

## Review Article

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# Review of thermal conductivity of gold nanoparticle composites

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**Abstract:** Nanocomposites are defined as a combination of two or more components, wherein at least one of these components is on the nanoscale. Applications requiring increased thermal conductivity are demonstrating interest in nanocomposites based on conductive nanoparticles, such as gold nanoparticles (AuNPs), that are currently the subject of intense investigation due to their amazing thermal, electronic, and optical properties. Because of their remarkable stability, high conductivity, and high ability to create strong chemical interactions with groups, including organic ligands or polymers. In addition, adjustable production, simple surface modification, nontoxicity, low resistance, and strong light interaction. All these numerous advantages made AuNPs ideally suited for a variety of application areas, including environmental science, biomedicine, sensors, and nano-electronics. Various factors affect the properties of AuNPs, including particle size and shape, concentration, temperature, surface functionalization, covalent functionalization, and base fluid material are responsible for the variation of thermal conductivity. There is a main gap in a broad analysis, offering a cohesive view on the preparation of thesis materials, their properties, their characteristics, the highest challenges, and future perspectives. This review aims to fill this gap and summarizes the recent developments in the thermal properties of AuNPs of nanocomposites and the mechanisms by which AuNPs enhance thermal conductivity and the heat transfer properties of various composite matrices, such as graphene and polymers. In addition, it discusses how interfacial thermal resistance is reduced and highlights how

the size, shape, concentration, and distribution of the nanoparticles affect the overall thermal conductivity of the composites. Comparative evaluation of Au nanocomposite with other NPs in enhancing conductivity and challenges and future directions will be carried out as well.

**Keywords:** gold nanoparticles; gold nanoparticles-graphene composite; transient hot-wire; thermal conductivity

## Abbreviations

AuNPs	gold nanoparticles
MMT–AuNP	montmorillonite/gold nanoparticle
PA–AuNPs	polyamide/gold nanoparticle
PA/MMT/AuNP	polyamide/montmorillonite/gold nanoparticle
CPs	conducting polymers
PEDOT	polyethylenedioxythiophene
PPy	polypyrrole
Au NSs	gold nanostructures

## 1 Introduction

Nanoparticles with sizes less than 100 nm can be either inorganic or organic in nature.

As a result of their distinctive properties, nanostructured materials have attracted substantial scholarly interest. These properties include an exceptionally high surface area-to-volume ratio, pronounced surface reactivity, the elimination of contact resistance in heat transfer, and the capacity to enhance heat transfer during phase transitions, such as melting, through the promotion of natural convection mechanisms [1].

Heat transfer-involving fluids in both laminar and turbulent flow regimes, as well as in boiling conditions, whether in motion or stagnant, is a fundamental aspect of numerous industrial processes. These operations often span a broad spectrum of pressures and temperatures. Enhancing the thermal conductivity of heat transfer fluids, thereby reducing their thermal resistance, would offer significant advantages across various applications. Such improvements could enable the design of more compact heat exchange systems, leading to lower capital investment and enhanced energy efficiency.

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Nanofluids, providing enhanced heat transfer fluids that a wide range of industrial sectors could benefit from, could diminish such thermal resistances. Transportation, electronics, healthcare, food, and various forms of industry are among them [2,3].

The nanofluid is known as a colloidal suspension composed of metallic or nonmetallic nanoparticles (NPs) with a size smaller than 100 nm; also, nanofluid can be nanofibers dispersed uniformly within a conventional base fluid [4]. Nanofluids have recently attracted extensive attention as a result of their markedly enhanced thermal conductivity [5], which has been recognized as an essential requirement for new technologies, particularly in aerospace and aeronautical technology [6].

It has been demonstrated that dispersing less than a 1 % volume fraction of carbon nanotubes or copper nanoparticles (CuNPs) in oil or ethylene glycol enhances thermal conductivity by approximately 150 % and 40 %, respectively [5]. By increasing a material's thermal conductivity, heat can be dispersed more quickly, preventing significant overheating that could cause the material to degrade prematurely [6, 7].

Additional potential benefits of nanofluid testing include reduced pressure drop and erosion, especially in microchannels, and improved long-term dispersion stability and thermal conductivity in comparison to suspensions containing particles with micrometer or millimeter in size [5].

Furthermore, the development of energy-efficient heat transfer systems depends heavily on the thermal fluid's conductivity [8]. For instance, it has been found that nanofluids with copper nanoparticles (CuNPs) directly dispersed in ethylene glycol exhibit noticeably better thermal conductivity increases than either nanoparticles that contain fluids or nanofluids with metal oxide nanoparticles [8].

It is widely recognized that colloidal suspension nanoparticles can greatly improve thermal conductivities, even at low volumes significantly under 1 %, this improvement is going down in the case of those found in conventional heat transfer fluids [9].

As a result, to the naturally high thermal conductivities of metal nanoparticles (MNPs) and metal oxide nanoparticles (MONPs) compared to traditional fluids, the incorporation of MNPs or MONPs into essential fluids has seemed like a hopeful approach for improving fluid thermal conductivity. Utilizing particles that are nanometers in size and have a significant specific surface area may enhance heat transfer while simultaneously increasing particle stability [9].

It is possible to create nanofluids by suspending several kinds of nanoparticles, including metallic and nonmetallic NPs, in various fluids with differing sizes and shapes. Many investigations on the thermal characteristics of nanofluids demonstrate significantly increased thermal conductivity.

