Model of Paired and Solitary Influence of Ingredients of Polymer Composition



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Abstract The problem of calculating the properties of a polymer-modified with a complex of active additives is considered. The proposed approach to solving the problem involves the use of already available information about polymer compositions to create a black-box model and further refine the interaction of composition components based on the laws of the subject area and a gradual transition to the gray box. This work is devoted to creating a black-box model based on two-component interaction and so far does not take into account the complex component. The proposed model also allows you to evaluate the unknown pair influence of the ingredients of the polymer composition from the known single effect of these ingredients. The resulting model has been tested on a specific example. Computations using fuzzy mathematics has been held. Based on the results obtained, a solution to the task is suggested.

Keywords Polymer composition · Active additive · Property · Fuzzy numbers · Decision making

1 Introduction

Composite materials (composites) are multicomponent systems, today it is one of the most filled scientific and technical niches. However, the number of developments related to the creation of new materials in this category is constantly growing, since it is here where it is expected to obtain significant results for technology and economics. Managing the properties and overall quality of composites during their design and development is a multicriteria problem solved by experimental and model methods. Moreover, in this category, special mixtures are distinguished by composite mixtures

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created on the basis of high molecular weight compounds—polymers, which can be either organic or inorganic in nature [3, 21].

It will not be superfluous to note that at present high-molecular compounds as individual substances are practically not processed today but are used to prepare technological mixtures, and the main tool for regulating the properties of the final material is, so to speak, a game with ingredients. Moreover, the use of active additives to solve technical problems for various materials is quite broad [13]. For the ingredient control of the properties of polymeric materials in the presence of a base high molecular weight matrix, various active and inactive fillers, modifying, and non-modifying additives are widely used. This is especially true for multi-ingredient (more than two) polymer compositions (PCs) and materials produced on their basis. At the same time, the goals are directed, efficient, economical regulation and optimization of both the composition of the system "polymer matrix (up to 100%) + filler (0–95%) + additives (0–45%)" and its processing technology, as well as a wide range of operational characteristics [1, 5, 18, 20].

In terms of theory, there is a need to solve the multifactor task of optimizing PCs, which is essentially fuzzy [4, 6, 8, 9, 11, 17], today it is based on the dependency platform "composition—property—quality—application" and provides reasonable (better quantitatively) selection of a certain amount and quality of ingredients with the necessary technical functions. Today's sets of ingredients are presented very widely today, so the result is ambiguous solutions that are unlikely to be unambiguous in the future.

Another theoretical prerequisite for this topic is that raw PCs (in other words, mixtures), abstracting from the specific content, must be considered as complex chemical-technological systems (CTS), in which the composition, physicochemical state of the system (PC and the interaction between the ingredients and the polymer matrix individually and in combination), depend not only on their nature of the elements of the system but also on the technological processes of their processing and are difficult to theoretically multifactor modeling and on this basis of identification and forecasting [10, 14]. Here, the research tool is system analysis methods and, in particular, modeling. And given the similarity of the structural models of materials of natural and artificial origin from the point of view of chemical composition and degree of ordering in the polymer structure [16], we can talk about the prospects for describing natural phenomena.

Turning specifically to the subject matter of this chapter, we have that the materials obtained on the basis of PCs also fall into this circle, being extremely complex heterogeneous systems that can be divided according to prevailing views according to processing technology and structural features [3, 21] about the following groups:

- fiber filled;
- dispersed filled;
- with interpenetrating structures of continuous phases;
- mixed;
- voluminous ("3D");
- layered ("2D").

Given the special modern scientific, technical, and consumer interest in nanotechnology, we separately distinguish PCs of the composition "polymer matrix—additive (filler)", which uses 2D and 3D nanoparticles—graphene, carbon nanotubes, and similar filler ingredients. They make it possible to obtain PCs with a number of unique properties [2]. Other technologies also make it possible to control fiber geometry [12] or nanoparticle reinforcement [15].

In this chapter, fillers are not considered in detail—their range is relatively limited, and there is an extensive and detailed set of theoretical and practical developments on that part [19]. Another thing is supplements. Today, their diversity, offered by the chemical industry and underdevelopment, is very large and constantly growing. Widely represented, for example, super concentrates (2–5% of the mass of the polymer matrix). However, at the same time, the number of problems associated with their properties and the properties of the PC as a system object is growing.

