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Influence of the concentration and ratio of components of hybrid nanofillers on the electrical conductivity of modified bitumen binders

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Abstract: Modifying additives are introduced into the composition of modern bituminous binders to meet the high modern requirements for the performance of road surfaces. The use of carbon nanomaterials as modifiers imparts antistatic properties to bituminous binders and opens up the possibility of applying an innovative approach to healing cracks in the asphalt pavement under the action of microwave radiation with its subsequent regeneration. A series of samples of nanocomposite materials based on BND 60/90 bitumen and hybrid filler containing graphene nanoplatelets (GNPs) and multi-walled carbon nanotubes (MWCNTs) were prepared using a simple mixing technique. The use of hybrid with a ratio of MWCNTs to GNP equal to 4:0.1 made it possible to achieve the maximum electrical conductivity of bitumen nanocompositions, the value of which was $4.82 \cdot 10^{-3}$ S·cm^{-1} at a filler concentration of 8 wt. %. The optimal ratio of MWCNTs to GNP components equal to 4:1 was experimentally found, at which the percolation threshold decreased by 50 and 100 % compared with nanocomposites having a MWCNT to GNP ratio of 4 : 0.1 and 1 : 1, respectively. A mechanism for the formation of a double percolation contour in a bitumen-based nanocomposite system containing GNP and MWCNTs in a ratio of 4 : 1 is proposed.

Keywords: bitumen; electrical conductivity; percolation; carbon nanotubes; graphene nanoplates.

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Влияние концентрации и соотношения компонентов гибридного нанонаполнителя на электропроводность модифицированных битумных вяжущих

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Аннотация: В состав современных битумных вяжущих вводят модифицирующие добавки для удовлетворения высоких современных требований, предъявляемых к эксплуатационным характеристикам дорожных покрытий. Использование углеродных наноматериалов в качестве модификаторов придает битумным вяжущим антистатические свойства и открывает возможность применения инновационного подхода по залечиванию трещин в асфальтовом покрытии под действием сверхвысокочастотного микроволнового излучения с его последующей регенерацией. Серии образцов нанокомпозиционных материалов на основе битума марки «БНД 60/90» и гибридного наполнителя, содержащего графеновые нанопластинки (ГНП) и многостенные углеродные нанотрубки (МУНТ), были изготовлены посредством простой методики смешения. Использование гибрида с отношением МУНТ к ГНП равным 4 : 0.1 позволило достичь максимальной электропроводности битумных нанокомпозиций, значение которой составляло $4{,}82 \cdot 10^{-3}$ См/см при концентрации наполнителя 8 масс. %. Экспериментально найдено оптимальное соотношение компонент МУНТ к ГНП равное 4 : 1, при котором порог перколяции снижался на 50 и 100 % по сравнению с нанокомпозитами, имеющими отношение МУНТ к ГНП равное 4 : 0.1 и 1 : 1 соответственно. Предложен механизм образования двойного перколяционного контура в нанокомпозиционной системе на основе битума, содержащей ГНП и МУНТ в соотношении 4 : 1.

Ключевые слова: битум; электропроводность; перколяция; углеродные нанотрубки; графеновые нанопластинки.

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1. Introduction

Modern bituminous binders are multicomponent systems containing a wide range of modifiers, such as polymers, rubbers, synthetic or natural resins, and inorganic salts. As a result of the use of modifying additives in bitumen, a composite with improved performance characteristics is obtained. A special class of modifiers are micro- and nano-sized electrically conductive fibers and particles (steel wool, carbon fibers, carbon black, multi-walled carbon nanotubes (MWCNTs), graphene nanoplates (GNPs)), the use of which makes it possible to ensure the susceptibility of bituminous binders to microwave radiation and the implementation of the process of healing cracks in an asphalt pavement with its subsequent regeneration [1]. However, metal fiber has a rather high cost, which significantly complicates the selection and manufacture of modified bitumen compositions due to the shape of the filler particles and reduced adhesion of bitumen to stainless steel [2].

The use of MWCNTs and other microwave-susceptible carbon nanostructures as modifiers provides an increase in the performance properties of bituminous binders at significantly lower concentrations compared to metal fiber. Microwave-susceptible carbon nanostructures make it possible to obtain bituminous binders, which have the effect of self-healing microcracks in the asphalt pavement when “warmed up” under the action of microwave irradiation [2].

To study the processes occurring under the action of microwave irradiation in nanocomposite bitumen systems containing carbon nanostructures, information on their electrical characteristics (specific electrical conductivity, volume fraction of the filler at the percolation threshold, critical electrical conductivity index) is needed.

The current literature completely lacks data on the electrical characteristics of bituminous binders modified with hybrid filler containing MWCNTs and GNPs, which are necessary for the design and development of innovative asphalt pavement compositions susceptible to microwave radiation.

