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Data Article

Data on the synergistic effect of a hybrid filler based on graphene nanoplates and multiwalled nanotubes for increasing the thermal conductivity of an epoxy composite

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a r t i c l e i n f o

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a b s t r a c t

The dataset contains raw files related to the manuscript "The synergistic effect of a hybrid filler based on graphene nanoplates and multiwalled nanotubes for increasing the thermal conductivity of an epoxy composite" (Shalygina T.A. et al., 2021). The study presents the values of the heat capacity used to calculate the coefficients of thermal conductivity of epoxy composites by combining one-dimensional multiwalled nanotubes (MWCNTs) and two-dimensional graphene nanoplates (GNPs) in the role of a heat-conducting filler. To determine the heat capacity of materials with different concentrations of hybrid filler (GNP/MWCNT), the method of differential scanning calorimetry in the mode of the heat flux modulation was used. The analysis of the heat flux modulation samples is presented in raw and processed form.

The materials scientists may apply the dataset to an in-depth study of the thermal conductivity formation mechanisms in composites doped with carbon-containing substances.

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Specifications Table

Value of the Data

- It is found that the nanocomposite exhibits improved heat-conducting properties when adding a hybrid filler GNP/MWCNT due to the manifestation of a synergistic effect compared with the use of its individual components (GNP or MWCNT). The data show a change in the value of the thermal conductivity coefficient for two different methods of obtaining a hybrid filler and the ratio of filler components (GNP/MWCNT).
- Validation is a method for evaluating the thermal conductivity of a nanocomposite using TMDSC.
- The manuscript provides an idea for modeling the final heat-conducting properties of epoxy composites with a hybrid filler.

1. Data Description

Compared to [\[1\],](#page-5-0) the data presented in this paper are the results of the Temperaturemodulated differential scanning calorimetry (TMDSC) analysis of apparent and specific heat capacity in the epoxy-based composite samples containing different amounts of the hybrid filler. The results are displayed in pictorial form of tables and figures. In addition, the article presents a method for calculating the value of thermal conductivity for polymer composite samples. The detailed raw data on heat capacity were deposited at Mendeley public repository as twelve excel spreadsheets: [https://data.mendeley.com//datasets/z7dsd269vg/2.](https://data.mendeley.com//datasets/z7dsd269vg/2) The tables show the data on the heat capacity of three samples of composites at different concentrations. The obtained values of the heat capacity were used to calculate the values of the thermal conductivity coefficients of composite samples. Further, these data were processed using the excel office program and presented in form of graphs in [Figs.](#page-4-0) 2–4.

2. Experimental Design, Materials and Methods

2.1. Materials

Samples based on T67 epoxy binder (AO INUMit, Moscow, Russia) with various concentrations (0.1, 1, 2.5, and 5 wt %) of a hybrid filler comprised of a mixture of GNPs and MWCNTs (OOO NanoTechCenter, Tambov, Russia) were objects of the study. The samples were produced by mixing the filler with the epoxy resin for 20 min. with subsequent curing in a drier. The multiwalled carbon nanotubes were coaxial carbon nanotubes of the Taunit-M series with a diameter of 8–15 nm and a length of more than 2 μ m, which are synthesized by the method of gas-phase chemical deposition (CVD). The GNPs were prepared by intercalation of natural graphite with a solution of ammonium persulfate in anhydrous sulfuric acid, subsequent thermal expansion, and treatment with gaseous ammonia; a detailed description of the technology for producing GNPs is given in Russian Federation patent no. 2 693 755. [\[2\].](#page-5-0)

Hybrid fillers of three series were used:

- A. 60% graphene, 40% carbon nanotubes, series D (dry) GNP/MWCNT(60/40)D;
- B. 80% graphene, 20% carbon nanotubes, series D (dry) GNP/MWCNT(80/20)D;
- C. 80% graphene, 20% carbon nanotubes, series W (watery)- GNP/MWCNT(80/20)W.

In addition to the composition, the fillers differ in the method of preparation, which is de-scribed in more detail in [\[1\].](#page-5-0)

[Fig.](#page-3-0) 1 shows a scanning electron microscopy (SEM) image of dry hybrid filler GNP/MWCNT(80/20)D. As can be seen from the electron microscopy data, the surface of graphene nanoplates is covered with carbon nanotubes with a length of about 500 nm [\(Fig.](#page-3-0) 1a). MWCNTs that are longer than 2 μ m remain as separate aggregates of nanotubes intertwined with each other. It is likely that short nanotubes were present in the Taunit-M initial carbon product as a small fraction; in this case, it is this fraction that is useful for graphene disaggregation.

