

A Numerical Approach For Determining Interactivity: A Case Study in Chemical Degradation

* Peter Sussner ^a and Vinícius Wasques ^{b,c} and Julio Cesar Fernandes ^d and Estevão Esmi ^a

^a Dept. of Applied Mathematics, University of Campinas, Campinas, SP, Brazil, sussner@unicamp.br

^b Dept. of Mathematics, São Paulo State University, Rio Claro, SP, Brazil, vwasques@outlook.com

^c Int. Sci. Teach. Center, National Center for Research in Energy and Materials, Campinas, SP, Brazil

^d Inst. of Ed. Sci., Fed. Univ. of Western Pará, PA, Brazil, julio.fernandes@ufopa.edu.br

^a Dept. of Applied Mathematics, University of Campinas, Campinas, SP, Brazil, eesmi@unicamp.br

Abstract

Interactivity naturally occurs in many real-world problems. The main goal of this paper is to propose a first approach toward characterizing the interactivity that is inherently present in the derivative of an unknown interval or fuzzy number valued function that is subject to certain restrictions. A series of experiments regarding isothermal degradation of L-ascorbic acid tablets serves as an important case study for our approach.

Keywords: Interval valued function, sup- J extension principle, interactive fuzzy arithmetic, fuzzy Euler method, L-ascorbic acid, chemical degradation.

1 Introduction

Fuzzy derivatives of fuzzy number valued functions are a very active research topic in academia. Among the concepts of fuzzy derivative in the literature, we chose to consider the concept of interactive derivative which is particularly interesting and applicable. On the one hand, the concept of interactive derivative encompasses other concepts of fuzzy derivative, namely the generalized and the generalized Hukuhara derivative of a fuzzy number valued function [13]. On the other hand, interactivity is certainly a natural phenomenon that can be observed in the development of market shares of large competing companies (e.g., in the airline sector) and in the quantities of reagents in a chemical solution. Roughly speaking, in a fuzzy dynamic system, the drift of the trajectory of the fuzzy solution is described by the corresponding fuzzy differential equation. The evolution of the uncertainty around the trajectory is associated with the existence of a certain local interactivity between the present state and the near future. Thus, the specificity of the obtained solution at every time, that can be measured by means of

the length of the α -levels, is tied to the choice of this local interactivity.

Most numerical methods for fuzzy initial value problems (FIVPs) are based on characterizing a notion of the fuzzy derivative in terms of α -cuts. Thus, using this characterization, an FIVP is converted into a classical system of initial value problems for every $\alpha \in [0, 1]$ and then some crisp numerical method is employed to determine the endpoint functions of the α -cuts of the fuzzy solution. A challenge when using this approach consists in verifying the hypotheses of the well-known Stacking Theorem [9], which establishes conditions for a family of intervals to be the α -cuts of some fuzzy number. In contrast, numerical methods for FIVPs that are based on an interactive arithmetic are guaranteed to produce a fuzzy number at each step.

This paper addresses the following questions:

1. How can one characterize the interactivity observed in the evolution of real-world data?
2. Can one provide some experimental evidence that interactivity occurs in an interval or fuzzy number valued function that describes a real-world phenomenon?
3. To what extent does interactivity occur?

To tackle these problems, we consider a family of parametrized numerical solutions of a fuzzy initial value problem (FIVP) that describes the degradation of L-ascorbic acid under isothermal stress [11]. To this end, we applied an interactive Euler Method that depends on the choice of an interactive fuzzy arithmetic associated with a parametrized possibility distribution [5]. The corresponding parameter indicates a degree of interactivity that is present in the evolution of amounts of L-ascorbic acid, modelled in terms of epistemic intervals that arise from experimental data and expert knowledge, from an initial time t_0 to a final time t_f .

In order to collect all data, L-ascorbic tablets were exposed to various temperatures and the percentage of undegraded ascorbic acid was measured at different times. In solid state, one can assume that thermal degradation of ascorbic acid follows an exponential rate, similar to what occurs with the decomposition of polymers [10]. Given a crisp initial value, the rate of degradation can be determined exactly if a final value is at hand. However, several sources of uncertainty exist in the data: Reactions in solid state are very complex. Furthermore, many processes occur simultaneously or sequentially such as melting, sublimation, polymorphism, chemical transformation or degradation [7]. Moreover, the amount of ascorbic acid in a tablet varies to a certain extent if the tablets are manufactured according to pharmaceutical tables [4]. In this paper, we employ (epistemic) intervals to cope with these inherent sources of uncertainty. Thus, given an interval $[\underline{a}_0, \bar{a}_0]$ corresponding to the initial percentage of ascorbic acid at time t_0 , the evolution over time of the uncertain percentages of ascorbic acid can be described in terms of a fuzzy initial value problem (FIVP) if one has the fuzzy derivative of this function as well as the underlying type of the fuzzy derivative at hand. If $[\underline{a}_f, \bar{a}_f]$ is an interval that corresponds to the final percentage of ascorbic acid at time t_f , then the rate of degradation k can be estimated by using a function of the form $\exp(-kt)$ to interpolate the nodes $(t_0, \frac{a_0 + \bar{a}_0}{2})$ and $(t_f, \frac{a_f + \bar{a}_f}{2})$. However, it is unclear how to select an appropriate concept of a fuzzy derivative. Thus, since the drift the trajectory is given by a differential equation and we focus on interactive fuzzy derivatives, the selection of an appropriate fuzzy derivative consists in determining a certain local interactivity such that the corresponding solution embraces and adjusts to the available imprecise and uncertain data of the phenomenon under consideration.

