

Influence of the type of filler distribution on the electrical and thermal conductivity of metal-filled polymer composite

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Received March 4, 2020

The electrical and thermal conductivity of the composite based on low density polyethylene filled with dispersed copper are investigated in a wide concentration range. The composite with random (*R*) and segregated (*S*) types of the filler distribution are studied. The electrical conductivity of the composites demonstrates the percolation behavior, with the value of the percolation threshold equals to 23.0 vol.% and 3.3 vol.% for the *R*-type and *S*-type, respectively. A segregated system is characterized by higher conductivity due to the high local filler concentration in the wall of framework. The concentration dependences of the density and thermal conductivity for the segregated system show a similar behavior: they increase to the value of the packing factor, followed by reaching a plateau. The thermal conductivity of the *R*-type composite was described using the Lichtenecker model and the value of thermal conductivity parameter of the dispersed phase $\lambda_f = 69$ W/(mK) in the composite was found. The *S*-type composite is characterized by higher thermal conductivity than the *R*-type composite at the same filler concentration.

Keywords: metal-polymer composites, electrical conductivity, thermal conductivity.

Вплив типу розподілу металевого наповнювача на електропровідність та теплопровідність металонаповненого полімерного композиту. *А.І.Місюра, О.В.Маруженко, Є.П.Мамуня, М.П.Куліш, А.М.Пилипенко.*

Досліджено електропровідність та теплопровідність композиту на основі поліетилену низької густини наповненого дисперсною міддю, у широкому інтервалі концентрацій. Розглянуто композит з випадковим (*R*) та сегрегованим (*S*) типами розподілу наповнювача. Електропровідність композитів має перколяційний характер зі значенням порогу перколяції 23.0 об.% та 3.3 об.% для *R*-типу та *S*-типу, відповідно. Сегрегована система характеризується вищим значенням електропровідності за рахунок високої локальної концентрації наповнювача у стінці каркасу, утвореного частинками наповнювача. Концентраційні залежності густини та теплопровідності для сегрегованої системи мають схожу поведінку, а саме, зростання до межі наповнення композиту з подальшим виходом на плато. Теплопровідність композиту *R*-типу описано за допомогою моделі Ліхтенекера та знайдено значення параметру теплопровідності дисперсної фази композиту $\lambda_f = 69$ Вт/(мК). Композит *S*-типу характеризується вищими значеннями теплопровідності, ніж композит *R*-типу при однакових концентраціях міді.

Исследованы электропроводность и теплопроводность композита на основе полиэтилена низкой плотности, наполненного дисперсной медью в широком интервале концентраций. Рассмотрены композиты со случайным (*R*) и сегрегированным (*S*) типами распределения наполнителя. Электропроводность композитов имеет перколяционный характер со значением порога перколяции 23.0 об.% и 3.3 об.% для *R*-типа и *S*-типа, соответственно. Сегрегированная система характеризуется более высоким значением электропроводности за счет высокой локальной концентрации наполнителя в стенке каркаса, образованного частицами наполнителя. Концентрационные зависимости плотности и теплопроводности для сегрегированной системы имеет схожее поведение, а именно, рост до предела наполнения композита с последующим выходом на плато. Теплопроводность композита *R*-типа описана с помощью модели Лихтенекера, найдено значение параметра теплопроводности дисперсной фазы композита $\lambda_f = 69$ Вт/(мК). Композит *S*-типа характеризуется более высокими значениями теплопроводности, чем композит *R*-типа при одинаковых концентрациях меди.

1. Introduction

Polymer composites are one of the advanced materials, because their properties can be controlled and modified depending on the conditions of further applying. One of the leading areas of research is the metal-filled composites, whose scope is gradually expanding [1–3]. The main application of such materials are electronics [2], where special attention is paid to new types of electrodes [4], supercapacitors [5], sensors [6] and shielding materials [7]. Polymer composites are used as biomaterials [8], in particular in prosthetics [9] and for the manufacturing of medical instruments [10]. Also, composite materials are used in the auto- [11, 12] and aerospace [13] industry.

