THE COMPLEX INVESTIGATION OF SEPARATE STRUCTURAL ELEMENTS OF COMPOSITIONAL MATERIAL

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The investigations of electric conduction of vulcanizing layer of multielectrode electric radiator are carried out by electron diffraction method. It is shown that the black carbon is the main phase of technical one. The average degree of dispersion which is equal to 93, 05% is obtained by investigations and measurements carried out by Leigh-dugmore method on the samples of vulcanizing agent with black carbon in chosen concentrations. The supposed method of regular thermal regime of first kind in plane biocalorimeter with alpha-blocks at cooling of equally heated sample is the closest to the given one in State Standard.

Keywords: compositional materials, optical microscopy, electron microscopy, granulometry, diffraction, rubber and non-vulcanized mixture, technical carbon, microelectronic diffraction pattern.
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INTRODUCTION

The known modern and widely used heating technique, having the disadvantages such as: dielectric constant, stable electric resistance, self-regulation possibility don’t supply the complex of properties necessary for reliable power-efficient functioning. That’s why the technique development of power-efficient surface-distributed electric heating with the given electro-, thermo-physical characteristics in conjunction with the work in the self-regulation regime and functioning possibility in wet and corrosive mediums allows us to solve the problem presenting the important scientific-technical value.

The multielectrode compositional electric radiator (MCER) is supposed in the aim of technique development of power-efficient surface-distributed electric heating with the given electro-, thermo-physical characteristics in the self-regulation regime.

The composites are the heterogeneous solid multicomponent materials consisting in two or more components among which we can emphasize the reinforcing elements supplying the necessary mechanical characteristics (high strength, hardness) of material and matrix which is plastic base supplying the joint work of reinforcing elements.

The electroconductive layer of MCER presents itself the dispersion-filled polymer butyl-caoutchouc matrix with nano-structural filler in the form of technical carbon. The technical carbon supplies the stable temperature on the surface of electroconductive layer without the possibility of its regulation in the dependence of surrounding conditions. The stearin is the plasticizing agent and the additional functional ingredients serve in the capacity of the filler.

The butyl-caoutchouc is the product of low-temperature copolymerization of isobutenyl and 1-5% of any diene, mainly isoprene.

The technical carbon (carbon-black), non-active mineral fillers (chalk, kaolin and etc), high-disperse SiO_2, their mixtures (50 – 70 mass particles on 100 caoutchouc mass particles) serve as fillers of rubber compounds on the base of butyl-caoutchouc. Only saturated compounds (for example, naphthenic and paraffinic oils, low-molecular polyethylene) are used in the capacity of plasticizers, as the non-saturated plasticizers decelerate the caoutchouc vulcanization.

The technical carbon is the high-disperse amorphous carbonic product produced in industrial scale. The technical carbon particles present themselves the globules consisting in degraded graphite structures. The interplanar spacing between graphite-like layers is 0,35-0,365nm (in comparison in graphite it is 0,335 nm). The particle size (13 – 120nm) defines the technical carbon “dispersion”.

The specific surface is the physic-chemical index characterizing the dispersion. The particle surface has the roughness because of the layers overlapping each other.

The technical carbon is applied in the capacity of the reinforcing component in production of rubbers and plastic masses. About 70% whole technical carbon is used in tyre production, about 20% of technical carbon is used in production of rubber-technical products.

EXPERIMENT TECHNIQUE

The composition electric radiator (CER) calculation model consists in long plate put in rectangular isolating material and body with plane surface. The following boundary conditions: the surface between the object of heat removal and isolating cover is isothermal one, surface between the isolating cover and air is impermeable one (adiabatic) for heat flux lines [1,2] are chosen at CER model choice.

The construction of MCER is shown in Fig.1.

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The given construction of MCER allows us to regulate the electric energy consumption on local heating in the dependence on heat exchange.

The directed projecting of resistive layer structure can supply MCER operation with both positive and negative temperature coefficients in the dependence on fields of application.

