Thermal conductivity modeling for composite polypropylene / vapor grown carbon fibers

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Abstract. According to the melt technology a polymer composite material based on polypropylene (PP) matrix and vapor grown carbon fibers (VGCF) as a filler was obtained. The dependences of the thermal conductivity on the filler mass fraction and temperature were experimentally obtained. In this article we demonstrate that the heat-conducting properties of the material don’t depend on the sample geometry and the dependences of the thermal conductivity on temperature and the filler mass fraction have a nearly linear character. It is established that when the VGCF mass fraction is more than 5%, the obtained composite material can be used to remove heat in electrical and electronic devices. Such materials have antistatic properties at the VGCF mass fraction 5%. The model based on the multichannel conduction was built and the task was solved. This method allows the heat transfer process description in composite materials with sharply differing heat-conducting properties of components taking into account such material characteristics as density, heat capacity, heat conduction and heat velocity in the material.

1. Introduction

One of the main problem of the electrical and electronic equipment operation is heating that affects the functionality. So heat removal or cooling is a very important task. There are different ways to solve the problem: air cooling, usage of substrates based on special materials, etc. In addition to heat dissipation properties, it is advisable that the materials have special electrical properties for antistatic effect, good elasticity and affordability.

It is well known that polymer materials have required mechanical properties. That’s why they are used in various devices. But polymer materials have a rather low value of the thermal conductivity: for polypropylene the thermal conductivity value λ=0.22 Wt/(m∙K) [1]. Loading of fillers in the polymer matrix makes a possibility to modernize the properties of materials, including the heat-conductivity [2-4]. Such composite materials may also have a high value of the electrical conductivity, improved mechanical properties. They are called transenergoplastics. Typical values of the thermal conductivity of transenergoplast are 5–15 Wt/(m∙K) which are less than the thermal conductivity of aluminum but under conditions of natural cooling. These values turned out to be quite enough to replace aluminum...
with composite plastics [2-6]. Besides the usage of polymers allows to reduce the price of devices drastically, to make them lighter and more compact.

In most cases, the existing protection is based on the control, as a rule, of effective values of currents and voltages [7-10]. This requires a relatively long observation of signals in emergency situations to make the right decision, which leads to a decrease in performance.

During the work we achieved an increase in the thermal conductivity of polypropylene by loading of carbon fillers such as VGCF.

2. Experimental study of the thermal conductive properties

Samples were obtained in the form of films and fibers by melt technology. VGCF specified amount was dispersed in a polypropylene melt in twin-screw microcounder DSM Xplore 5 ml. For the formation of film and fiber materials were used slotted and round extrusion nozzles. The mass fraction of VGCF were 0, 3, 5, 8, 10, 15 and 20%.

The thermal conductivity was determined using a bridge connection. The operation principle is to measure the thermal resistance in comparison with reference samples. Then the coefficient of thermal conductivity was calculated by the formula

\[ \lambda = \frac{l}{RS} \]

where \( l \) – the sample length, \( S \) – cross sectional area, \( R \) – the thermal resistance.

We experimentally found dependences of the thermal conductivity on the filler mass fraction at temperatures of 10 and 70°C for samples of all geometries (see Figure 1). As expected the thermal conductivity value doesn’t depend on the sample geometry. The low thermal conductivity coefficient of the polypropylene matrix \( \lambda = 0.22 \text{ Wt/(m} \cdot \text{K)} \) with the VGCF floating increased to \( \lambda \approx 5.5 \text{ Wt/(m} \cdot \text{K)} \).

![Figure 1. Experimental and theoretical dependences between the thermal conductivity and the filler mass fraction and temperature: 1 – fibers PP/VGCF, T=10°C; 2 – fibers PP/VGCF, T=70°C; 3 – films PP/VGCF, T=10°C; 4 – films PP/VGCF, T=70°C.](image)

Light emitting diodes need the thermal conductivity value in the range \( \lambda \approx 4÷10 \text{ Wt/(m} \cdot \text{K)} \) [7]. Thus, our obtained composite PP/VGCF can be used as the heat removing material. There is another feature of the studied composite material. Such materials also have antistatic properties in addition to heat removing properties with the VGCF mass fraction of 5-10% [6].

3. Heat transfer modeling

There are various approaches that allow describing the heat transfer in composite materials: models of the mixing rule, the generalized conductivity theory, the Fourier law [8–12]. However all of these approaches have disadvantages.
The mixing rule and the generalized conductivity theory are empirical models and well describe isotropic and homogeneous mixtures. VGCF particles are anisotropic structures. Therefore the VGCF thermal conductivity value is four orders of magnitude higher than the value of the PP thermal conductivity. The PP/VGCF structure cannot be considered as a homogeneous.

