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Computer-aided Design of Polymer-based Composites Using Multi-scale Models

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Abstract. The study is devoted to computer-aided design of particulate polytetrafluoroethylene-based composites for increasing their thermal conductivity values (as much as possible) with minimal deterioration of the mechanical properties. To solve the issue, an algorithm has been developed and applied using a multi-scale model. It has enabled to fabricate such composites, characterized by both high thermal conductivity and acceptable mechanical properties, via loading the polymer matrix with copper particles of two size ranges, differing by more than an order of magnitude.

Keywords: multi-scale model, polymer composite, particulate filler, stress-strain state, thermal conductivity, polytetrafluoroethylene (PTFE).

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Introduction

Polymer-based composites have become widespread because of the possibility to vary their functional characteristics over a wide range [1, 2]. As a rule, loading with fillers, including particulate ones, changes their both structure and properties but does not affect the chemical nature of the polymer matrix [3]. For tribological applications, numerous composites are based on polytetrafluoroethylene (PTFE) [4], which is an advanced high-molecular polymer, suitable for manufacturing of various products [5, 6]. In particular, PTFE-based composites are used for the fabrication of seals in flange connections, since they meet all requirements for such components. They include prolonged service life, great resistance to wear, negative environmental impacts, as

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well as chemical, aggressive and abrasive substances. At the same time, PTFE has a very low friction coefficient [7, 8]. Its loading with graphite enhances the seal durability by many times due to the formation of a strengthened transfer film on the surface of tribological counterparts (shafts, rods, gears, fittings, etc.) [9–11]. The functional characteristics of PTFE are determined by its specific chemical and supermolecular structure [12–15], among which the most outstanding are high thermal resistance at temperatures up to 260 °C, as well as flexibility and elasticity in the range from -70 to +270 °C [16]. The PTFE-based composites are also applied for the manufacturing of piston rings for gas compressors, sealing gaskets, rings and cuffs for connecting equipment units, operating both stationary and at relatively low sliding speeds in the chemical, as well as oil and gas industries. The required set of the functional characteristics of the PTFE-based composites for various operating conditions is ensured by variations in their composition and production routes [17]. As most polymers, PTFE possesses low thermal conductivity that does not enable to dissipate heat properly when seals are operated at elevated temperatures. One of the methods to solve this issue is the addition of fillers, primarily metal powders. In this case, rising the dimensions of their particles substantially increase this parameter for such composites [18], but simultaneously deteriorates their mechanical properties, since such inclusions act as stress concentrators under applied loads. Ceteris paribus, stress peaks near inclusions contribute to the initiation of cracks in the composites, leading to their preliminary failure. It should be noted that the stress peaks enhance with increasing the dimensions of particulate filler. Thus, the problem arises of searching for a compromise between increasing thermal conductivity of the PTFE-based composites and reducing their mechanical properties. This research proposes an approach for solving this problem, considered in relation to the design of ones, characterized by both increased thermal conductivity and acceptable mechanical properties.

1. Problem statement and research method

The objects of the study were the PTFE-based composites that included the F-4 polymer matrix (according to the Russian state standard GOST 10007-80) and loaded with two industrially available fine copper powders with particle sizes of 1.5 µm (met the technical specification TU 1793-011-50316079-2004, designated as "filler 1") and 60 µm (met the Russian state standard GOST 4960-2009, designated as "filler 2") [19]. The key physical properties of the composites components in Tab. 1.

Table 1. The key physical properties of the composites components

Parameter	Matrix (PTFE)	Filler 1 (1.5 µm copper powder)	Filler 2 (60 µm copper powder)
Elastic modulus, MPa	410	129800	110000
Poisson's ratio	0.37	0.32	0.32
Yield point, MPa	11.8	–	–
Ultimate tensile strength, MPa	20	–	–
Stress at break, MPa	27	314	–
Elongation at break, %	250-500	–	–
Density, kg/m3	2200	8960	8930
Specific heat capacity, J/(kg °C)	1040	–	385
Thermal conductivity, W/(m °C)	0.200	–	401

