



Article **Tunable Head-Conducting Microwave-Absorbing Multifunctional Composites with Excellent Microwave Absorption, Thermal Conductivity and Mechanical Properties**

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Abstract: Developing composite materials with both thermal conductivity and microwave absorption is an effective strategy to solve the problems of heat dissipation burden and microwave radiation interference caused by the development of miniaturization and high performance of portable electronic equipment. However, these properties are not easy to simultaneously implement due to the limitation of single type fillers with a single particle size, inspiring the possibility of realizing multifunctional composites with the introduction of composite fillers. In this work, using alumina (Al₂O₃) and zinc oxide (ZnO) as head-conducting fillers, carbonyl iron (Fe(CO)₅) as microwave-absorbing fillers, silicone rubber (SR) composites (Al₂O₃/ZnO/Fe(CO)₅/SR) with enhanced microwave absorption, high thermal conductivity and good mechanical properties were successfully mass prepared. It was found that the composites can achieve a thermal conductivity of $3.61 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$, an effective microwave absorption bandwidth of 10.86–15.47 GHz. Especially, there is an effective microwave absorption efficiency of 99% at 12.46–14.27 GHz, which can realize the integration of electromagnetic shielding and heat dissipation. The compact microstructure, formed by the overlapping of large particle size fillers and the filling of their gaps by small particle size fillers, is helpful to enhance the thermal conduction path and weaken the microwave reflection. The heat-conducting microwave-absorbing Al₂O₃/ZnO/Fe(CO)₅/SR composites also have the advantages of thermal stability, lightness and flexibility, providing a certain experimental basis for the research and development of high-performance and diversified composites.

Keywords: composite material; head-conducting; electromagnetic shielding; reflection loss

1. Introduction

With the development of communication technology, the requirement for miniaturization and high performance is a powerful driving force for the development of integrated portable electronic equipment. However, integrated circuit chips, RF chipsets and microprocessors are used in most devices, and they generate microwaves, resulting in electromagnetic radiation and interference. It is easy to cause mutual interference between electronic devices, such as TV screen flicker, murmur on the audio system, and information leakage, and even more seriously, it may affect human health [1,2]. Some scholars have shown that the long-term impact of electromagnetic radiation will increase the risk of cancer, heart disease and migraine [3,4]. The solutions to microwaves by different devices are mainly to use shielding materials to react to incident microwaves. The mechanisms includes reflection, absorption and transformation, and multiple reflections [5]. The shielding material attenuates the incident microwave through any or more of the three mechanisms, but the reflection of microwave is difficult to achieve completely. Therefore, not only reflection



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Copyright: © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). was needed, but also internal loss was carried out, which was converted into heat in the form of emission. On the other hand, based on the internal high heat flow in the power and miniaturization equipment, the burden of heat accumulation was increased [6–8]. It will also increase the radiation of microwaves, resulting in a more complex practical application. Therefore, the electromagnetic interference problem and the overheating problem are two major loss problems that must be considered in devices [9].

In traditional applications, conductive metal is the most common material to solve the problem of electromagnetic interference and heat dissipation. Its internal free electron motion makes have good heat dissipation performance and can realize microwave shielding. However, it is difficult for metals to absorb microwaves mainly through reflection, and their high cost, heavy weight, poor flexibility, biodegradable additives and high stiffness are not conducive to large-scale applications [10]. Fortunately, it is well known that conventional polymers with lightweight and flexible properties have low internal phonon transport capacity, resulting in low thermal conductivity. Therefore, there is an urgent need for polymer based composite materials that are lightweight, flexible, thermal stability, and especially both microwave-absorbing and head-conducting [6,11-13]. From the research and development process of polymer matrix composites with single function of thermal conductivity and microwave absorption, thermal conductivity and microwave absorption composites can also be based on polymer matrix and filler. The polymer has the advantages of light weight, high flexibility, good durability, and simple processing. The fillers absorbed by microwaves include carbon black, carbon fiber, ferrite, graphene [14–17], and metal powders such as iron, cobalt, and nickel [18-20]. The heat-conducting fillers are mainly pure metal [21], ceramic materials including zinc oxide (ZnO), alumina (Al_2O_3), silicon carbide (SiC), and boron nitride (BN) [22–26], carbon materials including graphene, carbon fiber (CF), and carbon nanotubes (CNT) [27–31]. However, it is difficult to achieve high thermal conductivity and electromagnetic shielding performance of materials simultaneously by adding a single head-conducting filler or microwave-absorbing filler. In addition, the fillers not only increase the functionality [32] but also improve the mechanical properties of the material. Nano fillers have been developed to improve the mechanical properties of polymer matrix composites [33], such as synthetic graphite [34], cellulose nanocrystals [35], melamine polyphosphate [36], and flame-retardant magnesium hydroxide [37]. Therefore, the method of combining fillers with different functions in polymers would be a good strategy.

