Research Article

Dielectric Properties of Epoxy-Matrix Composites with Tungsten Disulfide Nanotubes

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Addition of conductive nanotubes to an insulating polymer matrix has been proven as an efficient strategy that can improve the electromagnetic shielding performance, due to the high aspect ratio of nanotubes. Herein, a set of epoxy-matrix composites filled with 0.15-1.6 vol% of tungsten disulfide (WS_2) nanotubes being of 30-120 nm in diameter and 5-20 μm in length has been produced. Electromagnetic properties of the prepared composites were probed in the frequency range from 20 Hz to 1 MHz in a temperature range from 250 K to 500 K. Broadband properties of these materials are controlled by the dynamics of epoxy resin molecules, and no electrical percolation was observed up to the highest concentration (1.6 vol%) of WS_2 nanotubes. The value of dielectric permittivity for all composites is not bigger than 6 at room temperature and 1 kHz frequency, and the electrical conductivity of composites is about 10^{-6} S/m at 500 K, which demonstrate that the composites are suitable for antistatic applications at higher temperatures. The relaxation time follows the Vogel-Fulcher law, and the Vogel temperature T_0 has the minimum for the WS_2 nanotube concentration 0.15 vol%. Above 410 K, the electrical conductivity determines the properties of the investigated composites due to nonzero electrical conductivity of epoxy resin. The value of DC electrical conductivity for pure epoxy at $T = 450$ K is 0.3 μS/m, while the DC conductivity of the composites slightly increases with the WS_2 concentration. Therefore, the electrical contacts between WS_2 nanotubes and polymer matrix are rather ohmic. Additionally, the activation energy is almost independent on the concentration of WS_2. However, it is higher in composites than in pure epoxy resin.

1. Introduction

Nanoparticles are very interesting objects for investigations due to the possibility to control properties at the nanoscale and to find new physical phenomena. The nanotubes are a very important type of nanoparticles, due to the high aspect ratio of these structures. The nanotubes are typical for all layered materials, including tungsten disulfide (WS_2). Recently, it was demonstrated that WS_2 nanotubes and composites with these inclusions can be applied in various fields, including lithium ion batteries [1], bone tissue engineering [2], ultrafast photonics [3], and solar cells [4]. A lot of papers was already published on mechanical [5] and tribological [6] applications of WS_2 nanotubes and composites with these inclusions [7–12].

Moreover, due to the needle-like shape of WS_2 nanotubes and the appearance of chemical bonds between WS_2 nanotubes and polymer matrix, preparation of composites with these inclusions should be simpler than the carbon-based polymer composite preparation [13, 14]. Chemical bonds between WS_2 nanotubes and polymer matrix and unique mechanical and thermal properties of WS_2 nanotubes [5] are responsible for improved mechanical behavior and thermal stability of polymer composites [14]. Moreover, the

functionalization of WS_2 for obtaining composites in various polymer matrices is not needed [15]. This is due to unique chemical properties of the WS_2 nanotubes surface.

Despite the fact that the electrical conductivity of WS_2 nanotubes is about 10^3 S/m [16, 17] and these nanotubes can be used in transistors, photodetectors, and other electronic devices [2, 18, 19], the amount of publications on electromagnetic properties of polymer composites with WS_2 nanotubes is still very small [20–22]. Therefore, being of nanosize, having excellent mechanical and electrical properties, and possessing good dispersion and adhesion to polymers, inorganic nanotubes of WS_2 could be a good candidate for broadband electromagnetic composite applications.

