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Creating a composite based on thermal paste and carbon nanotubes to improve heat-conducting properties

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Abstract: Efficient heat transfer is essential in many industrial sectors to ensure safe and efficient operation of equipment. Thermal pastes are materials used to improve heat transfer between different surfaces. In this study, the influence of carbon nanotubes on the heat-conducting properties of thermal pastes was studied in order to create a new, more efficient and economical composite. The study included an experiment consisting in selecting the optimal type of thermal paste, as well as determining the most effective amount of carbon nanomaterial, in order to assess the thermal stability and improve the characteristics of thermal pastes when adding carbon nanotubes. The results of the study showed that the addition of carbon nanotubes makes it possible to improve the distribution and transfer of heat, namely, when carbon nanotubes were introduced at a concentration of 0.01 %, the processor temperature remained lower by 15 °C with preliminary exposure to nanotubes with ultrasound, and by 10 °C without pretreatment in an ultrasonic stirrer. The results highlight the potential of carbon nanotubes as a promising additive for improving the thermal conductivity properties of thermal pastes in various applications.

Keywords: thermal paste; carbon nanotubes; thermal conductivity; interactions; structural modification; nanoelectronics and microsystems engineering.

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Создание композита на основе термопасты и углеродных нанотрубок для улучшения теплопроводных свойств

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Аннотация: Эффективная передача тепла имеет важное значение во многих промышленных отраслях для обеспечения безопасной и эффективной работы оборудования. Термопасты являются материалами, применяемыми для улучшения теплопередачи между различными поверхностями. В данном исследовании было изучено влияние углеродных нанотрубок на теплопроводные свойства термопаст в целях создания нового, более эффективного и экономичного композита. При выполнении работы был проведён эксперимент, заключающийся в подборе оптимального типа термопасты, а также определения наиболее эффективного количества углеродного наноматериала, для оценки термической стабильности и улучшения характеристик термопаст при добавлении углеродных нанотрубок.Результаты исследования показали, что добавление углеродных нанотрубок позволяет улучшить распределение и передачу тепла, а именно при введении углеродных нанотрубок в концентрации 0,01 % температура процессора оставалась ниже на 15ºС при предварительном воздействии на нанотрубки ультразвуком, и на 10 °С без предварительной обработки в ультразвуковой мешалке. Полученные результаты подчеркивают потенциал углеродных нанотрубок как многообещающей добавки для улучшения теплопроводных свойств термопаст в различных приложениях.

Ключевые слова: термопаста; углеродные нанотрубки; теплопроводность; взаимодействия; новые композиты; наноэлектроника и микросистемная техника.

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1. Introduction

In today's world, with the growth of the computing power of computers, servers, the gradual transition to electric motors and the growth of production capacity, many industrial sectors require efficient heat transfer to ensure the safe and efficient operation of equipment. Due to the air gap and uneven surface, heat transfer from the heating part to the cooling part is not good enough (because air is a heat insulator), and it is not always possible to perfectly polish the surfaces of the radiator and the heat-producing part. In this case, thermal paste is the ideal solution.

Thermal pastes are materials that are used to improve heat transfer between different surfaces. They usually consist of a thermally conductive base and a filler that improves the properties of the thermal paste. Thermal pastes are used in a wide range of applications including electronics, automotive and industrial applications.

In electronics, thermal pastes are used to improve heat transfer between a computer's processor and its cooler. They can also be used to improve heat transfer between transistors, diodes, and other electronic device components. Without proper heat transfer, processors can overheat, resulting in reduced efficiency or damage. This is what causes increased interest in the possibility of improving the existing heat-conducting properties of thermal pastes.

In the industrial sector, thermal pastes are used to improve heat transfer between various equipment components such as motors, pumps and compressors. They can also be used to improve heat transfer in refrigeration and air conditioning systems and in industrial processes that require precise temperature control. Thermal pastes are also widely used in medical applications for medical equipment such as lasers, magnetic resonance imaging and other devices where precise and efficient heat management is required.

Based on the above, thermal pastes are an important material for efficient heat transfer in various industries and applications.