In this paper, we attempt to remedy this situation by considering the interactive arithmetic based on a parametrized family of joint possibility distributions J_γ , where $\gamma \in [0, 1]$ [5]. For every parameter $\gamma \in [0, 1]$, one can formulate a numerical method such as the γ -interactive fuzzy Euler method, i.e., the one that employs a J_γ based interactive arithmetic for some $\gamma \in [0, 1]$, in order to generate a numerical solution to an FIVP with a fuzzy initial value such as $[\underline{a}_0, \bar{a}_0]$. Recent studies and computational simulations performed by the authors of [5] indicate that the γ -interactive fuzzy Euler method converges, under some mild conditions, to the solution of an FIVP for some concept of an interactive fuzzy derivative as $h \rightarrow 0$. After selecting a small step size h , the approach introduced in this paper estimates an appropriate interactivity parameter γ by approximating the parameter γ that yields the length of

the interval $[\underline{a}_f, \bar{a}_f]$ at time t_f .

The paper is organized as follows: In the next section, we present a brief description of the investigations on isothermal degradation of L-ascorbic acid tablets and the initial value problems that arise from these investigations if one has a crisp initial value at hand and a crisp estimate of the rate of degradation at hand. Then we explain several sources of uncertainty (due to measurement errors and errors in the fabrication of L-ascorbic acid tablets [4]) and how to deal with these sources of uncertainty using intervals, i.e., special cases of fuzzy numbers. Section 3 reviews the concept of a joint possibility distribution (JPD), in particular addressing the parametrized family of JPDs J_γ , of fuzzy numbers and how it can be applied to define an interactive fuzzy arithmetic and a fuzzy Euler method. Section 4 presents our approach toward determining an appropriate interactivity parameter γ such that, for any given $w \geq 0$, the width of the resulting fuzzy number at time t_f is approximately equal to w in an application of a J_γ based fuzzy Euler method of a certain form within a time interval $[t_0, t_f]$. In Section 5, we apply this approach to the aforementioned investigations on ascorbic acid degradation under isothermal stress. We finish with some concluding remarks.

2 A Brief Description of Some Kinetic Studies on Isothermal Degradation of L-Ascorbic Acid

L-ascorbic acid in solid state degrades exponentially over time during thermal decomposition. Formally, we have $a(t) = a_0 \cdot e^{-k(t-t_0)}$, where $a_0 \in \mathbb{R}$ is the amount of L-ascorbic acid at time t_0 and $k > 0$ is the degradation constant. This can be seen by considering the following initial value problem (IVP):

$$\begin{cases} a' &= -ka, \\ a(t_0) &= a_0 \end{cases} \quad (2.1)$$

The higher the temperature to which L-ascorbic acid is exposed, the faster its degradation, i.e., the larger the constant k [1]. However, the constant k is generally unknown and needs to be determined experimentally. In theory, the constant k can be exactly determined if one has a_f , i.e., the value of a at a time $t_f > t_0$ at hand. Specifically, k can be chosen so that $a_0 \cdot e^{-k(t-t_0)}$ interpolates (t_f, a_f) .

In practice, diverse sources of uncertainty diminish the applicability of this approach. In some previous experiments [11], individual tablets containing L-ascorbic acid were exposed to a number of different but constant temperatures. Each tablet was fabricated according to an elapsed patent [8] so as to contain $92.49 \pm 5\%$

of L-ascorbic acid. Degradation of solid compounds follows a first-order reaction when subjected to temperatures that are close to their melting points [12]. Therefore, there is a time delay before the exponential degradation of L-ascorbic acid sets in. For these reasons, we considered a time of thirty minutes of exposure of the tablets to a certain temperature as the initial time t_0 in this paper. We are interested in the percentage of L-ascorbic acid contained in a tablet at times $t_0 = 30$ and $t_f = 90$ which corresponds to ninety minutes of exposure of the same temperature, i.e., 175 deg and 180 deg Celsius for the purposes of this paper. To compute $A(t)$, the percentage of L-ascorbic acid contained in a tablet for $t = 30$ and $t = 90$, we used the following formula:

$$A(t) = \left[\frac{c_p \cdot B(t)}{A_p} \cdot d \right] \cdot \frac{w}{M}, \quad (2.2)$$

where

- the constant c_p is the molar concentration of the standard ascorbic acid solution employed to calibrate the spectrophotometer,
- A_p is the absorbance value measured with the spectrophotometer at 243 nm of the standard ascorbic acid solution,
- $B(t)$ is the absorbance of the ascorbic acid solution that was obtained at time t from a tablet subjected to thermal stress,
- $d = 2500$ is the dilution factor, adjusted to percentage, of the L-ascorbic acid solution that is suitable to take measurements using a spectrophotometer,
- w is the molecular weight of L-ascorbic acid, whose value is $176.13 \text{ g mol}^{-1}$,
- M is the initial mass of L-ascorbic acid contained in the tablet before exposure to thermal stress.

