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导热复合材料降低填料之间界面热阻研究进展

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摘要：复合材料热导率增强的低效率源于其内部存在界面热阻——填料与树脂基体之间的界面热阻及填料之间的界面热阻。目前大多数研究都集中于降低填料与树脂基体之间的界面热阻，而高填充量下填料之间的界面热阻才是影响复合材料热导率的关键因素。文中从增加填料之间的接触面积和提高填料之间的键接强度两方面综述了近年来降低填料之间界面热阻的研究进展，为高导热复合材料的设计和制备提供参考。

关键词：复合材料；热导率；填料；界面热阻
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随着电子元器件的微型化和越来越高的集成度，其发热量和热流密度越来越高，有研究表明，约50%的电子元器件的失效与工作温度过高有关。由聚合物树脂基体和导热填料组成的热界面材料应用在电子元器件与散热器之间，能起到热连接和降低二者之间接触热阻的作用。然而填料对复合材料的热导率增强效率通常较低，因此为了实现高热导率，往往需要在树脂基体中填充大量的导热填料(质量分数大于80%甚至90%)。过多导热填料的使用不仅会提高成本，还会使得膏体状的热界面材料黏度过高，缺乏流动性；凝胶状的热界面材料缺乏柔韧性和适形性，导致电子元器件的散热效率降低[1]。

复合材料热导率增强效率低主要源于其内部的界面热阻，复合材料内部的界面热阻一般由两部分组成：低填充量下，填料无规分布在聚合物基体中，形成海-岛结构，彼此之间的接触很少(Fig.1(a))，此时填料和聚合物基体之间的界面热阻是影响复合材料热导率的关键因素；高填充量下，填料开始相互接触而形成导热通路(Fig.1(b))，此时填料之间的界面热阻才是影响复合材料热导率的关键因素。

降低填料与聚合物基体之间的界面热阻一直是制备导热复合材料的研究热点，相关研究主要集中

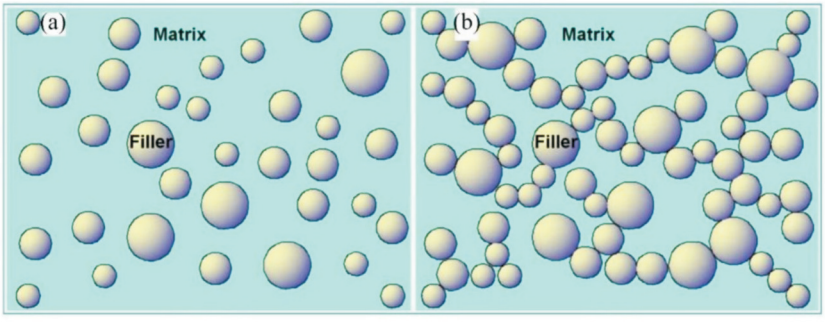


Fig. 1 Dispersion of fillers among resin matrix at different filler loadings: (a) low loading; (b) high loading[2]

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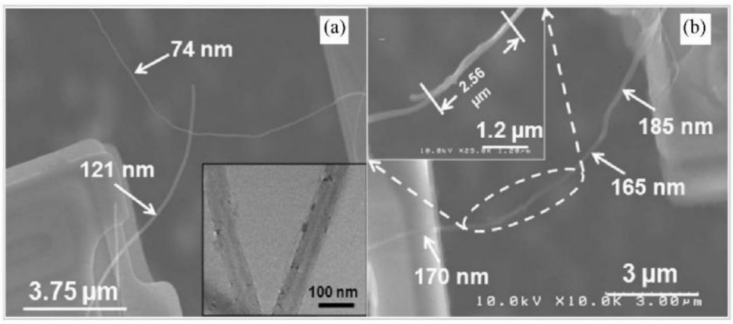


Fig. 2 SEM micrographs of two individual CNTs forming a contact between two suspended membranes: (a) a cross contact; (b) an aligned contact with an overlap of 2.56 μm [9]

在填料的表面改性上,如接枝偶联剂,能降低无机填料的表面能,改善填料与聚合物基体之间的界面相容性,降低二者之间的接触热阻[3,4];或者包覆中间层,缓和填料与树脂基体之间模量的偏差,降低二者之间的边界热阻[5,6]。相比之下,通过降低填料之间界面热阻来提高复合材料热导率增强效率的报道则较少,迄今为止也未见相关的综述。基于此,本文首先分析了影响填料之间界面热阻的因素,然后综述了近年来文献中报道的降低填料之间界面热阻的方法及机理,并对未来需要攻克的重点难点进行了展望。

1　填料之间界面热阻的影响因素

1.1　填料之间的接触面积

填料之间的接触热阻与二者之间的实际接触面积紧密相关[7,8]。Yang等[9]测试了不同接触形式的碳纳米管之间的热阻。结果表明,当2个碳纳米管之间由交叉接触变为排列接触时(如Fig.2(a,b)所示),由于接触面积的增大,界面热传导增大了2个数量级(10^{-8} W/K增长到10^{-6} W/K)。此外,Liu等[10]的研究也表明,当填料的接触区域从几个原子增加到几平方纳米时,每单位接触面积的界面热传导会随着接触面积的增大而提高。原因在于随着接触面积的增加,每个原子在另一界面处有更多的邻近原子,这增加了声子的传输通道。

1.2　填料之间的键接力

填料之间的键接力也是影响界面热阻的重要因素[11,12]。键接力一般分为4种,由弱到强分别为分子间作用力(范德华力)、氢键、共价键及金属键。键接强度的提高可以减少界面处声子或电子的散射,从而增强界面热传导。如Fig.3所示,共价键和金属键键接的界面热传导明显要高于分子间作用力,尤其是金属键键接比分子间作用力要高1个数量级左右[13]。

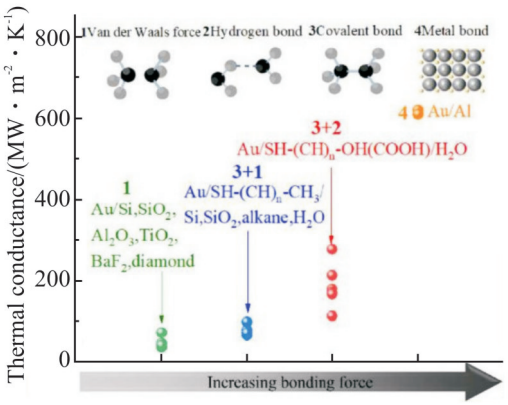


Fig. 3 Interfacial thermal conductance between Au and multiple materials[13]