Also recently, carbon nanotubes (CNTs) have become widely used in the field of electronics and microelectronics due to their unique physical and chemical properties. One area where CNTs can be effectively applied is to improve the thermal conductivity of electronics. A group of scientists previously conducted a comparative study of the thermal conductivity of high-conductivity thermal paste ($k = 8.9$ W·(mK)$^{-1}$) when modified with coal-derived multilayer graphene (coal-MLG) with thermal conductivity achieved using other nano-additives, such as: carbon nanotubes (MWNTs), graphene nanoplatelets (GNPs), graphite-based multilayer graphene (g-MLG) [1].

This articleconsiders the influence of CNTs on the heat-conducting properties of the KPT-8 brand thermal paste, the thermal conductivity of which is in the range of 0.8–1.0 W·(mK)$^{-1}$, which can be quite relevant to improve the quality of thermal pastes presented on the Russian market of components for electronic equipment.

Previously, it was found that carbon nanotubes have outstanding heat-conducting properties, which are due to the unique structure and electronic properties:

1. CNTs are made up of carbon atoms that are held together by strong covalent bonds. These bonds are strong and ensure the structural integrity and stability of the nanotubes. CNTs have high strength and stability, which allows them to maintain their structure even at high temperatures and mechanical stress. Due to these bonds, they have high thermal conductivity, as they allow efficient transfer of heat between carbon atoms. All of the above makes it possible to ensure high thermal conductivity of carbon nanotubes and composites based on them [2–5].

2. Carbon nanotubes have a high degree of crystallinity; carbon atoms are strictly ordered. This allows phonons (elementary excitations of the lattice) to move through the structure of nanotubes without significant energy loss, contributing to thermal conductivity. The crystal structure of CNTs has a direct effect on their electronic, optical, and mechanical properties. Different types of CNTs with different chiral vectors and twist angles have different electronic structures and energy bands [6–8].

3. CNTs have very small sizes in one dimension, which leads to a limitation of the free path of phonons. This means that phonons rarely have defects and impurities, which contributes to the efficient heat transfer [9, 10].

4. Carbon nanotubes have the shape of a cylinder and are a structure with one-dimensional properties. This structure allows them to exhibit effective thermal conductivity, electrical conductivity and mechanical strength. The one-dimensional structure of CNTs also provides them with high surface activity and accessibility for interaction with other materials [11–13].

5. Nanotubes have good electrical conductivity due to the presence of conductive electronic states; the interaction between electrons and phonons contributes to effective thermal conductivity [14–17].

Thus, this study aims to analyze the possibility of using carbon nanotubes as a doping element in thermal paste, which is a fairly common product on the Russian market, to determine the effectiveness of this method to improve an existing material and to select the required concentration of CNTs to determine and obtain the most optimal properties of thermal paste.

2. Materials and Methods

2.1. Materials

The KPT-8 thermal paste, which is quite popular on the market, was chosen as the test sample. At present, *Elox-Prom CJSC* stands out of the manufacturers of this brand of thermal paste, which is one of the few manufacturing companies in Russia that manufactures thermal paste in full compliance with Russian Standard 19783-74. Also, a fairly popular company producing the KPT-8 brand thermal paste is *SOLINS*, which makes goods for radio electronics and electrical works. For this study, the *Connector* thermal paste was used, which is also manufactured in compliance with Russian Standard 9783-74, while having a low cost and sufficient efficiency.

Thermal paste was prepared by thickening polydimethylsiloxane liquid with zinc oxide powder and aerosil and has the characteristics shown in Table 1.

Polydimethylsiloxane (PDMS) fluid provides thermal stability thermal paste to ensure its stability in high temperature conditions, the hydrophobicity of this compound allows you to protect the processor from moisture in adverse environments. This property will also prevent doping carbon nanomaterials from attaching water molecules to their surface. In addition, PDMS has excellent electrical insulating properties and low viscosity, which affects the uniform distribution of carbon nanotubes in the mass of thermal paste and uniform application of the composite material on the surface.

Pyrogenic silicon dioxide (PSC) is a highly dispersed powder consisting of amorphous silicon oxide nanoparticles. Its main properties, such as a large surface due to the small size and high porosity of the particles, the ability to withstand high temperatures without a significant change in its properties, and dispersibility in polymer matrices, make it possible to evenly distribute MPC particles in the material and improve its mechanical, physicochemical, and heat-conducting properties.