To account for measurement errors, the absorbances A_p and $B(t)$ were measured three times. The measurements of the absorbance A_p of the standard solution yielded the following values:

- For 175° : 1.404, 1.399, and 1.406;
- For 180° : 0.796, 0.795, and 0.794.

Since $B(t)$ varies with time, $B(t)$ was measured thrice for $t = 30$ and $t = 90$. Specifically, the following measurements were taken after 30 minutes and 90 minutes of exposure to the temperatures 175 and 180 degrees Celsius:

1. For 175° :
 - (a) Measurements of $B(30)$: 1.225, 1.227, 1.232;
 - (b) Measurements of $B(90)$: 0.560, 0.557, 0.558.
2. For 180° :
 - (a) Measurements of $B(30)$: 1.169, 1.164, 1.162
 - (b) Measurements of $B(90)$: 0.185, 0.186, 0.189.

Let us emphasize that measuring the mean absorbance of ascorbic acid at time t_0 requires destroying the tablet. Therefore, another tablet had to be used in order to take the three measurements of $B(t_f)$, which increases the uncertainty. Another source of uncertainty is M , the mass of L-ascorbic acid at the time of fabrication. As mentioned before, each tablet was assumed to have an L-ascorbic acid content anywhere between 87 and 97 %. Multiplying the interval $[87, 97]$ by the weight of the tablet, whose weight was determined without any significant measurement error, at the time of fabrication yields $M \in \mathbb{I}_+^*$, where \mathbb{I}_+^* denotes $\{[a, \bar{a}] \mid 0 < a \leq \bar{a}\} \subset \mathbb{R}$. Moreover, we also modelled A_p and $B(t)$, where $t \in \{t_0, t_f\}$, as subintervals of \mathbb{R}_+ by using the minima and the maxima of the measured values as their left and right endpoints, respectively.

Since there is no relationship between A_p , $B(t)$ and M , the arithmetic operations in Equation (2.2) can be executed using the usual interval arithmetic yielding an initial epistemic interval $A(t_0) = [a_0, \bar{a}_0]$ and a final epistemic interval $A(t_f) = [a_f, \bar{a}_f]$. However, interactivity does play a role in the process of transiting from $A(t_0) \in \mathbb{I}_+^*$ to $A(t_f) \in \mathbb{I}_+^*$ by means of an interval-valued derivative $A' = -kA$. Since intervals can be viewed as a special case of fuzzy numbers, let us proceed by briefly reviewing the concepts of interactive fuzzy numbers, arithmetic and fuzzy derivatives.

3 A Brief Review of Joint Possibility Distributions and Concepts of Interactivity

To begin with, recall that every fuzzy subset A of a universe $X \neq \emptyset$ is uniquely determined by its membership function $\mu_A : X \rightarrow [0, 1]$. For convenience, we simply write $A(x)$ instead of $\mu_A(x)$ for $x \in \mathbb{R}$. The symbol $\mathcal{F}(X)$ denotes the class of fuzzy sets of X . The class $\mathcal{F}(X)$ together with the partial ordering of fuzzy set inclusion is a complete lattice that is isomorphic to the complete lattice given by $[0, 1]^X = \{f : X \rightarrow [0, 1]\}$ together with the usual partial ordering of functions.

Note that $[0, 1]$, which is the value set of these functions, is a complete chain with the usual total ordering. Therefore, the supremum and the infimum, denoted respectively using the symbols \bigvee and \bigwedge , of any subset of $[0, 1]$ exist in $[0, 1]$.

From now on, let us focus on fuzzy subsets of \mathbb{R} . An element A of $\mathcal{F}(\mathbb{R})$ is called a *fuzzy number* if for every $\alpha \in [0, 1]$ the α -cut of A , denoted $[A]^\alpha$, is a non-empty, bounded, closed interval and the (topological) closure of its support is compact. Recall that $[A]^\alpha$ is defined as follows:

$$[A]^\alpha = \begin{cases} \{x \in \mathbb{R} : A(x) \geq \alpha\} & , \text{ if } \alpha \in (0, 1] \\ cl\{x \in \mathbb{R} : A(x) > 0\} & , \text{ if } \alpha = 0 \end{cases}$$

where $cl(Y)$ stands for the (topological) closure of the set Y [2]. Given an arbitrary fuzzy number A and $\alpha \in [0, 1]$, the interval $[\underline{a}_\alpha, \bar{a}_\alpha]$ denotes $[A]^\alpha$. For simplicity, we also write $[\underline{a}, \bar{a}]$ instead of $[\underline{a}_0, \bar{a}_0]$ in this paper. Note that this notation is consistent with our notation for elements of $\mathbb{I}^* \subset \mathbb{R}_{\mathcal{F}}$. Note that any non-empty and closed interval $[\underline{a}, \bar{a}] \subset \mathbb{R}$ can be identified with a unique fuzzy number via its characteristic function $\chi_{[\underline{a}, \bar{a}]} : \mathbb{R} \rightarrow \{0, 1\}$. Let us use the symbols $\mathbb{R}_{\mathcal{F}}$ and \mathbb{I}^* to denote the classes of fuzzy numbers and intervals in \mathbb{R} , respectively. Moreover, the symbol $\mathbb{R}_{\mathcal{F}_c}$ denotes the class of fuzzy numbers with continuous endpoints. More precisely, $A \in \mathbb{R}_{\mathcal{F}_c}$ if and only if the functions $[0, 1] \rightarrow \mathbb{R}$ given by $\alpha \mapsto \underline{a}_\alpha$ and $\alpha \mapsto \bar{a}_\alpha$ are continuous.