2　降低填料之间界面热阻的方法及机理

2.1　增加填料之间的接触面积

Choi等[14]使用了不同金属颗粒与碳纳米管协同填充来 改善聚苯乙烯的热导率。结果表明,与使用铝或铜相比,使用银时复合材料的热导率要高得多(Fig.4(c))。更高的热导率源于银颗粒本身的低模量和优异的延展性,当银颗粒与高黏度热塑性树脂混合成型时会发生变形,邻近的银颗粒之间可以形成面接触(Fig.4(b)),热量可以通过接触面快速传输。而铜和铝的模量较高,混合成型过程中变形很小,热传输只能发生在一些金属-金属接触点上,而没有直接接触的区域就会扰乱热传输,使得热阻增大(Fig.4(a)),热传输效率降低。

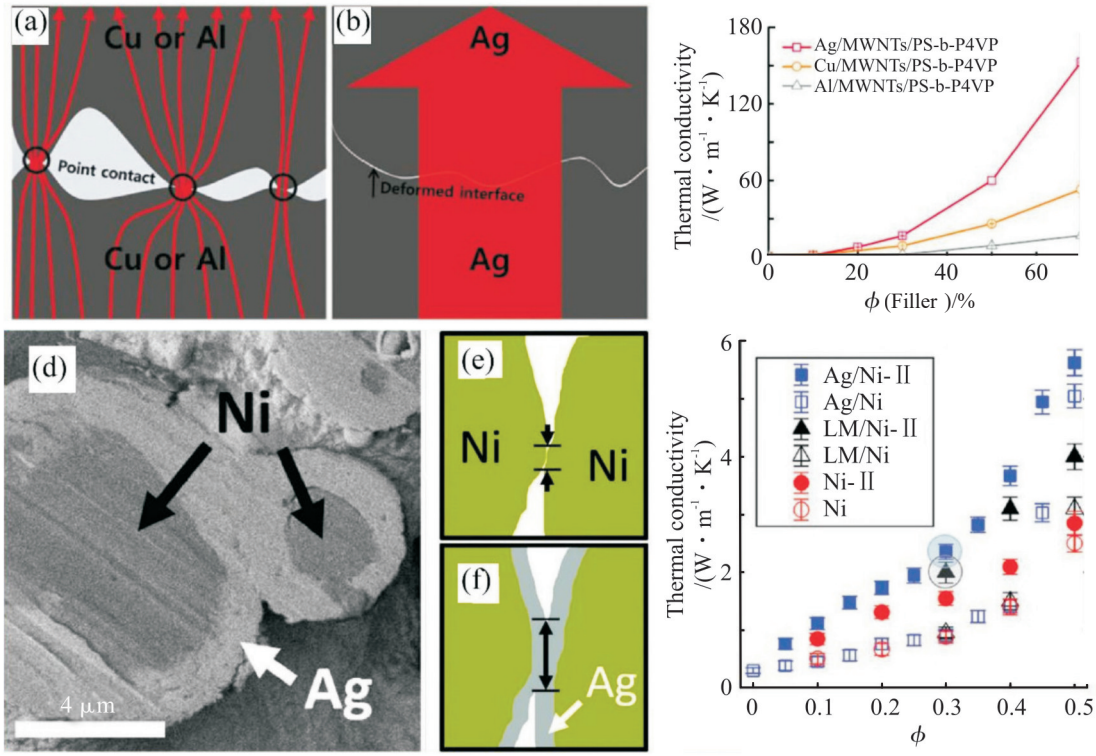


Fig. 4 Schematic illustrations of heat conduction through (a) rarely deformable Cu or Al particles and (b) deformed Ag particles; (c) thermal conductivities (TC) of hybrid composites with various metal particles of Ag, Al and Cu[14]; (d) SEM of the cross-section of the silver-coated nickel powder; illustration showing the small interface area between (e) two nickel particles compared to (f) the larger interface of the deformed/fused silver – silver interface; (g) TC of nickel particle composites as well as silver-coated nickel composites[15]

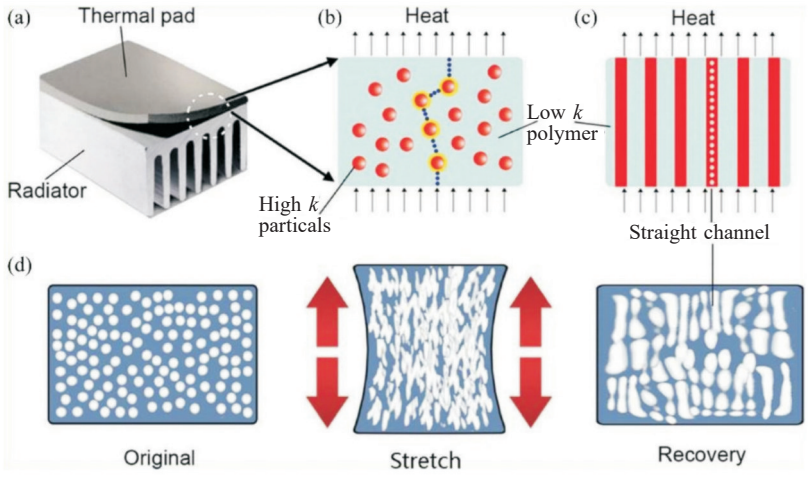


Fig. 5 (a, b) Schematic illustration of the typical TIM pad-heat sink heat dissipation system; (c) schematic of ideal thermal interface materials; (d) schematic representation of the LM/PDMS composite TIM[16]

由于银的价格较贵，Ralphs等[15]使用银包覆镍颗粒作为导热填料(Fig.4(d)),与表面未包覆银的镍颗粒相比，在相同填充量下复合材料的热导率明显更高(Fig.4(g))。如Fig.4(e,f)所示，镍颗粒的银外壳在压力作用下会发生变形，从而形成了更好的热接触；而表面未包覆银的镍颗粒，接触形式则以点接触为主，填料之间直接接触面积很小。