In the earlier studies, it was established that the percolation threshold in WS_2/epoxy composites can be close to 25 vol% [20] and in polyurethane/WS_2 composites-larger than 2 wt% [21]. No electrical percolation for polyvinylidene fluoride (PVDF) composites with WS_2 was observed in reference [20]; however, in this work was established that pellets of WS_2 nanotubes are quite conductive. Such high values of electrical percolation, established in the previous works, are quite surprising, because of the WS_2 nanotube shape comparable to one of the carbon nanotubes, where percolation threshold in epoxy resin can be 0.0025 wt% [23]. Namely, WS_2 nanotubes have a high aspect ratio (30-120 nm in diameter and 5-20 μm in length [13]); therefore, the electrical percolation in composites with WS_2 nanoinclusions should be low enough, while electromagnetic properties of these composites are quite high. The comparison with carbon nanotube composites [24], where a wide range of percolation thresholds was observed depending on composite preparation technology, suggests that the composite with WS_2 nanotube preparation technology and WS_2 nanotube dispersion inside the polymer matrix should be important for electrical percolation in WS_2 nanotube composites as well. So, the open question was as following: is it possible to make the percolation threshold value in WS_2 nanotubes as low as in carbon nanotube composites? Other challenging tasks were to study the dielectric properties of composites with WS_2 nanotubes and the properties of conductive polymers modified with WS_2 nanoparticles. For example, to investigate an impact of WS_2 nanotubes on the electromagnetic properties of epoxy-matrix composites in a wide temperature and frequency ranges. These questions are crucial for further development of the WS_2 composite engineering and promising applications. Therefore, this paper is focused on the above questions, answers to which were not given hitherto. In order to address these points, an investigation of the broadband (20 Hz–1 MHz) electromagnetic properties of WS_2/epoxy-matrix composites in the broad temperature range (250 K–500 K) presented.

2. Materials and Methods

WS_2 nanotubes were produced through the chemical reaction of WO_{3-x} nanoparticles with H_2S and H_2 gases at high temperatures. Actually, the process consists of two main parts: formation of suboxide whiskers, by which majority is 5-20 μm in length and 30-120 nm in diameter, and their sub-

sequent sulfidization into WS_2 nanotubes. More details about the WS_2 nanotube preparation mechanism are in [13]; according to the formation mechanism of nanotubes, the sizes of WS_2 nanotubes repeat those of suboxide whiskers, being in average of 20 μm in length and 60 nm in diameter.

As synthesized, these nanotubes at different concentrations (0.15, 0.3, 0.94, and 1.6 vol%, which is corresponding to 0.5, 1, 3, and 5 wt%) were dispersed in epoxy resin diglycidyl ether of bisphenol-A (DGEBA) (D.E.R. 331 product of Dow Chemical, Midland, MI, USA), and further, polyetheramine, used as a curing agent (JEFFAMINE T-403 product of Huntsman), was added, taken in a ratio of 100 : 40 w/w. The mixture of WS_2 nanotubes and DGEBA was sonicated for 1 hr under controlled temperature and constant mechanical stirring. The sonicator was a high-intensity ultrasonic processor with a tip diameter of 19 mm that resonates at 20 kHz ± 50 Hz (VCX 400 (ultrasonic processor) and CV26 (tip), Sonics & Materials Inc., Newtown, CT, USA). The sonication process was performed in a pulsed on/off mode of 6 and 4 sec, respectively. The curing agent was added to the epoxy/WS_2 nanotube mixture and mixed manually. The curing conditions were 100°C for 6 hours; before the curing process took place, all the mixtures were degassed for 20 minutes at 45°C. All composite preparation technology conditions were varied in order to obtain the biggest complex dielectric permittivity value of samples at room temperature; it was determined that the above listed conditions are optimal. These conditions are different from those, which applied for epoxy/WS_2 composite preparation in [21].

To make sure of the morphological and structural quality of the nanotubes, a transmission electron microscope (TEM, Philips CM 120 operated at 120 kV) and scanning electron microscope (E-SEM, model FEI XL-30) were used. The crystallographic phase of the reaction product was confirmed by an X-ray powder diffractometer (XRD, Ultima III, Rigaku, Japan) operated at 40 kV and 40 mA (not shown). TEM, SEM, and XRD analyses were carried out after each synthesis and before impregnation into the polymer matrix. The dispersion of the nanoparticles inside the polymer was characterized by E-SEM analysis of sample's cross section.

The dielectric properties of the samples were investigated using a LCR meter (HP4284 A) in the frequency range 20 Hz-1 MHz. The measurements were done by heating and cooling in the temperature interval of 300 K–500 K at the constant temperature rate of 0.5 K/min. No noticeable hysteresis in experimental results was observed in both temperature change modes. The picture of measurement equipment is presented in Figure 1. The dielectric measurement accuracy was as better as 1%.

