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New algorithm for the elucidation of functional properties of gelatin-based materials

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ABSTRACT

In the present work, fish gelatin was employed to develop renewable and biodegradable materials, reducing environmental problems associated with conventional petroleum-based materials. Glycerol was used as plasticizer and gallic acid was added in order to enhance the functional properties of the material. *L*-fuzzy concept analysis was applied for modelling the formulations and properties of the films. These new methodology is used in the design of the material avoiding the traditional inefficient trial an error approach usually employed. Two applications of the developed bio-based material were analyzed: fatty food packaging application in food area and wound healing in the biomedical field. The functional properties requirements of water contact angle (CA), water vapour transmission rate (WVTR), colour L^* and b^* values, tensile strength (TS), elongation at break (EB) and gloss values were specified for both applications. Applying the proposed algorithm, the required formulations were estimated and the experimental results showed a high accordance with the predicted values of the final properties, as well as with the requirements. This analysis allowed finding the required formulations in a highly cost-effective way.

1. Introduction

Materials design is a common task in laboratories and industry, and many times its efficiency may be conditioned by the complexity of the formulation and/or the limitations of the time or money required for the analysis. The adjustment of a manifold of parameters with a view to achieving the desired final properties of a material can be performed on a basis of previous knowledge, but in any case, the complex interactions among these parameters make the process a challenge. The designer is commonly blind exploring a wide multidimensional space of combinations, and he/she must bear in mind that, for a given set of chemical constituent species, the properties that he/she is trying to achieve may be out of hand.

In order to address this problem, computer-aided mathematical treatment is a suitable tool that allows systematic and rational materials design. Doing so, unnecessary tasks and their attached costs are avoided, since the mathematical analysis greatly reduces experimental labor by focusing on only indispensable measurements. In this context, fuzzy sets theory (Klir and Yuan, 1995) helps the designer when the requirements that guide design are imprecise, in the sense that either values for different magnitude may be acceptable in the proximity of ideal objective values; or the desirability criteria are formulated in a textual form, e.g. “it is recommended”, or “it is very important that” (Balakrishna et al., 2011; Liao, 1996).

In this scenario, the research on alternative solutions to conventional plastics is promoted by serious environmental problems and the rising consciousness about sustainability that population and governments are acquiring. Bio-based materials are an interesting approach to be exploited, because of their versatility and increas-

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ing functionality when applied to a wide range of requirements. In this regard, bio-based materials showed promising properties as wound dressing (Bhowmik et al., 2017; Jiang et al., 2019; Khan et al., 2016; Liu et al., 2017; Singh et al., 2017) and food packaging (Haghighi et al., 2020; Pandey et al., 2020; Priyadarshi and Rhim, 2020), among many others.

In reference to wound healing application, to accelerate the wound healing process, an ideal wound dressing should display adequate gas permeability, biocompatibility, provide an antibacterial environment (Ding et al., 2017; Jayakumar et al., 2011) as well as maintain a moist environment at the wound surface (Zilberman et al., 2015).

Within the field of food packaging, food oxidation, that is considered a major cause of quality deterioration affecting both the nutritional and sensory quality and safety of foods, may be accelerated by moisture, light, oxygen or exposure to high temperatures (Carrizo, 2016). Packaging for fatty food requires a particular attention, since auto-oxidation, which is the oxidation in the absence of light, can be responsible for the deterioration of olive oil flavor termed “oxidative rancidity” (Gargouri et al., 2014). Unsaturated fatty acids are generally susceptible to lipid oxidation and to development of undesirable odor (Ali et al., 2020; Muhammed et al., 2015; Sae-leaw and Benjakul, 2017). The prevention or inhibition of these processes during fatty food storage are thus required for maintaining their quality standard.

An efficient design of bio-based materials for both applications of wound dressing and fatty food packaging is needed. In order to tackle this problem, we propose the use of a methodology based on the analysis of *L*-fuzzy concepts (Burusco and Fuentes-González, 1994; 1998; 2000) obtained from initial requirements that will provide the designer with a set of formulation parameters which will plausibly lead to the desired properties values. Or, to be precise, and attending to the fuzzy nature of the method, values which will lie *near* to some target values.

