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A FUZZY LOGIC APPROACH FOR MATHEMATICAL MODELING OF THE EXTRACTION PROCESS OF BIOACTIVE COMPOUNDS

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Abstract. The aim of the present study was to optimize the extraction process of bioactive compounds from berries and wastes from the agro-food industry (grape marc). Mathematical models of the extraction process of biologically active compounds based on algorithms of artificial intelligence: fuzzy logic and neuro-fuzzy algorithms have been established. The mathematical models, which use the experimental average values of uncertain models, as well as of some predictive models, offer values of the sizes with a large prediction horizon. It was established, that mathematical models, which use the experimental average values of uncertain models, the experimental data, as well as of some predictive models offer values of the sizes with a large prediction horizon. The existence of various interactions between the influence factors (ethanol concentration, extraction temperature, pretreatment method) and the measured parameters (total polyphenol index, quantity of tannins extracted and antiradical activity, DPPH) was established. The great diversity of processes at different products and various parameters, as well as the existence of non-linear dependencies between sizes, allow credible extrapolations of the results only within the experimental limits.

Keywords: *fuzzy mathematical model, neuro-fuzzy mathematical model, berries, extraction, bioactive compounds.*

Introduction

Classical statistics is based on the law of large numbers, which requests many experimental values. In the case of costly experiments with practical values less numerous, the formulated results can be questionable, because classical statistics offers a single prediction horizon, which is a disadvantage in terms of the conclusions credibility. Various algorithms of artificial intelligence can be applied to establish mathematical models. Thus, it is possible to call on the fuzzy sets, neural networks, neuro-fuzzy algorithms, genetic algorithms, etc. [1].

Fuzzy logic is a type of logic with a series of values specified as a degree of truth instead of true or false binary values [2]. It is considered that the most important application of fuzzy logic is in uncertainty management [3]. Fuzzy logic is a powerful and appropriate tool for managing complex problems in a position where data is incomplete or not very accurate [13]. There are many applications in the field of life sciences, for example, in the risk analysis of some diseases [4, 5], in the analysis of genetic expression data [6-8], in the modeling of enzymatic kinetics [9, 10]. The fuzzy reasoning (the fuzzy algorithm / logic) supposes the execution of rules that link the values of the factorial size (influence factors or independent variables) to those of the resultant size (experimentally measured parameters or dependent variables). These rules are usually created deductively either by man or by a calculation algorithm. Regardless of the system, there are three specific basic steps of establishing a fuzzy model. These are the fuzzification of the factorial and resultant sizes, the generation of the rules base and the convergence of the result (defuzzification) [2, 3]. It is well known that one of the trends of the modern food industry is the complex valorisation of bioactive compounds in natural products and the decrease of the synthetic additives rate [11, 12]. The aim of the present study was to optimize the extraction process of bioactive compounds from berries and wastes from the agro-food industry (grape marc). These raw materials are rich in bioactive compounds - polyphenols, carotenoids, which are of particular interest for the food and pharmaceutical industries [13-16].

The purpose of mathematical modeling consisted in establishing some predictive elements, which allow a good interpolation of the data, assures the highest credibility of the experimental results, including for the values of the influence factors on the extraction process (concentrations of ethyl alcohol), which cannot be found experimentally, as well as establishing the most accentuated and the weakest interdependencies between the measured parameters.

Materials and methods

The experimental research aimed at 3 products (extracts of bioactive compounds from berries and agro-food wastes) and a maximum number of 3 experimental parameters determined [17]. The 3 parameters, symbols and units of measurement used for them are the following:

- 1 – total polyphenols index, symbols P4;
- 2 – the antiradical activity, DPPH, in acidic medium, symbols P5 [%];
- 3 - the quantity of tannins extracted, symbols P9 [mg·3g⁻¹].

The 3 targeted products and the used symbols are:

- 1 – aronia melanocarpa, symbol "a";
- 2 – grape marc, symbol "d";
- 3 – hawthorn, symbol "p".

The experimental data represent finite discrete series, obtaining 3 values of the 3 parameters at each concentration of ethyl alcohol; in 2 products there are 5 concentrations of alcohol (20%, 40%, 50%, 60%, 80%) and in the grape marc there are 6 concentrations (20%, 40%, 50%, 60%, 80%, 96%).

Lotfi A. Zadeh introduced for the first time in 1973 the fuzzy linguistic model, which is a set of written rules in general form. Thus, for two finite discrete sizes some x and y:

$$R_i: \text{IF } x \text{ is } A_i \text{ THEN } y \text{ is } B_i, i=1, 2, \dots, k \quad (1)$$

where: x is the input linguistic variable (of the factorial size),

A_i - the linguistic term (a constant) of the input,

y - the linguistic variable of output (of the resultant size),

B_i - the linguistic term (a constant) of the output. The language terms A_i and B_i are predefined, for example by the form {small (Mc), medium (M), large (Ma)}.