For instance, thermal conductivity enhancements of about 160 % and 70 % have been experimentally shown in nanofluids having 1 vol% multiwalled carbon nanotubes (MWCNTs), dispersed in oil and 1 vol% ultra-dispersed diamond in ethylene glycol, respectively [9]. In spite of these important improvements in thermal performance, the colloidal stability of such suspensions is still limited, even at enough nanoparticle concentrations [9]. In addition, the heat transfer capacity of base fluids such as oil, ethylene glycol, and water is poor as a result of their low thermal conductivity, causing limitations in heat transfer procedures [10, 11]. This limitation underscores the necessity for further systematic investigations into alternative base fluids and stabilization strategies to ensure both high thermal conductivity and long-term dispersion stability [9]. According to Abbas et al. [12], nanofluids are the best alternative to increase the thermal conductivity of conventional heat transfer fluids and showed that the properties of the fluid can be changed depending on the nanoparticle mixture, with the thermal conductivity of NPs being much higher than in the case of the base fluid. The importance of nanofluid has increased due to the convective heat transfer factor and improved thermal conductivity [12].

Moreover, because nanocomposites can display distinct properties, they can be used as a foundation for many different applications [13]. Recently, polymer nanocomposites incorporating conductive nanoparticles, such as carbon, gold, and silver, have emerged as promising materials for applications demanding boosted thermomechanical and electrical properties, including super-capacitors and structural power composites [14].

As a result, to their numerous advantages, including great thermo-mechanical performance, excellent chemical resistance, and low shrinkage, epoxy polymers are commonly employed as the matrix phase in a wide range of composite materials [15]. These advantageous properties render epoxy polymers indispensable components of structural composite materials, particularly in the aerospace and automotive industries. Epoxy resins currently dominate the thermoset polymer market, representing approximately 70 % of the total thermosetting polymers [16].

Nanoparticles exhibit stimulating electrical, optical, mechanical, and thermal properties that are utilized in improving multifunctional nanocomposites. Among various nanoparticles (NPs), gold nanoparticles are extensively utilized in a broad range of applications, including biomedical applications [17] and photonics [18–20].

The extensive deployment of AuNPs stems from their exceptional physicochemical attributes, like greater resistance to corrosion and high thermal and electrical conductivities. For example, distinct other MNPs such as CuNPs and

AgNPs, which are prone to oxidation when contacted with air, AuNPs show remarkably greater stability against oxidative degradation [21–23]. Likewise, some active functional groups can be attached to the surface of AuNPs due to the simplicity of functionalization of the AuNPs' surface. This advantage will open up new opportunities for multifunctional applications of AuNPs [24, 25]. The addition of AuNPs into the epoxy polymer is shown to enhance the glass transition temperature, possibly for the reason that of a reduction in the chain flexibility and mobility of the polymer matrix [26].

There are limited studies that have studied the effects of AuNPs on the thermal enhancement of AuNP-composites. This review will focus on the relationship between sizes, shapes, and concentration of AuNPs on the thermal conductivity of AuNPs-composite.

## 1.1 Nanocomposites

Polymers generally exhibit low thermal conductivity, primarily due to their inherently weak chemical interactions, limited bonding strength, and disordered crystal structures, high molecular vibrations, and comparatively low atomic density [27]. It has been confirmed that the attachment of polymer chains to the surfaces of AuNPs of varying sizes enables the investigation of the effects associated with chain flexibility when the chain ends are not constrained [28].

Polymers are extensively applied in electronic packaging, adhesives, and thermal conductors thermally due to their worthy insulation, good chemical stability, and abundant industrial production. While polymers offer many advantageous properties, their low thermal conductivity usually ranges between  $0.1$  and  $0.5 \text{ Wm}^{-1}\text{K}^{-1}$ , which remains a significant obstacle to their broader application across various fields [29]. Enhancing the polymer composites with good thermal conductivity continues to be a challenge and focuses on contemporary materials research. Currently, polymer materials can be produced to be more thermally conductive in two approaches.

One approach to enhancing the thermal conductivity of polymers is to modify the composition and arrange the polymer chains to raise their intrinsic thermal conductivity. This can be achieved, for example, by creating polymers with extensive conjugated  $\pi$ -bond structures and promoting electronic thermal conduction. The second and more broadly adopted method involves the synthesis of polymer composites by combining the fillers with high thermal conductivity. For example, boron nitride, carbon fibers, alumina, and silver nanoparticles are incorporated into the polymer matrix in this method. In the past decades, research

has concentrated on developing the thermal conductivity of composites and polymers. While the field has presented major developments in recent years, various unresolved issues remain that need to be overcome for expected industrial uses [29].

Most studies are focused on devolving polymers' thermal conductivity by adding thermally conductive fillers. These fillers are classified according to their geometry: one-dimensional (1D), which includes families of metal, ceramic, or carbon-based material [30]; two-dimensional (2D) [31]; and three-dimensional (3D) materials [32]. Lately, metallic fillers like gold and copper nanoparticles have been added to carbon nanotubes (CNTs), graphene, and graphite as carbon-based fillers, displaying high electrical conductivity. These materials are typically used in applications demanding remarkable thermal and electrical conductivity. Demand both high thermal and high electrical conductivity [33]. In addition, compared to traditional heat shield materials, the increase in thermal conductivity might rise as much as 500 %. Composite materials that are flexible and lighter cause a decline in weight and improve the maneuverability [34].

Types of gold nanoparticle composites with thermal conductivity are broadly classified according to the matrix materials, structural configurations, and functionalization methods used. Below are some of the principal categories and examples:

### 1.1.1 Gold-polymer nanocomposite

The integration of polymer-linked nanoparticle networks is highly promising and has become a focal point of research for diverse applications, including phononics, thermoelectric devices, molecular electronics, and autonomous computing materials. According to the literature, incorporating AuNPs into polymer matrices such as polyethylene, polystyrene, or epoxy can enhance thermal conductivity [28].