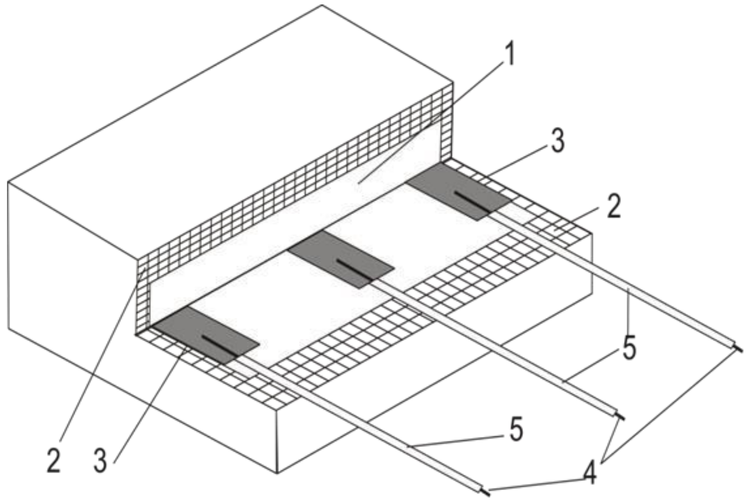

Fig.1. Multielectrode composition electric radiator.
1 is fuel electroconductive layer; 2 are isolating layers; 3 is system of electrodes from metal gauze (copper or brass); 4 are flexible shunts; 5 is isolating membrane of shunts.

The additional function is necessary to introduce into two-dimensional Laplace equation for successful decision of theoretical estimation of complex state of electric and heating parameters of CER. This function corresponding to two-dimensional Laplace equation and boundary conditions of first kind, expresses the angle value formed by strength vector of plane-parallel field with the one of the axis of Cartesian coordinate system which is harmonic one.

The calculation of MCERs is carried out by the scheme which is given in Fig.2.

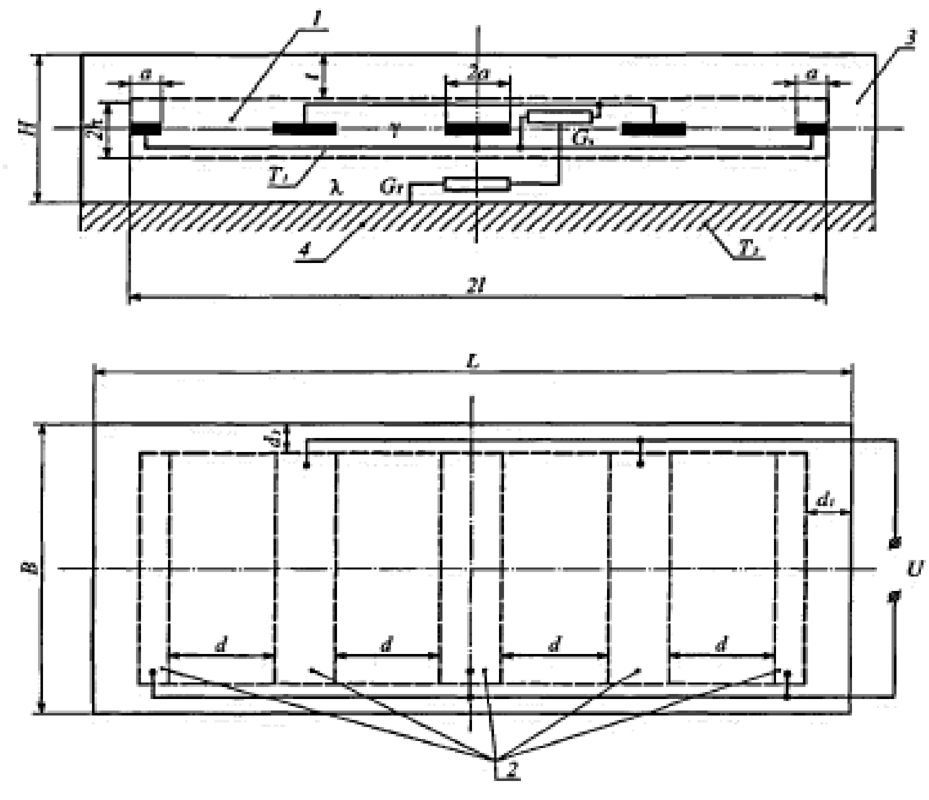

Fig.2. The scheme of 4-electrode plate electric radiator.
1 is resistive material; 2 are electrodes; 3 is isolating layer; 4 is surface of heating recoil.

