



Thermally conducting composites for power electronics

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October 12, 2019

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Abstract — The thermal conductivity of polyvinylchloride (PVC) containing graphite (Gr) micro fillers have been studied because of their potential applications in power electronics devices, batteries, electromagnetic shielding, antistatic and corrosion resistant coatings and other functional applications. Micro – composites containing (2, 4, 6, 8, 10, and 12%) volume fractions of graphite were fabricated. The thermal conductivity (K) of composites is enhanced by ~420% as concentration is increased from 0 to 12vol. %. Composites with a higher concentration show a stronger dependence in the thermal conductivity.

It was revealed that, upon heating and cooling of these heterogeneous macro systems, hysteresis phenomena based on the effect of the “asymmetry” of the temperature evolution of the electrically active defects appear at the temperatures $T < 80^{\circ}\text{C}$.

During the heating –cooling cycle, the relative changes in the ε' and $\tan\delta$ with the temperature were about 33 and 61%, respectively.

Keywords— thermal conductivity, polyvinylchloride, composite, graphite, power electronics, dielectric permittivity, dielectric losses

I. INTRODUCTION

Heat dissipation in electronic devices becomes very important because the power density of the state of the electronic devices is extremely high due to their highly integrated circuits and small size. Effective heat dissipation is also crucial in maintaining the performance devices [1]. One of the most limiting factors in heat dissipation is thermal contact resistance between different components. In order to reduce the contact resistance, various materials have been synthesized and tested for a wide variety of applications including microprocessors, optoelectronics, and power electronics [2].

In the power industry, inorganic fillers are usually incorporated into electrical insulating polymers to archive specific electrical, mechanical, and thermal properties. More recently carbon nanoparticles, graphite, nanotubes, graphene are used as fillers to improve thermal conductivity of polymers, although their high price limits the use of these materials to special applications [3]. In this context, the design and fabrication of multifunctional materials with 1) relatively low mass-density; 2) coefficient of thermal expansion similar to metals; 3) lower electrical conductivity than most metals; 4) higher thermal conductivity than polymers; 5) high compressive moduli are needed.

The most important polymer that has Gr micro-and nano composites is polyvinylchloride (PVC). Graphite and carbon offer the benefit of low density and cost when compared to metallic substances used for the same function. Graphite and carbon also have an advantage in that they are typically inert and compatible with most if not all polymer systems. Many of carbons, and especially graphite, have thermal and electrical conductivities many orders of magnitude higher than most polymers or plastics. Graphite attracts attention because of their potential applications in various high technology aspects, for example sensors, catalysis, super capacitors, electromagnetic shielding, radar evasion, rechargeable batteries, conductive inks, and antistatic textiles, thermistors, aerospace as well as in secondary battery and bipolar plates in the polymer electrolyte membrane fuel cell [4].

It should be noted that when choosing a material for power electronics devices, it is necessary to have information about the processes occurring in the material, as will be affected by this or that material on the operation of other parts of the device depending on the temperature, frequency of the applied field, including due to electrical redistribution fields. It is necessary to take into account the mechanical, thermal, electro physical, physic-mechanical, magnetic, optical and other properties of both active and passive polymeric composite materials. These studies will open up new opportunities for the effective management of a complex of physical properties of polymer composite materials by exposing them to temperature and an alternating electric field.

Many researches have studied the thermal, electrical and dielectric properties of polymer-graphite composites [3-10].

I. K. Tawman and T. Ergin [3] investigated various thermophysical properties of HDPE/graphite nanocomposites, such as thermal conductivity, specific heat capacity, electrical conductivity, mechanical properties. Thermal conductivity increased from 0.53W/m·K for pure HDPE to 2.64 W/m·K for 30Wt% of graphite particles. The percolation threshold for electrical conductivity is 16Wt% of graphite particle filler; the electrical conductivity increased from $8.07 \times 10^{-14} \text{S}\cdot\text{cm}^{-1}$ for neat HDPE to $2.02 \times 10^{-4} \text{S}\cdot\text{cm}^{-1}$ for composites with 30Wt% of graphite particles.