Later, more advanced forms of fuzzy language models emerged. For example, in the Takagi-Sugeno model the set of rules has the general form [18]:

$$R_i: \text{IF } x \text{ is } A_i \text{ THEN } y_i = f_i(x), i = 1, 2, \dots, k \quad (2)$$

The simplest Takagi-Sugeno model is the one in which the functions f_i are straight and so the expression (2) becomes:

$$R_i: \text{IF } x \text{ is } A_i \text{ THEN } y_i = a_i x + b_i, i = 1, 2, \dots, k \quad (3)$$

Based on the above, applying the Takagi-Sugeno algorithm and using the experimental average values, nominal fuzzy models were obtained. Because the systems examined have a multitude of influence factors and many interdependencies, the combined use of neural networks and fuzzy sets was applied to establish the mathematical model (neuro-fuzzy models). For this, the ANFIS (Adaptive NeuroFuzzy Inference System) algorithm was applied using the Matlab program toolbox [19].

Results and discussions

The first step in establishing a mathematical model using fuzzy logic is the fuzzification of the factorial (independent variables) and resultant (dependent variables) sizes of the target process. This is achieved by constructing a function correlated to each from the factorial/resultant sizes. Theoretically, there is infinity of possible forms for these functions, more commonly the triangular, Gaussian or trapezoidal ones being used. Figure 1 shows the example of a triangular shape function to describe the concentration of ethyl alcohol in the 20-80% range; the purpose is to establish the model of type $P4 = f(C_a)$ at the aronia melanocarpa.

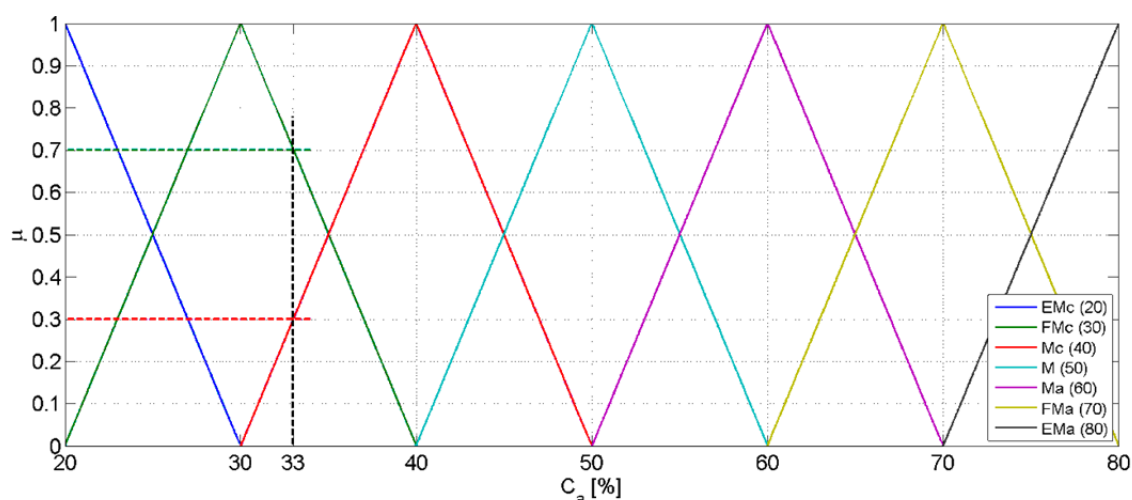


Figure 1. Triangular fuzzy sets (7) for the concentrations of ethyl alcohol (C_a) from aronia fruits in the range 20% - 80%.

The number of corresponding functions can be any number of linguistic variables. The 7 linguistic variables corresponding to the example in figure 1 are: extremely small (EMc, at

which C_a varies around 20%), very small (FMc), small (Mc), medium (M), large (Ma), very high (FMa) and extremely high (EMa), to which are appropriately assigned the values 20%, 30%, 40%, 50%, 60%, 70% and 80%; in the graph, μ represents the multiplication function. Once the correlative function is established for each size, the actual inputs/outputs are fuzzified. First, the input is read as a fixed value. It is considered, for example, that the input is introduced as 33% concentration of ethyl alcohol (figure 1). Then a vertical line is drawn on the axis of the abscissa near $C_a = 33\%$ to indicate its point of intersection with each component of the correlated function. The vertical line intersects on "Mc" to the value 0.3 and "FMc" at 0.7. In linguistic terms, an input of 33 is considered to be 30% small (Mc) and 70% very small (FMc). These are the dispersed values of the C_a input; once this process was completed for all values of the input size, the fuzzification step ended.

To generate the rule base, the correlative function P4 of the outputs must be defined in predetermined. For example, the correlative function in the form of a triangle for the total polyphenols index P4 in aronia extracts, as in Figure 2b. It was found that the linguistic variables corresponding to the output (parameter P4) are defined as: Very Small (FMc), Small (Mc), Medium (M), High (Ma), Very High (FMa) to which are associated values 7; 9.25; 11.5; 13.75 and 16. These values are shown by horizontal lines and in Figure 2a, where both the fuzzy sets for the concentrations of hydroalcoholic extracts (those in Figure 1) and the values of parameter P4 appear. Figure 2 shows the rule base sought, as a series of logical statements "IF-THEN". For example:

$$\text{IF } C_a = \text{FMc} \quad \text{THEN} \quad P4 = \text{Mc} \quad (4)$$

or, otherwise expressed: if C_a is around 30%, then parameter P4 has values around 9.25%.

