Available online at www.sciencedirect.com
SciVerse ScienceDirect
Scripta Materialia 66 (2012) 931–934
Scripta MATERIALIA
www.elsevier.com/locate/scriptamat

Silver nanoparticle-based thermal interface materials with ultra-low thermal resistance for power electronics applications

Hui Yu, Liangliang Li* and Yujun Zhang

Department of Materials Science and Engineering, Key Laboratory of Advanced Materials, Tsinghua University, Beijing 100084, People's Republic of China

Received 15 November 2011; revised 2 February 2012; accepted 20 February 2012
Available online 26 February 2012

We studied the thermal conduction of thermal interface materials (TIM) using silver nanoparticles (AgNP) and achieved ultra-low thermal resistance. The experimental data show that silver nanoparticles are very good candidates for TIM in power electronics applications in terms of the reduction in thermal resistance. The ultra-low thermal resistance of the AgNP-based TIM originates from the thinness, high thermal conductivity of silver and low temperature sintering properties of AgNP.

Keywords: Thermal interface material; Thermal conductivity; Bonding; Sintering

Heat dissipation in power electronics devices such as ultra-fast computer chips, high power light emitting diodes, high power lasers and insulated gate bipolar transistors has become an urgent issue due to the increase in their power density, because the heat generated by these power electronics devices increases the device working temperature and dramatically degrades device reliability and lifespan [1,2]. In high power devices thermal interface materials (TIM), which are used at interfaces such as those between computer chips and heat sinks, are attracting a lot of interest due to the benefits of fast heat dissipation [3–5]. A low thermal resistance is required for TIM and many kinds of materials have been used in this role, including thermal pastes [6], phase change materials [7], polymer-based composite materials [8–12], carbon-based materials [13–21] and solders [22]. However, these materials have their own drawbacks, such as low thermal conductivity or poor thermal contact [3–5].

Metallic nanoparticles could be potential candidates for TIM. Firstly, metals generally have a high thermal conductivity that ensures a low thermal resistance. Secondly, the melting points of metallic nanoparticles are much lower than those of bulk materials [23,24], which makes their use for many applications requiring a low process temperature feasible. Thirdly, strong metal–metal bonding can be achieved with metallic nanoparticles [25,26]. Among metallic nanoparticles silver nanoparticles (AgNP) are the most interesting. They can be used in metal–metal bonding [25,26], printing electronics [27–32], die-attached interconnection [33] and solar cells [34] due to their low sintering temperature, good electrical properties and good mechanical strength. Since the thermal conductivity of silver at room temperature is as high as 429 $W\,m^{-1}\,K^{-1}$ and AgNP can be sintered at a temperature below 300 °C [25–34] AgNP could be a good candidate for TIM. In this study we first report a TIM based on AgNP and demonstrate its better performance in terms of thermal conduction.

We synthesized uniform AgNP according to a reported procedure [28]. The particle size was controlled by tuning the ratio of the silver ions (silver acetate) to the surfactant (sodium polyacrylate) in the reaction vessel. Silver acetate (>99% pure, Sinopharm Chemical Reagent Co. Ltd), ascorbic acid (>99.7% pure, Sinopharm Chemical Reagent Co. Ltd), sodium polyacrylate (mol. wt. 8000, 45 wt.% solution in water, Sigma–Aldrich Chemical Co.) and triple distilled water (TDW) were used as the silver precursor, reducing agent, surfactant, and solvent, respectively. In a typical experiment 2.31 g sodium polyacrylate, 1.60 g silver acetate and 1.23 g ascorbic acid were added to 100 g TDW sequentially and the mixture was stirred using a magnetic stirrer at 95 °C. The as-synthesized nanoparticles were centrifuged at 15,000 r.p.m. for 10 min and then washed

* Corresponding author. Tel.: +86 10 62797162; fax: +86 10 62771160; e-mail: liliangliang@mail.tsinghua.edu.cn

1359-6462/$ - see front matter
doi:10.1016/j.scriptamat.2012.02.037

six times with TDW before use. The scanning electron microscopy (SEM) (Zeiss LEO 1530) images and transmission electron microscopy (TEM) (FEI TECNAIG^{2}20) image in Figure 1 show the morphology of AgNP of different sizes. Figure 1a–c shows the AgNP with sizes of 20–30, 30–40, and 100–120 nm, respectively; they will be referred to as AgNP-20, AgNP-30, and AgNP-100, respectively. The lattice fringe in the inset in Figure 1d was 0.235 nm, corresponding to the silver {1 1 1} interplanar spacing (JCPDS file No. 02-1098). In addition, the X-ray diffraction (Rigaku D/max 2500 diffractometer with monochromatic Cu K_α radiation) patterns of the AgNP are shown in Supplementary Figure S1 in Supplementary material, demonstrating a pure silver phase. Weight loss of the AgNP was monitored using a thermogravimetric analyzer (TGA) (TA Instruments Q5000, under N_2) and is shown in Figure 2a. Some organic material adsorbed to the surface of the AgNP was debonded at ~200 °C. The percentage by weight of organic material was 1.5–2.5%. Figure 2b shows differential scanning calorimetry (DSC) (TA Instruments Q2000) profiles of the AgNP under N_2. AgNP-20 and AgNP-30 show some exothermic peaks in the temperature range 200–280 °C. These peaks are similar to the data reported in Moon et al. [27] and are due to the surface sintering reaction of the AgNP (to reduce the surface energy) or recrystallization of strained nanoparticles by heating. More analysis on these exothermic heat peaks can be found in Moon et al. [27]. For AgNP-100 there is no exothermic peak in the DSC curve, and it can be concluded that surface sintering is more difficult for larger AgNPs. Smaller AgNP can readily coalesce at low temperatures, suggesting the possible formation of a good thermal conduction path for their use as a TIM. For example, Supplementary Figure S2 shows the morphology of AgNP-30 baked in Ar at different temperatures, with obvious coalescence at 200 °C.

