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Segregated Network Polymer Composites with High Electrical Conductivity and Well Mechanical Properties based on PVC, P(VDF-TFE), UHMWPE, and rGO

Kseniya A. Shiyanova, Maksim V. Gudkov, Arkady Ya Gorenberg, Maxim K. Rabchinskii, Dmitry A. Smirnov, Maria A. Shapetina, Tatiana D. Gurinovich, Galina P. Goncharuk, Demid A. Kirilenko, Sergey L. Bazhenov, and Valery P. Melnikov*



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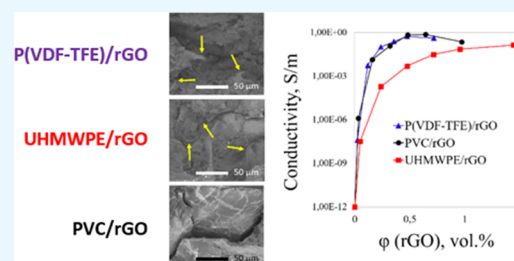


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ABSTRACT: The formation of a segregated network structure (wittingly uneven distribution of a filler) is one of the most promising strategies for the fabrication of electrically conductive polymer composites at present. However, the simultaneous achievement of high values of electrical conductivity with the retention of well mechanical properties within this approach remains a great challenge. Here, by means of X-ray photoelectron spectra (XPS), near-edge X-ray absorption fine structure (NEXAFS) spectra, scanning electron microscopy (SEM), dielectric spectroscopy, and compression engineering stress–strain curve analysis, we have studied the effect of a segregated network structure on the electrical conductivity and mechanical properties of a set of polymer composites. The composites were prepared by applying graphene oxide (GO) with ultralarge basal plane size (up to 150 μm) onto the surface of polymer powder particles, namely, poly(vinyl chloride) (PVC), poly(vinylidene fluoride-co-tetrafluoroethylene) (P(VDF-TFE)), and ultrahigh-molecular-weight poly(ethylene) (UHMWPE) with the subsequent GO reduction and composite hot pressing. A strong dependence of the segregated network polymer composites' physical properties on the polymer matrix was demonstrated. Particularly, 12 orders of magnitude rise of the polymers' electrical conductivity up to 0.7 S/m was found upon the incorporation of the reduced GO (rGO). A 17% increase in the P(VDF-TFE) elastic modulus filled by 1 wt % of rGO was observed. Fracture strength of PVC/rGO at 0.5 wt % content of the filler was demonstrated to decrease by fourfold. At the same time, the change in strength was not significant for P(VDF-TFE) and UHMWPE composites in comparison with pure polymers. Our results show a promise to accelerate the development of new composites for energy applications, such as metal-free supercapacitor plates and current collectors of lithium-ion batteries, bipolar plates of proton-exchange membrane fuel cells, antistatic elements of various electronic devices, etc.



INTRODUCTION

One of the most important tasks of modern materials science is the production of electrically conductive polymer composite materials (ECPCM). These materials with high electroconductive and antistatic characteristics based on industrial polymers have been widely used to solve a number of technical problems in recent years.¹ Compared with metal-based composite materials, ECPCM have several advantages, such as easy molding and processing, low density, a wide range of conductivity values, corrosion resistance, and so on. ECPCM are used in electronics, radio equipment, for removing static electricity, etc. Such composites are usually filled with conducting copper, silver, and carbon particles.¹

One of the most promising electrically conductive fillers is graphene that possesses an attractive combination of low price, high conductivity, and mechanical strength due to its two-dimensional (2D) structure.^{2–7} The synthesis of graphene is cheap if particles are made by the reduction of graphene oxide

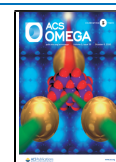
(GO).⁸ It is known that GO can be reduced by heat treatment^{9–11} or by exposure to chemical reducing agents, the most common of which is hydrazine.^{12–17} The reduction process allows us to convert GO from a dielectric into an electrically conductive material (rGO), which is graphene with a large number of structural defects and residues of oxygen-containing groups.⁸

In this paper, we investigate ECPCM with a segregated network structure in which the conductive filler has been located only at the interface between the polymer matrix particles.^{18,19} This approach for the composite material