3. Results and Discussion

SEM and TEM images of WS_2 nanotubes are presented in Figures 2(a) and 2(b)), respectively. SEM images of epoxy/WS_2 nanotube cross section are presented in Figure 3. The WS_2 nanotubes are very well dispersed in the
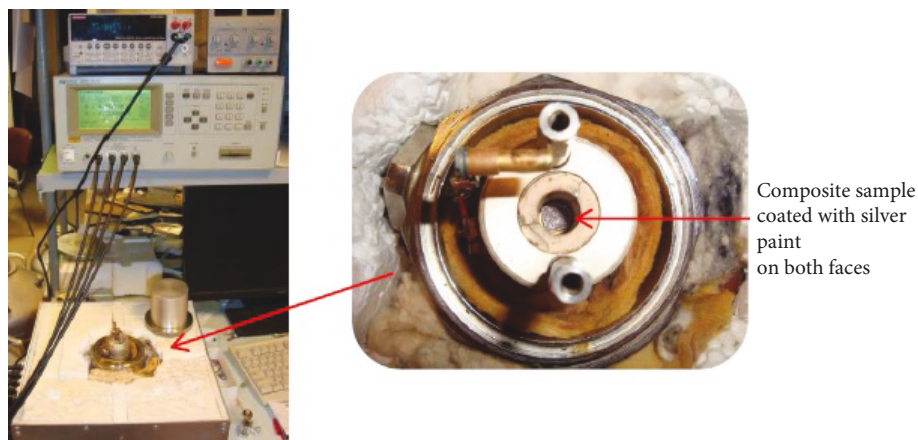
FIGURE 1: View of the LCR meter and the sample holder for measuring dielectric properties of composites in the 20 Hz–1 MHz frequency range.
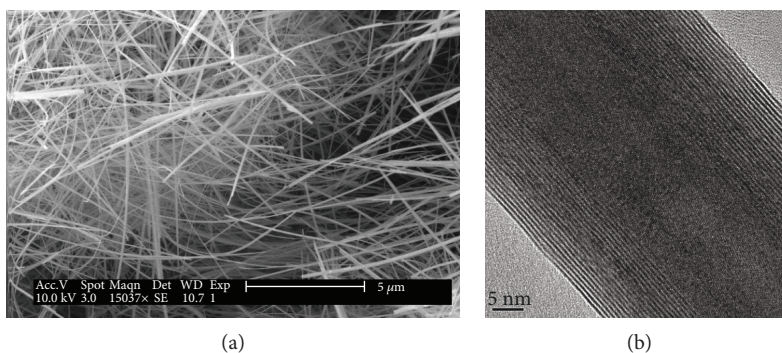

(a)　　　　　(b)

FIGURE 2: Scanning electron microscopy (SEM) (a) and transmission electron microscopy (b) images of WS$_2$ nanotubes.
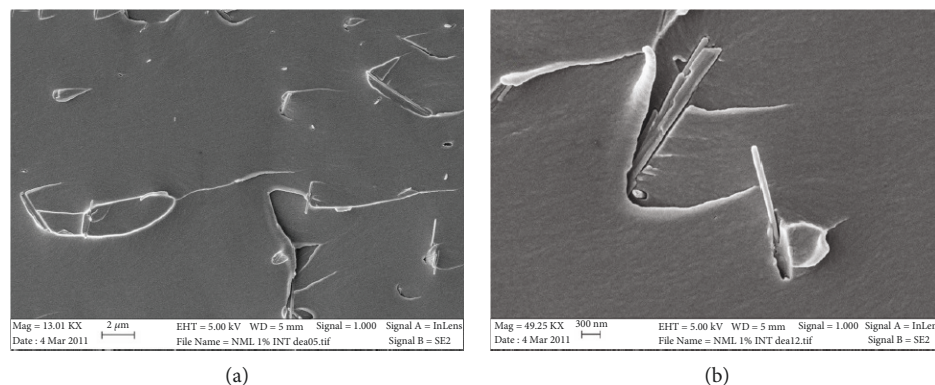

(a)　　　　　(b)

FIGURE 3: SEM images of epoxy/WS$_2$ nanotubes.

polymer matrix and no agglomerates of the WS$_2$ nanotubes are observed.

Temperature dependencies of complex relative dielectric permittivity for all composites at 1 kHz are presented in Figure 4. The complex relative dielectric permittivity strongly increases with WS$_2$ concentration; however, its value at room temperature remains very low even for the biggest concentration (1.6 vol%, $\varepsilon' < 6$, $\varepsilon'' < 0.3$). Therefore, all composites are below the percolation threshold.