The preparation method of composite materials is relatively easy, and the blending method is generally used. However, the incorporation of head-conducting and microwaveabsorbing mixed fillers in the polymer can easily cause interfacial effects such as agglomeration and uneven dispersion. When the filler content is high in the composite materials, it will bring problems such as increased hardness, uneven mixing and unstable performance, which increases the difficulty and cost for the development of materials [38]. The orientation of fillers in matrix also affects the properties of composites [39]. Therefore, it is necessary to consider the synergistic effect of composite materials in performance and structure design.

By comparing the effects of different factors on the molded composites, researchers tried to obtain the ratio and preparation methods for guiding significance for practical production and application [40]. Chen et al. [41] successfully prepared silicone rubber-based composites filled with holey graphene nanosheets with only 1 wt% graphene loading, which has significantly high microwave absorption with return loss reaching -32.1 dB at 13.2 GHz with a thickness of 2 mm. Ren et al. [42] successfully prepared Ag/NWF/FeCo@rGO/WPU composites through Ag/NWF conductive grid and WPU resin encapsulation. By controlling the loading of Ag at 10.5 wt%, the thermal conductivity can reach to 1.65 W·m⁻¹·K⁻¹, with excellent electromagnetic loss ability. The loss path of microwaves was extended by layered structure, reaching to -77 dB, and the reflectivity is reduced to 0.77. The mechanical properties of the composites are excellent, and the performance is stable. Zhang et al. [43] introduced low melting point alloy (LMPA) into epoxy resin to construct composite materi-

als with layered structure. The study showed that when the load amount of LMPA reached to 20 vol%, the thermal conductivity was $1.23 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$, and the electromagnetic loss efficiency could reach to -35.56 dB at 30 GHz. It can be seen from the above research that there are many factors that affect the thermal conductivity and microwave absorption performance of composite materials. It is necessary to study many factors such as filler ratio, particle size, shape, and molding process parameters. In the composite materials, the microwave-absorbing and heat-conducting fillers are required to be uniformly dispersed to play the intrinsic characteristics, and the continuous network chain structure is also required. Thus, it has the coordinated development of structure and performance and plays the advantages in electromagnetic loss and thermal conductivity. In general, the research system of both thermal conductivity and electromagnetic shielding composite materials is becoming more and more perfect, and the application of the composite materials in the fields of communication and navigation is also becoming more and more common. It not only protects the working space, environment, and equipment from the influence of microwave radiation, but also improves the efficiency and life of the device.

In this work, spherical ZnO and Al_2O_3 composite fillers were selected as heat-conducting fillers, because they all have the characteristics of relative economy, strong applicability and convenient operation; and carbonyl iron ((Fe(CO)₅) was selected as microwave-absorbing filler, because it has the advantages of low cost, high saturation magnetization and permeability, wide absorbing frequency band, and good absorbing effect. A facile approach of mass production of $Al_2O_3/ZnO/Fe(CO)_5/SR$ composites by mixing and vacuum pressure technology was reported. This method is relatively simple and can be widely used in practical mass production as described in our previous work [24]. The electromagnetic properties, thermal conductivity, thermal resistance, and mechanical properties of as-prepared $Al_2O_3/ZnO/Fe(CO)_5/SR$ composites were investigated. To reveal the thermal conductivity enhancement and microwave absorption mechanisms, the cross-section microstructures of the composites were analyzed.

2. Experimental

2.1. Materials

The Al_2O_3 powder and ZnO powder used as heat-conducting fillers were purchased from Guangdong Letu New Material Co., Ltd., Dongguan, China. The Fe(CO)₅ powder used as microwave-absorbing filler was purchased from Shenzhen Guangan Chemical Industrial Co., Ltd., Shenzhen, China. The properties of powder materials is shown in Table 1. The two-component silicone oil (including vinyl silicone oil and hydrogencontaining silicone oil) used as polymer matrix was purchased from CHT Tianjin Co., Ltd. Tianjin, China, which undergoes an addition reaction in the presence of a platinum catalyst to improve its mechanical properties. In addition, the catalysts mentioned above and small quantities of curing agents and inhibitors (ethynyl cyclohexanol) for preparation were all purchased from Guangzhou Siyou New Materials Co., Ltd., Guangzhou, China. All chemicals (analytical grade) were used as purchased without further treatment.