Dispersed metals are widely used as a filler for polymer composites. They are available in a wide range of particle sizes and shapes, and easily combined with polymers. Metal-filled polymer composites are less expensive, lighter, have better corrosion resistance and typically require simpler processing than products based on solid metals. The main problem of metal-filled polymer composites is that in order to achieve high conductivity values, these materials require a considerable amount of filler within polymer matrix, which adversely affects their processing parameters.

For polymer composites with a random type of metal filler distribution, the percolation threshold can be obtained at rather high filler content. The data of [14] demonstrate that the percolation threshold (φ_c) for a polymer composite with a random filler distribution is 21 vol.%, 24 vol.%, and 29 vol.% for PE–Fe, POM–Fe and PA–Fe composites, respectively. For composites based on metal fillers such as cobalt (Co) and nickel (Ni) embedded in epoxy resin (ER), silicone (Si) and polyurethane (PU),

the percolation thresholds were: $\varphi_c = 19$ vol.% in the ER–Co composite, $\varphi_c = 35$ vol.% in the ER–Ni composite, $\varphi_c = 27$ vol.% in the Si–Co composite, $\varphi_c = 17$ vol.% in the Si–Ni composite and $\varphi_c = 22$ vol.% in the PU–Co composite [15]. High percolation thresholds were also found in low density polyethylene (LDPE)–Cu composite, where $\varphi_c = 18.7$ vol.% [16]. For the composite PA6–Zn $\varphi_c = 18$ vol.% [17]. The composites based on HDPE and different metals have a high φ_c value as well: in HDPE–Al — 30 vol.%, HDPE–Fe — 40 vol.%, HDPE–Cu — 50 vol.% [18], as well as in HDPE–Al — 55 vol.% and HDPE–Ag — 45 vol.% [19].

One of methods to reduce the value of the percolation threshold in the composite is to change the type of filler distribution in the polymer matrix. One option is creation of the segregated systems [20–22], which is possible when the filler particles are smaller than the polymer particles. First, a mechanical mixture of two components should be obtained, in which small metal particles cover the surface of the polymer particles; next, the samples are formed by hot compacting. In this case the filler is localized at the interface between the polymer particles and creates the conductive framework in the polymer matrix [23].

For polymer composites with a segregated filler distribution, the value of the percolation threshold is lower than for a random filler distribution. For example, in the PE–Cu composite, the value of the percolation threshold is 10 vol.% [24]. In the PMMA–ITO composite, $\varphi_c = 3$ wt.% [25], and the PMMA–Al system has the percolation threshold of 10 vol.% [26]. For the system PMMA–Cu with a filler size 3.3–4.7 μm , $\varphi_c = 9.28$ vol.%, and for particles size 78 nm, $\varphi_c = 13.95$ vol.% [27].

The introduction of a metal filler in the polymer matrix also affects the thermal conductivity of the composite. In [28], the thermal conductivity of the PE–Ag composite with a random filler distribution was investigated. In the region up to 15 vol.%, the results are in a good agreement with the Lichtenecker and Bruggeman models, at $\varphi > 15$ vol.% only the Lichtenecker model correctly describe the experimental data. The thermal conductivity (λ) of the composite with 22 vol. % of silver reaches as much as 1.5 W/(mK). The authors of [29] investigated the PP–Cu composite with particles of different sizes, 30 μm and 300 μm . The results show that the value of thermal conductivity is higher for the composites with a smaller filler particle size, namely $\lambda = 2.25$ W/(mK) versus 1.7 W/(mK) for bigger particles. In [30] it was shown that the thermal conductivity of the HDPE–Al composite increases with decreasing filler particles size.

