VOL. 9, NO. 11, NOVEMBER 2014 ISSN 1819-6608
ARPN Journal of Engineering and Applied Sciences

www.arpnjournals.com

CEMENT-QUARTZ ELECTRICALLY CONDUCTIVE COMPOSITES BASED ON GRAPHITE DISPERSIONS

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ABSTRACT

Specific electric conductivity of composite materials based on graphite dispersions was studied. The aim of the work is to optimize electrical properties of composites. It was found that with increase of graphite mass fraction from 0.06 to 0.2 electrical conductivity of the system cement-quartz-graphite increases from 0.85 to 13.11 $\text{ohm}^{-1}\cdot\text{cm}^{-1}$. Temperature dependences of the model systems cement-graphite, quartz-graphite, cement-quartz-graphite on conductive phase mass fraction were analyzed. To obtain effective compositions with stable electrical characteristics threshold concentration of a conductive component for cement-quartz composites equal to 0.06 was determined; any excess of this value leads to a great increase in the electrical conductivity due to formation of continuous chain structures. Temperature dependence of an electric conductivity logarithm in the model system cement-quartz-graphite is described by rising curves. Activation energies of electrical conductivity in the model systems cement-graphite, quartz-graphite, cement-quartz-graphite were calculated at different mass fraction of graphite.

Keywords: conductivity, activation energy, specific electrical conductivity.

INTRODUCTION

Intensive development of the construction industry requires the use of effective composite materials with high rates of construction and technology, functional properties and durability. In modern construction technologies composite conductive materials [1] which are complex multicomponent disperse systems are widely used. The main problem of these composites technology is the control of the process of structure formation and assurance of a uniform conductive phase distribution over the volume in order to ensure stable electro-physical characteristics of the material. A relevant method of conductive dispersed systems formation is the method of cold dry pressing [2].

Fine metal powders [3], metal oxides, graphitized carbon materials are used as an electrically conductive phase in composite materials. Depending on the distribution of the conductive component in the system its electrical, physical, mechanical and thermal properties are changed [4]. Stability of these characteristics is the main criterion of conductive additives suitability. Dispersed conductive components along with chemical inertness should have high thermophysical parameters with sufficient mechanical strength [5]. In this paper the use of graphite as a conductive component is based upon its high physical-chemical and performance characteristics: inactivity; high temperature of the start of oxidation, thermal and electrical conductivity; fire-resistance; environmental friendliness, etc.

A complicated complex of requirements applicable to composite materials with conductive properties is fulfilled by choosing and optimizing characteristics of the materials used. In order to select effective compositions of functional conductive materials the condition of the modern market of building composites was analyzed. The carried out patent search showed that conductive systems based on various forms of carbon and

Portland cement are widely used in modern civil and industrial construction, power industry and agriculture. It was revealed that researchers from different countries place special emphasis on the development of a technology of low-temperature heating systems production.

METHODOLOGY

Samples were formed using a laboratory hydraulic press at a pressure of 5 MPa in order to investigate the physical and mechanical characteristics that reflect structural properties of systems based on graphite dispersions. For the purpose of the experiments sets of cylindrical samples were produced in a purpose made cylindrical die. Temperature dependences of electrical conductivity of graphitized carbon composite materials were investigated. Portland cement CEM I 42, 5N manufactured by JSC Belgorodskiy cement was used as the cement matrix. Sand of Nizhne-Olshanskoye field was used as filler in the composition under test, and graphite of GL-1 brand of Zaval'evsky field was used as a conductive phase.

Using the cylindrical samples the effect of conductive phase concentration on electrical properties of pressed composites was analyzed. Measurements of electrical conductivity were carried out using the following procedure. Constant potential difference of 100 V was applied to end face parts of the sample, current was measured using a micro-ammeter M 2018 (volt-ampere line was plotted), resistance was calculated and specific electrical conductivity was calculated according to geometrical parameters of the samples [6, 7, 8]. The activation energy of the electrical conductivity E was determined according to the slope of the line passing through the experimental points. Approximation of the experimental results using the least-square method gives a linear dependence. The correlation coefficient is 0.91-0.98. To calculate E according to the experimental data the integral form of the equation of Van't- Hoff-Arrhenius was used.

RESULTS AND DISCUSSIONS

After holding of the samples for one month their specific electrical conductivity was determined by way of measuring of samples voltage and current passing through the sample. The measurement results are shown in Table-1.

Table-1. Specific electrical conductivity of graphite dispersions in model systems.