Table 1. Properties of powder materials.

Raw Materials	Mean Diameter (µm)	Particle Size Distribution D50 (μ m)	Thermal Conductivity W/(m \times K)	Microwave-Absorbing Properties
ZnO/Al ₂ O ₃ power	20.3	15.799	29.98	moderate
Fe(CO) ₅ power	1.9	1.799	7.85	strong

2.2. Preparation of the Al₂O₃/ZnO/Fe(CO)₅/SR Composites

The $Al_2O_3/ZnO/Fe(CO)_5/SR$ composite slurry was prepared by a simple blending method. First, the vinyl silicone oil and hydrogen-containing silicone oil were mixed in a molar ratio of 1:1 to obtain a silicone rubber matrix after stirring for 20 min. To prolong the reaction time between the silicone oils, a small amount of inhibitor was added before stirring. Next, heat-conducting powders (Al_2O_3 and ZnO) and microwave-absorbing

powders (Fe(CO)₅) in a certain proportion were slowly filled into the silicone rubber matrix, while stirring for about 1 h during the addition until completely mixed. The total amount of the powder fillers is 94% of the silicone rubber matrix. The mass ratio of the head-conducting powders and microwave-absorbing powders is 1:2, 1:1 and 2:1. And the mass ratio of Al₂O₃ and ZnO is 1:1. The composites containing only microwave-absorbing fillers and only head-conducting fillers in the silicone rubber matrix were marked as AM and TC, respectively. The composites containing both head-conducting fillers and microwave-absorbing fillers with the mass ratio of 1:2, 1:1 and 2:1 were marked as TA-1, TA-2 and TA-3, respectively. Then, a trace amount of catalysts and curing agents was added and stirred for 1 h. Finally, the mixture was put into a vacuum defoaming machine to remove air bubbles completely, and the composite slurry was prepared.

The Al₂O₃/ZnO/Fe(CO)₅/SR composites were prepared with the composite slurry by the coating method. First, the composite slurry was poured naturally in the center of the PET film on a coating machine, avoiding mixing in gas. The slurry was sandwiched between the upper and lower PET films, and coated into sheets with a thickness of 2 mm. Then, the sheets were baked at 120 °C for 12 min in a multi-sectional tunnel oven to produce Al₂O₃/ZnO/Fe(CO)₅/SR composite sheets. The overall preparation process is shown in Figure 1.



Figure 1. The schematic illustration of preparation process for Al₂O₃/ZnO/Fe(CO)₅/SR composite sheets.

2.3. Characterization of the *Al*₂O₃/ZnO/Fe(CO)₅/SR Composites

Scanning electron microscopy (SEM, Zeiss supra55, Oberkochen, Germany) was used to observe the morphology of the functional powder fillers and the cross-sectional morphology of the composite sheets brattled by liquid nitrogen. In addition, the analytical energy dispersive spectrometer (EDS) equipped on the SEM was used to characterize the chemical composition of the composite sheets. For the SEM test, the applied accelerating voltage is 3.00 kV, the low vacuum mode was operated, the working depth is 1.5 nm@10 kV and 1.8 nm@3 kV. To characterize the crystal structure changes of the fillers before and after compounding with rubber, the X-ray diffraction (XRD) patterns of the fillers and the composites were collected by an X-ray diffractometer (D8ADVANCE-A25, Bruker, Salbrücken, Germany) with scanning angles ranging from 10° to 80°.

The vector network analyzer (E4991B, KEYSIGHT, Santa Clara, CA, USA) and test fixtures (16453A, 16452) were manipulated to measure the electromagnetic parameters of the composites, including permittivity and permeability. The frequency range of the microwave was set from 1 MHz to 1 GHz. The composite sheets were cut into rings with an inner diameter of 15 mm and an outer diameter of 26 mm. The microwave reflection

loss was measured by the bow-shaped method. The measurement frequency ranges from 2 to 18 GHz, and the dimensions of the test composite sheets is $18 \times 18 \times 2$ cm. At least three specimens were tested for each composite sheet to obtain reliable data.

