MATERIALS PHYSICS AND MECHANICS RESEARCH ARTICLE

Submitted: March 25, 2025 **Revised: April 18, 2025** **Accepted: May 20, 2025**

Conductive and mechanical properties of graphene-filled polymer composites

D.N. Tonkov ✉, **M.I. Kobylyatskaya**, **E.S. Vasilyeva, V.E. Gasumyants**

Peter the Great St. Petersburg Polytechnic University, St. Petersburg, Russia

✉ wtk_dima@mail.ru

ABSTRACT
The conductive and mechanical properties of polymer composites based on styrene-butadiene rubber with different concentrations of graphene as a filler are studied. Based on the dependence of the resistivity of composites on the graphene content, the range of its values corresponding to the hopping conductivity mechanism was determined. For composites of compositions from this range, the sensitivity of the resistance to the degree of uniaxial stretching, as well as mechanical properties were investigated. The values of the gauge factor were determined, and it was found that with an increase in graphene content in the composite, its value first increases and then decreases. It is found that even under minimal stresses in composites, along with elastic deformations, plastic deformations also develop, and the proportion of the latter increases with the filler concentration. As a result, complete mechanical relaxation of the composites does not occur after the applied stress is removed.

KEYWORDS
polymer composite materials • styrene-butadiene rubber • graphene • resistivity • gauge factor • deformations

Citation: Tonkov DN, Kobylyatskaya MI, Vasilyeva ES, Gasumyants VE. Conductive and mechanical properties of graphene-filled polymer composites. *Materials Physics and Mechanics*. 2025;53(2): 48–54. http://dx.doi.org/10.18149/MPM.5322025_5

Introduction

In recent years, there has been a significant increase in the number of studies devoted to polymer conductive composite materials with graphene-based fillers, as well as other structures in which graphene is used as one of the materials [1–7]. This is due to the unique physical and chemical characteristics of graphene-containing materials and the prospects for their application in various fields of science and technology. Polymer nanocomposite films represent a promising basis for developing sensor devices for use as elements of wearable electronics [8–10]. These materials are characterized by unique structural and mechanical properties, which makes it possible to achieve such important characteristics of composites from a practical point of view as lightness, flexibility, wear resistance and corrosion resistance, as well as the ability to form products in various configurations. Devices based on polymer nanocomposites are distinguished by their miniature size, making them nearly invisible to users when worn continuously. The development and application of new sensor materials contribute to progress in various fields such as medicine [11–14], the sports industry [15], professional activities in hazardous conditions where health monitoring is necessary [16], as well as in game design and fashion [17]. In addition, polymer nanocomposites are actively used as triboelectric generators [18].

Publisher: Peter the Great St. Petersburg Polytechnic University

One of the promising areas for the use of flexible conductive polymer composites is their use for the creation of strain sensors [19,20], for which a conductive filler is introduced into the polymer matrix. The conductive network formed by the filler particles (e.g., metallic micro- or nanoparticles) is partially destroyed during deformation, which leads to an increase in the resistance of the composite. However, traditional composites based on metallic fillers have low flexibility and elasticity, as well as sensitivity to stretching in a very limited range [21]. In this sense, polymer composites with carbon-based fillers exhibit better characteristics, which is why composites based on various polymers with fillers such as graphite [22], graphene [23] or carbon nanotubes [24] have been actively studied in recent years. However, the properties of the resulting composites with these fillers depend significantly on the synthesis method, the type of polymer matrix and filler, as well as the filler content, which initiates further research. It should be noted that graphene is an excellent choice as a filler material for such composites due to its conductivity, thermal conductivity, and mechanical properties [25].

This study presents an experimental investigation of the conductive and mechanical properties of flexible polymer composites based on styrene-butadiene rubber (SBR) as the matrix and graphene as the filler. The aim of the study was to determine the dependence of the strain sensitivity of the obtained composites, their elongation under uniaxial static tension, ultimate strength and relaxation properties on the graphene content.

Materials and Methods

Styrene-butadiene rubber (SBR) from Sigma-Aldrich was used as the non-conductive polymer matrix, and powdered graphene was used as the filler.

