Fuzzy logical-linguistic model for assessing the qualitative composition of carbon nanomaterials

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Abstract. The paper considers the development of methodological approaches to assessing the qualitative composition of carbon nanomaterials (nanotubes) and predicting their behavior in processes based on the theory of fuzzy sets. The state of the nanotechnology industry is analyzed, and the problematic moments of its development are highlighted. The necessity of proposing new approaches to evaluate nanomaterials due to the specificity of their properties is substantiated. The trends in the industrial use of carbon-based nanomaterials are identified, and the importance of their rational application based on an objective description of the qualitative composition of carbon nanomaterials for the development of the nanotechnology industry is shown. In this regard, it is proposed to use the mathematical apparatus of the theory of fuzzy sets, linguistic variables, and corresponding mathematical methods considering expert opinions and experimental results. A mathematical model created allows to predict the behavior of the nanotubes in various processes and solve the problem of the latters’ optimization.

1. Introduction

The pace of formation of innovative economy and the active implementation of advanced scientific achievements in the manufacturing sector determine the level of the country’s economic security and the quality of life of its citizens. The formation of the sixth technological structure necessitates structural changes in the national economy based on the use of research results in nanotechnology.

The intensive development of the scientific component in Russia’s nanotechnology industry and the increasing volumes and ranges of nanostructured materials update the task of their large-scale use in the product manufacturing for final consumption. The solution for this task involves the acceleration of the transition from the laboratory searching of technologies for taking advantage of the unique properties of nanoscale objects to their universal applications in the key sectors of economy. In the development of nanoindustry, the following problematic points can be highlighted:

- insufficient theoretical basis for designing processes at the nanolevel, both in the production of materials and their use in industry; as a result, the predominantly empirical way of overcoming this issue is through getting new knowledge;
- it is difficult to obtain data on the properties of single nanoparticles and describing the cumulative properties of the entire system.
The formation of nanoindustry and the active use of nanomaterials to improve the quality of products require, among other things, developing methods and measurement tools, and searching for adequate options to assess the qualitative composition of such products.

Employing advanced diagnostic methods [1], standardization and certification of nanomaterials produced on an industrial scale will make it possible to control the quality of products thereof manufactured, and ensure national and international markets entry for nanoproducts [2,3]. The need for developing new approaches to the evaluation of nanomaterials is due to their special properties, considerable variation in the parameters of single components, and, accordingly, to the inability to completely apply the regulations of chemicals to these materials.

The significant gap between the quality of scientific research, scientific and technical groundwork and insufficient infrastructure of the country’s nanoindustry and the incompliance with the requirements on the availability and quality of work on standardization, metrology and assessment of the nanomaterials conformity pose a rather serious barrier to the innovative renewal of the country. It is necessary to bring the systemic classification of nanotechnology and products thereof based to a new level, and to develop mechanisms for assessing the quality of the nanomaterials obtained.

At present, the analysis of the state and development trends of nanotechnology objects makes it possible to conclude that synthesizing carbon nanomaterials (CNMs) – fullerene-like structures that represent a new allotrope of carbon in the form of closed, frame, macromolecular systems – is one of the most promising areas of nanotechnology [3-6]. Among these materials, carbon nanotubes (CNTs) occupy a special place, which, with a diameter of 1-50 nm and a length of several microns, form a novel class of quasi-one-dimensional nanoobjects. CNTs possess a number of unique properties due to the ordered structure of their nanofragments: good electrical conductivity and adsorption characteristics, the ability for cold electron emission and gas storage, diamagnetic characteristics, chemical and thermal stability, and high strength combined with high values of elastic deformation.

Materials developed on the basis of CNTs can be successfully used as structural modifiers of construction materials, hydrogen accumulators, radioelectronic elements, additives to lubricants, varnishes and paints, highly efficient adsorbents, gas distribution layers of fuel cells. The use of carbon nanostructures in fine chemical synthesis, biology and medicine is being widely discussed.