The combination of polymers and gold nanoparticles encompasses many application areas, including electron storage, energy, and catalysis. This combination leads to several property transformations, impacted by the shape, size, and size distribution of the NPs. For instance, increasing the particle size leads to a peak in the absorption at higher wavelengths [35, 36]. The ability and tendency of inorganic nanoparticles to aggregate is mainly because of their high surface energies, normally between  $500 \text{ mJ/m}^2$  and  $2,000 \text{ mJ/m}^2$ . Polymers have much lower surface energies, which range from  $20$  to  $50 \text{ mJ/m}^2$ . When MNPs are combined with the softening properties of a heated polymer, this property causes the particles to become immersed in the polymer matrix, consequently reducing their surface energies; this phenomenon is known as the embedding [36]. Metal colloids stabilized

polymers are typically prepared from appropriate metal sources, where the size and size distribution of the produced nanoparticles are highly sensitive to the conditions of reaction, with the concentration of the protective polymer playing a particularly crucial role [37]. For instance, both polyethylene oxide (PEO) and polyvinyl alcohol (PVA), as hydrophilic polymers, are used in the green approach to silver nanoparticles (AgNPs) because of their mechanical performance, excellent biocompatibility, and low toxicity. Where polymer-based NPs act as scaffolds for coordinating silver ions ( $\text{Ag}^+$ ) and as electron donors to enable reduction of  $\text{Ag}^+$  to ( $\text{Ag}^0$ ). The functional groups of the polymer, such as phenols, amines, and carbonyls, play a remarkable part in determining the speed of the reduction process (reduction kinetics) and the stability of the produced colloidal nanoparticles [38].

Additionally, the polymer chains attached to the AuNPs' surfaces not only improved the gold cores' stability but also introduced additional functional properties. These properties result from the characteristics of the outer polymer layers. In particular, the smart nanocomposites that include AuNPs and advanced polymers display remarkable features through their effects in a straightforward manner [39]. According to the latest study, spherical gold/poly(methyl methacrylate) (gold/PMMA) hybrid nanocomposites were successfully synthesized. They showed that the sizes and dispersions of the AuNPs are adjusted by controlling the concentration of the polymer. Furthermore, gold/PMMA nanoball composites are produced in the presence of  $\text{NaBH}_4$  at polymer concentrations of  $35 \mu\text{M}$ . However, without using the polymer, significant macroscopic precipitation of gold particles occurs, confirming the critical role of PMMA in particle stabilization [37]. Likewise, polymers containing primary, secondary, or tertiary amine groups are among the most common types of polymers. Recent research has shown that a key requirement for the formation of gold nanoparticles with a specific structure is the presence of electron-donating groups in the polymer, which ultimately leads to spontaneous reactions. From a thermodynamic perspective, this is possible when the oxidation potential of these groups falls within the range of the oxidation potential of gold from Au(0) to Au(I) and the reduction potential from Au(III) to Au(0), as is the case for low-molecular-weight amines [40].

It has been observed that the type of combining polymer and the AuNPs can affect the heat conductance inside dimers, thereby influencing the conductivity [28]. Numerous experimental investigations have been conducted on the fabrication of polymer–AuNP systems [39, 41–43]. Nevertheless, only a few studies have focused on the synthesis, dimensional control, and spatial arrangement of inorganic/organic hybrid materials [37].

The dielectric properties of the AuNP–epoxy composite were experimentally characterized by Fraga et al. who observed that both ionic conductivity and capacitance improved with increasing concentrations of AuNPs [43].

According to Zhang et al. [44], conductive polymer nanocomposite embedded with AuNPs was successfully formed via a one-step synthesis technique at R.T. In this technique, the hydrogen tetrachloroaurate ( $\text{HAuCl}_4$ ) was used to prepare AuNPs as precursor salt in a poly(3,4-ethylenedioxythiophene)polystyrene sulfonate (PEDOT:PSS) medium (see Figure 1). The stabilization of the AuNPs/PEDOT:PSS nanocomposites is done via electrostatic interactions between the PEDOT:PSS polymer chains and the surfaces of AuNPs [44].

Furthermore, AuNPs synthesized at the lowest concentration of  $\text{HAuCl}_4$  ( $2 \mu\text{M}$ ), exhibit relatively uniform sizes and predominantly spherical shapes (see Figure 1a–d), with diameters ranging from approximately 2–7 nm and an average diameter of 4.1 nm. Whereas, at a higher concentration ( $20 \mu\text{M}$ ) (see Figure 1b–e), the NPs remain largely spherical, with diameters between ~5 and 11 nm and an average size of 7.5 nm. At an even higher concentration ( $200 \mu\text{M}$ ), while a significant portion of the AuNPs retains a spherical morphology, a variety of other shapes, including pentagonal, triangular, hexagonal, and rod, begin to arise (see Figure 1c,f).

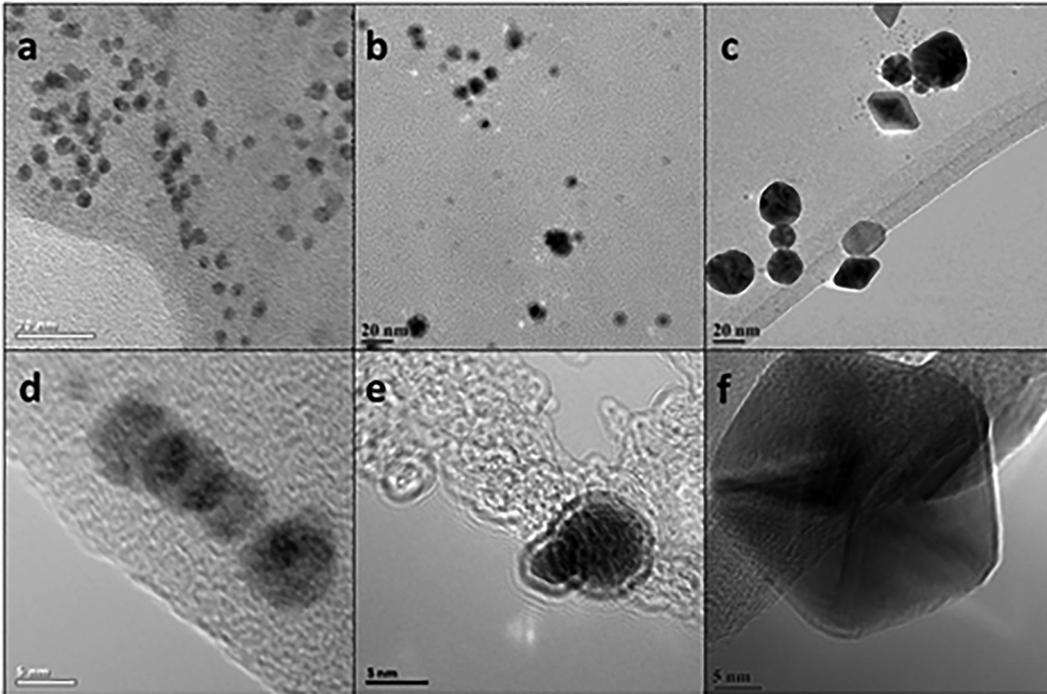
From Figure 2, it can be seen that larger AuNPs were generally obtained by increasing the concentration of  $\text{HAuCl}_4$  or plasma treatment time. Moreover, smaller and sparsely distributed AuNPs were effectively embedded within a continuous polymer matrix, displaying good dispersion [44].