Yu Chao Li et al. [5] investigated the electrical conductivity and dielectric properties of PVDF/EG composites (the content of fillers was 2-12vol.%) in a wide range of frequencies (from

10^2 to 10^8 Hz) and temperatures (from 20 to 190 °C). Their results showed that the AC conductivity (σ) and dielectric constant (ϵ') of the PVDF/EG composites near the percolation threshold were frequency depended and obeyed the power law relations ($\sigma \sim \omega^u$ and $\epsilon' \sim \omega^{-v}$). V. Panwar et.al. [6] studied and analyzed the dielectric properties of HDPE-Gr composites in low and radio frequency ranges for capacitor and electromagnetic interference shielding application. The highest dielectric constant (3.12×10^6) at 20 Hz and highest dissipation factor (5.58×10^4) at 150Hz for composites with $\Phi = 0.187$ makes there composites suitable for charge storing devices, decoupling capacitors and electromagnetic interference shielding applications.

Authors of [9,10] studied the dielectric properties and dc and ac electrical conductivity of PVC/Gr composites in a wide range of temperatures and frequencies . They noted that the temperature dispersion of the ac conductivity of PVC/Gr composites has two regions of positive temperature coefficient and two regions of negative temperature coefficient of resistivity and the temperature dependences of ϵ' and $\tan \delta$ are nonmonotonic.

It has been shown that the frequency dispersion of the ac conductivity of the PVC/Gr composites exhibits two linear portions with $n_1=0.48$ and $n_2=0.39$ for the PVC-1%Gr composite and $n_1= 0.46$ and $n_2=0.37$ for the PVC-2%Gr composite. The percolation threshold of the composites was estimated to be $\sim 6\text{vol.}\%$. the main mechanism of ac conductivity in insulators of the PVC-Gr composite system is barrier hopping.

This research work aims to produce graphite filled PVC composites with high thermal conductivity by exploiting properties of PVC and Gr . In this work we report here that homogenous PVC/Gr composites can be prepared using a simple method, i.e. by dispersing commercial Gr powder in PVC matrix via powder technology method followed by hot pressing, and report our preliminary experimental results on the dependence of the thermal conductivity and dielectric characteristics (ϵ' and $\tan \delta$) on the filler content and temperature.

II. EXPERIMENTAL SETUP

A. Materials

The matrix polymer used in this work is a commercial white powder polyvinyl chloride (PVC) with particle sizes in the range of 150-200 μm manufactured in South Korea. The various physical parameters of PVC are as follows: density 1,42g/cm³, melting temperature =180°C, heat deflection temperature =147°C, glass transition temperature = (80-87°C), dielectric constant at 1 kHz = 6, dissipation factor at 1 kHz = 0.0468 and volume resistivity = $5 \cdot 10^{13}$ Ohm·sm. As the electrical conducting filler natural graphite (EUZ-M, Ukraine) with average particle size ranging from 50 to 63 μm was used without treatment. Some of the properties of graphite are as follows: color is black, density=(2,25~2,30)g/cm³, maximum usable temperature=3600, electrical resistivity = $4,5 \cdot 10^{-4}$ Ohm·m.

B. Composite sample preparation

In the former case, graphite powder in the desired amount was mixed thoroughly with the PVC powder in an agate pestle mortar for 1h. This process coated the conducting graphite powder on the surface of the PVC particles, so it is referred to as prelocalization of the conductive phase. Prolonged mixing improved the homogeneity of the spatial distribution of the conductive particles and their uniform coating thickness on the PVC particles. The tumble mixed prelocalized powders was prepared for different graphite contents of 2-12vol. %. The resultant powder was further dried at 120°C for 6h prior to compression molding. Then, the PVC/Gr composites were prepared by hot - pressing the powder mixture technique. The dried powder was filled in a tool steel die having diameter 15mm. The powder was heated at an average heating rate of 6°C/min under pressure of 15MPa to a maximum temperature of 180°C. After soaking period of 10 minutes, the dick samples of 15mm diameter and thickness 1,2mm were punched from the plates and cooled down to 0°C in a water-ice mixture (quenching mode) at atmospheric pressure to eliminate porosity, bubbles or blisters. Thus, the 0-3 composites containing 2,4,6,8,10 and 12vol.% Gr in the PVC matrix were prepared. The specimens were sealed in air free polyethylene bags prior to measurements to avoid atmospheric and humidity effects that may induce same changes in the conductivity of the specimens.