Being synthesized in reactors on a commercial scale, CNTs represent a multi-component mixture containing single-, two- and multi-walled formations of various lengths and diameters in a fairly wide range. For instance, the diameter of single-walled carbon CNTs ranges from 0.3 to 5 nm, whereas the diameter of two-walled CNTs varies from 1.8 to 7.1 nm. It is possible to make any CNTs type dominant by creating various technological synthesis modes in the reactors, but the synthesis procedure rarely yields highly homogeneous structures. They often represent mixtures of various nanocarbon formations possessing obviously different properties.

Developing multi-walled CNTs (Taunit series) with a predominantly conical shape of graphene layers, having an outer diameter of 10-60 nm, an inner diameter of 10-20 nm, and a length of more than 2 microns, is one of the promising areas of commercial CNTs production. These CNTs have shown a significant effect when used in various sectors of the economy (e.g., [3]). At the same time, the resulting product is quite heterogeneous, some of the parameters of its constituents – multi-walled CNTs – are inferior to those of single-walled ones; therefore, a certain problem lies in describing the nanoproduct properties by means of an integral characteristic.

Considering the aforementioned, the actual task is to assess the qualitative composition of CNTs based on the known characteristics: length, diameter, and number of layers. To construct the corresponding model, it is required to combine expert opinions presented in a natural language and experimental results with specific values of those characteristics.

Implementing the mathematical apparatus of the theory of fuzzy sets is one of the well-known and widely recommended approaches to the construction of the above-mentioned model [7,8]. For the expert opinion formalization within the framework of the chosen theory, linguistic variables are employed [7-9]. Taking into account the need for analyzing a significant amount of textual information, appropriate mathematical methods are actively used [10-15].
2. Materials and methods

Let us build a model for assessing the qualitative composition of carbon nanomaterials and predicting the result of their participation in further reactions using the theory of fuzzy sets. For this, several linguistic variables, described below, should be introduced.

Let \( L = \langle l, T_l, R, Gl, Ml \rangle \) be a linguistic variable that formalizes the nanotube length, where:
- \( l = \text{"length of a tube"} \) is the name of the linguistic variable \( L \);
- \( T_l = \{ \text{"short"}, \text{"medium"}, \text{"long" (tube)} \} \) is the term-set of \( L \) values;
- \( Gl \) is the syntactic rule generating new \( L \) values;
- \( Ml \) is the rule specifying the semantics for new, \( Gl \)-generated, statements.

Let us consider the use of the syntactic rule. If we take the arbitrary element \( svl \in Sv \), and two arbitrary values \( t_1 \) and \( t_2 \in T_l \), then the syntactic rule for constructing a new value will be as follows: \( t_1 \text{ svl } t_2 \). That is, for instance, let \( t_1 = \text{"short"}, t_2 = \text{"medium"} \) and \( svl = \text{"or"} \); then \( t_1 \text{ svl } t_2 = \text{"short or medium (tube)"} \). Now, we consider the arbitrary element \( modl \in Modl \). The semantic rule for the arbitrary term \( r \in T_l \) will be as follows: \( modl \ r \). For instance, when \( t = \text{"short"} \) and \( modl = \text{"not"} \), the semantic rule builds \( \text{"not short (tube)"} \).

To construct statements, their consistent application to each other is admissible. For instance, the connective \( \text{"or"} \) with respect to \( t_2 \) is applied to the expression obtained in the previous example. Then, we will get the following statement: "not short (n)or medium (tube)".

\( Ml \) is the semantic rule that assigns a membership function to each value of a fuzzy variable [10]. The membership function formalizes the meaning of this value; that is, for each \( Y \in Gl(Tl) \), it defines: \( \mu_Y : R \rightarrow [0;1] \).

Let us construct the membership functions \( \mu_L \) for \( Y = \text{"short"} \) and \( Y = \text{"long"} \). For this, it is required to check 4 sizes with the expert: \( a_1, b_1, a_2, b_2 \), where \( a_1 \) (nm) is the size of a very short or even extremely short tube; \( b_1 \) (nm) is the minimum size of a tube, which under no circumstances can be called short; \( a_2 \) (nm) is the size of a very long or even extremely long tube; \( b_2 \) (nm) is the maximum size of a tube, which under no circumstances can be called long.