However, the complex relative dielectric permittivity increases with temperature and has two anomalies: first, below room temperature, which is related to α relaxation
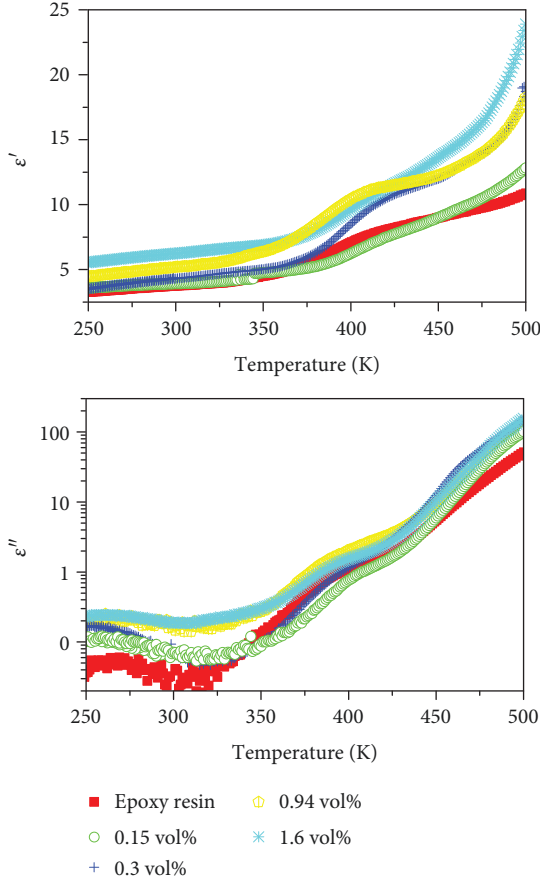
FIGURE 4: Temperature dependence of complex dielectric permittivity for epoxy/WS$_2$ composites at 1 kHz frequency.

FIGURE 5: Temperature dependence of complex dielectric permittivity for epoxy with 1.6 vol% WS$_2$ inclusions at different frequencies.

[25], and second, in the temperature range of 350-400 K, which is related to the Maxwell-Wagner relaxation [26] and the onset of electrical conductivity [27]. Both anomalies are strongly affected by the presence of WS$_2$ nanotubes. Temperature dependencies of complex relative dielectric permittivity for composites with 1.6 vol% of WS$_2$ at different frequencies are presented in Figure 5. Below room temperature, the maximum of losses is frequency-dependent and it is related to α relaxation [25]. Above 350 K, the step-like anomaly is related to the Maxwell-Wagner relaxation [26]. At higher temperatures, the electrical conductivity strongly affects results [27]. The temperature of the maximum of the dielectric losses (T_m) is strongly frequency-dependent (Figure 6). The behavior was characterized by the Vogel-Fulcher-Tammann law [28]:

$$\nu = \nu_0 \exp \frac{-E_f}{k(T_m - T_0)}, \qquad (1)$$

where k is the Boltzmann constant, ν_0 is the frequency approached with $T_m \to \infty$, E_f is the pseudoactivation energy, and T_0 is the Vogel temperature.
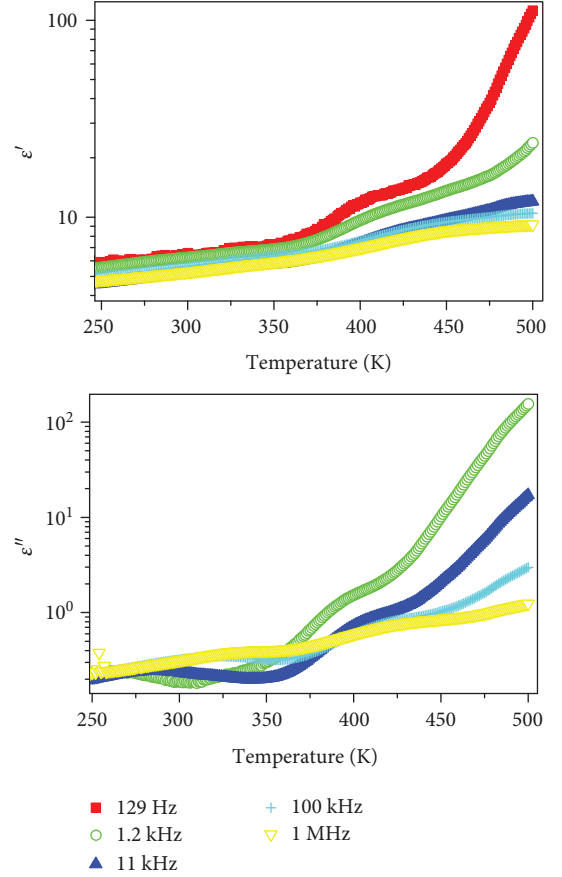
Obtained parameters are listed in Table 1.