Enlarged additives for PCs located on the production market can be divided into two classes: technological (processing) and modifying (functional), associated with the operation of the polymer product. We note the main most common purposes of their use in a PC (the most relevant for modern conditions are indicated in bold):

- generation and satisfaction of new needs of society;
- technology optimization;
- giving specific effects;
- extension of durability (weather resistance, water resistance, frost resistance, crack resistance, fatigue, chemical resistance, etc.);
- increase fire resistance;
- expansion of the intervals of physical and mechanical characteristics;
- expansion of technical and ergonomic functions;
- regulation of shrinkage;
- improvement of surface properties;
- improvement of environmental friendliness;
- improvement of aesthetic properties;
- acceleration of drying and a set of standardized properties;
- saving resources.

The most important types of additives (without details) are presented in Table 1.

The formation of a complete and current formulation, taking into account the goals and properties listed above, has a peculiar feature that, for example, the virtual (pre-design) composition of a PC, like the real one, is also a multicomponent and multifactor complex, often intelligent, system with uncertain the number and quality of communications, which is incomplete and requires engineering support in the technology for the implementation of the full algorithm "PC design—practice" through further design and experimental work. This, in addition to the above, maybe the definition of additional conditions, the nature of co-ingredients, their concentration, technological and operational characteristics, etc.

We give a tabular example (Table 2) showing only a partial variety of PC ingredients (mainly of polymer nature in modern technologies).

N	ю	Type of additive	Content, mass % to the polymer matrix
1		Retardants	0–3.0
2		Antiseptics	0–2.0
3		Antistatics	0–3.0
4		Inhibitors	0–2.0
5		Dyes (pigments)	0–2.0
6		Hardeners	0–10.0
7		Plasticizers	0–45.0
7		Rheological	0–3.0
8		Greases	0–2.0
9		Stabilizers	0.1–5.0

Table 1	Types of additives in
PC	

Table 2	Additives	in poly	vmer	composite	s
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1 2	1	
Type of additive	Technical function	Active substance
Polyolefins	Ethylene propylene copolymer, ethylene propylene diene vinyl copolymer, ethylene–vinyl acrylate copolymer	Polyolefins
Polypropylene	Ethylene propylene divinyl copolymer, terephthalic ether, ethylene methyl acrylate, polybutadiene	Polypropylene
Polystyrene, styrene plastics	A copolymer of butadiene and styrene, a copolymer of ethylene, butadiene, and styrene	Polystyrene
Polyamides	Methyl disulfide, methyl acrylonitrile copolymer, ethylene, butadiene, and styrene copolymer, grafted copolymers methyl acrylonitrile	Polyamides
Polyesters	Acrylates, polyethylene glycidyl dimethacrylate	Polyesters
Polycarbonate	A copolymer of methyl methacrylate, butadiene, and styrene, ABC (PS/ABC mixture)	Polycarbonate
Polyphenylene sulfide	Polyethylene glycidyl dimethacrylate	Polyphenylene sulfide
Anti-blocking	Sticking prevention	Silicic acid, amide waxes
Antioxidant	Thermal oxidation prevention	Phenols, phosphides
Flame retardant	Flammability reduction	Inorganic oxides, hydroxides metals halogen-containing, phosphorus-containing, etc

In connection with the foregoing, we point out that the management of PC properties for the creation of competitive technologies is possible only in a combination of theoretical and empirical methods while taking into account the multicomponent nature of the PC and the variety of technologies for its processing, the following design work vectors are possible:

- pre-experimental heuristic identification of PCs as a system and selection of material properties;
- design of the polymer matrix and the technology for its production (polymerization, polycondensation, modification, crystallization, structuring, etc.).
- the choice of composition and structure by varying the ingredients;
- regulation of the technical functions and concentration of ingredients, taking into account possible environmental damage and the interests of society.

Naturally, such a cumbersome task and its solution must be considered in stages, starting, for example, with a two-component system, which is done in the present work.

In connection with the foregoing, it is clear that there is always a need to develop new accurate methods for predicting and constructing properties of PCs, which will allow us to create qualitatively new materials in the future. Therefore, the possibility of designing effective materials based on adequate models, which make it possible to predict with a high degree of accuracy the properties of newly created composites and not only polymer ones, is important.