2.2. Methods

2.2.1. Method for determining the thermal conductivity of a polymer composite

To obtain the values of thermal conductivity, formulas 1, 2 were used. The heat capacity of the samples was obtained using TMDSC. The thermal conductivity coefficient of the sample is calculated by the formula:

$$
\lambda = \frac{\left[\lambda_0 - 2D + (\lambda_0^2 - 4D\lambda_0)^{1/2}\right]}{2} \tag{1}
$$

where λ_0 - the observed heat conductivity factor, (W/K·m);

D - calibration constant, W/(m·K).

The observed heat conductivity factor λ_0 (W/K·m) was estimated as

$$
\lambda_0 = (8LC^2) \cdot (C_p m d^2 P) \tag{2}
$$

Fig. 1. SEM image of GNP/MWCNT hybrid filler (a) and TEM images initial multilayer GNP with the number of layers varies in the range from 5 to 25 (b,c).

where *L - sample length, mm;*

C is the apparent heat capacity, μJ/K; *^С*^р is the specific heat capacity, J/(g·K); m is the mass of the sample, mg; d - sample diameter, mm; P-period, s.

The calibration constant applied to the thermal conductivity factor *D*, W/(m·K) is calculated by the formula:

$$
D = (\lambda_0 \lambda_r)^{1/2} - \lambda_r \tag{3}
$$

The coefficient of thermal conductivity α is calculated by the formula:

$$
\alpha = \frac{(\pi \lambda d^2 L)}{4C_p m}.
$$
\n⁽⁴⁾

2.2.2. TMDSC-analysis of polymer samples with hybrid filler

The thermal conductivity of epoxy composites with a hybrid filler was measured using a DSC25 differential scanning calorimeter with the possibility of heat flux modulation [\[3\].](#page-5-0) After adding the hybrid filler to the epoxy binder, it was dispersed by mechanical stirring for 10 min. After that, the mixture was poured into TZero aluminum crucibles with a diameter of 5 mm and a height of 3 mm, and placed in a climatic chamber for curing according to the following preset program: 3 h at 80 °C and 6 h at 120°C. After curing, the crucibles were sealed using a hand press. The identical sealed empty crucible was used as a reference sample. The measurements were carried out in a pure nitrogen atmosphere in the temperature range from 15 to 70 \degree C at a

Fig. 2. Thermal conductivity of epoxy composite with filler A (29 (60-40) D).

Fig. 3. Thermal conductivity of epoxy composite with filler В (38 (80-20) D).

heating rate of $1^{\circ}C/\text{min}$; the heat flux modulation was characterized by a period of 40 s and an amplitude of ± 0.5 °C.

2.2.3. Change in the thermal conductivity of the epoxy composite

Figs. 2–4 show the thermal conductivity vs. concentration curves for a range of the hybrid filler series and various contents in the epoxy composites. The thermal conductivity was estimated from the input data presented in the Mendeley public repository.

Fig. 4. Thermal conductivity of epoxy composite with filler С (39 (80-20) W).

Ethics Statement

The authors read and follow the ethical duties of authors.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT Author Statement

T.A. Shalygina: Conceptualization, Methodology, Formal analysis, Writing – original draft; **S.Yu. Voronina:** Conceptualization, Methodology, Formal analysis, Writing – original draft; **A.G. Tkachev:** Methodology, Investigation; **N.N. Grotskaya:** Writing – original draft; **V.D. Voronchikhin:** Conceptualization, Methodology; **A.Yu. Vlasov:** Conceptualization, Methodology.

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- [1] T. A. Shalygina, A. V. Melezhik, A. G. Tkachev, S. Y. Voronina, A. Y. Vlasov, V. D. Voronchikhin, Technical Physics Letters, 2021, Vol. 47, No. 4, 317–320 doi[:10.1134/S1063785021040143.](https://doi.org/10.1134/S1063785021040143)
- [2] RU2693755C1.
- [3] ASTM Е1952-11 "Standard test method for thermal conductivity and thermal diffusivity by modulated temperature differential scanning calorimetry", MOD.