The best-fit value of ν_0 was determined as 1 THz for all investigated composites. The value is consistent with phonon spectra of epoxy resin [29]. The Vogel temperature T_0 has the minimum for concentration 0.15 vol%. The decrease of T_0 is related to the intensified polymer molecule dynamics due to the additional free space at the polymer-filler junction. A similar change of the glass transition temperature (which is related to the Vogel temperature [30]) in polymeric composites with nanoinclusions was observed in reference [31]. On the other hand, the increasing of the Vogel temperature can be clarified by the strong interplay between epoxy resin and WS$_2$ nanotubes. Furthermore, the density of the composite could be higher than the pure polymer density, and consequently, the increasing of the glass transition temperature with the concentration of inclusions could be observed [32].

Frequency dependencies at different temperatures of complex relative dielectric permittivity for composites with 1.6 vol% inclusions are presented in Figure 7.

At low temperatures, the maximum of dielectric losses is observed, which shifts to the higher frequencies with temperature. The mean relaxation time was calculated as $\tau = 1/\nu_{max}$, where ν_{max} is the frequency at which dielectric losses have the maximum. The temperature dependence of the mean relaxation time is presented in Figure 8.
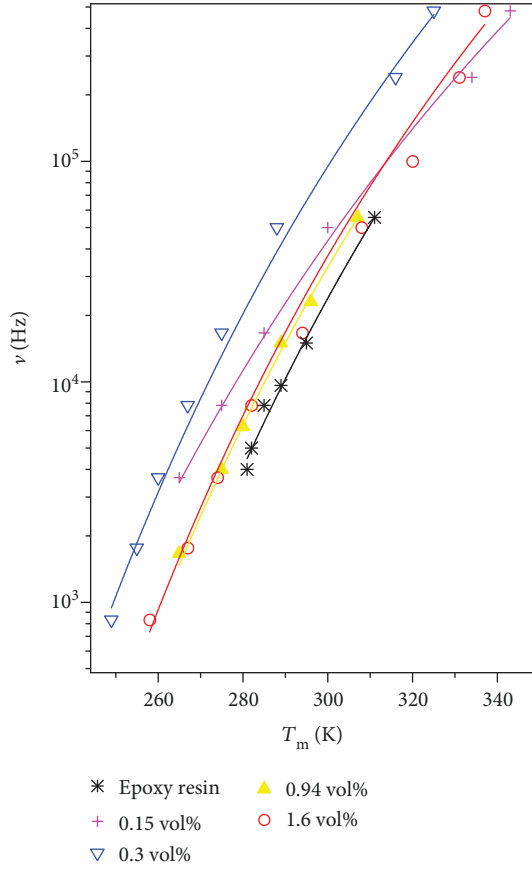
FIGURE 6: Measured frequencies ν versus T_m of WS$_2$/epoxy composites.

FIGURE 7: Frequency dependence of complex dielectric permittivity for epoxy with 1.6 vol% WS$_2$ inclusions at different temperatures.

TABLE 1: Parameters of the Vogel-Fulcher relationship (equation (1)) for WS$_2$/epoxy composites.

	E_f/k_B (K(eV))	T_0 (K)
Epoxy	3839 (0.33) ± 209 (0.01)	81 ± 11
0.15 vol%	4651 (0.4) ± 135 (0.01)	25 ± 7
0.3 vol%	3660 (0.31) ± 114 (0.01)	72 ± 6
0.94 vol%	4044 (0.35) ± 77 (0.01)	65 ± 4
1.6 vol%	4047 (0.35) ± 126 (0.01)	65 ± 7