2 Formulation of the Problem

Let polymer *P* be given. Based on this polymer, it is planned to create a composition *C* in order to provide the best possible values of some *m* properties $Q_1, Q_2, ..., Q_m$. The values of these polymer properties are denoted by $x_1, x_2, ..., x_m$, respectively. Here we will assume that the parameters x_j are normalized so that an increase in its value corresponds to an improvement in the corresponding property. Some clarification is required here, which is understood as "best properties".

When buying polymer products, the consumer pays attention to a number of properties. These properties should meet the expectations of the consumer to the maximum extent, i.e., one of the main requirements for the polymer composition is the user's needs. Another important source of requirements for the properties of the polymer composition is the technological process (for example, the features of the technological process can lead to polymer degradation, then stabilizers will be additionally needed, or softening of the composition may be required in the production process, then plasticizer is required, etc.). Other sources of polymer composition requirements may arise. In any case, a list of requirements for the property values (in our case, their number is indicated by m) of the polymer composition is obtained. Hereinafter, we will consider those values that are least different from property values according to the formulated requirements to be considered the best.

Let polymer composition *C* consist of a polymer matrix *P* and ingredients s_i , i = 1, ..., n, where *n* is the number of ingredients in *C*. It is required to evaluate how the properties of Q_j for *C* will change compared to P, j = 1, ..., m. To do this, we construct a mathematical model of the interaction of the components of the composition and obtain a procedure for calculating its properties.

3 Mathematical Model of the Interaction of Components

Denote by x_{ij} the value of the Q_j property of the polymer *P* after modification with the compound s_i , i = 1, ..., n. Suppose that additives s_i interact in pairs with each other, exerting a cumulative effect on *P* according to the following scheme (Fig. 1). So s_{i_1} and s_{i_2} ($i_1 \neq i_2 = 1, ..., n$) together give the property Q_j the value $x_{i_1i_2j}$, j = 1, ..., m. Then the effect of the additives s_{i_1} and s_{i_2} on *P* can be expressed by the formula

$$x_{i_1i_2j} = a_{i_1i_2j}x_j$$

where $i_1 \neq i_2 = 1, ..., n$ are the numbers of active additives, $a_{i_1i_2j}$ is the coefficient of influence of additives s_{i_1} and s_{i_2} on the Q_j property of the polymer composition, $x_{i_1i_2j}$ is a manifestation of the Q_j property polymer composition after modification. From that we have

$$\frac{x_{i_1i_2j}}{x_j} - 1 = a_{i_1i_2j} - 1 = q_{i_1i_2j},$$

where $q_{i_1i_2j}$ is the reduced coefficient of influence of the additives s_{i_1} and s_{i_2} on the property Q_j of the polymer composition, the feature of which is that when the property worsens, it takes negative values, and with improvement, it takes positive values.

The total effect of the entire complex of additives $s_1, s_2, ..., s_n$ will be expressed by the ratio





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$$q_j = \frac{x'_j}{x_j} - 1,$$
 (1)

where q_i is a certain reduced coefficient that describes the combined effect of all nadditives on the property $Q_j, j = 1, ..., m$, resulting in the value x'_j .

For the upper and lower bounds (1) in [8] it is suggested to take

$$q_j \le \sum_{i_1 < i_2 = 1}^n q_{i_1 i_2 j} = \overline{q}_j,$$
 (2)

$$q_j \ge \frac{1}{C_n^2} \sum_{i_1 < i_2 = 1}^n q_{i_1 i_2 j} = q_j,$$
(3)

It is believed that additives are selected so as not to impair the properties of the polymer composition, i.e.

$$0 \leq q_j \leq \overline{q}_j.$$

As a result, we obtain that the cumulative effect on the property Q_j is expressed by an odd number

$$\hat{q}_{j}(x_{j}) = \exp\left(-\frac{\left(x_{j} - \tilde{q}_{j}\right)^{2}}{\delta_{j}^{2}}\ln 2\right),\tag{4}$$

where $\tilde{q}_j = (\bar{q}_j + q_j)/2$, $\delta_j = (\bar{q}_j - q_j)/2$, j = 1, ..., m. The overall assessment \hat{q} of the impact of the whole complex of additives is expressed by the formula

$$\hat{q} = \sum_{j=1}^m \alpha_j \hat{q}_j,$$

where $\alpha_j \ge 0$ reflects the importance of the property Q_j in comparison with others and for which the relations

$$\sum_{j=1}^{m} \alpha_j = 1.$$

4 Computing Experiment

Consider two polymer compositions (Table 3) and evaluate them in terms of compliance with consumer needs.