Let \( FL = \{ f : R \rightarrow R \mid f(x) \downarrow \text{on} \ R, f(a_1) = 1, f(b_1) = 0 \} \), and \( FR = \{ f : R \rightarrow R \mid f(x) \uparrow \text{on} \ R, f(a_2) = 1, f(b_2) = 0 \} \). Then, the corresponding membership functions can be built with respect to the element \( FL \) or \( FR \):

\[
\mu_{sh}(x) = \min \{ 1, \max \{ 0, f(x) \} \} \forall x \in R, \text{ where } f \in FL, \\
\mu_{la}(x) = \min \{ 1, \max \{ 0, f(x) \} \} \forall x \in R, \text{ where } f \in FR.
\]

The choice of a specific function from a given wide class will be carried out at the final stage of model building. At the same time, it is easy to see that the membership function (1) will be equal to 1 for all values \( x < a_1 \), to 0 for all \( x > b_1 \), and continuously decrease between them. Similarly, the membership function (2) will be 0 for all values \( x < b_2 \), and 1 for all \( x > a_2 \), and continuously increase between them.

It is advisable to choose Archimedean \( T,S \)-norms and an operation of fuzzy negation as a semantic formalization of the syntactic connectives "and" and "or" and the modifier "not" [7]. On the one hand, such a choice is classical for the theory of fuzzy sets, and on the other, it provides a wide range of \( T,S \)-norms, thereby allowing to select a specific function type at the final stage, which will increase the adequacy of the model.

Thus, let \( t_1 \) and \( t_2 \) be some values of the linguistic variable with the membership functions \( \mu_{t_1} \) and \( \mu_{t_2} \), respectively. Then, \( \mu_{t_1 \text{ or } t_2} = S(\mu_{t_1}, \mu_{t_2}), \mu_{t_1 \text{ and } t_2} = T(\mu_{t_1}, \mu_{t_2}), \) and \( \mu_{\text{not}} = n(\mu_L) \), where \( T(.,.) \) and \( S(.,.) \) are the Archimedean \( T,S \)-norms, respectively, and \( n(.) \) is the negation expressed as follows:

\[
n(\mu(x)) = 1 - \mu(x), \forall x \in R.
\]
It should be noted that number squaring is classically used to create the semantic rule for the modifier "very". However, in our opinion, it is more expedient to take a more generalized form of this modifier. Thus, the functions $\text{pow}(\cdot)$ and $\text{pow}2(\cdot)$ will be chosen herein as a semantic rule formalizing the syntactic modifier "very" and its superlative form "extremely":

$$\text{pow}(\mu(x)) = (\mu(x))^\alpha, \forall x \in R;$$

$$\text{pow}(\mu(x)) = (\mu(x))^\beta, \forall x \in R,$$

where $\alpha$ and $\beta \in \{r \in R : r > 1\}$.

Like in the case of the $FL$ and $FR$ function types, specific values of $\alpha$ and $\beta$ will be determined at the final stage of model creation based on experimental data.

It should be noted that for complete definition of the linguistic variable $L$, setting the membership function to formalize the term "medium" is left. The tube length is referred to as medium if the tube is not short and not long, which, according to the accepted semantic rules, means:

$$\mu_{\text{medium}}(x) = \begin{cases} n(\mu_{\text{short}}(x)), & n(\mu_{\text{short}}(x)) \leq R \\ n(\mu_{\text{long}}(x)), & \text{otherwise} \end{cases} \forall x \in R. \tag{6}$$

Let $D = <d, Td, R, Gd, Md>$ be the linguistic variable for representing the tube diameter in the mathematical apparatus of fuzzy sets, where

$d = \text{"diameter of a tube"}$ is the name of the linguistic variable $D$;

$Td = \{\text{"small"}, \text{"medium"}, \text{"large"}\}$ is the term-set of $D$ values;

$Gd$ is the syntactic rule generating new $D$ values and consisting of the syntactic connectives $Svd = \{\text{"and"}, \text{"or"}\}$ and the modifiers $Modd = \{\text{"extremely"}, \text{"very"}, \text{"not"}\}$;

$Md$ is the rule specifying the sense for $Gd$-generated statements. This rule is identical to $Ml$ in the case of the linguistic variable $L$, and is given by formulas (1)-(6), with the only difference that formula (1) is related to the function describing the meaning "small", whereas formula (2) – to "large".