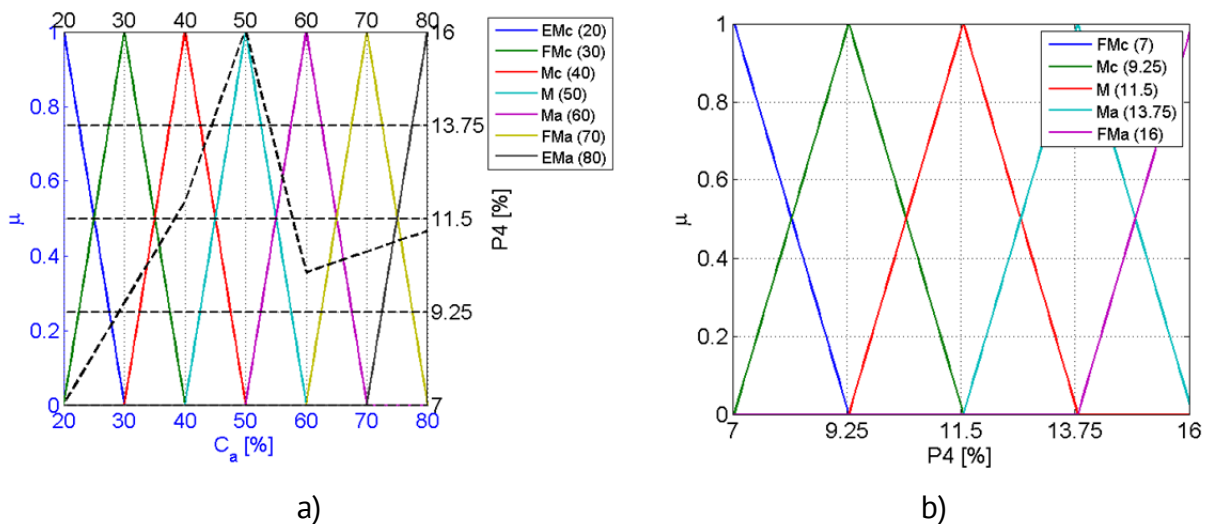


Figure 2. Triangular fuzzy sets (7) for: a) the concentrations of ethyl alcohol (C_a) from aronia fruits in the range 20% - 80%; b) the total polyphenols index P4 in aronia extracts.

Or, another example:

$$\text{IF } C_a = \text{M} \quad \text{THEN} \quad P4 = \text{FMa} \quad (5)$$

otherwise expressed: if C_a is around 50%, then parameter P4 has values around 16% (the maximum for P4 in Figure 2a).

Convergence (defuzzification) is the process of converting dispersed outputs into a single or fixed output value. This process can be accomplished by a few convergence methods. Some common methods include the principles of maximum correlation, centroid method and multiplication method. To identify the fixed value of output y^* by the multiplication method, it is calculated the sum of the multiplications of each multiplication function, μ_y , with the corresponding maximum correlation value and is divided it by the sum of the multiplication functions:

$$y^* = \frac{\sum [\mu_y(\bar{y}) \cdot \bar{y}]}{\sum \mu_y(\bar{y})} \quad (6)$$

It is now considered that the dynamics of a process / system is described on the input-output relationship by the nonlinear regressive model written in the general form [3]:

$$y(k+1) = f(y(k), y(k-1), \dots, y(k-na+1), u(k), u(k-1), \dots, u(k-nb+1)) \quad (7)$$

where: u is input size (factorial variable);

y - output size (resultant variable);

$f(\bullet)$ - nonlinear function.

Expression (7) represents the NARX model (Nonlinear AutoRegressive with eXogenous input), the correspondent of the linear model ARX (AutoRegressive with eXogenous input).

In this case the fuzzy language model has the set of form rules:

$$\begin{aligned} R_i: \text{ IF } y(k) \text{ is } A_{i_1} \text{ and } y(k-1) \text{ is } A_{i_2} \text{ and } \dots y(k-n+1) \text{ is } A_{i_n} \\ \text{ and } u(k) \text{ is } B_{i_1} \text{ and } u(k-1) \text{ is } B_{i_2} \text{ and } \dots u(k-m+1) \text{ is } B_{i_m} \\ \text{ THEN } y(k+1) \text{ is } C_i \end{aligned} \quad (8)$$

For the case of the Takagi-Sugeno algorithm the mathematical model from the expression (8) becomes:

$$y^i(k+1) = \sum_{j=1}^{na} A_j^i y(k-j+1) + \sum_{j=1}^{nb} B_j^i u(k-j+1) \quad (9)$$

Consequently, the one-step prediction of the output size is:

$$y(k+1) = \sum_{i=1}^c \beta_i(u(k)) y^i(k+1) \quad (10)$$

where c represents the number of rules, and β_i the weight of rule i .