Table 1. Characteristics of the KPT-8 thermal paste used in the study

Color	White
Safety	Explosion-proof, non-flammable, chemically inert, does not have an irritating or toxic effect on humans
Corrosive effect	No green on copper plate within 24 hours
Working temperature range	From −60 to +180 °C
Density	2.6–3.0 g·cm^{-3}
Specific volume electrical resistance	Not less than 1012 Ohm·cm
Electrical strength	2.0–5.0 kV·mm^{-1}
The dielectric constant	50 Hz – 6.0; 1 MHz – 4.0; 10 MHz – 4.8
Penetration	110–175 mm
Dissipation tangent at 10 MHz	Not less than 0.005
Dynamic viscosity at 20 °C	130–180 Pa·s
Radiation resistance	Allowable integral radiation dose – 1.25 × 10^{8} Rad
Thermal conductivity, W·(mK)$^{-1}$, not less than	–50 °C – 1.0; 20 °C – 0.7; 100 °C – 0.65

Zinc oxide (ZnO) has good conductive properties, because it is a wide bandgap semiconductor and also has high thermal stability, making it resistant to high temperatures.

As a doping element, multi-walled carbon nanotubes of the Taunit-M brand manufactured by LLC NanoTech-Center (Tambov) were chosen.

2.2. Preparing CNTs to add to thermal pastes

To improve the quality of the carbon nanomaterial, an ultrasonic cleaner PS-G20 (ultrasonic cleaner PS-G20) was used. The impact of ultrasound on carbon nanotubes was carried out in several stages with duration of 20 minutes and a frequency of 40 kHz. In this experiment, the effect of acoustic flow on CNTs was important for the destruction of the formed lumps and the formation of nanotube conglomerates. Previously, nanotubes were placed in special sealed containers to prevent water from getting inside them.

To determine the effect of CNTs on the thermal conductivity of thermal paste, the following steps were taken in practice:

1. *Preparation of thermal paste samples.* Three modifications of the KPT-8 thermal paste were made with different concentrations of CNTs in them by conventional stirring with a plastic spatula and subsequent filling of the container (syringe) for more convenient storage, transfer and dosage of the thermal paste:

– 0.0005 g of CNTs per gram of thermal paste (0.05 %);

– 0.001 g of CNTs per gram of thermal paste (0.1 %);

– 0.005 g of CNTs per gram of thermal paste (0.5 %).

2. The surfaces of the heat sinks adjacent to the processor cover were wiped to a shine and degreased. These manipulations were carried out in accordance with the requirements of thermal paste manufacturers to ensure optimal interaction of the heat-conducting material with the surface, the absence of air gaps and contaminants. After cleaning the surfaces, a layer of thermal paste with a volume of 0.2 ml was applied to them.

3. Built-in sensors were used to measure the processor temperature, the heat flow was directed from top to bottom, and the access to air was free.

4. Thermal paste tests were carried out indoors at the same ambient temperature (25 °C) and under the same load on the processor (100 %).

5. Comparison tests with base materials or other types of thermal pastes are often carried out to evaluate the effectiveness of modified thermal paste and its thermal conductivity properties. This is exactly what was done, the thermal paste with the addition of CNTs was compared with the same thermal paste, but without the addition of CNTs.

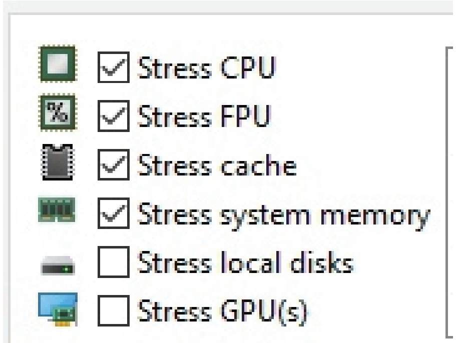

Fig. 1. Program Settings AIDA 64

Testing was carried out on 2 personal computers that had the following configurations:

1) CPU – Intel(R) Core(TM) i3-2120 CPU @ 3.30GHz TDP 65W;

2) CPU – Pentium(R) dual core E6800 @ 3.30GHz TDP 65W.

Testing was carried out in the AIDA64 program with the following settings (Fig. 1).

Temperature measurements were taken both in the AIDA64 program and in the OCCT program for confirmatory control of measurements and elimination of measurement errors. The testing time was 15 min. During the study, the temperature was recorded at the beginning of testing (0 min), after 3 min, 5, 10 and 15 min. Also, to obtain more reliable results, the processors' relaxation time between tests was 10 min.

