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E.A. LYSENKOV, O.V. KOZLOV

Petro Mohyla Black Sea National University
(10, 68 Desantnykyv Str., Mykolaiv 54003, Ukraine; e-mail: ealysenkov@ukr.net)

STUDY OF MELTING TEMPERATURE BEHAVIOR OF POLYMER NANOCOMPOSITES USING FUZZY LOGIC-BASED APPROACH OF ARTIFICIAL INTELLIGENCE

This work presents a fuzzy logic-based artificial intelligence approach for predicting the melting temperature of polymer nanocomposites based on polylactic acid and carbon nanotubes (CNTs). A Mamdani-type fuzzy inference model was developed using the degree of crystallinity, carbon nanotube concentration, and nanotube diameter as input parameters. The constructed model reproduced nonlinear relationships between the structural characteristics and the thermal behavior of the nanocomposites and demonstrated good agreement with experimental calorimetric data. The resulting response surfaces revealed the existence of an optimal CNT concentration range associated with the maximum nucleating effect of the nanotubes. The predictive capability of the model was confirmed by an adjusted coefficient of determination $R^2 = 0.86$, indicating the applicability of fuzzy logic methods for intelligent modeling of polymer nanocomposite systems.

Keywords: polylactic acid, polymer nanocomposites, carbon nanotubes, melting temperature, artificial intelligence, fuzzy logic models, property prediction.

1. Introduction

Polymer nanocomposites filled with carbon nanostructures have attracted significant scientific and technological interest due to their enhanced mechanical, electrical, and thermal properties [1, 2]. Among various nanofillers, carbon nanotubes (CNTs) are considered particularly promising because of their high aspect ratio, exceptional mechanical strength, and high thermal conductivity. The incorporation of CNTs into polymer matrices enables the formation of multifunctional materials for applications in electronics, energy storage systems, sensors, biomedical engineering, and additive manufacturing [3]. At the same time, increasing attention is being devoted to biodegradable polymer matrices, especially polylactic acid (PLA), owing to its renewable origin, biocompatibility, biodegradability, and favorable processing characteristics [4]. PLA-based nanocomposites reinforced with CNTs represent a promising class of materials with improved functional performance compared to neat PLA [5].

One of the key scientific and practical challenges in the development of PLA/CNT nanocomposites is the prediction and control of their physicochemical and thermophysical properties [6]. The ability to design materials with predetermined characteristics is critically important for modern materials science because it significantly reduces the number of costly experimental iterations and accelerates the development cycle of advanced functional materials. In this context, special attention is devoted to thermal characteristics, particularly the melting temperature of the nanocomposite system. The melting temperature is an essential parameter governing processing conditions, thermal resistance, dimensional stability, crystallization behavior, and the operational reliability of polymer products [7]. In technologies such as extrusion, injection molding, and additive manufacturing, accurate prediction of the melting behavior is necessary for optimizing processing conditions and preventing thermal degradation of the polymer matrix [8].

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The melting temperature of PLA/CNT nanocomposites depends on multiple interconnected factors, including the degree of crystallinity of the polymer matrix, filler concentration, and geometric characteristics of carbon nanotubes [9, 10]. CNTs can simultaneously promote crystallization by acting as nucleating agents and restrict polymer chain mobility due to interfacial interactions [11]. As a result, the overall thermal behavior of such systems becomes highly nonlinear and difficult to describe using conventional analytical models. Despite numerous experimental studies, there is currently no universal model capable of simultaneously accounting for the combined influence of the degree of crystallinity, CNT concentration, and nanotube diameter on the melting temperature of PLA-based nanocomposites. In recent years, artificial intelligence (AI) methods and data-driven approaches have become increasingly important tools in polymer science and materials engineering [12, 13]. Machine learning algorithms are actively employed for the prediction of mechanical, thermal, rheological, and electrical properties of polymer composites, optimization of material compositions, identification of hidden dependencies in multidimensional datasets, and acceleration of materials discovery [14, 15]. AI-based models are especially attractive for systems characterized by nonlinear interactions among numerous variables, where classical analytical methods become insufficient. By learning from experimental data, such models can establish functional relationships between processing parameters, structural characteristics, and resulting material properties [16].