Graphite content, w	Model systems; specific electrical conductivity Ohm^{-1}·m^{-1}		
	Cement-graphite	Quartz- graphite	Cement-quartz-graphite
0,06	0,06±0,12	1,12±0,027	0,85±0,049
0,1	0,87±0,085	6,92±0,089	6,56±0,099
0,15	4,95±0,092	9,95±0,097	8,57±0,043
0,2	6,37±0,097	13,27±0,053	13,11±0,081

Mathematical treatment of the results of measurements was carried out as follows. Thus, when the content of graphite was 0.06 (w), values of specific electrical conductivity were: 0.06; 0.08; 0.04 Ohm^{-1} • m^{-1}. The mean value was 0.06 ohm^{-1} • min^{-1}. The dispersion S^2 and the standard deviation *S* were determined:

$$S^2 = \frac{1}{N-1}\sum\left(x-\bar{x}\right)^2 = \frac{1}{4-1}\left(0+4\cdot10^4+4\cdot10^4\right)=2{,}7\cdot10^4; S=1{,}63 10^2$$

With the reliability of 0.99 at degrees of freedom equal to two, Student's coefficient is 9.925, so the expectation value for the specific electrical conductivity is within the range of ± 0.12:

$$\bar{\sigma}-\frac{S\cdot t_\alpha}{\sqrt{N}} \le \sigma_x \le \bar{\sigma}+\frac{S\cdot t_\alpha}{\sqrt{N}}; \frac{S\cdot t_\alpha}{\sqrt{N}} = \frac{0{,}02\cdot 9{,}925}{\sqrt{3}} = 0{,}12$$

The specified confidence interval value was the highest (the lowest accuracy of measurements) for this run of measurements, Table-1, so the subsequent results were evaluated for the given confidence interval, despite the fact that the other runs of measurements were carried out with a higher accuracy.

To obtain effective models of electrically conductive composites with stable electrical characteristics threshold concentration of the conductive component was defined; any excess of this value leads to a great increase in the electrical conductivity due to formation of continuous chain structures - current lines. [7, 8].

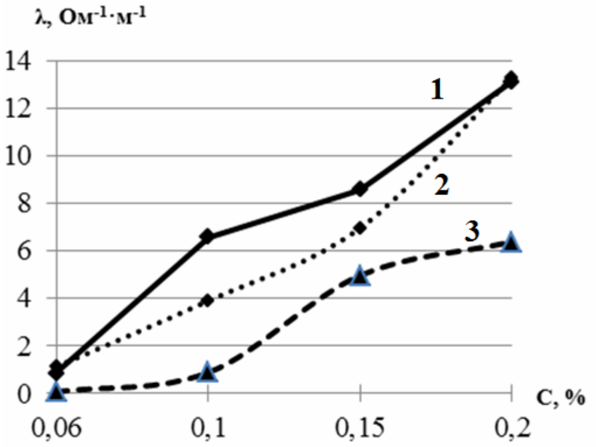

Figure-1. Dependence of the electrical conductivity on the mass fraction of the conductive phase for the following model systems: 1 - cement-graphite; 2 - graphite-quartz; 3 - cement-quartz-graphite, T = 293 K.

The obtained results indicate that conductive phase concentration threshold in the composites under test is 0.06% (Figure-1). The electrical conductivity in the range of 0, 85-13, 11 ohm^{-1}•cm^{-1} is observed in the model system "cement-quartz-graphite". As can be seen from Figure-2 the temperature dependence of the specific conductivity logarithm in the model system cement-quartz-graphite is described by rising curves.

Based on the data obtained a diagram of dependence of the specific conductivity λ on the inverse temperature 1/T was plotted in a semi-logarithmic scale. It was found that with increase of mass fraction of the conductive component from 0.06% to 0.2%, the curves retain their behavior and shift to higher values of electrical conductivity.

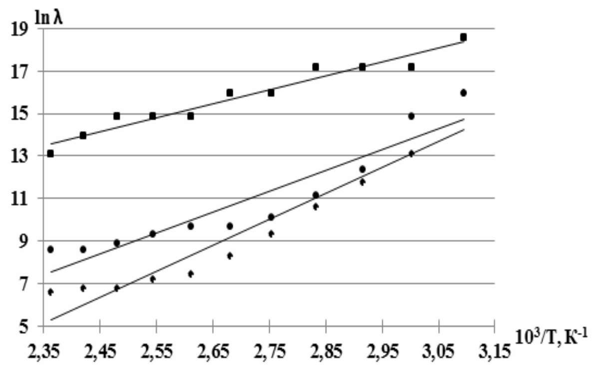

Figure-2. Temperature dependence of the electric conductivity logarithm of the model system cement-quartz with different mass concentration of graphite: 1- 0.1%; 2 - 0, 15%; 3 - 0.2%.

Studies of the structure of composites carried out using the microscopy method confirm presence of graphite chain structures, which conduct electric current (Figure-3).