The process of forming the composite film involved the preliminary dispersion of the graphene powder in the CCl_4 solvent using magnetic stirring in a sealed flask to prevent the solvent from evaporating and air from entering the system. SBR granules were then added to the obtained dispersion in an amount of 5.9 wt. % relative to the mass of the solvent, after which mixing was continued until the polymer was completely dissolved. The resulting solution was poured into a PTFE mold and dried in an exhaust system for 1 h until the solvent was completely removed.

Samples with graphene content ranging from 13 to 33 wt. % relative to the total composite mass were synthesized. The resistivity ρ of the obtained films was measured using the Van der Pauw method. To determine the ρ dependence on the degree of mechanical stretching, the samples were fixed in a measurement cell with a movable part, and electrical contacts were connected to them. After this, step-by-step stretching was carried out by registering the resistivity values at each stage.

The mechanical properties of the composite films were analyzed using the same setup without connecting electrical contacts. The strength and relaxation properties were studied by repeatedly stretching the samples in 10 mm steps. At the end of each cycle, the sample was held in a stretched state for 5 sec, after which the load was removed, and after 25 sec the residual length of the sample was measured. The relaxation time was determined by stretching the sample to 400 % of its initial length and then recording the change in length every 20 sec until the relaxation process was complete.

Results and Discussion

To analyze the change in the conductive properties of the composites under deformation, it is necessary to take into account the percolation threshold for the conductive filler particles introduced into the dielectric polymer matrix. The main parameter that determines the properties of the resulting composite is the degree of particle overlap [26]. At high particle concentrations, a continuous conductive cluster is formed, where the particles directly contact with each other. As the particle concentration decreases, their arrangement becomes sparser, and the conduction mechanism changes to a hopping form. With a further decrease in the particle concentration, taking into account their average size, the probability of such "jumps" significantly reduced resulting in a sharp decrease in the conductivity of the composite to nearly zero levels. It is the samples with filler concentrations at which hopping conductivity is realized that are the most promising for use as active elements in sensor devices. The probability of "jumping" (and, consequently, the conductivity value) depends exponentially on the distance between the conductive particles, which makes such composites the most sensitive to stretching.

Thus, to obtain samples with high sensitivity to deformation, it was necessary to determine the range of graphene concentrations corresponding to the hopping conduction mechanism. Figure 1 shows the dependence of the resistivity of the investigated samples at room temperature on the filler mass fraction. From the data presented it is evident that at graphene concentrations below 33 wt. % a sharp increase in the resistivity of the samples is observed. This indicates the destruction of the continuous conductive cluster and the transition to hopping conductivity. For this reason, samples with graphene concentrations of 30, 24, and 16 % were selected for further measurements.

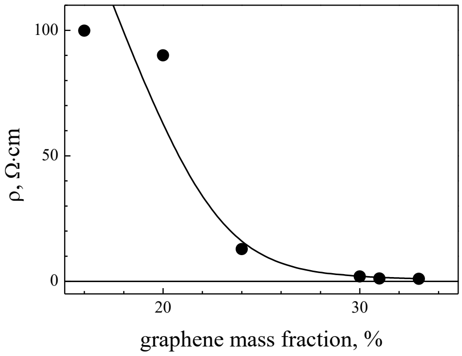

Fig. 1. The dependence of the resistivity of composites at T = 300 K on the graphene content

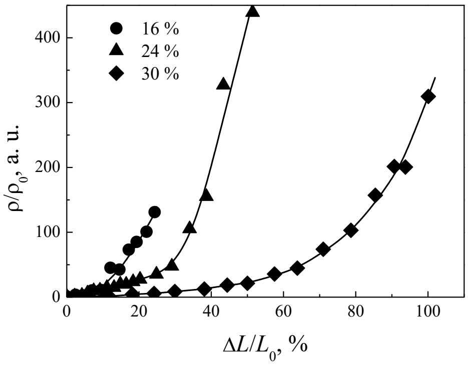

Fig. 2. Changes in the resistivity of composites under their stretching

For the three composites of the above compositions, Fig. 2 shows the dependence of the resistivity, normalized to its value in the absence of applied stress, ρ_0, on the relative elongation of the sample under the influence of uniaxial stress, $\Delta L/L_0 \cdot 100\%$, where L_0 is the initial length of the sample, ΔL is the change in its length under stretching. For the composites of each composition, the data are provided for the stress ranges in which they exhibit conductivity.

