

Thermally Conductive Adhesive in Electronic Packaging

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Contributor: M Abdullah

The application of epoxy adhesive is widespread in electronic packaging. Epoxy adhesives can be integrated with various types of nanoparticles for enhancing thermal conductivity. The joints with thermally conductive adhesive (TCA) are preferred for research and advances in thermal management. The rapid advancement of microelectronic technology has resulted in the progressive transformation of electronic components from isolated to highly integrated. It produces a lot of heat while they are functioning. The materials and structures used to attach the semiconductor chip to other electronic components involve the sector of electronic packaging. Conductive adhesives represent a significant advancement in electronic packaging for advanced devices, where effective heat dissipation and enough electrical conductivity are critical. The TCA joint is a crucial choice for the thermal management of the device. The advances in TCA are essential because of future demands for inexpensive and handy electronic devices. The adhesive applications have become more important due to their remarkable versatility and unparalleled compatibility. The electronic industry has indeed developed, especially in consumer electronics, which depends on conductive adhesives. It would not be possible to identify the most modern electronic devices without conductive adhesives.

Keywords: epoxy adhesive ; electronic packaging ; thermal conductivity ; conductive filler ; thermally conductive adhesive (TCA)

1. Formulation of Thermally Conductive Adhesive and Heat Transfer Mechanism

A polymer matrix and thermally conductive filler make thermally conductive adhesives that are formed as composites. Nanoparticles are having a diameter of less than 100 nm exhibit unique physical and mechanical properties. These fillers have a large specific surface area, high surface energy, and interfacial area with the polymeric matrix ^[1]. Thermal conductivity of epoxy is inadequate to remove heat produced from electronic devices. Dispersing nanoparticles inside an epoxy adhesive matrix can dramatically improve the adhesive's thermal properties ^[2] and the performance of adhesively bonded joints ^[3]. The high thermal conductive solid particles like—graphene ^[4], carbon nanotubes ^[5], carbon fiber, carbon black are the most common carbon-based fillers. Metallic (gold, copper, silver, aluminium) ^[6], and ceramic (aluminium nitride, boron nitride, silicon carbide, aluminium oxide) ^[7] fillers have been blended with matrix resin to increase the thermal conductivity. Carbon-based and metallic filler adhesive has received significant attention because of its excellent thermal conductivity and specific surface area ^[8]. These conductive filler particles can alter polymer matrix isolation properties by making them very heat conductive. The thermally conductive adhesive is a composite material, and it follows the same formulation process of nano-composite ^[9]. Most common formulation process starts with the filler's dispersion with organic solvents. An example of graphene nanoparticle (GNP)-based nano-composite ^[10] is shown in **Figure 1**. The process until the application, i.e., (a) to (h), is maintained for all thermally conductive adhesive according to the filler type. Bubbles and voids reduce the bonding quality of the adhesive ^[11]. To avoid the bubble and void in the adhesive joint the vacuum degassing is important in thermally conductive adhesive production process.

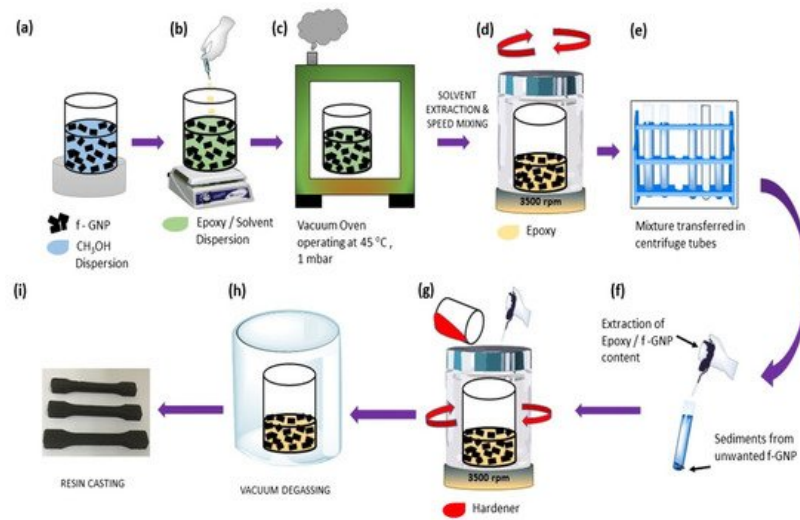


Figure 1. Schematic of the dispersion (a) to application (i) process of conductive filler in nano-composite [10].