Recall that the *width* of a fuzzy number A is given by

$$width(A) = \bar{a} - \underline{a}.$$

If $A \in \mathbb{I}^*$, then we speak of the *length* of the interval A and write $length(A)$ instead of $width(A)$.

Definition 1. [6, 14] Let $A_i \in \mathcal{F}(X_i)$, where X_i are non-empty sets for $i = 1, \dots, n$. A fuzzy set, also called *fuzzy relation*, J on $X_1 \times \dots \times X_n$ is said to be a joint possibility distribution (JPD) of the fuzzy sets A_i , $i = 1, \dots, n$ if the following condition is satisfied for every $i \in \{1, \dots, n\}$:

$$A_i(y) = \bigvee_{\mathbf{x} \in \mathbb{R}^n : x_i = y} J(\mathbf{x}), \quad \forall y \in \mathbb{R}, \quad (3.3)$$

In this case, $A_1, \dots, A_n \in \mathbb{R}_{\mathcal{F}}$ are called J -interactive or, simply, interactive and each A_i is called a *marginal possibility distribution* of J .

Given $A_1, \dots, A_n \in \mathbb{R}_{\mathcal{F}}$, any t-norm t gives rise to the following joint possibility distribution J_t of these fuzzy numbers:

$$J_t(x_1, \dots, x_n) = A_1(x_1) t \dots t A_n(x_n) \quad \forall (x_1, \dots, x_n) \in \mathbb{R}^n$$

The fuzzy set J_t is called a t -norm-based joint possibility distribution. For the particular case where t is

the minimum t-norm we say that $A_1, \dots, A_n \in \mathbb{R}_{\mathcal{F}}$ are *non-interactive*. Note that the notion of interactivity of fuzzy numbers depends on the joint possibility distribution J under consideration. Moreover, J can be used to define interactive arithmetic operations via Zadeh's extension principle [3].

Definition 2. Let J be a joint possibility distribution of given fuzzy numbers A_1 and A_2 . The interactive sum $A_1 +_J A_2$ and the interactive difference $A_1 -_J A_2$ are defined as follows:

$$(A_1 +_J A_2)(y) = \bigvee_{(x_1, x_2) : x_1 + x_2 = y} J(x_1, x_2),$$

$$(A_1 -_J A_2)(y) = \bigvee_{(x_1, x_2) : x_1 - x_2 = y} J(x_1, x_2).$$

Several parametrized families of JPDs have already appeared in the literature. This paper focuses on the parametrized family of joint possibility distributions $\{J_\gamma \mid \gamma \in [0, 1]\}$ that was proposed by Esmi et al. for any pair of fuzzy numbers $A_1, A_2 \in \mathbb{R}_{\mathcal{F}_c}$ and whose explicit construction is rather technical [5]. For illustration, the family of joint possibility distributions proposed by Esmi et al., in the interval case, can be seen in the following figure for the $\gamma = 0$, $\gamma = 0.5$ and $\gamma = 1$. Note that for $\gamma = 1$ the intervals are non-interactive.

For the purposes of this paper, it suffices to characterize the α -cuts of the J_γ -interactive sum of $A, B \in \mathbb{R}_{\mathcal{F}_c}$ [5]. For convenience, we will use the symbols $+_\gamma$ and $-_\gamma$ to denote respectively $+_J$ and $-_J$ for $J = J_\gamma$.

Theorem 1. Let $A, B \in \mathbb{R}_{\mathcal{F}_c}$, $\gamma \in [0, 1]$ and $\alpha \in [0, 1]$. If $C = A +_\gamma B$, $a = 0.5(\underline{a}_1 + \bar{a}_1)$ and $b = 0.5(\underline{b}_1 + \bar{b}_1)$, then we have

$$[\underline{c}_\alpha, \bar{c}_\alpha] = \left[\bigwedge_{\beta \geq \alpha} \underline{h}(\beta, \gamma), \bigvee_{\beta \geq \alpha} \bar{h}(\beta, \gamma) \right]$$

where

$$\underline{h}(\beta, \gamma) = [\underline{a}_\beta + \bar{b}_\beta - \gamma(\bar{b}_\beta - \underline{b}_\beta)] \wedge [\bar{a}_\beta + \underline{b}_\beta - \gamma(\bar{a}_\beta - \underline{a}_\beta)] \wedge [(1 - \gamma)(a + b) + \gamma(\underline{a}_\beta + \underline{b}_\beta)]$$

and

$$\bar{h}(\beta, \gamma) = [\underline{a}_\beta + \bar{b}_\beta + \gamma(\bar{a}_\beta - \underline{a}_\beta)] \wedge [\bar{a}_\beta + \underline{b}_\beta + \gamma(\bar{b}_\beta - \underline{b}_\beta)] \wedge [(1 - \gamma)(a + b) + \gamma(\bar{a}_\beta + \bar{b}_\beta)].$$

Moreover, $A -_\gamma B$ is equal to $A +_\gamma (-1) \cdot B$.

