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Reduction of thermal resistance of Ag-coated GFs/copper structure using nano-Ag paste as interconnection

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Abstract. Reduction of the thermal resistance between graphene films (GFs) and substrate is crucial to the application of GFs in thermal management. GFs/copper structures were prepared using nano-Ag paste as interconnection material. The effect of the thickness of nano-Ag paste on thermal resistance of GFs/copper structure was investigated. A thin layer of Ag was coated on GFs by physical vapor deposition (PVD) to further reduce thermal resistance. The thermal resistance of GFs/copper structure using Ag-coated GFs is 5.84% lower than that using raw GFs. The thermal resistance of GFs/copper structure decreases first and then increases with the increase of coating temperature and thickness of Ag layer. The minimum thermal resistance of 1.64 $mm^2 \cdot K \cdot W^{-1}$ was gained for GFs/copper structure using GFs coated Ag at 300 ℃ for 60 min.

1. Introduction

The power density is increasing with the development of packaging integration of electronic devices, which will lead to more heat accumulation in electronic products[1]. Traditional interconnection materials cannot meet the requirements of high operating temperature for high power density devices[2]. Due to its high thermal conductivity, electrical conductivity and melting point, nano-Ag paste has been considered as a promising thermal interface material to replace the traditional thermal interface materials, such as lead-free solder, conductive adhesive and so on[3]. The lower glass transition temperature of conductive adhesive and the lower melting point of solder limit their application at high temperature. Cao et al. evaluated the thermal performance of IGBT modules connected with sintered nano-Ag, SAC305 and SN100C, and concluded that the thermal resistance of IGBT modules connected with sintered nano-Ag is 12.1% lower than that of SAC305 and SN100C[4].

The interface between metal and carbon material plays crucial role in determining the thermal properties of composites[5-6]. The poor wettability between sintered nano-Ag and GFs is the key problem to obtain composites with high heat dissipation performance. The thermal resistance of the composite is increased due to voids formed at the interface between sintered nano-Ag and GFs. An effective way to solve this problem is to coat metal on the surface of GFs[7-9]. Yu Huang et al. prepared graphite film/Cu/Al composites by coating copper on the surface of graphite film. The out-of-plane thermal conductivity of graphite film reinforced aluminum matrix composites is increased by more than 20 times[10].

In this study, the surface of GFs was modified by coating a layer of Ag using PVD technology. GFs/copper structure was prepared by screen printing process with nano-Ag paste as interconnection material. The interfacial microstructures and thermal resistance of GFs/copper structure using Ag-coated GFs and raw GFs were studied. In order to optimize the coating process, the effects of coating temperature and coating thickness on the thermal resistance of the structure were also investigated.

2. Experimental

Nano-Ag paste was prepared firstly. Nano-Ag particles, triethylene glycol, terpineol and Polyvinyl butyral (PVB) were used as fillers, dispersant, thinner and organic vehicle. The proportions of them were 82 wt.%, 1 wt.%, 7 wt.% and 10 wt.%, respectively. The diameter of the nano-Ag particles is about 20 nm. Firstly, PVB was mixed with ethanol slowly by stirring. After cooling to room temperature, PVB and terpineol were added to the solution. Then, nano-Ag particles were added with stirring slowly for 15 min. Finally, the mixture was subject to ultrasonic treatment at 20 °C for 30 min to make the particles distributed evenly.

A sandwich structure of GFs, sintered nano-Ag and copper substrate was prepared for thermal resistance testing. The specific processes are listed as follows: (1) A ultra-thin layer of Ti (20-30nm) was coated on the surface of GFs (40 μm) by PVD technology to enhance the adhesion between nano-Ag paste and GFs. (2) Ag layer was coated on the surface of Ti layer at 25 ℃, 100 ℃, 200 ℃, 300 ℃ and 400 ℃, respectively. The coating time is set to 5, 15, 30, 60 and 90 min at 25 ℃, respectively. The contact between GFs and sintered nano-Ag was transferred to the contact between Ag coating and sintered nano-Ag. (3) The nano-Ag paste was printed on the Ag-coated GFs and raw GFs by screen-printing process. The coating thickness of nano-Ag paste were 15 μm, 30 μm, 50 μm, 80 μm and 100 μm, respectively. Then, a copper substrate ($10mm \times 10mm \times 0.5mm$) is gently placed on the nano-Ag paste layer. (4) The structure was dried at 120 °C for 15 minutes to volatilize the alcohol in the nano-Ag paste. Then, the samples were sintered at 280 for 40 min with a pressure of 0.625 MPa. A pretreatment at 200 °C for 15min was introduced to volatilize triethylene glycol completely in nano-Ag paste before sintering.