The main difference between the linguistic variable defining the number of tube layers (walls) is that the determination domain for the membership functions, specifying the semantic rules, lies in $R$, and due to the apparent discreteness of the number of layers, the determination domain for the corresponding functions of the considered variable will be $N$.

At the same time, it should be noted that for simplicity of treatment using processor devices, we will use continuous approximation of the membership functions formalizing the semantic rules. Let $\mu$ be a certain membership function that formalizes the arbitrary semantic rule of the linguistic variable describing the number of layers. Then, to set $\mu, \mu : N \rightarrow [0; 1]$, we will use the approximation of $\mu_{st}, \mu : N \rightarrow [0; 1]$ being such that $\forall i \in N \mu(i) = \mu_{st}(i)$. Due to the fact that the transformations made using formulas (1-6) for the continuous functions are identical to the corresponding transformations made for their values at each point of the determination domain, the approximation proposed herein will not affect the result of calculations. In other words, instead of saving a table of membership function values in integer arguments, we save a function $\mu_{st}$ with real arguments, which has the same values in integer numbers.

Let $S = <s, Ts, R, Gs, Ms>$ be a linguistic variable to formalize the number of layers in the language of the mathematical apparatus of the theory of fuzzy sets, where:

$s = \text{"number of tube layers"}$ is the name of the linguistic variable $S$;

$Ts = \{\text{"few"}, \text{"medium"}, \text{"many"}\}$ is the term-set of $S$ values;

$R$ is the determination domain for the semantic rules.

$Gs$ is the syntactic rule generating new $S$ values and consisting of the syntactic connectives $Svd = \{\text{"and"}, \text{"or"}\}$ and the modifiers $Modd = \{\text{"extremely"}, \text{"very"}, \text{"not"}\}$;

$Ms$ is rule specifying the sense for $Gs$-generated statements. This rule is also given by formulas (1)-(6), with the only difference that formula (1) is related to the function describing the meaning “few”, whereas formula (2) – to “many”.

It is obvious that the constants, $a_1, b_1, a_2, b_2$ will be unique for each of the linguistic variables considered, since the very nature of the characteristics described by these variables is different. Then, for convenient sharing of these constants, we will denote them regarding each linguistic variable $L, D$.
and \( S \) as \( a_1^b, b_1^c, a_1^d, b_1^e, a_2^f, b_2^g, a_2^h, b_2^i, a_3^j, b_3^k \), respectively. On the other hand, the semantic rules that formalize the connectives and the modifiers, on the contrary, should be unified to create formulas, in which several different linguistic variables are present simultaneously.

Considering the expert opinions, we will make up a list of rules in a natural language using the linguistic variables values and the syntactic connectives between them from the set \( S_v = \{ \text{"and", "or"} \} \).

We will develop a model simulating the behavior of a mixture of CNTs in subsequent chemical reactions based on the theory of fuzzy sets.

3. Results and discussion

Let us develop a fuzzy logical-linguistic model to assess the behavior of a CNTs mixture in subsequent chemical reactions based on the theory of fuzzy sets. The base of fuzzy production rules is one of the key components of this model [10].

The rules created by the experts will be of two types:
1) “if \( V \), then the reaction rate is acceptable”, designated as \( Y(V) \),
2) “if \( V \), then the reaction rate is not sufficient”, designated as \( X(V) \),
where \( V \) represents an expression composed of the values of the linguistic variables \( L, D \) and \( S \) connected with the modifier \( S_v \).