The relation (10) can be written as:

$$y(k+1)^T = \sum_{i=1}^c \beta_i(u(k)) [\Phi(k) I_{1 \times na}] \theta_i^T \quad (11)$$

in which $\Phi(k)$ represents the regression matrix (the regression matrix for the input and output sizes):

$$\Phi(k) = [y(k), \dots, y(k-na+1), u(k), \dots, u(k-nb+1)]^T \quad (12)$$

and θ_i the matrix of the parameters of the local model i (rule i):

$$\theta_i = [A_1^i, \dots, A_{na}^i, B_1^i, \dots, B_{nb}^i] \quad (13)$$

Based on the relationships presented, the values for the parameters of the fuzzy model are obtained, using for example the least squares method:

$$\theta_i = [\Phi^T \Psi_i \Phi]^{-1} \Phi^T \Psi_i Y \quad (14)$$

In the expression (14) it was noted:

$$\Phi = [\Phi(1) \quad \Phi(2) \quad \dots \quad \Phi(n)]^T; Y = [y(2) \quad y(3) \quad \dots \quad y(n+1)]^T \quad (15)$$

and respectively:

$$\Psi_i = \begin{bmatrix} \beta_i(1) & 0 & \dots & 0 \\ 0 & \beta_i(2) & \dots & 0 \\ \dots & \dots & \dots & \dots \\ 0 & 0 & \dots & \beta_i(n) \end{bmatrix} \quad (16)$$

According to the presented relations, it turns out that the values of the resulting size are calculated with a written expression in compact form as follows:

$$Y = \Psi \theta \quad (17)$$

The following is a mathematical model that offers the interdependence between parameter P4 (resultant size, total polyphenols index), concentration of ethyl alcohol C_a and parameter P5 (the antiradical activity, DPPH, in acidic medium, AAA). The analysis of the experimental data showed that there are dependencies between the various measured parameters.

Being two factorial sizes, we adopt fuzzy spatial sets, here triangular, which has on the right side the graduated scale in values and colors with μ_i values.

As shown in Figure 3, 6 fuzzy sets along the C_a axis and 6 sets along the P5 axis were adopted for the mathematical model $P4 = f(C_a, P5)$, so a total of 36 sets; as a result, the fuzzy model will have 36 coefficients Θ in expressions (13) and (17).

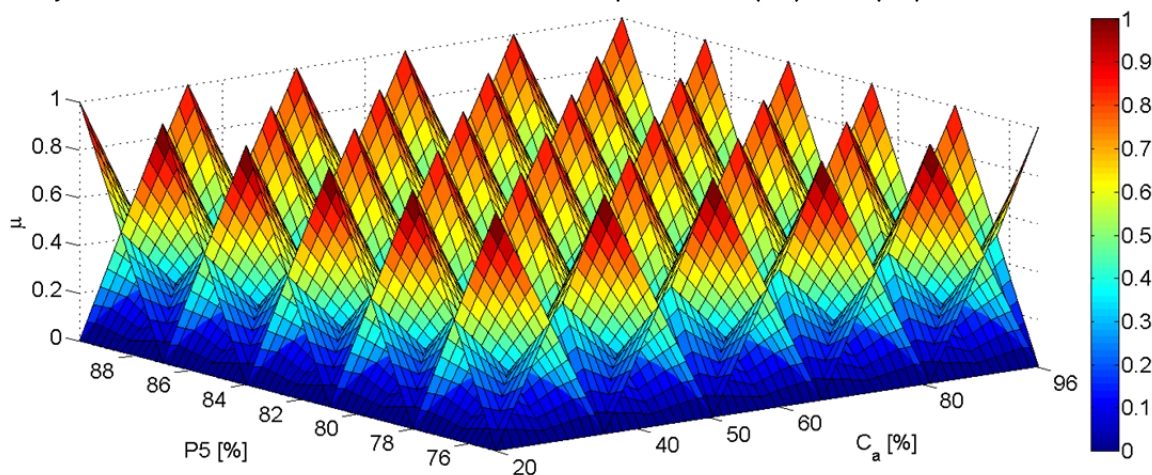


Figure 3. Fuzzy mathematical model $P4 = f(C_a, P5)$ at the grape marc, fuzzy triangular sets (6 and 6).

Figure 4 shows the fuzzy calculation surface, on which the points with the experimental values of parameter P4 are arranged.