液态金属是室温呈液态的金属材料，它既具有一般金属的高热导率，又具有很低的模量和很好的流动性，近年来作为一种新型导热填料逐渐得到应

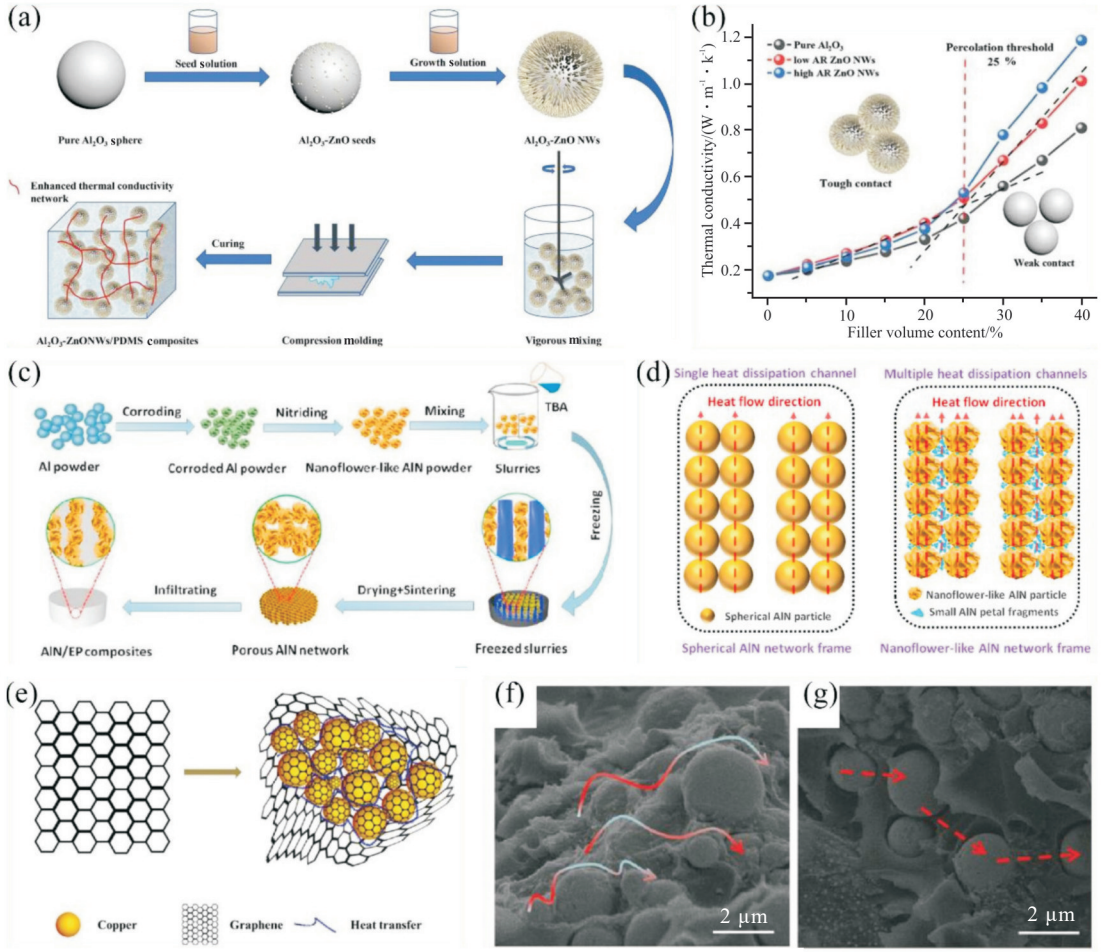


Fig. 6 (a) Flow chart of the fabrication of Al_2O_3- ZnO NWs/PDMS thermally conductive composites; (b) variation of thermal conductivity for Al_2O_3/PDMS and Al_2O_3 – ZnO NWs/PDMS hybrid composites[21]; (c, d) schematic of the preparation process of the nanoflower-like AlN-epoxy composites and thermally conductive paths for epoxy composites with vertical AlN channels[22]; (e) schematic of the interfacial conductive pathways in the Cu@rGO/epoxy composites; cross-sectional images of (f) Cu@rGO/EP and (g) Cu/EP[23]

用[16~18]。如Fig.5(a,b)所示,传统的导热复合材料通常使用高模量的导热填料,其内部的界面热阻使得热传输很慢。而使用液态金属微滴作为导热填料时,在受到拉伸应力时液态金属微滴会发生变形并相互融合,形成类似Fig.5(c)中那样连续的没有界面热阻的线状导热微通道。而且这些导热微通道一旦形成,即使在应力消失后,也会保持不变(Fig.5(d))。这得益于这些线状导热微通道,使复合材料的热导率高达8.3 W/(m·K)[16]。此外与银一样,液态金属除了可以直接作为导热填料以外,还可以包覆在其它高模量导热填料表面以改善填料之间的接触[19,20]。

考虑到球形填料之间单个接触点的实际面积很小,而接触点的数量又较少,Drummer等[21]通过水热法在球形氧化铝表面垂直生长了氧化锌纳米线(Fig.6(a))。氧化铝表面的氧化锌纳米线可以显著增加临近填料之间的接触点,增加导热通路的数量,从而降低界面热阻。

Xiang等[22]以金属铝作为铝源,通过腐蚀-氮化的方式制备了纳米花状氮化铝颗粒(Fig.6(c)),氮化铝表面特殊的纳米花瓣结构可以显著增加邻近填料之间的接触面积,并增加导热通路的数量。

Fu等[23]通过静电自组装法在铜微球表面包覆了还原氧化石墨烯(Fig.6(e)),铜微球表面的三维结构石墨烯可以作为桥梁连接邻近的铜微球,从而改善填料之间的接触,建立连续性更好、热阻更低的导热通路(Fig.6(f))。

