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ROBUST FUZZY MODELS FOR ULTRASONIC POLYMER DEGRADATION

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Abstract: Fuzzy Logic Models are practical solutions to reach a definite conclusion in data sets with uncertain, complicated, and incomplete input data. Owing to these models, achieving the desired outputs with very low error in large data sets obtained theoretically or experimentally is possible. In this study, a subtractive clustering based fuzzy model approach has been presented to analyze the ultrasonic polymer degradation. Fuzzy models include obtaining cluster centers from the data set, preparing a fuzzy rule-based linear equation system, and optimizing parameters for the least error. The designed fuzzy models have high accuracy and clearly express ultrasonic degradation behavior.

Keywords: Ultrasound, Polymer Degradation, Subtractive Fuzzy Clustering, Fuzzy Inference System, Fuzzy Modeling.

1. INTRODUCTION

According to frequency range, ultrasound has various usage areas. In the 16 kHz—1 MHz frequency range, ultrasound can interact with the material and cause physical and chemical changes in the relevant material. In this range, known as the sonochemistry region, ultrasound is utilized in many physical and chemical applications [1].

Ultrasonic polymer degradation is a degradation process that occurs due to the mechanical, chemical and thermal effects created by high-frequency sound waves on polymer chains. [2]. This process is based on the acoustic cavitation phenomenon, which is caused by microscopic gas bubbles that form in a liquid medium and collapse rapidly. The sudden release of energy as the bubbles collapse causes the polymer chains to break by creating excessive heat and pressure [3]. This method has the advantages of reducing the need for chemical reactors and providing an environmentally friendly alternative, while providing controlled degradation of polymers. This technique, which is especially preferred in biomedical applications, plastic recycling processes and chemical synthesis studies, stands out as a more sustainable option in cases where the use of solvents is not required [4-5].

Simulating reactions such as ultrasonic polymer degradation is of great importance in providing a better understanding of the process and reducing experimental costs. While experimental studies are often time-consuming and costly, computer-aided simulations can help determine how polymer chains are broken down, which parameters affect the degradation rate, and optimum operating conditions. In addition, simulations can help predict the effects on different polymer types, preventing unnecessary trials. This provides a great advantage, especially in polymer recycling processes critical for biomedical applications and environmental sustainability. In addition, free radical mechanisms and acoustic cavitation effects that occur during ultrasonic degradation can be analyzed in detail, allowing more controlled and efficient processes to be designed [6-8]. Cluster analysis is a collection of methods for separating data stacks whose groupings are unknown in a data set into similar subsets. Considering the distance between each data in a large data set, collecting similar data in the same clusters, and estimating which group the newly added data belongs to constitute cluster analysis's basis. Clustering aggregates data, increasing process speed and allowing to evaluate data set under a more general structure. Clustering procedures aim to determine the most suitable cluster centers to represent the data set using the distances between the data [9-10].

Fuzzy models are solution tools for defining the relationships between input and output variables with the help of fuzzy rules. The most important difference between fuzzy models and other black box models (e.g., artificial neural networks, genetic algorithms) is that they simplify system identification and allow for transparent analysis [11]. Due to the success of usage in plenty of areas, there has been an increase in data-based fuzzy modeling studies in recent years. Fuzzy models are mostly designed using the Fuzzy Inference System (FIS) technique called Takagi–Sugeno–Kang (TSK) type-1 FIS. FIS is a collection of numerical methods based on membership functions, fuzzy rules, and fuzzy thinking, divining the relationship between input and output data [12-13]. Moreover, there is Mamdani-type FIS, in which if-then rules are used with linguistic antecedents and consequences [14]. However, TSK type-1 FIS is more suitable than Mamdani-type fuzzy modeling because it allows local predictions in system control and can be used with clustering algorithms [15]. TSK type-1 FIS rules include linguistic antecedents, but the result is obtained as a linear function of the input variables. Therefore, the result of each rule has design dimensions as a linear function of our design definitions. It has been shown that models using TSK type-1 FIS rules can accurately describe complicated behavior with only a few rules, thus significantly reducing the complexity of the system [16].

Subtractive Fuzzy Clustering (SFC) is a fuzzy logic technique in which fuzzy rule assignment is performed from cluster centers. Each cluster center obtained by clustering numerical data groups is a system feature in this method. In this way, a linear equation system depending on the input variables is constituted from the cluster centers, which correspond to the fuzzy rules. With the use of SFC, the processing intensity is significantly reduced [17].