However, conventional machine learning methods typically require large experimental datasets, which are often unavailable in polymer nanocomposite research. Fuzzy logic modelling has demonstrated significant potential in solving materials science problems involving poorly formalized dependencies and partially uncertain experimental data [17]. In polymer composite systems, fuzzy inference methods enable the integration of heterogeneous parameters into a unified predictive framework and allow estimation of material properties even when exact analytical relationships are unknown. This creates favorable conditions for constructing predictive models capable of describing the thermal behavior of nanocomposite systems with high flexibility and sufficient accuracy [18].

Therefore, the present work is focused on the development of a fuzzy logic model for predicting the melting temperature of PLA/CNT nanocomposites as a function of the matrix degree of crystallinity, carbon nanotube concentration, and nanotube diameter. Such modeling is expected to contribute to the optimization of nanocomposite formulations and to improve the understanding of the complex interactions governing the thermal behavior of polymer systems reinforced with carbon nanotubes.

2. Theoretical Approach for Predicting of Melting Temperature

2.1. A fuzzy logic-based AI approach for modeling the properties of polymer nanocomposites

A major challenge in the modeling and prediction of polymer nanocomposite properties is the limited availability of experimental data, which reduces the effectiveness of conventional machine learning and deep neural network methods under small-data conditions [12]. Therefore, considerable attention is focused on fuzzy logic methods capable of operating under conditions of uncertainty and incomplete experimental information [17]. Fuzzy models can preserve satisfactory predictive capability even for relatively small datasets [19]. In addition, fuzzy inference systems possess high interpretability because their decision-making structure can be represented through understandable linguistic IF–THEN rules, while also demonstrating robustness to noisy experimental measurements [20, 21].

In this study, an intelligent model for predicting the melting temperature of polymer nanocomposites was developed based on the Mamdani-type fuzzy inference mechanism [22]. The input and output parameter spaces were partitioned into corresponding linguistic terms (LT) with predefined membership functions (MF), while the fuzzy rule base (RB) was constructed using experimentally observed dependencies and known physicochemical regularities governing nanocomposite behavior. Due to the transparent structure of the fuzzy inference system, the developed model preserves high interpretability while reproducing complex nonlinear relationships between the parameters affecting the melting temperature of the polymer nanocomposite.

2.2. Construction of the core logic of the fuzzy model

In the proposed fuzzy logic model, the melting temperature of a polymer nanocomposite was considered as a function of three key parameters: the degree of crystallinity of the polymer matrix (χ), the concentration of carbon nanotubes (C), and their diameter (D). These parameters determine the supramolecular organization of the polymer and the interfacial interactions in the "matrix–nanofiller" system, directly affecting the thermodynamic stability and melting temperature of the composite. To ensure the universality of the model under different conditions, all input and output variables were represented in normalized relative units within the interval from 0 to 1.

For the construction of the fuzzy inference model, each input parameter was partitioned into several characteristic subdomains described by corresponding linguistic terms. The degree of crystallinity of the polymer matrix was divided into three subdomains corresponding to low, intermediate, and high structural ordering levels. Such partitioning is sufficient for reproducing the general influence of crystallinity on the thermal behavior of the nanocomposite while preserving the interpretability of the fuzzy rule base. In contrast, the concentration of carbon nanotubes required a more detailed partitioning because of its strongly nonlinear influence on the thermophysical properties of the material. The nanotube diameter was described using a moderate number of fuzzy subdomains sufficient for distinguishing between different geometric scales of nanofillers and their influence on interfacial interactions within the polymer matrix. The output parameter corresponding to the melting temperature was also partitioned into several subdomains, enabling the fuzzy inference system to reproduce gradual nonlinear transitions in the thermophysical response of the polymer nanocomposite.