These studies were carried out in order to create a synthetic load on the processor to achieve maximum temperature and to test the effectiveness of heat dissipation with modified thermal paste.

3. Results and Discussion

The results of the study showed that the thermal paste with CNT additives had a higher thermal conductivity than the thermal paste without additives. This suggests that CNTs can be effectively used as additives to improve the thermal conductivity of thermal pastes.

The graphs (Fig. 2 and Fig. 3) show that thermal paste with concentrations of 0.05 and 0.1 % are more stable to cope with the distribution and removal of heat.

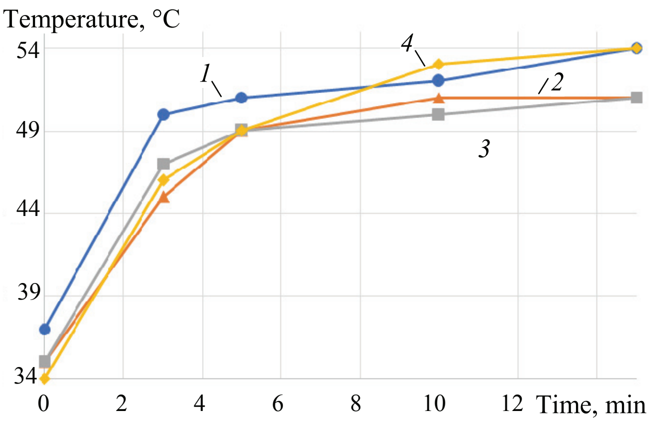

Fig. 2. Pentium (R) dual core E6800 processor temperature plot, %:
1 – 0; *2* – 0.05; *3* – 0.10; *4* – 0.50

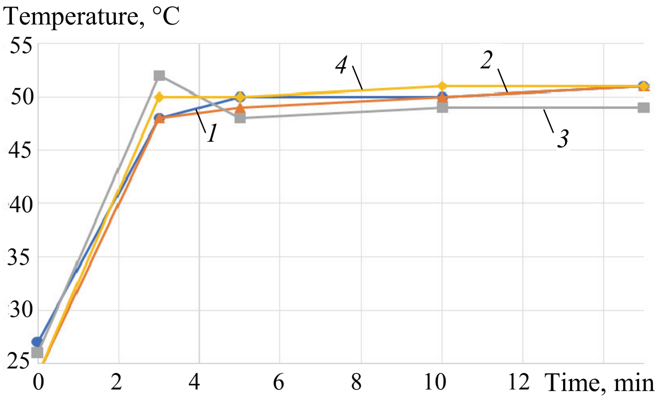

Fig. 3. Intel (R) Core (TM) i3-2120 CPU temperature plot, %:
1 – 0; *2* – 0.05; *3* – 0.10; *4* – 0.50

However, it should be noted that certain problems may arise when CNTs are used as additives in thermal pastes, such as clustering of CNTs in the thermal paste. This can lead to an uneven distribution of the nanomaterial in the thermal paste and, as a result, to inhomogeneous thermal conductivity. Figure 4 clearly shows the zones of CNT grouping in thermal paste with a concentration of 0.5 %, due to which heat transfer occurs unevenly.

Thus, the study showed that CNTs can be effectively used as additives to improve the thermal conductivity of thermal pastes. Thermal paste with CNT additives has a higher thermal conductivity than thermal paste without additives. However, for the effective use of CNTs as an additive in thermal pastes, it is necessary to solve the problem of CNT grouping, since during normal mixing with a plastic spatula, the thermal paste itself does not get between the carbon nanotubes and, in fact, air bubbles are formed, and the air, in turn, is a heat insulator.

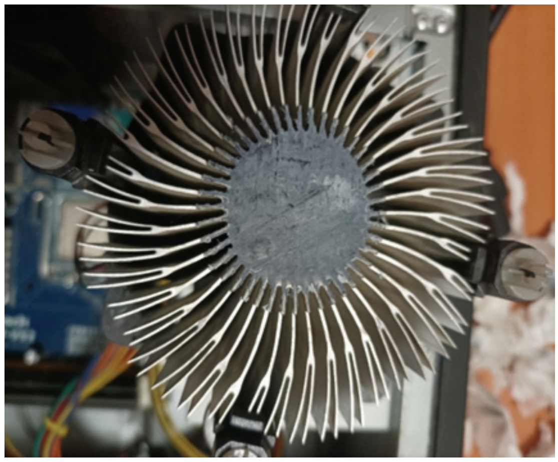

Fig. 4. CNT grouping zones

In order to increase the efficiency of carbon nanomaterial and improve the quality of thermal paste, it is necessary to treat nanotubes with ultrasound.

