Наносистеми, наноматеріали, нанотехнології
Nanosistemi, Nanomateriali, Nanotehnologii
2024, т. 22, № 3, сс. 637–642
https://doi.org/10.15407/nnn.22.03.637

PACS numbers: 65.80.Ck, 66.70.Lm, 72.80.Tm, 81.05.ue, 81.07.Wx, 84.32.Tt

Electrophysical Properties and Thermal Conductivity of Composite Based on Zinc Oxide and Reduced Graphene Oxide (1 vol.%)

B. Turko[1], V. Vasil'ev[1], B. Sadovyi[1,2], R. Bihun[1], I. Pylypiv[3], V. Kapustianyk[1], and D. Leonov[4]

[1]*Ivan Franko National University of Lviv,*
50, Drahomanov Str.,
UA-79005 Lviv, Ukraine
[2]*Institute of High Pressure Physics P.A.S.,*
29/37, Sokolowska Str.,
01-142 Warsaw, Poland
[3]*Systems Engineer II, SiTime,*
10[a] B. Khmelnytsky Str.,
UA-81100 Pustomyty, Lviv Region, Ukraine
[4]*Technical Centre, N.A.S. of Ukraine,*
13, Pokrovs'ka Str.,
UA-04070 Kyiv, Ukraine

The thermal conductivity of the composite materials based on the commercial ZnO micropowder with reduced graphene oxide (1 vol.%) powder dispersed in the polymethylsiloxane (silicone oil) is measured using the radial heat-flow method. The thermal conductivity of the composite material based on the commercial ZnO micropowder with an average particle size of 50 μm and reduced graphene oxide is found to be 9.4 W/(m·K). At room temperature, the values of the dielectric permittivity at the measuring electric-field frequencies of 50 Hz and 1 MHz and the specific volume electrical resistance for the composite are obtained. An increase in the values of both the coefficient of thermal conductivity and the dielectric constant as well as a decrease in the specific volume electrical resistance due to a change in the volume fraction of reduced graphene oxide in the composite from 0.5 vol.% up to 1 vol.% are recorded.

Методом радіяльного теплового потоку вимірювали теплопровідність композиційних матеріялів на основі комерційного мікропорошку ZnO з відновленим порошком оксиду графену (1 об.%), дисперґованого у поліметилсилоксані (силіконова олія). Встановлено, що теплопровідність композиційного матеріялу на основі комерційного мікропорошку ZnO із середнім розміром частинок у 50 мкм і відновленого оксиду графену становить 9,4 Вт/(м·К). За кімнатної температури одержано значення діелектричної проникности на частотах вимірювального електричного поля у 50 Гц та 1 МГц і питомого об'ємного електричного опору для композиту. Було зафіксовано збільшення значень коефіцієнта теплопровідности й діелектричної проникности та зменшення питомого об'ємного електроопору за рахунок зміни об'ємної частки відновленого оксиду графену в композиті від 0,5 об.% до 1 об.%.

Key words: reduced graphene oxide, zinc oxide, composites, thermal conductivity, dielectric constant, specific volume electrical resistance.

Ключові слова: відновлений оксид графену, оксид Цинку, композити, теплопровідність, діелектрична проникність, питомий об'ємний електричний опір.

(Received 25 April, 2024)

1. INTRODUCTION

There is a problem with effective heat removal from heat-generating working elements of electronic devices, in particular processors, chipsets, computer video accelerators, high-power LEDs and lasers. If heat is not removed, this can lead not only to a significant deterioration in the performance of devices, but also to their failure [1]. To remove heat from the heat-generating working elements of electronic devices, thermal greases are widely used, which consist of a substance in a liquid state (for example, silicone oil, polyol, *etc.*) and a wide variety of fillers-thickeners (for example, Ag, SiO_2, ZnO, BN, AlN, Al_2O_3, graphite, graphene, carbon nanotubes, diamonds, *etc.*) [2].

Graphene has a record value of the coefficient of thermal conductivity among all known materials [3]; that is why it has attracted considerable scientific interest for its application in thermal management [1–7]. Graphene-containing commercial thermal greases are already available. For example, 'Baircool', 'Scythe Thermal Elixer G' and 'Hi-G Thermal Grease'. The thermal conductivity coefficients of the above thermal greases are in the range from 4 to 11 W/(m·K), and their composition is a trade secret. On the other hand, reduced graphene oxide (*r*GO) is more stable than graphene [8]. The thermal conductivity of the *r*GO can reach 2600 W/(m·K) and depends on the concentration of oxygen atoms in *r*GO [9]. In addition, according to [10], silicone thermal greases with *r*GO and *h*BN (hexagonal boron nitride) are promising.

In this work, in the continuation of our previous studies [3], we investigated the effect of increasing the volume fraction of reduced graphene oxide from 0.5 vol.% to 1 vol.% on the specific volume resistance, dielectric permittivity, and thermal conductivity of the composite material based on ZnO, *r*GO, and polymethylsiloxane.