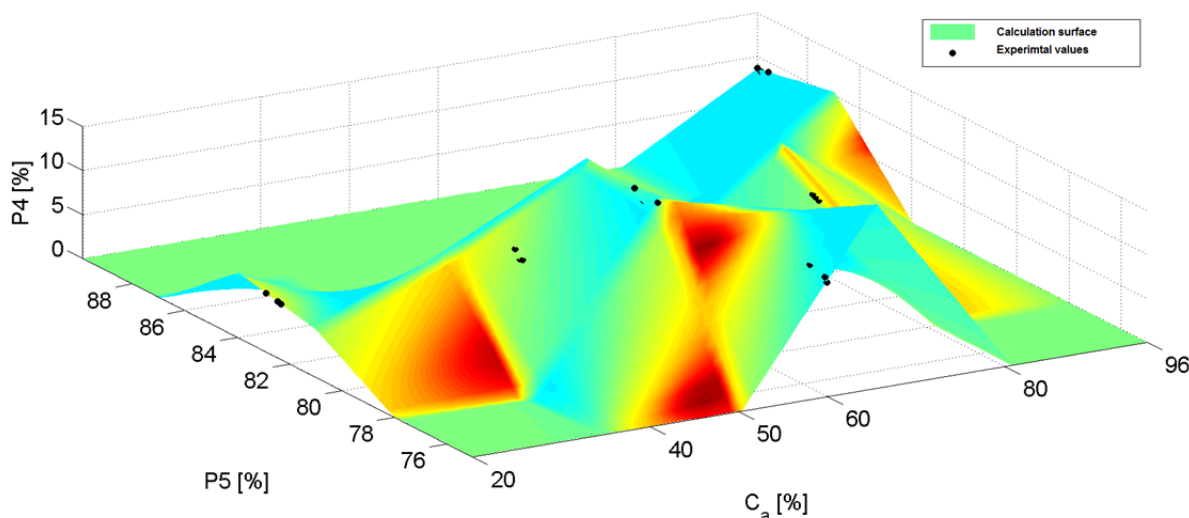


Figure 4. Fuzzy mathematical model $P4 = f(C_a, P5)$ at the grape marc, experimental values and calculation surface.

The graph in Fig. 5a contains the weights of the fuzzy sets β_i from the expression (11), which is the matrix Ψ_i from the relation (16), respectively Ψ from the general formula (17) of the mathematical model.

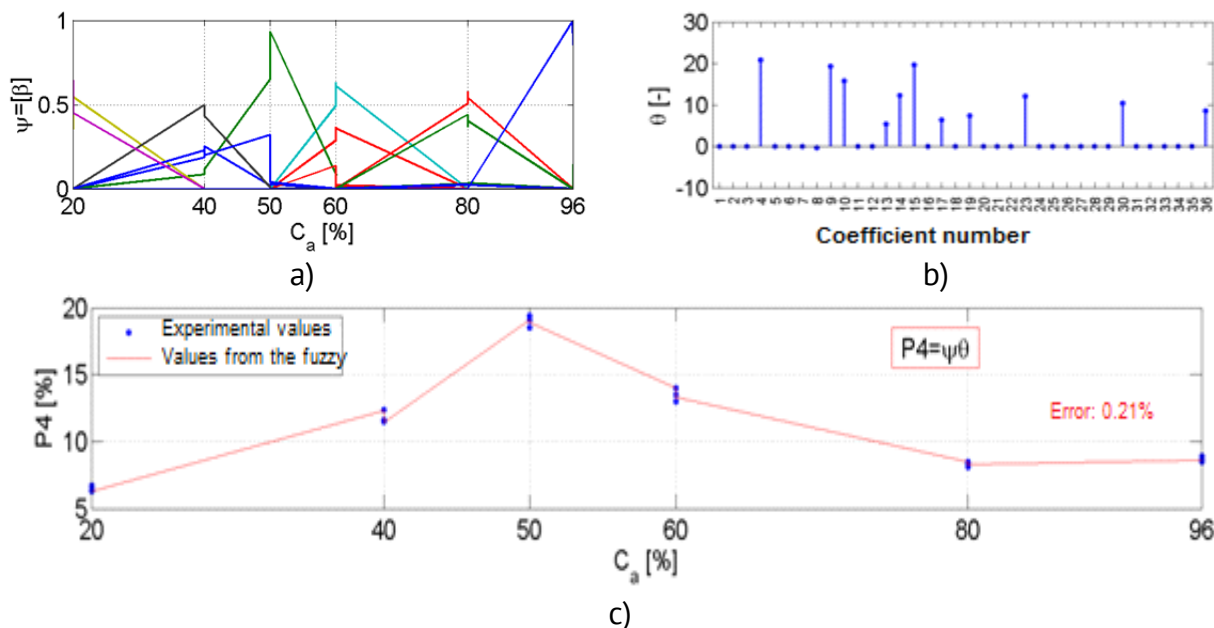


Figure 5. Fuzzy mathematical model $P4 = f(C_a, P5)$ at the grape marc, weights, coefficients, experimental values from the fuzzy model.

Figure 5b shows the values of the 36 coefficients θ_i from the expression (14) of the fuzzy model, so the vector Θ from the relation (17) that also appears on the graph in Figure 5c, where the experimental and fuzzy model values are presented, as well and modeling error, which is acceptable.