The mean relaxation time increases on cooling according to the Vogel-Fulcher law [28]:

$$\tau = \tau_0 \exp \frac{E_f}{k(T - T_0)}, \tag{2}$$

where τ_0 is the relaxation time at very high temperatures, E_f is the activation energy, and T_0 is the Vogel-Fulcher temperature. Obtained parameters are listed in Table 2. The behavior is more diffused in comparison with the data presented in Table 1.
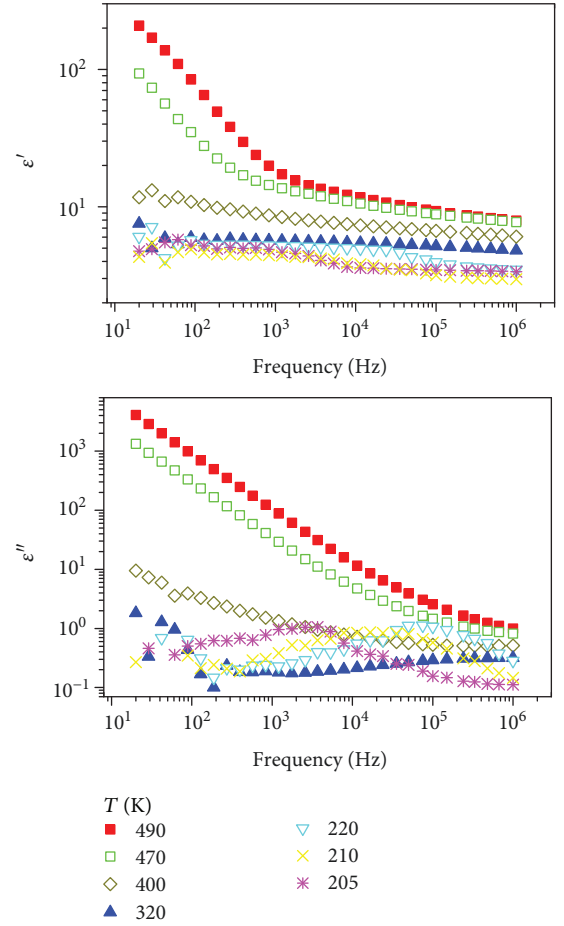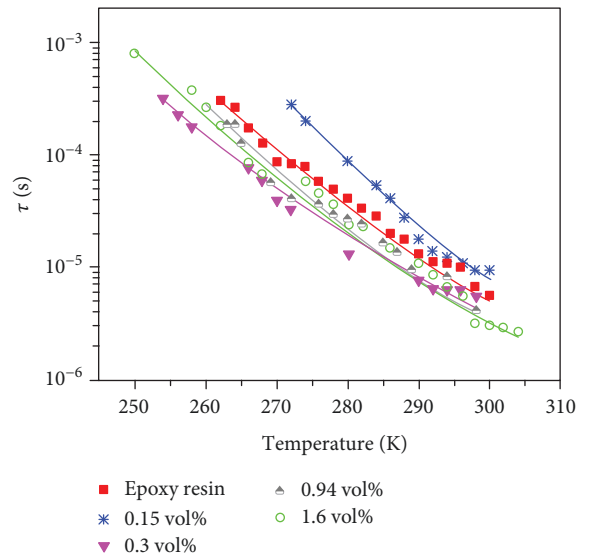

FIGURE 8: Temperature dependence of the mean relaxation time of WS$_2$/epoxy composites. The solid lines are calculated according to equation (2).

TABLE 2: Parameters of the Vogel-Fulcher law (equation (2)) for WS$_2$/epoxy composites.

	E/k_B (K(eV))	T_0 (K)
Epoxy	2981 (0.26) ± 61 (0.01)	109 ± 3
0.15 vol%	2363 (0.2) ± 88 (0.01)	150 ± 5
0.3 vol%	3167 (0.27) ± 111 (0.01)	90 ± 6
0.94 vol%	2877 (0.25) ± 114 (0.01)	111 ± 6
1.6 vol%	2826 (0.24) ± 75 (0.01)	114 ± 4

Spectra of the electrical conductivity for epoxy-matrix composites with 1.6 vol% (5 wt%) inclusions of the WS$_2$ nanotubes are presented in Figure 9. Above 420 K, the accidental distribution of the electrons according to energies causes a frequency-independent conductivity (DC conductivity) (Figure 9). The electrical conductivity of composites is caused by the electrical conductivity of the pure epoxy resin matrix. Above some critical frequency, the electrical conductivity strongly increases with frequency. The spectra of σ have been fitted according to the Almond-West type power law [33]:

$$\sigma = \sigma_{DC} + A\omega^s, \qquad (3)$$

where σ_{DC} is the DC conductivity and $A\omega^s$ is the AC conductivity. The model fits the electrical conductivity spectra of the investigated composites quite well; only at lower frequencies, the divergence is observed due to the nonohmic contact impact.