The non-conductive properties can be transformed into conductive properties with the addition of fine conductive filler particles in polymer matrices, resulting in a continuous thermal conductive network [12]. The thermal interface material (TIM) required high thermal conductivity. The thermal conductivity of polymers ranged from 0.1 to 0.5 Wm⁻¹ K⁻¹ [13]. The simple form of polymer matrix is insufficient to meet the thermal conductivity requirement. Conductive filler particles increase heat conductivity while conserving polymer matrix characteristics [14]. Before integrating the epoxy with the filler, it is essential to know the thermal conductivity of the filler. A list of widely used conductive fillers is presented in **Table 1**.

Table 1. Thermal conductivity (TC) of materials used as conductive fillers [15][16][17][18][19].

Material	TC (Wm ⁻¹ K ⁻¹)	Material	TC (Wm ⁻¹ K ⁻¹)
Aluminum oxide	20–30	Graphite	100–400 (on plane)
Molybdenum	142	Silver	450
Tungsten	155	Copper	401
Nickel	158	Silicon carbide (SiC)	490
Aluminum	204	Diamond	2000
Beryllium oxide	260	Boron nitride	~2000 (in-plane); ~380 (out-of-plane)
Carbon fiber	260	Multiwalled carbon nanotube (MWCNT)	~3000
Aluminum nitride (AlN)	200–320	Graphene	~5300
Gold	345	Single-walled carbon nanotube (SWCNT)	~6000

Electrons and phonons are responsible to heat transfer in metal. Due to the lack of free electrons in non-metals, the phonon is responsible for heat transmission. When a polymer surface comes into contact with a heat source, heat is transferred via vibrations to the nearest atom, then to the next, and so on [20]. The formulation of thermally conductive adhesives consists of a polymer matrix, filler, and specific additives that are uniform and isotropic at the macro scale.

The thermal conductivity of the polymer matrix is less than 0.5 Wm⁻¹ K⁻¹. Thermal conductivity is improved by incorporating a highly thermally conductive filler into the insulating polymer matrix. The filler has a significant influence on the thermal conductivity of polymer composites. In the case of the filler network, it can speed heat transmission along the continuous and integrated filler network with less phonon scattering. Significantly, optimizing the filler structure (including size and aspect ratio) and dispersion can improve thermal conductivity by arranging the filler in a thermally conductive network within the polymer matrix [21]. Therefore, heat transmission across TCA occurs when filler particles establish a conductive path. A general heat transfer mechanism of TCA is shown in **Figure 2**.

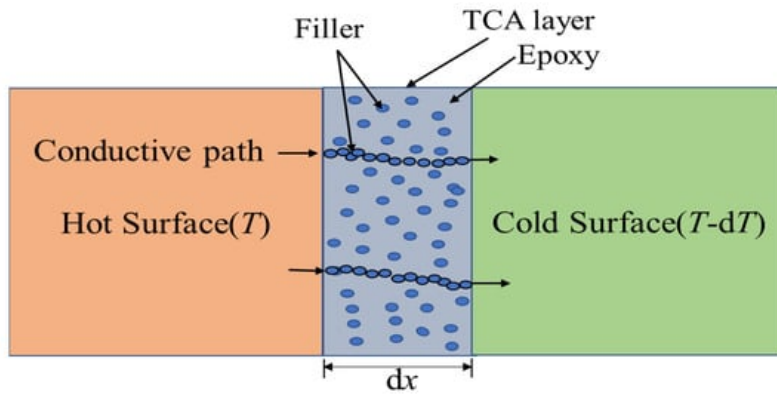


Figure 2. Heat conduction mechanism of TCA.