However, the effect of hybrid carbon fillers on the properties of polymer nanocomposites is currently being actively studied. Many authors present findings that report a significant increase in the electrical conductivity of polymer nanocomposites containing hybrid carbon filler due to the realization of a synergistic effect [3–10].

The authors of [3] studied the influence of MWCNTs and GNPs ratio on the mechanical and electrical properties of hybrid epoxy composites. The study shows that the combination of CNTs to GNPs in a ratio of 8 : 2 synergistically increases the bending strength and reduces the electrical percolation threshold of epoxy composites due to the easier formation of a conductive network.

Liu et al. studied the effect of hybrid filler containing CNTs to GNPs in a 3 : 1 ratio on the electrical conductivity of thermoplastic polyurethane (TPU) [4]. The paper shows an increase in electrical conductivity by 7 orders of magnitude for hybrid nanocomposites compared to composites containing only CNTs. The authors attribute this synergistic effect to the formation of efficient electrically conductive paths when using hybrid filler, which confirms the low percolation threshold that amounted to 0.012 wt. %.

It was reported in [5] that the combination of CNTs with GNPs in polymer composites can have a synergistic effect on increasing the electrical properties of composites. The resulting polyvinylidene fluoride compositions filled with a CNT/GNP hybrid demonstrate higher electrical conductivity compared to nanocomposites containing only CNTs or GNPs.

The authors of [6] obtained multifunctional epoxy composites reinforced with a three-dimensional hybrid consisting of reduced graphene oxide (r-GO) and CNTs, in which the carbon network was constructed by depositing r-GO and CNTs on the framework of a polyurethane (PU) sponge through wall-by-wall assembly. The r-GO/CNTs hybrid filler significantly changed the surface properties of the polyurethane increasing its ability to be wetted by epoxy resin and also contributed to the suppression of crack formation and thus inhibited the mechanical destruction of the composition. These composites had good electrical conductivity (0.01 $S \cdot cm^{-1}$), an ultra-low percolation threshold (0.0034 wt. %), and increased shielding efficiency from electromagnetic radiation.

Bagotia et al. used CNTs, GNPs, and hybrid filler based on them as a polycarbonate/ethylene methyl acrylate filler [7]. The conductivity of compositions based on CNTs and GNPs was $1{,}56 \cdot 10^{-3}$ and $5{,}7 \cdot 10^{-3}$ $S \cdot cm^{-1}$, respectively. The authors also note that the use of hybrid filler in various ratios increases the electrical conductivity by about fifteen orders of magnitude compared to the polymer matrix. A nanocomposite sample with a mass ratio of GNP to CNT equal to 1 : 3 exhibited a synergistic effect and had a maximum conductivity of $1{,}913 \cdot 10^{-1}$ $S \cdot cm^{-1}$.

In [8], the study of nanocomposites based on hybrid filler containing CNTs and GNPs is righteous. The authors note an increase in the electrical conductivity of nanocomposites filled with hybrid filler compared to compositions based on a monofiller. The percolation threshold for hybrid composites was observed in the region of 10^{-7} $S \cdot m^{-1}$, which corresponds to an increase in electrical conductivity by six orders of magnitude compared to the original epoxy matrix. The addition of small amounts of CNTs (10 % compared to GNPs) as an auxiliary conducting phase makes it possible to reduce the mass content of the filler corresponding to the percolation threshold by 50 % and obtain hybrid nanocomposites with an increased electrical conductivity by several orders of magnitude. The authors explain this synergistic effect by the formation of highly efficient electrically conductive hybrid networks based on CNTs and GNPs.

The results of experimental studies of the electrical conductivity of polymer nanocomposite systems containing hybrid carbon fillers (controlled-length CNTs and GNPs) are summarized in [9]. The authors report a significant increase in the electrical conductivity of polymer composites when using the optimal ratio of CNTs to GNPs, and not when using only one modifying filler. Moreover, it was found that GNPs are more effective in increasing thermal conductivity and tensile strength, while CNTs significantly increase electrical conductivity.

Theoretical studies of synergistic effects of hybrid polymeric nanocomposites were carried out by the authors of [10]. The study proposes and applies a model for predicting the conductivity of hybrid nanocomposites based on the Bethe lattice method. The model innovatively combines the geometric and electrical parameters of the filler and matrix with the overall electrical conductivity of polymer nanocomposites containing hybrid filler, which is impossible or only partially achieved by the traditional phenomenological model and the nonphenomenological binary conductivity model.

Thus, the study of the effect of hybrid carbon fillers on the electrical conductivity of bituminous binders is an important scientific and practical task.

In this regard, the aim of the work was to develop a methodology for obtaining and studying the effect of the concentration and ratio of the components of hybrid filler containing GNPs and CNTs on the electrical conductivity of modified bitumen binders.