As expected, an increase in graphene concentration leads to an expansion of the applied stress range in which the composite retains its resistivity. A noticeable change in the resistivity of the composite at the minimum graphene concentration (16 %) occurred at a stretch of 12 %, while at its maximum concentration (30 %), it occurred at a stretch of 24–30 %. At the same time, the composite with the highest graphene content among those studied has lower sensitivity to stretching: to noticeably change the resistivity, it had to be stretched over a significantly greater length. This effect of filler concentration on stretch sensitivity can be explained by the fact that stretching increases the distance between particles, which, from the perspective of conductivity values, is equivalent to a decrease in the filler concentration. Additionally, as noted by the authors of [27], strong stretching of graphene-containing composites leads to a change in the dimensionality of the filler particles (transition from a three-dimensional to a two-dimensional case), which should lead to an increase in the values of the percolation threshold and, consequently, to a decrease in the effect of stretching on the resistivity value.

For composites of different compositions, the values of the gauge factor were calculated using the equation:

$$GF = (\Delta\rho_{max}/\rho_0)/(\Delta L_{max}/L_0), \quad (1)$$

where $\Delta\rho_{max}$ and ΔL_{max} are the maximum changes in resistivity and length, respectively. This coefficient characterizes the sensitivity of the resistivity of composites to stretching and can be used to compare data obtained for different composites [28–33]. As the filler concentration increases, the gauge factor first rises and then decreases. For the sample with 16 % graphene content, the gauge factor is $GF = 534.7$, for the sample with 24 % content – $GF = 2661$, and for the sample with 30% content – $GF = 340.4$. This can be explained by the fact that increasing the filler concentration leads to two different effects. The first of these is due to the fact that the range of stress in which the composite retains its conductive properties increases (see Fig. 2), which should lead to a decrease in the GF value. The opposite effect on the GF value is caused by the decrease in the resistivity of the composite in the absence of stress that occurs with an increase in the graphene concentration (see Fig. 1).

To initially estimate the mechanical properties of the synthesized composites, an assessment of their tensile strength, an analysis of the nature of the deformations developing in them under uniaxial static tension, and a study of their relaxation properties were carried out. The samples were repeatedly stretched with a successive increase in their extension in 10 mm increments until they failed. After each stretching step, the sample length L was measured 25 sec after the stress was removed. The obtained results are presented in Fig. 3 as the dependence of the ratio of the sample length L to its initial length L_0 on its relative elongation under stretching.

As seen from the presented data, the tensile strength decreases with an increase in filler content and is 565, 542, and 401 % for composites with graphene contents of 16, 24, and 30 %, respectively. Additionally, it is evident that none of the synthesized composite samples retain their initial length even under minimal applied stress, which indicates the occurrence the irreversible plastic deformations in them, along with elastic ones. The slope of the dependences shown in Fig. 3 increases consistently with increasing filler concentration, which suggests an increase in the proportion of irreversible deformations, i.e., the occurrence of the effect of their accumulation under repeated stretching of the sample.

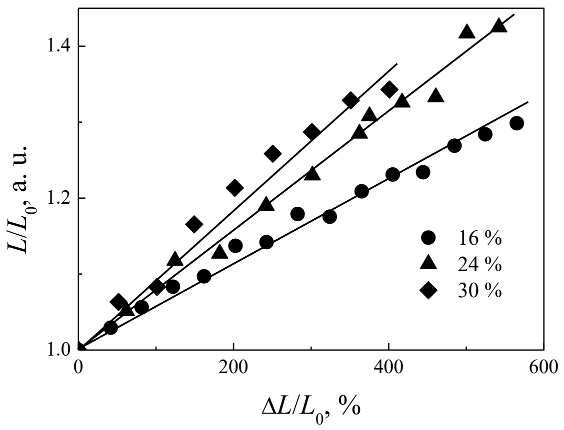

Fig. 3. The dependence of relative elongation of composite samples after stress removal on the degree of their stretching