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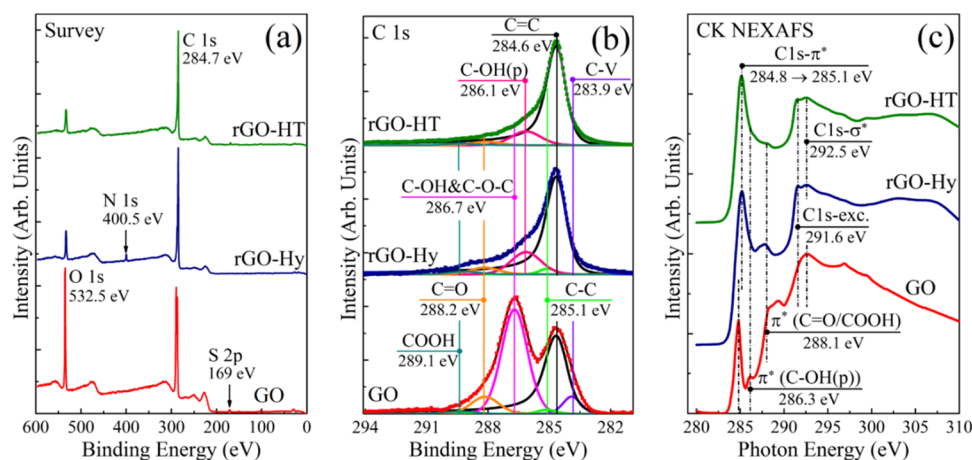


Figure 1. (a) Survey and (b) high-resolution C 1s XPS spectra and (c) C K-edge NEXAFS spectra of the initial GO and GO reduced by hydrazine treatment (rGO-Hy) and thermal annealing (rGO-HT).

production is promising since despite the small amount of electrically conductive filler used, it allows us to achieve high values of electrical conductivity.²⁰ To obtain a segregated network structure composite, it is necessary to apply a thin coating of an electrically conductive filler on polymer powder particles, and then subject the powder to hot pressing to form a unified electrically conductive network into the bulk of composite material.

Polymer/rGO composites with a segregated network structure have been developed and studied in some works.^{21–24} In some of them, GO and a polymer powder were dispersed in a water/alcohol solution and then GO was reduced by hydrazine directly in the mixture^{21,24} followed by evaporation of the liquid phase and hot pressing of a coated polymer powder. In the other one, the authors used preliminarily reduced graphene oxide, which was dispersed in alcohol by ultrasonic treatment and evaporated in the presence of the polymer powder.²² The thermal reduction of graphene oxide approach is also used to produce such composites. Poly(vinylidene fluoride) polymer well wetted by water and thermally stable at 200 °C was used to obtain polymer/rGO composites with a segregated network structure.²³ Such an approach ensures the immobilization of GO monolayer particles on the surface of polymer powder particles and allows in situ thermal reduction of GO. The disadvantage of this approach is the fact that in the process of GO reduction, a rather large amount of gaseous products is released into the atmosphere²⁵ directly during hot pressing, including CO, which is a limiting factor for using this method as the main one for the production of such materials. In addition, the minimum temperature of effective GO reduction is 180–250 °C,²⁶ which is too high for most of the polymers.

One of the most attractive feature is the approach that utilizes the formation of a polymer powder coated with graphene oxide, which can be converted into an electrically conductive material by the treatment with hydrazine vapor at room temperature.²⁰ This approach allows us to immobilize the initial monolayer GO flakes on the surface of the polymer powder particles, convert them into an electrically conductive form, and then press the powder into a sample of the required shape and size at a temperature corresponding to the optimum value for a particular polymer. The absence of reduction heat treatment can significantly expand the number of polymers that can be used in the framework of the proposed approach.

In this paper, we obtained ECPCM with a segregated network structure based on the GO reduced by hydrazine at moderately elevated temperature and set of polymers, explored the structure features of obtained composites, and investigated in detail the effect of a polymer matrix from poly(vinyl chloride) (PVC), poly(vinylidene fluoride-*co*-tetrafluoroethylene) (P(VDF-TFE)), and ultrahigh-molecular-weight poly(ethylene) (UHMWPE) on the electrically conductive properties. Also, we have conducted the study of mechanical properties of obtained ECPCM with a segregated network structure, which, despite the abundance of work in this direction, had been investigated superficially. The results of this study are incredibly important for understanding the possibilities of using such composites to develop electrically conductive elements for energy application based on polymer composites, such as metal-free supercapacitor plates, current collectors of lithium-ion batteries, bipolar plates of proton-exchange membrane fuel cells, antistatic elements of various electronic devices, and many others.

