



Advanced Thermal Interface Materials for Thermal Management

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Suitable temperature is a necessary condition for the normal operation of many devices, especially microelectronic devices. Therefore, heat dissipation has become a bottleneck in many fields. Furthermore, the largest thermal resistance in the process of heat transfer occurs between two solid surfaces due to the poor thermal conductivity of air that exists in the gaps. Replacing air with thermal interface materials (TIMs) is the fundamental way to solve the problem of heat dissipation. Consequently, TIMs are widely used in LED lighting,^{1,2} solar energy,³ microelectronics,⁴⁻⁶ electrical and electrical engineering,⁷ aerospace,⁸ defense and other fields (Figure 1a.).

In the case of using TIMs, the interfacial thermal resistance consists of three parts (Figure 1b): two boundary thermal resistances (R_{c1} , R_{c2}) associated with the TIM contacting either side of the surface and a thermal resistance relative to the inherent properties of the TIMs (R_{BLT}).⁹ An excellent TIM should possess high intrinsic thermal conductivity, small effective bond line thickness (BLT) and low boundary thermal resistance. The effective thermal performance of a TIM is generally expressed in terms of total thermal contact resistance (TCR). The most straightforward method for determining the heat dissipate at the interface is to characterize experimentally. There have been many methods to characterize the TCR such as the steady-state heat flow method, the T-type method, micro-thermometry methods, Raman-based techniques, infrared thermography measurements, laser-flash measurements, photo-acoustic techniques, the 3ω method and transient thermoreflectance techniques.¹⁰⁻¹² A proper implementation of characterization need to consider the material characteristics and material level of the testing sample.

Carbon nanomaterials occupy a unique position in TIMs due to their excellent thermal transport properties (Figure 2). The implementation of vertically aligned CNT arrays and CNT fibers as TIMs requires the optimization of two distinct thermal properties: the intrinsic thermal conductivity of the CNT materials and the contact resistance (R_c), which locates at intertube for CNT fibers and between the array and substrate for CNT array. Much research has focused on enhancing the intrinsic thermal conductivity of the CNT materials, while significantly less for contact resistance. The thermal conductivity is primarily limited by crystalline quality of the CNTs, while the contact resistance will be limited by non-uniformity in the array height and undesired amorphous carbon situated at the top of the canopy for CNT array, and limited by CNTs' interfacial resistance for CNT fibers. To boost the thermal transport performance of CNT assembled materials, the main-stream studies center around the following approaches, i.e., increase the height uniformity of CNT array¹³ (Figure 2b), control the tube diameter, array length and reduce the amount of amorphous carbon¹⁴ (Figure 2c) and increase the contact area based on flexible horizontally-aligned CNT array¹⁵ (Figure 2d) for reducing the R_c between CNT array-heat sink interface. In addition, the inter-tube thermal transport of CNT fibers can be greatly improved with the functionalization and densification of inter-bundle interface¹⁶ (Figure 2e,f).

Phase change materials (PCMs) used in TIMs mainly includes liquid metal (LM) and paraffin based composites. LM is an amorphous metal and can be regarded as a mixture of positive ion and free electron gas. The interest in the research of LM is rapidly warming up because of its superior thermo-physical properties, like high thermal conductivity, low interface thermal resistance, excellent fluidity, low toxic and melting temperatures. However, LM with large surface tension may leak out and easy to oxidize. The oxidation/corrosion can be mitigated by providing a hermetic seal,¹⁷ and the formation of intermetallic compounds can be prevented by using a diffusion barrier coating.¹⁸ Lithium ion (Li-ion) batteries suffer from a high temperature rise during operation due to high energy and power density, thus affecting their life span and efficiency.¹⁹ Paraffin based composites can prevent Li-ion batteries from rising temperature, because it stores heat in the form of sensible and latent heat, mainly in the latent heat form owing to the large latent storage capacity. It is also worth mentioning that the thermal conductivity of paraffin based composites is very important, which effected the heat storage rate. Furthermore, many techniques about paraffin filled