Computer-aided design of the PTFE-based composites with the required properties was carried out as follows. Initially, it was necessary to find a combination of the control parameters that would ensure the maximum thermal conductivity value ($\geqslant 0.3$ W/m °C) and elongation at break of $\geqslant 0.02$. To solve this problem, an approach proposed the authors of [20–22] was modified in relation to loading the polymer matrix with particles whose sizes differed by more than an order of magnitude. An algorithm of computer-aided design of the composites with the required set of properties is shown in Fig. 1. At the first stage of computer-aided design, both mechanical and thermophysical properties were assessed for the composites. In this way, both stress-strain state (SSS) parameters and thermal conductivity values were calculated for various applied loads and thermal fluxes using the equations of mechanics of a deformable solid and solving thermal conductivity problems. In general, the functional characteristics, the filler sizes and shapes, the filling degree and other parameters determining both composition and properties of the composites could be optimized. In the studied cases, the filling degree was varied for both loaded copper powders. The problems were solved using the Abaqus CAE software package based on the finite element method.

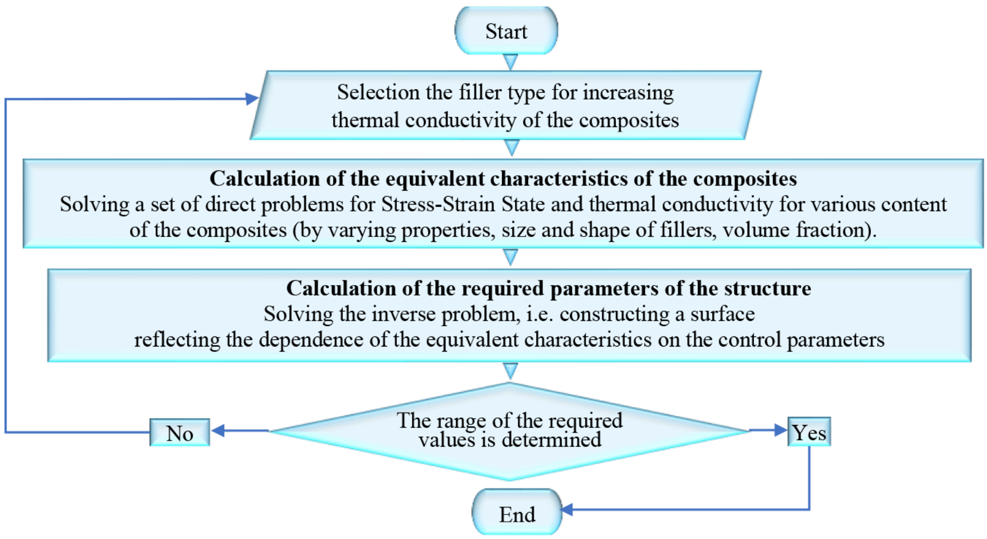

Fig. 1. The algorithm of computer-aided design of the PTFE-based composites with the required set of properties

At the second stage, inverse problems of computer-aided design were solved, generally aimed at manufacturing the PTFE-based composites with the specified properties. A solution was based on constructing response surfaces of the functional characteristics to the control parameters in a space of states. To achieve this goal, a number of reference points, obtained from solving linear equations, were used. The presence of particles with sizes differing by more than an order of magnitude complicated the considered algorithm (Fig. 1) when solving the direct problems, since they involved several structural levels. Their number (two for the studied cases) depended on the size of fillers [22, 23]. At the first level, solving the direct problems made it possible to calculate the functional characteristics of the matrix loaded with filler 1 (1.5 µm). Then, such a composite was considered as the matrix loaded with filler 2 (60 µm) at the second level. As a result, the functional characteristics were assessed for the composites as a whole. An implemented multi-scale algorithm is presented in Fig. 2.

Fig. 3 shows computational domains implemented for determining the SSS parameters under uniaxial tension and for solving the thermal conductivity problems in relation to the PTFE-based composites loaded with copper particles of different sizes.