The basic principle of heat conductance through a thermally conductive adhesive layer dx (**Figure 2**), also known as Fourier's law [22], states that the rate of heat transfer through a material is proportional to the negative gradient in the temperature and to the area at right angles to that gradient through which the heat flows and can be written as follows:

$$\frac{dQ}{dt} = -\lambda A \frac{dT}{dx} \quad (1)$$

where Q is the quantity of heat energy (J), t is time (s), λ is a thermal conductivity ($\text{Wm}^{-1} \text{K}^{-1}$), dT/dx is the temperature gradient in the heat flow direction (K/m), x is the distance along the direction of heat flow (m), A is the area of the cross-section (m^2). The thermal conductivity is given by:

$$\lambda = \frac{1}{3} (C_e V_e L_e + C_{ph} V_{ph} L_{ph}) = \lambda_e + \lambda_{ph} \quad (2)$$

where C_e and C_{ph} are the heat capacities per unit volume ($\text{J/m}^3 \text{K}$) of electrons and phonons, respectively, V_e and V_{ph} are their root-mean-square velocities and L_e , L_{ph} are their mean free paths. The thermal conductivity of electron type λ_e is dominant for metals, and one can roughly assume that:

$$\lambda \cong \lambda_e \quad (3)$$

In such a case, the ratio of thermal conductivity λ_e and electrical conductivity σ_e , according to Wiedemann–Franz's law, can be expressed as:

$$\frac{\sigma_e}{\lambda_e} = LT \quad (4)$$

where T is temperature (K) and L is the Lorenz constant, the theoretical value of which is $2.44 \times 10^{-8} \text{ W}\cdot\Omega/\text{K}^2$. The transport of heat in nonmetals occurs mainly by phonons. For insulators, thermal conductivity increases linearly with temperature, and the efficiency of phonon heat conduction is size dependent. In nanostructures, the thermal conductivity of a phonon type λ_{ph} may be drastically different than in macrostructures. When the size of a nanostructure approaches or exceeds the mean-free path of a phonon, phonons collide with the border more frequently than in bulk materials. This extra collision mechanism increases the resistance to heat transfer, hence lowering the effective thermal conductivity of thin films, wires, nanotubes, and other nanoparticles [29].

2. Adhesives with Improved Conductivity

Epoxy is inferior in heat conductivity. However, research in electronic packaging is improving the heat conductivity of nanoelectronics epoxy adhesion. The conductivity range of TCAs is reported as $1\text{--}30 \text{ Wm}^{-1} \text{K}^{-1}$ for sufficient heat transfer [23]. The thermal conductivity of epoxy has been increased using ceramic [24], metallic [25], and carbon-based [26] conductive fillers to increase the thermal conductivity of the epoxy, which are described below.

2.1. Ceramic-Based Fillers

Chip power density is a significant factor in the performance of electronic devices. TCAs minimize the thermal resistance between the electronic devices' cooling medium and the outside environment. A ceramic substance is used to insulate to prevent electrical shorts [27] and enhance electronic equipment's effectiveness [28]. Highly thermally conductive ceramic fillers have been added in epoxy adhesive systems, such as boron nitride (BN), aluminium nitride (AlN), alumina (Al_2O_3), and silicon carbide (SiC) particles. **Figure 3** shows various types of thermally conductive ceramic used in TCAs.



Figure 3. Commonly used thermally conductive ceramics filler.

2.1.1. Boron Nitride

Boron nitride (BN) has the same number of boron (B) and nitrogen (N) atoms and is isoelectronic to carbon structures [29]. These particles have received considerable interest because of their excellent characteristics in all aspects. It is also referred to as “white graphene” because of its honeycomb composition. It has an insulating property with a bandgap of 5.2 eV and possesses considerable thermal conductivity (experimentally determined $360 \text{ Wm}^{-1} \text{ K}^{-1}$ and theoretically $2000 \text{ Wm}^{-1} \text{ K}^{-1}$ [30]. The findings are outstanding since the dielectric characteristics and the anti-oxidation resistance were found to be excellent. Electronic devices require better thermal management to match the rising demand for BN and epoxy mixed composites. The hexagonal form of BN (h-BN) [31] is comparable to graphite, the cubic form (c-BN) is like diamond [32], and the amorphous form (a-BN) is similar to amorphous carbon [33]. Several BN/epoxy composites have been created. However, a hybrid BN solution was unable to be found since the interfacial thermal resistance was too high [34]. BN nanoplates filler affects thermal conductivity of the composite with the increase in filler concentrations. It is found that the 2D-BN nanoplates filler at 14 wt% with silicone can reduce the mechanical strength, but the thermal conductivity is 30% better than the without filler sample. The reduction in the mechanical strength can be due to agglomeration of BN nanoparticles, whereas the increase in the thermal conductivity is due to the acceleration of phonons transmission by the BN nanoplates [35][36].