3. Results and discussion

3.1. Effect of thickness of nano-Ag paste on thermal resistance

The cross-sectional of GFs/copper structure using raw GFs and Ag-coated GFs were observed by scanning electron microscopy (SEM). Fig. 1(a) and (b) show that a layer of organic residues is agglomerated in the interface between GFs and the sintered nano-Ag. The organic residues with low thermal conductivity will hinder the heat transfer from the sintered nano-Ag to GFs, resulting in the increase of the thermal resistance of GFs/copper structure. The SEM images of GFs/copper structure samples with Ag-coated is shown in Fig. 1 (c) and (d). The good wettability between the Ag coating and the sintered nano Ag makes the organic residues unable to agglomerate, so that the heat could transfer via the interface smoothly, and therefore the thermal resistance is reduced.

The thermal resistance test results of GFs/copper structure using coating nano-Ag paste with different thickness is shown in Fig. 2. It can be found that the thermal resistance of GFs/copper structure increases with the increase of nano-Ag paste thickness, whether coated or not. The increase of thermal resistance of GFs/copper structure with Ag coating is much lower than that of uncoated samples. The thermal resistance of GFs/copper structure with Ag-coated increased by 5.36% with the thickness of nano-Ag paste increased from 15 μm to 80 μm, while that of GFs/copper structure using raw GFs increased by 12.03%. The thermal resistance difference of samples using Ag-coated GFs and raw GFs further increases as the thickness of nano-Ag paste increases to 100 μm. It indicates that the thickness of nano-Ag paste has a greater effect on the thermal resistance of GFs/copper structure using raw GFs.

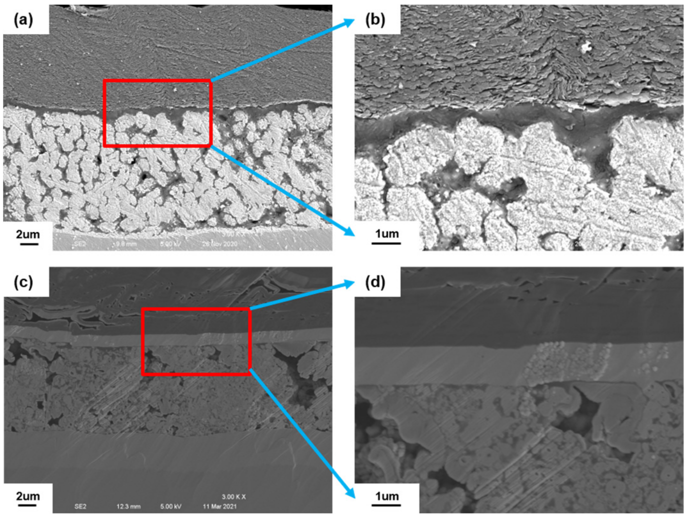

Figure 1. SEM images of cross-section of GFs/copper structure using raw GFs and Ag-coated GFs. (a)& (b) raw; (c) & (d) Ag-coated.

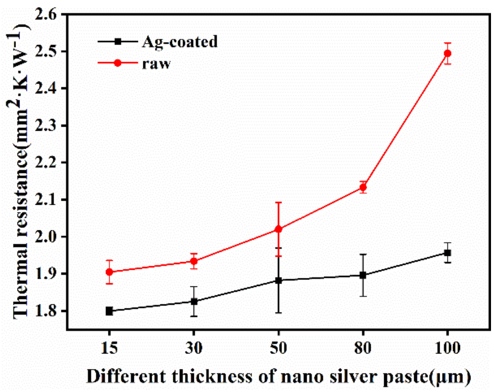

Figure 2. Thermal resistance of GFs/copper structure using Ag-coated GFs and raw GFs.

3.2. Effect of coating temperature on thermal resistance

Fig. 3 shows that the SEM images of the surface of GFs coated Ag at 25 ℃, 100 ℃, 200 ℃, 300 ℃ and 400 ℃ for 30 minutes. It can be seen that the size of nano-Ag particles on GFs increases significantly with the increase of coating temperature in Fig. 3.