At the first structural level, the rectangular computational domain was characterized by a side of 25 µm and particles with a radius of 1.5 µm (Fig. 3, right). The bottom edge of the domain

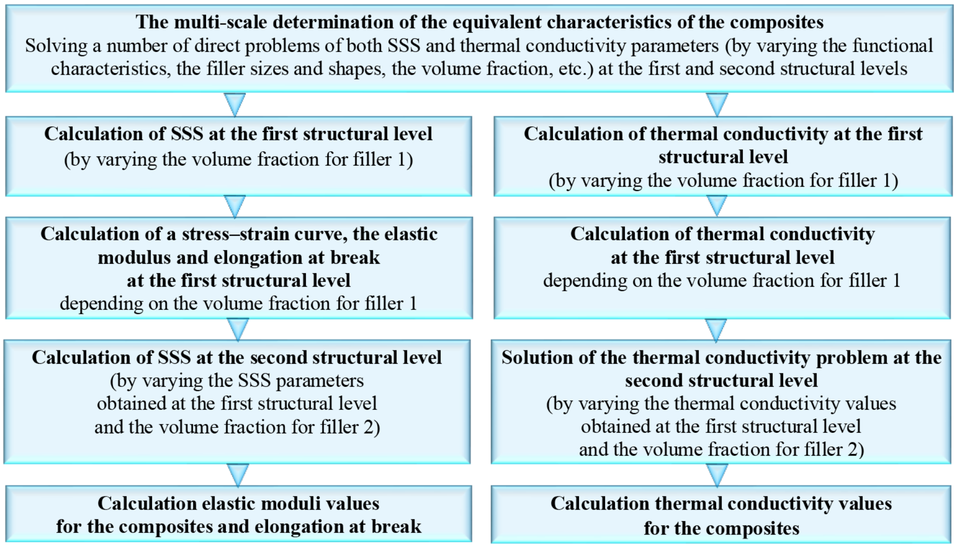

Fig. 2. The multi-scale algorithm for solving the direct problems to determine the physical and mechanical properties of the PTFE-based composites

was rigidly fixed. On the top edge, a tensile load was applied along the normal axis, while the side edges were stress-free. At the "matrix-particle" boundaries, ideal adhesion conditions were assumed. The problems were solved in a flat statement. Upon loading, the non-linear PTFE properties were taken into account. The calculated results, obtained according to the algorithm shown in Fig. 2, are given below.

2. Results and discussion

The calculated stress-strain properties. After solving the problems of determining the SSS parameters via averaging procedures [22, 23], the mechanical properties of the composites were calculated at the first structural level. Tab. 2 presents dependences of their values on the filling degree for the composite loaded with filler 1 (1.5 µm).

Table 2. The mechanical properties calculated at the first structural level for the composite loaded with filler 1 (1.5 µm)

Volume fraction, φ_m	10 vol.%	20 vol.%	30 vol.%
Elastic modulus, MPa	471	487	495
Poisson's ratio	0.370	0.381	0.376
Elongation at break	0.068	0.064	0.045

Rising the volume fraction expectedly led to an increase in the elastic modulus and a decrease in the average stresses over the computational domain (Fig. 4), corresponding to the crack initiation stage (marked by an elliptical contour).

Fig. 5 shows stress-strain curves obtained by stretching composites with different degrees of filling, characterizing the effective properties of PTFE loaded with filler 1 (1.5 µm).

Further, the mechanical properties obtained at the first structural level were used to determine the SSS parameters for the composite loaded with filler 2 (60 µm). In this way, they were

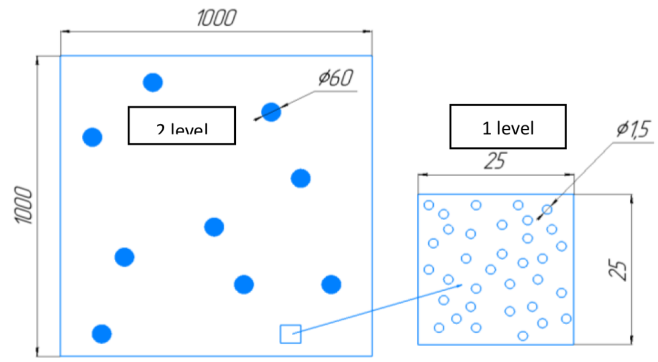

Fig. 3. The computational domains for determining the physical and mechanical propertiesat the first and second structural levels (dimensions are in microns)