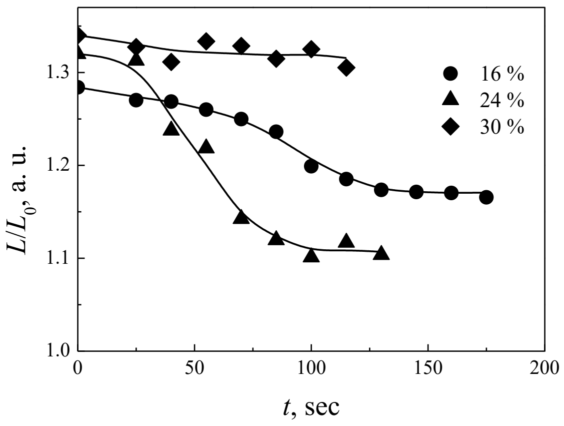

Fig. 4. The dependence of the relative change in the length of composites after stress removal on time

The results of the study of the relaxation properties of the synthesized composites after stress of 400 % are presented in Fig. 4. Composites of all studied compositions do not return to their original length even 2–3 min after the stress is removed. This can be due to the formation of jams of flat filler particles inside the polymer matrices during stretching [34–36]. The sample with 30 % graphene content practically does not relax at all, and the sample with 16 % graphene content relaxes, but rather weakly. The composite with 24 % graphene content demonstrates the best relaxation properties, both in terms of time and degree of relaxation. Thus, a filler concentration of 24 % is close to the critical point for the studied composites, above which the improvement of the mechanical properties is replaced by their deterioration. It should be noted that this result indirectly corresponds to the data obtained in [37] when analyzing the properties of composites based on polyvinyl acetate with graphene filler. The authors of [37] studied samples in a lower range of graphene concentration range than in our study (below its possible critical value), but found that a consistent increase in the graphene content in composites initially leads to a significant improvement in their mechanical properties, but then this improvement slows down.

Thus, the graphene content of 24 % is optimal for the studied SBR-based polymer composites, as it corresponds to both the maximum strain sensitivity and the best relaxation properties.

Conclusions

In this study, we investigated the electrical and mechanical properties of polymer composites based on styrene-butadiene rubber with variable content of graphene as a conductive filler. It was found that increasing the graphene concentration in the SBR matrix leads to an increase in the range of value of applied uniaxial stress in which the composite slightly changes its resistivity. A gradual increase in graphene concentration first leads to an increase in the sensitivity of composite resistivity to stretching, and then to a decrease. At the same time, the tensile strength of the composites decreases slightly, and repeated uniaxial stretching leads to the accumulation of plastic deformations preventing complete relaxation of the samples after deformation.

The maximum change in the resistivity under deformation was observed in the composite with a graphene concentration of 24 %, for which the value of the gauge factor reaches *GF* = 2661. This graphene concentration also corresponds to the best relaxation properties. The obtained data indicate the potential of using the synthesized composites for the development of sensor devices for flexible wearable electronics.

Conflict of interest

The authors declare that they have no conflict of interest.

