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Study of thermal conductivity of polymer materials with carbon nanotubes using laser flash method

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Abstract. The thermal conductivity of nanocomposite polymer materials based on polyvinyl alcohol and epoxy resin with the inclusion of multi-walled carbon nanotubes was studied using the laser flash method. The possibility of using the laser flash method to determine thermal conductivity in thin-film polymer materials is demonstrated and its features are revealed. It has been shown that the presence of carbon nanotubes in polymer matrices leads to an increase in the coefficients of thermal diffusivity and thermal conductivity by several times compared to pure polymer materials.

Keywords: carbon nanotubes, polymers, thermal conductivity, laser flash method

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Материалы конференции
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Исследование теплопроводности полимерных материалов с углеродными нанотрубками методом лазерной вспышки

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Аннотация. Методом лазерной вспышки исследована теплопроводность нанокомпозитных полимерных материалов на основе поливинилового спирта и эпоксидной смолы с включением многостенных углеродный нанотрубок. Продемонстрирована возможность и выявлены особенности использования метода лазерной вспышки для определения теплопроводности в тонкопленочных полимерных материалах. Показано, что наличие в полимерных матрицах углеродных нанотрубок приводит к увеличению коэффициентов температуропроводности и теплопроводности в несколько раз по сравнению с чистыми полимерными материалами.

Ключевые слова: углеродные нанотрубки, полимеры, теплопроводность, метод лазерной вспышки

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Introduction

Polymer nanocomposites with carbon nanotubes (CNTs) serve as the basis for creating microelectronic sensors, optical elements, protective coatings and other products [1]. One of the characteristics of polymer nanocomposite materials that determines their functional properties is thermal conductivity [2]. It is expected that the use of CNT additives in polymeric materials can significantly improve their temperature characteristics. The purpose of this work was to study the thermal conductivity of polymer materials with different concentrations of CNTs using a promising non-contact laser flash method (LFM). In LFM, the upper surface of the controlled object is exposed to a short pulse of heating focused laser radiation, and at certain points on the surface on the back side of the object, a thermal imager or IR camera measures the change in temperature over time. It is also of interest to determine the possibilities and features of using LFM for studying polymeric materials, including in the form of thin films.

Materials and Methods

In this study, we used multi-walled carbon nanotubes (MWCNTs) with a diameter of 40-80 nm, synthesized by the MOCVD method at the experimental facility of the Ulyanovsk State Technical University [3]. MWCNTs were subjected to multi-stage liquid-phase treatment to remove metallic impurities and improve the compatibility of MWCNTs with the polymer matrix [4]. Films based on polyvinyl alcohol (PVA) were manufactured using the method described in [5], with different mass concentrations of MWCNTs.

Plates of polymer nanocomposites with a thickness of 2–5 mm and a diameter of 16 cm were obtained on a base of EDP grade epoxy resin, into which, after mixing with a hardener, multi-walled carbon nanotubes (MWCNTs) were introduced, dispersed by ultrasonic treatment [6].

When implementing the LFM using an IR camera as a temperature sensor, a measurement point is selected on the lower surface of the film on the image obtained from the IR camera, located at a distance l from the center of the laser radiation spot on the upper surface of the film (Fig. 1, *b*). For a point opposite the center of the laser spot on the top surface of the film, this distance is equal to the film thickness d. Based on the measurement results at the selected point, a graph of the dependence of the temperature change $\Delta T(t)$ on time is constructed (Fig. 2). The graph is used to determine the maximum value of the temperature increment ΔT_{max} and the temperature increment ΔT after exposure to a laser radiation pulse (section *b* in Fig. 2). Next, using formula (1):

$$m_0 = \int_{t_{0.1}}^{t_{0.8}} \frac{\Delta T(t)}{\Delta T_{max}} dt, \quad m_{-1} = \int_{t_{0/1}}^{t_{0.8}} \frac{1}{t} \frac{\Delta T(t)}{\Delta T_{max}} dt, \tag{1}$$

we calculate the partial moments of time m_0 and m_{-1}, and using formula (2):

$$F(m_{-1}) = -0.0819 + 0.305 m_{-1}, \tag{2}$$

we calculate the approximating function $F(m_{-1})$.

And finally, using formula (3):

$$\alpha = \frac{d^2 F\left(m_{-1}\right)}{m_0}, \tag{3}$$

the thermal diffusivity coefficient α is calculated, and using formula (4):

$$\lambda = \alpha \rho C_p. \tag{4}$$

the thermal conductivity coefficient λ is calculated, where ρ is the density and C_p is the specific heat capacity of the material.

A laser with a wavelength of 960 nm and a power of 50 mW was used as a source of the heating pulse, and a PI640 Optris IR camera operating in the spectral range of 8–14 μm with a temperature resolution of 0.04 degrees and a frame refresh rate of 32 Hz was used as a sensor.