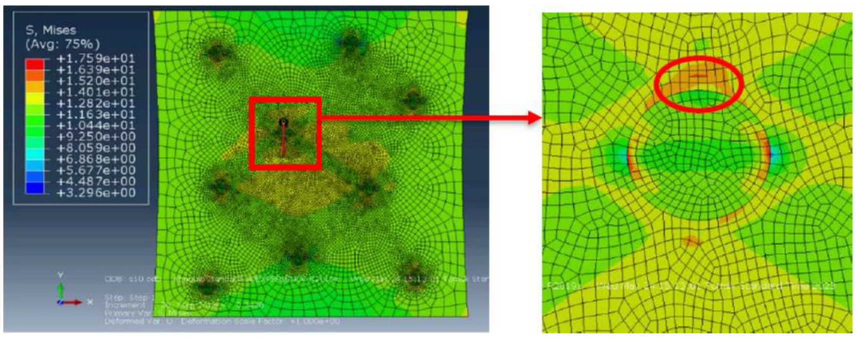

Fig. 4. A tensile stress distribution for the composite loaded with 10 vol.% filler 1 at the crack initiation stage

designated as values for the "modified matrix". Fig. 6 shows a stress distribution in the computational domain during uniaxial tension of the composite loaded with 30 vol.% filler 1 (60 µm) at the crack initiation stage (represented by a horizontal segment inside the elliptical contour).

Tab. 3 shows dependences of elongation at break versus the volume fraction and the elastic modulus of the modified matrix. It should be noted that the data obtained at the second structural level were similar to those for the first one. In particular, rising the volume fraction was accompanied by enhancing the elastic modulus and lowering the strain level, corresponding to the beginning of fracture under the tensile load.

Calculated thermophysical properties of the composites. An approach applied previously for particulate composites [23, 24] was to a certain extent similar to the method of determining the mechanical properties in the studied cases. The static thermal conductivity problem was solved in relation to the non-uniform computational domain, taking into account the specific characteristics of both matrix and filler materials. As a result, a temperature distribution over the computational domain was calculated, on the basis of which thermal conductivity was assessed according to the method proposed in [24]. The computational domains were the same as those used to determine the SSS parameters shown in Fig. 3. The lowest preset temperature level was 0 °C, while the highest one was 20 °C. The obtained thermal conductivity values are presented in Tab. 4 for the modified matrix loaded with filler 2 (60 µm) at its different volume fractions. With rising the contents of thermally conductive copper particles, the corresponding parameter enhanced for the composites as a whole. At the second structural level, the thermal conductivity

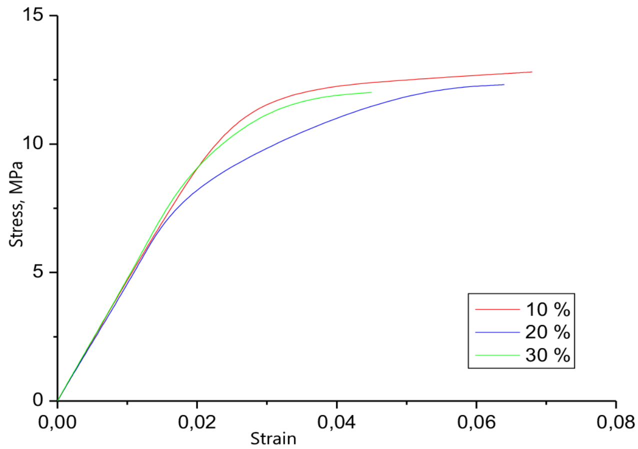

Fig. 5. The stress-strain curves for the composite loaded with filler 1 (1.5 µm) at its different filling degrees