Mathematical models based on neuro-fuzzy algorithms

In this case we resort to the combined use of neural networks and fuzzy sets to establish the mathematical model (neuro-fuzzy models). For this, the ANFIS (Adaptive

NeuroFuzzy Inference System) algorithm is applied using the Matlab software toolbox; this toolbox uses the previously presented fuzzy sets (membership function) as well as others [19]. The principle diagram of the ANFIS algorithm is presented in Fig.6, where its main elements are shown.

The ANFIS algorithm applies neuro-adaptive learning techniques, which provide the needed data for modeling with the help of fuzzy sets.

Using input and output data (factorial variables and resultant sizes), a system is

constructed whose fuzzy sets adjust the coefficients of the mathematical model by a neural networks specific algorithm. For this purpose, the number of activation functions (used in neural networks - figure 7) must be equal to that of the fuzzy rules, and the algorithm is based on the fuzzy neuron. The ANFIS architecture is similar to the Takagi-Sugeno algorithm. It is considered, that the system is characterized by two input sizes u_1 and u_2 and an output size y . Consequently, if, for example, the basis of Sugeno-type rules of the first order (linear variation) is adopted, then it results:

$$\text{IF } u_1 \text{ is } A_1 \text{ and } u_2 \text{ is } B_1 \text{ THEN } y_1 = c_{11}u_1 + c_{12}u_2 + c_{10} \tag{18}$$

and respectively:

$$\text{IF } u_1 \text{ is } A_2 \text{ and } u_2 \text{ is } B_2 \text{ THEN } y_2 = c_{21}u_1 + c_{22}u_2 + c_{20} \tag{19}$$

in which c_{ij} are the coefficients of the mathematical model, i. e. the managed parameters by the fuzzy sets that adjust their values through a neural networks specific algorithm.

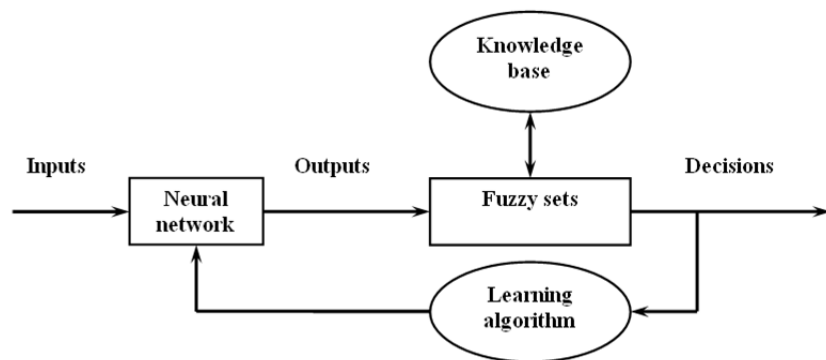


Figure 6. Principle diagram of the ANFIS algorithm.

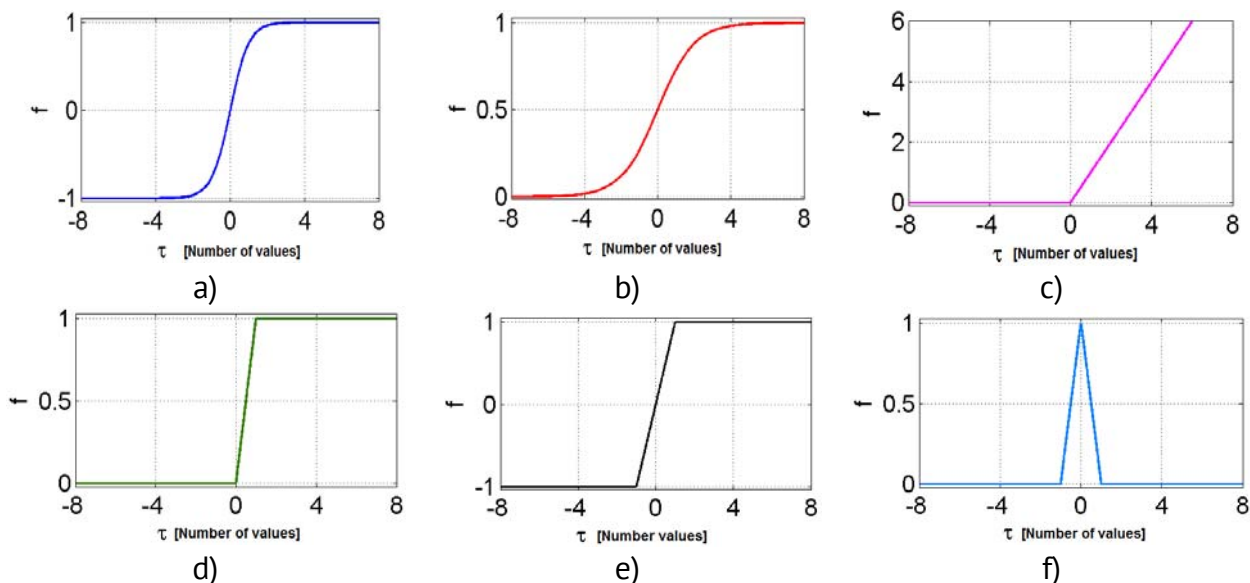


Figure 7. Discrete transfer functions used in neural networks and neuro-fuzzy algorithms: a) tansig type; b) logsig type; c) poslin type; d) satlin type; e) satlins type; f) tribas type.