In this study, subtractive clustering-based fuzzy models were established using experimental data of ultrasonic polymer degradation. Then, the accuracy of each model was tested by performing error analyses, and the results were compared with the experimental results from the literature to show their validity. As a result of the comparison, the success of the models is at a satisfactory level.

2. SPECIFICATION OF CLUSTER CENTERS BY EMPLOYING THE SFC METHOD

Clustering is a technique for finding similar groups in large data. It aims to define the system's behavior simply by classifying large data collections. To determine the cluster numbers and centers, a simple and effective algorithm called the Mountain Method (MM) has been introduced [18]. The proposed method is based on generating a mesh network in the data space, determining the grid points, and achieving the potential values of these grid points according to their distances from the real data points.

This study used Subtractive Fuzzy Clustering (SFC), an improved version of the Mountain Method. In the SFC method, data points are specified as potential cluster centers instead of grid points, and the number of data points is worked up to equal the number of grid points. In this way, calculations based on grid points are unnecessary, and the processing density is remarkably reduced [19].

SFC is a technique that fulfills fuzzy rule assignments by automatically specifying groups of similar data. This method acquires the most appropriate cluster centers representing the data sets by considering the distance between the data groups. Utilizing SFC, the cluster centers are determined according to the potential of the data points in the dataset. As a result of the potential computation around the data points, the points with the highest potential are selected as cluster centers. SFC algorithm is started by evaluating the distances of each data point to other data points and calculating the potential value resulting from this process. The potential value is usually computed with an equation similar to Gaussian function as follows:

$$P_i = \sum_{j=1}^{N} e^{(-\frac{4}{r_\alpha^2}\|x_i - x_j\|^2)}, i = 1, 2, \ldots, N \tag{1}$$

In this procedure, $\|.\|$ represents the Euclidean distance, while r_α is a positive constant denoting the neighborhood radius. This process selects the data point with the highest potential as the first cluster center. The data point with the maximum P_1^* the potential value is designated as the initial cluster center x_1^*. To eliminate the impact of the first cluster center when identifying subsequent cluster centers, the potential formula for the data points is adjusted as follows:

$$P_i' = P_i - P_k^* e^{(-\frac{4}{\eta r_\alpha^2}\|x_i - x_k^*\|^2)}, i = 1, 2, \ldots, N \tag{2}$$

The squash factor, represented by η, expresses the separation between cluster centers and ensures that they are not close to each other. This procedure continues until a sufficient number of cluster centers are attained, as stated by the specified criterion [19].

3. DEVELOPMENT OF FUZZY MODEL BASED ON SFC

In fuzzy logic systems, fuzzy rules that include linear functions are used to connect the antecedent and consequent parts. These rules link input and output data through IF-THEN statements. In this research, a type-1 Takagi-Sugeno-Kang (TSK) Fuzzy Inference System (FIS) was employed to model ultrasonic degradation data. Models based on TSK-type rules have proven effective in accurately capturing complex behaviors with a minimal number of rules, thereby greatly simplifying the system's complexity [20]. The structure of the FIS is depicted in Figure 1.