The thermal conductivity (λ) of the composites was measured by a thermal conductivity tester (DRL-III, Xiangtan Instruments, Xiangtan, China) according to ASTM D5470-2005 test standard. Three samples per configuration were tested and averaged for the thermal conductivity. The thermal loss of the pure silicone rubber and composite sheets was analyzed by thermogravimetric analyzer (TGA, Diamond TG, Hitachi, Tokyo, Japan) at a heating rate of 10 °C/min in Ar atmosphere. The real-time heating and temperature images of the composites were recorded by an infrared thermal imaging camera to obtain the thermal diffusion properties. The dimensions of the composite test specimens are 25 mm \times 20 mm \times 2 cm. The composites were placed directly on the heating table at a constant temperature of 80 °C.

According to GB. T 1040.1-2006 standard, the tensile properties and 50% compressive strength of the composites were tested by universal material testing machine (5960, Instron, Boston, MA, USA) at room temperature. Shore 00 hardness tester was used to measure the hardness of the composites according to ASTM D2240 standard.

3. Results and Discussion

3.1. Structure and Composition

The morphology of the filler is very important for the performance of the formed composites. The SEM diagrams and corresponding particle size distribution curves of the absorbing powder and head-conducting powder used in the experiment were measured, as shown in Figure 2. In general, in various forms of fillers, one-dimensional fibrous and two-dimensional sheet fillers were filled in polymers. When they have a certain orientation, the thermal conductivity of composites can be improved effectively. However, the matrix type and filler orientation method need to be carefully selected. For the spherical or quasi-spherical particles, the flowability is good, resulting in no selectivity for the matrix. The contact area between the spherical powder and the polymer is very small, leading to the realization of easier larger fillings, which is conducive to the formation of a coherent thermal conductivity network, thereby improving the thermal conductivity of the composites. Therefore, spherical fillers were selected in this work. It can be seen from Figure 2a,c that the particle morphology of the two kind of powders is regular spherical.



Figure 2. SEM images and particle size distribution curves of the functional powders. (**a**,**b**) Absorbing fillers, (**c**,**d**) head-conducting powders.

Figure 2b,d shows the particle size distribution curve of the fillers measured by particle size distribution instrument. The median diameter (D50) of the absorbent is 1.76 μ m, and the particle size is small and uniform. The D50 of head-conducting powders is 15.87 μ m, the span of particle size is large, and the distribution range is wide. This indicates that the particle sizes of the two functional powders are quite different. Generally, the contact of spherical particle surfaces will generate gaps in the matrix. The effect of mutual filling can be achieved by filling particles of different sizes, resulting in a very tight filling, thereby achieving rapid heat transfer. According to the theory of heat conduction, to achieve the synergistic effect between fillers as much as possible, the proportion of powders with different sizes should be uniform.

Figure 3 shows the cross-sectional SEM images of the composites AM, TC and TA-2. From the comparison, when different powders with the same mass were filled in the polymer, the cross-sectional morphologies of the composites are different. For composite AM filled with microwave-absorbing powder only as seen from Figure 3b, all particles were encapsulated in the polymer, indicating a good combination effect between absorbent powder and polymer. For composite TC filled with head-conducting powder only as seen from Figure 3a, pits formed by powder falling off and large particle protrusions appear in the cross section, which indicates that the adhesion of the polymer matrix to the headconducting powder is not strong. Figure 3c,d shows the cross-section SEM of the composite TA-2, in which the filling amount of the head-conducting powder and the absorbent is 1:1. As seen from its low-magnification SEM image (Figure 3c), the head-conducting powder particles with large particle sizes were uniformly dispersed in the polymer matrix. As seen from its high magnification SEM image (Figure 3d), the spherical powders have smooth surfaces, which were tightly wrapped by the polymer, and exhibited the morphology characteristics of large particles surrounded by small particles. This shows the advantage of compounding spherical powders with different particle sizes that small particles can enter the gaps among large particles, forming relatively dense packing system in the polymer. The gaps visible in the SEM images are caused by the slight movement of the large powder particles when the sample was cut with the knife. Figure 3e shows the EDS elemental analysis of the selected cross-section of the composite TA-2. It clearly exhibits that the main elements in the composite TA-2 are O, C, Si and a large amount of Al, Zn and Fe, which are all the elemental components in fillers and polymers, and there are no other impurities mixed.