Figure 8 presents the results of applying the ANFIS algorithm for establishing a mathematical model that offers the values of parameter P2, hawthorn, depending on the concentration of ethyl alcohol. It can be seen that in the upper graphs 13 fuzzy Gaussian sets were used, and in the lower trapezoidal ones.

In this case it followed to establish the nominal model, because it operates with the average values at each concentration of ethyl. Modeling accuracy is all the better as the number of fuzzy sets adopted is higher. The results of the mathematical modeling carried out can be presented in tabular form, for all 6 products concerned, for all the measured parameters, with a calculation step of 2 - 4% of the concentration of the ethyl alcohol with the best precision (the smallest modeling error).

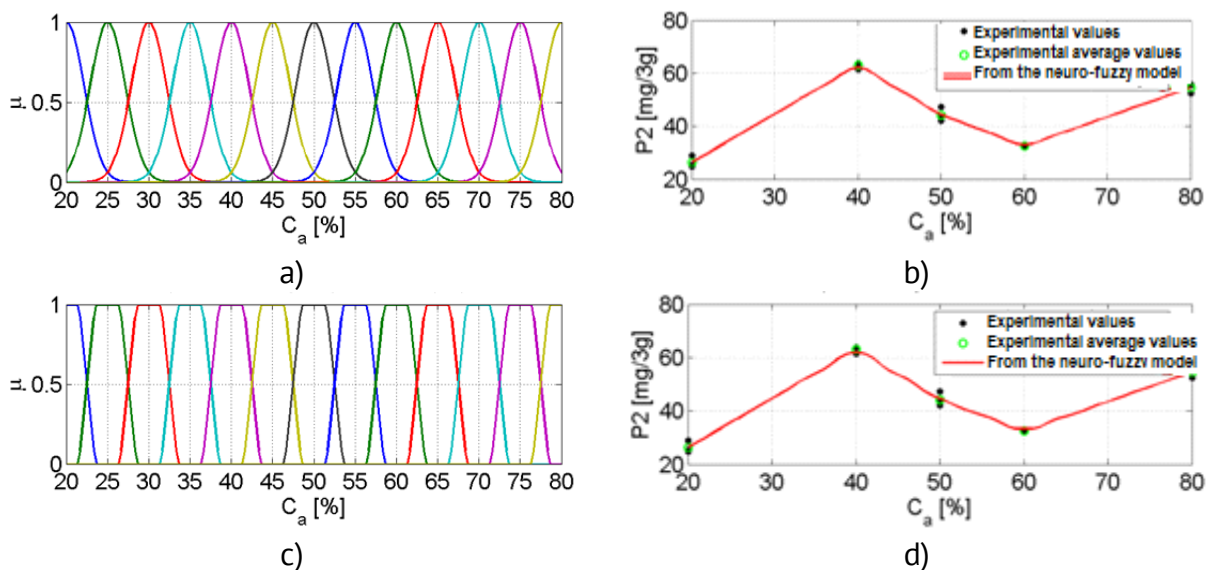


Figure 8. Neuro-fuzzy mathematical model (ANFIS algorithm), parameter P2 from hawthorn depending on the concentration of ethyl alcohol.

For example, Figure 9 shows the nominal / average values (index n) of the tannin mass $P9_n$, the extracted tannin mass $P9_e$ and the extracted total mass $P9_t$ depending on the number of extractions k (with $k \in Z^+$, with Z^+ the set of positive integers).