Having treated CNTs in an ultrasonic bath of the PS-G20 model (ultrasonic cleaner PS-G20), in several stages with a duration of 20 minutes and a frequency of 40 kHz, we prepared and tested 3 more samples of thermal paste, to which carbon nanomaterial was added in the same percentage ratio, as in the experiment described above. The mixing method and ambient temperature (25 °C) also remained unchanged.

The results of repeated testing (Fig. 5 and Fig. 6) showed that thermal paste with addition of sonicated CNTs had higher thermal conductivity than thermal paste with non-sonicated CNTs.

At the same time, CNT conglomerates in samples with a high concentration of nanotubes were observed much less frequently, while in samples with a lower concentration, they were not observed at all.

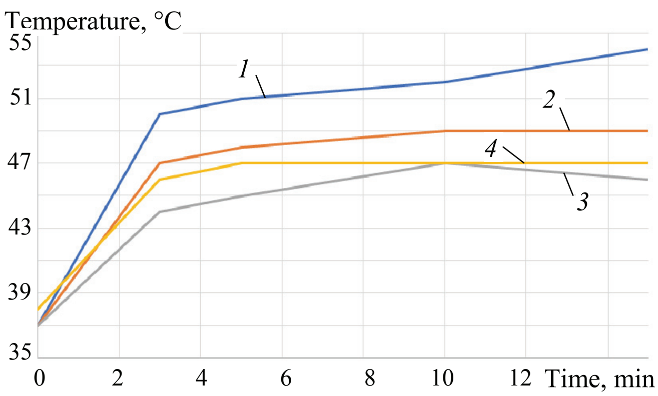

Fig. 5. Pentium (R) dual core E6800 processor temperature plot *vs.* time after injection of sonicated CNTs, %:
1 – 0; *2* – 0.05; *3* – 0.10; *4* – 0.50

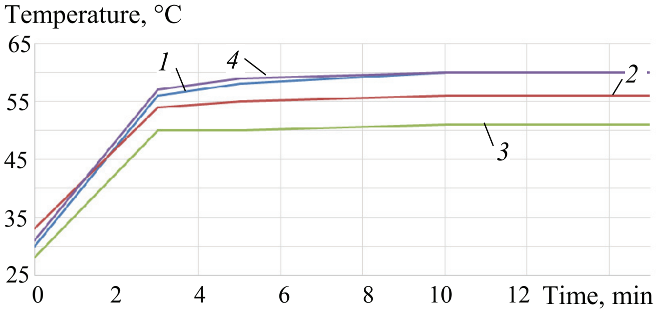

Fig. 6. Intel (R) Core (TM) i3-2120 CPU temperature plot *vs.* time after injection of sonicated CNTs,%: *1* – 0; *2* – 0.05; *3* – 0.10; *4* – 0.50

This suggests that the preliminary exposure of CNTs to ultrasound is a method of preventing the presence of groups and is of great importance.

4. Conclusion

The paper analyzed the results of measuring the temperature of Intel(R) Core(TM) i3-2120 CPU and Intel(R) Core(TM) i3-2120 CPU processors, in which the KPT-8 thermal paste was used without the addition of carbon nanomaterial and the proposed new composite, based on thermal paste mixed with CNTs in various concentrations. The presence of nanomaterial, taken in each of the percentage options, had a positive effect on the thermal conductivity of thermal paste, increasing it. At the same time, the concentration of nanotubes taken in the amount of 0.1% of the total weight of thermal paste was optimal for achieving the effect of better heat removal. It was also found that prior to the introduction of carbon nanotubes into thermal paste, it is necessary to pre-treat the nanomaterial with ultrasound, which ensures better dispersion of CNTs in the mass of thermal paste and avoids the subsequent formation of air cavities in the thickness of the layer deposited on the cooled surface.

Thus, it can be concluded that CNTs are effective modifiers of thermal paste, and can improve its heat-removing characteristics. The application of such nanocomposites based on the KPT-8 thermal paste will improve the efficiency of computer equipment and extend its service life.

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6. Conflict of interests

The authors declare no conflicts of interest.