### 1.1.2 Gold–graphene nanocomposite

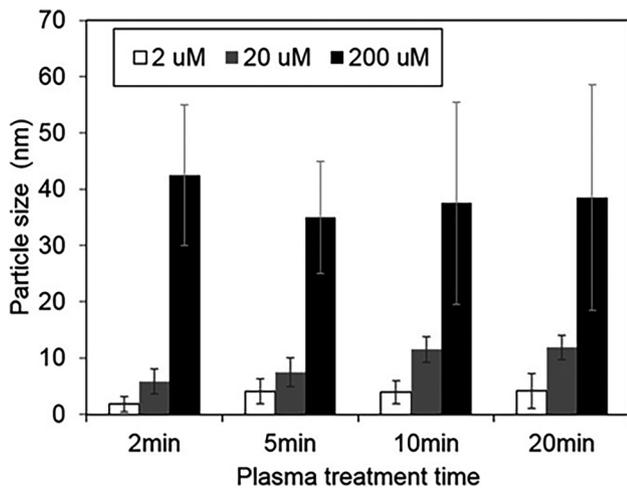
Graphene nanoparticles exhibit superior mechanical strength, electrical, and thermal conductivity. The advantageous thermophysical properties of graphene make it excellent for applications in nanofluids. Furthermore, synthesizing graphene nanoparticles is relatively easy and cost-effective [45].

Combining the distinctive conductivity advantages of graphene with the high reactivity of metal nanoparticles (MNPs), such as AuNPs, enables the manipulation of key phenomena, including the spatial confinement of surface plasmon resonance on MNPs, the development of graphene's light absorption capabilities, and the bio-functionalization with various molecules for biosensor fabrication, as well as, the improving of nanoparticles' catalytic activity [46].

In addition, the combination of graphene-based materials with AuNPs has yielded a synergistic effect that remarkably enhances sensor sensitivity. This effect stems



**Figure 1:** Transmission electron microscopy (TEM) pictures of AuNP/PEDOT:PSS nanocomposites prepared from varying concentrations of the gold salt precursor (a and d) exhibit 2–7 nm sizes and spherical shapes of AuNPs when synthesized at low concentration (2 μM of HAuCl<sub>4</sub>); while (b and e) showed spherical sizes of 5–11 nm at a high concentration (20 μM). Images (c and f) showed sizes of approximately 35 nm with different shapes at the highest concentration (200 μM), including pentagonal, triangular, hexagonal, and rod shapes beginning arise [44].



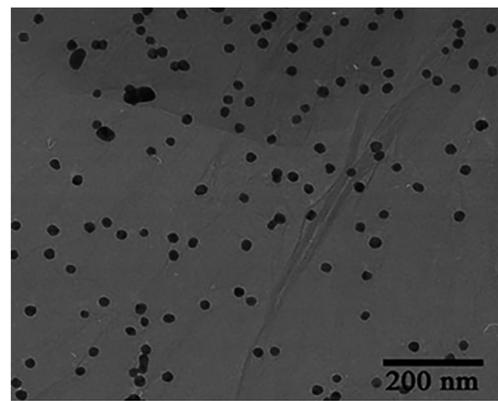
**Figure 2:** Mean AuNP particle size obtained from different precursor concentrations and plasma processing time [44].

from the complementary functions of these materials. AuNPs provide active catalytic sites, while graphene increases electron transfer kinetics and provides a supporting matrix for the dispersion of the nanoparticles [47].

Gold nanoparticles attached to graphene sheets have the potential to generate hybrid materials with distinctive photothermal properties that are advantageous for applications in sensing, catalysis, smart polymer composites, and

optoelectronic devices. It has been demonstrated that a straightforward, reagent-free method at room temperature can produce a few layers of graphene nanoplatelets (GNPs) coated with colloiddally stable gold nanoparticles [48].

Furthermore, AuNPs were efficiently synthesized on functionalized graphene sheet surfaces through a straightforward chemical approach conducted using graphene oxide in an aqueous solution. The homogeneous distribution of AuNPs–graphene composite exhibited spherical particles with sizes of  $21.3 \pm 1.8$  nm, as illustrated in Figure 3, based on



**Figure 3:** TEM image of graphene/gold nanocomposite synthesized from graphene oxide [49].

the literature [49]. In addition, the graphene sheets exhibit a thin structure and smooth surface, with slight corrugations observed, indicating their flexible nature [49].

According to the literature, graphene/AuNP hybrids were produced by noncovalent assembly, or *in situ* reduction of  $\text{HAuCl}_4$  on graphene oxide (GO). High and consistent AuNP coverage on graphene was confirmed by transmission electron microscopy (TEM), UV–vis spectroscopy, and powder X-ray diffraction (XRD). However, the effectiveness of these techniques is limited by the inability to control the size of AuNPs and by the weak interactions between both graphene and AuNPs [50].