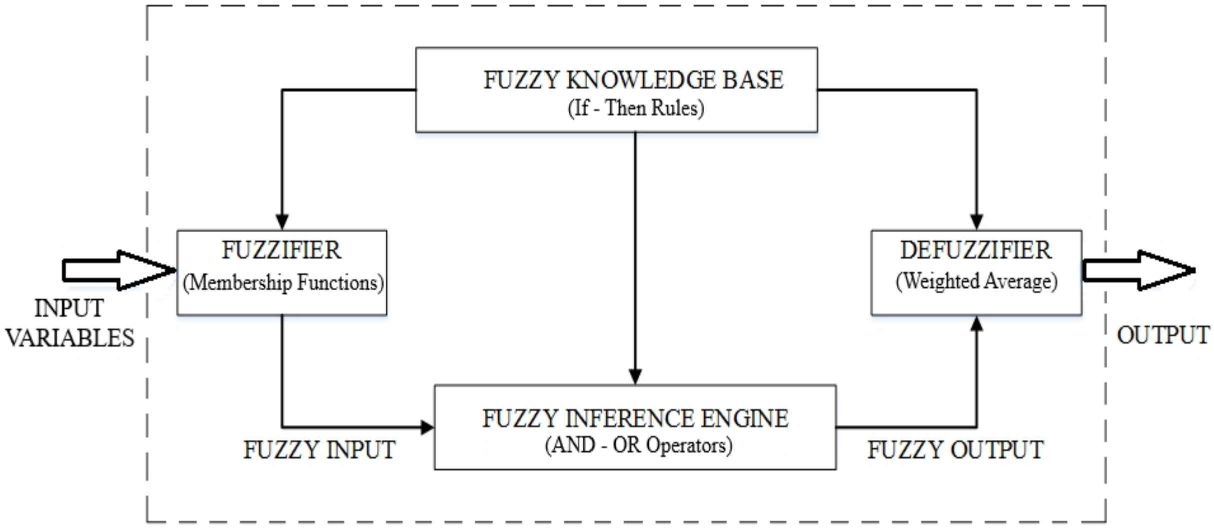

Figure 1. Fuzzy inference system architecture

As can be seen from the block diagram above, FIS consists of six main components:

Input Variables: Raw inputs are received into the system.

Fuzzification: Input variables are transformed into fuzzy sets using appropriate membership functions.

Fuzzy Knowledge Base: This section consists of fuzzy rules in IF – THEN.

Fuzzy Inference Engine: This part generates fuzzy results by evaluating the rules. The rules' weights are determined with AND (min) or OR (max) operators.

Defuzzifier: Since the results in Sugeno-type systems are usually described as linear functions, a crisp output is achieved by directly computing the weighted average, unlike classical fuzzy systems.

Output: In Sugeno type FIS the output is attained as a crisp value.

In such systems, the outputs of the rules are no longer represented by fuzzy sets but are instead defined as linear functions. As a result, the relationships between inputs and outputs, as well as the output functions, can be described using the following equations:

$$\begin{aligned} &IF\ x_1\ is\ A_1^k\ \&\ x_2\ is\ A_2^k\ \&\ldots\&\ x_j\ is\ A_j^k\ THEN \\ &\quad y_1\ is\ B_1^k\ \&\ y_2\ is\ B_2^k\ \&\ldots\&\ y_j\ is\ B_j^k \end{aligned} \tag{3}$$

$$B_j^k = c_0 + c_1 x_1^k + c_2 x_2^k + \ldots \tag{4}$$

Here, the jth input variable is denoted as x_j, and the jth output variable is represented as y_j, with k indicating the total number of rules. The linear equation system, referred to as B_j^k, is formulated based on the input variables. By implementing the SFC algorithm, a specific number of cluster centers ($x_{(1)*}, x_{(2)*}, ..., x_{(k)*}$) are determined. The input fuzzy set A_i^k is defined using a Gaussian membership function for the *k*th rule, as depicted in the following form:

$$\mu_{A_i^k}(x) = e^{-\frac{4}{r_\alpha^2}\left\|x_i - x_{(k)*}\right\|^2}, i = 1, 2, ..., N \tag{5}$$

To identify the most appropriate FIS configuration, it is essential to calculate the Gaussian function's center and standard deviation values. The output of the FIS is obtained by computing a weighted average of the outputs from each rule, similar to the centroid defuzzification method, as illustrated below:

$$y_j = \frac{\sum_{i=1}^{c} \mu_i y_{ij}}{\sum_{i=1}^{c} \mu_i} \tag{6}$$

Here, c represents the number of rules, j denotes the index of the data pair, and y_{ij} corresponds to the linear equation. In the final stage, the coefficients of the linear equation are determined by implementing the least squares approximation to the SFC method. The neighborhood radius r_α, the squash factor η, and the parameters for deciding new cluster centers (ε_{up} and ε_{down}) significantly impact the number of rules and the error values in the designed fuzzy system. The SFC algorithm concludes by identifying the optimal parameter values for the fuzzy system and computing the coefficients of the linear equation [19]. As a result of integrating the SFC algorithm with the least squares method, both the equation coefficients that capture the problem's characteristics are achieved, and a fast, high-accuracy fuzzy model is built.

4. FUZZY MODEL DESIGNS FOR ULTRASONIC POLYMER DEGRADATION

In this study, first, a single input—single output fuzzy model design illustrated in Figure 2 was made using experimental data on ultrasonic polymer degradation from a reference [21]. 3439 experimental data pairs giving the time-dependent viscosity change were evaluated to establish a model.