The nature of the filler also affects the value of thermal conductivity. The value of λ of the composites is 0.9 W/(mK), 0.85 W/(mK), 0.77 W/(mK) and 1.0 W/(mK) at a filler content of 20 vol.% for HDPE filled with Zn, Cu, Fe and Cu/Sn, respectively [31]. In a segregated system based on UHMWPE filled with a hybrid filler BN–AlN (6:1), the value of $\lambda = 7.1$ W/(mK) was reached, at $\varphi = 50$ wt.%. This value is higher than for composites with one type of filler, λ (UHMWPE–BN) = 4.6 W/(mK) and λ (UHMWPE–AlN) = 1.16 W/(mK) [32]. In [33], two types of the segregated PS–Cu composites were investigated: (a) with polystyrene spheres covered by a thin layer of Cu; (b) PS–Cu composites with copper powder. The thermal conductivity was 26.14 W/(mK) for type (a) and 0.45 W/(mK) for type (b) of the composites at 23 vol.% Cu.

As can be seen from the observed articles, the introduction of a metal particles into the polymer matrix affects the electrical and thermal conductivity of the composites. The aim of this work is to investigate the effect of the type of filler distribution (random or segregated) on the electrical and thermal conductivity of polyethylene-based composites, filled with Cu, in a wide range of copper concentrations from 0 to 100 vol.%.

2. Experimental

The polymer composites were produced on the base of low density polyethylene (type 168-03-070) in the powder form with

a melting temperature $T = 112^\circ\text{C}$ and a melt flow rate of 7.5 g/10 min. As a filler, dispersed copper, grade PMS-1, dendritic form, with a particle size of 35–50 μm (DSTU 4960-75) was used. Two types of samples were manufactured: 1) composites with random filler distribution (*R*-type); 2) composites with segregated filler structure (*S*-type).

The polymer composites with random filler distribution (*R*-type) were manufactured using a single screw laboratory extruder ($D = 17$ mm and $L/D = 20$), into which a mixture of polymer and copper powders was loaded. For better dispersion of the filler, the mixture was extruded 2 times. The obtained strands were ground to a size of 0.5–1.0 mm, and then pressed at a temperature of 130°C and a pressure of 40 MPa in a closed steel mold. Samples had a shape of discs 30 mm in diameter and ~ 2 mm thick. The preparation of the composites with a filler concentration of more than 30 vol.% Cu was not possible by the extrusion method because of a significant increase in the viscosity of the polymer/filler mixture in the melt state.

For the preparation of the segregated composites (*S*-type), the mixture of polymer powder and filler was first mechanically mixed and then hot-pressed under the conditions described above. The concentration interval for the *R*-type composite is 0–30 vol.% Cu, and for *S*-type one is 0–100 vol.% Cu.

To study the structure of the composites, 20 μm slices were prepared with a microtome; the structure images were obtained using a Primo Star optical microscope (Carl Zeiss, Germany). The packing density coefficient of fillers (packing factor) F was determined by the vibrational compression method [34]. All the measurements were performed at $T = 20 \pm 1^\circ\text{C}$. The density of the composites was determined by the method of hydrostatic weighing (GOST 15139-69). The samples were first weighed in the dry state, and second, being immersed in water. The sample density (ρ) was calculated using the equation:

$$\rho = \frac{M}{M - m} \rho_w, \quad (1)$$

where M is the weight of the dry sample; m is the weight of the sample in the liquid; ρ_w is the density of distilled water (at a temperature of 20°C , $\rho_w = 0.9981$).