The fuzzy rule base was constructed according to known physicochemical regularities governing polymer nanocomposites filled with carbon nanotubes and based on trends identified during the analysis of experimental data for the PLA/CNT system [6,9,11,23–29]. The model assumed that increasing crystallinity of the polymer matrix leads to an increase in melting temperature due to the higher ordering of the macromolecular structure and the greater energy required for crystalline phase destruction. The influence of nanotube concentration was defined as nonlinear: low and moderate concentrations improve crystallization and interfacial interactions, resulting in increased melting temperature, whereas excessively high concentrations promote nanotube aggregation and structural heterogeneity, reducing the reinforcing effect. In addition, smaller nanotube diameters were associated with higher melting temperatures because of the increased specific interfacial surface area and stronger interaction between the nanofiller and the polymer matrix.

3. Results and Discussion

3.1. AI-based fuzzy model for melting temperature prediction of polymer nanocomposites

A Mamdani-type fuzzy model is characterized by a rule base in which both the antecedent and consequent parts are represented by fuzzy propositions defined over the corresponding input and output variables [19]. Each fuzzy rule generally has the following form:

$$\text{if "}x_1 \text{ is } A_1^i\text{" and ... and "}x_n \text{ is } A_n^i\text{" then "}y \text{ is } B_i\text{"}, \quad (1)$$

where A_1^i and B_i denote fuzzy sets associated with the corresponding linguistic terms of the input and output parameters. The computational procedure of the Mamdani inference mechanism consists of five distinct stages: fuzzification, aggregation, rule activation, accumulation, and defuzzification. During fuzzification, crisp input variables are transformed into degrees of membership using predefined membership functions. In the aggregation stage, the fuzzy antecedent sets are prepared and combined according to the structure of the RB, forming the initial fuzzy representation of input-driven conditions. In the rule activation stage, the firing strength of each rule is computed by evaluating the antecedent part using a minimum operator as a standard t-norm for conjunction. In the accumulation stage, the contributions of all activated rules are merged into a single integrated fuzzy output set, typically using the maximum operator to ensure a coherent representation of the system response. Finally, defuzzification is performed to transform the resulting fuzzy set into a crisp numerical value, most commonly using the centroid (center of gravity) method [22].

Owing to its transparent rule-based structure and capability to reproduce complex nonlinear dependen-

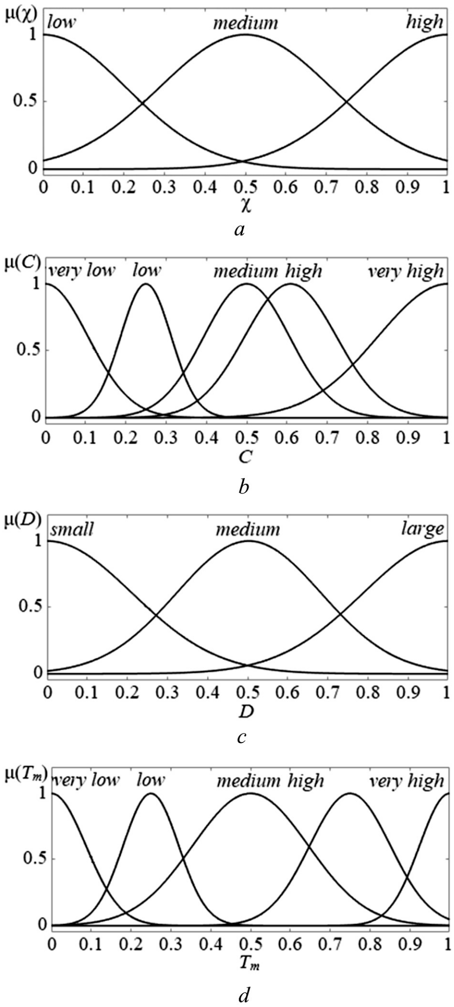


Fig. 1. Appearance of configured linguistic terms of input and output variables for the fuzzy model

cies under conditions of uncertainty, the Mamdani fuzzy model is particularly suitable for intelligent modeling tasks involving limited and partially uncertain experimental datasets.