2. Materials and methods

2.1. Materials

Fish gelatin was purchased from Healan Ingredients (East Yorkshire, UK). Glycerol and gallic acid were obtained from Panreac (Barcelona, Spain). All chemicals were used as received without further purification.

2.2. Film preparation

Fish gelatin films were prepared by mixing gelatin and gallic acid in distilled water. The acid contents employed in this work were 5, 10 or 15 wt. % on gelatin basis. Solutions were heated at 80° C for 30 min and stirred at 200 rpm. Then, 0, 5, or 10 wt. % glycerol (on gelatin basis) was added as a plasticizer and solution pH was adjusted with 1 N NaOH; the pH values used in the present work were 4.50, 7.25 and 10.00. The heating procedure was repeated and finally, solutions were poured into Petri dishes and allowed to cool for 48 h at room temperature. All films were conditioned in a controlled environment.

2.3. Mechanical behavior

Tensile tests were performed in an electromechanical testing system (MTS Insight 10) in order to determine tensile strength (TS) and elongation at break (EB). Tests were carried out according to ASTM D638-03.

2.4. Colour and gloss

Colour values of films were measured using a portable colorimeter (CR-400 Minolta Croma Meter). Film specimens were placed on a white plate, and the CIELAB color scale was used to measure color: L*= 0 (black) to L*= 100 (white) and -b*(blueness) to +b*(yellowness). Standard values for the white calibration plate were L*= 97.39 and b*= 1.77. With these values and considering standard light source D65 and standard observer 2 degrees color parameters L* and b* were measured.

Gloss of the films was measured with a Minolta gloss meter (MultiGloss 268 Plus). Gloss was measured at a 60° incidence angle, according to ASTM D523-14. Results were expressed as gloss units, relative to a highly polished surface of black glass standard with a value near to 100.

2.5. Contact angle

A contact angle system (model Oca20, Dataphysics Instruments) was used to measure the contact angle of water in air on the surface of gelatin films. A film sample (20 mm × 80 mm) was put on a movable sample stage and leveled horizontally; then a drop of about 3 μL of distilled water was placed on the surface of the film using a microsyringe. The contact angle was measured in a conditioned room by recording contact angle values. Image analysis were carried out using SCA20 software.

2.6. Water vapor permeability

WVTR of the films was determined according to ASTM E96-00. The sample film was cut into a circle of 7.40 cm diameter and the test area was 33 cm^2. The setup was subjected to a temperature and relative humidity of 38° C and 90%, respectively. Water vapor transmission rate (WVTR) was calculated as

$$WVTR = \frac{G}{t \times A}$$

where, G is the change in weigh (g), t is the time (day), and A is the test area of the film (m^2).

3. Description of the problem

3.1. Description of the materials designing conditions

When a novel material is designed, formulation and processing conditions play a vital role in the properties of the material during service. These properties are related with different magnitudes (chemical, physical, mechanical, thermal, optical,...). For each application, we know which target output values these magnitudes should show in an optimal performance, and the question is to find the formulation that will provide us with it.

In order to achieve this goal, not only the constituent chemical species that optimize properties must be identified, but also their quantity. These quantities will be input values to be determined in an optimization study. Usually, there is a previous knowledge that helps the designer determining the approximate ranges where the ideal input values might lie in order to obtain the desired performance output values.

When finding the desired input values in their already known approximate ranges, the traditional trial and error approach with many involved variables usually require high costs of energy, time and raw materials. There are so many combinations and variation to study that this is an inefficient protocol for analyzing the influence of each parameter on the performance of the design material. In addition to this, usually the analysis is carried out via one-variable-at-a-time test procedure, which does not consider the

Table 1
Material requirements for fatty food packaging and wound healing applications. Studied material properties are water contact angle (CA), water vapour transmission rate (WVTR), L* and b* colour values, tensile strength (TS), elongation at break (EB) and gloss values at 60° incidence angle.

	CA (°)	WVTR $\left(\frac{g}{m^2 \cdot day}\right)$	L*	b*	TS (MPa)	EB (%)	Gloss$_{60°}$(Gloss units)
Fatty food packaging	90		95	40	60	5	50
Wound healing		1300	60	50	90	3	50

cross-interactions between the different input values of the formulation.