Moreover, the synthesis of (AuNPs/graphene nanoplatelets) composite based on plasmonic AuNPs and graphene nanoplatelets, in the absence of reducing agents, is conducted simply under ambient conditions. Au acetate [ $\text{Au}(\text{C}_2\text{H}_3\text{O}_2)_3$ ] is directly reduced to create these hybrid architectures, in which AuNPs were homogeneously coated on the surface of graphene nanoplatelets, and their dimension is controlled by adjusting the Au acetate concentration [48].

According to the scanning electron microscope (SEM) images (see Figure 4a), AuNPs were homogeneously distributed on the graphene nanoparticle surface (GNPs). This may suggest that Au acetate, a reduction, usually involves the addition of a reducing agent or exposure to high temperatures. Research employing high-resolution transmission electron microscopy (HR-TEM) provided further evidence that AuNPs were successfully produced on the surface of GNPs. AuNPs are evenly dispersed throughout the surface of graphene nanoparticles, as seen in Figure 4b, and display clearly defined lattice fringes [48].

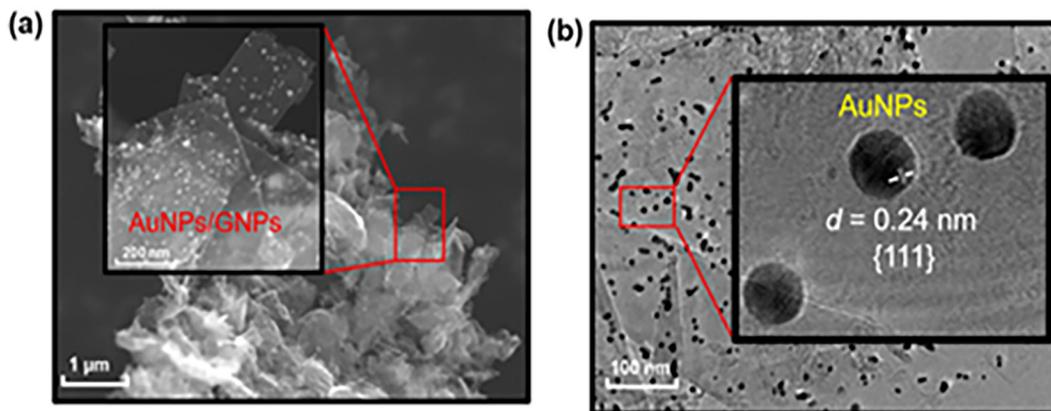
Furthermore, the formation and growth of gold nanoparticles on functionalized graphene are controlled by the oxygen functionalization present on the graphene surface. Where

AuNPs are not observed on completely reduced graphene surfaces, as the lack of oxygen groups prevents their formation. In contrast, the presence of ligand functions ligands containing oxygen atoms on graphene oxide significantly promotes the nucleation and growth of AuNPs. The resulting graphene/gold nanocomposite is considered a resource for surface-enhanced Raman scattering (SERS), where the AuNPs enhance the Raman signals of molecules near active nanostructures [49].

An earlier study indicated that the thermal conductivity of graphene nanosheets–AuNPs/ethylene glycol (GNs–AuNPs/EG) hybrid nanofluid was notably higher, reaching 0.41 W/mK at a temperature range of 25–45 °C. This value exceeds that of GNs/EG (0.35 W/mK), AuNPs/EG (0.39 W/mK), and (0.33 W/mK) of the EG base fluid. The GNs–AuNPs/EG nanofluid composite showed a remarkable improvement (about 26 %) in thermal conductivity. This is because of the synergistic impact between the graphene layer sheets and AuNPs, where both have inherently great thermal conductivity. Consequently, GNs–AuNPs/EG nanofluid nanocomposite shows great promise for improving heat transfer applications [51].

Compared to other types of nanomaterials, boron nitride nanosheets (BNNS), often referred to as white graphene, have quite a similar structure to graphene and exhibit outstanding thermal conductivity [52]. Research by Hou X et al. demonstrates that the BNNS nanofluids exhibit superior heat transfer properties compared to water, indicating that the fabricated BNNS nanofluids possess exceptional thermal conductivity and heat transfer capabilities [52]. Nonetheless, the improvement of nanofluids' thermal conductivity relies on multiple issues, such as volume fraction, temperature, particle size, pH values, concentration, viscosity, and more [52].

Likewise, graphene-based nanofluids and carbon nanotubes (CNT) emerge as the best improved heat transfer fluids, in line with their low mass density and high thermal



**Figure 4:** Visualization and characterization of AuNPs/GNPs composite at very high magnifications and resolutions. (a) SEM images of AuNPs/GNPs composite; (b) TEM images of AuNPs with insets showing high magnification images and specific details such as d-spacing [48].

conductivity in comparison to other NPs, such as copper, aluminum oxide ( $\text{Al}_2\text{O}_3$ ), and titanium dioxide ( $\text{TiO}_2$ ) [51]. The use of carbon nanotubes (CNTs) and graphene (GR) to enhance the interfacial properties in fiber-reinforced composites has been studied. These nanomaterials can increase the effective thermal conductivity by forming three-dimensional heat transfer networks within the matrix. However, surface modification of CNTs or graphene, which is necessary to improve their compatibility with the resin, may alter their original structural properties, thus reducing their thermal conductivity. Compared to using CNTs or graphene separately, CNTs have a limited surface area, subsequent in poor thermal conductivity. Graphene, on the other hand, tends to agglomerate, hindering its distribution and reducing its thermal conductivity. However, combining carbon nanotubes and graphene (CNTs–GR) can create a multidirectional network for heat conduction, enhancing thermal conductivity and improving the mechanical characteristics of the composite material [53].

Similarly, it has been illustrated that the thermal conductivity of both carbon nanotubes and graphene can be developed via the functionalization of their surfaces using MNPs. Torres-Mendieta et al. established an operative method for creating graphene–metal composites utilizing a femtosecond radiation procedure, where they attached AuNPs to the surfaces of GO sheets in deionized water, using laser radiation directed at an Au disk immersed in a GO suspension [46].