C. Xiaoa et al. [37] used hollow boron nitride microbeads (BNMB) at 65.6 vol% for improving thermal conductivity of epoxy resin. The result shows that the maximum thermal conductivity reached $17.61 \text{ W/m}^{-1} \text{ K}^{-1}$ in-plane direction and $5.08 \text{ W/m}^{-1} \text{ K}^{-1}$ for out-plane direction of BN.

2.1.2. Alumina

Alumina (Al_2O_3) is used in electronics packaging as a low-cost filler. It has high thermal conductivity ($30 \text{ Wm}^{-1} \text{ K}^{-1}$) and electrical insulation properties. Because of these unique properties become an essential filler in the thermal management of electronic devices [38]. It has been used in light-emitting diode (LED) packaging to avoid voltage drop, short circuits, or noise reduction. Alumina nanoparticles can significantly improve the mechanical properties of epoxy adhesives and interfacial wettability with an aluminum substrate [39]. A mixture of Bisphenol-F epoxy resin with 80% of $30 \mu\text{m}$ and 20% of $5 \mu\text{m}$ spherical Alumina (S- Al_2O_3) particle can increase thermal conductivity up to $1.364 \text{ Wm}^{-1} \text{ K}^{-1}$ [40].

A study by Mai et al. [41] reported that epoxy adhesive with Alumina contained hybrid filler can improve the thermal conductivity of the composite materials. Another experimental and simulation work was performed using Al_2O_3 and BN in the epoxy polymer (EP) matrix. The thermal conductivity of the BN/ Al_2O_3 /EP composite is higher than that of the Al_2O_3 /EP

composite. The thermal conductivity increases from $2.77 \text{ Wm}^{-1} \text{ K}^{-1}$ to $3.35 \text{ Wm}^{-1} \text{ K}^{-1}$ for the composite without BN loading (shown in **Figure 4**). Graphene coated alumina was used as a thermal conductivity enhancer in epoxy composite and was found to be a potential filler [42].

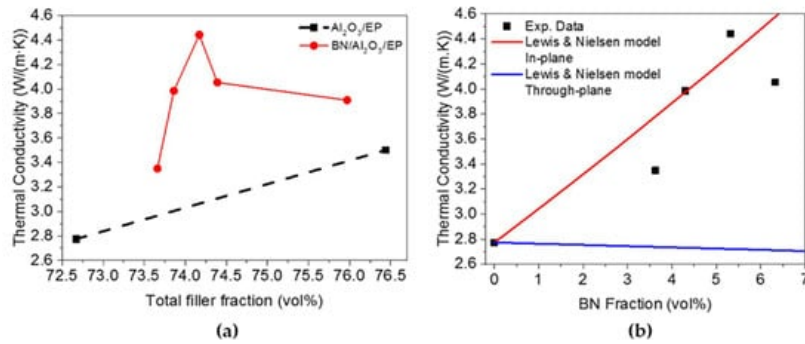


Figure 4. (a) Thermal conductivity vs. total filler fraction graph (b) Experimental and simulated data comparison graph [41].

2.1.3. Aluminum Nitride

Aluminum nitride (AlN) particles are a promising filler material in electronics packaging. The size of the AlN filler determines the characteristics of AlN-filled epoxy composites [43]. The thermal conductivity, based on AlN's particle size, ranges from 200 to $320 \text{ Wm}^{-1} \text{ K}^{-1}$ at room temperature. The maximum amount of particle in epoxy composite does not ensure the highest thermal conductivity. Research result shows that 1 wt% nano-AlN sample has superior electrical insulation and thermal conductivity among the pure epoxy, 0.5 wt%, 1 wt% and 2 wt% of AlN/epoxy solution [44]. However, magnetic-aligned AlN/epoxy composite at low filler content can effectively generate thermal transport channels and enhance thermal conductivity from $0.915 \text{ Wm}^{-1} \text{ K}^{-1}$ to $1.754 \text{ Wm}^{-1} \text{ K}^{-1}$.