2.2　提高填料之间的键接强度

Yuan等[24]将球形氧化铝高温烧结变为支化结构

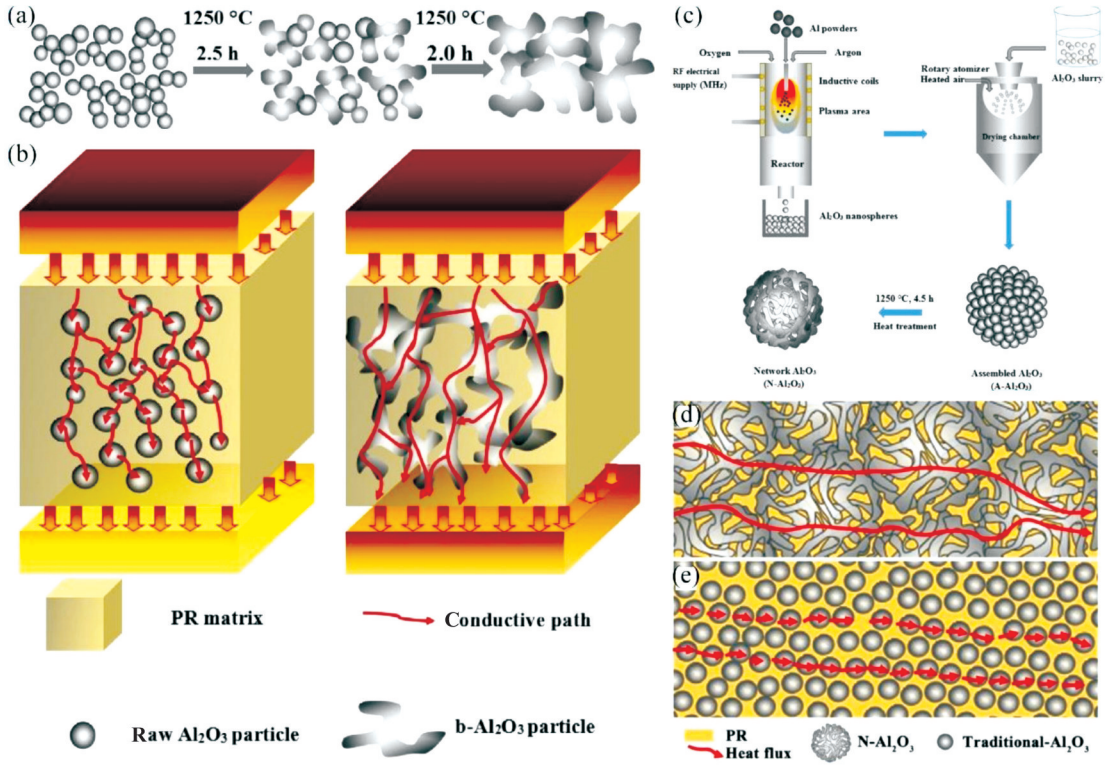


Fig. 7 (a) Schematic of evolution process of branched structures during heat treatment; (b) heat flow models of raw Al_2O_3/PR and b-Al_2O_3/PR composites[24]; (c) illustration of the preparation process of N-Al_2O_3 spheres; heat flow models of (d) N-Al_2O_3/PR composite and (e) PR composite with traditional fillers[25]

氧化铝(Fig.7(a)),高长径比的支化结构氧化铝相较于球形氧化铝更容易相互接触形成导热通路,更重要的是,高温烧结过程中部分氧化铝由物理接触(范德华力作用)变为了共价键键接,使得导热通路中填料之间的界面热阻大幅降低(Fig.7(b))。Yuan等[25]还制备了一种新型的网络状氧化铝:首先通过射频感应等离子体法制备了氧化铝纳米球,再将氧化铝纳米球通过喷雾干燥和高温烧结工艺制备成为网络状微球(Fig.7(c))。与氧化铝纳米球相比,氧化铝网络状微球具有多维连续性更好的长程声子传输通道(Fig.7(d,e))。

制备陶瓷泡沫可以显著增强填料之间的键接强度,常见的方法有凝胶灌注烧结法[2]、蛋白质泡沫法[26,27]、发泡法[28]和盐模板法[29]等。例如Tian等[27]通过蛋白质发泡法结合高温烧结制备了氧化铝泡沫,然后抽滤得到了氧化铝泡沫/环氧树脂复合材料(Fig.8(a))。无规分布氧化铝/环氧树脂复合材料中声子在界面处存在很强的散射(Fig.8(b)),而氧化铝泡沫/环氧树脂复合材料中声子传输界面散射很小,声子平均自由程要高得多(Fig.8(c));此外,烧结过程中晶界的合并使得氧化铝之间的界面热阻大大降低(由物理接触变为共价键键接),因此氧化铝泡沫/环氧复合材料具有更高的热传导效率。除了陶瓷泡沫以外,通过氢气泡模板法制备多孔金属也是增强金属填料键接强度的常用方法[30]。

金属颗粒表面的原子彼此间键接较少,它们离开颗粒的表面只需要很低的能量,因此金属颗粒的熔点会随着粒径的减小而降低,尤其是当粒径达到纳米级别的时候,熔点降低的速率会越来越快(Fig.9(a))[31],因此可以利用纳米金属颗粒的低熔点来提高金属填料之间的键接强度。Theodorian等[32]使用PVP包覆粒径20 nm左右银颗粒作为导热填料填充环氧树脂,如Fig.9(b)所示,该复合材料固化分为3个阶段:(1) 20 ℃左右纳米银粉在树脂基体中团聚;(2) 100 ℃纳米银粉表面包覆的PVP有机层去除;(3) 150 ℃左右纳米银颗粒发生烧结同时环氧树脂发生固化。烧结过程中纳米银颗粒之间由范德华力相互作用变为金属键键接,并形成高长径比的树状导热