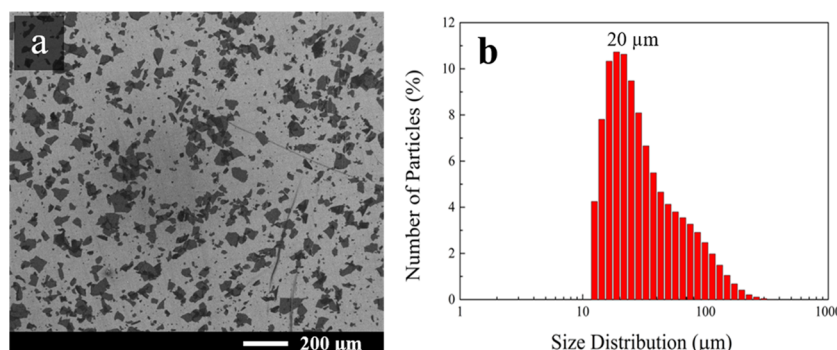
RESULTS AND DISCUSSION

Chemistry of GO and rGO Fillers. The chemical composition of GO and rGO-Hy was analyzed using X-ray photoelectron spectra (XPS) and near-edge X-ray absorption fine structure (NEXAFS) techniques. For comparison of differences in the chemical composition, the spectra of rGO obtained traditionally by thermal reduction at $T = 550$ °C (rGO-HT) are also shown in Figure 1. Figure 1a shows the survey XPS spectra of the initial GO (red curve) and rGO samples obtained via hydrazine treatment (blue curve) and high-temperature annealing (green curve). The obtained spectra are dominated by C 1s and O 1s lines with the absence of any prominent features related to any contaminants. Only almost indiscernible S 2p peak is detected in the GO spectrum, indicating the presence of about 0.5–1 atom % of the sulfur species retained after the synthesis. After the GO reduction, the O 1s peak substantially decreases for both rGO-Hy and rGO-HT samples, evidencing the elimination of the oxygen-containing functionalities.

To obtain more detailed information on the composition of the functional groups in the studied materials, high-resolution C 1s spectra were measured and analyzed (Figure 1b). The spectra were fitted using the set of seven Gaussian–Lorentzian sum peaks (Gaussian by 70% and Lorentzian by 30%) and an

Table 1. Relative Concentration of Functional Groups and the C/O Ratio of GO and rGO Samples Determined by Deconvolution of the Corresponding C 1s XPS Spectra

component	C–V	C=C	C–C	C–OH and C–O–C/C–OH(p)	C=O	COOH	C/O ratio
binding energy (eV)	283.9	284.7	285.1	286.7	288.2	288.9	
GO	0.078	0.361	0.103	0.406	0.040	0.012	2.12
rGO-Hy	0.037	0.847	0.018	0.070	0.016	0.012	10.2
rGO-HT	0.027	0.889	0.000	0.059	0.017	0.008	11.8

**Figure 2.** SEM images of GO particles (a) and size distribution (b) according to both laser diffraction and SEM analysis.

asymmetric Doniach-Sunjić function (DS). The peaks at 283.9 eV (C–V), 284.7 eV (C=C), and 285.1 eV (C–C) are related to the carbon atoms in the unfunctionalized areas of the graphene network.^{27,28} The origin of the C–V peak is indeterminate, with it being discussed to be related to either carbohydrate groups or carbon atoms near to the vacancy defects and holes.²⁹ After the reduction, this component diminishes, suggesting that it is more probably related to the presence of C–H and C–H₂ functionalities. Asymmetry of the C=C peak arises due to the π -conjugation in the graphene network and increases along with the GO reduction due to the restoration of the perfect graphene lattice.³⁰

The other four peaks positioned at 286.1, 286.7, 288.2, and 289.1 eV correspond to various oxygen functionalities: phenols (C–OH(p)), hydroxyls and epoxides (C–OH and C–O–C), carbonyls (C=O), and carboxyls (COOH), respectively. The C 1s XPS spectrum of the initial GO is dominated by the C–OH and the C–O–C peaks, assuming high oxidation of the obtained material. The quantitative analysis of the fitted spectra demonstrates that the C/O ratio of the initial GO is of about 2.12 (Table 1). The C–OH and the C–O–C peaks drastically decrease after the reduction along with the C=O and COOH peaks, with the rise of the C/O ratio to 11.8 and 10.2 for rGO-Hy and rGO-HT, respectively. These values are very close to those of rGO commonly prepared by various reduction methods. At the same time, the presence of the C–OH(p) peak in both C 1s spectra of the rGO-Hy and rGO-HT indicates retention of a certain amount of phenols, known to be highly stable in the reduction processes,³¹ in the obtained materials.

The successful restoration of the graphene network and elimination of the oxygen-containing groups were additionally verified by NEXAFS spectra (Figure 1c). As seen, after the treatment of the peak at 284.8 eV, corresponding to C 1s– π^* in the carbon atoms of the pure graphene,³² it becomes more prominent and shifts to higher energies, 285.1 eV. Simultaneously, the peak at 291.6 eV, corresponding to the excitonic states in the restored sp²-conjugated graphene network,³³ arises, whereas spectral features related to the carboxyls and carbonyls (288.1 eV) decrease and further disappear.