The heat distribution can be described by the Fourier law according to which the heat flow is proportional to the temperature gradient. The Fourier law describes quite accurately the heat transfer process at the macroscopic level for systems located near the thermodynamic equilibrium. But the heat distribution in a one-dimensional crystal is difficult to describe using the Fourier law. In addition Fourier’s law cannot describe the instantaneous heat distribution in the system. Therefore significant deviations are observed at small time intervals. The description of multilayer and composite materials is complicated by the specification of boundary conditions.

We proposed a mathematical model in the form of an equation system describing the process of the heat transfer taking into account the flow structure [13]. It was considered the absence of heat exchange with the environment. The heat transfer is carried out through channels each of which has a certain value of the heat transfer velocity $c_n$ and frequency. The quantity of heat in each channel determines the heat flow structure.

The heat transfer process was considered as a one-dimensional problem. Since the thermal energy can propagate in forward and reverse directions along $N$ channels we entered heat flow densities in both directions ($q_n(x,t)$ – the forward direction, $q_n(x,t)$ – the reserved direction) as a local parameters. The heat flow density can be proposed as a system

$$q(x,t) = \sum_{n=1}^{N} q_n(x,t) = \sum_{n=1}^{N} \left[ q_n(x,t) - q_n(x,t) \right]$$  \hspace{1cm} (1)

The transferred part of the internal energy $U(x,t)$ is equal to the sum of the energies in each channel

$$u(x,t) = \sum_{n=1}^{N} \left[ u_n(x,t) - u_n(x,t) \right] = \sum_{n=1}^{N} \frac{1}{c_n} \left[ q_n(x,t) - q_n(x,t) \right].$$  \hspace{1cm} (2)

To derive the heat transfer equations in the general case it suffices to write a system of $2N$ continuity equations for the linear energy density on each channel in accordance with the energy conservation law

$$\frac{du_n}{dt} = - \frac{\partial q_n}{\partial x} - \sum_{m=1}^{N} \left[ a_{nm} + b_{nm} \right] q_n + \sum_{m=1}^{N} b_{mn} q_m + \sum_{m=1}^{N} a_{nm} q_m,$$

$$\frac{du_n}{dt} = \frac{\partial q_n}{\partial x} - \sum_{m=1}^{N} \left[ a_{nm} + b_{nm} \right] q_n + \sum_{m=1}^{N} b_{mn} q_m + \sum_{m=1}^{N} a_{nm} q_m.$$  \hspace{1cm} (3)

Here $a_{nm}$ is the reflection coefficient, i.e. a piece of thermal energy transferred from one flow to another with a transfer direction change, $b_{mn}$ is the transition coefficient, thermal energy transfer from one flow to another without a transfer direction change, $b_{nn} = 0$.

Equations of the system expressed the energy transfer taking into account the internal exchange between the channels. Following notation were introduced

- scattering matrix $\Theta_{kn} = \sum_{m=1}^{N} \left[ a_{km} + b_{km} \right] \delta_{kn} + (a_{kn} + b_{kn})$;

- energy transfer matrix $G_{kn} = c_k \left\{ \sum_{m=1}^{N} \left[ a_{km} + b_{km} \right] \delta_{kn} - (a_{kn} + b_{kn}) \right\}$.

The system of equations (3) takes the form:

\begin{align*}
\frac{du_n}{dt} &= - \frac{\partial q_n}{\partial x} - \sum_{m=1}^{N} \left( a_{nm} + b_{nm} \right) q_n + \sum_{m=1}^{N} b_{mn} q_m + \sum_{m=1}^{N} a_{nm} q_m, \\
\frac{du_n}{dt} &= \frac{\partial q_n}{\partial x} - \sum_{m=1}^{N} \left( a_{nm} + b_{nm} \right) q_n + \sum_{m=1}^{N} b_{mn} q_m + \sum_{m=1}^{N} a_{nm} q_m.
\end{align*}
The first group of \( N \) equations is the continuity equations with internal energy transfer from one channel to another (energy conservation law). The second group of \( N \) equations expresses the generalized Fourier law.