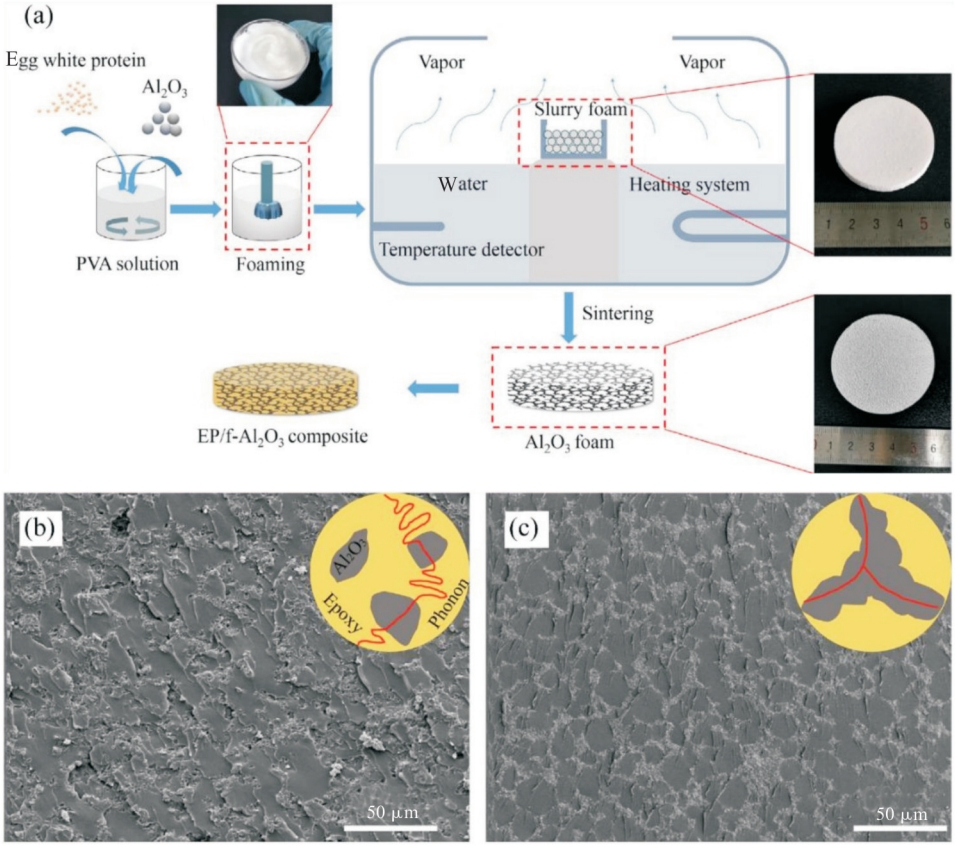


Fig. 8 (a) Flow chart of the preparation of porous Al_2O_3 framework (f-Al_2O_3) and EP/f-Al_2O_3 composite[27]; SEM fracture morphologies of (b) EP/Al_2O_3 composites and (c) EP/f-Al_2O_3 composites, respectively. Schematic diagrams in (b) and (c) present the different phonon transmission path

网络,这使得复合材料的热导率与树脂基体相比提高了近200倍。

除了直接将纳米银颗粒作为导热填料使用以外,在填料表面沉积银纳米颗粒也是降低填料之间界面热阻的常用方法。例如,曾小亮等[33]通过液相还原法在六方氮化硼纳米片表面沉积了银纳米颗粒,银纳米颗粒的桥联作用有效降低了六方氮化硼纳米片之间的界面热阻(Fig.9(c))。Kim等[34]在碳纳米管表面沉积了银纳米颗粒,一维碳纳米管在微米银片之间构筑了有效导热通路,二者之间的接触界面则通过银纳米颗粒和微米银片的融合形成金属键键接来增强热传输。该方法制备得到的环氧复合材料的热导率高达160 W/(m·K),与并联模型预测值的上限很接近(Fig.9(d))。

碳纤维和碳纳米管是常用的碳基导热填料,但碳材料之间的接触界面处往往存在很高的热阻,在其表面镀一层金属是降低界面热阻的常见方法[35~40]。例如Wang等[36]通过电镀的方法在碳毡表面包覆了一层铜,表面镀铜碳毡/环氧复合材料热的导率高达30.69 W/(m·K),而碳毡/环氧复合材料的热导率仅为0.28 W/(m·K)。碳毡/环氧复合材料热导率较低的主要原因是碳毡中碳纤维之间界面处存在很大的热阻,热阻主要源于临近碳纤维之间明显的缝隙(实际接触面积很小),如Fig.10(a),而表面镀铜碳毡/环氧复合材料中,碳纤维表面均匀紧凑的铜层可以连接邻近的碳纤维,热量可以通过电子的运动沿着三维铜层导热通道快速传递(Fig.10(b))。

除了金属纳米颗粒以外,金属纳米线也是常用的导热填料。Ramanath等[41]使用平均直径5 nm的金纳米线作为导热填料,结果在填充体积分数不足3%时,复合材料的热导率便增强了近30倍,这一结果甚至超过了理论模型的预测值。其原因在于直径小于10 nm的金纳米线只需在低应力作用下表面原子就会快速扩散而发生冷焊[42],在此过程中纳米线

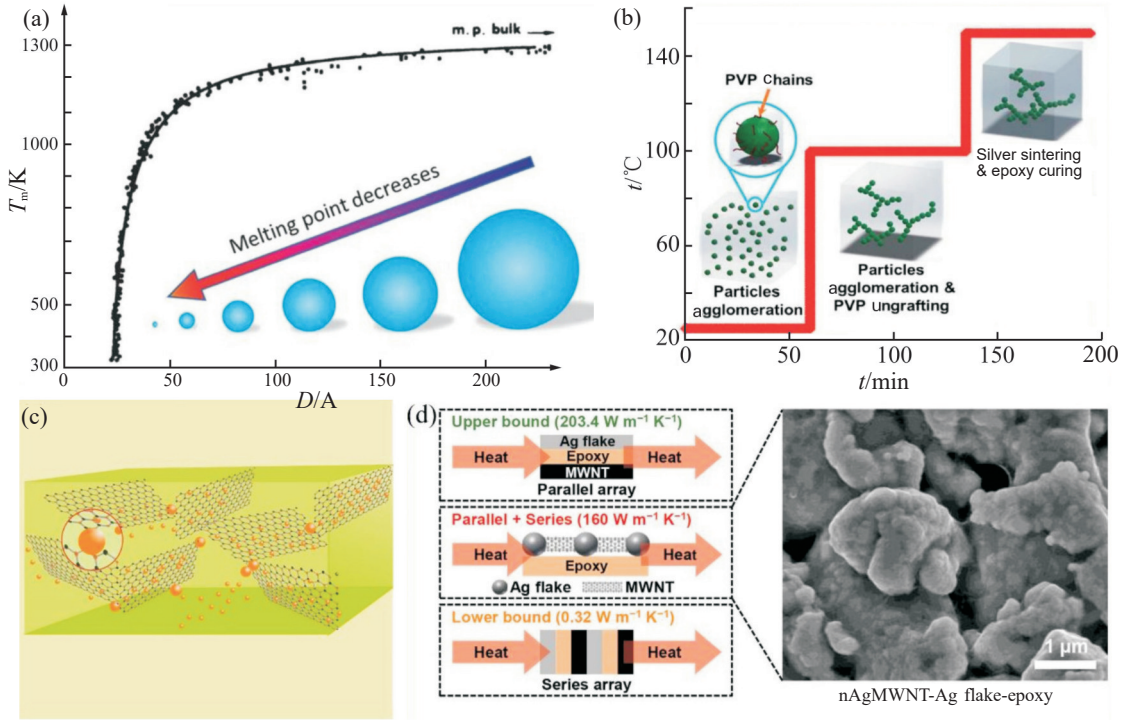


Fig. 9 (a) Melting point vs. particle diameter of gold nanoparticles[31]; (b) three-step optimum processing for sintering of AgNPs[32]; (c) schematic to show how heat to be dissipated between BN layers and AgNPs[33]; (d) SEM image of nAgMWNT – Ag-flake – epoxy composites and the theoretically estimated thermal conductivities provided with model schematics