2. Materials and Methods

2.1. Initial materials

Road bitumen of the BND 60/90 brand (Ural Bitumen Plant Ltd., Yekaterinburg, Russia) was used as the basis for the composites. Taunit-M CNTs, as well as Taunit GM GNPs in the form of an aqueous paste (7.2 wt. % dry residue), produced by NanoTechCenter LLC, Tambov, obtained by a modified Hummers method, was used as an electrically conductive modifier.

2.2. Technology for obtaining modified bitumen samples

To eliminate aggregation and remove adsorbed water, MWCNTs were preliminarily dried in a vacuum oven at 150 °C for 4 h. After drying, MWCNTs were mechanically activated in a WF-20B blade mill for 3 min at a grinding body rotation speed of 25000 rpm in order to reduce the size of agglomerates and improve their dispersion in the polymer matrix, as was shown in [11].

GNPs in its original form was a water paste, which prevented its combination with bitumen. In this regard, GNPs was freeze-dried in a SCIENTZ-10N dryer (SCIENTZ, China). Drying consisted of two stages. At the first stage, the sample of GNPs was frozen to a temperature below −30 °C for 20 hours. The sample was frozen until the temperatures of the freezing chamber and the sample being frozen were equalized. At the second stage, the frozen sample was treated with vacuum for 20 h.

Table 1. Designation of the series of nanocomposite samples

Designation of the series of samples	Ratio of MWCNTs to GNPs in hybrid filler	Concentration of hybrid filler, wt. %
1	1 : 1	0.2, 0.4, 0.6, 0.8, 1, 2, 3, 4, 5, 6, 7, 8
2	4 : 0.1	
3	4 : 1	

After freeze drying, GNPs was mechanically activated under the same conditions as MWCNTs.

To obtain modified bitumen samples with hybrid filler containing MWCNTs and GNPs, the technique described in detail in [1] was used. Initially, hybrid modifier was mixed with Nefras S2-80/120 gasoline (NK Rosneft, Russia) using a vertical rotary mixer HT-120DX (Daihan, Korea) and processed with an I-10 ultrasonic generator (Ultrasonic technology – INLAB, Russia) for 30 min. Gasoline heated to 110°C and lumpy bitumen were placed in a separate metal container. The previously prepared dispersion of the modifier in gasoline was introduced into the resulting melt. When obtaining a series of samples of nanocomposites, hybrid filler containing GNPs and MWCNTs at the concentrations indicated in Table 1 was added to 5 g of bitumen.

2.3. Determination of the properties of nanofillers and specific volumetric electrical resistance of modified bitumen compositions

The study of the morphology and structure of MWCNTs and GNPs was carried out using scanning (SEM) and transmission microscopy (TEM) methods implemented in Merlin (Carl Zeiss, Germany) and JEM 2100F (JEOL, Japan) instruments, respectively.

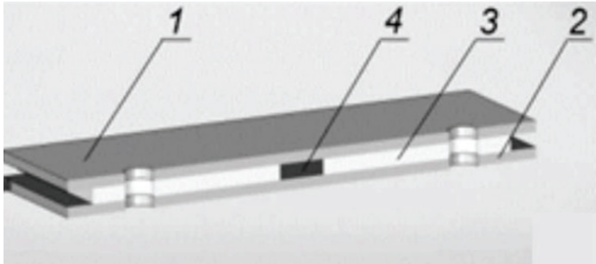


Fig. 1. Measuring cell of the volume resistivity of bituminous binders in the section: *1*, *2* – measuring electrodes; *3* – matrix; *4* – composite

To study the specific volumetric electrical resistance of modified bitumen compositions, a measuring cell was used (Fig. 1).

A detailed description of the operation principle of the measuring cell is given in [1]. Sample resistance was measured by connecting the upper and lower measuring electrodes to an E6-13A teraohmmeter (PunaneRet, Estonia) with an upper measurement limit of 10^{14} Ω. Electrical conductivity was calculated according to formula (1) [12, 13]:

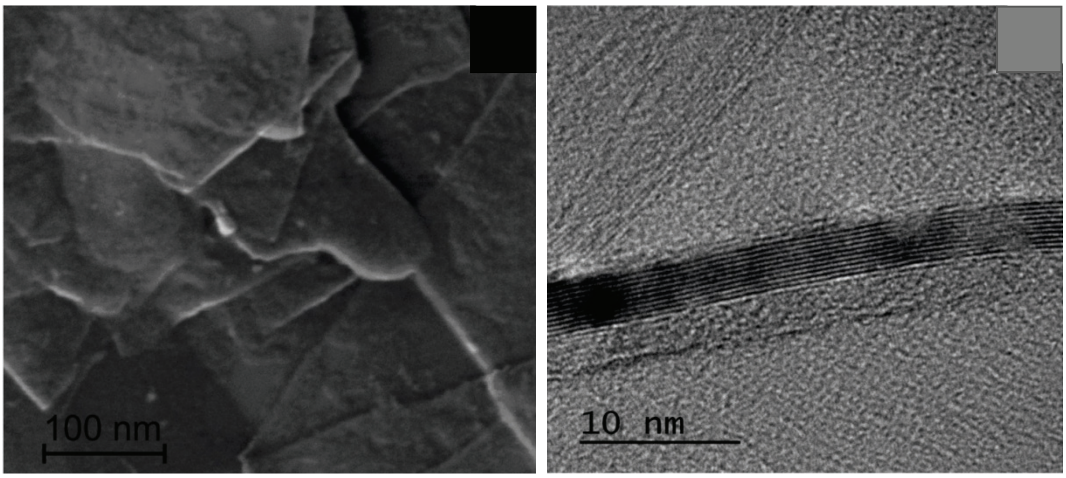
$$\sigma = \frac{4h}{\pi d^2 R}, \quad (1)$$