In the same way, Fu et al. created nanocomposites from GO and AuNPs to produce solar steam when exposed to sunlight. Study indicated that the effectiveness of solar vapor generation with graphene oxide–gold nanofluids improved by 10.8 % with just 15.6 wt% of AuNPs, confirming an increase in steam generation efficiency [54].

Yarmand et al. prepared graphene nanoplatelets by decorating them with AgNPs in deionized  $\text{H}_2\text{O}$  using a chemical synthesis method [45]. The experimental results from this nanofluid indicated enhancements in heat transfer efficiency and thermal conductivity in comparison to other related base fluids. As well as the extent of enhancement depended on both temperature and NPs' weight concentration [45].

## 1.2 Thermal Stability of Nanocomposite

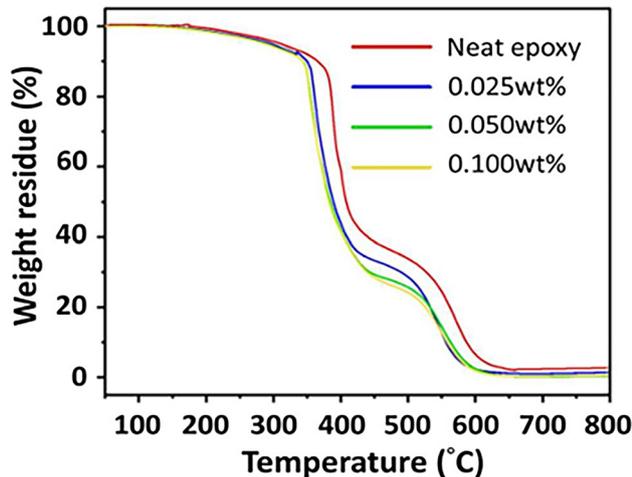
The findings from some studies showed that the combined effect of nanoclay and AuNPs loaded in the polyamide (PA) matrix. The char yield of pure polyamide was determined to be less than 20 %. However, as nanofiller loading increased, char yield from nanocomposites increased. With an increase in montmorillonite/gold nanoparticle (MMT–AuNP) nanofiller

loading ranging from 0.1 to 0.5 wt%, the char yield is also increased from 37 to 60 %. The nanocomposite containing 0.5 wt% loading of PA/MMT/AuNP 0.5 exhibited a more significant enhancement in thermal properties, indicating improved thermal stability. These findings clearly exhibited that the thermal stability and flame-retardant properties of processing-friendly aromatic polyamide are improved after the addition of AuNPs and MMT [55].

Enhanced interfacial interactions and improved interfacial thermal transport (ITT) between carbon nanotubes (CNTs) are critical factors in optimizing the performance of CNT-based thermo-functional instruments and materials, including thermal interface materials and energy transfer media. In this regard, the integration of AuNPs onto CNT fibers offers a compelling strategy, providing a multifunctional platform with significant potential for advancing thermal management technologies [56]. For example, incorporating CNT into silicon chips is found to improve the heat dissipation and, therefore, increase the effectiveness of nano instruments. Nonetheless, a bottleneck persists in harnessing the excellent electrical and thermal characteristics of CNTs for applied applications. Electrical conductivity was improved when Au and Pt nanoparticles were incorporated, demonstrating a clear advantage for electron transport. Conversely, the contribution of AuNPs to improving thermal transport did not achieve a definitive consensus. Traditionally, gold nanoparticles are considered to serve as phonon scattering centers when situated at the surfaces or interfaces between different materials, thereby significantly impeding thermal transport. A notable example is the reduced interfacial thermal transport (ITT) observed in Au/ $\text{SiO}_2$  structures, where the crystalline–amorphous interface disrupts interactions between phonons and phonons [57]. In a different instance, various layered graphene nanoplatelets experience a decrease in phonon transport when adorned with AuNPs, because of the disparity in the phonon vibrational density of states (VDOS) between graphene and AuNPs [56].

A new composite nanostructure, AuNPs–CNTs, was developed, achieving experimentally a remarkable increase in thermal transport of up to 70 % in CNT bundle-like fibers.

Figure 5 shows the thermal degradation behavior of the cured neat epoxy and the AuNP/epoxy composite, which was evaluated over a temperature range of 50–800 °C. The incorporation of AuNPs considerably reduced the thermal stability in comparison to the cured neat epoxy. This decline in stability is likely due to the enhanced thermal conduction introduced by the presence of AuNPs within the composite matrix. Numerous groups have documented comparable findings in the literature [26]. For example, Gorghiu and colleagues [58] proposed that metals might accelerate the thermal degradation procedure through interactions between metal particles and polymers on their surface, leading



**Figure 5:** Thermogravimetric analysis (TGA) results for both the cured unmodified epoxy and the epoxy composites incorporated with gold nanoparticles [26].

to an earlier initiation of thermal decomposition in the composites [58].

Indeed, in certain aerospace applications, metallic materials are progressively being substituted with high-performance polymers and composites [6]. However, there are some challenges in expressively enhancing the thermal conductivity of these materials to meet the demands of such applications. This necessity has prompted researchers to explore and reevaluate the mechanisms governing thermal conductivity in nanocomposites, aiming to identify effective strategies for improvement [6].

### 1.2.1 Thermal conductivity in nanocomposites

To enhance the thermal conductivity of nonconductive compounds like polymers, researchers have concentrated on adding various fillers or modifying the structural characteristics of the material. Additionally, the degree of enhancement in thermal conductivity is impacted by several factors, including the filler kinds, filler characteristics, and composition. Various types of thermally conductive nanofillers (crystalline) have been investigated in recent years, yielding comparable results, including carbon-based fillers, graphite, CNT, graphene, and metallic fillers (such as Ag, Al<sub>2</sub>O<sub>3</sub>, Cu, TiO<sub>2</sub>) [6]. Several studies demonstrated similar rates of improvement when employing different combinations of fillers [59–61], aiming to achieve a synergistic effect. Han et al. [62] provided a summary of how the thermal conductivity of CNT nanocomposites is enhanced. Following initial pristine experimental findings regarding the distribution of pure carbon nanotubes within a polymeric matrix, major enhancements are noted in electrical and mechanical properties [6]; however, the consequences regarding

thermal conductivity were lower than expected. Recently, with modified 2D thermally conductive fillers like graphene [63–65], the thermal conductivity of composites can achieve levels comparable to metals; however, the processing methods vary for each individual case and may influence the overall thermal conductivity [6].