Corollary 1. In the special case where $A, B \in \mathbb{I}^* \subset \mathbb{R}_{\mathcal{F}_c}$, we obtain the following representation of the

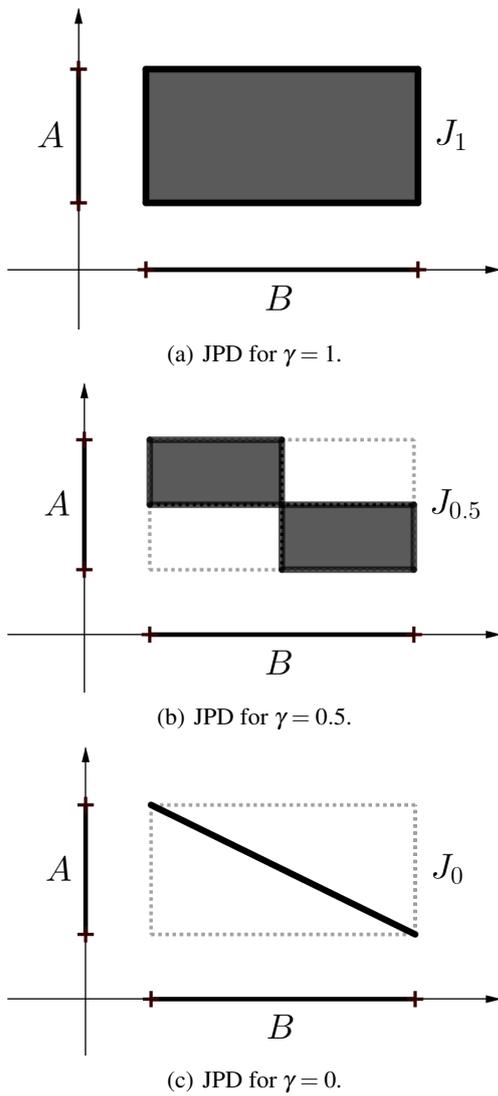


Figure 1: The joint possibility distributions J_γ of the intervals A and B are represented by the dark regions. The intervals A and B are represented by the black lines.

left and right endpoints \underline{c} and \bar{c} of $C = A +_\gamma B$ for any $\gamma \in [0, 1]$:

$$\underline{c} = [\underline{a} + \bar{b} - \gamma(\bar{b} - \underline{b})] \wedge [\bar{a} + \underline{b} - \gamma(\bar{a} - \underline{a})] \wedge [(1 - \gamma)(a + b) + \gamma(\underline{a} + \underline{b})],$$

and

$$\bar{c} = [\underline{a} + \bar{b} + \gamma(\bar{a} - \underline{a})] \wedge [\bar{a} + \underline{b} + \gamma(\bar{b} - \underline{b})] \wedge [(1 - \gamma)(a + b) + \gamma(\bar{a} + \bar{b})],$$

with $a = 0.5 * (\underline{a} + \bar{a})$ and $b = 0.5 * (\underline{b} + \bar{b})$.

In addition, we will make use of the following results that can also be found in [5]:

Theorem 2. Let $A, B \in \mathbb{R}_{\mathcal{F}_\phi}$. The following properties hold true:

- a) $A +_\gamma B \in \mathbb{R}_{\mathcal{F}_\phi}$ for all $\gamma \in [0, 1]$.
- b) $A +_{\gamma_1} B \subseteq A +_{\gamma_2} B$ if $0 \leq \gamma_1 \leq \gamma_2 \leq 1$.
- c) The mapping $\gamma \mapsto \text{width}(A +_\gamma B)$ is continuous and increasing.

Note that an interactive arithmetic can be used as follows to generalize a numerical method such as the Euler method for solving IVPs in order to deal with FIVPs. Specifically, consider the following simple IVP, where $y, f : \mathbb{R} \rightarrow \mathbb{R}$ are functions of t :

$$\begin{cases} y' = f(t, y) \\ y_0 = y(t_0) \end{cases}, \quad (3.4)$$

whose numerical solution via the Euler method at times t_0, t_1, \dots, t_N is given by the recursion

$$y_{k+1} = y_k + hf(t_k, y_k). \quad (3.5)$$

Here, $t_{k+1} - t_k = h > 0$ for $k = 0, 1, \dots, N - 1$, where $N \in \mathbb{N}$, and (t_0, y_0) is the initial condition.