where h is the height of the test sample, cm; d is the diameter of the test sample, cm; R is the electrical resistance Ω.

3. Results and Discussion

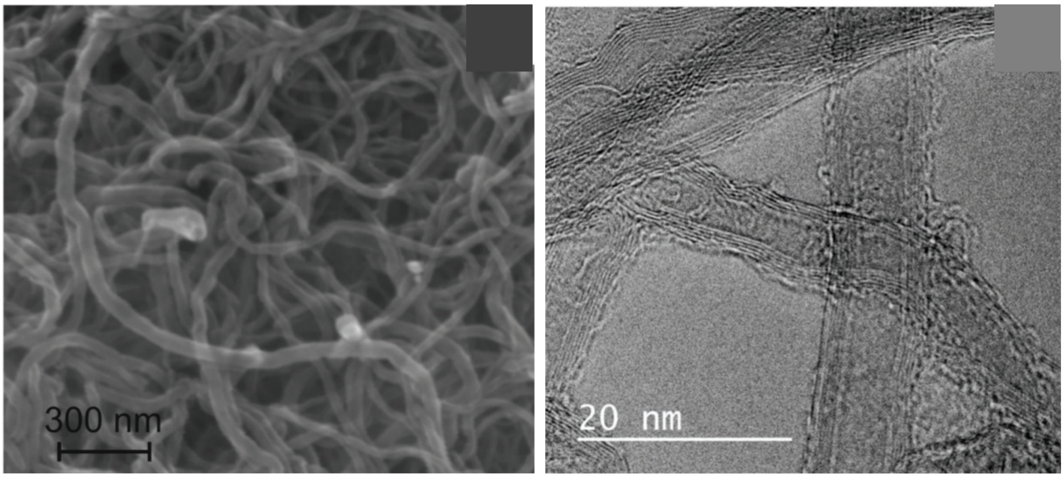
3.1. Characterization of nanofillers

The structure of the GNPs "Taunit-GM" is shown in Fig. 2.



(*a*) (*b*)

Fig. 2. *a* – SEM; *b* – TEM images of GNPs "Taunit GM"



(*a*) (*b*)

Fig. 3. *a* – SEM; *b* – TEM images of GNPs "Taunit-M"

Table 2. Characteristics of the nanomaterial "Taunit GM" [14]

Parameter	GNPs
Number of graphene walls	3–5
Thickness of nanoplates, nm	2–3
Size of nanoplates in plane, μm	2–10
Content of nanoplates, wt. %	10–12
Oxygen content, wt. %	9–13
Sulfurcontent, wt. %	≤ 0,7
Specific absorption coefficient, $(g·cm)^{-1}$	58–63

Table 3. Characteristics of MWCNTs "Taunit-M" [15]

Characteristic	Taunit-M
Outer diameter, nm	10–30
Inner diameter, nm	5–15
Length, μm	≥ 2
Total amount of impurities, %	
Primary	≤ 5
After cleaning	≤ 1
Specific surface, $m^2·g^{-1}$	≥ 270
Bulk density, $g·cm^{-3}$	0,025–0,060

The characteristics of the GNPs are presented in Table 2.

MWCNTs "Taunit-M" are quasi-one-dimensional, nanoscale, filamentous formations of polycrystalline graphite, predominantly cylindrical in shape with an internal channel (Fig. 3).

Structural characteristics of MWCNTs"Taunit-M" are presented in Table 3.

3.2. Electrical conductivity of bitumen nanocomposites containing hybrid filler

The electrical conductivity of Series 1 nanocomposites increased with increasing mass content of hybrid filler and reaches a maximum value of $2.18·10^{-4}$ $S·cm^{-1}$ at its concentration of 8 wt. % (Fig. 4).

The use of hybrid filler with an MWCNTs/GNPs – 4 : 0.1 ratio also led to an increase in the electrical conductivity of the nanocomposites. The maximum electrical conductivity was $4.82·10^{-3}$ $S·cm^{-1}$ at a concentration of 8 wt. %, which is almost an order of magnitude higher than the electrical conductivity of Series 1 nanocomposites (Fig. 5). A decrease in GNPs concentration in hybrid filler makes it possible to increase the maximum electrical conductivity of bitumen nanocomposites, which is consistent with the results of [9].