References

1. Skiba NV. Deformation mechanism of bimodal metal-graphene composites with nanotwinned structure. *Materials Physics and Mechanics.* 2021;47(5): 676–680.
2. Zhang B, Sun F, Li Y, Wu D, Yang C, Wang Z, Gao J, Zhao G, Sun S. Changing the potassium-based activation path to prepare coal-based porous carbon with more graphitic or graphene structures for high-performance organic supercapacitors. *Carbon.* 2024;219: 118812.
3. Li W, Liu M, Cheng S, Zhang H, Yang W, Yi Z, Zeng Q, Tang B, Ahmad S, Sun T. Polarization independent tunable bandwidth absorber based on single-layer graphene. *Diamond and Related Materials*. 2024;142: 110793.
4. Kopac T. Evaluation of recent studies on electrochemical hydrogen storage by graphene-based materials: Impact of modification on overall effectiveness. *International Journal of Hydrogen Energy*. 2024;69: 777–803.
5. Fan X, He C, Ding J, Gao Q, Ma H, Lemme MC, Zhang W. Graphene MEMS and NEMS. *Microsystems & Nanoengineering.* 2024;10(1): 154.
6. Tonkov DN, Kobylyatskaya MI, Vasilyeva ES, Semencha AV, Gasumyants VE. Conductive properties of flexible polymer composites with different carbon-based fillers. *Journal of Physics: Conference Series.* 2022;2227(1): 012022.
7. Tonkov D, Vasilyeva E, Tolochko O, Gasumyants V. Conductive and tensoresistive properties of flexible polymer composites: Experimental study and modeling. In: *2024 International Conference on Electrical Engineering and Photonics (EExPolytech)*. Saint Petersburg: Russian Federation; 2024. pp. 410–413
8. Khayal A, Nami SAA. Next-generation wearable technologies: The rise of multifunctional hydrogels in smart electronics. *Journal of Cleaner Production.* 2025: 145265.
9. Li J, Zhao Y, Fan Y, Chen J, Gong J, Li WJ. Flexible wearable electronics for enhanced human-computer interaction and virtual reality applications. *Nano Energy.* 2025;138: 110821.
10. Li Y, Li Q, Yin H, Jiang C, Chen Y, Jiang L, Guo Y. Super P carbon nanoparticles activated hydrogel sensors for developing fully integrated wearable electronics via an in-situ writing strategy. *Chemical Engineering Journal.* 2025;508: 161060.
11. Bonato P. Advances in wearable technology and applications in physical medicine and rehabilitation. *Journal of NeuroEngineering and Rehabilitation.* 2002;2: 2.
12. Avcu E, Bastan FE, Guney M, Yildiran Avcu Y, Ur Rehman MA, Boccaccini AR. Biodegradable polymer matrix composites containing graphene-related materials for antibacterial applications: A critical review. *Acta Biomaterialia.* 2022;151: 1–44.
13. Das TK, Prusty S. Graphene-based polymer composites and their applications. *Polymer-Plastics Technology and Engineering.* 2013;52(4): 319–331.
14. Mousavi SM, Hashemi SA, Kalashgrani MY, Omidifar N, Bahrani S, Vijayakameswara Rao N, Babapoor A, Gholami A, Chiang W-H. Bioactive graphene quantum dots based polymer composite for biomedical applications. *Polymers.* 2022;14(3): 617.
15. Aroganam G, Manivannan N, Harrison D. Review on wearable technology sensors used in consumer sport applications. *Sensors.* 2019;19(9): 1983.
16. Chutturi M, Kelkar BU, Yadav SM, Wibowo ES, Bhuyar P, Naik BP, Sinha A, Lee SH. Exploring nanolignin as a sustainable biomacromolecule in polymer composites: Synthesis, characterization, and applications: A review. *International Journal of Biological Macromolecules.* 2025;304(2): 140881.