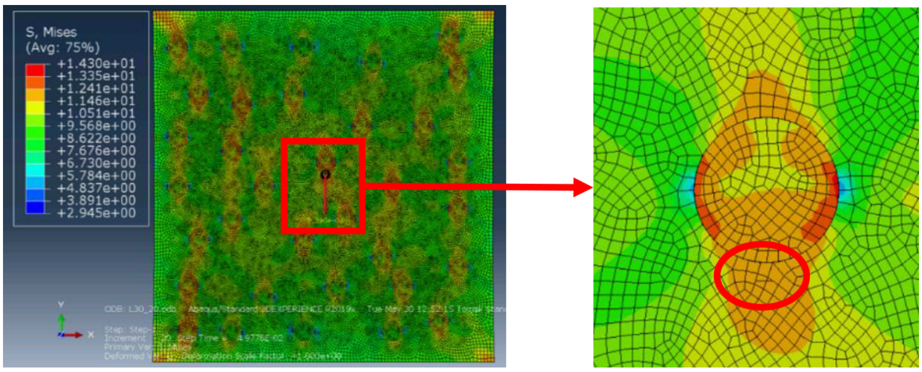

Fig. 6. The stress distribution in the computational domain during uniaxial tension of the composite loaded with 30 vol.% filler 1 (60 µm) at the crack initiation stage

problem was solved in the same way.

As an example, Fig. 7 shows a temperature distribution in the composite loaded with 20 vol.% filler 2 (60µm)

Tab. 5 presents the thermal conductivity values for the composite loaded with filler 2 (60 µm), obtained at the second structural level depending on both its volume fraction and thermal conductivity of the modified matrix. It could be concluded based on these data that an increase in the volume fraction expectedly caused rising the thermal conductivity values.

Considering that the thermal conductivity value of PTFE was taken as 0.20 W/(m °C), the data presented in Tab. 5 indicated the possibility of its rising up to 0.33 W/(m °C), i.e. by more than 60%. In this case, as shown above, the limiting elongation at break level decreased, but not as significantly as after loading with the only filler 2 (60 µm). So, loading with both fillers, corresponding to two structural levels that enabled to improve the functional characteristics of the PTFE-based composites, gave the much greater positive effect. The data presented in Tabs. 3 and 5 are also shown in Figs. 8 and 9, respectively, as surfaces and ranges of the control parameters that satisfied the specified boundary conditions. Since the thermal conductivity and elastic modulus values were obtained at the first structural level for different volume fractions of

Table 3. The dependences of the elongation at break values on the Em elastic modulus of the modified matrix and the φm volume fraction for the filler 2 (60 µm)

Elastic modulus E_m, MPa	Volume fraction, φ_m		
	10 vol.%	20 vol.%	30 vol.%
471	0.0220	0.0226	0.0203
487	0.0279	0.0260	0.0245
495	0.0191	0.0187	0.0176

Table 4. The thermal conductivity values for the modified matrix obtained at the first structural level

Volume fraction, φ_m	10 vol.%	20 vol.%	30 vol.%
Thermal conductivity, W/(m °C)	0.240	0.254	0.277

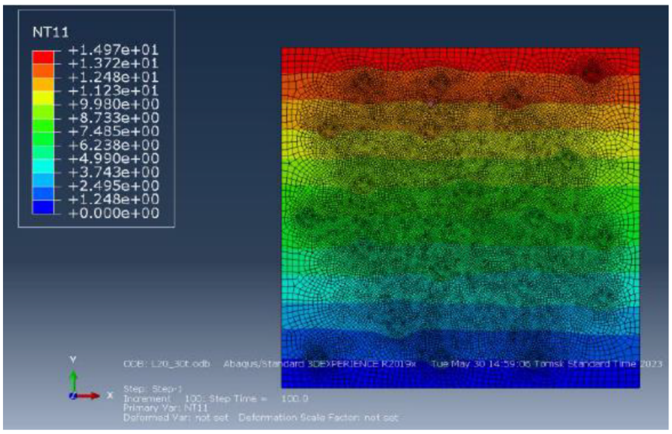

Fig. 7. The temperature distribution in the composite loaded with 20 vol.% filler 2 (60 µm)

Table 5. The dependence of thermal conductivity W/(m °C) of the composites on both fillers volume fraction and thermal conductivity of the modified matrix

Thermal conductivity of the modified matrix, W/(m °C)	Volume fraction, φ_k 10%	20%	30%
0.240	0.250	0.266	0.290
0.254	0.266	0.280	0.305
0.277	0.290	0.307	0.330

filler 1, the horizontal scale corresponds to this parameter, while the axis values are normalized for drawing the continuous surfaces in Figs. 8 and 9.

Fig. 10 shows the overlapped areas that determined the control parameters and the required functional characteristics, i.e. thermal conductivity of $\geqslant 0.3$ W/(m °C) and elongation at break $\geqslant 0.02$.