The compositions of the composites and their fillers were further characterized by XRD, as shown in Figure 4. Figure 4a shows the XRD patterns of the pure head-conducting composite TC and its head-conducting powder fillers. It can be clearly seen that the XRD peak of composite TC is basically consistent with that of its head-conducting powder fillers and corresponding to the standard cards Al₂O₃-PDF#10-0173 and ZnO-PDF#36-1451. The 20 angles of 25.584°, 35.136°, 43.362°, 52.552° and 57.518° correspond to the (012), (104), (113), (024) and (116) crystal planes of α -Al₂O₃ crystal, respectively. Meanwhile, the 2 θ angles of 31.769°, 34.421°, 36.252°, 56.602° and 62.862° correspond to the (100), (002), (101), (102), (110) and (103) crystal planes of ZnO crystal, respectively. However, the absorption peak intensity of ZnO crystal is higher than that of α -Al₂O₃ crystal in the composite TC, indicating that the crystallinity of ZnO is better than that of α -Al₂O₃ in the composite. Figure 4b shows the XRD patterns of the pure microwave-absorbing composite AM and its microwave-absorbing powder fillers, which all correspond to the standard card α -Fe-PDF#06-0696. In the pattern, there is a sharp large bulge near 44.67° and a sharp small peak near 65.02°, which correspond to the (110) and (200) crystal planes of α -Fe, respectively. The other diffraction peaks fluctuate greatly, indicating that the microwave-absorbing powder filler is mainly amorphous Fe, containing a small fraction with crystalline features of Fe. However, the fluctuation of the diffraction peaks of the composites decreased, and the intensity of the diffraction peaks was reduced, which originated from the coating of the absorbent by the polymer.



Figure 3. Cross-sectional SEM images of composites: (**a**) TC; (**b**) AM; (**c**,**d**) TA-2. (**e**,**f**) Element content maps for EDS analysis of selected portions of composite TA-2.

0

Fe

Al

Zn

Si

0

С

<u>30 µm</u>

TA-2

Figure 4c shows the XRD patterns of the composite TA-2, which basically corresponds to the diffraction peaks of ZnO, α -Al₂O₃ and α -Fe. As seen from the comparison of XRD patterns of the three composites (Figure 4d), the XRD patterns of the composite TA-2 combines the XRD patterns of the composites TC and AM. However, compared with composite TC, the diffraction peak intensity of α -Al₂O₃ crystal in the composite TA-2 decreased. This is owing to the fact that the particle fillers with different sizes were closely contacted with each other, and the coating phenomenon of "large ball" surrounded by "small ball" in the polymer matrix was formed. The large particles of Al₂O₃ were coated by the small particles of ZnO, resulting in a decrease in the diffraction intensity of Al₂O₃.

Moreover, it was further verified that the composition of the head-conducting powder was Al_2O_3 and ZnO, and the composition of the microwave-absorbing powder was $Fe(CO)_5$, which was consistent with the EDS element analysis results in Figure 3e. The results indicate that the preparation method used in the experiment has no effect on the elemental composition and structure of the raw material. After the filler was mixed with the polymer matrix to prepare the composite, no chemical reactions such as corrosion and oxidation occurred, which would not affect the crystallization performance of the filler, and the internal crystal structure was not destroyed. Therefore, it would not affect the thermal conductivity provided by the head-conducting powders and the intrinsic electromagnetic properties provided by the microwave-absorbing powders.



Figure 4. XRD patterns of the composites and their fillers: (**a**) TC; (**b**) AM; (**c**) TA-2; (**d**) comparison of the three composites.

3.2. Electromagnetic Properties

For the wave-absorbing properties of head-conducting microwave-absorbing multifunctional composites, it needs to be considered from many aspects, including intuitive microwave reflection loss and electromagnetic parameters. Reflection loss (RL) is the absorption of incident microwave, which is the synergistic effect of reducing reflection and internal attenuation after microwave incident. When RL < -10 dB, the frequency width of the material is called the effective absorption width, which means that the reflected wave energy is only 10^{-1} of the incident wave energy, that is, 90% of the energy is absorbed; while RL < -20 dB, which means that the reflected wave energy is only 10^{-2} of the incident wave energy, that is, 99% of the energy is absorbed. That is, the effective loss microwave efficiency reaches to 90%, which can be called the effective absorption loss of microwave.