In the case of rGO-Hy, the N 1s peak also appears, indicating nitrogen doping of the obtained material during the reduction using hydrazine treatment.³⁴ The concentration of the incorporated nitrogen is estimated to be around 4.3 atom %. To determine the form of the nitrogen species presented in the sample, high-resolution N 1s XPS and N K-edge NEXAFS spectra were measured (Figure S1).

SEM Analysis of GO- and rGO-Coated Polymer Powders. Scanning electron microscopy (SEM) analysis of graphene oxide images on a silicon substrate and laser diffraction (Figure S2, Table S1) of water dispersion allow us to talk about ultralarge-size basal plane (10–150 μ m) of GO particles in the dispersion with a predominant content of 20–100 μ m (Figure 2). Note, the SEM analysis demonstrates the presence of a large number of particles with the lateral sizes smaller than 5 μ m. These particles are not diagnosed via laser diffraction measurements, where the collected size distributions demonstrate a sharp decrease at the small particles' sizes. We attribute this to the features of the diffraction pattern processing within the frame of the mathematical model used in the laser diffraction particle analysis. The smaller particles show the less resolved scattering pattern with broadband maxima in the range of 0–90° angles.³⁵ In the range of lateral sizes of less than 1 μ m, the scattering pattern becomes a smooth curve without any detectable features. Hence, in the diffraction patterns of polydisperse systems such as GO suspensions, it is extremely difficult to detect the presence of small particles, even in the multimodal model. Flakes of both GO and rGO-Hy exhibit high structural quality without any holes and defects, which are determined by the means of TEM imaging (Figure S3a,b). Selected-area electron diffraction patterns and atomic force microscopy (AFM) imaging (Figure S3) demonstrate that the monolayer content of GO was used.

SEM images of P(VDF-TFE), UHMWPE, and PVC particles coated with rGO are shown in Figure S4. Note the quality of the P(VDF-TFE)/rGO composite. The particles are fibrous in shape. However, the coated surface structure is rather uniform, without high number of defects (Figure S4b,c). This is explained by polar polymer C–F bonds interacting with polar GO prior to the reduction. PVC particles are rather

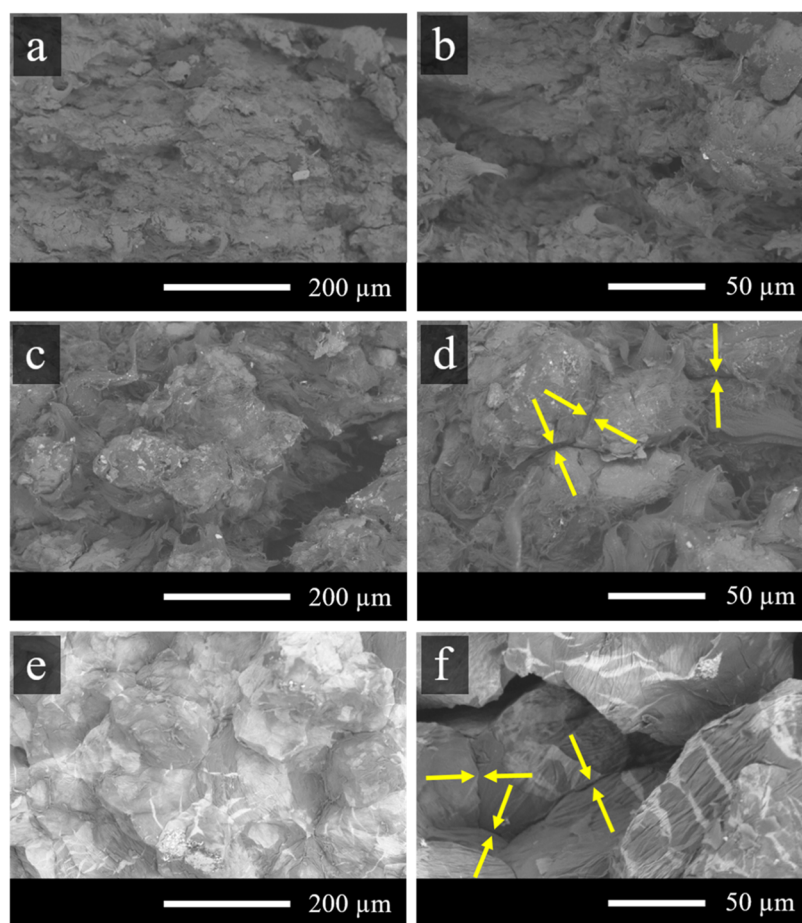


Figure 3. SEM images of brittle fractures of composite samples: (a, b) P(VDF-TFE)/rGO, (c, d) UHMWPE/rGO, and (e, f) PVC/rGO with 1 wt % of rGO content.