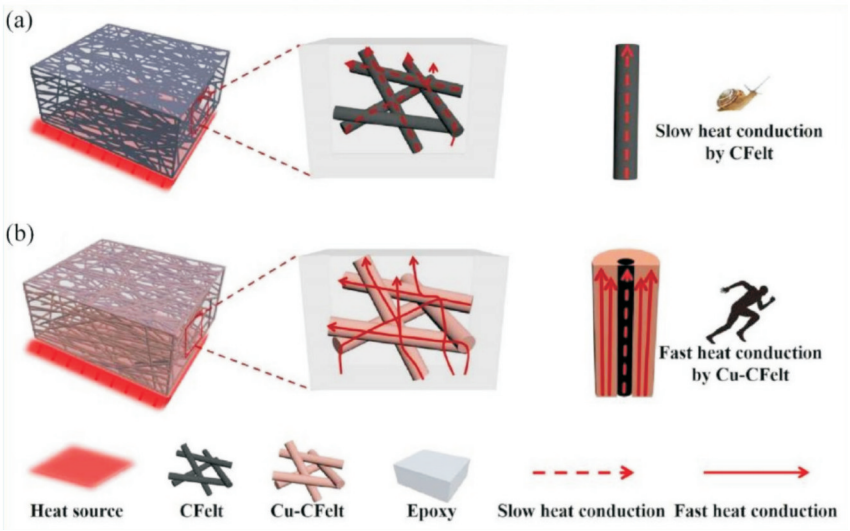


Fig. 10 Schematic illustration of the heat conduction in the (a) CFelt/epoxy and (b) Cu-CFelt/epoxy composites[36]

之间的相互作用由机械接触转变为金属键连接，界面热阻大幅降低，热传输效率大大增强。

对于直径大于10 nm，难以发生冷焊的金属纳米线则可以采用微波焊接[43,44]的方式来降低界面热阻。例如Simon等[44]使用不同的方法制备了3种复合材料：复合材料A是直接将铜纳米线通过剪切分散在硅橡胶中制备而成；而复合材料B则首先将铜纳米线溶液冷冻干燥后形成海绵状逾渗网络，然后再将硅橡胶注入其中，形成复合材料；复合材料C则是对复合材料B进行微波辐射处理制备得到(Fig.11(a))。从Fig.11(d)中可以看出，复合材料C的热导率明显高于A和B，尤其是在高填料含量下。复合材料

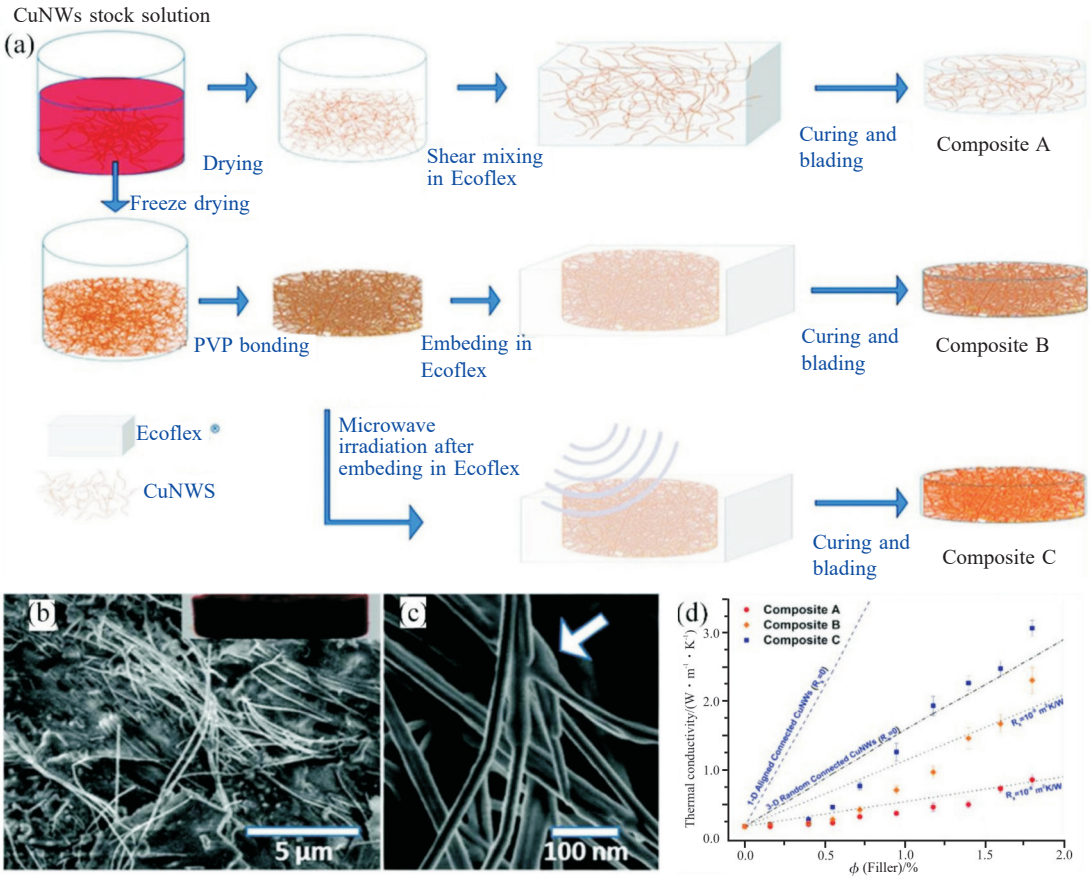


Fig. 11 (a) Schematic of fabrication of composites A, B, and C; (b, c) magnified SEM image of the composite highlighting welding of CuNWs; (d) thermal conductivity of composites A, B, and C with respect to filler loadings[44]