For this, it is necessary to use the above model and determine how additives interact in pairs with each other and how they affect the corresponding property of the polymer composition (Tables 4 and 5).

Here j = 1, 2, 3 means the sustainability of the polymer composition to combustion, oxidation, photodestruction, respectively.

For composition C_1 , according to (2) and (3), we obtain

$$\overline{q}_1 = \sum_{i_1 < i_2 = 1}^3 \overline{q}_{i_1 i_2 1} = 0.4 + 0.05 + 0.02 = 0.47,$$
$$q_1 = \frac{1}{C_3^2} \sum_{i_1 < i_2 = 1}^3 q_{i_1 i_2 1} = \frac{1}{3} (0.2 + 0.02 + 0.01) \approx 0.077$$

where $\overline{q}_{i_1i_21} = \sup q_{i_1i_21}, q_{i_1i_21} = \inf q_{i_1i_21}$.

Carrying out similar calculations, we obtain the results given in Table 6.

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No	Designation	Polymer, %	Flame retardant (%)	Antioxidant (%)	Light stabilizer (%)
1	<i>C</i> ₁	86.8–92.9	6–10	1–2	0.4–0.6
2	<i>C</i> ₂	82.1–91.8	7–14	1–3	0.1–0.3

Table 3 Examples of polymer compositions' contents

j	Designation	Complex of two additives, $i_1 i_2$			
		12	13	23	
1	$q_{i_1i_21}$	0.2–0.4	0.02-0.05	0.01-0.02	
2	$q_{i_1i_22}$	0.01–0.02	0.3–0.5	0.4–0.7	
3	$q_{i_1 i_2 3}$	0.00-0.01	0.1-0.18	0.4–0.7	

Table 4 The effect of pairs of additives on the polymer composition C_1

Table 5 The effect of pairs of additives on the polymer composition C_2

J	Designation	Complex of two additives, $i_1 i_2$			
		12	13	23	
1	$q_{i_1i_21}$	0.20-0.35	0.03-0.04	0.02-0.05	
2	$q_{i_1i_22}$	0.02–0.06	0.2–0.44	0.2–0.4	
3	$q_{i_1i_23}$	0	0.1–0.3	0.4–0.7	

j	<i>C</i> ₁		<i>C</i> ₂	
	q_j	\overline{q}_{j}	q_j	\overline{q}_{j}
1	0.077	0.47	0.083	0.44
2	0.24	1.22	0.14	0.90
3	0.17	0.89	0.17	1.00

Table 6 The results of the calculation of the upper and lower estimates of changes in the properties of the compositions C_1 and C_2

According to (4), we obtain the following cumulative effect of the complex are additive on the polymer composition C_1

$$\hat{q}_1(x_1) = \exp\left(-\frac{(x_1 - 0.27)^2}{0.039}\ln^2\right), \\ \hat{q}_2(x_2) = \exp\left(-\frac{(x_2 - 0.73)^2}{0.24}\ln^2\right), \\ \hat{q}_3(x_3) = \exp\left(-\frac{(x_3 - 0.53)^2}{0.13}\ln^2\right)$$

and to the composition C_2

$$\hat{q}_1(x_1) = \exp\left(-\frac{(x_1 - 0.26)^2}{0.032}\ln^2\right), \\ \hat{q}_2(x_2) = \exp\left(-\frac{(x_2 - 0.52)^2}{0.14}\ln^2\right), \\ \hat{q}_3(x_3) = \exp\left(-\frac{(x_3 - 0.59)^2}{0.17}\ln^2\right).$$

Further, we assume that the most important thing for the customer is the incombustibility of the material and the high resistance to light exposure and put $\alpha_1 = 0.5$, $\alpha_2 = 0.2$, $\alpha_3 = 0.3$ and calculate \hat{q} using the formulas obtained in [7] for the polymer composition C_1

$$\hat{q} = \sum_{j=1}^{3} \alpha_j \hat{q}_j = \exp\left(-\frac{(x-0.44)^2}{1.1} \ln 2\right),$$

and to the composition C_2

$$\hat{q} = \sum_{j=1}^{3} \alpha_j \hat{q}_j = \exp\left(-\frac{(x-0.41)^2}{0.93} \ln^2\right).$$

5 The Model with Unknown Interaction of Some Components

Let's review the case when some additives are not known for how they interact with other components, and this introduces an additional factor of uncertainty. Suppose the result of the interaction of additives 2 and 3 is unknown.