Figure 5 shows the reflection loss at 2-18 GHz and the bandwidth results of effective absorption loss of composite materials TA-1, TA-2 and TA-3. Figure 5a shows that the peak reflection loss of composite TA-1 is -14.9 dB at 9.8 GHz, the peak reflection loss of composite TA-3 is -12.1 dB at 16.8 GHz, and the peak reflection loss of composite TA-2 can reach to -24.7 dB at 13.4 GHz. The results indicate that the frequency of the microwave reflection loss peak increases with the decrease of the filling amount of absorbent filler. The effective absorption efficiency of the composite TA-2 can reach to 99% at 12.46–14.27 GHz, namely, the reflection loss is below -20 dB. Figure 5b shows that the electromagnetic frequency bandwidth of the three composites under -10 dB reflection loss is 3.54, 4.61 and 2.16 GHz. The three composites all show a certain degree of broadband absorbing ability, and the composite TA-2 has a wider absorbing band. The above results show that the filling ratio of absorbing powder and head-conducting powder has a significant influence on the absorbing frequency band of the composites and has a certain influence on the absorbing effect. The three composites have a certain absorbing effect at different frequencies. The composite TA-1 with the largest amount of absorbing powder filling has the best absorbing performance at relatively low frequencies, while the composite TA-3

with the least amount of absorbing powder filling has the best absorbing performance at relatively high frequencies. The possible reason is that the mixing degree of powders with different particle sizes is inconsistent with the different filling amount of the absorbing powder, which leads to the extension of the microwave absorption path of the composite material, or the different filling ratios make the impedance matching energy levels inside the composite material show different degrees of attenuation [44].



Figure 5. Absorbing properties of composite materials: (a) reflection loss, (b) effective loss bandwidth.

In order to achieve better wave absorbing effect for the composite materials, two conditions need to be met: efficient reflection loss ability and good impedance matching performance. In addition, electromagnetic parameters include dielectric constant and permeability (ε' , ε'' , μ' , μ''), which can characterize the microwave absorption properties of materials [45,46]. To obtain better microwave absorption performance, good complementarity must be exhibited between high real part values (ε' and μ') and low imaginary part values (ε'' and μ'') of the electromagnetic parameters.

Figure 6a shows the ε' value curves of the composites. With the decrease of the filling amount of the absorbent, the ε' value decreases relatively. The value of ε' represents the polarization ability of the material. Generally, the higher the filling amount of the absorbing power, the better the polarization ability of the material, and more energy can be stored after the microwave incidence. However, the content of absorbent in the composite AM is the highest, which is dependent on frequency. Therefore, its numerical fluctuation is large in different frequencies. Meanwhile, other composites with smaller filling content showed good frequency stability. Figure 6b shows the ε'' value curves of the composites, and the composite AM with the highest absorbing power content has the strongest power attenuation ability and strong dependence on frequency. Moreover, the ε'' values of all composites have an upward trend when the frequency exceeds 1 GHz, indicating that the composites may also achieve attenuation of microwaves at a higher frequency. The absorbent used in the prepared composites is Fe(CO)₅ powder. It is easy to agglomerate in the polymer matrix, leading to the increase of complex permittivity. However, after mixing with head-conducting powder, the agglomeration phenomenon was reduced, resulting in a decrease in the complex permittivity.

Figure 6c shows the variation curves of the real part μ' of the complex permeability of different composites at the frequencies ranging from 1 MHz to 1 GHz. The magnetic permeability is also relatively small with the decrease of the filling amount of the absorbing power. Moreover, at the frequency of nearly 100 MHz, the μ' values of all composites decrease with the increase of frequency, and the fluctuation of the μ' values is great. The μ' values decrease sharply to about 1, and then increase slightly, obviously showing a typical relaxation phenomenon caused by dipole motion [47]. Figure 6d shows the curves of the imaginary part μ'' of the complex permeability. At the frequency of 1–30 MHz, the μ'' only shows a small amount of fluctuation, indicating that all the composites did not undergo polarization reaction, and could not achieve the effect of absorbing and shielding microwaves at this frequency. With the increase of frequency, the μ'' value of the composites increased with the amount of absorbent filler added, and the magnitude of improvement increased. Meanwhile the magnetic energy attenuation ability of the composites increased. Generally, if the impedance matching is not considered, larger dielectric constant and permeability indicate better absorption performance of the materials. However, too high dielectric or magnetic permeability of the material will cause too high impedance, resulting in the inability of microwaves to enter the material and be absorbed. Therefore, the real part and the imaginary part of the electromagnetic parameters should be complementary.



Figure 6. Electromagnetic parameters of composite materials: (**a**) real permittivity, (**b**) imaginary permittivity, (**c**) real permeability, (**d**) imaginary permeability.