When developing the model based on the core logic outlined in subsection 2.2, the following linguistic terms were selected for each input and output variable: 3 LTs (low, medium, high) for the degree of crystallinity of the polymer matrix χ; 5 LTs (very low (v. low), low, medium, high, very high (v. high)) for the concentration of carbon nanotubes C; 3 LTs (small, medium, large) for the diameter of nanotubes D; 5 LTs (very low (v. low), low, medium, high, very high (v. high)) for the melting temperature T_m.

For all linguistic terms of all (input and output) variables 1st type Gaussian membership functions were chosen. In turn, this function $\mu(\chi)$ for the first input variable χ is described by expression (2),

$$\mu(\chi) = \exp\left[-\frac{(x-b)^2}{2a^2}\right], \tag{2}$$

where a, b are adjustable MF parameters.

The appearance of the input and output variables' LTs with adjusted parameters of their MFs are presented in Fig. 1.

Using the presented LTs (Fig. 1) and based on the above physical considerations, a complete fuzzy rule base of the Mamdani-type model was formed, describing the interaction between the degree of crystallinity, concentration and diameter of nanotubes. This approach allowed us to account for the complex nonlinear nature of the influence of structural parameters on the melting temperature of a polymer nanocomposite and to provide a physically justified description of the system even with a limited amount of experimental data.

For convenience, the synthesized rule base is presented in a shortened matrix form in Table 1. This RB consists of 45 rules, each of which is given in the form of expression (1). In turn, the internal cells of Table 1 contain the LTs of the output variable T_m, which act as the corresponding consequents.

Table 1. **Rule base of the fuzzy model**

χ	C				
	v. low	low	medium	high	v. high
			D is small		
Low	V. low	Medium	Medium	Medium	Low
Medium	V. Low	Medium	High	High	Medium
High	V. low	High	V. high	V. high	High
			D is medium		
Low	V. low	Low	Medium	Medium	Low
Medium	V. low	Medium	Medium	High	Medium
High	V. low	Medium	High	High	Medium
			D is large		
Low	V. low	V. low	Low	Low	V. low
Medium	V. low	Low	Medium	Medium	Low
High	V. low	Medium	Medium	High	Medium

For example, the first rule in full form in accordance with Table 1 is given by the following expression:

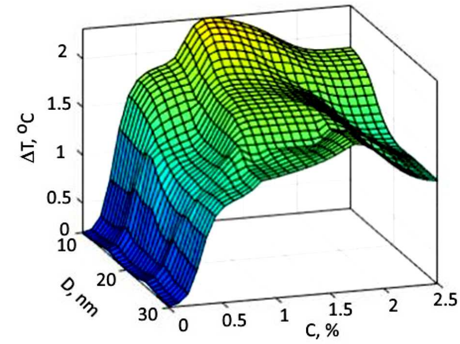
If “χ is low” and “C is very low” and

“D is small” then “T_m is very low” (3)

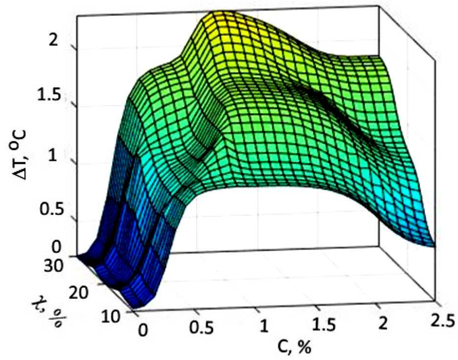
3.2. Theoretical modeling results and their physical interpretation

The obtained response surfaces (Fig. 2), constructed using a Mamdani-type fuzzy rule system for PLA/CNT composites, demonstrate the complex nonlinear nature of the relationship between the degree of crystallinity of the polymer matrix, the concentration and diameter of carbon nanotubes, and the melting temperature of the composite. Analysis of the resulting surfaces indicates that the proposed fuzzy model adequately reproduces the main physical regularities characteristic of PLA-based polymer nanocomposites obtained by the melt-blending method [23–29].