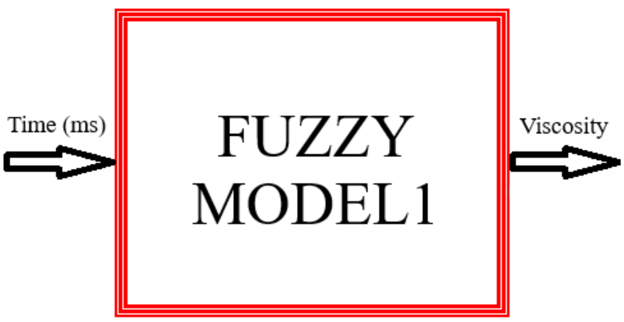

Figure 2. Fuzzy model for ultrasonic polymer degradation data [21] (One input, one output)

Out of the total 3439 experimental data pairs, 2000 were utilized for training the fuzzy model, while 1439 were employed to confirm the model's accuracy. Root mean square error (RMSE) was used to assess the errors associated with the training and control data pairs. To optimize the system, the main parameters r_α and η along with the cluster center criteria ε_{up} and ε_{down}, were carefully analyzed. In order to determine the optimum error value in this model, the error change depending on the r_α value of both training and control data was examined at fixed values of η, ε_{up} and ε_{down} parameters as depicted in Figure 3. The r_α value that gives the lowest common error value of training and control data was determined as the most suitable parameter value.

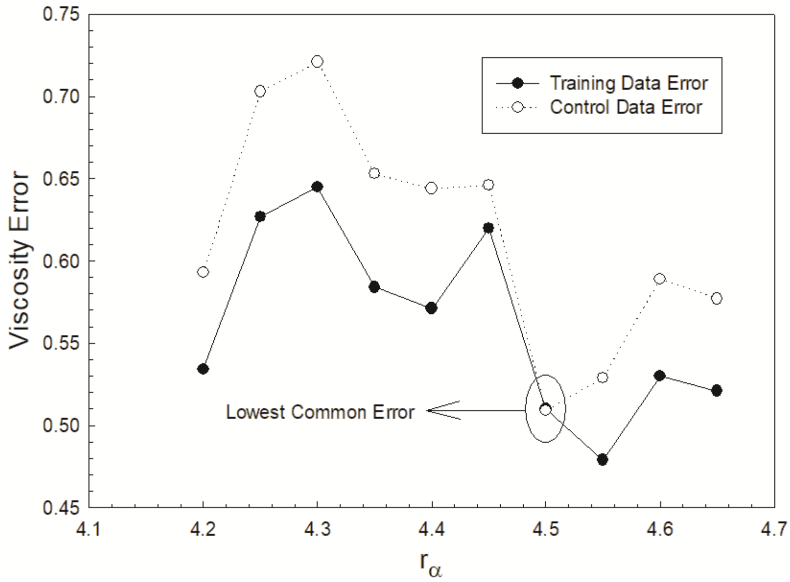

Figure 3. Distribution of viscosity error for training and control data of ultrasonic polymer degradation model depending on r_α while $\eta = 0.25$, $\varepsilon_{up} = 0.45$ and $\varepsilon_{down} = 0.2$

RMSE errors for both the training and control data obtained using the optimal input criteria for the ultrasonic polymer degradation model are presented in Table 1. Additionally, this table displays the neighborhood radius, squash factor, and input criteria values associated with the optimal result, along with the corresponding number of rules. The time-dependent distribution of the model output and the real experimental results at the r_α radius value corresponding to the optimum value are illustrated in Figure 4. It is seen that the fuzzy model results shown with symbols coincide with the experimental data shown with lines.

Table 1. Rms error values together with optimum input criteria and number of rules

RSML Model Error Values && Optimum Input Criteria		
RMS Error Values	Train Data	Control Data
	0.510	0.509

Optimal Parameter Values and Number of Rules	r_α	η	ε_{up}	ε_{down}	Number of Rules
	4.5	0.25	0.45	0.2	20