A中剪切分散的铜纳米线之间缺少直接接触,形成的导热通路数量少导致热导率较低。复合材料B中铜纳米线虽然可以直接接触形成较多的导热通道,但它们之间的接触面积很小,界面处依旧存在很高的热阻。复合材料C中铜纳米线直径小于微波长度,因而可以吸收微波的能量受热而相互融合,这不仅增加了铜纳米线之间的接触面积,而且使得铜纳米线之间由物理接触变为金属键键接。Fig.11(c)中铜纳米线接触结点处的膨胀可以证明铜纳米线之间发生了融合。值得注意的是,微波辐射时间不宜过长,若时间过长则会导致金属纳米线的断裂,从而降低铜纳米线的长径比,造成导热网络的不连续,复合材料热导率的下降。

3　总结和展望

降低填料之间的热阻是提高复合材料热导率增强效率的有效方法。本文介绍了影响填料之间界面热阻的因素及机理,综述了降低各种填料(如陶瓷填料、金属填料(包括液态金属、纳米颗粒和纳米线)和碳基材料)之间界面热阻的方法。但还需要解决以下几个理论和技术问题,这对于完善复合材料的导热理论和将学术研究转化为工业化应用非常重要。

(1)目前已有的各种复合材料的热导率预测模型几乎都不涉及填料之间的界面热阻,因此发展新的涉及填料之间接触面积和键接强度的热导率预测模型对于高导热复合材料的设计和制备具有理论指导意义。

(2)使用银粉和液态金属是提高填料之间接触面积,降低界面热阻的常用方法。但银粉价格较贵,在廉价填料表面包覆一层银(如化学镀银等)可以有效降低成本。液态金属本身的流动性使得它很容易从树脂基体中泄露,从而腐蚀金属散热器,降低散热效率,因此液态金属作为导热填料时必须解决其易泄露的问题。

（3）制备陶瓷泡沫可以提高填料之间的键接强度，从而降低界面热阻，但其制备工艺较为复杂，目前在大规模应用中还存在困难。此外这种方法制备的导热复合材料作为热界面材料应用时也存在较大的问题，这是因为热界面材料通常要求具有一定的压缩性和适形性。

参考文献:

[1] Chung D D L. Performance of thermal interface materials[J]. Small, 2022, 18: 2200693.

[2] Hu Y, Du G, Chen N. A novel approach for Al_2O_3 /epoxy composites with high strength and thermal conductivity[J]. Composites Science and Technology, 2016,124: 36-43.

[3] Guo Y, Ruan K, Shi X, *et al*. Factors affecting thermal conductivities of the polymers and polymer composites: a review [J].Composites Science and Technology, 2020, 193: 108134.

[4] Xu X, Chen J, Zhou J, *et al.* Thermal conductivity of polymers and their nanocomposites[J]. Advanced Materials, 2018, 30: 1705544.

[5] Cui W, Du F, Zhao J, *et al*. Improving thermal conductivity while retaining high electrical resistivity of epoxy composites by incorporating silica- coated multi- walled carbon nanotubes[J]. Carbon, 2011, 49: 495-500.

[6] Jiang Y, Li M, Chen C, *et al.* Effect of elastic modulus mismatch of epoxy/titanium dioxide coated silver nanowire composites on the performance of thermal conductivity[J]. Composites Science and Technology, 2018, 165: 206-213.

[7] Su Y, Li J J, Weng G J. Theory of thermal conductivity of graphene- polymer nanocomposites with interfacial Kapitza resistance and graphene- graphene contact resistance[J]. Carbon, 2018, 137: 222-233.

[8] Zhou S, Xu J, Yang Q, *et al.* Experiments and modeling of thermal conductivity of flake graphite/polymer composites affected by adding carbon- based nano- fillers[J]. Carbon, 2013, 57: 452-459.

[9] Yang J, Waltermire S, Chen Y, *et al.* Contact thermal resistance between individual multiwall carbon nanotubes[J]. Applied Physics Letters, 2010, 96: 23109.

[10] Liu C, Wei Z, Wang J, *et al*. The contact area dependent interfacial thermal conductance[J]. AIP Advances, 2015, 5: 127111.

[11] Harikrishna H, Ducker W A, Huxtable S T. The influence of interface bonding on thermal transport through solid – liquid interfaces[J]. Applied Physics Letters, 2013, 102: 251606.

[12] Wei X, Zhang T, Luo T. Thermal energy transport across hard-soft interfaces[J]. ACS Energy Letters, 2017,2: 2283-2292.

[13] Zhang X D, Yang G, Cao B Y. Bonding - enhanced interfacial thermal transport: mechanisms, materials, and applications[J]. Advanced Materials Interfaces, 2022,9: 2200078.

[14] Choi J R, Yu S, Jung H, *et al*. Self- assembled block copolymer micelles with silver- carbon nanotube hybrid fillers for high performance thermal conduction[J]. Nanoscale, 2015,7: 1888-1895.

[15] Ralphs M, Kong W, Wang R Y, *et al.* Thermal conductivity enhancement of soft polymer composites through magnetically induced percolation and particle – particle contact engineering [J]. Advanced Materials Interfaces, 2019, 6: 1801857.

[16] Zhao L, Liu H, Chen X, *et al.* Liquid metal nano/micro-channels as thermal interface materials for efficient energy saving[J]. Journal of Materials Chemistry C, 2018, 6: 10611-10617.

[17] Shu M, Zhang P, Ding X, *et al*. Low melting point alloy segregated network construction in thermosetting polymer matrix for the electromagnetic interference shielding and thermal conductivity enhancement[J]. Materials Research Express, 2019, 6: 115352.

[18] Bartlett M D, Kazem N, Powell- Palm M J, *et al*. High thermal conductivity in soft elastomers with elongated liquid metal inclusions[J]. Proceedings of the National Academy of Sciences, 2017,114: 2143-2148.

[19] Ralphs M I, Kemme N, Vartak P B, *et al.* In situ alloying of thermally conductive polymer composites by combining liquid

and solid metal microadditives[J]. ACS Applied Materials & Interfaces, 2018, 10: 2083-2092.

[20] Uppal A, Kong W, Rana A, *et al.* Enhancing thermal transport in silicone composites via bridging liquid metal fillers with reactive metal co- fillers and matrix viscosity tuning[J]. ACS Applied Materials & Interfaces, 2021, 13: 43348-43355.