At a low filler concentration in the composites (up to 30 vol.%) the conductivity was measured at constant current using a

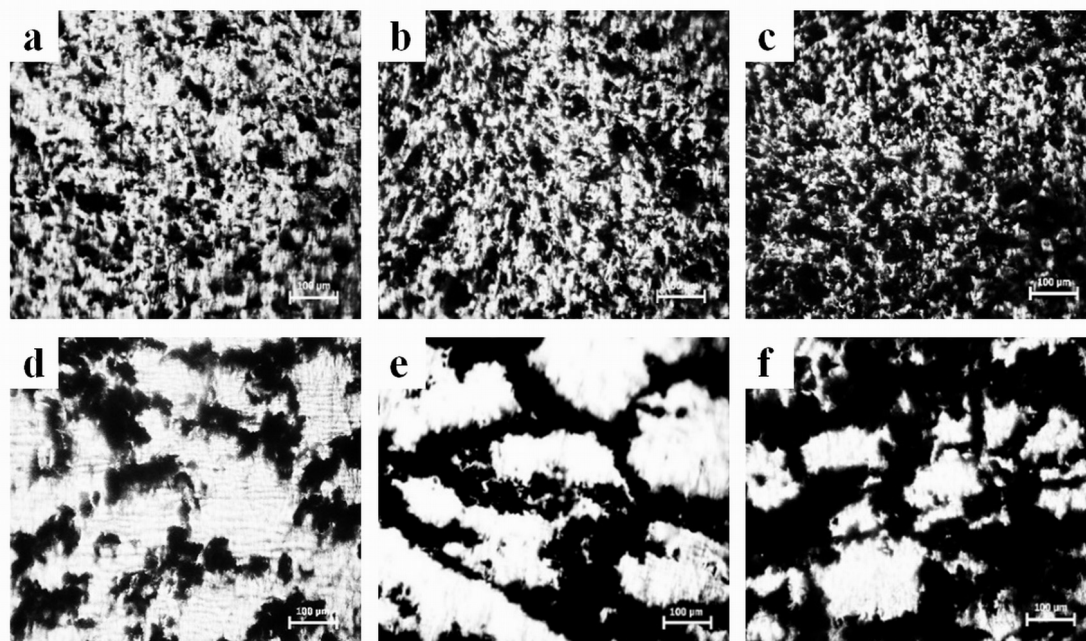


Fig. 1. Optical images of the LDPE-Cu composites structure with two types of the filler distribution: random (a–c) and segregate (d–f). Microphotos (a, d) correspond to 5 vol.%; (b, e) to 10 vol.% and (c, f) to 15 vol.% of Cu.

E16-13A teraohmmeter (RadioTechnik, Latvia) with a two-electrode method and voltage on the electrodes of 100 V, the diameter of the electrodes was 2 cm. For the filler concentration $\varphi > 30$ vol.%, electrical conductivity was measured with the P4833 DC bridge (Kharkiv, Ukraine) designed to measure low resistances. The experimental values of the electrical conductivity (σ) of the composites were calculated as follows:

$$\sigma = \frac{h}{RS}, \quad (2)$$

where h is the thickness of the sample; R is the resistance of the sample; S is the area of the electrode.

The thermal conductivity of the composites was measured using a method of Hot Disc with a TPS 2200S instrument (HotDisk AB, Sweden).

3. Results and discussion

3.1 Structure of composites

The structure of the composites, namely the distribution of filler in the volume of a polymer matrix, plays an important role in the determining material properties. The random type of the distribution is achieved by mixing the filler powder with the polymer in the molten state during the composite extrusion (*R*-type composite). In this case, the filler is dispersed in a random

manner over the whole volume of the polymer matrix, the particles can be single or in the form of aggregates.

The formation of a composite with the segregated type of the filler distribution (*S*-type composite) requires filler particles of smaller size than the polymer ones [22]. During the mechanical mixing of the two powders, the filler covers the surface of the polymer particles. Subsequently, during the hot compacting, an ordered structure of the filler occurs within the polymer matrix. In this case, the filler is localized at the interface between the polymer particles and, with an increase in its concentration, forms a conducting framework. With this approach, it is possible to form a composite with a high local concentration of filler in the framework wall, which defines electrical and thermal properties of composites.