Note the next state in Equation (3.5) is given by adding the current state and a multiple of the approximate derivative at the current time t_k . Using joint possibility distributions, the conventional Euler method can be extended to fuzzy number valued functions $Y : \mathbb{R} \rightarrow \mathbb{R}_{\mathcal{F}}$ and $F : \mathbb{R}_{\mathcal{F}} \rightarrow \mathbb{R}_{\mathcal{F}}$ by means of J -interactive sums. In the special case where $Y : \mathbb{R} \rightarrow \mathbb{R}_{\mathcal{F}_\phi}$ and $F : \mathbb{R}_{\mathcal{F}_\phi} \rightarrow \mathbb{R}_{\mathcal{F}_\phi}$, one can apply a fuzzy version of the Euler method as follows for every $\gamma \in [0, 1]$, yielding (t_k, Y_k) for $k = 1, \dots, N \in \mathbb{N}$:

$$Y_{k+1} = Y_k +_\gamma hF(t_k, Y_k). \quad (3.6)$$

4 An Approach Toward Determining γ for the γ -Interactive Euler Method

Let us consider a family of γ -interactive fuzzy Euler methods given by Equation (3.6). We have strong reasons to believe that each of these γ -interactive fuzzy Euler methods yields a numerical solution to an interactive FIVP with the same fuzzy initial condition $Y_0 = Y(t_0)$. However, the width of the fuzzy trajectory produced by a γ -interactive fuzzy Euler method depends on the choice of γ . Some additional a priori information is necessary to determine a suitable γ .

In this paper, we assume that we are given the width of $Y(t_N)$. In our opinion, this assumption only constitutes

a minor amount of additional information and the approach presented below does not constitute a straightforward generalization of numerical approaches for boundary value problems.

Specifically, given a family of γ -interactive fuzzy Euler methods described by Equation (3.6) and $Y(t_0) = Y_0$, the goal of this section is to determine a parameter $\omega \in [0, 1]$ such that performing the recursive steps of Equation (3.6) for $\gamma = \omega$ yields $\text{width}(Y_N) \approx w$. To this end, we employ the following properties of the family J_γ :

1. The joint possibility distribution J_γ exists for every pair of elements $A_1, A_2 \in \mathbb{R}_{\mathcal{F}_\phi}$;
2. If $A_1, A_2 \in \mathbb{R}_{\mathcal{F}_\phi}$, then $A_1 + \gamma A_2 \in \mathbb{R}_{\mathcal{F}_\phi}$
3. For every pair of elements $A_1, A_2 \in \mathbb{R}_{\mathcal{F}_\phi}$, we have that the function that maps $\gamma \in [0, 1]$ to $\text{width}(A_1 + \gamma A_2) \in \mathbb{R}$ is continuous and increasing.

For convenience, let us from now on use the symbol Y_N^γ instead of Y_N to denote the approximation of $Y(t_N)$ obtained via Equation (3.6). Note that one can view Y_N^γ as a function of γ , that is, we might as well write $Y_N(\gamma)$ instead of Y_N^γ . This observation implies that a numerical method such as the secant method can be used to approximate a root of $Y_N^\gamma - w$, i.e., a solution of $Y_N^\gamma = w$. The third property mentioned above (cf. item c) of Theorem 2) implies that $\text{width}(Y_N) : [0, 1] \rightarrow \mathbb{R}$ is continuous and increasing. Therefore, the function $\text{width}(Y_N) - w$ is also continuous and increasing. Thus, $\text{width}(Y_N) - w$ has a unique zero which can be approximated to any precision $\varepsilon > 0$ using a numerical method such as the secant method. Note that applications of these numerical methods do not require calculating the derivative of $\text{width}(Y_N)$.

5 A Case Study in L-Ascorbic Acid Degradation under Isothermal Stress

Let us return to the problem of isothermal degradation of L-ascorbic acid that we discussed in Section 2. Our focus is on the the percentage of L-ascorbic acid contained in a tablet that can be determined using the formula of Equation (2.2). For crisp values of A_p , $B(t)$ and M , this formula gives rise to a crisp value of $A(t)$. However, in practice, A_p , $B(t)$ and M are inherently subject to uncertainty. In order to account for this uncertainty, we modelled A_p , $B(t)$ and M as elements of $\mathbb{I}^* \subset \mathbb{R}_{\mathcal{F}_\phi}$ using the following approach:

- We computed $A_p, B(30), B(90) \in \mathbb{I}^*$ by taking respectively the minima of the three measurements

corresponding to $A_p, B(30)$, and $B(90)$ as the left endpoints and the maxima of the three measurements corresponding to $A_p, B(30)$, and $B(90)$ as the right endpoints. In other words, $A_p, B(30)$, and $B(90)$ were modelled as the smallest intervals containing their measured values. This strategy yielded the following intervals:

- For 175° : $A_p = [1.399, 1.406]$, $B(30) = [1.225, 1.232]$, and $B(90) = [0.557, 0.560]$;
- For 180° : $A_p = [0.794, 0.796]$, $B(30) = [1.162, 1, 169]$, and $B(90) = [0.185, 0.189]$.

- The percentage of L-ascorbic acid contained in the tablet at the end of the manufacturing process is uncertain. The manufacturing process of the tablets considered for the purposes of this paper followed the guidelines of the elapsed patent [8], implying that errors of up to 5% are possible. Since 92.49 was the mean percentage of L-ascorbic acid determined using three different tablets, we assume that the percentage of L-ascorbic acid initially (at time $t = 0$) contained in the tablet is $92.49 \pm 5\%$, which amounts to the interval $[0.8749, 0.9749]$. Since the mass of an individual tablet can be determined with high precision, multiplying the aforementioned interval by the mass of an individual tablet yields another interval which corresponds to the initial mass M . Specifically, we obtained:

- For 175° : $M(30) = [0.5550, 0.6184]$ and $M(90) = [0.5573, 0.6210]$;
- For 180° : $M(30) = [0.5724, 0.6378]$ and $M(90) = [0.5484, 0.6130]$.