The surface $\Delta T = f(\chi, D)$ is characterized by a predominantly monotonic variation in melting temperature (Fig. 2, *a*). With increasing crystallinity, a gradual increase in T_m is observed, which is consistent with classical concepts of the thermodynamic stability of the crystalline phase of the polymer. An increase in the proportion of ordered regions in the PLA structure leads to the formation of more perfect lamellar structures, the destruction of which requires more thermal energy. Within the constructed surface, this effect is manifested in the form of an upward gradient of the melting temperature along the crystallinity axis. At the same time, a clear influence of the nanotube diameter is observed. For systems with a smaller CNT diameter, the melting temperature is higher than for composites with larger nanotubes. This behavior is explained by an increase in the specific surface area of the nanofiller with a decrease in the CNT diameter, which leads to an increase in the interfacial interaction between the polymer matrix and nanotubes [30]. As a result, the effectiveness of restricting the segmental mobility of PLA macrochains increases and the crystalline structure of the polymer stabilizes. At the same time, the nature of the surface indicates that the influence of diameter is less pronounced than the influence of the degree of crystallinity, which is in good agreement with experimental data for PLA/CNT systems, in which



a



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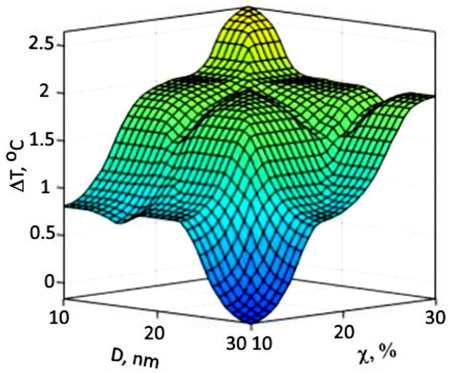

c

Fig. 2. Characteristic dependence surfaces $\Delta T = f(\chi, C, D)$ for polymer nanocomposites

changes in melting temperature usually do not exceed 0.5–2 °C [23, 26, 29].

The surface $\Delta T = f(C, D)$ demonstrates the most complex and physically significant variation in melting temperature (Fig. 2, *b*). In the central region of the surface, a local maximum is observed, which corresponds to small diameters of nanotubes and moderate concentrations of CNTs. Such a surface shape in-

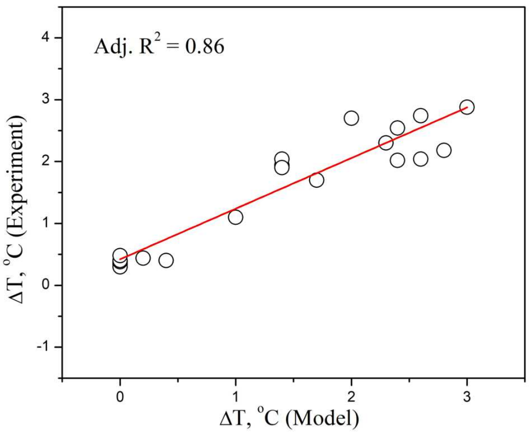

Fig. 3. Comparison of predicted and experimental T_m values obtained using the fuzzy model