spherical and also uniformly coated by rGO sheets (Figure S4h,i). The C–Cl bond is also quite polar, which provides excellent interaction with the GO planes. Cracks, rGO layers without adhesion to the polymer particles, and uncoated pure polymer zones are observed for UHMWPE (Figure S4e,f). Despite this, UHMWPE particles are simpler in shape and surface morphology than P(VDF-TFE); the absence of polar bonds in the molecules of this polymer does not allow for a satisfactory quality of the coating. The globules characteristic of the UHMWPE structure is clearly visible in Figure S3f. Structural features characteristic of PVC and P(VDF-TFE) were not detected in SEM images, which indicates the superior quality of the rGO coating.

The thickness of the rGO layer was estimated, assuming the spherical shape of polymer particles. The volume fraction of a graphene-like layer is

$$\varphi = \frac{4\pi R^2 h}{4\pi R^3 / 3} = \frac{3h}{R}$$

where h is the thickness of the rGO layer and R is the polymer particle radius. The thickness of the rGO layer between polymer particles is double the value determined by the above equation, and the thickness of rGO in the composite is

$$2h = \frac{2}{3}VR$$

where V is the volume fraction of rGO. For $R = 100 \mu\text{m}$ and $V = 0.5\%$, the thickness of the rGO layer is estimated as $0.33 \mu\text{m}$.

SEM images of brittle fractures of P(VDF-TFE)/rGO, UHMWPE/rGO, and PVC/rGO composites are shown in Figure 3. It can be seen that the fracture structures of the UHMWPE/rGO and PVC/rGO composites are granular, which indicates the segregation of polymer particles by rGO layers, which is also clearly seen in the images of the general plan of fractures (Figure S5a–c). The interfaces of the polymer granules with the rGO layers are marked with yellow arrows, as shown in Figure 3d,f. The P(VDF-TFE)/rGO pattern is quite different. The structural units that are characteristic of the segregated structure are not obvious imaged. This indicates the formation of a finer structure upon pressing the sample due to the highly developed surface and fibrous shape of the initial P(VDF-TFE) polymer powder.

Electrical Conductivity of Polymer/rGO Composites.

Figure 4 shows electrical conductivity plotted against the volume fraction of the rGO. All composites show a 10^4 – 10^{12} -fold increase in conductivity in comparison with that of a pure polymer. The conductivity increased to 10^{-4} S/m at $\varphi = 0.5 \text{ wt } \%$, and the maximum conductivity for PVC and P(VDF-TFE) composites is at $\varphi = 2 \text{ wt } \%$ (0.7 and 0.5 S/m, respectively). The increase in the rGO content leads to the layer thickening, and the quality of coating of particles decreases, cracks and chips become more frequent, as well as places of coating with a loose (expanded) structure (Figure S6). The thicker the coating film, the lower the diffusion velocity of released

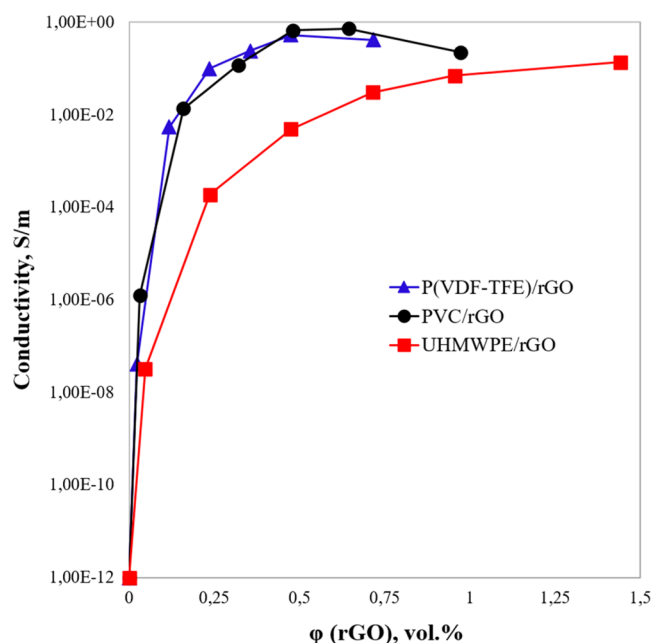


Figure 4. Electrical conductivity plotted against the volume fraction of the rGO in composites.

gaseous products of GO reduction. Violation of the uniformity of the coating leads to a breach of the electrically conductive network unity during a hot pressing procedure of the coated powder into the composite material. This leads to a slowdown in the growth of electrical conductivity at filling from 0.25 to 0.5 vol % for PVC/rGO and P(VDF-TFE)/rGO composites and a further decrease in values with an increase in the rGO content. In case of UHMWPE/rGO, the rGO coating initially has a large number of defects, which provides fewer diffusion difficulties for the release of gaseous products during the reduction of graphene oxide, and determines the value of electrical conductivity.