It is mentioned that composites containing carbon nanotube materials are attractive; the reason is related to their excellent conductivity and strength. After CNTs are incorporated into composite materials, they lead to improve their mechanical performance and exceed the boundaries of what is possible with other conventional composites. The sturdy van der Waals forces between separate tubes of CNTs lead to clusters, which hinder their regular diffusion within a matrix, giving CNs exceptional features. Further, the static nature of the CNT surfaces complicates their interaction with other matrices, limiting mechanical load transfer. Other bigger challenges are their contamination and low affinity toward metals [66].

Ming et al. [67] have revealed that the thermal conductivity of graphite-embedded metal–organic frameworks (MOFs) was two to four times higher in directions perpendicular to the stress axis. Meanwhile, Chen et al. [68] highlighted the influence of processing approaches on thermal conductivity, emphasizing the role of anisotropic composite structures. They showed that the processing technique affects the distribution of fillers within the matrix, which can seriously affect the thermal conductivity of the final product [6].

### 1.2.2 Thermal conductivity of gold nanoparticle composites

AuNPs possess outstanding characteristics, including good biocompatibility, exceptional conductivity, high density, a large surface-to-volume ratio, as well as notable magnetic and catalytic behaviors, distinguishing them from their bulk form [69].

Recently, significant studies have been devoted to the development of graphene-based nanocomposites, including graphene/polymer, graphene/metal nanoparticles, and graphene/ceramic systems. These hybrid materials demonstrate exceptional mechanical, electrical, magnetic, thermal, catalytic, and optical properties, which can never be achieved individually from separate components [70].

For example, sodium acetate trihydrate combined with modified graphene oxide aerogel achieved an outstanding solar thermal conversion efficiency of 86.3% due to its outstanding light absorption characteristics. Graphene nanoplatelets with higher light absorption capability provided Au/TiO<sub>2</sub>@n-octadecane phase change composites (PCCs), which showed a notable photo-thermal conversion efficiency of 97%.

In this system, graphene served as both a supporting matrix and a light-absorbing material. While oxygen-deficient titanium dioxide acted as an enhancer of thermal and optical performance, resulting in a photothermal conversion efficacy of up to 89.9% for the PCCs. Individual fillers frequently experience insufficient photo-thermal stability and a restricted range of light absorption, which presents obstacles for satisfying various application requirements. Consequently, creating synergistic composite approaches that incorporate various materials is anticipated to address the shortcomings of individual materials and enhance the photo-thermal conversion efficiency [71]. For instance, copper foam integrated with graphene aerogel has improved shape stability, thermal conductivity, and light absorption properties. However, these approaches face challenges such as inefficient material distribution and incompatibility between layers, which lead to poor stability of the composite materials under heat and light exposure, and reduced energy conversion efficiency, as illustrated in the literature [71].

Studies have shown that the conductivity of AuNPs aggregates in solution was increased as much as increase upon exposure to light or heat and reverts to its original state once the stimulus is removed. The extent of this conductivity change is directly influenced by the intensity of the applied light or heat. This reversible behavior is tentatively attributed to the dissociation of ions from the surfaces of NPs triggered by thermal or photonic stimulation [72]. Hybrid nanofillers with enhanced photothermal capabilities are fabricated by loading gold nanorods (AuNRs) onto graphene nanoparticles. The *in situ* growth technique enables tight bonding and uniform distribution of nanomaterials compared to the conventional physical mixing, significantly improving photothermal performance and dispersion stability [71].

In addition, it is discovered that gold–water nanofluids, at a concentration of 0.026 vol%, exhibit an enhancement in thermal conductivity ranging from 5% to 21% within the temperature range of 30 °C–60 °C [1].

### 1.3 Factors influencing thermal conductivity in AuNP composites

The thermal conductivity (TC) of a material plays a critical role in determining its melting and solidification behavior, often referred to as the charging and discharging degree. Notable enhancements in mechanical properties and thermal responsiveness have been detected in poly(N-isopropyl amide) hydrogels incorporated with dispersed gold nanoparticles [1].

Moreover, the final properties of nanocomposites are strongly influenced by the characteristics of the nanoparticles, such as their chemical composition, shape, size, and

volume, as well as their stability within the polymer matrix. Based on the literature results, various metal NPs, such as CuNPs, AgNPs, or AuNPs, are incorporated into an epoxy system, and their stability was examined through the phase transition, from the aqueous to the organic phase. The epoxy curing process was analyzed by UV–vis spectroscopy and TEM as well. The findings indicated that the development of a crosslinked epoxy structure created a polymer network that effectively confined the nanoparticles, reducing their tendency to agglomerate and thereby inhibiting further particle growth [26].

#### 1.3.1 Effect of the size of AuNPs-composite on thermal conductivity

The investigation of the effects of shape and size on nanomaterials has garnered significant consideration as results to their impact on both scientific research and industrial applications. Most of the properties associated with NPs have been attributed to their size of the NPs. It is clearly shown that nanoparticle size with a range of 10–20 nm has a direct influence on the thermal and electrical conductivity, as well as the stability of the polymer phase and its behavior. When the size of NPs decreases, the magnitude of cohesive forces between nanoparticles reduces in a hyperbolic manner. However, an exception to this movement is observed in systems where silver nanoparticles (AgNPs) are incorporated into a nickel-containing bulk matrix, wherein an inverse relationship is reported. This occurs because of the strong attractive force between silver and nickel, in contrast to AgNPs' low surface energy. Furthermore, the size of the nanoparticle has a direct effect on the surface and interfacial characteristics of NPs. NPs with small diameters generally exhibit enhanced stability due to their increased surface energy and stronger interaction with the surrounding matrix, while the tendency to agglomerate can be caused by steric, electrostatic, and van der Waals forces. Consequently, achieving homogeneous dispersed nanoparticles is essential for optimizing the overall performance and required properties of the composite material [1].