The percolation threshold for samples of Series 2 was observed at a lower content of hybrid filler (3 wt. %,) (Fig. 5) in comparison with samples of Series 1, for which the percolation threshold was observed at 4 wt. % hybrid filler (Fig. 4). Therefore, the use of hybrid filler with MWCNTs/GNPs – ratio of 4 : 0.1 contributes to a better formation of a percolation network in bitumen.

In the case of using hybrid filler MWCNTs/GNPs – 4 : 1, a double percolation

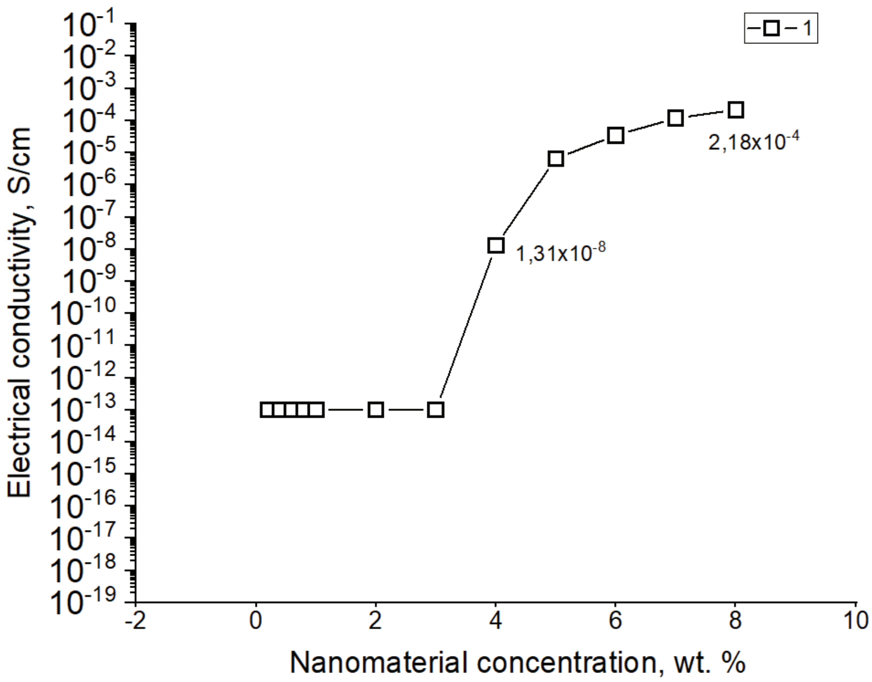


Fig. 4. Dependence of the electrical conductivity of Series 1 nanocomposites on the concentration of hybrid filler

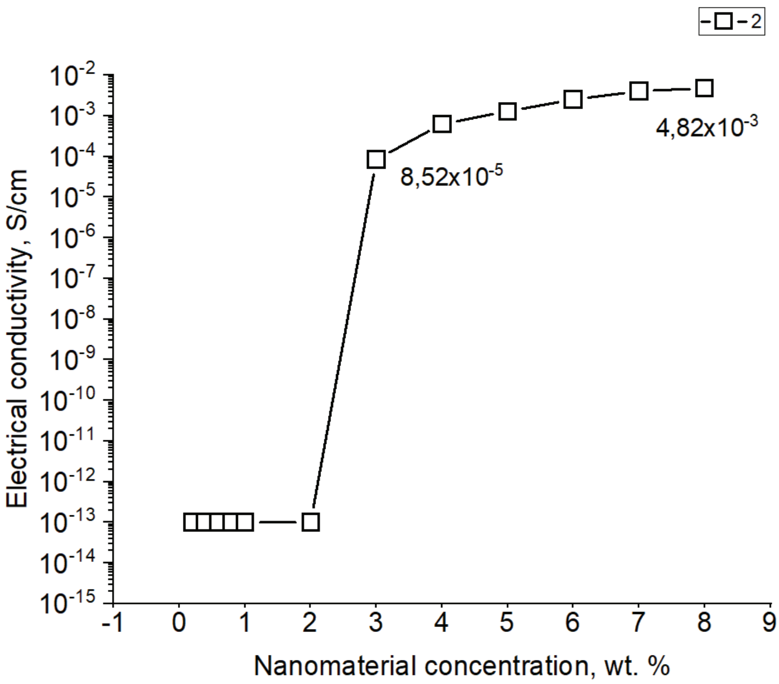


Fig. 5. Dependence of the electrical conductivity of Series 2 nanocomposites on the concentration of hybrid filler

threshold was observed. The first percolation threshold was observed at 2 wt. %, and the second – at 5 wt. %. The maximum value of electrical conductivity of $5.65{\cdot}10^{-4}$ $S{\cdot}cm^{-1}$ was observed after the second percolation threshold at 8 wt. % hybrid filler in samples of Series 3 (Fig. 6).