17. Wilson S, Laing R. (2018). Wearable technology: Present and future. In: *Proceedings of the 91st Textile Institute World Conference, Vol. 1*. Leeds: United Kingdom; 2018. p. 272–286.
18. Yang S, Larionova T, Kobykhno I, Klinkov V, Shalnova S, Tolochko O. Graphene-doped thermoplastic polyurethane nanocomposite film-based triboelectric nanogenerator for self-powered sport sensor. *Nanomaterials.* 2024;14(19): 1549.
19. Ma L, Lu W. Carbon nanotube film based flexible bi-directional strain sensor for large deformation. *Materials Letters.* 2020;260: 126959.
20. Ren J, Wang C, Zhang X, Carey T, Chen K, Yin Y, Torrisi F. Environmentally-friendly conductive cotton fabric as flexible strain sensor based on hot press reduced graphene oxide. *Carbon.* 2017;111: 622–630.
21. Trivedi DN, Rachchh NV. Graphene and its application in thermoplastic polymers as nano-filler – A review. *Polymer.* 2022;240: 124486.
22. Takei K, Takahashi T, Ho JC, Ko H, Gillies AG, Lew PW, Fearing RS, Javey A. Nanowire active-matrix circuitry for low-voltage macroscale artificial skin. *Nature Materials.* 2010;9: 821–826.
23. Xia Y, Zhang Q, Wu XE, Kirk TV, Chen XD. Practical and durable flexible strain sensors based on conductive carbon black and silicone blends for large scale motion monitoring applications. *Sensors.* 2019;19(20): 4553.
24. Shi G, Zhao Z, Pai J-H, Lee I, Zhan, L, Stevenson C, Ishara K, Zhang R, Zhu H, Ma J. Highly Sensitive, Wearable, Durable Strain Sensors and Stretchable Conductors Using Graphene/Silicon Rubber Composites. *Advanced Functional Materials.* 2016;26(42): 7614–7625.
25. Dalmas F, Dendievel R, Chazeau L, Cavaille J-Y, Gauthier C. Carbon nanotube-filled polymer composites. Numerical simulation of electrical conductivity in three-dimensional entangled fibrous networks. *Acta Materialia.* 2006;54(11): 2923–2931.
26. Phiri J, Gane P, Maloney TC. General overview of graphene: Production, properties and application in polymer composites. *Materials Science and Engineering: B.* 2017;215: 9–28.
27. Hu N, Karube Y, Arai M, Watanabe T, Yan C, Li Y, Liu Y, Fukunaga H. Investigation on sensitivity of a polymer/carbon nanotube composite strain sensor. *Carbon.* 2010;48(3): 680–687.
28. Chen Y, Li Y, Xu D, Zhai W. Fabrication of stretchable, flexible conductive thermoplastic polyurethane/graphene composites via foaming. *RSC Advances.* 2015;5(100): 82034–82041.
29. Shchegolkov AV, Shchegolkov AV, Kaminskii VV, Chumak MA. Polymer composites with nanomaterials for strain gauging: A review. *Journal of Composites Science.* 2025;9(1): 8.
30. Zhang W, Piao S, Lin L, Yin Y, Guo J, Jiang Z, Cho Y, Li R, Gao J, Pang H, Piao Y. Wearable and antibacterial HPMC-anchored conductive polymer composite strain sensor with high gauge factors under small strains. *Chemical Engineering Journal.* 2022;435(2): 135068.
31. Zhang Y, Zhang Y, Deng W, Li Q, Guo M, Chen G. Skin-like soft thermoelectric composites with a “J-shaped” stress–strain behavior for self-powered strain sensing. *Advanced Functional Materials.* 2025;35(14): 2420644.
32. Costa P, Nunes-Pereira J, Oliveira J, Silva J, Moreira JA, Carabineiro SAC, Buijnsters JG, Lanceros-Mendez S. High-performance graphene-based carbon nanofiller/polymer composites for piezoresistive sensor applications. *Composites Science and Technology.* 2017;153: 241–252.
33. Yang H, Yao X, Zheng Z, Gong L, Yuan L, Yuan Y, Liu Y. Highly sensitive and stretchable graphene-silicone rubber composites for strain sensing. *Composites Science and Technology.* 2018;167: 371–378.
34. Ader F, Sharifzadeh E, Azimi N. Unveiling the impact of percolation phenomenon on the microstructure of polymer nanocomposites based on the combination of scaling and percolation theories. *Polymer Composites.* 2024;45(17): 16280–16292.
35. Yusuf J, Sapuan SM, Rashid U, Ilyas RA, Hassan MR. Thermal, mechanical, thermo-mechanical and morphological properties of graphene nanoplatelets reinforced green epoxy nanocomposites. *Polymer Composites.* 2024;45(3): 1998–2011.
36. Lee SJ, Yoon SJ, Jeon I-Y. Graphene/polymer nanocomposites: Preparation, mechanical properties, and application. *Polymers.* 2022;14(21): 4733.
37. Zhang Q, Chen D. Enhanced mechanical properties of graphene-based poly(vinyl alcohol) composites. *Macromolecules.* 2010;43(5): 2357–2363.