With an increase of concentration of the conductive phase, a change of the electrical conductivity activation energy due to an increase of the number of contacts between particles is observed.

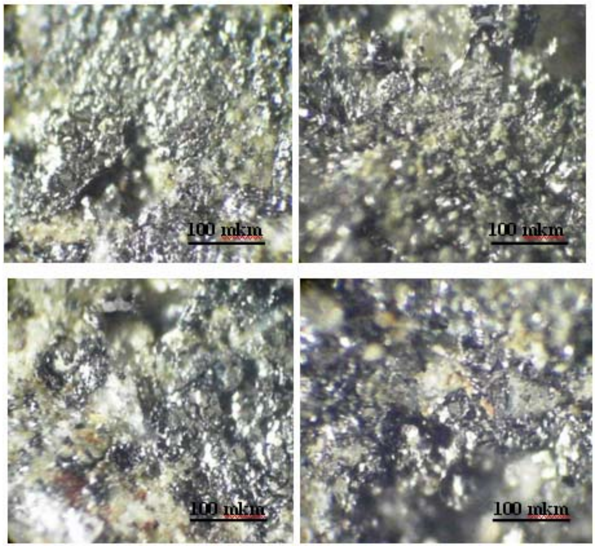

Figure-3. Photomicrographs of graphite particles in the system "cement-quartz" at their content: 1 - 0.05%; 2 - 0.1%; 3 - 0.15%; 4 - 0.2%.

There is a maximum of the electrical conductivity activation energy at the mass fraction of the conductive component of 0.1% (see Figure-4) on the curve of the samples "cement-quartz-graphite", and in the range of 0.1 to 0.2% a reduction of the electric conductivity activation energy from 20.48 to 6.91 kJ/mol takes place.

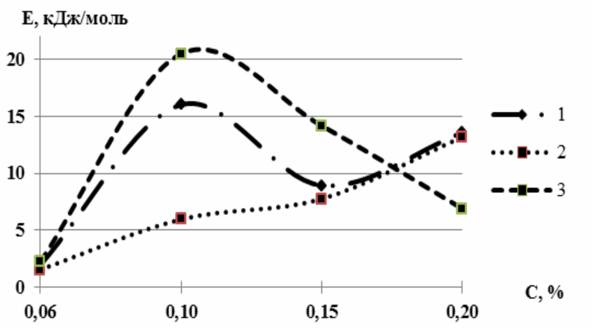

Figure-4. Dependence of the electrical conductivity activation energy on the mass fraction of the conductive phase for the following model systems: 1- cement-graphite; 2-graphite-quartz; 3- cement-quartz-graphite, T = 293 K.

A decrease of volume resistivity is observed in the studied model systems after reaching of a certain temperature; this fact indicates a negative temperature coefficient inherent in dispersion-filled semiconductor materials [9]. The optimum mass fraction of graphite filler is in the range of up to 20%; it is impractical to increase this value due to a slight change of the electrical characteristics of the composite. At graphite dispersion phase concentrations of less than 0.06 (w) an aggregation of particles in the cement-quartz composite takes place. The method of dry-pressing used in the work provides optimum distribution of the conductive component [10] and the degree of compaction of the samples. This technology of composite mixture forming allows reducing capital costs by reducing the conductive phase concentration while maintaining the quality characteristics of products. Use of innovative low-temperature composites and heating systems based on them allows the mass adoption of energy saving technologies in modern systems of radiant panel heating.

CONCLUSIONS

The results of the studies allow concluding that it is effective to use pressed construction composites based on graphite dispersions as conductive composite materials. It is found that the threshold concentration of the conductive phase of cement-quartz composites is 0.06%. A change of the electric conductivity activation energy is caused by an increase of the number of contacts between particles.

Temperature dependences of the model systems cement-graphite, quartz-graphite, cement-quartz-graphite on the mass fraction of the conductive phase were studied. The temperature dependence of the logarithm of the specific conductivity in the model system cement-quartz-graphite is described by rising curves. The electrical conductivity activation energies in the model systems cement-graphite, quartz-graphite, cement-quartz-graphite at different mass fraction of graphite were calculated. Electrical parameters of a material allow producing heating elements operating at given temperature specifications [5, 9]. It is planned to define the regularities of formation of structure and properties of pressed composites depending on formulation and technology factors. The development of a technology of low-temperature heating systems based on graphite dispersions is of vital importance.

ACKNOWLEDGEMENTS

The research was made within the framework of the grant No. B-26/12 dtd. 10.04.2014, which is enforced since April 10, 2014 till December 31, 2014, in the framework of implementing of the Strategic Development Program of BSTU named after V.G. Shoukhov for 2012-2016 on the subject “Development of functional conductive construction composites based on silicates and various forms of carbon for low-temperature electric heating systems”.

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