The value of electrical conductivity, which was $1{\cdot}10^{-13}$ $S{\cdot}cm^{-1}$ for nanocomposites with a low content of hybrid filler (< 3 wt. % for Series 1, < 2 wt. % for Series 2 and < 1 wt. % for Series 3) does not differ from the original bitumen (Figs. 4–6). Therefore, an increase in the electrical conductivity of nanocomposites occurs only with the formation of electrically conductive network formed by hybrid filler in a bitumen matrix, which is consistent with the results of [3–10].

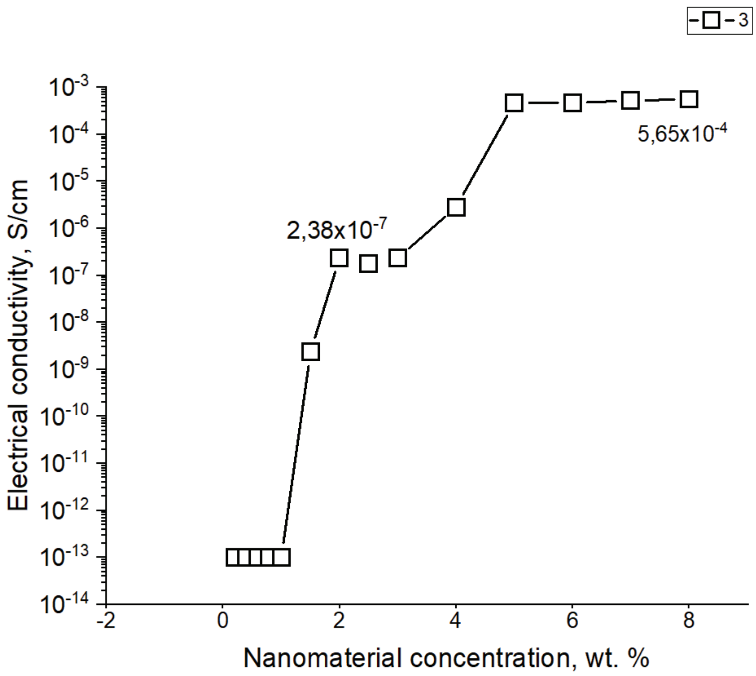


Fig. 6. Dependence of the electrical conductivity of Series 3 nanocomposites on the concentration of hybrid filler

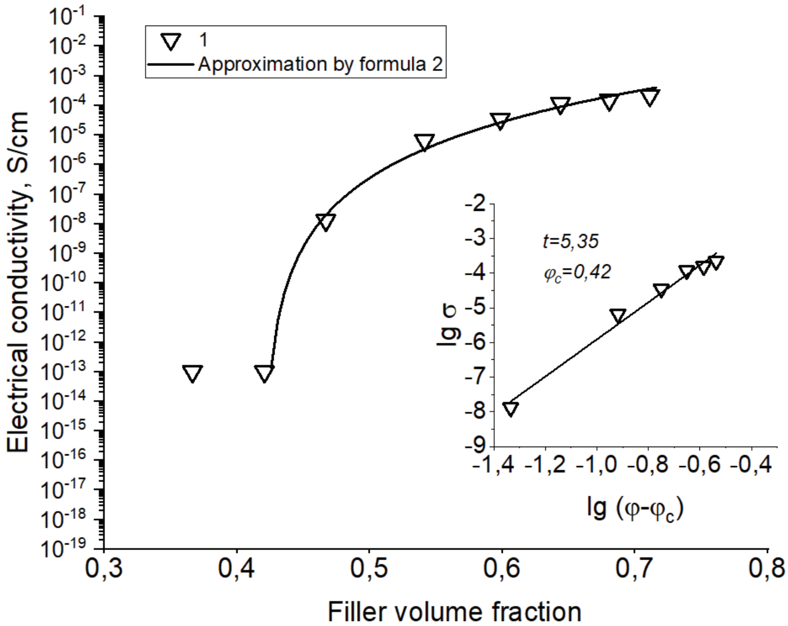


Fig. 7. Approximation of the dependence of the electrical conductivity of Series 1 nanocomposites

Besides, the findings presented in Figs. 4–6 show that dependences of the electrical conductivity of nanocomposites on the concentration of hybrid filler are percolative in nature. The use of various ratios of hybrid filler had a positive effect on the formation of the percolation contour of nanocomposites. The percolation thresholds for nanocomposites of Series 2 and 3 were observed at a lower mass content of hybrid filler, in comparison with samples of Series 1 (Figs. 4–6). The obtained dependences of electrical conductivity are described by the classical equation of percolation theory 2 [16]:

$$\sigma = \sigma_f (\varphi - \varphi_c)^t , \quad (2)$$

where φ_c is the volume fraction of the filler corresponding to the percolation threshold; t is the critical electrical conductivity index; and σ_f is the electrical conductivity of the filler.