Figure 1 shows the structure of the LDPE–Cu composites with different filler concentrations and types of its distribution. As can be seen from Fig. 1 (a–c), in the *R*-type composites, the copper particles are distributed in a random manner and form a number of aggregates. As the proportion of Cu in the composite increases, a conductive filler cluster is formed that imparts the conductivity to the composite. In the segregated system (*S*-type composites) shown in Fig. 1 (g–e), a cell structure is formed, in

which the filler is localized at the interfaces between the polymer particles creating a conductive framework. As the concentration of the filler increases, the walls of the framework become thicker.

One of the important parameters that determines the structure of a composite is the packing factor (F), which takes into account the shape of the filler particles and their distribution in the polymer matrix. This parameter defines the filling limit of the system and can be represented as follows [35]:

$$F = \frac{V_f}{V_f + V_p}, \quad (3)$$

where V_f is the volume occupied by filler particles at their maximum content, V_p is the volume of free space between the filler particles (occupied by polymer). For spherical particles, the theoretical value is $F = 0.64$ (64 vol.%) in the case of their statistical (random) distribution. The larger the shape of the particles differs from the spherical one, the lower the value of the packing factor. In [35] it was shown that the value of F increased from 0.22 to 0.32 with an increase in the average particle size of copper from 10 to 105 μm due to a change in the shape of the particles with their size. It should be noted that the method of production of the dispersed copper significantly affects the shape of its particles. For the copper particles used in this work, the experimental value $F = 0.36$ (36 vol.%) was obtained.

As the copper concentration increases above the filling limit, i.e. above 36 vol.% the composite becomes a three-component system: filler particles, polymer in the space between them and air pores. An increase in the filler concentration up to 100 vol.% is formal, since the true filler concentration cannot exceed the value of F , which is the filling limit. The remaining part, i.e. $(100-F, \text{vol.}\%)$, will be occupied by polymer and pores. When the formal copper concentration goes towards 100 vol.%, the polymer volume decreases, the part of pores increases, and at 100 vol.%, the sample consists of the copper particles and the pores between them.

3.2. Electrical conductivity of composites

The dependences of the electrical conductivity on the filler content in the LDPE–Cu composites with different types of the filler distribution are presented in Fig. 2. To de-

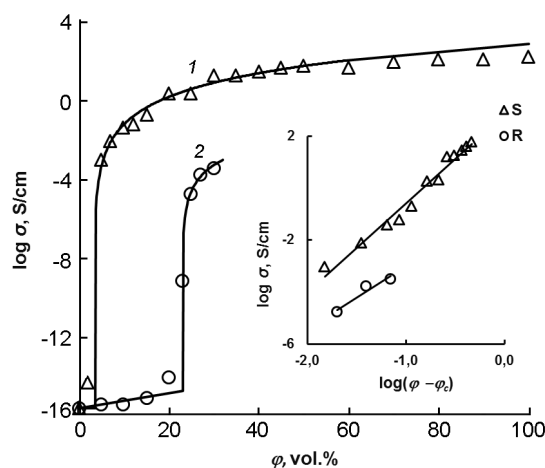


Fig. 2. Concentration dependences of electrical conductivity: points are experimental values; lines are calculated curves in accordance with Eq. 4 for the composites with segregated (1) and random (2) types of the filler distribution. The inset is a fit of the experimental data.

scribe the obtained experimental results, the equation of the percolation theory in the concentration region $\varphi > \varphi_c$, was used [21]:

$$\sigma = \sigma_0(\varphi - \varphi_c)^t, \quad (4)$$

where σ_0 is the parameter that determines the conductivity of the dispersed phase; φ is the filler concentration in the composite; φ_c is the value of the percolation threshold; t is the critical index.

The calculated curve based on the equation (4) describes the concentration dependence of the electrical conductivity of the composite and demonstrates very good agreement with experimental data (Fig. 2). The parameters obtained from the fitting of the experimental data are presented in Table. Although at $\varphi > F$ the composite becomes a three-component system, it is still possible to use the percolation equation up to $\varphi = 50$ vol.%, since the percolation theory is based on the fact that the conductive particles are immersed in a non-conductive media, which is both polymer and air.