EXPERIMENTAL PART

The system with concrete boundary conditions is chosen for calculation of electric conduction of multielectrode systems with definite parameters in Fig.2. On the base of this system, the calculative model is chosen so that the system requirements allow us to calculate their parameters with necessary delicacy.

For realization of self-regulation mode the electroconductive layer of MCER should have the negative temperature coefficient of resistance αρ that corresponds to decrease of specific bulk resistance of electroconductive layer ρ_v with temperature increase on MCER surface. The character of obtained dependences ρ_v=f (T) after the output in the operation regime evidences about the negative temperature coefficient corresponding to polymer semiconductor materials. This circumstance makes MCER operation possible in power-efficient self-regulation regime.

According to this, MCER made from composition material where the obligatory base of which is butyl-caoutchouc, the filler is technical carbon, stearin is plasticizing agent and additional functional ingredients are supposed.

On the first stage the electroconductive layers are vulcanized at temperatures 172-174^{0}C and under the pressure 12-13 MPa during 0,5 – 1 minutes. On the second stage they are vulcanized at temperatures 165-167^{0}C under the pressure 11-11.5 MPa during 35 minutes.

The dependence graph of MCER layer sample electric conduction on temperature is shown in Fig.3.

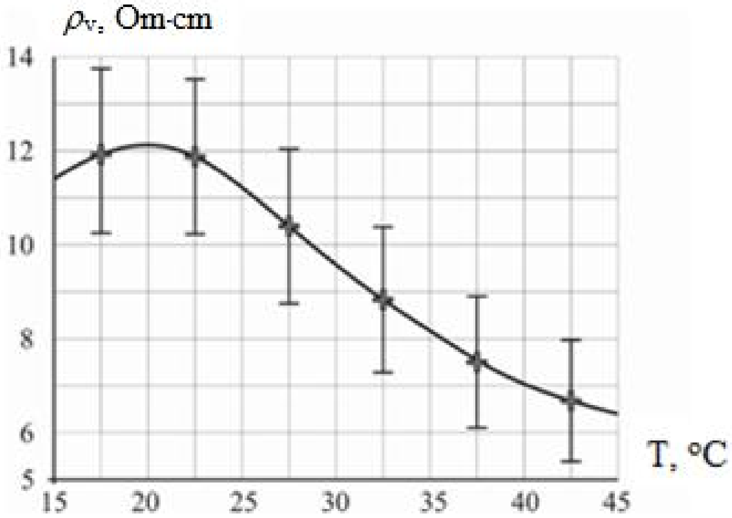

Fig.3. The dependence of electric conduction ρ_v of MCER sample layers on temperature.

The series from 5 MCERs is investigated with the aim of the analysis of the dependence of electric conduction ρ_v of MCER sample layers on temperature (T) in order to obtain the correspondence with the supposed method.

The measurements are carried out at temperature of environment 18 - 20 °C. The sample is put on the wood base at voltage 220v and 50hz. The current strength, voltage and internal temperature of electric radiator are measured each 5 minutes during first hour and each 10 minutes during the second hour.

The investigations are carried out by the means of comprehensive microscopic methods in the different large-scale levels including the scanning microscopy and microscopy of transmission electron, beginning from dimensions of black particles of technical carbon 29 ÷ 42 nm which are used for the series of MCER production.

Using the electron diffraction method, the investigations of electric conduction of MCER vulcanizing layer with the following carbon particles: N-245, N-330, having the diameter 29-32nm are carried out. The investigations are carried out on transmission microscope EM - 125 sec with increasing up to 125kVt.

The analysis carried out by electron diffractometer shows that the main phase is black carbon taken in the correspondence with exact copy method.

The crystallite sizes are 20÷25 nm. The electron diffractometer gives the evidence to carbon black particles of typical nano-crystalline structure.

The typical image of hexagonal crystal lattice is shown by electron microscope in Fig.4.(a). The carbon black particles are shown by arrows in Fig.4.(a). The image of carbon black particles is shown by electron diffractometer in Fig.4.(b).

The qualitative estimation of dispersion and homogeneity of filler nano-particle distribution in polymer material is carried out by Leigh-dugmore method. The dispersion average degree 93,05% is obtained by investigations and measurements carried out by above mentioned method on 10 samples of vulcanizing butyl caoutchouc (BK) agent - 1675 with black carbon N- 245 in chosen concentrations.