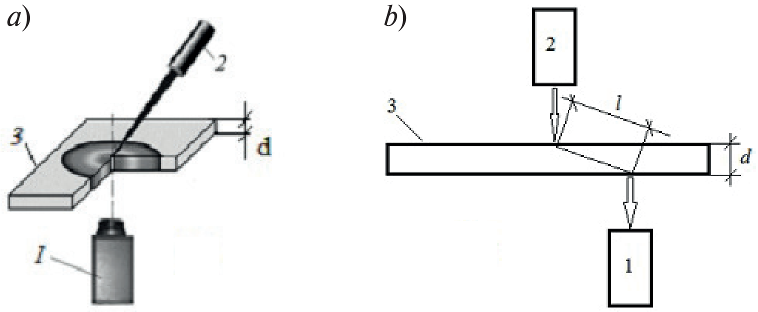

Fig. 1. Scheme (*a*) and geometry (*b*) of the arrangement of the IR camera *1*, laser *2* and object *3* when implementing the LFM

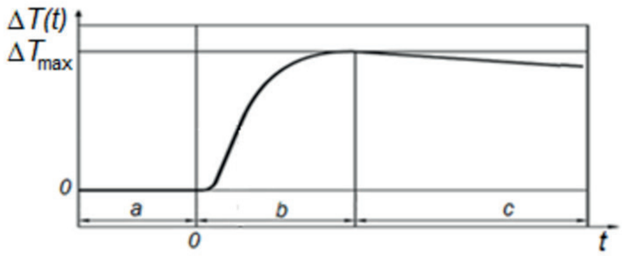

Fig. 2. Temperature change at a given point on the lower surface of the film after exposure to a laser pulse

Results and Discussion

Fig. 3 shows the dependence of the obtained values of the thermal diffusivity coefficient α of PVA films and epoxy resin plates on the concentration of MWCNTs. It can be seen that the addition of MWCNTs to the polymer leads to an increase in the thermal diffusivity and thermal conductivity coefficients several times compared to samples without MWCNTs.

At low concentrations of MWCNTs, the dependence of the thermal diffusivity coefficient α of the polymer nanocomposite on the concentration of MWCNTs C for both polymers is well described within the framework of the partial influence model:

$$\alpha = (1 - C)\alpha_{POL} + C \cdot \alpha_{CNT}, \tag{5}$$

where α_{POL} is the thermal diffusivity of the pure polymer, α_{CNT} is the thermal diffusivity of MWCNTs and $\alpha_{CNT} \approx 2.6...3.2$ m^2/s. One of the possible explanations for the saturation of the dependence of PVA-based composites at MWCNTs concentrations greater than 2% is the process of MWCNTs clustering in the polymer matrix and an increase in contact resistance at the MWCNTs – polymer interface.

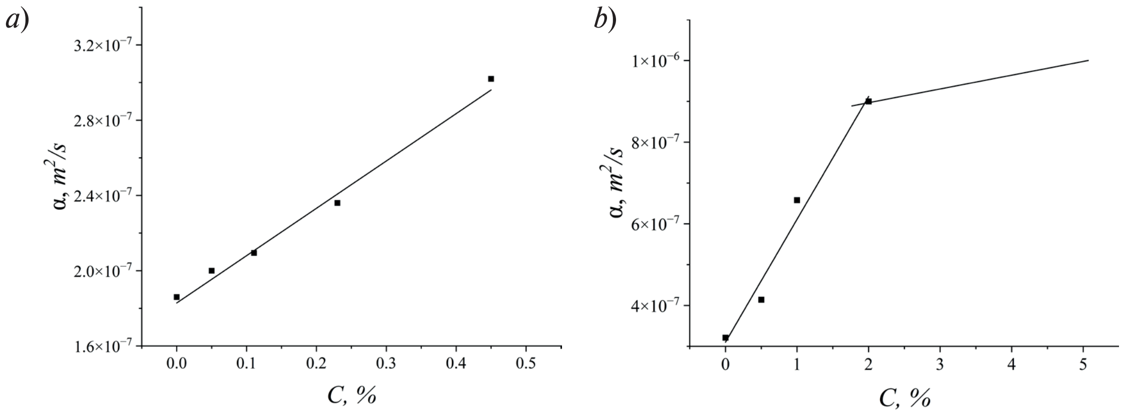

Fig. 3. Dependence of the thermal diffusivity coefficient α of epoxy resin plates (*a*) and PVA films (*b*) on the concentration of MWCNTs

Conclusion

The studies carried out showed a significant effect of the addition of MWCNTs on the thermal conductivity of the resulting polymer composite materials. In our opinion, the LFM for determining the thermal conductivity of polymer nanocomposite materials is a promising method that can be further improved by using, for example, not one, but four measurement points located at the vertices of a square. This orthogonal arrangement of measurement points will make it possible to detect and evaluate the anisotropy of thermal conductivity in composites with ordered nanotubes.

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