Often, these inputs and outputs are *blurry*: the designer would accept input values *at around* some particular input values, because he/she is willing to admit output values*at around* some particular output values. In this case, we can ascribe desirability fuzzy variables to them, and perform a study based on the fuzzy concepts theory.

Within this approach, and with a strict analysis of the involved variables, a set of plausible input values of the formulation can be provided in an efficient way reducing the consumption of time or resources, avoiding an inefficient protocol with unnecessary tasks and their attached costs.

Many researchers tried to optimize the material design and the properties of the materials using techniques based on the design of experiments such as response surface methodology (Astray et al., 2016; Leceta et al., 2018; Garrido et al., 2019). This type of tools are very useful, but they are based on curves or hypersurfaces fitting methods. This requires a previous knowledge of the curve or hypersurface to which the experimental points are going to be fitted and only works in the cases in which an adequate fitting is possible. Usually due to the complexity of the system, or owing to its instability having significant changes in properties with slight variations in formulation values, the techniques based on approximation are not always valid.

In this context, in the case of the developed material, it was not find any analytic relation between formulation values and functional properties of the materials, or adequate fitting. Due to this, the aim is to analyse each formulation independently, and L-fuzzy context analysis provides a tool for obtaining this objective.

3.2. Properties required for the material applications

Gelatin-based films display interesting characteristics that make them suitable for being applied to both wound healing and food packaging. Barrier properties, mechanical behaviour, and optical properties are vital properties when designing food packaging materials (Leceta et al., 2013). Regarding wound healing application, the layer is designed to cover the wound, and to give mechanical strength to the dressing. In addition, it needs to control moisture transmission to prevent fluid loss and dehydration, while allowing exudate removal (Garcia-Orue et al., 2019).

Bearing this in mind, the following output properties were chosen for further analysis: contact angle (CA), water vapour transmission rate (WVTR), L* and b* (as far as colour properties are concerned), tensile strength (TS), elongation at break (EB) and gloss.

Considering the expertise in the field of the BIOMAT-biopolymeric materials group, a set of target output properties for the candidate formulations for both wound healing and food packaging applications has been proposed, which takes into account the specificities of gelatin-based films. This is the first milestone in order to launch the *L*-fuzzy concept analysis. These target properties are listed in Table 1.

In the case of water vapor transmission rate, for fatty food packaging application no target value was selected because it is not a key property for this application. The same happened for contact angle in wound healing. It may be the case that the target properties are impossible to be achieved simultaneously for a given material, given the trade-off among them. In this case, the proposed estimation method will provide us with a compromise solution in which the estimate properties lie near the target properties. The basic concepts of the *L*-fuzzy contexts theory and the way of applying it to the material formulation selection are discussed in the following section.

4. Modeling of the experiment using *L*-fuzzy contexts

4.1. L-fuzzy concept analysis

Formal concept analysis is a mathematical tool that was developed by R. Wille in 1982(Ganter and Wille, 1999; Wille, 1982) with the aim of processing conceptual knowledge and representing it in a formal way.

A *formal context* is a mathematical structure formed by a triple (X, Y, R), where X and Y are finite sets of *objects* and *attributes* respectively, and $R \subseteq X \times Y$ represents the binary relation among them. Information involved in the context is provided by pairs (A, B) with $A \subseteq X$ and $B \subseteq Y$, called *formal concepts*. The set A is the *extension* of the formal concept and B is its *intension*, and they verify that $A^* = B$ and $B^* = A$, being $(\cdot)^*$ the *derivation operator* that associates objects and attributes as follows: A^* is the set of attributes common to the objects in A and B^* the set of objects having all the attributes in B.

$$A^* = \{y \mid xRy, \forall x \in A\},$$
$$B^* = \{x \mid xRy, \forall y \in B\}.$$

Along these lines, a formal concept can be interpreted as a group of objects A sharing all the attributes of B.

In order to admit different relationship grades among objects and attributes, Burusco and Fuentes-González extended Wille's formal concepts analysis to the fuzzy case with the definition of *L-fuzzy context* (Burusco and Fuentes-González, 1994; 1998; 2000). An *L*-fuzzy context is defined as a tuple (L, X, Y, R) where L is a complete lattice, X and Y are respectively the set of objects and attributes, and the incidence relation $R \in L^{X\times Y}$ takes values in the lattice $(L, \leq)$.

