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連続繊維/BN/シリコーンゴム熱伝導複合材料の創製および性能評価
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論 文 内 容 要 旨

Currently, with the development of industries such as electronics, information technology, communications, and energy, there is an increasing demand for high-density electronic components in integrated circuits. This trend has led to the miniaturization of electronic devices while increasing their power consumption. The rising power density makes electronic devices susceptible to temperature increases due to heat accumulation, which can significantly impact their performance, operational safety, and lifespan. In light of this situation, composites with high thermal conductivity and electrical insulation properties hold great promise in various fields, including electronics, information technology, and energy. Silicone rubber (SR) is widely used in the preparation of flexible thermal-insulating composites due to its high flexibility, excellent resistance to extreme temperatures, thermal stability, and outstanding electrical insulation properties. SR serves purposes such as heat dissipation, shock absorption, puncture resistance, and sealing. However, existing thermal-insulating SR composites still face challenges, including low thermal conductivity and instability in performance when the filler content is high. These issues prevent them from simultaneously meeting the requirements of high thermal conductivity, electrical insulation, and flexibility. Furthermore, many of the methods currently used to enhance the thermal conductivity of composites are not suitable for large-scale production and industrial applications.

This thesis addresses the aforementioned issues by attempting to build a thermal conductivity network through the combined use of thermally conductive fillers of different forms. Simultaneously, it leverages the varying electrical conductivity of these fillers to interrupt the conductive network. The aim is to create silicone rubber composites that exhibit high thermal conductivity while retaining electrical insulation properties. Additionally, this research explores the continuous production of thermal-conductive silicone rubber composites by employing continuous, highly thermally conductive fiber materials as fillers. This approach seeks to achieve a balance between composite performance and the continuous, large-scale production of thermally conductive silicone rubber.

In chapter 1, the main introduction includes the background of the research, various thermal conduction principles, thermal conductive filler materials, methods to enhance the thermal conductivity of composite materials, some reported thermal conductive silicone rubber composite materials, and the purpose of this study.

In chapter 2, the main introduction includes the materials used in the experiments, the testing apparatus, and the general preparation method of silicone rubber in the experiments.

In chapter 3, various forms of boron nitride (BN), graphite, and carbon nanotubes (CNT) were used as fillers. These fillers were individually or in combinations dispersed in SR through mechanical mixing to prepare thermally conductive silicone rubber. The study investigated the enhancement effect of different sizes and shapes of single fillers or composite fillers on the thermal conductivity of SR. The influence of filler content and composition on the curing process, thermal conductivity, and mechanical properties of silicone rubber was discussed. Ultimately, the most suitable individual powder filler and mixed powder filler combination were determined. When the total powder filler content was 20 wt%, the highest thermal conductivity was achieved with solely filled BN, reaching up to 0.526 W m^{-1} K^{-1}. When BN and graphite were mixed as fillers, the thermal conductivity reached up to 0.685 W m^{-1} K^{-1}. The composite also exhibited satisfactory mechanical properties, meeting the requirements of relevant applications. Additionally, incremental curing allowed silicone rubber to be used as an insulating coating. Incremental curing had a minor impact on the mechanical properties of SR. The coating effectively bonded with the composite without the need for adhesives, and the coating interface had a minimal effect on the thermal conductivity of BN/SR. Through this method, we successfully prepared and controlled the electrical conductivity of high thermal conductivity silicone rubber composites.

In chapter 4, continuous carbon fiber (CF), (BN, and SR were used as raw materials. The CF/SR or CF/BN/SR composites with vertical alignment were prepared by infiltrating CF with SR first or mechanically winding BN/SR around CF. Under the influence of traction and CF compression, BN was distributed along the CF direction. This arrangement facilitated the orientation of BN along the fibers, enabling the formation of independent thermal pathways within SR. With the increase in BN content, when CF content was 30 vol% and BN content was 10 vol%, the thermal conductivity of CF/BN/SR composite reached 2.789 W m^{-1} K^{-1}. Additionally, an insulating BN/SR coating was prepared on the surface of the CF/BN/SR composite through incremental curing. This significantly increased the volume resistivity of the composite while maintaining high thermal conductivity.

In chapter 5, a method involving continuous fiber winding was employed to prepare PBO/BN/SR thermally conductive insulating SR using inherently electrically insulating continuous PBO fibers and BN as fillers. Low viscosity LSR was used as a transition material to reduce defects in the PBO/BN/SR composite. When the PBO content was 20 vol% and BN content was 10 vol%, the thermal conductivity of PBO/BN/SR was 7.853 W m^{-1} K^{-1}. The PBO/BN/SR composite heated from 35°C to 100°C in just 19.4 seconds and cooled from 100°C to 35°C in 28 seconds. Using continuous fibers in this method facilitates continuous production. Furthermore, by controlling factors such as traction speed and LSR viscosity, the PBO fiber content can be precisely regulated.

In Chapter 6, all the experimental results were summarized. SR composites with high thermal conductivity were prepared by filling continuous fibers and boron nitride.

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論文審査結果要旨

本論文では、柔軟性を有する熱伝導複合材料を開発するために、シリコーンゴム(SR)の母材に窒化ホウ素(BN)粒子や連続繊維を充填させ、高い熱伝導性を有する複合材料の創製方法とその性能を検討している。本論文は全６章で構成されている。

第１章は緒論として、既存の研究から熱伝導材料の背景を紹介し、本研究の背景を述べ、本研究の目的を示している。第２章では、材料および試験方法として、熱伝導シリコーンゴム複合材料の作製方法とその性能評価方法を説明している。第 3 章では、粉末熱伝導フィラーを添加することにより、シリコーンゴム複合材料の熱伝導率が向上することを示している。さらに、選択した BN フィラーを用いて、増量硬化法で BN/SR 絶縁コーティングし、その界面の力学的性能と熱伝導率への影響を評価した。第４章では、第３章で作製した BN/SR 材料をマトリクスとして、開発した巻き付け方法で、連続炭素繊維（CF）で充填された CF/BN/SR 熱伝導複合材料の作製に成功し、その複合材料の熱伝導率を大きく向上させた。また、絶縁コーティングにより、CF/BN/SR 複合材料の導電性の上昇を抑えている。第５章では、より柔軟的で電気絶縁性も有するポリビフェニレンオキシビフェニレンテトラゾール繊維（PBO）を CF と置き換え、同じ巻き付け方法で PBO/BN/SR 熱伝導複合材料を作製した。その結果、PBO/BN/SR 複合材料の熱伝導率が $7.853\ \mathrm{W\,m^{-1}\,K^{-1}}$ まで大きく向上している。第６章は結論であり、研究結果をまとめている。

この論文の成果は、連続繊維を充填した柔軟性を有する高熱伝導複合材料の開発に関する理論と技術において重要な学術的貢献を果たすだけでなく、その高い性能により優れた工学的価値を持っている。さらに、研究成果として、査読付きの学術論文４編、国際会議１件、国内会議３件を公表している。

よって、本論文は博士（工学）の学位論文として合格と認める。