The conductivity versus the rGO volume fraction curves for P(VDF-TFE)/rGO and PVC/rGO practically coincide, and the curve for UHMWPE/rGO is lower. The lower electrical conductivity of UHMWPE/rGO is explained by the difference in the chemical composition. In the set C–H, C–Cl, and C–F, the polarity of the bond increases, and, consequently, the adhesion with GO in the consequence UHMWPE–PVC–P(VDF-TFE) these groups should also increase because GO is

a strongly polar compound due to the presence of oxygen-containing groups. UHMWPE does not have polar groups, and graphene oxide has weaker interaction with the surface of the polymer, which reduces the quality of the rGO layer and, thus, electrical conductivity in comparison with PVC and P(VDF-TFE) at fixed rGO content.

Mechanical Properties of Polymer/rGO Composites with a Segregated Network Structure. Figure 5 shows the elastic modulus plotted against the volume fraction of rGO. Elastic modulus for all composites changes insignificantly. For composites with PVC and UHMWPE, the elastic modulus decreases insignificantly. In contrast, in P(VDF-TFE)/rGO it increases. This may be explained by the fibrous shape of P(VDF-TFE) particles, which provides the formation of a finer structure. The probability of rGO penetration into the bulk of the polymer during the hot pressing is much higher than with the spherical shape of the polymer particles. This contributes to the formation of new polymer–polymer-type contacts and the construction of the interpenetrating network structure. The polymer may be reinforced with rGO particles in this case.

Figure 6 shows the compressive strength plotted against the volume fraction of rGO particles. The strength of P(VDF-TFE)/rGO and UHMWPE/rGO slightly decreases with an increase in rGO volume content. However, for PVC/rGO, even at a filler content of $\varphi = 0.5$ wt %, a fourfold decrease in strength is observed. In contrast to pure PVC, the composite is rather brittle (Table 2). Evidently, the composite is weak in the plane of graphene particles and this causes shear failure. As a result, the failure strain of the composite decreases (Table 2). This effect for PVC is explained by the almost complete absence of the formation of polymer–polymer contact boundaries formed during the pressing of the sample. The SEM images of fractures of P(VDF-TFE)/rGO and UHMWPE/rGO composites clearly show the sites of rupture of such polymer–polymer boundaries, marked with yellow arrows, as shown in Figure S5d–f and Figure 3. In the case of PVC/rGO, the destruction of the composite occurs absolutely brittly along the rGO interfaces with no clear signs of polymer–polymer rupture sites.

For ductile UHMWPE and P(VDF-TFE) matrices with strong strain hardening, the material remains ductile despite the presence of weakness in the planes of rGO particles, and the yield stress decreases insignificantly. Still, the mechanical properties of composites are determined by not too high adhesion of neighboring graphene particles. Evidently, the

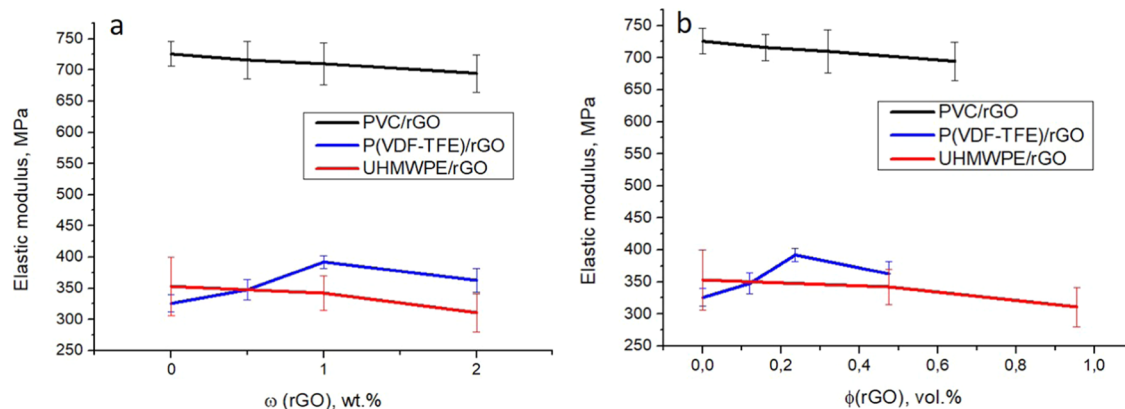


Figure 5. Elastic modulus of polymer/rGO composites at compression plotted against weight (a) and volume (b) content of rGO.