To extract information from these *L*-fuzzy contexts, the derivation operators were defined for all sets $A \in L^X$ and $B \in L^Y$ by means of the following expressions (Burusco and Fuentes-González, 2000):

$$A_1(y) = \underset{x \in X}{inf}\{I(A(x), R(x, y))\}, \forall y \in Y,$$
$$B_2(x) = \underset{y \in Y}{inf}\{I(B(y), R(x, y))\}, \forall x \in X.$$

being I a fuzzy implication operator defined on L.

L-fuzzy concepts constitute the tool that allows to visualize the stored information in the context. These concepts are pairs (M, M_1) where the set $M \in L^X$ is a fixed point of the *constructor operator* φ which is defined using the derivation operators as $\varphi(A) = (A_1)_2 = A_{12}$ for any $A \in L^X$.

Equivalently, the *L-fuzzy concepts* can be defined from the point of view of the attributes as pairs (N_2, N) being $N \in L^Y$ a fixed point of the constructor operator ϕ that is defined $\phi(B) = (B_2)_1 = B_{21}$ for all $B \in L^Y$.

Given a subset of objects $A \in L^X$ (or, a subset of attributes $B \in L^Y$), it is possible to calculate the associated L-fuzzy concept by repeatedly applying the constructor operator φ (or ϕ) until a fixed point is obtained. We can get a fixed point just applying twice the derivation operators if the used implication operator is residuated (Bělohlávek, 1999; Pollandt, 1997). Thus, if I is defined from a t-norm T such that for all $(a, b) \in [0, 1]$

$$I(a, b) = sup\{x \in [0, 1] \mid T(a, x) \leq b\},$$

then the pair (A_{12}, A_1) is the L-fuzzy concept associated with A. Similarly, the L-fuzzy concept associated with $B \in L^Y$ will be the pair (B_2, B_{21}).

The L-fuzzy concept associated with a subset of objects A (or attributes B) provides the “stable” situation in the context closest to the initial conditions fixed by A (or B).

In this work, and in order to simplify the calculations, the implication operator chosen to obtain the derivated sets was Lucasiewicz implication:

$$I(a, b) = min\{1, 1 - a + b\}, \forall a, b \in [0, 1]$$

which is a residuated implication operator.

4.2. Proposed methodology to extract information from experimental data

Methodology proposed in this work takes advantage of the potential that L-fuzzy context analysis have to extract information from relationship tables with double entry, and can be described in the following steps:

1. Determine the sets of objects and attributes involving in the problem. The relation among them will be given by the experimental data.
2. Normalizing the experimental values, construct the L-fuzzy context related to the properties of the analyzed material.
3. Establish the set of attributes that represents the situation that we want study. Properties for which there are no requirements will initially be assumed to be zero.
4. Calculate the L-fuzzy concept closest to the starting situation, which will be given by the fixed point of the constructor operator.
5. Rescale and interpret the obtained values to extract the required information.

Algorithm 1 describes the calculations to be done in order to obtain the estimated values for material properties.

```
Algorithm 1 Estimation of material properties.
Inputs:
  $\{x_1, x_2, \ldots, x_n\}$: set of objects $X$.
  $\{y_1, y_2, \ldots, y_m\}$: set of attributes $Y$.
  $E(x_i, y_j)$ for all $(x_i, y_j) \in X \times Y$: experimental values.
  $\{r_1, r_2, \ldots, r_k\}$: required values for attributes $\{y_{j_1}, y_{j_2}, \ldots, y_{j_k}\}$.
Output: $\{e_1, e_2, \ldots, e_m\}$: estimated values for all the attributes.
Steps:
▶ Construct the relation of the context.
for $i = 1$ to $n$ do
    for $j = 1$ to $m$ do
        $R(x_i, y_j) \leftarrow \frac{E(x_i, y_j) - \min_{1 \leq h \leq n} \{E(x_h, y_j)\}}{\max_{1 \leq h \leq n} \{E(x_h, y_j)\} - \min_{1 \leq h \leq n} \{E(x_h, y_j)\}}$
    end for
end for
▶ Construct the initial set of attributes to represent the required information.
for $j = 1$ to $m$ do
    if $j \in \{j_1, j_2, \ldots, j_k\}$ then
        $B(y_j) \leftarrow \frac{r_j - \min_{1 \leq h \leq n} \{E(x_h, y_j)\}}{\max_{1 \leq h \leq n} \{E(x_h, y_j)\} - \min_{1 \leq h \leq n} \{E(x_h, y_j)\}}$
    else
        $B(y_j) \leftarrow 0$
    end if
end for
▶ Calculate the associated concept.
while $B \neq \phi(B)$ do
    $B \leftarrow \phi(B)$
end while
▶ Obtain the estimated values for the properties.
for $j = 1$ to $m$ do
    $e_j \leftarrow \min_{1 \leq h \leq n} \{E(x_h, y_j)\} + B(y_j) \left( \max_{1 \leq h \leq n} \{E(x_h, y_j)\} - \min_{1 \leq h \leq n} \{E(x_h, y_j)\} \right)$
end for
```