For instance, let \( V = \text{"the tube length is short and the tube diameter is small or medium, and there are many tube layers (number of layers)"} \). Then, \( Y(V) = \text{"if the tube length is short and the tube diameter is small or medium, then the reaction rate is acceptable"} \), and \( X(V) = \text{"if the tube length is short and the tube diameter is small or medium, and there are many tube layers (number of layers), then the reaction rate is not sufficient"} \). It should be noted that due to setting the semantic rules for each variable and formalizing the connectives "and" and "or" between the variables, we can put the expression \( V \) in accordance with the following membership function:

\[
\mu_V(x, y, z) = T\left[\mu_{\text{short}}(x), T\left[\mu_{\text{small}}(y), T\left[\mu_{\text{many}}(z), T\left[\mu_{\text{short}}(y), \mu_{\text{many}}(z)\right]\right]\right]\right],
\]

where \( x, y, z \) are the length and the diameter of tubes, and the number of tube layers, respectively. The resulting membership function is multidimensional [5], since its determination domain is \( R^3 \).

In the same way, one can compose a membership function specifying the semantics of the arbitrary expression \( V \) created by the elements of term sets of linguistic variables, syntactic connectives and modifiers.

Let us suppose that according to the results of the expert survey, \( n \) rules of the first type, \( Y(V_1), Y(V_2), \ldots, Y(V_n) \), and \( m \) rules of the second type, \( X(V_{n+1}), X(V_{n+2}), \ldots, X(V_{n+m}) \), were obtained, and the semantics of the expressions \( V_1, V_2, \ldots, V_{n+m} \) is formalized by the membership functions \( \mu_{V_1}(x, y, z), \mu_{V_2}(x, y, z), \ldots, \mu_{V_{n+m}}(x, y, z) \), respectively.

Then, by virtue of the syntax and the semantics of the rules, the latter can be represented as two generalized rules:
1) “if \( V_1 \) or \( V_2 \) or \( \ldots \) or \( V_n \), then the reaction rate is acceptable”;
2) “if \( V_{n+1} \) or \( V_{n+2} \) or \( \ldots \) or \( V_{n+m} \), then the reaction rate is not sufficient”.

The membership function (\( \mu_V \)), evaluating the degree of truth of the expression “\( V_1 \) or \( V_2 \) or \( \ldots \) or \( V_n \)” is as follows:

\[
\mu_V(x, y, z) = S(\mu_{V_1}(x, y, z), \mu_{V_2}(x, y, z), \ldots, \mu_{V_n}(x, y, z)),
\]

By analogy, the degree of truth of the expression “\( V_{n+1} \) or \( V_{n+2} \) or \( \ldots \) or \( V_{n+m} \)” has the following form:

\[
\mu_V(x, y, z) = S(\mu_{V_{n+1}}(x, y, z), \mu_{V_{n+2}}(x, y, z), \ldots, \mu_{V_{n+m}}(x, y, z)).
\]
the properties of associativity and commutativity [7], it is possible to generalize the $S$-norm as a function of several variables:

$$S_{\lambda,m}(x) = \begin{cases} S(x_1, x_2), & m = 2, \\ S(x_m, S(x_1, x_2, \ldots, x_{m-1})), & m > 2. \end{cases}$$

(9)

It is worth mentioning that due to the semantics of generalized rules, the solution to the problem of predicting the reaction is carried out by calculating the values of expressions (7) and (8). Thus, in the ideal case, if $\mu_r(x,y,z) = 1$, and $\mu_s(x,y,z) = 0$, then the reaction rate is acceptable. The reaction rate is not sufficient if $\mu_r(x,y,z) = 0$, and $\mu_s(x,y,z) = 1$.

In practice, the values of $\gamma$ and $\gamma_0$ will lie in the range from 0 to 1, and, according to one of the main classes of fuzzy implication [11], the decision on the acceptability of the reaction rate should be made on the basis of the following rule:

$$S_{\lambda,m}(x) = \begin{cases} \mu_r(x,y,z) > \mu_s(x,y,z), \text{reaction rate acceptable}, \\ \mu_r(x,y,z) \leq \mu_s(x,y,z), \text{reaction rate not sufficient}. \end{cases}$$

(10)

It remains to decide on some parameters, namely:
- select functions from $FL$ and $FR$ to build a semantic rule using formulas (1) and (2);
- determine the values of the real numbers $\alpha$ and $\beta$ to formalize the modifiers “very” and “extremely”;
- choose the $T$ and $S$ norms.

