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Highly thermally conductive polymer composite nanomaterials based on exfoliated boron nitride and graphite nanosheets

Relevance of the study

Rapid development of integrated circuits and advanced technologies (internet of things, high-precision scanning systems, 5G/6G, etc.), along with the miniaturization and increased autonomy of electronics, are leading to increased heat dissipation and exacerbating the problem of dissipating of the excess heat generated by electronic devices. An increase in the average temperature of an electronic device, and especially local overheating, leads to deterioration in its performance, and exceeding a certain critical temperature can lead to device failure.

The most common thermal interfaces are thermal pads, which are flexible polymer composite materials (PCM) and have following advantages over their analogues: low density and cost, easy manufacturing and installation process, high electrical insulation properties, and ability to produce materials with controlled anisotropy. The anisotropy of the thermal conductivity of polymer composites in cooling systems provides for optimal heat distribution and resolves the problem of "hot spots" – areas with temperatures significantly higher than the device average. Presence of these hot spots determines the overall reliability and efficiency of electronic devices.

For the anisotropic polymer composites manufacture, it is necessary to use anisometric particles (1D or 2D) and/or structure and align filler particles within the polymer matrix (3D structures). First method is the simplest, and use of a 2D filler being preferable due to its smaller specific surface area (which allows for the production of PCMs with a higher filler content and, consequently, with an increased thermal conductivity) and lower thermal resistance at the filler-filler interface due to in-plane contact between the particles. Among the wide variety of 2D materials, the most common for thermally conductive PCMs production are graphene and boron nitride nanosheets (BNNS). Despite graphene's higher thermal conductivity (3500-5300 W/m·K), its utilization as a filler for PCMs in cooling systems is limited by its high electrical conductivity, which is unacceptable in a number of applications, such as high-voltage and high-frequency power electronics and closed integrated circuits. BNNS, in turn, have excellent dielectric properties and a relatively high intrinsic thermal conductivity (over 300 W/m·K).

Numerous methods for synthesizing 2D materials have been described in the literature, including chemical vapor deposition (CVD), hydrothermal synthesis, ball milling, chemical, electrochemical, and ultrasonic (US) exfoliation and others. Among these methods, ultrasonic liquid-phase exfoliation appears to be the most promising, characterized by its simple implementation, versatility, scalability, and the ability to produce low defect 2D materials due to the absence of chemical treatment steps (intercalation, oxidation).

One of the most important factors influencing the yield and quality of the resulting BNNS and exfoliated graphite (EG) during ultrasonic exfoliation is the choice of dispersion medium (DM). Currently, literature does not provide a comprehensive explanation of the mechanism of ultrasonic exfoliation; however, most researchers choose a thermodynamic approach based on the concept of minimizing the Gibbs energy of mixing of particles and DM. Thus, a relevant task is to study the thermodynamics of the exfoliation process, develop simple and effective methods for obtaining 2D materials, create heat-conducting polymer composites based on them, and study their thermophysical and electrophysical properties.

Aim of the work

Development of highly thermally conductive polymer composite nanomaterials based on exfoliated boron nitride and graphite nanosheets and methods for producing boron nitride and graphite nanosheets with a high yield (over 10%) through the rational selection of a dispersion medium and ultrasonic exfoliation process parameters.

Objectives of the study:

1) Selecting a dispersion medium for ultrasonic exfoliation that ensures a high yield and lateral dimensions with a small thickness of the resulting nanosheets;

2) Determining the influence of the nature and concentration of the dispersed phase, as well as the exfoliation duration, on the yield of exfoliated particles;

3) Obtaining polymer composite materials with high thermal conductivity coefficients and thermal conductivity anisotropy values;

4) Obtaining polymer composite materials with a mixed filler consisting of microparticles and nanosheets to improve thermal properties;

5) Improving the thermal stability and thermal stability of the thermophysical properties of polymer composite materials through the use of high concentrations of filler microparticles and nanosheets.

Scientific novelty

1) For the first time, it has been demonstrated that exfoliation in an ultrasonic bath allows for the production of highly concentrated suspensions (up to 3.3 g/l) of boron nitride and graphite nanosheets in water-alcohol medium with a yield of 8-14%.

2) For the first time, the effect of the composition of a mixed filler consisting of microparticles and nanosheets obtained by ultrasonic exfoliation on the thermophysical properties of polymer composite materials has been established. It has been shown that the maximum thermal conductivity corresponds to a nanosheet content of 5-10% in a mixture with microparticles.

3) Dependence of the nature of the dispersion medium, the initial concentration of the layered crystal on the yield and concentration of boron nitride and graphite nanosheets, and the exfoliation duration has been established.

4) Highly thermally conductive polymer composite materials with highly anisotropic thermal conductivity (based on graphite) and high dielectric properties (based on hexagonal boron nitride) have been obtained. Their properties meet the requirements for modern electronic devices cooling and exceed published literature data.

Theoretical and practical significance

1) Results of the study of the influence of the nature of the dispersion medium for exfoliation of hexagonal boron nitride and graphite, exfoliation time, the nature and initial concentration of particles on the yield and concentration of nanosheets after exfoliation are of theoretical significance for improving and scaling up ultrasonic exfoliation methods of layered particles and developing a theoretical foundation for the exfoliation process.

2) Developed method of ultrasonic exfoliation of hexagonal boron nitride and graphite in low-boiling dispersion media without surfactants utilization with a high yield has practical significance for production organization of nanosheets.

3) Results of the development of polymer composite films, including those using nanosheets with high thermal conductivity and thermal stability, as well as high dielectric characteristics, are of practical importance for the use of these materials as thermal interface materials for modern heat-loaded electronic devices cooling.

Provisions submitted for defense:

1) Influence of the nature of the dispersion medium during ultrasonic exfoliation on the morphology (lateral dimensions, thickness) and yield of the resulting nanosheets;

2) Influence of the concentration of hexagonal boron nitride and graphite and their exfoliation time on the yield of exfoliated particles;

3) Conditions for producing polymer composite materials with high thermal conductivity and thermal stability;

4) Conditions for producing polymer composite materials with a mixed filler consisting of microparticles and nanosheets, and the influence of the mixed filler composition on the thermophysical properties of the resulting composite materials;

5) Patterns of temperature influence on the thermophysical properties of the resulting composite materials.