The thermal conductivity of adhesive is mainly determined by the filler heat transfer capacity, density of thermal network, as well interfacial thermal resistance. Thus, the formation of effective thermal flow 3-D percolating network through synergistic effect in matrix is a crucial criterion, dominating the thermal conductivity. Yuan et al. [45] worked on different sized (5 μm , 2 μm and, 50 nm) AlN- with graphite and graphene oxide(GO) as a hybrid filler to observe the improvement of thermal conductivity of composite. The results demonstrate that large particles of AlN with epoxy are more heat conductive than small particles. Similarly, GO can improve the thermal conductivity of epoxy resin more effectively than natural graphite. In the case of a single filler, adding 70 wt% 5 μm -AlN particles to the epoxy resulted in the maximum conductivity which is 10.8 times that of pure epoxy (shown in **Figure 5**).

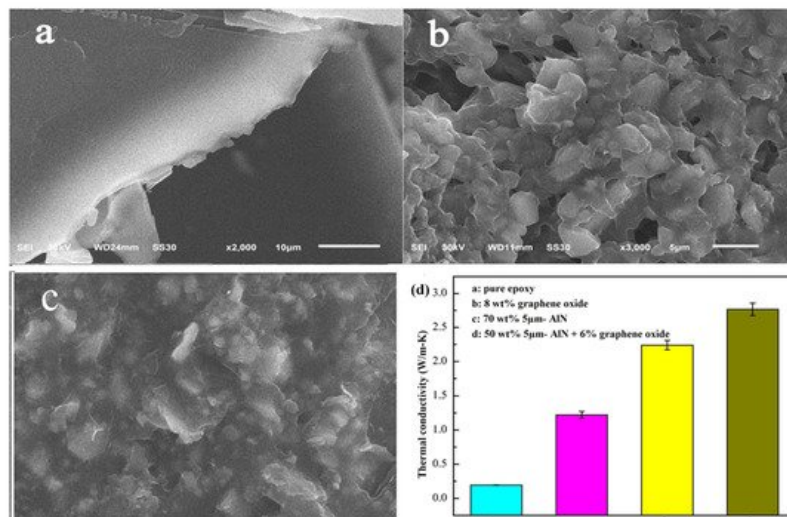


Figure 5. SEM images of epoxy adhesives containing (a) only epoxy resin, (b) epoxy with AlN particles, (c) epoxy with AlN and GO hybrid filler and (d) Thermal conductivity of different test samples [45].

The heat conductivity of epoxy adhesive containing GO is more effective than natural graphite. For single fillers, 10.8 times higher than pure epoxy is the most excellent conductivity AlN's thermal expansion coefficient (CTE) is low. It is relatively low-cost, non-toxic, and can provide a stable crystalline structure [46].

2.1.4. Silicon Carbide

The researchers are interested in the Silicon Carbide Filler (SiC) as it has greater hardness and strength, good resistance to corrosion and oxidation. SiC has high intrinsic thermal conductivity ($490 \text{ Wm}^{-1} \text{ K}^{-1}$), more than three times higher than silicon and 10 times higher than gallium arsenide and sapphire [47]. Nanowire form of SiC is a primarily familiar shape for dispersing with epoxy. Most of the recent works are focused on hybridization with SiC nanowire. According to Danyu Shen et al. [48], the thermal conductivity of 3 wt% SiC nanowire with epoxy is $0.449 \text{ Wm}^{-1} \text{ K}^{-1}$, which is 1.06 times higher than plain epoxy. Another experimental output found thermal conductivity $0.43 \text{ Wm}^{-1} \text{ K}^{-1}$ at 3.91 vol% of SiC nanowire epoxy composite [49]. BN and SiC hybrid filler with vertical alignment was examined, and thermal conductivity enhancement was found [50][51]. Carbon fibre (CF) was also used with SiC to improve the heat transfer properties and saw satisfactory improvement [52]. A summary of research output from recently published works on ceramic fillers are given in Table 2.

Table 2. Summary of the recently published works on ceramic fillers.