Fig. 4 shows that the thermal resistance of GFs/copper structure decreases first and then increases with the increase of coating temperature. The minimum thermal resistance of GFs/copper structure using GFs coated with Ag at 300 ℃ is 1.67 $mm^2·K·W^{-1}$. It can be seen from Fig. 3 (A-D) that Ag particles on the coating surface begin to coarsen with the increase of coating temperature. The heat transfer efficiency of Ag coating is improved because the coarsened Ag particles can reduce the amount of interface between particles, so as to reduce the thermal resistance of GFs/Copper structure. Some cracks appear on the surface of the Ag-coated GFs at 400 ℃ as shown in Fig. 3 (E). These cracks will hinder the heat transfer in the Ag coating, resulting in the increase of the thermal resistance of GFs/copper structure.

Fig. 5 shows that cross-sectional SEM images of GFs/copper structures using GFs coated with Ag at 25 ℃ and 300 ℃. It can be seen that a tight connection is formed between the coating and sintered nano-

Ag. Fig. 5 (c) and (d) show that the coating thickness at 25 ℃ is thicker than the sample with the coating temperature of 300 ℃.

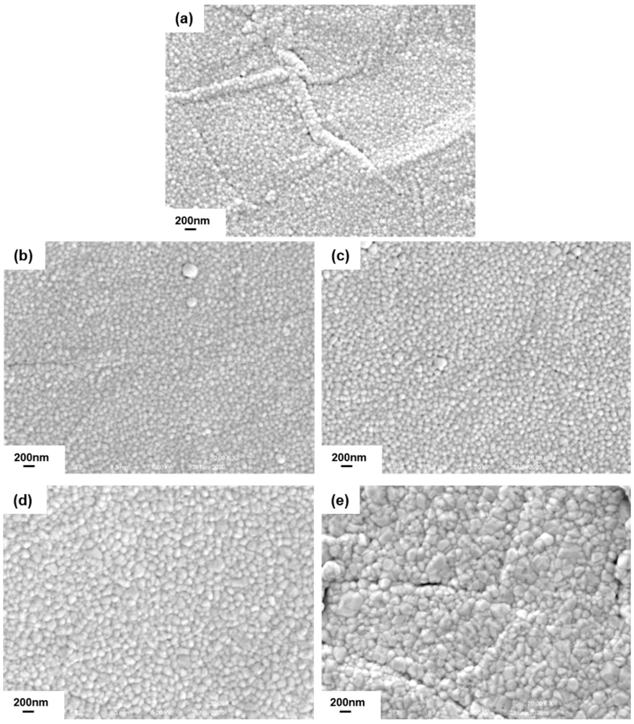

Figure 3. SEM images of GFs coated with Ag at different temperatures. 25℃; (a) 100℃; (b) 200℃; (c) 300℃; (d) 400℃.

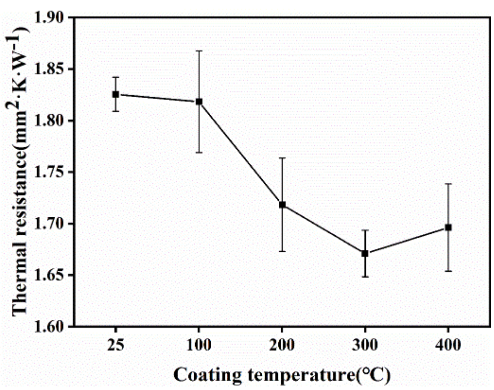

Figure 4. Thermal resistance of GFs/copper structure using GFs coated with Ag at 25 ℃, 100 ℃, 200 ℃, 300 ℃ and 400 ℃.

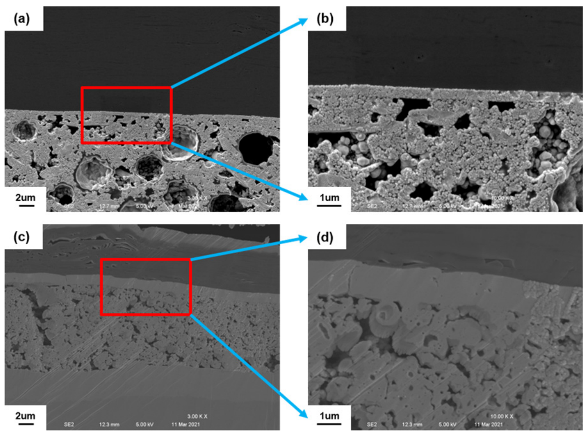

Figure 5. SEM images of cross-section of GFs/copper structure using GFs coated with Ag at 25 ℃ and 300 ℃. (a) & (b) 300 ℃; (c) & (d) 25 ℃.