Note that $A_p, B(t), M \in \mathbb{I}_+^* \subset \mathbb{R}_+$. Moreover, A_p , i.e., the absorbance of the standard ascorbic acid solution, $B(t)$, i.e., the absorbance of the L-ascorbic acid solution at time t , and M , i.e., the initial mass of L-ascorbic acid contained in a tablet, are completely unrelated which implies that non-interactive fuzzy arithmetic can be used to perform an arithmetic operation with a pair of elements of $\{A_p, B(30), M\}$ or $\{A_p, B(90), M\}$. In other words, one can perform arithmetic operations with a pair of these intervals using the standard Moore interval arithmetic. In particular, for $A, B \in \mathbb{I}_+^*$, we obtain:

$$A \cdot B = [\underline{a} \cdot \underline{b}, \bar{a} \cdot \bar{b}], \quad A/B = [\underline{a}/\bar{b}, \bar{a}/\underline{b}]. \quad (5.7)$$

In light of Equation 2.2 and using the fact that the percentage of L-ascorbic acid cannot exceed 100 %, we computed the following intervals $A(30)$ and $A(90)$, i.e., the percentages of L-ascorbic acid $A(30)$ and $A(90)$, for the temperatures 175° and 180° :

- For 175° : $A(30) = [0.9203, 1.0282 \wedge 1] = [0.9203, 1]$ and $A(90) = [0.4167, 0.4654]$;
- For 180° : $A(30) = [0.7728, 0.8615]$ and $A(90) = [0.1284, 0.1454]$.

As mentioned at the beginning of Section 2, the isothermal degradation of L-ascorbic acid can be described in terms of the crisp IVP given by Equation (2.1) if the values of the percentages of L-ascorbic acid are crisp. Note that $a(t) = a_0 \cdot e^{-k(t-t_0)}$ is a solution of Equation (2.1). Furthermore, if $a(t)$ is assumed to interpolate (t_f, a_f) for some $t_f > t_0$ and some $a_f > 0$, then k is uniquely determined as follows:

$$k = \frac{\log(a_0) - \log(a_f)}{t_f - t_0}. \tag{5.8}$$

However, here we are dealing with interval-valued $A(t) \in \mathbb{I}_+^* \subset \mathbb{R}_{\mathcal{F}_\epsilon}$. Thus, it is unclear what concept of derivative to use in order to generalize Equation (2.1). It is reasonable to believe that there exists a suitable interactive FIVP that extends Equation (2.1) and that there exists a γ -interactive Euler method yielding numerical solutions that approximate the analytical solution of this interactive FIVP as the step size h tends toward 0. In this paper, we assume that $k \in \mathbb{R}_+$ because there should be an FIVP that extends Equation (2.1) using the same choice of k and some notion of fuzzy derivative. We adopted the following numerical approach for each temperature in $\{175^\circ, 180^\circ\}$:

1. We set $A_0 = A(30)$ and $A_f = A(90)$ and defined a_0 and a_f as $\frac{\bar{a}_0 + \underline{a}_0}{2}$ and $\frac{\bar{a}_f + \underline{a}_f}{2}$, respectively. In addition, we denoted the width of A_f , i.e., $\bar{a}_f - \underline{a}_f$ using the symbol w .
2. After calculating k using Equation (5.8), we considered the family of γ -interactive Euler methods given by (cf. Equation (3.6)):

$$A_{k+1}^\gamma = A_k^\gamma + \gamma(-kA_k^\gamma) = A_k - \gamma k A_k, \tag{5.9}$$

where $A_0^\gamma = A_0$.

3. After selecting a number of iterations $N \in \mathbb{N}$ and defining the step size h as $\frac{t_f - t_0}{N} = \frac{60}{N}$, we applied the secant method with a given precision $\epsilon > 0$ to determine an approximate solution of

$$A_N(\gamma) - w = 0, \text{ where } A_N(\gamma) = A_N^\gamma. \tag{5.10}$$

In practice, we adopted the values $h = 0.06$ and $\epsilon = 0.0001$, that is, we stopped generating approximate values of the unique solution γ^* of Equation (3) if the current approximation γ_k satisfies $|A_N(\gamma_k) - w| < \epsilon$ or

$|\gamma_k - \gamma_{k-1}| < \epsilon$. Using $\gamma_0 = 0.25$ and $\gamma_1 = 0.75$ as initial values, $h = 0.06$ and $\epsilon = 0.0001$, we obtained the following approximate solutions $\bar{\gamma}^*$ of Equation (3):

- For 175° : $\bar{\gamma}^* = 0.1875$;
- For 180° : $\bar{\gamma}^* = 0.0371$.

Figures 2 and 3 illustrate the applications of the $\bar{\gamma}^*$ -interactive Euler method for the two temperatures under consideration.