References

1. Bharadwaj B, Singh P, Mahajan PL. Thermal performance of different carbonaceous nanoparticles as additives to thermal paste as an interface material. *ASME 2021 International Technical Conference and Exhibition on Packaging and Integration of Electronic and Photonic Microsystems2021, 26-28 October 2021, Virtual, Online.* 2021;IPACK2021-69254. DOI:10.1115/IPACK2021-69254

2. Kumanek B, Janas D. Thermal conductivity of carbon nanotube networks: a review. *Journal of Materials Science*. 2019;54:7397-7427. DOI:10.1007/s10853-019-03368-0

3. Yang L, Chen ZG, Dargusch MS, Zou J. High performance thermoelectric materials: progress and their applications. *Advanced Energy Materials.* 2018;8:1-28. DOI:10.1002/aenm.201701797

4. Blackburn JL, Ferguson AJ, Cho C, Grunlan JC. Carbon-nanotube-based thermoelectric materials and devices. *Advanced Materials.* 2018;30:1704386. DOI:10.1002/adma.201704386

5. Cheung KY, Segawa Y, Itami K. Synthetic strategies of carbon nanobelts and related belt-shaped polycyclic aromatic hydrocarbons. *Chemistry*. 2020;26(65):14791-14801. DOI:10.1002/chem.202002316

6. Parker CB, Raut AS, Brown B, Stoner BR, Glass JT. Three-dimensional arrays of graphenated carbon nanotubes. *Journal of Materials Research.* 2012;27(7):1046-1053. DOI:10.1557/jmr.2012.43

7. Gupta N, Gupta SM, Sharma SK. Carbon nanotubes: synthesis, properties and engineering applications. *Carbon Letters*. 2019;29:419-447. DOI:10.1007/s42823-019-00068-2.

8. Farhat S, Scott CD. Review of the Arc process modeling for fullerene and nanotube production. *Journal of Nanoscience and Nanotechnology*. 2006;6(5):1189-1210. DOI:10.1166/jnn.2006.331

9. Zhang Z, Ouyang Y, Cheng Y, Chen J, Li N, Zhang G. Size-dependent phononic thermal transport in low-dimensional nanomaterials. *Physics Reports*. 2020;860:1-26. DOI:10.1016/j.physrep.2020.03.001

10. Anufriev R, Maire J, Nomura M. Review of coherent phonon and heat transport control in one-dimensional phononic crystals at nanoscale. *APL Materials*. 2021;9:070701. DOI:10.1063/5.0052230

11. Collins PG, Avouris P. Nanotubes for electronics. *Scientific American*. 2000;28(6):62-69. DOI:10.1038/scientificamerican1200-62

12. Karousis N, Tagmatarchis N, Tasis D. Current progress on the chemical modification of carbon nanotubes. *Chemical Reviews*. 2010;110(9):5366-5397. DOI:10.1021/cr100018g

13. Eatemadi A, Daraee H, Karimkhanloo H, Kouhi M, Zarghami N, Akbarzadeh A, Abasi M, Hanifehpour Y, Sang WJ. Carbon nanotubes: properties, synthesis, purification, and medical applications. *Nanoscale Research Letters*. 2014;9(1):393. DOI:10.1186/1556-276X-9-393

14. Tarhini A, Tehrani-Bagha AR. Advances in preparation methods and conductivity properties of graphene-based polymer composites. *Applied Composite*

Materials. 2023;184:107797. DOI:10.1007/s10443-023-10145-5.

15. Mishra S, Jena S, Mohapatra S, Das S, Das K. Mechanical behavior and texture evolution of integrated MCNTs/Cu composites with balanced electrical/thermal conductivity. *Powder Technology*. 2023;427:118751. DOI:10.1016/j.powtec.2023.118751

16. Si R, Zhang Z, Liu C, Peng Y, Bai X, Feng B, Chen J, Gao J, Miao L. Decoupled electron and phonon transport in thermoelectric GeTe compounded with multi-walled carbon nanotubes. *Materials Today Physics*. 2023;34:101081. DOI:10.1016/j.mtphys.2023.101081.

17. Muchuweni E, Mombeshora ET. Enhanced thermoelectric performance by single-walled carbon nanotube composites for thermoelectric generators: A review. *Applied Surface Science Advances*. 2023;13:100379. DOI:10.1016/j.apsadv.2023.100379

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