Filler	Conditions/Process	TC ($\text{Wm}^{-1} \text{ K}^{-1}$)	References
BN	Platelet-shaped Boron Nitride(BN) particles	3.5	[53]
BN	At 70 wt% functionalized and mix with epoxy resin	2.8	[29]
BN	Admicellar-treated BN particles.	2.7	[54]
BN	30 wt% of BN particles modified by 3-aminopropyl triethoxysilane	1.178	[55]
BN	Hexagonal BN/epoxy composites at 44 vol% (densely packed and vertically aligned).	9	[56]
BN	Hexagonal, cubic, and conglomerated -BN.	2.91, 3.95, and 10.1	[57]
BN	Hexagonal boron nitride laminates	20	[58]
BN	Untreated and OTAB-treated BN/epoxy composites.	1.9 and 3.4	[59]
BN	88 wt% of BN loading.	32.5	[60]
AlN	58.4 vol% of large-sized Aluminum nitride (AlN) with small-sized Al_2O_3	2.842 and 3.4	[61]
AlN	29 wt% of MWCNTs/AlN	1.04	[62]
AlN	20 vol% AlN particles (magnetically aligned)	1.8	[63]
AlN	50 wt% of 5 μm -AlN particles and 6 wt% of GO	2.77	[45]
AlN	67 vol% of AlN particles (large-sized silane-coated).	14	[64]
AlN	Cycloaliphatic epoxy/trimethacrylate system	0.47	[65]
AlN	At 47 vol% nano-whiskers AlN	4.2	[66]
Al_2O_3	At 80 wt% of Alumina (Al_2O_3)/epoxy, filled with 5 wt% of graphene oxide (GO) and 5 wt% of $\text{Al}(\text{OH})_3$ -coated GO	3.5 and 3.1	[67]
Al_2O_3	Al_2O_3 /GFRP (amino group grafted)	1.07	[68]
Al_2O_3	At 60 vol% of micron-sized alumina	4.3	[69]
SiC	Magnetically aligned BN and Silicon Carbide (SiC) filler system	5.77	[50]
SiC	Nano-sized SiC particles with triethylenetetramine (TETA) functionalized MWCNTs, (at 30% vol%)	2.00	[70]
SiC	At 20 vol% of SiC particles (magnetically aligned Fe_3O_4 coated)	1.681	[71]

2.2. Metallic Fillers

Metal nanoparticles (NPs) have recently gained popularity due to their unique properties such as low melting temperature and high diffusion coefficient [72]. Research has shown that epoxy conductive adhesives enhance their heat conductivity when integrated with metal fillers. Sphere, fibre, granules, or flakes may be metallic particles. The optimal shape can be such that the filler levels of the surrounding metallic parts are the lowest crucial, well interacted, and the best matrix-resin adhesion can be found. These criteria are suitable for metallic flakes because of their high aspect ratio [73]. However, polymers loaded with metal are required with the required reinforcement. The amount of filler may be readily changed to adjust thermal conductivity. The material for heat sinking requires a low coefficient of thermal expansion (CTE) since the

semiconductor chips have low CTE content. Thus, it has good thermal conductivity, and low CTE needed for thermally conductive material. Cu is usually not utilized in TCAs. Because Cu is thermally good yet has a high CTE content, **Figure 6** shows some metal fillers used to manufacture TCAs.

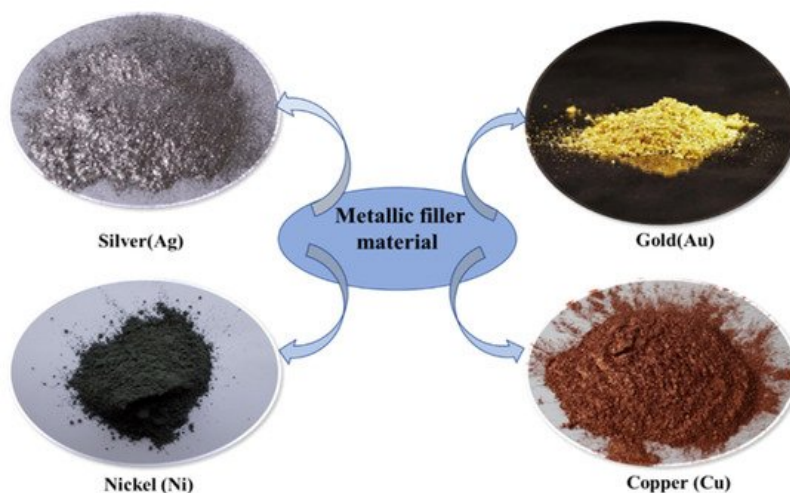


Figure 6. Commonly used thermally conductive metallic filler.