3.3. Heat Transfer Properties

To evaluate the heat transfer properties of the as-prepared composites with high electromagnetic shielding properties, the thermal conductivity of the composites with different contents of head-conducting fillers were measured, as shown in Figure 7a. The thermal conductivity of the composite AM without head-conducting filler is $1.43 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$, showing relatively low heat transfer performance. However, the thermal conductivity of the composite TA-2 reached to $3.61 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$. In addition, with the increase of the head-conducting filler content, the thermal conductivity of the composites increased exponentially. The result indicates that the addition of head-conducting powder has a significant influence on the thermal conductivity of the composite.

To further demonstrate the heat dissipation performance of the composites, the temperature variation trend of different composites was observed on a constant temperature heating platform at 80 °C, and recorded by an infrared thermal imager, as shown in Figure 7b,d. As seen from the real-time temperature curves of the composites with heating time (Figure 7b), when the amount of head-conducting filler increases, the surface temperature rise rate of the composites increases faster with the heating time. Figure 7d shows the infrared thermal images selected at different heating time, which are consistent with the temperature rise curves. During the same heating time, the surface temperature of the composite AM without head-conducting filler is about 60 °C, while the surface temperature of the composite TC with head-conducting filler can reach to 70 °C, showing a difference of 10 °C. This further illustrates that the composites with higher thermal conductivity, the heat from the heating element is more likely to diffuse uniformly in the plane. In summary, the infrared thermal imaging results are consistent with the thermal conductivity variation, indicating that the addition of head-conducting fillers to the composites with high electromagnetic shielding properties can contribute to the improvement of the thermal conductivity and heat dissipation capacity of the device.



Figure 7. Heat transfer properties of composites: (**a**) thermal conductivity; (**b**) temperature rise curve; (**d**) TGA curve of composites; (**c**) infrared thermal imaging.

The TGA curves of the selected composites are shown in Figure 7c. All the composites and polymer matrix did not react within 200 °C, and there was almost no mass loss. The polymer matrix began to decompose until about 200 °C, and finally at 800 °C, with a weight loss rate of about 74%. The polymer matrix used in this work is a two-component silicone rubber cross-linked by an addition reaction. When the temperature reaches 390–410 °C, the silicon hydrogen bond formed by the addition reaction began to break, and the polymer volatilized rapidly. When the temperature reaches to 800 °C, the downward trend of the DTG curve of the polymer matrix was flattened gradually. However, the composites began to undergo a slight change in quality until about 500 °C, and finally decomposed completely at about 650 °C. The total mass loss was in the range of 5–5.5%, which was equivalent to the mass of the remaining solid residue after heating and volatilization of the polymer matrix in the composites. The composites was excellent, which could be used not only in common electronic equipment, but also in some equipment with high temperature requirements.

3.4. Mechanical Properties

In practical application, the composite material with excellent mechanical properties and stronger resistance to external force can better meet the assembly requirements. For example, when the hardness of the composites is lower, the compression rate is higher, and the internal thermal conduction path is shorter, resulting in shorter heat diffusion time and better thermal conductivity. Figure 8a shows the hardness data of the composites. The increase of the head-conducting powder loading leads to an increase in the hardness of the composites. Figure 8b shows that the tensile strength of the composites increased firstly, and then decreased with the increase of the head-conducting powder loading leads. The tensile strength of the composite TA-2 reached to the maximum value of 200 Pa, which is higher than that of the composite TC with higher proportions of head-conducting fillers. In addition, as shown in Figure 8c, the breaking elongation rate decreases with the increase of the filling amount of head-conducting powder. The variation trend of the breaking elongation rate is consistent with that of the tensile strength of the composites. Since the tensile properties of materials can evaluate the degree of bonding between materials, it indicates that the addition of head-conducting fillers with larger size can reduce the bonding between the fillers and the matrix. However, as shown in Figure 8d, the increase of the head-conducting powder loading promoted an increase in the compressive strength of the composites under 50% compression. This indicates that the filler with large particles is beneficial to the improvement of compressibility, thus showing the advantages of large particle fillers. In short, the results showed that the composite TA-2 expressed good mechanical properties. Combined with the cross-sectional SEM images of the composites in Figure 2, the binding ability of the head-conducting powder to the polymer matrix was not as good as that of the microwave-absorbing powder. The head-conducting powder of large particles with large content would be separated from the polymer matrix, resulting in insufficient internal bonding of the composite materials.