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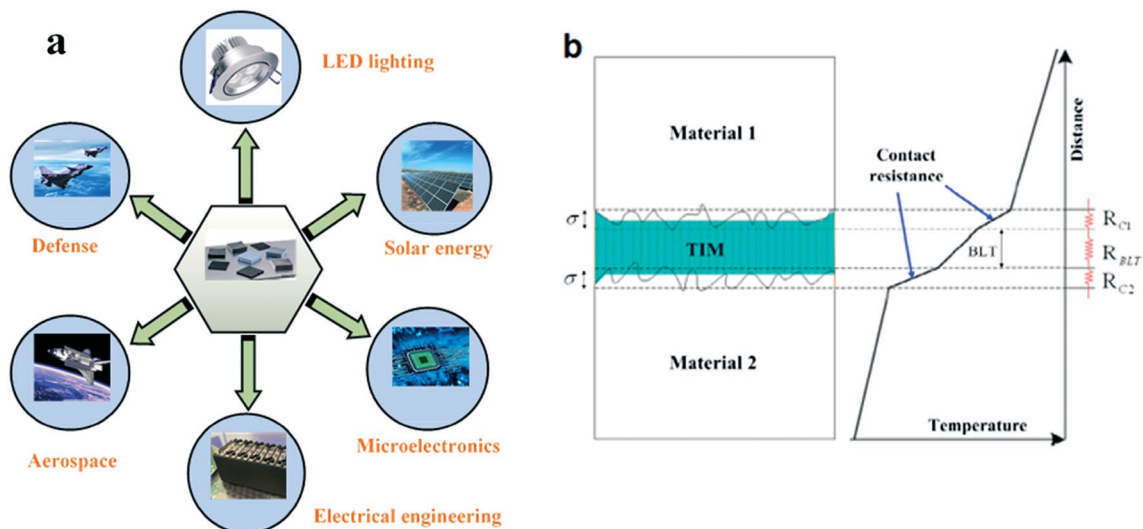


Fig. 1 a. Application fields of thermal interface materials; b. Various components of thermal contact resistances after the insertion of a TIM (reprinted with permission from⁹ Copyright Elsevier Composites Part A: Applied Science and Manufacturing).

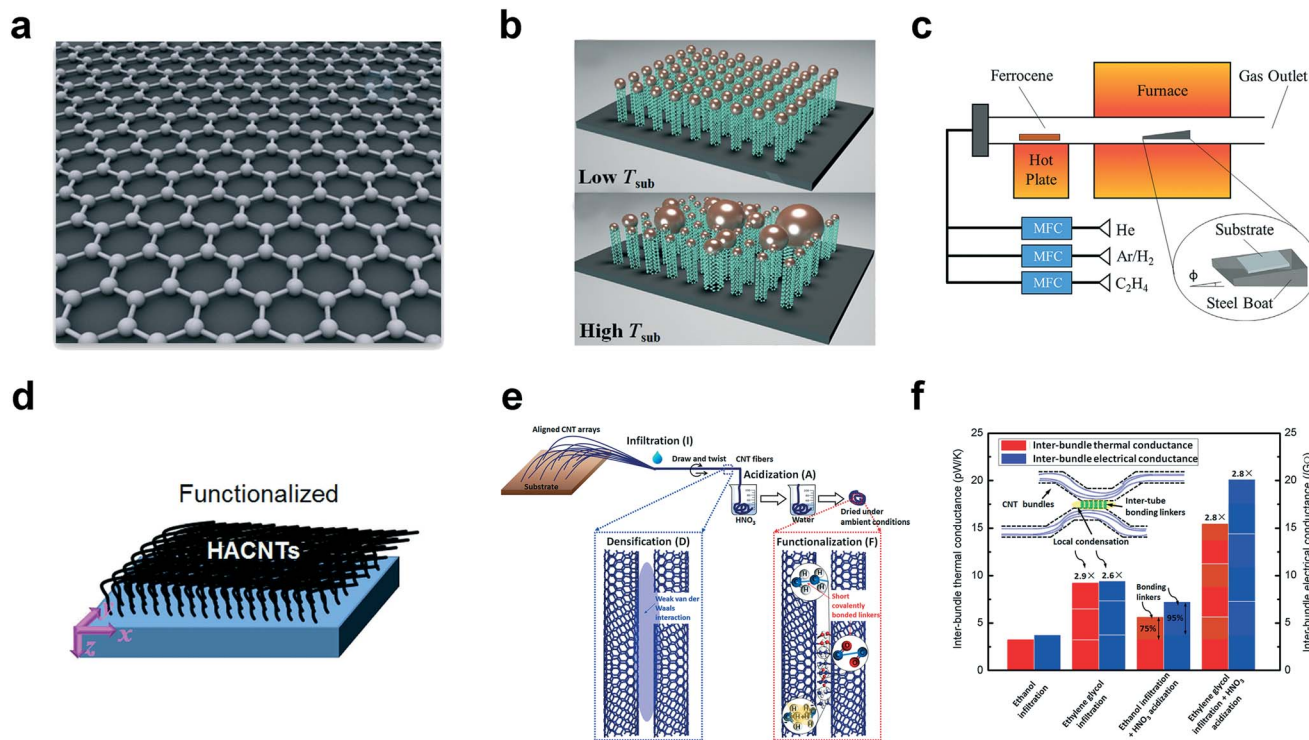


Fig. 2 The thermal properties enhancement of carbon nanomaterials-based TIMs. Scheme of **a.** graphene, **b.** producing uniform height for CNT arrays (reprinted with permission from¹³ Copyright Elsevier Carbon), **c.** controlling the tube diameter, array length and reduce the amount of amorphous carbon for CNT array synthesis during chemical vapor deposition process, **d.** horizontally aligned CNT arrays (reprinted with permission from¹⁴ Copyright John Wiley and Sons Physics Status Solidi A-Applications and Materials Science), **e.** functionalization and densification of inter-bundle interface (reprinted with permission from¹⁶ Copyright Elsevier Carbon). **f.** boosted inter-bundle interfacial thermal and electrical conductance for functionalized and condensed CNT fibers (reprinted with permission from¹⁶ Copyright Elsevier Carbon).