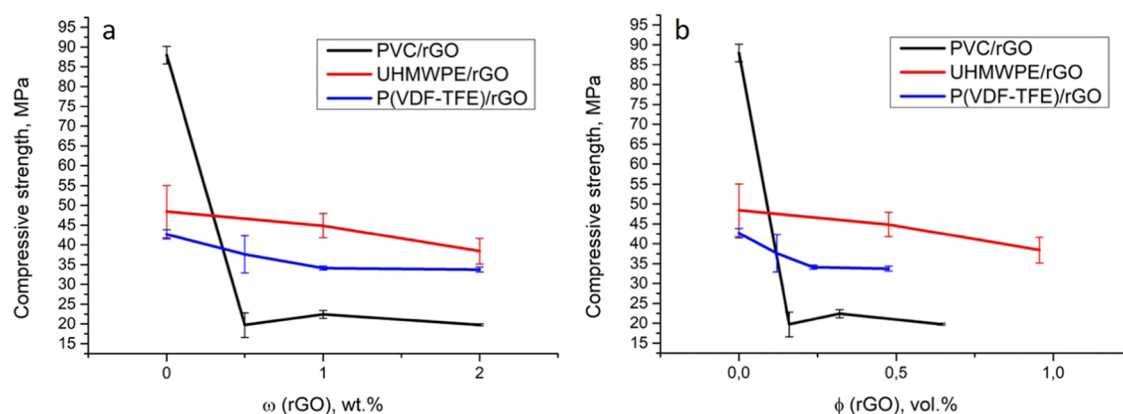


Figure 6. Compressive strength of polymer/rGO composites plotted against weight (a) and volume (b) content of rGO.

Table 2. Value of the Main Parameters during the Compression Test of Composite Samples

material	$\omega(\text{rGO})$, wt %	$\phi(\text{rGO})$, vol %	max. load, N	max. pressure, MPa	failure strain, %
PVC/rGO	0	0	1727 ± 34	87.9 ± 2.2	17.0 ± 1.4
	0.5	0.12	388 ± 38	19.7 ± 3.1	2.8 ± 0.2
	1	0.24	470 ± 43	34.8 ± 8.8	5.7 ± 1.2
	2	0.48	408 ± 26	20.8 ± 1.3	4.7 ± 0.7
UHMWPE/rGO	0	0	739 ± 39	116.2 ± 20.1	44.7 ± 2.7
	1	0.48	854 ± 10	92.1 ± 9.2	47.2 ± 5.0
	2	0.96	700 ± 10	47.8 ±	36.6 ± 5.5
P(VDF-TFE)/rGO	0	0	760 ± 60	44.7 ±	49.2 ± 2.6
	0.5	0.16	738 ± 60	37.6 ± 4.7	36.3 ± 3.3
	1	0.32	708 ± 47	36.2 ± 2.0	43.9 ± 2.8
	2	0.64	672 ± 22	34.2 ± 1.1	47.8 ± 2.3

plane of rGO particles is a weak shear plane, and this causes some decrease in composite strength.

CONCLUSIONS

The effect of polymer matrices on electrical conductivity and mechanical properties of polymer/rGO composites with a segregated network structure was investigated. For a nonpolar UHMWPE matrix, the electrical conductivity at fixed filler content was lower than that in composites with polar PVC and P(VDF-TFE). However, with all matrices, the pressing of a powder coated with rGO proposed approach provides an increase in conductivity by at least 8 orders of magnitude compared to a pure polymer already at 0.25 vol % fillings. It has been shown that already 0.5 wt % of the filler content has a critical effect on the glassy polymer matrix PVC; the strength decreases by 78% compared with the pure polymer, and the failure strain decreases from 17 to 2.8%. For the P(VDF-TFE) matrix, a filler content of 1 wt % leads to an increase in the elastic modulus by 17% and a reinforcing effect is observed. The mechanical properties of segregated structure composites based on UHMWPE changed insignificantly over the entire interval of filler content. The examined approach to the production of electroconductive composites with a segregated network structure is universal, which can be applied to a wide range of different polymers, and the data presented in this work will expand the understanding of the possibilities of using such materials in various manufacturing industries.