dicates the existence of an optimal region of nanofiller content, in which the maximum nucleating effect of CNTs is realized. The introduction of a small amount of nanotubes into the PLA matrix promotes the formation of additional crystallization centers, which accelerates the ordering of macromolecular chains and leads to a partial increase in the melting temperature [24, 27, 29]. The most pronounced effect is observed for concentrations of about 1–1.5%, which is fully consistent with experimental DSC data [26]. However, a further increase in the CNT concentration leads to a gradual decrease in the melting temperature, which manifests itself in the form of a decrease in the surface at high values of C. This behavior may be associated with the aggregation of nanotubes and the deterioration of their dispersion in the polymer matrix. At high CNT concentrations, the probability of the formation of local agglomerates increases; these agglomerates act as structural defects and prevent the formation of a regular crystalline morphology of PLA [24, 28]. In addition, this may be accompanied by local limitation of the diffusion mobility of macrochains during crystallization, which reduces the efficiency of nucleation [29]. An increase in the diameter of nanotubes also leads to a decrease in the melting temperature, since with a decrease in the specific surface area of CNTs, the interfacial interaction with the polymer weakens and their ability to act as crystallization centers decreases.

The surface $\Delta T = f(\chi, C)$ also has a pronounced nonlinear character (Fig. 2, *c*). With an increase in the degree of crystallinity, the melting point naturally increases, which confirms the decisive role of the supramolecular organization of PLA in the formation of the thermal properties of the composite [25]. At the same time, the dependence on the CNT concentration is nonmonotonic. In the range of low and moderate concentrations, an increase in the melting point is observed, while a further increase in the CNT content is accompanied by a gradual decrease in T_m. Such a surface shape indicates the competition of two physical mechanisms. On the one hand, nanotubes act as effective centers of heterogeneous crystallization, contributing to the formation of a more ordered PLA structure [27, 29]. On the other hand, an excess of CNTs can lead to the formation of an inhomogeneous morphology of the composite, the emergence of local packing defects of macromolecules and a partial disruption of the regularity of the crystalline structure. Therefore, the maximum melting point is realized only in a certain range of nanotube concentrations.

In general, the resulting response surfaces confirm that the melting point of PLA/CNT composites is determined by a complex interaction between the degree of crystallinity of the polymer matrix, the geometric characteristics of the nanotubes and their concentration. The Mamdani fuzzy model allowed the experimental trends not only to be generalized but also to reflect the physically justified nonlinear nature of the influence of structural parameters on the thermal properties of nanocomposites. It is especially important that the model adequately reproduces the existence of an optimal CNT concentration, at which the maximum nucleation effect is realized without significant development of aggregation processes.

3.3. Comparison between model predictions and experimental data

The developed rule base and the selected parameters of the linguistic membership functions provided sufficiently high predictive accuracy of the AI-based model. The predictive performance of the Mamdani fuzzy inference system used to predict the melting temperature variation in PLA/CNT nanocomposites is presented in Fig. 3. Fig. 3 demonstrates a clear positive correlation between the model-predicted and experimentally determined values of ΔT, obtained from [6, 9, 11, 23–29]. Most experimental points are located close to the regression line, indicating good agreement between the fuzzy model and the experimental DSC data over the investigated parameter range. The obtained adjusted coefficient of determination (Adj. $R^2 = 0.86$) confirms that the model adequately reproduces the main regularities governing the thermal behavior of the PLA/CNT systems.

At the same time, a certain scatter of the experimental points relative to the regression line is observed, especially in the region of higher ΔT values. Such deviations are expected for polymer nanocomposite systems and may be associated with several physical factors that are difficult to describe explicitly within a simplified fuzzy inference framework. In particular, the melting behavior of PLA/CNT composites is strongly affected by local heterogeneity of nanotube dispersion, partial CNT agglomeration, variations in crystal morphology, and differences in interfacial interactions between the polymer matrix and the nanofiller [31]. In addition, the crystallization behavior of PLA is highly sensitive to thermal prehistory and processing conditions, which can also contribute to experimental variability [6, 32].