[21] Liu C, Wu W, Drummer D, *et al*. ZnO nanowire-decorated Al_2O_3 hybrids for improving the thermal conductivity of polymer composites[J]. Journal of Materials Chemistry C, 2020, 8: 538-5388.

[22] Liu L, Xiang D, Wu L. Improved thermal conductivity of ceramic- epoxy composites by constructing vertically aligned nanoflower- like AlN network[J]. Ceramics International, 2022, 48: 10438-10446.

[23] Liu S, Zhao B, Jiang L, *et al.* Core-shell Cu@rGO hybrids filled in epoxy composites with high thermal conduction[J]. Journal of Materials Chemistry C, 2018, 6: 257-265.

[24] Ouyang Y, Hou G, Bai L, *et al.* Constructing continuous networks by branched alumina for enhanced thermal conductivity of polymer composites[J]. Composites Science and Technology, 2018, 165: 307-313.

[25] Ouyang Y, Ding F, Bai L, *et al.* Design of network Al_2O_3 spheres for significantly enhanced thermal conductivity of polymer composites[J]. Composites Part A: Applied Science and Manufacturing, 2020, 128: 105673.

[26] Yin L, Zhou X, Yu J, *et al.* Fabrication of a polymer composite with high thermal conductivity based on sintered silicon nitride foam[J]. Composites Part A: Applied Science and Manufacturing, 2016, 90: 626-632.

[27] Xiao C, Chen L, Tang Y, *et al.* Three dimensional porous alumina network for polymer composites with enhanced thermal conductivity[J]. Composites Part A: Applied Science and Manufacturing, 2019, 124: 105511.

[28] Tian Z, Sun J, Wang S, *et al.* A thermal interface material based on foam- templated three- dimensional hierarchical porous boron nitride[J]. Journal of Materials Chemistry A, 2018, 6: 17540-17547.

[29] Pan D, Li Q, Zhang W, *et al.* Highly thermal conductive epoxy nanocomposites filled with 3D BN/C spatial network prepared by salt template assisted method[J]. Composites Part B: Engineering, 2021, 209: 108609.

[30] Lin C H, Izard A G, Valdevit L, *et al.* Mechanically compliant thermal interfaces using biporous copper- polydimethylsiloxane interpenetrating phase composite[J]. Advanced Materials Interfaces, 2021, 8: 2001423.

[31] Yang C, Wong C P, Yuen M M F. Printed electrically conductive composites: conductive filler designs and surface engineering[J]. Journal of Materials Chemistry C, 2013, 1: 4052.

[32] Pashayi K, Fard H R, Lai F, *et al.* Self- constructed tree- shape high thermal conductivity nanosilver networks in epoxy[J]. Nanoscale, 2014, 6: 4292-4296.

[33] Wang F, Zeng X, Yao Y, *et al.* Silver nanoparticle- deposited boron nitride nanosheets as fillers for polymeric composites with high thermal conductivity[J]. Scientific Reports, 2016, 6: 19394.

[34] Suh D, Moon C M, Kim D, *et al.* Ultrahigh thermal conductivity of interface materials by silver- functionalized carbon nanotube phonon conduits[J]. Advanced Materials, 2016, 28: 7220-7227.

[35] Yu S, Park B, Park C, *et al.* RTA-treated carbon fiber/copper core/shell hybrid for thermally conductive composites[J]. ACS Applied Materials & Interfaces, 2014, 6: 7498-7503.

[36] Xu F, Cui Y, Bao D, *et al*. A 3D interconnected Cu network supported by carbon felt skeleton for highly thermally conductive epoxy composites[J]. Chemical Engineering Journal, 2020, 388: 124287.

[37] Sun Y, Zhang M, Zhang Y, *et al.* Fabrication of high thermal and electrical conductivity composites via electroplating Cu network on 3D PEEK/CF felt skeletons[J]. Composites Communications, 2021, 28: 100909.

[38] Yan F, Liu L, Li M, *et al.* One-step electrodeposition of Cu/CNT/CF multiscale reinforcement with substantially improved

thermal/electrical conductivity and interfacial properties of epoxy composites[J]. Composites Part A: Applied Science and Manufacturing, 2019, 125: 105530.

[39] Carlberg B, Ye L, Liu J. Polymer-metal nanofibrous composite for thermal management of microsystems[J]. Materials Letters, 2012, 75: 229-232.

[40] Ganguli S, Reed A, Jayasinghe C, *et al*. A simultaneous increase in the thermal and electrical transport in carbon nanotube yarns induced by inter-tube metallic welding[J]. Carbon, 2013, 59: 479-486.

[41] Balachander N, Seshadri I, Mehta R J, *et al.* Nanowire-filled polymer composites with ultrahigh thermal conductivity[J]. Applied Physics Letters, 2013, 102: 93117.

[42] Lu Y, Huang J Y, Wang C, *et al.* Cold welding of ultrathin gold nanowires[J]. Nature Nanotechnology, 2010, 5: 218-224.

[43] Seshadri I, Esquenazi G L, Borca-Tasciuc T, *et al*. Multifold increases in thermal conductivity of polymer nanocomposites through microwave welding of metal nanowire fillers[J]. Advanced Materials Interfaces, 2015, 2: 1500186.

[44] Bhanushali S, Ghosh P C, Simon G P, *et al.* Copper nanowire-filled soft elastomer composites for applications as thermal interface materials[J]. Advanced Materials Interfaces, 2017, 4: 1700387.

Progress in Decreasing the Thermal Interfacial Resistance Between Fillers for Thermally Conductive Composites

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ABSTRACT: The low efficiency of thermal conductivity enhancement of composites is due to the existence of interfacial thermal resistance: the interface thermal resistance between fillers and resin matrix, and the interface thermal resistance between fillers. Most of the existing studies were focused on decreasing the thermal interfacial resistance between fillers and resin matrix. While the interfacial thermal resistance between fillers is the key factor to determine the thermal conductivity of composites. This paper discussed the recent research results of reducing the interface thermal resistance between fillers from two aspects of increasing the contact area and improving the bond strength between fillers. This review offered some guidance for the design and fabrication of highly thermally conductive composites.

Keywords: composite; thermal conductivity; filler; thermal interfacial resistance