Copper nanoparticle paste is used for bonding chips and different metallic substrates at low temperatures. The result shows that good bonds are mainly achieved with Cu and Au surfaces [74]. It provided good thermal conductivity and shear strength (20 MPa) when sintered at 350 °C [75]. The thermal conductivity of the composite was three times higher than pure epoxy at 15 wt% of MWCNT, and 34 wt% of copper nanoparticles [76], TiO₂ coated copper nanowires 0.2–1.12 Wm⁻¹ K⁻¹ [77] and 2.59 Wm⁻¹ K⁻¹ [78] were found for a length of up to ~40 µm and diameter of ~20 nm.

Silver (Ag) particles exhibit maximum thermal conductivity among functional metallic fillers. Silver nanoparticles have a vast application in catalysis, conduction, antibacterial and electrical devices [79]. Silver nanoparticles have different forms and sizes to obtain a high heat conductivity and decrease total product costs. Silver's thermal conductivity is 450 Wm⁻¹ K⁻¹, although its conductivity improvement with an epoxy adhesive is lower. Silver/epoxy self-constructed nano-structured networks led to two times better thermal conductivity than plain epoxy [80]. Ag particle at 25.1 vol% of Ag-decorated BN nanosheets provides 3.06 Wm⁻¹ K⁻¹ [81] and silver-decorated MWCNT/epoxy adhesive 0.88 Wm⁻¹ K⁻¹ [82]. Ni decorated MWCNTs as a reinforcement provides 0.30 Wm⁻¹ K⁻¹ [83]. The best part of Ag nano particle is that it improves the thermal conductivity and the shear strength of the adhesive joint [84], which is essential for the electronic packaging reliability [85]. Additionally, some composites of silver-coated copper, silver-coated reduced, graphene oxide nanoparticles, and graphene nanosheets embedded in the epoxy resin provide good thermal and mechanical properties [86]. Another research observed the thermal conductivity of epoxy adhesive filled by eight different filler- ZnO powders, BN powders, Al₂O₃ powders, graphite flake, Al powders, Cu powders, diamond powders and Ag powder. The results indicate that each sample is capable of significantly increasing the thermal conductivity of the epoxy resin. The highest thermal conductivity (1.68 Wm⁻¹ K⁻¹) was obtained in the graphite-epoxy adhesive at 44.3 wt%. Meanwhile, the layer-shaped filler and sharp corner-shaped fillers are preferable for improving the thermal conductivity of epoxy resin [87]. The morphologies of the eight fillers samples after spray-gold treatment were observed by SEM images (shown in **Figure 7**).

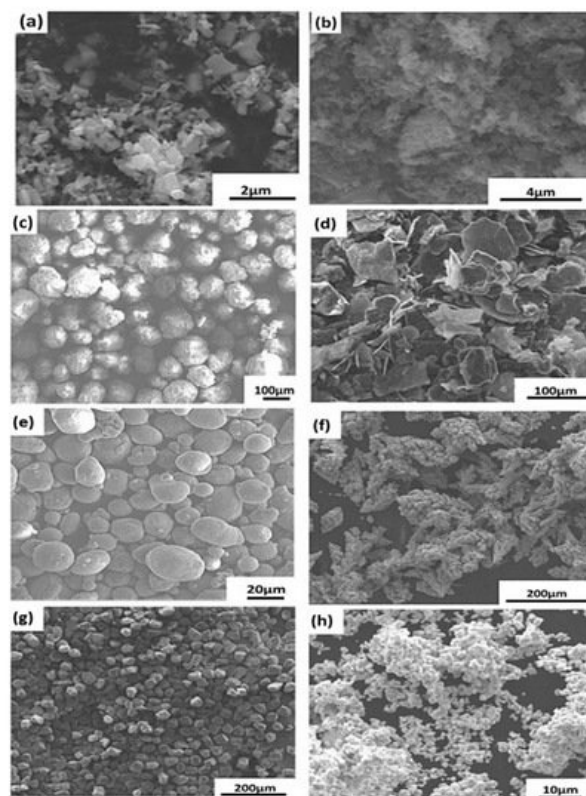


Figure 7. SEM images of fillers sample as powder form. (a) ZnO, (b) BN, (c) Al₂O₃, (d) graphite flake, (e) Al (f) Cu (g) diamond and, (h) Ag powders [87].

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