Nevertheless, the overall trend reproduced by the model indicates that the fuzzy logic approach successfully captures the nonlinear relationship between crystallinity, CNT concentration, nanotube diameter, and the resulting melting temperature variation. The obtained predictive accuracy demonstrates that the developed Mamdani fuzzy model can be considered an effective tool for describing complex structure–property relationships in PLA/CNT nanocomposites, particularly under conditions where conventional analytical models become difficult to apply due to the multivariate and nonlinear nature of the system.

4. Conclusions

A fuzzy logic-based artificial intelligence approach for predicting the melting temperature of PLA/CNT polymer nanocomposites was developed in this work. The proposed Mamdani fuzzy inference model was constructed using experimentally established physicochemical relationships between the degree of crystallinity of the polymer matrix, CNT concentration, nanotube diameter, and the resulting thermal behavior of the nanocomposite system.

The developed fuzzy model demonstrated the ability to adequately reproduce complex nonlinear dependencies characteristic of PLA/CNT composites. The obtained response surfaces revealed that the melting temperature is governed by the combined influence of the supramolecular organization of PLA and the structural characteristics of carbon nanotubes. An increase in the crystallinity degree led to a systematic increase in melting temperature due to the formation of more thermodynamically stable crystalline regions. At the same time, the influence of CNT concentration exhibited a pronounced nonlinear behavior associated with the competition between the nucleating effect of nanotubes and aggregation-induced structural heterogeneity at elevated filler contents.

The modeling results also showed that smaller nanotube diameters contribute to a partial increase in melting temperature owing to the larger specific interfacial area and enhanced interfacial interaction between the polymer matrix and the nanofiller. The fuzzy model successfully reproduced the existence of an optimal CNT concentration range corresponding to the maximum nucleating efficiency of nanotubes without significant aggregation effects.

Comparison between the model predictions and experimental DSC data demonstrated good agreement, with an adjusted coefficient of determination $R^2 = 0.86$. The obtained predictive accuracy confirms that the developed fuzzy inference system is capable of capturing the main regularities governing the thermal behavior of PLA/CNT nanocomposites even under conditions of limited experimental datasets and partially uncertain information.

The proposed AI-based fuzzy modeling approach may therefore be considered a promising tool for intelligent prediction and optimization of the thermophysical properties of polymer nanocomposites. The developed methodology can potentially be extended to other multifunctional polymer systems reinforced with nanoscale fillers and may contribute to accelerating materials design in modern polymer engineering and nanocomposite science.

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Е.А. Лисенков, О.В. Козлов

ДОСЛІДЖЕННЯ ТЕМПЕРАТУРИ ПЛАВЛЕННЯ ПОЛІМЕРНИХ НАНОКОМПОЗИТІВ З ВИКОРИСТАННЯМ НЕЧІТКО-ЛОГІЧНОГО ПІДХОДУ ШТУЧНОГО ІНТЕЛЕКТУ

У роботі запропоновано нечітко-логічний підхід із застосуванням штучного інтелекту для прогнозування температури плавлення полімерних нанокомпозитів на основі полімолочної кислоти й вуглецевих нанотрубок (ВНТ). Розроблено модель нечіткого висновку типу Мамдані, в якій ступінь кристалічності, концентрацію та діаметр ВНТ використано як входові параметри. Побудована модель дала змогу відтворити нелінійний характер взаємозв'язку між структурними параметрами й термічними властивостями нанокомпозитів і продемонструвала хорошу узгодженість з експериментальними калориметричними даними. Отримані поверхні відгуку виявили існування оптимального діапазону концентрацій ВНТ, в якому реалізується їхній максимальний нуклеаційний ефект. Прогностична здатність моделі підтверджується значенням скоригованого коефіцієнта детермінації $R^2 = 0{,}86$, що свідчить про перспективність застосування нечіткої логіки для моделювання полімерних нанокомпозитних систем.

Ключові слова: полімолочна кислота, полімерні нанокомпозити, вуглецеві нанотрубки, температура плавлення, штучний інтелект, моделі нечіткої логіки, прогнозування властивостей.