EXPERIMENTAL SECTION

Materials and Methods. Poly(vinyl chloride) (trade mark PVCS-7059M), poly(vinylidene fluoride-co-tetrafluoroethy-

lene) (trade mark F-42V), and ultrahigh-molecular-weight poly(ethylene) (UHMWPE) GUR-4120 (Celanese Corporation) with a molecular weight of 4.5 million Da were used as polymer matrices. Isopropyl alcohol (chemical grade) was used as a dispersant. A dispersion of graphene oxide with C = 12 mg/mL was provided by GraphOx company (www.graphox.ru).

The X-ray photoelectron spectra (XPS) and near-edge X-ray absorption fine structure (NEXAFS) spectra were obtained at the Russian-German beamline of an electron storage ring BESSY II (Helmholtz-Zentrum Berlin). The samples for the XPS and NEXAFS measurements were prepared via drop-casting of the studied GO or rGO suspension onto the surface of silica wafers with the formation of ~100 nm thick uniform films. The excitation energy in the case of survey XPS spectra was 736 eV, whereas C 1s and N 1s XPS spectra were obtained with the radiation energy of 850 eV. The quantification and curve fitting of the obtained XPS spectra were performed using standard CasaXPS software. The shift of the GO high-resolution C 1s and N 1s XPS spectra, arising due to the effect of the surface charging, was corrected by the alignment of the measured spectra with respect to the ones obtained for well-conducting rGO films. The position of the latter ones was corrected according to the position of the Au 4f_{7/2} line (84.0 eV). The NEXAFS spectra measurements were performed in the total electron yield (TEY) mode realized by sweeping the incident photon energy and simultaneously recording the sample drain current. The thus-obtained TEY XAS spectra were then subjected to appropriate normalization and smoothing.

The surface of the polymer particles with an rGO layer was studied using a JSM-7001F high-resolution scanning electron

microscope. Basal particle size in GO dispersion was also studied with a JSM-7001F microscope and a laser diffraction particle size analyzer, Malvern Mastersizer 2000.

To analyze the structure quality of GO flakes, the transmission electron microscopy (TEM) and the corresponding electron diffraction studies were performed with a point-to-point resolution of 0.19 nm at an accelerating voltage of 200 kV (Jeol JEM-2100F, Japan).

The atomic force microscopy of GO flakes was carried out by a Solver P47 AFM instrument (NT-MDT, Zelenograd, Russia) in the semicontact mode on a silicon substrate at a scan rate of 1 Hz.

The electrical conductivity of composites was measured with a wide-range BDS-40 dielectric spectrometer (Novocontrol). The diameter and the thickness of the test samples were 12 and 2 mm, respectively.

For mechanical compression tests, cylindrical samples with a diameter of 5 mm and a height of 10 mm were prepared. Rod samples were compressed in a Shimadzu AGS machine.

Preparation of Segregated Network Structure Composites. Polymer particles were coated by graphene oxide. With this aim, GO water dispersion was diluted to a concentration of 3 mg/mL, the calculated amount of which was mixed with isopropyl alcohol (10:1 relative to the water) and a polymer powder followed by the evaporation of the liquid phase on a rotary evaporator. The obtained material was dried in air at a temperature of 60 °C for 12 h.

After coating, the powder was treated with hydrazine vapors for 2 h at a temperature of 100 °C so that graphene oxide was reduced to graphene-like form (rGO-Hy). The reduction conditions were selected in a way that the GO reduction degree was maximum since the electrical conductivity of composites directly depends on this parameter. As was shown, the degree of GO reduction by hydrazine depends on the temperature³⁶ and the high efficiency occurs at 100 °C. Then, the disks with a diameter of 12 mm and a thickness of 2 mm were hot pressed. Composites with P(VDF-TFE), PVC, and UHMWPE were pressed with the required pressures and temperatures: 200 °C and 140 kg/cm², 120 °C and 300 kg/cm², 185 °C and 100 kg/cm², respectively.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsomega.0c02859>.

High-resolution N 1s and N K-edge NEXAFS spectra of the rGO-Hy sample (Figure S1); the particle size distribution in GO water dispersion according to laser diffraction (Figure S2); laser diffraction data of GO particle size distribution measurements (Table S1); TEM images and corresponding SAED patterns of the GO and rGO-Hy samples, the typical AFM image of the GO sample, and the topological profile (Figure S3); SEM images of polymer powders coated with 1 wt % of the rGO (Figure S4); SEM images of the brittle fracture general plan of composite samples and respective fault zones at higher magnification (Figure S5); SEM images of polymer powders coated with 2 wt % of rGO (Figure S6) (PDF)

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Notes

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