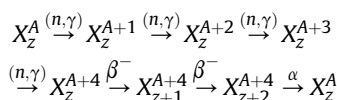


Fig. 1. Normalized Concentration vs time, for the isotopes given in (28).

4.2. Case 2: repeated decay constants

There are two situations where repeated decay constants appear in (1): when there are repeated isotopes in a chain, and when there are two different isotopes with the same decay constant. The first situation is related to the concept of cyclic chains [30], for example:



The second situation, where two different isotopes have the same value for their decay constants, is common for half-lives that are estimated with theoretical considerations [37]. As it was mentioned before, it is adequate to carry-out numerical experiments using decay chains where only the alpha decay appears. Nevertheless, it is not possible to propose a real decay chain whose fulfills some of the mentioned conditions. Nonetheless, from a mathematical point of view, it is useful to propose a hypothetical decay scheme to carry-out comparisons, which will be based in the case that was studied by Dreher [37]. In the present work, such scheme will be reduced to four different isotopes, with only two different decay constants:



The initial concentration for the first isotope will be set equal to  $6.023 \times 10^{23}$  atoms, and the values of the proposed decay constants will be given by  $\lambda_1 = \lambda_2 = \ln(2)/2 \text{ seconds}^{-1}$ , and  $\lambda_3 = \lambda_4 = \ln(2)/3 \text{ seconds}^{-1}$ . A time interval from 0.1 to 10 s will be used, which is convenient to appreciate the behavior of the solutions. The

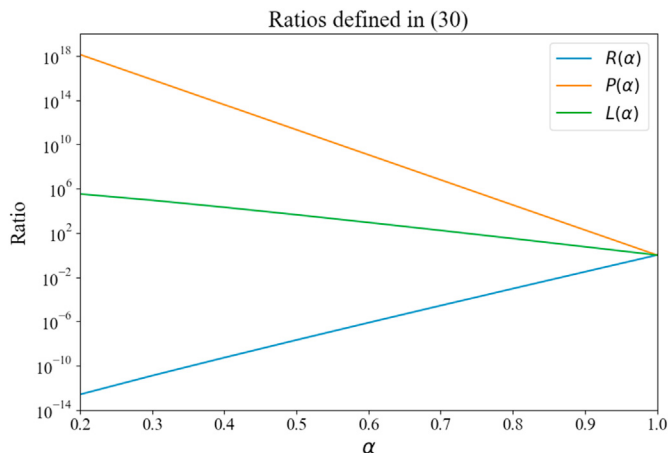


Fig. 2. Numerical Analysis of the ratios defined in (30).

standard and the fractional solutions for the first two isotopes in (31) are lack of interest for comparative purposes, because for the first case there are not repeated isotopes before the first position, and for the second one the solution is very similar to the exponential case. Therefore, only the concentration of the last two isotopes will be studied. The results of such isotopes are given in Fig. 3.

For the isotope  $X_3$ , the concentration of the fractional case is greater than the integer order one, for the time interval given between 0.1 s and 1 s. Such behavior is very similar to the one that was described for  $^{234}\text{U}$  in Fig. 1. Unlike the isotopes of  $^{230}\text{Th}$  and  $^{226}\text{Ra}$ , in this case there are not problems about the magnitude order of the results, which can be explained considering that the decay

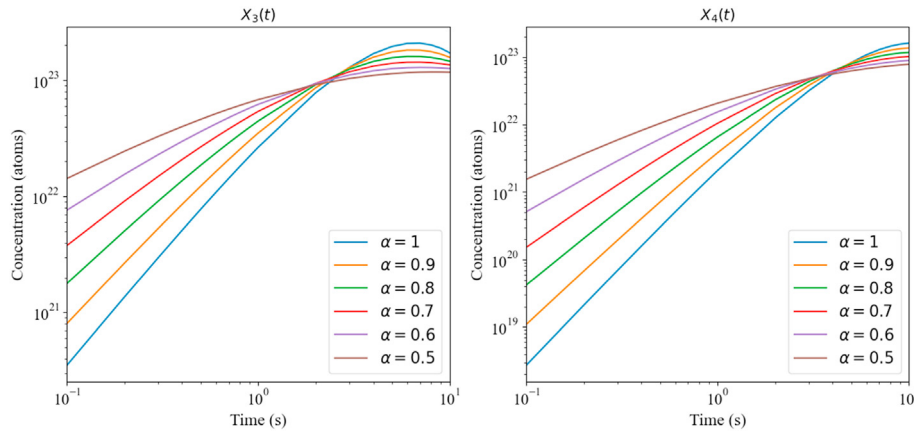


Fig. 3. Concentration of the isotopes  $X_3$  and  $X_4$ , given in (31).

constants do not differ significantly in the hypothetical decay chain.

For  $X_4$  there are two different decay constants in the associated decay chain, each of them appearing two times. The behavior of the graph is similar to the ones that were described for  $X_3$ . It is possible to observe that the point where the graphs are crossing is moved to the right, which corresponds to the position where the isotope appears in the chain given in (31). Essentially, it is possible to observe that the fractional model can describe in a proper way the behavior of the successive transformations, and as the value of the fractional order tends to 1, the fractional model tends to the standard Bateman one.