For the convenience of approximating experimental dependences of electrical conductivity, we take the logarithm of both parts of equation (2). As a result we get:

$$\log \sigma = \log \sigma_f + t \log(\varphi - \varphi_c). \quad (3)$$

The volume fractions of hybrid filler at the percolation threshold φ_c and critical electrical conductivity values t were determined using linear regression of $\log\sigma$ vs. $\log(\varphi - \varphi_c)$ plot (in set in Figs. 7–9).

For Series 1, and t were 0.42 and 5.35, respectively (Fig. 7). The use of hybrid filler MWCNTs/GNPs with a ratio of 4 : 0.1 led to a change in φ_c and t, which became equal to 0.53 and 3.73, respectively (Fig. 8).

Series 3 nanocomposites have a double percolation threshold (Fig. 9).

For the first percolation threshold, φ_c and t were 0.31 and 5.46, respectively (Fig. 9). Probably, the first percolation transition in Series 3 nanocomposites is formed by MWCNTs, since the value of the filler

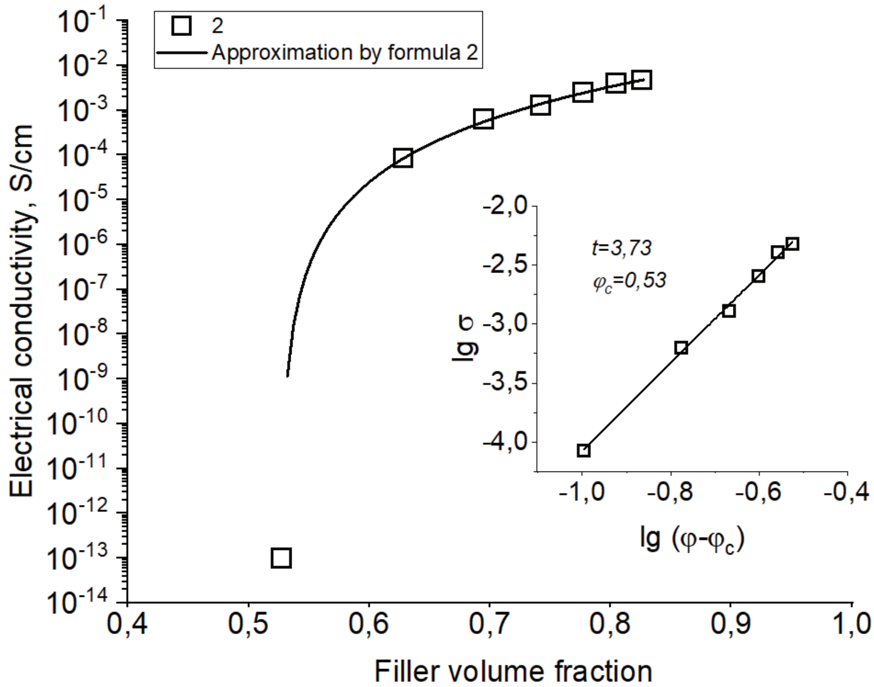


Fig. 8. Approximation of the dependence of the electrical conductivity of Series 2 nanocomposites

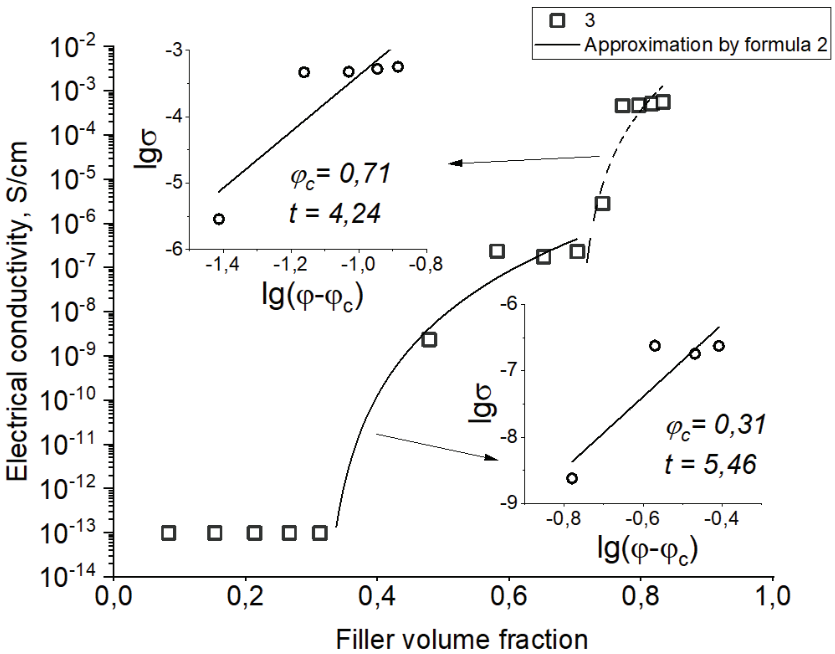


Fig. 9. Approximation of the dependence of the electrical conductivity of Series 3 nanocomposites