We apply the approach described in [8] and estimate the value q_{23j} , j = 1, 2, 3. To do this, we need data on the effect of each of additives 2 and 3 separately on polymer compositions, which are given in Tables 7 and 8.

For composition C_1 , according to (2) and (3), we obtain

$$\overline{q}_1 = \sum_{i=2}^{3} \overline{q}_{i1} = 0.15 + 0.03 = 0.18,$$

 $q_1 = \frac{1}{2} \sum_{i=2}^{3} q_{i1} = \frac{1}{2} (0 + 0.01) = 0.005$

where $\overline{q}_{i1} = \sup q_{i1}, q_{i1} = \inf q_{i1}$

Carrying out similar calculations, we obtain the results given in Table 9.

According to (4), we obtain the following cumulative effect of the additive 2 and 3 on the polymer composition C_1

$$\hat{q}_{231}(x_1) = \exp\left(-\frac{(x_1 - 0.0925)^2}{0.0875}\ln^2\right), \\ \hat{q}_{232}(x_2) = \exp\left(-\frac{(x_2 - 0.825)^2}{0.475}\ln^2\right), \\ \hat{q}_{233}(x_3) = \exp\left(-\frac{(x_3 - 0.85)^2}{0.35}\ln^2\right)$$

Table 7	The effect of
additives	2 and 3 on the
polymer	composition C_1

J	Designation	Number of additives, <i>i</i>		
		2	3	
1	q_{i1}	0–0.15	0.01-0.03	
2	q_{i2}	0.5–0.8	0.2–0.5	
3	<i>q</i> _{<i>i</i>3}	0.4–0.5	0.6–0.7	

Table 8The effect ofadditives 2 and 3 on thepolymer composition C_2

J	Designation	Number of additives, <i>i</i>		
		2	3	
1	q_{i1}	0.03–0.06	0.01-0.04	
2	q _{i2}	0.2–0.4	0.3–0.4	
3	<i>q</i> _{<i>i</i>3}	0.6–0.8	0.3–0.5	

j	C_1		<i>C</i> ₂	
	q_j	\overline{q}_{j}	q_j	\overline{q}_{j}
1	0.005	0.18	0.02	0.10
2	0.35	1.30	0.25	0.80
3	0.50	1.20	0.45	1.30

Table 9 The results of the calculation of the upper and lower estimates of changes in the properties of the compositions C_1 and C_2

Table 10Real estimates ofthe effect of the complex ofadditives 2 and 3 on polymercompositions C_1 and C_2

J	Designation	Composition	
		<i>C</i> ₁	C_2
1	<i>q</i> ₂₃₁	0.005-0.17	0.02-0.1
2	<i>q</i> ₂₃₂	0.35–1.3	0.25–0.8
3	<i>q</i> ₂₃₃	0.5–1.2	0.45–1.3

and to the composition C_2

$$\hat{q}_{231}(x_1) = \exp\left(-\frac{(x_1 - 0.06)^2}{0.04}\ln^2\right), \\ \hat{q}_{232}(x_2) = \exp\left(-\frac{(x_2 - 0.525)^2}{0.275}\ln^2\right), \\ \hat{q}_{233}(x_3) = \exp\left(-\frac{(x_3 - 0.875)^2}{0.425}\ln^2\right).$$

Fuzzy estimates were obtained for unknown parameters q_{23j} , j = 1, 2, 3. In order to use the computational model proposed above, it is necessary to defuzzification these estimates. As real values of the parameters q_{23j} , j = 1, 2, 3, we take a range of values at which the value of the membership function does not exceed 0.5. The obtained values are presented in Table 10.

Now there is all the necessary information and, repeating the calculations given in paragraph 4, we obtain a similar result, adjusted for the approximate values of the complex effect of additives 2 and 3 on polymer compositions.

6 Conclusion

It should be noted that the results obtained are oriented only on a specific consumer. Nevertheless, the proposed model allows us to evaluate polymer compositions and make an informed choice. In the above example—this is composition C_1 .

The proposed approach to solving the problem involves using the already available information about polymer compositions to create a black-box model and further clarify the interaction of the composition components based on the laws of the subject

area and a gradual transition to the gray box. The approach used here allows us to identify the direction of research in the subject area to refine and refine the model.

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