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ТЕПЛОМАСООБМІН

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Thermal diffusivity of polymeric composite cured in a magnetic field

Curing of the composite in a constant magnetic field of low intensity (H~10^4 *A/m) leads to the anisotropic spatial distribution of the filler particles - carbonyl nickel powder. Thermal diffusivity of the composite with orthotropic structure was measured using "pulsed heat source" method. Increase of thermal diffusivity value was found in the direction of the orthotropic axis.*

Introduction. Polymeric composite materials (PCM) uniquely can combine mechanical, electrical and thermo physical properties (TPP). Required in many cases for practical purposes increase of the composite's thermal conductivity is usually carried out by increasing the concentration ofthermal conductivity filler (soot, carbon fibers, metal powder, etc.). Limitations of this method of increasing the thermal conductivity of the material is associated with a significant deterioration of the mechanical properties of PCM with an excess of filler. More opportunity is provided by creating anisotropy in PCM and simultaneously increasing of the filler particles content. For example, in the manner previously proposed for pyrotechnical compositions [1, 2] and subsequently then used [3] to PCM – by creating an anisotropic spatial distribution of the metal powder particles in a polymeric binder.

The purpose of the work was to determine the ability to control the value of thermal diffusivity of PCM by changing filler concentration and creating an anisotropic (orthotropic) spatial distribution of the particles.

Preparation of test specimen. Such anisotropic (orthotropic) structure of PCM was created, as in [3], by polymerization of powdered (average particle size 100 nm) nickel carbonyl (GOST 9722-71) polymer/filler mixture) in a magnetic field of a solenoid. However, instead of oligomer used in [3], epoxy resin EDR-20 (made in Czech Republic) was selected as a binder in the present test. This polymer provides high durability and hardness (necessary for machining the cured composite in obtaining samples for measurement) in a wide temperature range and good thermodynamic compatibility with excipient. Thoroughly stirred and degassed mixture of resin (hardener PEPA, TU-2413-357-00203447-99 and isobutyl glycidyl) with filler in a plastic cylindrical container was placed in a solenoid with inductance of 3.4 H. To prevent sedimentation of the filler particles in the liquid mixture, container was rotated about its axis directed along the magnetic field lines during whole polymerization period (~ 120 min).

Note that the curing of the epoxy resin (without filler) in the magnetic field (intensity H=6.4*10^5 A/m) leads [4] to changes in its structure at the molecular and su-

Fig.1. Radiographs of sample PCM disks with "fibers" oriented: 1) in the disk plane and 2) normal to disk.

permolecular levels, and, due to the appearance of orderly structures, to anisotropy. The polymerization of the composite in the field with the magnetic induction (B = 200 mT) changes the processes of structure formation in the boundary layers of the polymer/filler particles, which also affects the properties of the compound [5].

Therefore, to establish effect on a heat transfer only of anisotropic spatial distribution of the filler particles in PCM and to eliminate the effects of the magnetic field on the binder properties curing was held in comparatively low field intensity (H=1.2·10^4 A/m). Magnetic field induction (B = 15 mT), selected by solenoid current, was enough to form in still liquid mixture of components extended, concentrated areas ($\varnothing \approx 0.1 \div 0.5$ mm) of powder particles - "fibers", that visualize the magnetic field lines. As a result of mixture polymerization, during its curing in PCM, this structurewas fixed (Figure 1. X-ray image). Due to increased concentration of the filler metal particles such "fibers" are characterized by high thermal and electrical conductivity. Therefore, it is expected that as the thermal conductivity [3], the heat transfer in composite along the direction of "fibers", defining the direction of the PCM orthotropic axis, is also at a maximum, and conversely, along the direction orthogonal to the axis –at a minimum.

To evaluate the effect of orthotropic axis direction on a heat transfer of a material, with respect to the heat flow, samples of 3 types for thermal measurements were cut from cured PCM. They were made in the form of discs (Ø~20 mm and h~2 mm) with "fibers" oriented: 1) in the disk plane and 2) normal to disk (Figure 1). Accordingly, as the heat flux in experiments with disks was directed along the normal to them, in the samples of type 1, it was orthogonal (⊥), and in the type 2 - parallel (||) to orthotropic axis. Samples of type 3 had the same geometry, and were cut isotropic PCM (prepared using same technology, but cured when the solenoid was turned off).

Measurement technique. To determine the TPP of manufactured samples a modified method of "pulsed heat source" was used, which allows to determine in a short time their thermal diffusivity – a, thermal capacity – c_p and thermal conductiv-