The last choice merits a separate discussion. The fact is that there exist whole families of $T,S$-norms [13] given by a formula with the real parameter $\lambda$. Each $\lambda$ value determined in the half-interval $[0, \infty)$ specifies its own pair of $T,S$-norms. For instance, let us consider the Hamacher $T,S$-norms [13] denoted as $T_h$ and $S_h$, respectively:

$$T_h(x,y) = \begin{cases} \frac{xy}{\lambda + (1 - \lambda)(x + y - xy)}, & \lambda \in [0, \infty), (\lambda, x, y) \neq (0,0,0), \\ 0, & (\lambda, x, y) = (0,0,0), \\ \lambda = \infty; \end{cases}$$

(11)

$$S_h(x,y) = \begin{cases} 1, & (\lambda, x, y) = (0,1,1), \\ \lambda = \infty; \end{cases}$$

(12)

where

$$T_d(x,y) = \begin{cases} 0, & (x,y) \in [0,1] \times [0,1], \\ \min(x,y), & x = 1 \text{ or } y = 1, \\ \lambda = \infty; \end{cases}$$

$$S_d(x,y) = \begin{cases} 1, & (x,y) \in [0,1] \times [0,1], \\ \max(x,y), & x = 1 \text{ or } y = 1. \end{cases}$$

The choice of the specific type of the parametric Archimedean $T$, $S$-norms (e.g., expressions (11) and 12)) and the value of the parameter $\lambda$ should be carried out to provide the best implementation of the prediction model (expressions (7)-(10)). To formalize such a choice, we will consider $A$ — the set of the Archimedean $T,S$-norms.

Let the result of a particular reaction rate experiment be given by the tuple $<(x, y, z), k>$, where $x$, $y$, $z$ is the length and the diameter of tubes, and the number of tube layers, respectively, and $k$ is the result of the experiment: $k \in \{0,1\}$, if $k = 0$, then the reaction rate was insufficient for the given value of the characteristics $(x, y, z)$; on the other hand, $k = 1$ means that the reaction proceeded at an acceptable
rate. Then, the error (E) of the prediction model for the particular experiment will be calculated as follows:

\[
E((x, y, z), k) = \left| k - \mu_{r}(x, y, z) + (1-k) - \mu_{s}(x, y, z) \right| / 2
\]

Let us suppose that \( q \) experiments were performed, then their results are represented by the set of the tuples \( K = \{(x_1, y_1, z_1), k_1\}, \{(x_2, y_2, z_2), k_2\}, \ldots, \{(x_q, y_q, z_q), k_q\}\). We want to find the parameters for the proposed model so that they minimize errors in all the experiments. In this case, it is convenient to minimize the maximum error. Let \( R:K \rightarrow [0,1] \) be the model error function, then

\[
R(K) = \max_{i=1}^{q} E((x_i, y_i, z_i), k_i)
\]

Since we have listed the model parameters that can be changed, and also have written down the total error, given by the model, in expression (14), it is advisable to set the following optimization problem for choosing the parameters:

\[
R(K) \rightarrow \min,
\]

\[
f \in FL, g \in FR, \alpha, \beta \in R, \alpha, \beta > 1, (T, S) \in A, \lambda \in [0, \infty)
\]

The general form of the optimization problem (expression (15)) is unlikely to have an analytical or iterative solution. Indeed, the number of functions in \( FL \) and \( FR \) is infinite, as well as the values of \( \alpha, \beta, \lambda \). In practice, when solving the optimization problem (expression (15)), it is worth considering functions of a particular type only (e.g., polynomial ones).

4. Conclusion
A fuzzy logical-linguistic model was developed for assessing the quality of carbon materials, taking into account the variation of the key parameters (diameter, length, and number of layers), and conceptual approaches were proposed for predicting the behavior of CNTs when conducting a reaction based on the theory of fuzzy sets. The proposed model combines two approaches: it aggregates expert opinions through the formalization of rules created using linguistic variables and is configured by selecting a number of parameters considering the results of experiments.

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