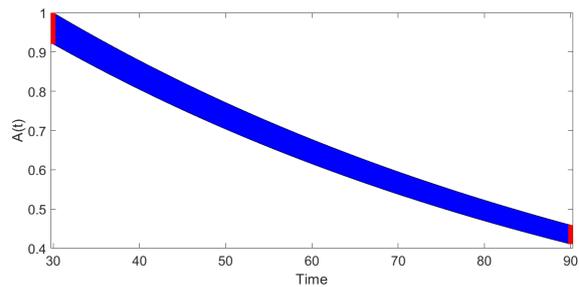


Figure 2: The $\bar{\gamma}^*$ -interactive Euler method for the temperature 175° . The blue region represents the numerical solution for $\bar{\gamma}^* = 0.1875$. The red intervals represent $A_0 = A(30)$ and $A_f = A(90)$.

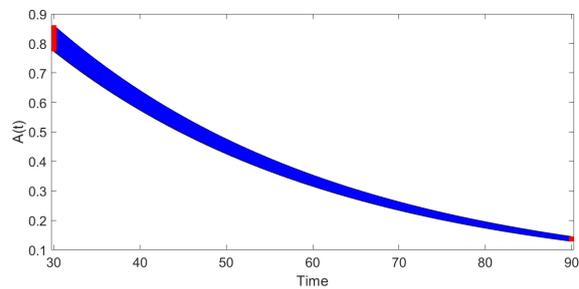


Figure 3: The $\bar{\gamma}^*$ -interactive Euler method for the temperature 180° . The blue region represents the numerical solution for $\bar{\gamma}^* = 0.0371$. The red intervals represent $A_0 = A(30)$ and $A_f = A(90)$.

6 Final Remarks

This paper introduces a novel numerical approach toward characterizing the phenomenon of interactivity. We conducted a case study using real world data that stem from a problem of chemical degradation of L-ascorbic acid and that we modeled in terms of intervals. These intervals intrinsically incorporate interactivity, which is associated with a family of joint possibility distributions J_γ . Using our approach, we determined an approximation for the parameter of the fuzzy numerical solution that yields the desired length at a

certain time t_f . Figures 2 and 3 provide a visual interpretation. Although the approach presented in this paper was applied to uncertain real-word data that was modelled using intervals, it can be easily extended so as to deal with uncertain data described by fuzzy numbers.

Acknowledgement

The authors would like to thank K. B. Rodrigues for her assistance in performing the experiments regarding L-ascorbic acid degradation under isothermal stress. In addition, the authors are grateful for the financial support of CNPq under grants nos. 315638/2020-6, 313313/2020-2, and 122297/2012-1, as well as FAPESP under grants nos. 2018/13657-1, 2020/09838-0, and 2007/55627-7.

References

- [1] P. Atkins, J. De Paula, *Physical Chemistry*, W. H. Freeman and Company, New York, 2006.
- [2] L. C. Barros, R. C. Bassanezi, W. A. Lodwick, *A First Course in Fuzzy Logic, Fuzzy Dynamical Systems, and Biomathematics*, 1st Edition, 347, Springer - Verlag Berlin Heidelberg, Berlin, 2017.
- [3] C. Carlsson, R. Fullér, P. Majlender, Additions of completely correlated fuzzy numbers, in: *Fuzzy Systems, 2004. Proceedings. 2004 IEEE International Conference on*, Vol. 1, IEEE, 2004, pp. 535–539.
- [4] U. S. P. Convention, *The United States Pharmacopeia - USP 29 - The National Formulary: NF 24*, United States Pharmacopeial Convention, Rockville, 2005.
- [5] E. Esmi, V. F. Wasques, L. C. de Barros, Addition and subtraction of interactive fuzzy numbers via family of joint possibility distributions, *Fuzzy Sets and Systems*, 2021, accepted for publication, available online.
- [6] R. Fullér, P. Majlender, On interactive fuzzy numbers, *Fuzzy Sets and Systems* 143 (3) (2004) 355–369.
- [7] A. Khawam, D. R. Flanagan, Basics and applications of solid-state kinetics: A pharmaceutical perspective, *Journal of Pharmaceutical Sciences* 95 (3) (2006) 472–498.
- [8] N. Kitamori, T. Makino, K. Hemmi, L-ascorbic acid tablets, U.S. Patent 4,036,948 (1977).
- [9] C. V. Negoită, D. A. Ralescu, *Applications of Fuzzy Sets to Systems Analysis*, Springer, 1975.
- [10] T. Ozawa, A new method of analyzing thermogravimetric data, *Bulletin of the Chemical Society of Japan* 38 (11) (1965) 1881–1886.
- [11] K. B. Rodrigues, J. C. B. Fernandes, Determinação da constante cinética de decomposição de vitamina c por ação do calor em formulação farmacêutica sólida, in: *65th Annual SBPC Meeting (SBPC'13)*, Recife, Brazil, 2013, in portuguese.
- [12] B. Tita, A. Fúlias, M. Stefanescu, E. Marian, D. Tita, Kinetic study of decomposition of ibuprofen under isothermal conditions, *Revista de Chimie* 62 (2) (2011) 216–221.
- [13] V. F. Wasques, E. Esmi, L. C. Barros, P. Susner, The generalized fuzzy derivative is interactive, *Information Sciences* 519 (2020) 93–109.
- [14] L. A. Zadeh, The concept of a linguistic variable and its application to approximate reasoning I, *Information Sciences* 8 (1975) 199–250.