5. Determination of the formulations of gelatin-based films

5.1. Experimental design for formulation optimization

The input values of formulation to be adjusted were chosen to be glycerol content, pH and gallic acid content. In order to feed the proposed methodology with data for the adjustment, a batch of samples was tested, in which the analyzed glycerol content values were 0%, 5% and 10%, the selected pH values 4.50, 7.25 and 10.00, whereas the gallic acid content values were 5%, 10% and 15%. The experimental values of the output properties obtained to feed the process for the construction of the L-fuzzy context are shown in Table 2.

5.2. Construction of the L-fuzzy context representing the experimental data

With the aim of modeling the process of material designing we constructed an L-fuzzy context (L, X, Y, R) in the lattice $(L = \{0, 0.01, 0.02, \ldots, 0.99, 1\}, \leq)$, considering the set of objects $X = \{x_1, x_2, \ldots, x_{13}\}$, where by x_i was represented the ith formulation. The set of attributes $Y = \{y_1, y_2, \ldots, y_{10}\}$ was formed by the different properties that were analyzed in each formulation:

- y_1: Glycerol content.
- y_2: Gallic acid.
- y_3: Solution pH.
- y_4: Water contact angle.
- y_5: Water vapour transmission rate
- y_6: L* colour value.
- y_7: b* colour value.
- y_8: Tensile strength.
- y_9: Elongation at break.
- y_{10}: Gloss value at 60° incidence angle.

The relation among objects and attributes $R \in L^{X \times Y}$ was calculated rescaling the elements of the matrix E formed by the average experimental values in Table 2 as follows:

$$R(x_i, y_j) = \frac{E(i, j) - \min_{1 \leq h \leq 13} \{E(h, j)\}}{\max_{1 \leq h \leq 13} \{E(h, j)\} - \min_{1 \leq h \leq 13} \{E(h, j)\}}$$

The obtained relation is shown in Table 3.

Table 2
Experimental values of functional properties for different formulations of the bio-based material. Parameters of material formulation are glycerol content (GLY), gallic acid (GA) and solution pH. Studied material properties are water contact angle (CA), water vapour transmission rate (WVTR), L* and b* colour values, tensile strength (TS), elongation at break (EB) and gloss values at 60° incidence angle.