Figure 8. Mechanical properties of different composites: (**a**) hardness; (**b**) tensile strength; (**c**) breaking elongation rate; (**d**) compressive strength.

In particular, the cross-sectional characterization of the composite TA-2 as seen from Figure 3c above showed that spherical particles were uniformly dispersed in the polymer, the particles with large sizes were overlapped, and the small particles were filled in gaps. On the one hand, the composite particles are used as physical cross-linking points to form a "skeleton" structure in the composite, which hinders the molecular chain movement of the polymer and improves its mechanical properties [48]. On the other hand, when the composites are subjected to external load, the particles with small particle size in the filler can appear at the crack position to play a pinning role and change the diffusion direction of the crack. In this situation, many small cracks appear on the path branch of the large crack, which can assist to buffer the energy transferred by external loads. However, when the

loading of fillers with small particle size is low, the pinning effect cannot be achieved [49]. If the fillers with small particle size is not enough, the tensile strength will be reduced, such as composites TA-3 and TC. However, the fillers with large particle size are not enough, the internal skeleton structure is difficult to support the polymer matrix, resulting in a decrease in compressive strength, such as the composite AM. Therefore, the combination of fillers with small particle size and large particle size needs a suitable proportion.

3.5. Mechanism Analysis

The relationship between the structure and properties of the composites can be summarized by characterizing the morphology of the composites and analyzing their thermal conductivity and microwave absorption properties. In this experiment, silicone rubber was used as the matrix, the additional reaction of silicone rubber occurred mainly under the catalysis of platinum catalyst. The Si-CH=CH₂ bond in vinyl silicone oil and the Si-H bond in hydrogen-containing silicone oil were broken, and the hydrosilylation reaction occurred, as shown on the left side of Figure 9. The two-component silicone rubber is mainly used in practical production, which has the advantages of room temperature curing, easy storage, long storage time, good intermolecular bonding after molding, and excellent mechanical properties.



Figure 9. The reaction process of two-component silicone resin and the thermal conduction and microwave-absorbing mechanism of the composites.

The filler-assisted phonon transmission and microwave reflection and absorption mechanism inside the composites can be seen from the right side of Figure 9. The polymer itself relies on phonon to transmit heat, while Al₂O₃ and ZnO among the head-conducting fillers are good phonon transmission media. Therefore, when the spherical particles are uniformly dispersed in the polymer, they can overlap into a coherent grid path, which promotes the composite materials to form a high-density heat transfer channel, and reduces the scattering of phonons during transmission, and increases the internal heat transfer path. Meanwhile, due to the close structure formed by the overlap of the internal filler particles, most of the incident microwaves are difficult to be reflected after entering, resulting in the microwave being effectively attenuated by repeated reflection. In addition, the incidence of high frequency microwaves can excite electrons in the absorbing power, forming electron migration, tunneling, and hopping mechanisms, which can increase the ohmic loss of

microwaves. Then, in the conduction process, electrons will generate current to cause a lot of electrons to transfer, transforming electromagnetic energy into heat energy, and further losing microwaves. In general, the head-conducting microwave-absorbing multifunctional composites are a magnetic microwave absorber with $Fe(CO)_5$ as the main part, which converts electromagnetic waves into thermal energy in the form of insulation loss, magnetic loss and impedance loss to reduce electromagnetic radiation. Therefore, the composites with dual function of thermal conductivity and microwave-absorbing can be achieved through blending two-component silicone rubber with thermal conductivity powders and absorbing powders.

4. Conclusions

In summary, Al2O₃/ZnO/Fe(CO)₅/SR composite materials with excellent microwave absorption, thermal conductivity and mechanical properties can be successfully mass-produced. For the Al₂O₃/ZnO/Fe(CO)₅/SR composite materials, its thermal conductivity can reach to 3.61 W·m⁻¹·K⁻¹, its effective absorption bandwidth is in the high frequency range of 10.86–15.47 GHz, and its effective absorption efficiency can reach to 99% at the frequency range of 12.46–14.27 GHz, indicating good heat dissipation and electromagnetic broadband loss properties. This is due to the compact microstructure formed by the overlapping of large particle size fillers and the filling of their gaps by small particle size fillers, which can contribute to enhance the heat conduction path and weaken the microwave reflection. Furthermore, the addition of composite fillers with different particle sizes can improve the thermal stability and mechanical properties of composite materials filled with single fillers. It provides a theoretical and experimental basis for the research of head-conducting microwave-absorbing multifunctional composites.

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