In order to separate effects of contacts and volume conductivity materials, we calculated the real part (ρ') and the imaginary part (ρ'') of the complex specific resistance $\rho^* = \rho' - i\rho''$:

$$\rho' = \frac{\varepsilon''}{\varepsilon'^2 + \varepsilon''^2} \frac{1}{\varepsilon_0 \omega},$$
$$\rho'' = \frac{\varepsilon'}{\varepsilon'^2 + \varepsilon''^2} \frac{1}{\varepsilon_0 \omega}, \qquad (4)$$

where $\omega = 2\pi\nu$ and ν is the measurement frequency. In the plot $\rho''(\rho')$, the half circles at higher frequencies are caused by volume conductivity of the composite, and the higher values of ρ^* are already influenced by contacts (Figure 10). The contact influence is playing an important role at higher temperatures and low frequencies.

The temperature dependence of $\ln(\sigma_{DC})$ of epoxy-matrix composites with various WS$_2$ concentrations is presented in Figure 11. The DC conductivity strongly increases with the WS$_2$ concentration. Therefore, electrical contacts between epoxy resin and WS$_2$ are rather ohmic. $\ln(\sigma_{DC})$ $(1/T)$ is the linear temperature dependence. Therefore, from this dependence, the activation energy E_B of the conductivity and the prefactor σ_0 were determined (the
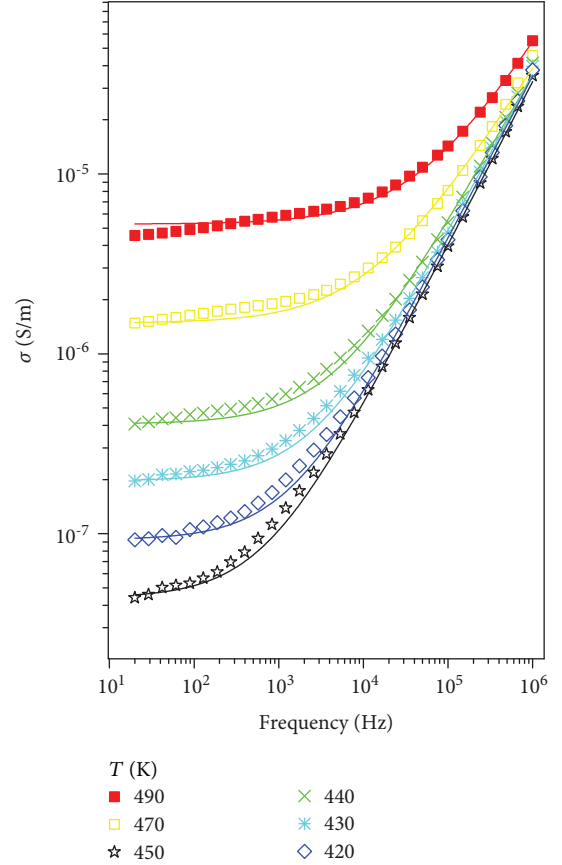

FIGURE 9: Frequency dependence of the electrical conductivity of WS$_2$/epoxy composites with 1.6 vol% inclusions at different temperatures.
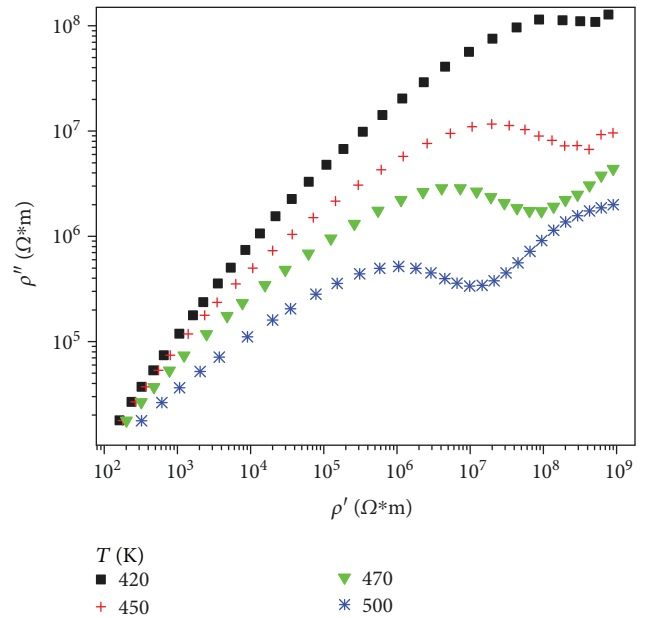