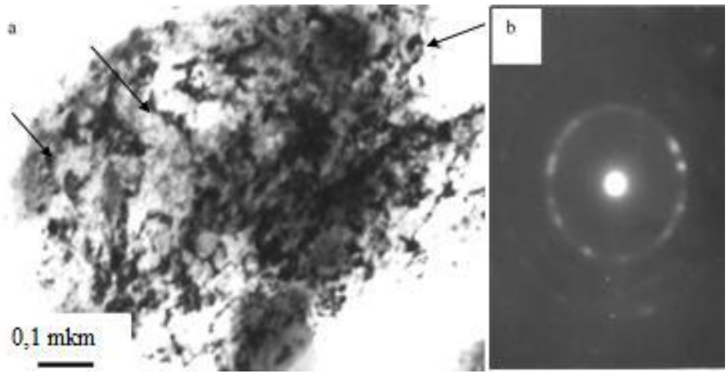

Fig.4. The sample image on electron microscope (a) and electron diffractometer (b) showing the hexagonal lattice of carbon black particles.

The purposeful change in composition of radiator layer electric conduction and its production process parameters allows us to change MCER resistance coefficient αρ in the dependence on temperature allowing to radiator to operate in self-regulating regime.

It is necessary to know the main material thermo-physical characteristics towards with the material electro-physical ones at construction and production of MCERs.

The main thermo-physical characteristics of material are the thermal capacity, heat conductivity and thermal conductivity. As the materials present themselves the filled polymers used at the MCER production, so State Standard 23630.1 – 79 "Plastics. The methods of specific heat" [3] should be applied to obtain their thermo-physical characteristics.

The three main thermo-physical characteristics of the substances are connected with the following dependence:

$$a = \lambda / \rho \cdot c \qquad (1)$$

where: *a* is thermal conductivity coefficient m²/c; λ is heat conductivity coefficient Vt/(m·°C); ρ is matter density kg/m³; *c* is specific heat capacity J/(kg·°C).

The definition of heat conductivity by [5] is carried out by the method of regular heat regime on cylindrical biocalorimeter based on heat regularities of metallic cylinder (nuclear) through the investigated matter layer.

The temperature change character in the space and time is established on the base of the first law of thermodynamics and Bio-Fourier law, which is expressed by heat conductivity differential equation [4]:

$$\frac{\partial T}{\partial \tau} = \frac{\lambda}{c\rho}\left(\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2}\right), \qquad (2)$$

where: *c* is specific heat capacity J/(kg·°C); ρ is density kg/m³; *T* is temperature ^{0}C; τ is time sec.

The definition of heat conductivity of the investigated materials is carried out in plane biocalorimeter with alpha-blocks [5]. The possibility of the investigation of the whole class of solid and bulk materials and also the fact that the thermostat becomes unusefull because of the big water discharge through cooling channels of alpha-blocks are the advantages of this method.

The samples for investigations present themselves the discs by diameter (142±5) mm and thickness (10±0,5) mm.

The thermal resistance of R_c, (m²·h·°C)/kcal sample is determined in the correspondence with technique [6] given to concrete values of core thermal capacity and biocalorimeter material, its geometric dimensions by the following formula:

$$R_c = \left(\frac{Y}{\Phi} - 5 \cdot 10^{-3}\right). \qquad (3)$$

where: *Φ* is the complex characterizing the radiator core heat exchange with sample surface in contact place , kcal/m²·h; *Y* is the parameter taking under consideration the rate of cooling.

Φ complex is calculated by the following formula:

$$\Phi = \Phi_0 \left[0,4 + \frac{0,6}{1 + 2\frac{\delta}{D}}\right]. \qquad (4)$$

where Φ_0 is biocalorimeter constant which is equal to 3,4 kcal/m² ·h;

D and δ are diameter and thickness of the sample m, correspondingly.

Y parameter is defined by the following formula:

$$Y = \frac{1}{\frac{m}{B} - A}. \qquad (5)$$

where: A is device constant which is equal to 0,32;

B is value depending on form factor and total nuclear heat capacity and the layer of the investigated matter.