From the data given in Table and Fig. 2 it can be seen that a composite with a random type of filler distribution has a percolation threshold at the value of 23 vol.% copper. Such a high value is due to the fact that the filler is distributed throughout the whole volume of the polymer matrix with a large distance between particles. Also, the extrusion of the composite facilitates good wetting of the filler particles by the poly-

Table. Parameters of the percolation equation for LDPE-Cu composites.

Type of distribution	σ_0 , S/cm	φ_c , vol.%	t
Random (<i>R</i>)	0.28	23.0	2.3
Segregated (<i>S</i>)	234	3.3	3.5

mer melt, which impedes the contacts between them and the formation of conductive chains.

The composite with a segregated filler distribution of *S*-type is characterized by a much lower value of the percolation threshold $\varphi_c = 3.3$ vol.% and a higher value of the conductivity $\sigma_0 = 234$ S/cm versus $\sigma_0 = 0.28$ S/cm for the *R*-type composite. This is due to the fact that the filler is concentrated in a layer between the polymer particles, and its local concentration φ_{loc} in the walls of the conductive framework is much higher than the average value φ calculated for the whole volume of the composite, $\varphi_{loc} > \varphi$. In the segregated system, filler particles directly contact each other; while in the case of a random filler distribution, a small polymer layer on the surface of the particles worsens the conditions for the transfer of electric charge.

The formal increase in the filler concentration in the region $\varphi > F$ slightly increases the composite conductivity. Thus, at 35 vol.% copper, $\sigma = 1.8 \cdot 10^1$ S/cm, whereas the pressed copper powder (100 vol.%) has a value only an order higher, $\sigma = 1.5 \cdot 10^2$ S/cm. This increase in conductivity is due to the gradual replacement of a part of the polymer by air pores with an increase in the calculated concentration of the filler. When the content of air pores predominates over the polymer content, the copper particles are packed more tightly when compressed, which leads to a slight increase in electrical conductivity.

3.3. Thermal conductivity of composites

The concentration dependences of the density of LDPE-Cu composites with the random and segregated types of filler distribution are shown in Fig. 3. The mixture rule was used to describe the experimental data:

$$\rho = (1 - \varphi)\rho_p + \varphi\rho_f, \quad (5)$$

where ρ_p is the density of the pure polymer, ρ_f is the density of copper, φ is the filler concentration. As can be seen from Fig. 3, in the concentration range from 0 to

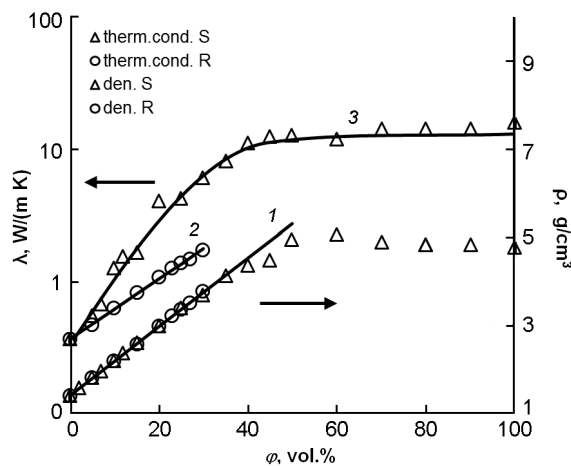


Fig. 3. Concentration dependences of the thermal conductivity and density of the LDPE-Cu composites: triangles and circles are the markers for *S*-type and *R*-type composites, respectively. Line 1 corresponds to the mixture rule (Eq. 5); line 2 is calculated in accordance to the Lichtenecker model (Eq. 6); curve 3 is guided for eyes.

30 vol.% copper, the densities of the *R*-type and *S*-type coincide and match with the values calculated using Eq. (5). It can be concluded that the type of filler distribution does not affect the density of the composite.