C. Measurements

To measure the thermal conductivity, we prepared the samples in a disk shape with a diameter of 15mm and a thickness of 1,2 mm. The measurement was carried out under stationary heat flow conditions with the thermal conductivity meter (model ИТ- λ -400, Russia)

The materials were examined using parallel-plate capacitors in a two-electrode system in the equivalent circuit of a resistor and a capacitor connected in parallel at temperatures of 20–150°C and a measuring voltage amplitude of $U = 1$ V, using a special shielded and grounded heated “sandwich” measuring cell with a system of a measuring and potential electrode with a diameter of 15 and 35 mm, respectively, in both the forward and backward directions, i.e., increasing and decreasing temperature T , respectively. Temperature measurements were estimated at a constant frequency 1 kHz. The samples were placed in the measuring cell with pressure-exerting stainless steel electrodes. Sample temperature was controlled by the microelectronic thermo regulator (type – MMX-400). The centering of the electrodes was provided by a special mandrel in a heated chamber. The distance between the electrodes was determined by the thickness of the test samples. The measurements of capacitance C , dielectric loss tangent $\tan \delta$ were recorded in a direction perpendicular to the plane of compression of the samples with the E7-20 broadband precision immittance meter. The measured values of these parameters and the geometric dimensions of the samples were used to determine real ϵ' ($\epsilon' = Cd/\epsilon_0 S$, where S is the area of the planar sample and d is the film thickness) part of the complex

dielectric permittivity. The dissipation factor was obtained directly from the instrument.

III. RESULTS AND DISCUSSION

Thermal conductivity (K) of PVC/Gr composites was measured as function of Gr concentration as presented in figure. It is seen from the Fig.1 that with the increase in the filler concentration, the thermal conductivity of the composite increases nonlinearly, it was similar as in the literature [11-13]. By adding 2% volume of graphite to polyvinylchloride, we obtain 0.24 value of thermal conductivity (W/mK) and with the increasing percent of graphite we get 0.96 values at the 12%. The experimentally observed value of thermal conductivity of PVC is 0.18 W/mK at room temperature. This value was in good agreement with [14]. The thermal conductivity data showed a behavior similar to the electrical conductivity and the dielectric permittivity [10]: the composite with graphite concentration had higher thermal conductivity, as shown in Fig.1. The sample with highest graphite concentration, 12vol. %, possessed a thermal conductivity of $K=0,96\text{W/mK}$. This value is 5 time higher than the thermal conductivity of the PVC matrix.

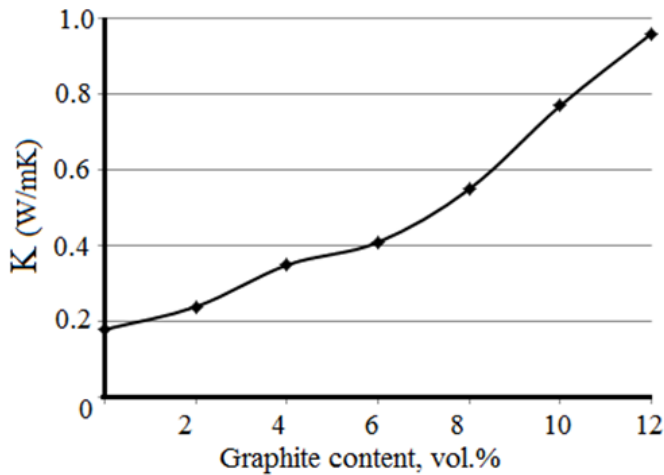


Fig. 1. Thermal conductivity of PVC/Gr composites as a function of Gr concentration

The increase in thermal conductivity was attributed to good dispersion of Gr loading. By increasing Gr loading, the thermal conductivity increases because particles begin to form conductive graphite chains, and heat flows not only through the formed conductive graphite chains, but also through the PVC itself, which caused the rapid increase in thermal conductivity [15].

Fig.2 shows the thermal behavior of the real part of the complex dielectric permittivity (ϵ') and $\tan\delta$ for the PVC+2%Gr micro composite, which was under thermal-cooling cycles at a hitting rate of 30C/min. Fig.2 indicates that the value of the ϵ' increases slightly until it reaches its maximum at 350C, and then decreases sharply and then abruptly decreases and achieves a minimum at the around glass transition temperature (80°C) of PVC; after that, as the temperature

increases, an increase in $\epsilon'(T)$ is observed again; after 125°C, after 125°C a value that is constant until 150°C.

