Investigation of the temperature dependences of the thermal conductivity of epoxy carbon-fibre-reinforced plastics

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SUMMARY

Investigations were made of the temperature dependences of thermal conductivity for specimens of polymer composite material in which epoxy novolac resin was used as the matrix and in which carbon nanotubes were used as the fillers. Investigations of the temperature dependences of the thermal conductivity of polymer composite materials containing carbon fibres were conducted on a special measuring system. A negligible increase in the thermal conductivity of the composites was shown.

Epoxy oligomers and polymers are widely used as matrices for the production of carbon-fibre-reinforced plastics characterised by a combination of high strength and rigidity with low density, a low temperature coefficient of friction, high thermal and electrical conductivity, wear resistance, and resistance to thermal and radiation effects. Coked and pyrocarbon epoxy carbon-fibre-reinforced plastics are resistant to thermal and thermooxidative degradation, have high strength characteristics, and possess good heat-shielding properties [1].

At present, in view of the creation of equipment radiating a broad spectrum of electromagnetic modes, the problem of the electromagnetic compatibility of technical equipment and the problem of ensuring ecological safety in residential areas have become increasingly acute. The most promising materials for use as electromagnetic radiation screens are polymer composite materials consisting of a non-conducting polymer matrix and a disperse conductive filler. Polymer composites with conductive fillers have reached a technical level at which they can stand up to competition with traditional conductive materials [1, 2].

Studies are known [2–7] where a detailed investigation is made of the properties of different composite materials comprising a polymer with the addition of a certain number of multilayer carbon nanotubes (CNTs).

In this work, experimental investigations of the temperature dependences of thermal conductivity were conducted on specimens of polymer composite materials, the matrix of which was epoxy novolac resin DEN 425 cured with dicarboxylic acid methylendic anhydride (MEA). As the accelerator in the composite, use was made of 2,4,6-tris(dimethylaminomethyl)phenol (UP 606/2), and multilayer carbon nanotubes (CNTs) in a solution of Laprolat-301 (Laprolat-301 = oligoester cyclocarbonate) [4] were used as the fillers.

The creation of epoxy composites modified with carbon nanotubes (CNTs) presents considerable difficulties in achieving a homogeneous distribution of the nanotubes in the matrix on account of high energetic activity and a tendency towards aggregation and sedimentation in the less dense oligomer medium [2].

The technology for preparing test specimens from composites was as follows. In the mixing vessel, to epoxy novolac resin was initially added a paste of nanoparticles in active diluent, then the curing agent, and finally the accelerator. The mixture was left to stand at room temperature for about 1 h to allow the air bubbles
to escape. The compositions were then poured into a preheated mould (about 100°C). The mould was placed in an oven heated to temperatures of 160–180°C. The holding time at temperature was 2 h.

Investigations of the temperature dependences of the thermal conductivity of polymer composite materials containing carbon fibres were conducted on a measuring system (MS) making it possible in a single short-time experiment to determine the temperature dependences of the thermal conductivity of solid materials in program-specified temperature intervals. To measure thermal conductivity, dynamic \( \lambda \)-calorimetry is used in the MS [8].

The MS was built as a result of substantial modification of the IT-400 thermal conductivity meter [8–11]. Signals from thermocouples are sent to the inputs of a PSI-1202H analogue/digital board (ADB: 12 bit, 44 kHz; DAB: 12 bit). The board has a programmed signal amplifier, which makes it possible to alter the range of the voltage supplied. Heating of the MS adiabatic jacket is realised by the system software via the DAB board output channels. The supply voltage of the main electric heater of the measuring cell ensures that conditions for a steady-state thermal regime of the second kind are observed during heating of the specimen. During the experiment, programmed control is carried out by Delphi software.

The MS is shown in Figure 1. The core of the MS is the measuring cell, which consists of an adiabatic jacket 1, a base 5, a thermometer 4, and a rod 2, between which the test specimen 3 is placed. The sensitive elements of the MS are thermocouples, the cold junctions of which are soldered to the inputs of the cold junction block 9. The inputs of the block are heated by a solid aluminium block, the temperature of which is determined by an incorporated gauge with potential output. The signal \( H \) from the gauge and also the signals \( A \) from the thermocouples are sent to the ADB/DAB boards 11 of the computer 10. Cold-junction temperature compensation is done by the MS software.

Control of the measurement process is done by the system software. The developed program processes data arriving from the MS thermocouples. Control of the measurement process is done by the software by sending signals \( G \) via the DAB to the supply and regulation block 12. Control of the heating process is done by changing the power released by the heaters 6 and 7.

In order to protect elements of the measuring cell against overheating, the MS includes an apparatus protection and commutation block 13. The main part of the block is a relay, which reacts to the signal \( D \) from the temperature gauge 8 and protects the MS against overheating. The block implements the commutation algorithm through its relay, and also controls the power relay 14 connecting the supply and regulation block to the circuit 15.

The MS was calibrated to certified specimens (glass KV, glass TF-1, copper), with determination of such constants as the coefficient of transformation of the thermometer and correction of the magnitude of the contact thermal resistances between the specimen and the surface of the thermometer (Figure 2).

Information is gathered during heating. Processing of measurement results is simplified owing to the developed software in the MS.

As a rule, the introduction of fillers or modifiers into a polymer affects the thermal conductivity of the latter, and here the numerical value of thermal conductivity (\( \lambda \)) of the composite material will be determined not only by the amount of additive introduced but also by the nature of its interaction with the polymer phase.

![Figure 1. Measuring system layout: 1 – adiabatic jacket; 2 – rod; 3 – specimen; 4 – thermometer; 5 – base; 6 – jacket heater; 7 – base heater; 8 – temperature gauge; 9 – cold junction box; 10 – PC; 11 – PCI-compatible ADB/DAB board; 12 – supply and regulation block; 13 – apparatus protection and commutation block; 14 – relay; 5 – 220 V, 50 Hz circuit; A – measuring signals of thermocouples; B – supply of base heater; C – supply of jacket heater; D – signal from base temperature gauge; E – power source for supply and regulation block; F – supply of cold junction block; G – heating control signal; H – signal from the thermocouple cold junction temperature gauge](image)

![Figure 2. The temperature dependences of the thermal conductivity of certified specimens of glass KV (1) and glass TF-1 (2)](image)
The temperature dependences of the thermal conductivity of composite materials based on epoxy novolac resin with CNT filler are presented in Figure 3. Each of the dependences 1 to 3 comprises the result of averaging five parallel tests.

The filling of epoxy novolac resin DEN 425 with carbon nanotubes in a quantity of 1–2% raises the thermal conductivity of the material slightly in the entire investigated range of temperatures (70–160°C), without changing the nature of the dependence.

Generalising the above, it can be said that, in spite of the high thermal conductivity of individual carbon nanotubes [2] entering the composition of the material, the thermal conductivity of composites produced with their use is raised negligibly. The reasons for this are possibly the ability of the CNTs to absorb gaseous and liquid substances (air, oligoester cyclocarbonate) and the non-uniform distribution of the carbon nanofibres in the polymer matrix [12].

Thus, the problem of creating methods for distributing carbon nanotubes in polymer materials remains pressing. The use, for example, of ultrasonic treatment improves considerably the dispersion of the CNTs in the polymer matrix [2]. Account must also be taken of the fact that the degree of homogeneity of composite material containing CNTs depends considerably on their concentration. With low concentrations, a high degree of homogeneity of the material is easier to achieve, because in this case it is possible to disperse the bundles containing the nanotubes. The properties of materials containing CNTs can also be enhanced by using additional manipulations with CNTs [2, 12].

REFERENCES