2. EXPERIMENT

The composite materials were produced by dispersing of ZnO micropowder with an average particle size of 50 μm (99.7%, UKRZINC, Kyiv, Ukraine) with *r*GO powder (0.5 vol.% or 1 vol.%, purchased from Sigma-Aldrich, Saint Louis, USA) in the polymethylsiloxane PMS 1000 (silicone oil, purchased from Sfera Sim, Lviv, Ukraine). The silicone oil and the fillers were taken in a volume ratio of 3:7. According to the certificate of analysis obtained from Sigma-Aldrich, the *r*GO contained 83% of carbon and 4% of nitrogen by mass.

Determination of the thermal conductivity of the composites was carried out by radial heat flow method [3, 11].

The values of dielectric permittivity and specific volume electrical resistance at room temperature and different frequencies of measuring electric field were obtained by LCR Meter IM3536-01 (HIOKI E. E. Corporation, Nagano, Japan).

The free software 'RealTemp' and 'CPU Burn-in v1.0' were used for testing of the thermally conductive composites. 'RealTemp' is a program for monitoring the temperature of computer processor cores. It was designed for Intel Single Core, Dual Core, Quad Core, and Core i7 processors. Each core in these processors has a digital thermal sensor. 'CPU Burn-in v1.0' is a program that 'heats' any processor with 'x86' architecture to the maximum possible operating temperature, accessible using a conventional software.

3. RESULT AND DISCUSSION

Figure 1 and Figure 2 presents the time dependences of the processor operating temperature. They were measured starting from the moment of turning on (Fig. 1)/off (Fig. 2) the stable load of the computer, using different layers of composite material between the surfaces of the processor and the copper heatsink used for heat dissipation. It has been found that heat was removed much better when using a composite with higher *r*GO concentration.

At room temperature, the obtained values of the dielectric permittivity ε at the frequencies of electric measuring field $\nu = 50$ Hz or $\nu = 1$ MHz and specific volume electrical resistance ρ ($\nu = 50$ Hz) for the two composites are given in Table.

The thermal-conductivity coefficient α [W/(m·K)] of the compo-

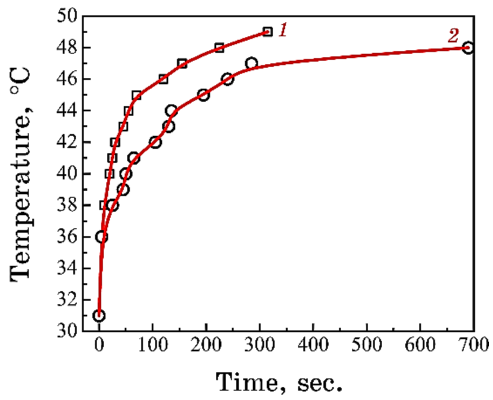

Fig. 1. The time dependences of the processor operating temperature, measured starting from the moment of turning on the stable load of the computer, using different layers of composite material between the surfaces of the processor and the copper heatsink used for heat dissipation: *1*—composite based on ZnO, *r*GO (0.5 vol.%) and polymethylsiloxane; *2*—composite based on ZnO, *r*GO (1 vol.%) and polymethylsiloxane.

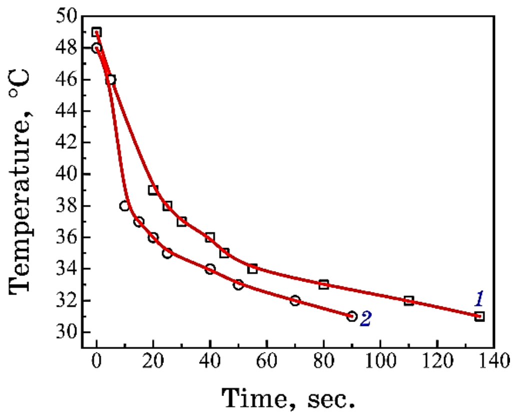

Fig. 2. The time dependences of the processor operating temperature, measured starting from the moment of turning off the stable load of the computer, using different layers of composite material between the surfaces of the processor and the copper heatsink used for heat dissipation: *1*—composite based on ZnO, *r*GO (0.5 vol.%) and polymethylsiloxane; *2*—composite based on ZnO, *r*GO (1 vol.%) and polymethylsiloxane.

sites is calculated on the basis of the following relation [3, 11]:

$$\alpha = K\frac{\ln(r_1/r_2)}{2\pi l(T_1 - T_2)}UI, \quad (1)$$

TABLE. Room-temperature electrophysical parameters of the studied materials.