### 5. Conclusions

The aim of this work was developed a more general formulation of the Bateman equations, which was carried out in a satisfactory way. This new approach considers the possibility of including memory effects through the use of fractional calculus. Using the Laplace transform method, it was possible to find two solutions to the developed fractional model: one considering that all the decay constants were different, and other one where repeated decay constants appear. As a notable result, the developed equations have the same mathematical shape that the standard ones, with only two differences: the proposed model contain the Mittag-Leffler function and its derivatives, instead of the exponential one, and that the decay constant and time variable have the fractional order as an exponent.

Due to the mentioned similarity between the standard and the fractional model, it is possible to extend several properties of the original Bateman equation to the fractional one. Through numerical experiments it was possible to conclude that the loss rate in the fractional model is faster than the standard one, at least for a first time's interval, then such tendence is reverted for large times. Finally, the results that were obtained from the fractional models show a huge potential for describing memory effects, which were traditionally ignored in the decay process models, representing an effective way to generalize the Bateman equations for successive transformations, as well as to study their properties from a fractional approach.

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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### Appendix A

#### A.1 Pseudo code for the 2-parameter Mittag-Leffler function

A.1 Pseudo code for the 2-Parameter Mittag-Leffler Function	
1	INPUT $z, \alpha, \beta, approx.$
2	OUTPUT $E_{\alpha, \beta}(z)$
3	$c = 0$
4	FOR $i = 0, 1, 2, \dots, approx :$
5	$c = c + z^i / \Gamma(\alpha \cdot i + \beta)$
6	RETURN $c$

#### A.2 Algorithm for fractional Bateman Equation

A.2 Algorithm for the fractional Bateman Equation	
1	INPUT $n, t, X_0, \alpha,$ $vec\_decay = [\lambda_1, \lambda_2, \dots, \lambda_n]$
2	OUTPUT $X_n^\alpha(t)$
3	$p = 1, s = 0, l = 1$
4	IF $n \neq 1 :$
5	FOR $i = 0, 1, \dots, (n - 1) :$
6	$p = p * \lambda_i^\alpha$
7	ELSE
8	$p = 1$
9	FOR $k = 1, 2, \dots, n$
10	FOR $j = 1, 2, \dots, n$ AND $j \neq k:$
11	$l = l \cdot (1 / (\lambda_j^\alpha - \lambda_k^\alpha))$
12	$s = s + E_{\alpha, \beta}(-\lambda_k^\alpha t^\alpha) \cdot l$
13	$X_n^\alpha(t) = X_0 \cdot p \cdot s$

A.3 Explicit expressions for equations (22) and (25) for case  $n = 3$  and  $n = 4$

For case  $n = 3$ :

$$X_3(t) = X_1(0)\lambda_1^2 \sum_{j=1}^2 \sum_{h=1}^{r_j} B_{j,r_j-(r_j-h)} \times \frac{t^{h-1}}{(h-1)!} \exp(-\lambda_j t)$$

Since  $r_1 = 2, r_2 = 1$ , it follows that:

$$X_3(t) = X_1(0)\lambda_1^2 \times \left( B_{1,2-(1)} \exp(-\lambda_1 t) + B_{1,2-(0)} t \exp(-\lambda_1 t) + B_{2,1-(0)} \exp(-\lambda_2 t) \right)$$

Using Eq. (24), the coefficients  $B_{1,2-(1)}, B_{1,2-(0)}$  and  $B_{2,1-(0)}$  and reducing these terms, leads to:

$$X_3(t) = X_1(0)\lambda_1^2 \times \left( -\frac{\exp(-\lambda_1 t)}{(\lambda_2 - \lambda_1)^2} + \frac{t \exp(-\lambda_1 t)}{\lambda_2 - \lambda_1} + \frac{\exp(-\lambda_2 t)}{(\lambda_1 - \lambda_2)^2} \right) \tag{A.1}$$

Using the same procedure, it follows that:

$$X_4(t) = X_1(0)\lambda_1\lambda_2^2 \times \left( \frac{t(\exp(-\lambda_1 t) + \exp(-\lambda_2 t))}{(\lambda_2 - \lambda_1)^2} - 2 \frac{\exp(-\lambda_1 t) - \exp(-\lambda_2 t)}{(\lambda_2 - \lambda_1)^3} \right) \tag{A.2}$$

Similarly, for the fractional case given in equation (25), under an equivalent procedure it follows:

$$X_3^\alpha(t) = X_1(0)(\lambda_1^\alpha)^2 \times \left( -\frac{E_{\alpha,1}(-\lambda_1^\alpha t^\alpha)}{(\lambda_2^\alpha - \lambda_1^\alpha)^2} + \frac{t^\alpha E'_{\alpha,1}(-\lambda_1^\alpha t^\alpha)}{\lambda_2^\alpha - \lambda_1^\alpha} + \frac{E_{\alpha,1}(-\lambda_2^\alpha t^\alpha)}{(\lambda_1^\alpha - \lambda_2^\alpha)^2} \right) \tag{A.3}$$

and:

$$X_4^\alpha(t) = X_1(0)\lambda_1^\alpha(\lambda_2^\alpha)^2 \times \left( \frac{t^\alpha (E'_{\alpha,1}(-\lambda_1^\alpha t^\alpha) + E'_{\alpha,1}(-\lambda_2^\alpha t^\alpha))}{(\lambda_2^\alpha - \lambda_1^\alpha)^2} - 2 \frac{E_{\alpha,1}(-\lambda_1^\alpha t^\alpha) - E_{\alpha,1}(-\lambda_2^\alpha t^\alpha)}{(\lambda_2^\alpha - \lambda_1^\alpha)^3} \right) \tag{A.4}$$

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