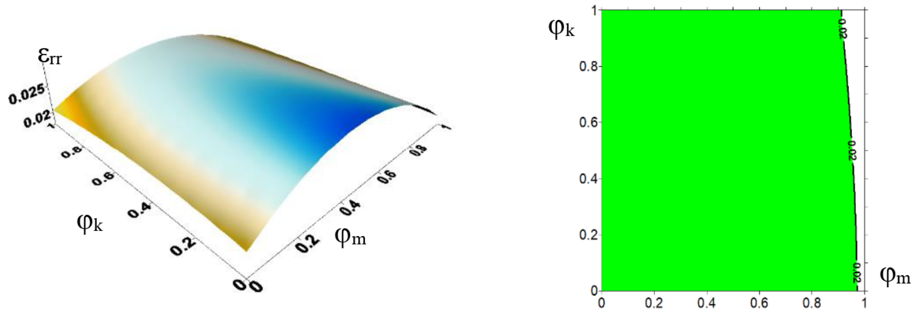

Fig. 8. The temperature distribution in the composite loaded with 20 vol.% filler 2 (60 μm)

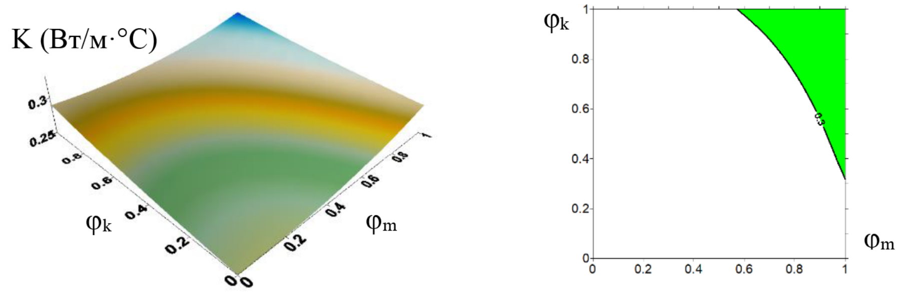

Fig. 9. The temperature distribution in the composite loaded with 20 vol.% filler 2 (60 μm)

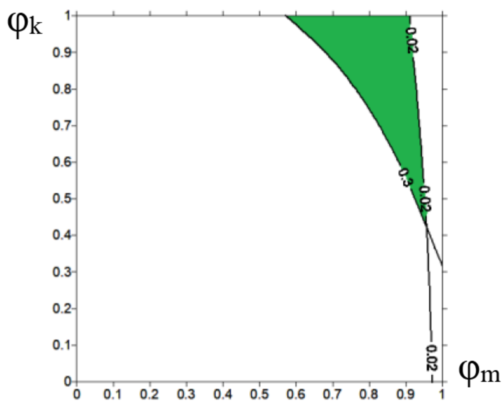

Fig. 10. The temperature distribution in the composite loaded with 20 vol.% filler 2 (60 μm)

Conclusions

In the study, the advanced approach was implemented to increase thermal conductivity of the PTFE-based composites with the minimal decrease in their mechanical properties. Loading with copper powders characterized by different particle sizes made it possible to solve this issue. Due to the large difference in the particle sizes, it was done at several structural levels. In this way, the sophisticated algorithm was developed and applied by implementing the multi-scale model.

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Компьютерное конструирование полимерных композитов с использованием многоуровневых моделей

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Аннотация. В работе решалась задача компьютерного конструирования дисперсно-наполненного полимерного композита на основе политетрафторэтилена (ПТФЭ) с целью повышения его теплопроводности (получения максимально возможного значения его коэффициента) с учетом ограничений на деформационно-прочностные свойства. Для решения задачи разработан и применен алгоритм с использованием многоуровневой модели. Показано, что за счет использования в качестве наполнителей частиц меди двух размерных диапазонов, отличающихся размерами более чем на порядок, можно получить материалы, которые характеризуются высокой теплопроводностью при сохранении приемлемых деформационно-прочностных свойств.

Ключевые слова: многоуровневое моделирование, полимерный композит, дисперсное наполнение, напряженно-деформированное состояние, теплопроводность, ПТФЭ.