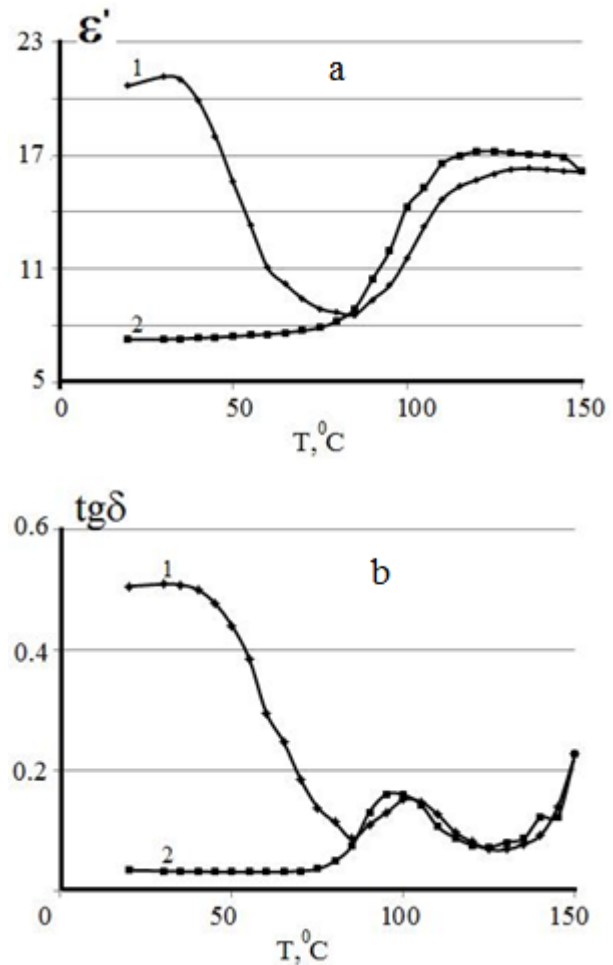


Fig. 2. The temperature dependence of the permittivity (a) and the tangent of the dielectric loss (b) of the PVC/Gr 2vol% composite materials: (1) heating; (2) cooling

However, ϵ' values under heating are higher than those under cooling. The thermal behavior of ϵ' and $\tan\delta$ of the samples during the cooling process temperature dependence of ϵ' and $\tan\delta$ of the samples from the high- temperature phase to the temperature of 80°C almost coincide with the heating curves. Temperature hysteresis effect appears at $T < 80^\circ\text{C}$, and the values of ϵ' and $\tan\delta$ for one and the some temperatures are smaller upon cooling than upon heating of the sample. As a result of the heating – cooling cycle, the dielectric characteristics of the samples take new values and at $T < 80^\circ\text{C}$ tends to stabilize at a definite value in a reversible process. At the end ϵ' and $\tan\delta$ does not return to it's until value. The value at the this point was 7,27 and 0,035, and before to start the thermal cycle was 20,67 and 0,057, respectively Such behavior of the composite material is related to the thermally stimulated process of the redistribution of the charge at the particle metrics interface. After cooling to room temperature, the samples of the composite material can stay in such a state for a long time. This peculiarity of composite materials causes hysteresis phenomena

that are based on the effect of “asymmetry” of the temperature evolution of electrically active defects [16, 17].

The $\tan\delta = f(T)$ dependence (Fig. 2b) has the following specific features: up to 80°C, the behavior of the $\tan\delta = f(T)$ function is almost identical to that of the $\epsilon' = f(T)$ function, and the region where the $\tan\delta$ value of the composites remains nearly constant decreases twofold compared with the analogous region for pure PVC. The positions of the $\tan\delta$ maximum and minimum observed at $T = 100$ and 130°C , respectively, do not depend on the content of graphite in the matrix.

During the heating –cooling cycle, the relative changes in the ϵ' and $\tan\delta$ with the temperature were about 33 and 61%, respectively.

Thus, these studies show that highly filled PVX / Gr composites can be used in natural cooling systems of power electronics devices and temperature-sensitive elements, etc.

IV. CONCLUSION

PVC micro composites were prepared by the mixing and the hot compression mold technique. The thermal conductivity of the composites is enhanced by ~420% compared with pure PVC as the filler concentration is increased from 0 to 12vol.% with continued improvement of Gr reduction processes and better linkage between, Gr/polymer, the thermal conductivity of Gr/polymer composites has potential to be improved further, making Gr/polymer composites such as PVC/Gr a good alternative for many thermal management applications in the future.

It was revealed that, upon heating and cooling of these heterogeneous macro systems, hysteresis phenomena based on the effect of the “asymmetry” of the temperature evolution of the electrically active defects appear at the temperatures $T < 80^\circ\text{C}$.

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