3.3. Effect of coating thickness on thermal resistance

Fig. 6 shows that the thermal resistance of GFs/copper structures using GFs coated with Ag at 25 ℃ for 5, 15, 30, 60 and 90 min, respectively. The thermal resistance of them decreases first and then increases with the increase of Ag coating thickness. The minimum thermal resistance of samples using GFs coated with Ag for 60 min is 1.66 $mm^2{\cdot}K{\cdot}W^{-1}$ The thermal resistances of GFs/copper structure using GFs coated with Ag for 5 min and 15 min were 1.73 $mm^2{\cdot}K{\cdot}W^{-1}$ and 1.72 $mm^2{\cdot}K{\cdot}W^{-1}$, respectively. Compared with

GFs/copper structure using GFs coated with Ag for 60 min, it increased by 0.7 $mm^2 \cdot K \cdot W^{-1}$ and 0.6 $mm^2 \cdot K \cdot W^{-1}$, respectively. The Ag coating is thin and cannot form an effective connection with sintered nano-Ag for the samples with Ag coating time of 5 and 15min. The decrease of thermal resistance for the samples using GFs coated with Ag for 30 min and 60 min is mainly due to the increase of Ag coating thickness and the corresponding improvement of coating quality, which can form an effective connection with sintered nano-Ag. However, the thermal resistance of GFs/copper structure increases to 1.72 $mm^2 \cdot K \cdot W^{-1}$ for the sample using GFs coated Ag for 90 min.

Fig. 7 shows that the SEM of the interface of GFs/copper structure using GFs coated with Ag at 25 ℃ for 30, 60 and 90 min, respectively. As shown in Fig. 7 (a), there are voids at the interface between the Ag layer and sintered nano-Ag. The heat transfer efficiency is reduced due to the existence of these holes, which leads to the increase of the thermal resistance. The heat transfer efficiency is improved because the Ag layer is combined closely with the sintered nano-Ag, as shown in Fig. 7 (b). Therefore, the thermal resistance of samples using GFs coated with Ag for 60 min is lower than that of samples using GFs coated with Ag for 30 min.

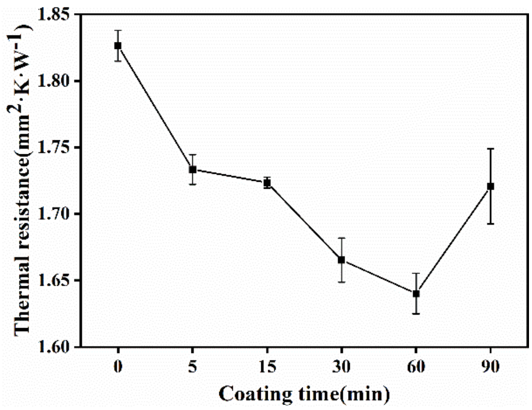

Figure 6. Thermal resistance of GFs/copper structure coated with Ag layer for different times.

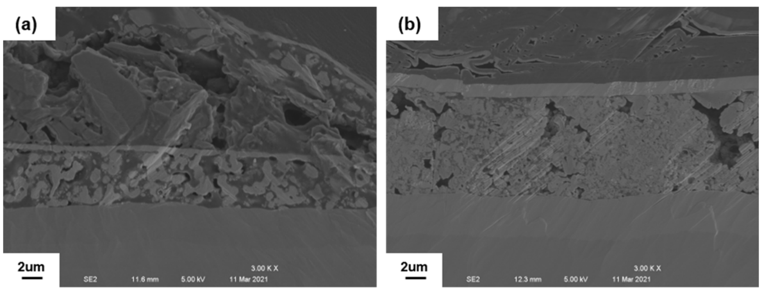

Figure 7. Cross sectional SEM of GFs/copper structure using GFs coated with Ag for 30min and 60min. (a) 30 min; (b) 60 min.

4. Conclusions

Nano-Ag paste was used to connect GFs and copper substrate to reduce thermal resistance effectively in this paper. A thin Ag layer was coated on GFs by physical vapor deposition (PVD) to improve the adhesion between GFs and copper substrate. The effect of the thickness of sintered nano-Ag on the thermal resistance of GFs/copper structure was investigated. Nano-Ag paste with a thickness of 15 μm is used as interconnection, and the thermal resistance of GFs/copper structure using GFs coated with Ag is 5.84% lower than that using raw GFs. The minimum thermal resistance of GFs/copper structure using GFs coated with Ag at 300 ℃ for 60 min is 1.64 $mm^2 \cdot K \cdot W^{-1}$. It can be concluded that nano-Ag paste as interconnection and GFs coated with Ag is an effective way to reduce the thermal resistance between GFs and copper substrate.

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