B value (m²·°C)/kcal is defined by the following formula:

$$B = \frac{1}{1 + 0,1\delta \cdot c \cdot \rho}. \qquad (6)$$

where: δ is the thickness of the investigated sample;

ρ is density of the sample material;

c is specific heat capacity of the sample material;

The heat capacity is defined with accuracy in the correspondence with [7]. The sample density is defined by the displaced water volume with the correction for its temperature.

The most character experimental dependences of temperature pressures on time for the sample groups from the materials of isolating production rubber source (PRS) and developed production rubber electroconductive (PRE) layers in different time moments are given in Fig.5.

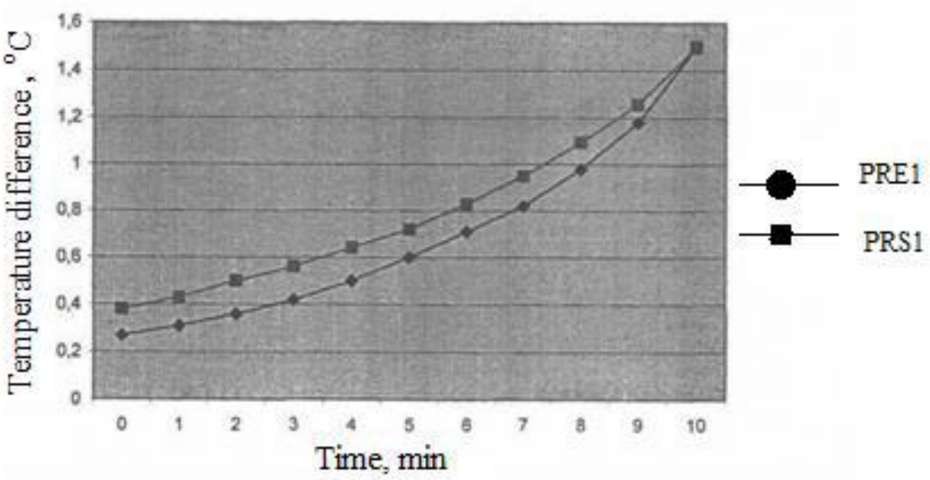

Fig.5. The experimental dependences of the temperature pressures PRS and PRE on time.

Thus, the supposed method of regular regime of the first kind in plane biocalorimeter with alpha-blocks at the cooling of uniformly heated sample is the closest to the given one in State Standard. The average temperature on electric radiator surface after its output on operating regime is obtained with the help of the software including the infrared imager complex and it is 39,1°C for MCER with the inner diameter 24mm at measuring inaccuracy ±0,1 °C.

The temperature inclination on the surface of electric radiator on average temperature defined on the base of measurements in the points uniformly distributed on the surface is 7,5% for MCER.

The calculations are carried out for MCER with resistive material on the base of technical carbon П-234 with concentration 52,5 mass particles on 100 mass particles of polymer. The obtained results allow us to estimate the thermal conductivity coefficient and specific heat capacity of developed composition materials and use the obtained data for the theoretical calculations.

CONCLUSION

- Using the electron diffraction method the investigations of electric conductivity of vulcanizing layer of MCER with the following carbon particles: N- 245, N- 330 having the diameter 29-32 nm are carried out;

- The diffraction method shows that the main phase is the black carbon taken in the correspondence with method of exact copy. Mainly the carbon black particles have the hexagonal lattice structure, which have the nano-crystalline structure;

- The dispersion quantitative estimation and homogeneity of filler nano-particle distribution in polymer material are carried out by the Leigh-dugmore method. The dispersion average degree 93,05% is obtained by investigations and measurements caused by above mentioned method on 10 samples of vulcanizing BK agent - 1675 with black carbon N-245 in chosen concentrations.

- The definition of the material thermal conduction is carried out by the method of regular heat regime on cylinder biocalorimeter based on the regularities of heating of metallic cylinder (nuclear) through the investigated matter layer. The supposed method of regular heat regime of the first kind in the plane biocalorimeter with alpha-blocks at the cooling of uniformly heated sample is the closest to the given one in State Standard. The average temperature on the electric radiator surface after its output on operating regime is defined with the help of software including the infrared imager complex and it is 39,1°C for MCER with the inner diameter 24mm at measuring inaccuracy ±0,1 °C.

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