with high thermal conductivity and superstructure materials have been suggested to improve thermal conductivity, like carbon material, metal foam.^{20–22}

Silver micro/nanoparticle-based TIMs are also good candidates for electronic power devices because of three important advantages including high intrinsic conductivity, lower melting points of metallic

micro/nanoparticles, ability of strong metal-metal bonding.²³ For high power devices ($\geq 100 \text{ W/cm}^2$), soldering is still a most effective method to reduce the TCR. Typically, Ag micro/nanopaste has become the most potential choice as the new type of lead-free interconnection materials since Ag micro/nanopaste can be sintered at a relative low temperature without the need of pressure loading²⁴ and can work at

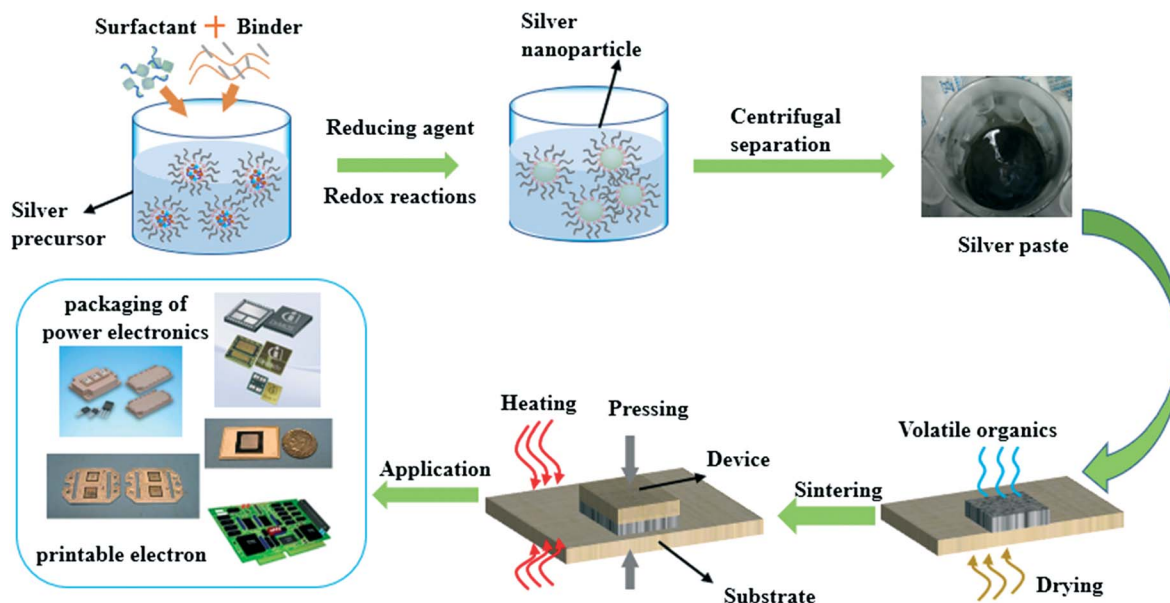


Fig. 3 Preparation and application of Ag micro/nanopaste.

a higher temperature than the sintering temperature with enhanced bond strength. The preparing methods and application of Ag micro/nanopaste is shown in Figure 3. It confirms the synthetic formula and key sintering process affecting the quality and sintering properties of Ag micro/nanopaste.

Speeding up heat transport is the fundamental goal of TIMs. Therefore, some types of TIMs with low TCR occupy the development direction. Many methods like functionalization, orientated arrangement, synergistic enhancement of heat transfer and three-dimensional material can reduce the interface thermal resistance between the fillers and form an efficient heat conduction network chain. Moreover, PCMs reduce the contact thermal resistance between heat source and TIMs. In addition, special heat dissipation field require high thermal conductivity and electrical insulation for TIMs. Finally, the TCR plays an importance role for devices in extreme engineering environments (e.g., in cryogenic engineering and aerospace engineering), while the research on these aspects as well as the reports about the TIMs for medium and high temperature applications are scarce. Thus, it still mainly adopts high-performance soldering as TIM to overcome the heat dissipation problem in these special conditions.

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