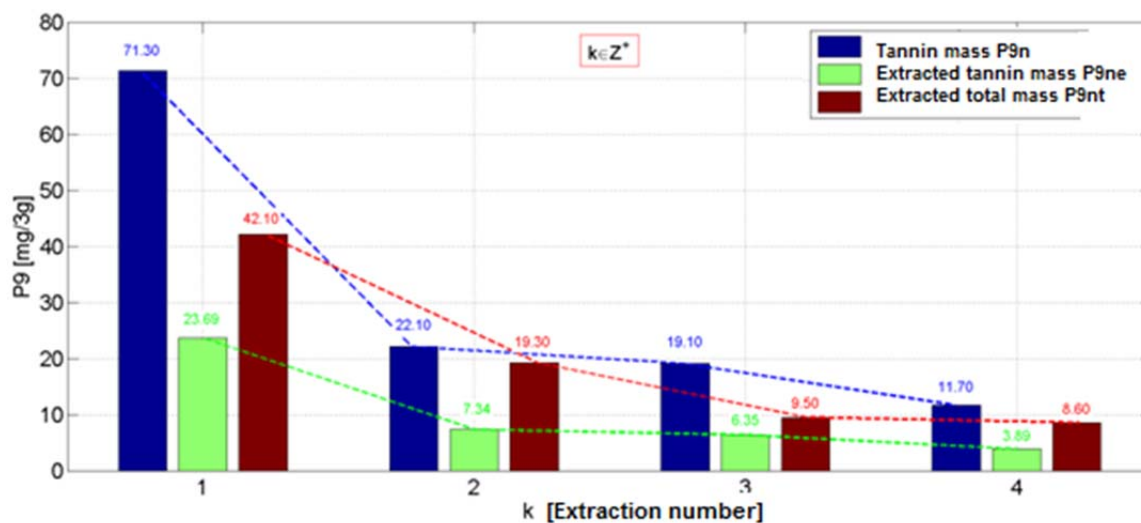


Figure 9. Nominal / average values (index n) of tannin mass $P9_n$, extracted tannin mass $P9_{ne}$ and extracted total mass $P9_{nt}$ depending on the number of extractions.

The values decrease with the order of extraction. This aspect is also confirmed in Figure 10, which shows the nominal and uncertain mathematical models, as well as the analytical expressions of the mathematical models on the 3 portions of the curve (indices 1, 2, 3), both nominal ones (index n), as well as and the uncertain upper (index s) and lower ones (index i). For example, to determine the total mass of tannins at the fourth extraction (at $k = 4$), then the expressions on the third portion are applied, where k is introduced as a difference from the left end, i. e. for $k=4-3=1$. As a result, for the nominal value: $P9_{m_3}(4) = 0.151 \cdot 1^3 + 0.597 \cdot 1^2 - 1.649 \cdot 1 + 9.5 = 8.6 \text{ mg} \cdot 3\text{g}^{-1}$, so exactly the experimental value.

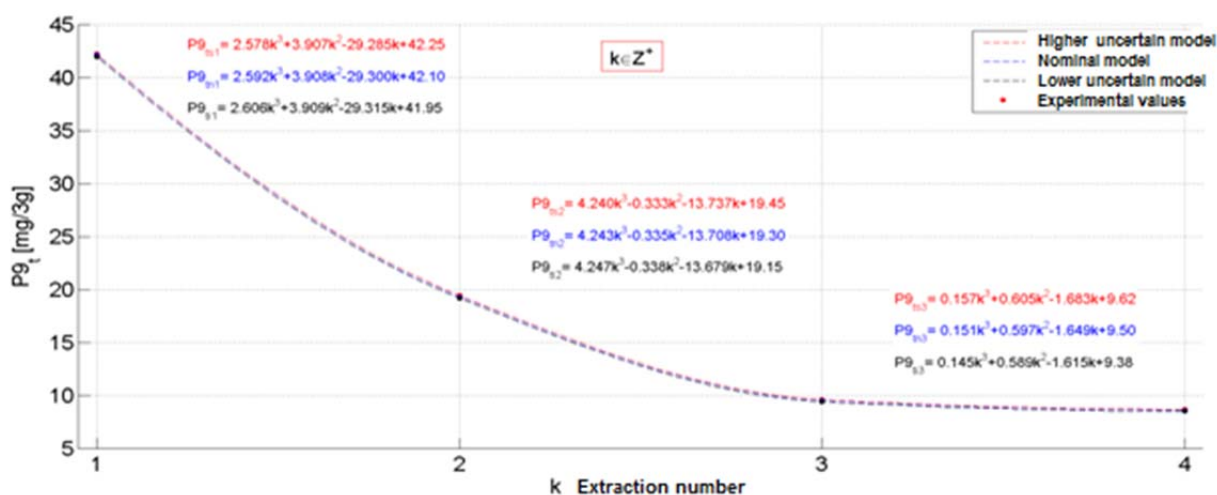


Figure 10. Mathematical modeling of the total mass of tannins according to the number of extractions.

Obviously, for the left end (at $k = 3$), in the previous expression the value $k = 0$ must be entered, where it results: $P9_{m_3}(3) = 0.151 \cdot 0^3 + 0.597 \cdot 0^2 - 1.649 \cdot 0 + 9.5 = 9.5 \text{ mg} \cdot 3\text{g}^{-1}$, so exactly the experimental value. Obviously, mathematical models can be established, including through value tables for any experimental duration, and for each of them other conditions other than experimental ones are adopted.

Conclusions

Establishing mathematical models based on experimental data allowed:

- ✓ deduction of mathematical models in analytical and graphical form;
- ✓ establishing the values of the targeted parameters and of the influencing factors (concentration of ethyl alcohol) that are not experimentally found;
- ✓ deduction of mathematical models based on artificial intelligence algorithms such as fuzzy logic and neuro-fuzzy algorithm;
- ✓ establishing some mathematical models that offer the values of the measured parameters according to the influence factors, as well as models that establish the interdependencies between these parameters;
- ✓ deduction of some nominal mathematical models, which use the experimental average values of uncertain models, which use all the experimental data, as well as of some predictive models, which offer values of the sizes with a large prediction horizon.

Mathematical models that use artificial intelligence algorithms, such as fuzzy logic and neuro-fuzzy algorithm, indicate the existence of diversified phenomena between influence factors and measured parameters.

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