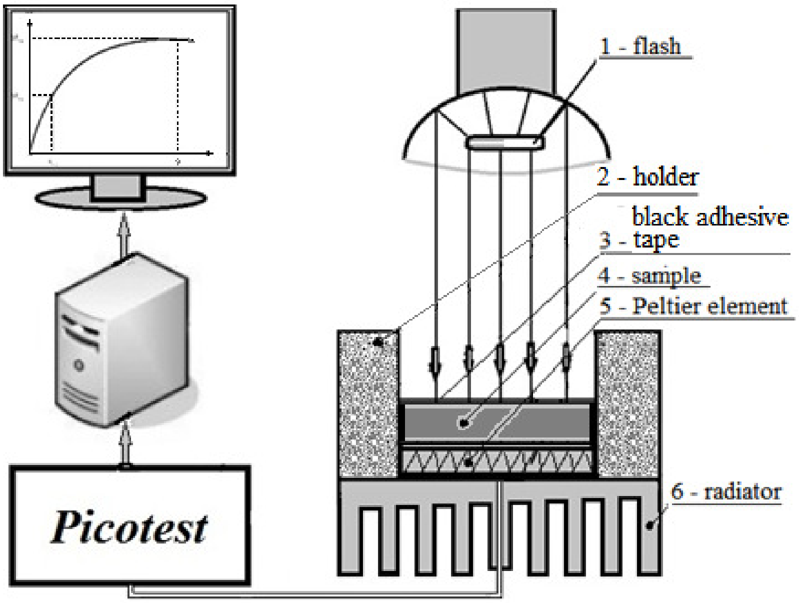

Fig.2. Setup for studying thermophysical properties of the samples - PCM disks - by " pulsed heat source" method.

ity- λ from single experiment. Flash lamps, lasers (with a single pulse or series), and others [6, 7] are used as "instantaneous" heat sources.

As it is known, the theoretical basis of this "non-stationary" method is the solution of the heat conductivity equation for the thermal distribution in the plate (thickness h), when its end ("face") surface is exposed to a short heat pulse.Solution shows that the time $\tau_{1/2}$ (s), during which the temperature ϑ (K) of the "back" surface of the plate(rising during time τ to the maximum – $\vartheta_{\max}$ (h, τ)) reaches half of its maximum value $\vartheta_{\max}/2 = \vartheta(h,\tau_{1/2})$, is associated with h(m) and the thermal diffusivityfactor of the material a(m^2/c) as follows:

$$(\pi^2 a/h^2)\tau_{1/2} = 1.38. \tag{1}$$

In an approved method for determining the type of $\vartheta(\tau)$ dependency, and it - finding temperatures ϑ_{max}, $\vartheta_{max}/2$ and time $\tau_{1/2}$, the signal the thermocouple on the back surface of the plate (thickness h) was fed to a storage oscilloscope with subsequent analysis of $\vartheta(\tau)$ graph on the screen. However, a significant inaccuracy in the determination ϑ_{max}, $\vartheta_{max}/2$ and $\tau_{1/2}$ values from the waveforms affected the measurement reliability. Preliminary results on estimating the influence of anisotropy of PCM on thermal diffusivity were obtained when the oscilloscope was replaced by a sensitive galvanometer (M195/3, $C = 1.5\cdot10^{-9}$ A/div). Current readings were recorded on video, followed by single frame analysis. Upon obtaineddata summary (~50) $\vartheta(\tau)$ dependencies were built and the values of its characteristic quantities were estimated, describing, in accordance with the (1), the thermal diffusivity of the sample. A significant shortcoming of this method is low thermopower (~40 µV/K) of thermocouples, and the difficulty of obtaining massive digital data.

Therefore, in the final version of the test setup and measurement scheme high temperature-sensitive calorimeter: Peltier element (TEC1-12706) was used as a temperature sensor (5, Fig. 2) of the bottom surface of the sample. Sample (4) is a PCM disc located in a holder (2). To ensure in the sample adiabatic heating process, holder is made of foam plastic with outer surface covered with reflective film. Samples with different content (concentration) and the type (orthotropic axis direction) of filler differ from each other with emissivityα. Therefore, to ensure the identity of the thermal pulse absorption black film (3) (adhesive tape, 1 μm, $\alpha = 0.99$) was applied on top of fatless polished disk surface with reliable (due to high adhesion) thermal contact. Lower - back polished surface of the disk coated with a thin layer of lubricant PPMS-4 (TU6-02-017-74), was in contact with the calorimeter, placed on the heat sink (6). YN560 gas discharge lamp (120 J/pulse) (1) served as the source of heat pulse (<1 ms).

Since the lamp flash - the value of Peltier element EMF, which is proportional to the increase of temperature of back (bottom in Figure 1) of the PCM disk for each consecutive short ($\Delta\tau \sim 20$ ms) intervals was recorded with multimeter Picotest M3510A. Formed discrete digital device data massive: voltage ΔU_i – time t_i was sent to the PC, which displayed the desired graph of increase of temperature with time. $\vartheta(\tau)$ graph allowed visual quality control of sample warm-up process. Such measurement technique enabled having ~200-500 ΔU_i values during the time of reaching maximum temperature ($\tau \sim 3 \div 16$ s).

The resulting digital data massive was processed by a special program, the result of which was the determining of desired values: the maximum temperature of "back" surface ϑ_{max}, the time $\tau$to achieve it, as well as the quantities $\vartheta_{max}/2$ and $\tau_{1/2}$. These values were used for calculation thermal diffusivity of samples and their thermal capacity and thermal conductivity.