In the concentration range $\varphi > F$, the density of the *S*-type composite is almost unchanged. This effect can be explained by the fact that the real filler concentration maintains at the level of $\varphi = F$ and the calculated increasing of the copper content is formal. In the region $\varphi > F$, the content of copper particles in the composite is constant, $\varphi = 36$ vol.%, while the substitution of a part of the polymer by the air pores leads to a nearly unchanged value of the density of the composite.

Concentration dependences of the thermal conductivity of the composites of *R*-type and *S*-type presented in Fig. 3. There are many models describing the thermal conductivity of a two-component system [36, 37]. For the *R*-type composite, the Lichtenecker model was used:

$$\lambda = \lambda_p^{(1-\varphi)}\lambda_f^\varphi, \quad (6)$$

where λ , λ_p , λ_f are the thermal conductivity of the composite, polymer matrix and filler, respectively. An excellent agreement of the experimental and calculated data at the value of $\lambda_f = 69$ W/(mK) can be seen.

The λ_f value is the conductivity of the dispersed filler phase at $\varphi = 100$ vol.%. This value is less than the thermal conductivity of copper λ_m in the solid state, $\lambda_m = 400$ W/(mK). The relation $k = \lambda_f/\lambda_m$ is the parameter defining the influence of high thermal resistance at the particle/particle and particle/polymer interfaces, which worsens the transfer of heat flux through the composite and limits the value of thermal conductivity [38, 39].

For the S-type composite with a segregated structure, the thermal conductivity increases more rapidly, and reaches the plateau in the region $\varphi > F$. As it is seen, the curves for thermal conductivity and density are similar (Fig. 3). A faster increase of thermal conductivity in the S-type composites is due to the existence of a framework consisting of the close-packed metal particles, which provide an increased heat flux transfer. However, the thermal conductivity in the polymer composites is provided both the filler phase and the polymer matrix, and their contribution is comparable [40]. At filler concentration $\varphi > F$, a third phase appears — air, which gradually increases with a thermal conductivity $\lambda(\text{air}) = 0.022$ W/(mK) much less than $\lambda(\text{LDPE}) = 0.37$ W/(mK). The air phase reduces the thermal conductivity of the composites and leads to the appearance of the plateau at $\varphi > F$. Such a behavior of the thermal conductivity correlates with concentration dependence of the density since the influence of air pores in the range of 36–100 vol.% is similar for both characteristics.

4. Conclusions

In this paper, the electrical conductivity and thermal conductivity of the composites with two types of metal filler distribution, random (R-type) and segregated (S-type), are studied in a wide concentration range. The electrical conductivity of the LDPE-Cu composites obeys the percolation equation and has a percolation threshold value of 23.0 vol.% for the random filler distribution and 3.3 vol.% for the segregated structure. The segregated system is also characterized by a higher value of electrical conductivity due to a high local filler concentration φ_{loc} in the walls of the conductive framework.

The type of filler distribution does not affect the density of the composites studied. When the calculated concentration of the filler is more than the value of packing-fac-

tor (F), $\varphi > F$, a third phase, air, appears in the composite, substituting the polymer phase. The content of air pores increases with an increase of the calculated filler content. At the filler concentrations $\varphi > F$, the density of the composite reaches a constant value, since the true filler concentration remains $\varphi = F$.

The thermal conductivity of a composite with a random filler distribution was described by the Lichtenecker model. The value of thermal conductivity of the dispersed phase $\lambda_f = 69$ W/(mK) was found, which is much less than the thermal conductivity of the solid copper (400 W/(mK)). This indicates the high thermal resistance at the particle/particle and particle/polymer interfaces, which worsens heat transfer conditions. The composites with a segregated structure are characterized by a higher value of thermal conductivity even at low filler concentrations. At a high filler concentration, the thermal conductivity of the segregated system reaches the constant value similar to the density behavior.

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