Formulation			Bio-based material properties						
GLY (%)	GA (%)	pH	CA (°)	WVTR $\left(\frac{g}{m^2 \cdot day}\right)$	L*	b*	TS (MPa)	EB (%)	Gloss$_{60^\circ}$ (Gloss units)
0	10	4.50	74.8 ± 4.2	1057.7 ± 11.5	95.3 ± 0.5	5.4 ± 0.4	78.7 ±12.5	3.8 ± 1.5	125.4 ± 3.2
5	15	4.50	74.6 ± 7.8	1147.3 ± 16.5	95.5 ± 0.2	5.4 ± 0.8	83.4 ± 8.4	2.7 ± 0.6	155.4 ± 10.3
5	10	7.25	52.8 ± 2.3	1272.1 ± 13.4	50.2 ± 6.6	44.6 ± 6.9	79.8 ± 3.7	3.1 ± 0.4	101.6 ± 8.5
5	5	4.50	58.9 ± 2.2	1097.3 ± 8.0	95.7 ± 0.4	4.8 ± 0.5	80.5 ± 2.1	3.9 ± 0.4	148.0 ± 9.9
0	10	10.00	38.3 ± 2.5	1256.3 ± 10.2	32.5 ± 1.3	20.7 ± 1.8	56.3 ± 5.4	2.7± 0.4	50.1 ± 5.6
10	10	10.00	44.6 ± 1.9	1198.0 ± 22.3	26.6 ± 0.5	8.8 ± 0.5	57.9 ± 1.2	2.6 ± 0.2	29.6 ± 3.7
5	5	10.00	44.7 ± 4.6	1227.0 ± 10.6	69.6 ± 5.7	59.1 ± 3.2	70.8 ± 5.6	3.1 ± 0.4	23.5 ± 2.6
0	5	7.25	56.7 ± 1.8	1206.0 ± 8.5	47.2 ± 0.7	38.6 ± 0.7	77.9 ± 10.4	3.5 ± 0.5	28.6 ± 2.5
10	15	7.25	99.3 ± 2.5	1310.3 ± 19.4	46.6 ± 1.8	42.8 ± 1.5	72.1 ± 6.2	2.7 ± 0.4	100.0 ± 6.4
5	15	10.00	40.8 ± 2.5	1001.7 ± 15.3	23.8 ± 0.9	4.0 ± 0.8	62.3 ± 4.0	2.6 ± 0.3	70.9 ± 3.2
10	10	4.50	101.0 ± 3.7	960.3 ± 22.5	95.2 ± 0.2	6.5 ± 0.9	81.8 ± 8.0	3.1 ± 0.6	148.4 ± 6.1
10	5	7.25	68.2 ± 4.7	1220.0 ± 20.7	52.8 ± 3.9	48.6 ± 1.3	77.5 ± 4.1	3.6 ± 0.4	97.6 ± 1.6
0	15	7.25	67.7 ± 4.2	1304.3 ± 9.3	54.1 ± 1.0	49.8 ± 1.5	86.4 ± 7.9	3.2 ± 0.6	100.0 ± 7.0

Table 3
Relation of the *L*-fuzzy context.

R	y_1	y_2	y_3	y_4	y_5	y_6	y_7	y_8	y_9	y_{10}
x_1	0.00	0.50	0.00	0.58	0.28	0.99	0.03	0.66	0.52	0.77
x_2	0.50	1.00	0.00	0.58	0.53	1.00	0.02	0.80	0.02	1.00
x_3	0.50	0.50	0.50	0.23	0.89	0.37	0.74	0.70	0.21	0.59
x_4	0.50	0.00	0.00	0.33	0.39	1.00	0.01	0.72	0.56	0.94
x_5	0.00	0.50	1.00	0.00	0.85	0.12	0.30	0.00	0.02	0.20
x_6	1.00	0.50	1.00	0.10	0.68	0.04	0.09	0.04	0.00	0.05
x_7	0.50	0.00	1.00	0.10	0.76	0.64	1.00	0.43	0.21	0.00
x_8	0.00	0.00	0.50	0.29	0.70	0.32	0.63	0.64	0.35	0.04
x_9	1.00	1.00	0.50	0.97	1.00	0.32	0.70	0.47	0.02	0.58
x_{10}	0.50	1.00	1.00	0.04	0.12	0.00	0.00	0.18	0.00	0.36
x_{11}	1.00	0.50	0.00	1.00	0.00	0.99	0.04	0.76	0.19	0.95
x_{12}	1.00	0.00	0.50	0.48	0.74	0.40	0.81	0.63	0.40	0.56
x_{13}	0.00	1.00	0.50	0.47	0.98	0.42	0.83	0.89	0.24	0.58

5.3. L-fuzzy concepts associated with the initial requirements

Once defined the *L*-fuzzy context (L, X, Y, R), the characteristics of the material were estimated from the *L*-fuzzy concepts associated with the set of attributes representing the desired requirements shown in Table 1.

In order to get values in *L*, the required values for the properties were normalized following the rescaling method used with relation *R*, and the membership values of the attributes representing the unknown properties were initially supposed to be 0.