FIGURE 10: Cole-Cole diagram of $\rho^* = \rho' - i\rho''$ for epoxy with 1.6 vol% WS$_2$ inclusions at different temperatures.
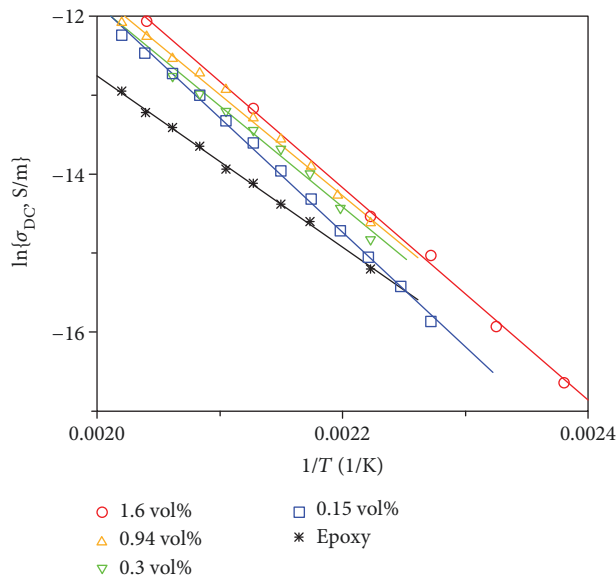
FIGURE 11: Temperature dependence of the DC electrical conductivity of WS$_2$/epoxy composites.

conductivity at very high temperatures) according to the Arrhenius law:

$$\sigma_{DC} = \sigma_0 \exp \frac{-E_B}{kT}. \tag{5}$$

The obtained parameters are presented in Table 3. The activation energy E_B and the conductivity σ_0 are practically not impacted by the concentration of nanoinclusions. However, the activation energy in composites is higher than in that in pure epoxy.

4. Conclusions

Broadband dielectric properties of tungsten disulfide (WS$_2$) nanotube epoxy-matrix composites over a wide temperature range (250 K-500 K) are presented for concentrations up to 1.6 vol% (5 wt%). The electrical percolation was not detected in the investigated composites at room temperature. The relative dielectric permittivity of composites with 1.6 vol% inclusions is almost 3 times higher than the relative dielectric permittivity of the pure polymer, and the DC electrical conductivity of composites is about 10^{-6} S/m at 500 K, which indicates that the composites are appropriate for antistatic applications at higher temperatures [34], similarly to WS$_2$/polyurethane composites [22]. Broadband electromagnetic spectra of the composites are largely governed by the dynamics of epoxy resin molecules. The Vogel temperature T_0 has the minimum for concentration 0.15 vol%. Above 410 K, the electrical conductivity is typical for WS$_2$/epoxy composites with the WS$_2$ inclusions as well. The DC conductivity increases with the WS$_2$ concentration, while its activation energy is almost uncontrolled by the concentration of nanoinclusions; however, in composites, it is higher than in pure epoxy resin. Therefore, the electrical transport occurs

TABLE 3: Arrhenius law fit parameters for WS$_2$/epoxy composites.

	ln $\{\sigma_0, \text{S/m}\}$	E_A/k_B (K(eV))
Epoxy	8.7 ± 0.1	$10727 (0.92) \pm 85 (0.01)$
0.15 vol%	16.9 ± 0.3	$14367 (1.24) \pm 132 (0.01)$
0.3 vol%	11.4 ± 0.3	$11702 (1.01) \pm 143 (0.01)$
0.94 vol%	13.7 ± 0.1	$12681 (1.1) \pm 74 (0.01)$
1.6 vol%	15.4 ± 0.1	$13457 (1.16) \pm 74 (0.01)$

between WS$_2$ nanotubes and epoxy matrix at higher temperatures (above 410 K).

Data Availability

The underlying data related to this article are available upon request.

Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

Acknowledgments

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