Parameter	Composite based on ZnO and *r*GO (0.5 vol.%) powders [3]	Composite based on ZnO and *r*GO (1 vol.%) powders
Specific volume electrical resistance at frequency of 50 Hz, Ohm·cm	$8\cdot10^9$	$2.5\cdot10^9$
Dielectric constant at frequency of:		
50 Hz	60	71
1 MHz	43	54

where K is the factor of the axial heat loss through the plugs of the measuring cell (depends on the plug material and is calculated by reference to a sample with known thermal conductivity); r_1 and r_2—the inner and outer radii of the cylindrical composite layer; T_1 and T_2—the temperatures of the internal and external surfaces of the composite layers; l—the length of the cylindrical composite layer; U—the voltage on the heater; I—the current in a heater.

The value of the thermal conductivity of the composite materials based on ZnO powders with a grain size of 50 μm with *r*GO (1 vol.%), calculated according to Eq. (1), was found to be equal to 9.4 W/(m·K). The relative measurement error did not exceed 10%.

An increase in the values of the coefficient of thermal conductivity and dielectric constant and a decrease in the specific volume resistance due to a change in the volume fraction of reduced graphene oxide in the composite from 0.5 vol.% up to 1 vol.% are associated with the physical properties of *r*GO ($\alpha = 2600$ W/(m·K), $\varepsilon = 1130$ ($\nu = 50$ Hz), $\rho = 1.4\cdot10^{-2}$ Ohm·cm), the Maxwell–Wagnare–Sillars interfacial polarization and more formation of the microcapacitor structures [3].

4. CONCLUSIONS

In summary, the coefficients of thermal conductivity of the composite material based on the ZnO with *r*GO (1 vol.%) powders dispersed in the polymethylsiloxane were determined by radial heat flow method. It was found to be equal to 9.4 W/(m·K). The differences in the electrophysical properties of *r*GO (0.5 vol.%)–ZnO–polymethylsiloxane and *r*GO (1 vol.%)–ZnO–polymethylsiloxane composites are explained by an increase in the Maxwell–Wagnare–Sillars interfacial polarization, the concentration of microcapacitor structures and reduced graphene oxide. The high performance of *r*GO–ZnO composite synthesized by a simple and facile process in this work shows promising potential in thermal control of the electronic devices.

ACKNOWLEDGMENTS

This work was done due to the supports of the Ministry of Education and Science of Ukraine (Project No. 0123U101880) and Long-Term Funding by the Polish Academy of Sciences and U.S. National Academy of Sciences (Project No. PAN.BFB.S.BWZ.369.022.2023).

REFERENCES

1. J. Khan, S. A. Momin, and M. Mariatti, *Carbon*, **168**: 65 (2020); https://doi.org/10.1016/j.carbon.2020.06.012
2. W. Xing, Y. Xu, C. Song, and T. Deng, *Nanomaterials*, **12**: 3365 (2022); https://doi.org/10.3390/nano12193365
3. B. Turko, V. Vasil'ev, and V. Kapustianyk, *Nanosistemi, Nanomateriali, Nanotehnologii*, **21**: 569 (2023); https://doi.org/10.15407/nnn.21.03.569
4. S. Ali, F. Ahmad, P. S. M. Yusoff, N. Muhamad, E. Onate, M. R. Raza, and K. Malik, *Composites Part A: Applied Science and Manufacturing*, **144**: 106357 (2021); https://doi.org/10.1016/j.compositesa.2021.106357
5. P. Huang, Y. Li, G. Yang, Z.-X. Li, Y.-Q. Li, N. Hu, S.-Y. Fu, and K. S. Novoselov, *Nano Materials Science*, **3**: 1 (2021); https://doi.org/10.1016/j.nanoms.2020.09.001
6. Q. Ge, J. Chu, W. Cao, F. Yi, Z. Ran, Z. Jin, B. Mao, Z. Li, and K. S. Novoselov, *Advanced Functional Materials*, **32**: 2205934 (2022); https://doi.org/10.1002/adfm.202205934
7. D. D. L. Chung, *Materials Chemistry and Physics*, **309**: 128432 (2023); https://doi.org/10.1016/j.matchemphys.2023.128432
8. R. Kamatchi and K. G. Kannan, *International Journal of Renewable Energy Research*, **8**: 313 (2018); https://doi.org/10.20508/ijrer.v8i1.6766.g7305
9. Y. Zeng, T. Li, Y. Yao, T. Li, L. Hu, and A. Marconnet, *Adv. Funct. Mater.*, **29**: 1901388 (2019); https://doi.org/10.1016/j.jallcom.2010.03.076
10. W. Liang, X. Ge, J. Ge, T. Li, T. Zhao, X. Chen, M. Zhang, J. Ji, X. Pang, and R. Liu, *Nanomaterials*, **9**: 938 (2019); https://doi.org/10.3390/nano9070938
11. B. I. Turko, V. B. Kapustianyk, V. P. Rudyk, and Y. V. Rudyk, *J. Nano-Electron. Phys.*, **8**: 02004 (2016); https://doi.org/10.21272/jnep.8(2).02004