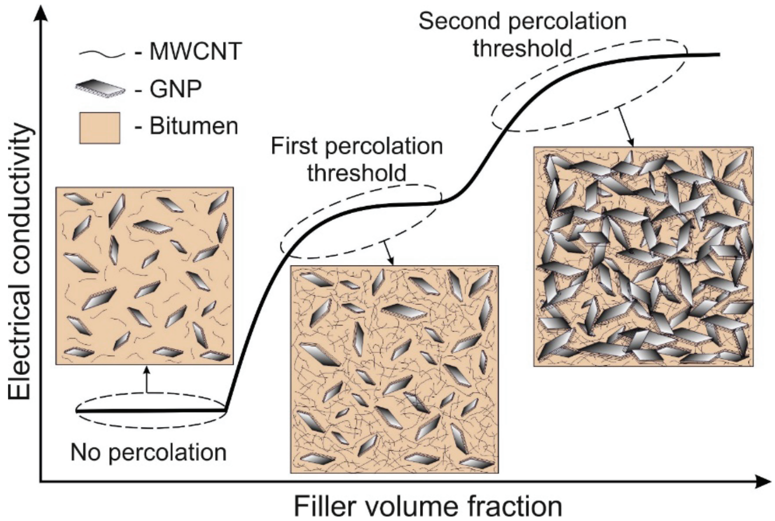


Fig. 10. Formation mechanism of a double percolation threshold in Series 3 nanocomposites

concentration at the percolation threshold approaches the value of φ_c equal to 0.22, obtained in [1] for bituminous compositions containing only MWCNTs.

For the second percolation threshold, φ_c and t were 0.71 and 4.24, respectively (Fig. 9). The appearance of the second percolation threshold in Series 3 nanocomposites occurs during the formation of a conducting loop formed by the second component of hybrid filler (GNPs). The value of the concentration of hybrid filler at the percolation threshold for the second threshold approaches value of φ_c equal to 0.63 obtained for bituminous compositions containing only GNPs [1].

The values of t obtained as a result of approximation of the experimental data by equation (3) (Figs. 7–9) range from 3.73 to 5.46 and are an indicator of good convergence of experimental results with the estimated values of the percolation theory for composites that have a three-dimensional conductive network [17, 18].

Figure 10 shows a hypothetical mechanism for the formation of a double percolation threshold in Series 3 nanocomposites, which was developed based on the analysis of the electrical conductivity dependence (Fig. 9).

When low concentrations of hybrid filler are introduced into the bitumen matrix, there is no change in the electrical conductivity of nanocomposites (Fig. 10), which is confirmed by the experimental results presented in Fig. 6. At the initial stage, the concentration of hybrid conducting phase is not enough to form a percolation contour, and the value of the electrical conductivity of compositions corresponds to the conductivity of the bitumen matrix. A further increase in the filler concentration leads to the formation of the first percolation contour formed by MWCNTs in the bitumen matrix. The concentration of GNPs at this stage is not enough for the formation of the second circuit. GNPs act here as an additional conductive component, which lowers the percolation threshold compared to samples of Series 1 and 2 (Figs. 4 and 5). With a further increase in the concentration of hybrid filler, the formation of the second percolation contour formed by GNP occurs (Fig. 10).

However, no synergistic effect was found in the considered series of compositions, which is probably due to the peculiarities of the method of their preparation. Despite this, the optimal ratio of MWCNTs to GNPs components equal to 4 : 1 of hybrid filler was experimentally found, at which the percolation prog is reduced by 50 and 100 % compared to nanocomposites with MWCNTs/GNPs ratios of 4 : 0.1 and 1 : 1, respectively. From a practical point of view, the replacement of CNTs by cheaper GNPs will lead to a decrease in the cost of bitumen nanocomposites by tens of percent, which is of great importance on the scale of road construction [19–22].

4. Conclusion

Thus, within the framework of the study, a simple scalable method for obtaining road bitumen modified with hybrid filler containing MWCNTs and GNPs has been developed. The optimal ratio of conductive components in hybrid filler for a bitumen matrix (4 : 1) was experimentally found, at which an early formation of the percolation threshold is observed. The formation of a double percolation contour in this nanocomposite system is also theoretically substantiated. The new knowledge obtained about the electrical conductivity of bitumen nanocomposites containing hybrid filler creates prerequisites for the design and production of optimized bitumen compositions with a given volumetric electrical conductivity and susceptible to microwave radiation, which will ensure the implementation of the process of healing cracks in the asphalt pavement with its subsequent regeneration. The cumulative socio-economic effect from the use of bituminous binders modified with hybrid filler will exceed by orders of magnitude the possible rise in the cost of these materials compared to "traditional" ones.

5. Acknowledgments

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6. Conflict of interests

The authors declare no conflict of interest.

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