Thus, material properties for fatty food packaging application were represented by the following fuzzy set of attributes:

$$B = \{y_1/0, y_2/0, y_3/0, y_4/0.82, y_5/0, y_6/0.99, y_7/0.65, y_8/0.11, y_9/1, y_{10}/0.20\}$$

And, after calculating the associated *L*-fuzzy concept, we focused on its intension:

$$B_{21} = \{y_1/0.63, y_2/0.60, y_3/0.63, y_4/0.89, y_5/0.81, y_6/0.99, y_7/0.65, y_8/0.98, y_9/1, y_{10}/0.70\}$$

The intension of a concept can be interpreted as a set of attributes that are always found together in the context. Hence, the obtained set of attributes B_{21} can be interpreted as the most similar set of attributes to the required set B that are given at the same time.

In the case of the material requirements for wound healing application, we considered the following set $B \in L^Y$:

$$B = \{y_1/0, y_2/0, y_3/0, y_4/0, y_5/0.97, y_6/0.50, y_7/0.64, y_8/1, y_9/0.16, y_{10}/0.20\}$$

The intension of the associated *L*-fuzzy concept was:

$$B_{21} = \{y_1/0.11, y_2/0.36, y_3/0.61, y_4/0.53, y_5/0.97, y_6/0.53, y_7/0.84, y_8/1, y_9/0.35, y_{10}/0.40\}$$

From the membership values of the intension of the obtained *L*-fuzzy concepts, reversing the rescaling process, we estimated that, for fatty food packaging and wound healing applications, the material properties should be as shown in Table 4.

6. Results and discussion

Given the experimental data for the *L*-fuzzy concept analysis, the estimated formulation values for an optimal performance are shown in Table 4. For a fatty food packaging application, this methodology suggests values of around 6.3% in glycerol content, 11% of gallic acid content and a pH of 8; whereas for wound healing use, the predicted values are approximately 1% of glycerol, 9% of gallic acid and a pH of 8.

In reference to fatty food packaging application, as can be observed in Table 5, the estimated achievable values were very similar to the target functional properties values, apart from gloss values and tensile strength, and the proposed values by the *L*-fuzzy concept analysis were higher than the selected objective values. As far as gloss values are concerned, according to the obtained estimation, obtaining values near to 50 is not possible taking into account the constraints in the rest of the properties, suggesting that gloss values must be greater. Since gloss values are inversely related to the roughness of the film surface (Sánchez-González et al., 2010), greater gloss values would indicate smoother surfaces. Something similar happened with tensile

Table 4
Estimated formulation and material functional properties values for fatty food packaging and wound healing applications.

	GLY (%)	GA (%)	pH	CA (°)	WVTR $\left(\frac{g}{m^2\cdot day}\right)$	L*	b*	TS (MPa)	EB (%)	Gloss$_{60^\circ}$ (Gloss units)
Fatty food packaging	6.28	10.96	7.95	93.94	1243.66	95.00	40.00	89.38	5.00	116.27
Wound healing	1.08	8.59	7.84	71.74	1300.01	61.83	50.00	90.00	3.44	75.96

Table 5
Material requirements, fuzzy estimation, experimental values and obtained error for fatty food packaging and wound healing applications. Parameters of material formulation are glycerol content (GLY), gallic acid (GA) and solution pH. Studied material properties are water contact angle (CA), water vapour transmission rate (WVTR), L* and b* colour values, tensile strength (TS), elongation at break (EB) and gloss values at 60° incidence angle.

	Formulation			Bio-based material properties						
	GLY (%)	GA (%)	pH	CA (°)	WVTR $\left(\frac{g}{m^2\cdot day}\right)$	L*	b*	TS (MPa)	EB (%)	Gloss$_{60^\circ}$ (Gloss units)
Food packaging - requirements				90		95	40	60	5	50
Estimated values	6.3	11	8	93.9	1243.7	95	40	89.4	5	116.3
Experimental values				74.3	1370.3	60.9	42.1	83.3	4.8	95.3
Error(%)				20.9	10.2	35.9	5.3	6.7	4.9	18
Wound healing - requirements					1300	60	50	90	3	50
Estimated values	1	9	8	71.7	1300	61.8	50	90	3.4	76
Experimental values				59.1	1321	60.6	42.6	80.6	4.6	96
Error(%)				17.6	1.6	2	14.8	10.4	33.5	26.4

strength values, estimated values are greater than the target. It is worth mentioning that the mechanical behavior of the material proposed by the estimation method is even better than that of the specification, having greater tensile strength values without decreasing elongation at break values. In the case of the water vapor transmission rate, as for fatty food packaging application no target value was selected due to the fact that it is not a key property for this application, for the proposed formulation the *L*-fuzzy concept estimates a value of 1243 $\frac{g}{m^2\cdot day}$ for this property.