For the reliability of the results of the samples on the TPP, the same measurements of thermal characteristics were carried out on known materials [8, 9]. For this purpose disks of the same geometry made of ebonite, textolite, polymethylmethacrylateand others were used. The choice of materials was based on the similarity of their thermal properties to the thermal properties of the studied PCM). There were also control experiments with standard samples (from the set to the device ITEM-1M). The relative error in the determination of the measured PCM thermal diffusivity of PCM did not exceed ~ 9%.

The measurement results confirmed [10] that the value of the heat transfer coefficients of the PCM in the direction of the axis orthotropic indeed higher than in the orthogonal direction to it. The influence of the content of nickel powder (its volume concentration) on the temperature diffusivity a (m^2/ c) of different types of samples – disks (different by "fibers" location in them) PCM is shown in Figure 3 with the introduction of polynomial trend lines. These lines reflect the concentration dependencies of the thermal diffusivity of samples of an anisotropic (and isotropic) PCM with the direction of the orthotropic axis in relation to the heat flow: 1, 3 - for the samples in parallel (||) to it, and perpendicular ($\perp$), respectively. Line 2 describes the same dependence measured in the samples of isotropic PCM cured outside the magnetic field.

As can be seen from Fig. 3, with the increase of the content of nickel powder in the composite thermal diffusivity of all 3 types of samples goes up. At the same time

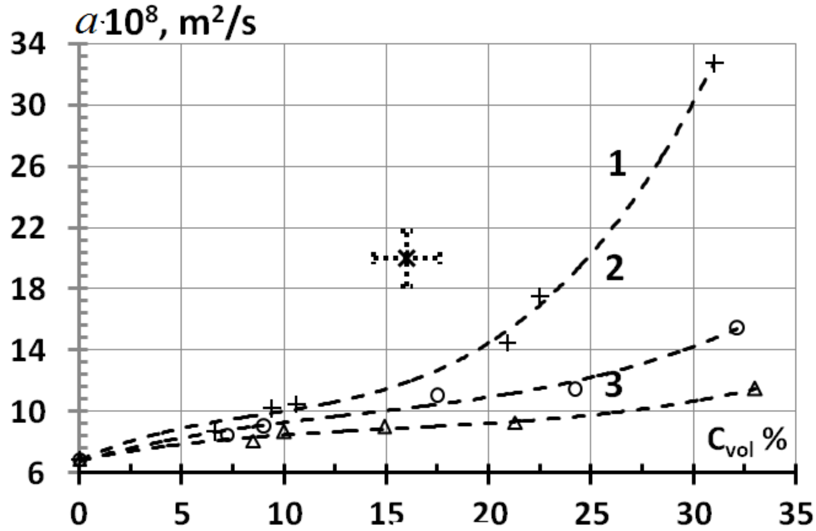

Fig. 3. The dependence of the thermal diffusivity of PCM with different orientation of "fibers" inits samples - drives on the concentration (C_{vol} %) in the EDR-20 nickel powder. Line 2 - sample of the isotropic PCM. The PCM samples of an anisotropic heat flow towards orthotropic axis is directed: 1 - in parallel, 3 - perpendicularly. T = 292-294 K.

the thermal diffusivity in the direction of heat flow ⊥ «fibers» is minimal, for isotropic samples - slightly higher (and by value and nature of the changes with the concentration corresponds to the data [8] for such PCM, and reaches maximum at the flow along the "fibers" - in samples with their || orientation). Thus, when the last coefficient of thermal diffusivity at filler content C_{vol}~ 25 % in comparison with the thermal diffusivity of the material in the direction ⊥ «fibers» ~ is above 2 times higher. Note that this difference increases significantly (electrical conductivity by several orders [3]) when curing the composite in a magnetic field of higher intensity, which is associated with the influence of the field on the structure of the binder and, to a greater extent – on increasing the number of "conductive fibers" and the concentration of metal particles in them. The latter explains that samples "fibers" oriented along the heat flow with increasing of filler concentration, starting at C_{vol}~ 20 %, (see. Fig. 3) a tendency of occurrence of percolation of the values of thermal diffusivity becomes noticeable.

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Температуропроводность полимерного композита, отвержденного в магнитом поле

Аннотация

Отверждение композита в постоянном магнитном поле малой напряженности ($H \sim 10^4$ А/м) приводит к анизотропному пространственному распределению частиц наполнителя – порошка карбонильного никеля. Методом «импульсного источника тепла» проведены измерения коэффициента температуропроводности такого композита с ортотропной структурой. Установлено повышенное значение этого коэффициента в направлении оси ортотропии.

Скоржевський А.В., Алтоіз Б.А., Савін С.Н.

Температуропровідність полімерного композиту, що затверджений у магнітному полі

Анотація

Затвердіння композиту в постійному магнітному полі малої напруженості ($H \sim 10^4$ А/м) призводить до анізотропного просторового розподілу часток наповнювача - порошку карбонільного нікелю. Методом «імпульсного джерела тепла» проведено вимірювання коефіцієнта температуропровідності такого композиту з ортотропной структурою. Встановлено підвищене значення цього коефіцієнта в напрямку осі ортотропії.