In order to evaluate the thermal conduction of AgNP-based TIM we fabricated copper plate/AgNP/copper plate sandwiched samples and measured their thermal resistance. The sandwiched samples were prepared as follows. First, copper plates with a diameter of 10.0 mm and a thickness of 1.60 mm were sequentially polished using 200, 400, 800 and 1500 grade sandpapers (Supplementary Fig. S3a). Second, a suspension of AgNP (10–15 wt.%) was added dropwise to one side

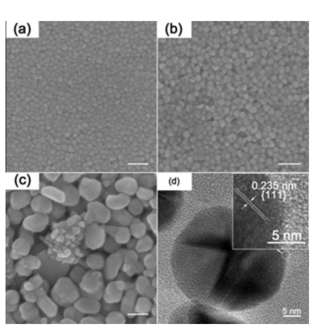

Figure 1. SEM images of AgNP with sizes of (a) 20–30, (b) 30–40, and (c) 100–120 nm. (d) TEM image of a typical AgNP. (Inset) The silver {1 1 1} interplanar spacing. Scale bars in (a–c) are 100 nm.

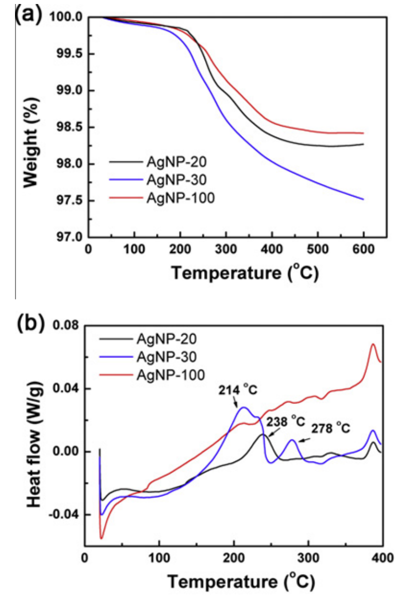

Figure 2. (a) TGA results and (b) DSC profiles of the AgNP of various sizes.

of the copper plates and dried in air. Third, two copper plates coated with AgNP were stacked to form a sandwiched sample with the AgNP in the middle. Finally, the sandwiched samples were hot pressed at various temperatures by spark plasma sintering at a pressure of 30 MPa for 30 min. Supplementary Figure S3c shows a schematic of a sandwiched sample. The total thickness of the sandwiched samples was ~3.20 mm, with each copper plate being ~1.60 mm thick. The typical thickness of the AgNP layer was 8–10 μm. Without the AgNP as a TIM an air gap forms between copper plates in direct contact, giving poor thermal conduction, due to the rough surface of the copper plates, as shown in Supplementary Figure S3b [20]. The thermal resistance R was obtained from $R = d/k$, where d is the measured thickness and k is the thermal conductivity. k was obtained using the equation $k = \rho \times \alpha \times C_p$, where α is the thermal diffusivity at room temperature determined with a Netzsch LFA427 Laser Flash apparatus (calibrated using Fe, Ta, and Fe–Ni standards), ρ is the density measured with a Mettler Toledo XS105DU analytical laboratory balance and C_p is the calculated heat capacity. In our calculations the ideal heat capacities of the copper plate and AgNP, $C_{p,Cu}$ and $C_{p,Ag}$, were 385 and 233 J kg$^-$ K^{-1}, respectively. The heat capacity of a sandwiched sample is calculated using the equation $C_p = C_{p,Ag} + (C_{p,Cu} - C_{p,Ag}) \times d_{Cu}/d_{sample}$, where d_{Cu} and d_{sample} are the measured thicknesses of the copper plates and the sample, respectively. Because the percentage by volume of the copper plates in the sandwiched samples is more than 99.0% we believe that the calculation of the heat capacity is fairly reasonable. The thermal resistance of the TIM, R_{TIM}, is equal to the thermal resistance of the sandwiched sample minus the thermal resistance of the two copper plates in that sample.

In our study AgNP-20, AgNP-30 and AgNP-100 were used as the TIM. The AgNP-20 samples were successfully hot pressed at 100, 150, 200 and 250 °C,