To verify the employed estimation method, a bio-based material was developed with the proposed formulation and the experimental responses of material functional properties were measured. As shown in Table 5, for the majority of the properties, the values given through the *L*-fuzzy context are reasonably similar to the experimental values. The obtained errors are less than 21% for all cases, apart from L* colour value in which the error is higher and the obtained experimental value is lower than the estimation. This gives rise to a higher luminosity of the film, greater than the specified in the requirement, which is adequate for the food packaging application. Moreover, it could be confirmed that, as the *L*-fuzzy concept predicted, gloss values were higher than the objective values and the mechanical performance of the material is even better than required for the estimated formulation.

Concerning wound healing application, the formulation proposed by our estimation method was approximately 1% glycerol content, 9% gallic acid content and a pH of 8. It is worthy of note that values of the functional properties of that formulation were very close to the defined objective values for the application. Gloss is again the principal magnitude that is not so close to the required value, and it was slightly higher than the estimation. In the case of the material application for wound healing, no target for contact angle was selected as it is not considered a key factor, and the estimation given by the *L*-fuzzy concept was 71.7° for this functional property.

As in the case of fatty food packaging application, for the validation of the estimation method, a material with the proposed formulation was developed and its functional properties were measured. Obtained material performance was very similar to the predicted by the *L*-fuzzy concept, as the measured values for most of the properties were very close to the values obtained by the calculated estimation. Obtained errors were lesser than 18% except in gloss values and elongation at break. For these properties, the errors were 33.5% and 26.4% respectively. Experimental values of elongation at break were higher than the estimated values provided by the *L*-fuzzy concept, giving rise to a slight improvement in the mechanical performance of the material, although tensile strength values were slightly lower than expected. Gloss values are related with the surface of the material. Having greater values in the bio-based material for wound healing application could lead to a decrease of the film roughness. Although the values were greater than the specification, the properties obtained lead to a better performance of the material.

7. Conclusions

In the present work, *L*-fuzzy concept analysis was employed as decision-making tool to support the material formulation decision for obtaining required functional properties for fatty food packaging and wound healing applications. Results obtained after the implementation of the proposed methodology revealed that values predicted by the *L*-fuzzy concepts were very similar to the target values for the majority of the properties for both applications. Moreover, after analyzing the material performance with the estimated formulations, the experimental values were very close to the proposed approximate ones. The error between the approximate values estimated by the *L*-fuzzy concepts and experimental values carried out with the proposed formulation were lesser than the 20% in most of the functional properties. In addition, in the cases in which the variation between predicted value and experimental value was greater than the 20%, this difference causes an improvement of the required properties, obtaining a better performance of the material.

Definitely, the formulations proposed by *L*-fuzzy concept analysis would be the best starting point for studying the effect of each parameter in the functional properties of the material. Instead of having a vast array of formulations to study, using this estimation method helps to identify approximately the values of the parameters in the formulation to obtain the desired properties, and to know if it is possible to obtain those functional properties for a specific system.

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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Itsaso Leceta: Data curation, Funding acquisition, Conceptualization, Investigation, Methodology, Supervision, Validation, Writing - original draft, Writing - review & editing. **Cristina Alcalde:** Conceptualization, Investigation, Formal analysis, Software, Methodology, Supervision, Validation, Writing - original draft, Writing - review & editing. **Marta Urdanpilleta:** Data curation, Funding acquisition, Conceptualization, Investigation, Methodology, Supervision, Validation, Writing - original draft, Writing - review & editing. **Pedro Guerrero:** Data curation, Funding acquisition, Conceptualization, Investigation, Methodology, Supervision, Validation, Writing - original draft, Writing - review & editing. **Koro de la Caba:** Data curation, Funding acquisition, Conceptualization, Investigation, Methodology, Supervision, Validation, Writing - original draft, Writing - review & editing. **Ana Burusco:** Formal analysis, Software, Conceptualization, Investigation, Methodology, Supervision, Validation, Writing - original draft, Writing - review & editing.

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