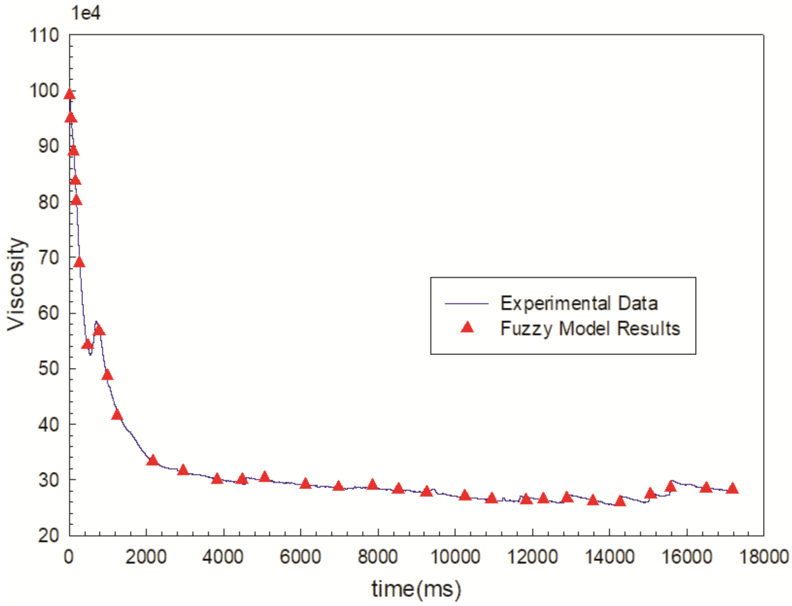

Figure 4. Comparison of polymer degradation model results with experimental data [21]

In the next stage, a fuzzy model design shown in Figure 5 was made in the form of three inputs - one output, evaluating the experimental data of ultrasonic polymer degradation taken from [22]. In the second fuzzy model, time, temperature and concentration were taken as inputs and specific viscosity as output. A total of 84 data pairs were utilized in model setup.

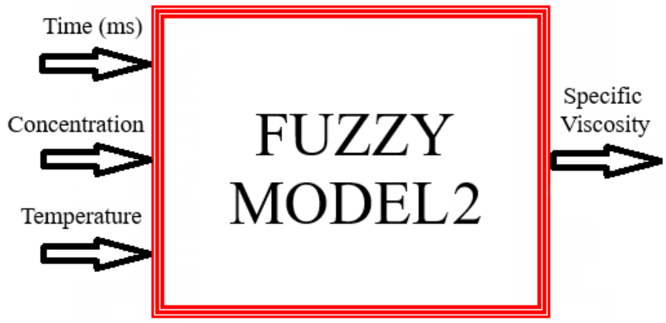

Figure 5. Fuzzy model for ultrasonic polymer degradation [22] (Three inputs, one output)

Out of the data pairs collected for model setup, 50 were utilized as training data, while 34 were employed to evaluate the model's validity. To identify the optimal parameter values for the second model, the specific viscosity error was computed in terms of RMSE by varying the neighborhood radius r_α, while keeping the parameters η, ε_{up} and ε_{down} fixed at specific values as depicted in Figure 6. Once again, the parameter values corresponding to the lowest common error of the training and control data were considered the most valid.

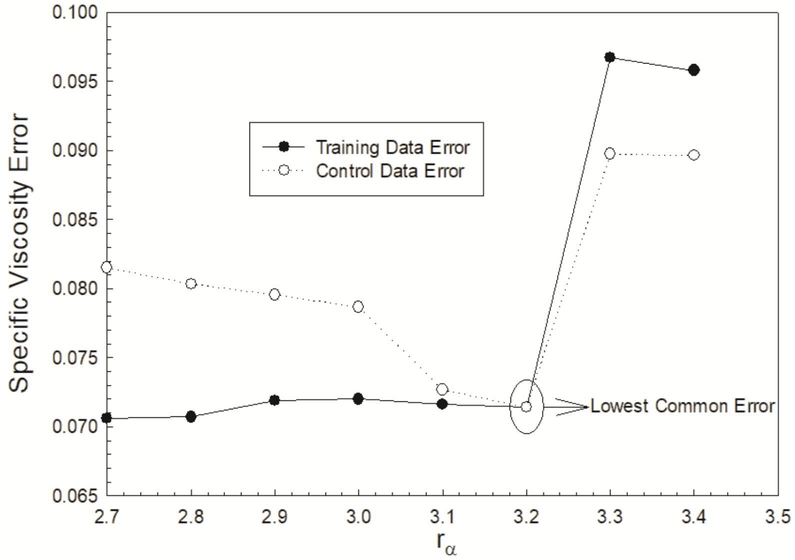

Figure 6. Distribution of viscosity error for training and control data of ultrasonic polymer degradation model depending on r_α while $\eta = 1$, $\varepsilon_{up} = 0.5$ and $\varepsilon_{down} = 0.25$

Table 2 displays the RMSE errors for both the training and control data, achieved utilizing the optimal input criteria for the ultrasonic polymer degradation model. Once again, this table includes the most appropriate parameter values together with the corresponding them number of fuzzy rules.