Let’s consider the solution of the obtained equation system (4) for the heat transfer process in a two-component system PP/VGCF in the stationary case. In the first approximation it is believed that there are two channels of heat distribution: the first is for PP, the second is for VGCF. The full transferred internal energy in the composite is the sum of the transferred internal energies in each channel.

\[
 u = u_1 + u_2 = \left[c_{p_1} \rho_1 (1 - \theta) + c_{p_2} \rho_2 \theta \right] T = (\omega^p_1 + \omega^p_2) T,
\]

where \( \omega^p_i \) is the part of energy in each channel (the “\( p \)” index means that an equilibrium is established between the channels, the system has a local temperature \( T(x) \)), \( c_{p_1}, c_{p_2} \) are the heat capacities of PP and VGCF, \( \rho_1, \rho_2 \) are the PP and VGCF densities, \( \theta \) is the VGCF volume fraction in the composite. Thus get the system

\[
 \sum_{m=1}^{N} \omega^p_m G_{mn} = 0
\]

\[
 \sum_{m=1}^{N} \omega^p_m = 1
\]

which determines the value \( \omega^p_i \).

The part of energy in the first channel

\[
 \omega^p_1 = \frac{c_{p_1} \rho_1 (1 - \theta)}{c_{p_1} \rho_1 (1 - \theta) + c_{p_2} \rho_2 \theta}
\]

The part of energy in the second channel

\[
 \omega^p_2 = \frac{c_{p_2} \rho_2 \theta}{c_{p_1} \rho_1 (1 - \theta) + c_{p_2} \rho_2 \theta}
\]

Scattering matrix and energy transfer matrix are

\[
 \Theta_{kn} = \begin{pmatrix}
 2a_{11} + a_{12} & a_{11} - b_{12} \\
 a_{21} - b_{21} & 2a_{22} + a_{21}
\end{pmatrix}
\]

\[
 G_{kn} = \begin{pmatrix}
 c_{1}(a_{12} - b_{12}) & -c_{1}(a_{12} - b_{12}) \\
 -c_{2}(a_{21} - b_{21}) & c_{2}(a_{21} - b_{21})
\end{pmatrix}
\]

The equation system (4) put on
\[
c_{p1} \rho_{c1} c_{p1} \rho_{c1} \frac{\partial T}{\partial x} + q_1 \Theta_{11} + q_2 \Theta_{21} = 0
\]
\[
c_{p2} \rho_{c2} c_{p2} \rho_{c2} \frac{\partial T}{\partial x} + q_1 \Theta_{12} + q_2 \Theta_{22} = 0
\]

(5)

where \( c_1 \) and \( c_2 \) are energy transfer speeds in PP and VGCF. The solving of the system (5) for \( q \) is

\[
q = q_1 + q_2 = \frac{(2a_{22} + a_{21})c_{p1} \rho_{c1} c_{p1} \rho_{c1} + (2a_{11} + a_{12})c_{p2} \rho_{c2} c_{p2} \rho_{c2} \frac{\partial T}{\partial x}}{4a_{11}a_{22} - b_{12}b_{21}}
\]

The expression in parentheses is the thermal conductivity. Simplification of this gives

\[
\lambda = \frac{(2a_{22} + a_{21})c_{p1} \rho_{c1} c_{p1} + (2a_{11} + a_{12})c_{p2} \rho_{c2} c_{p2} \frac{\partial T}{\partial x}}{4a_{11}a_{22} - b_{12}b_{21}} = \alpha_{c1} \lambda_1 + \alpha_{c2} \lambda_2 + \alpha_{c1} \lambda_1 \frac{b_{12}}{2a_{22}} + \alpha_{c2} \lambda_2 \frac{b_{21}}{2a_{11}},
\]

where \( \lambda_1, \lambda_2 \) – the thermal conductivity of PP and VGCF, \( a_{11} = \frac{c_{p1} \rho_{c1} c_{p1}}{2\lambda_1} \), \( a_{22} = \frac{c_{p2} \rho_{c2} c_{p2}}{2\lambda_2} \) – heat diffusion. Coefficients \( b_{12}=a\theta, b_{21}=a(1-\theta) \) are inversely proportional to the effective scattering cross section.

Figure 1 shows the curve of the solution to the multichannel conductivity problem. Experiment and theory agree quite well at different temperatures.

4. Conclusions
1. Samples of composite materials based on polypropylene and carbon fibers were obtained. Such materials are applicable for heat removal in electrical and electronic devices.
2. The dependences of the thermal conductivity on the filler mass fraction were experimentally determined at various temperatures for PP / VGCF. It is shown that such dependencies have a close to linear character.
3. It was offered a model which allows describing the heat transfer in composites with sharply differing heat-conducting properties of components taking into account material characteristics such as density, heat capacity, thermal conductivity, the rate of propagation of heat flow in the material.

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