Table 2. RMS error values together with optimum input criteria and number of rules

RSML Model Error Values && Optimum Input Criteria					
RMS Error Values	Train Data			Control Data	
	0.0714			0.0715	
Optimal Parameter Values and Number of Rules	r_α	η	ε_{up}	ε_{down}	Number of Rules
	3.2	1	0.5	0.25	3

Figure 7 illustrates the changes in both model output and experimental data over time for different temperature values . As can be clearly seen from the graph, although very few rules were used in our second model, the model results were acquired at values very close to the experimental data.

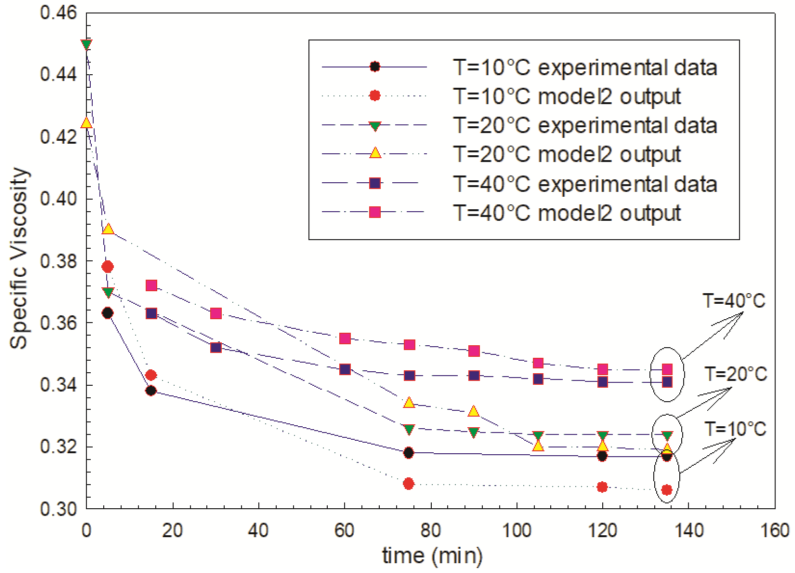

Figure 7. Comparison of polymer degradation model results with experimental data [22]

5. CONCLUSION

This study applied a SFC-based fuzzy modeling approach to analyze experimental data from two different ultrasonic polymer degradation samples available in the literature. The developed fuzzy models obtained linear equations describing polymer degradation with fast, reliable, and satisfactory error values. Unlike classical methods commonly used in the literature, the proposed SFC-based fuzzy modeling approach offers significant advantages, such as eliminating the need to determine the number of clusters before clustering and reducing system complexity. With these features, the study addresses the uncertainty and complexity challenges encountered in ultrasonic polymer degradation analysis, filling a gap in the literature and providing an original contribution.

The primary contribution of the proposed method to the literature is to enhance the effectiveness of fuzzy logic-based approaches in the analysis of experimental data and to offer a faster and more practical alternative compared to classical methods. Additionally, it has been demonstrated that SFC-based fuzzy modeling can better capture nonlinear relationships in the modeling of complex physical processes, such as ultrasonic degradation, and facilitates the interpretation of experimental data. In this context, the study highlights the potential of fuzzy logic-based modeling methods in ultrasonic polymer degradation analysis, offering a new perspective for researchers in this field.

For future studies, it would be beneficial to comprehensively examine the performance of the proposed method under different polymer types, ultrasonic frequencies, and various experimental conditions. Furthermore, to improve the accuracy and generalization capability of the fuzzy modeling method, the development of hybrid approaches (e.g., integrating fuzzy logic with artificial neural networks or machine learning algorithms) is recommended. To evaluate the method's applicability in real-time applications, testing it on larger datasets and different sensor data is also crucial. Additionally, using advanced optimization techniques, such as genetic algorithms, particle swarm optimization, or differential evolution, for optimizing fuzzy modeling parameters could further enhance the method's performance. Finally, investigating the applicability of the proposed method in different engineering applications (e.g., material fatigue analysis, ultrasonic non-destructive testing methods, and quality control processes of polymeric materials) would strengthen its position in the literature and expand its application areas. Studies conducted in line with these suggestions are expected to contribute significantly to the theoretical and practical development of fuzzy logic-based modeling methods.

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