Chon et al. [73] utilized the transient hot wire technique to evaluate the thermal conductivity of aluminum oxide nanofluids. Their findings revealed that thermal conductivity decreases with increasing nanoparticle size. Notably, smaller nanoparticles led to an enhancement in thermal conductivity of approximately 28% under conditions of maximum temperature and concentration. Another study investigated using the steady-state technique to define whether the nanoparticle size significantly affects the thermal conductivity of Al<sub>2</sub>O<sub>3</sub>/H<sub>2</sub>O nanofluid. Findings indicated that the increase in size of NPs from 36 nm to 47 nm

decreases the enhancement in conductivity by 2% at a constant temperature [73–75].

The increase in the percentage of thermal conductivity has shown a strong correlation with the grain size of MNPs. Typically, a reduced size results in the highest thermal conductivity ratio. For instance, copper nanoparticles have main particle sizes that vary from 50 nm to 100 nm with square and spherical shapes. Thermal conductivity is improved by up to 40% for a nanofluid consisting of 0.3 vol% of CuNPs with a size smaller than 10 nm. The CuNPs exhibiting the greatest development in thermal conductivity are produced using a high concentration of copper acetate  $[\text{Cu}(\text{CH}_3\text{COO})_2]$  as a precursor with hydrazine hydrate as a reducing agent [8].

Similarly, based on their shape and size, AuNPs possess the ability to scatter light across a broad spectrum of the visible and near-infrared (NIR) regions. Photothermal effects arise when surface electrons on AuNPs interact with light of a suitable wavelength, triggering localized heating through plasmonic excitation, which leads to electronic transitions on the NP surfaces being converted into heat, subsequently increasing the particles' temperature [70].

The aqueous dispersed AuNPs with spherical shapes and a variety of sizes (from 2 nm to 45 nm) are synthesized using triethyleneglycolmono-11-mercaptoundecylether (EGMUDE) as a stabilizing ligand. The resulting exhibited the maximum improvement in thermal conductivity (1.4%) at a size diameter of 40 nm and concentration of 0.11 vol%. However, no thermal conductivity enhancement was detected for 45 nm dispersed AuNPs stabilized by silica at 0.4 vol% [9] (see Table 1). In addition, some studies illustrate that with the increase in the size of nanofluids, aggregation was increased and leads to an enhancement of the nanofluids' thermal conductivity, while other studies presented the opposite that the thermal conductivity of nanofluids was increased as well as the size decrease [76]. Researchers recently conducted a study on the dependence of the thermal conductivity of nanofluids on the nanoparticle material. They argued and demonstrated that the thermal conductivity of nanofluids is directly related to the density of the nanoparticle material, although they did not emphasize the thermal conductivity of the NPs [76].

**Table 1:** Reported thermal conductivity of gold colloids based on their size at a variety of temperature from 25 °C to 40 °C [9].

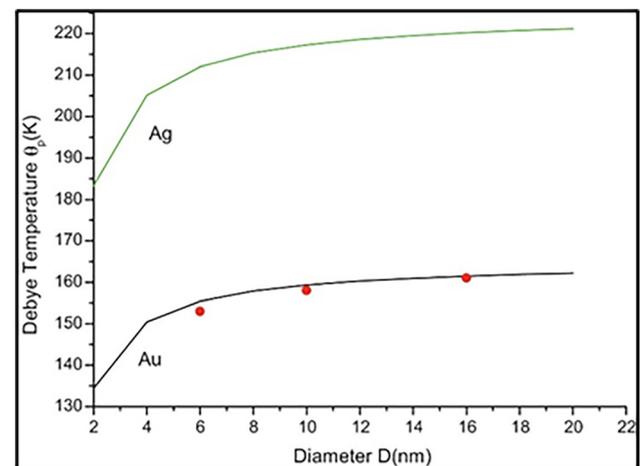
Particle size	Relative thermal conductivity (W/m K)
2 nm	0.952–1.01 (39 °C)
4 nm	0.997 (37 °C)
17 nm	0.972–1.015 (25–40 °C)
40 nm	1.006–1.014 (37 °C)
45 nm	1.002 (45 °C)

It is noted that the thermal conductivity declined by reducing polyethylene chain length in PE–AuNPs nanocomposite, and there was no effect shown by the size of AuNPs. The thermal conductivity of PE rises markedly during stretching. It has been confirmed that free PE chains with their related low  $k$  values correspond to the no-stretch regime [28]. It is known that thermal conductivity is an essential property of Au and Ag nanosolids, which directly affects their applications. Also, nanomaterials with different morphologies, such as spherical nanosolids, nanofilms, and nanowires with thermal conductivity, are investigated in the previous study, where the thermal conductivity reduced as the grain size declined, indicating that the formulation utilized was appropriate, as shown in Figure 6 [77].

In this figure, it shows the variation of Debye temperature of pure gold and silver nanoparticles upon their size. The Debye temperature is a fundamental physical parameter that characterizes the highest temperature at which specific heat in a solid, due to lattice vibrations (phonons), follows the T<sup>3</sup> law described by the Debye model. [78]. Debye temperature is extensively used in solid-state physics to analyze thermal conductivity, specific heat, and sound velocity of solids.

When nanoparticles such as gold or silver are added to a matrix (polymer, ceramic, metal), they can change the vibrational properties of the system, often resulting in various effects on the effective Debye temperature. For example, it stated that the temperature of the Au–Al<sub>2</sub>O<sub>3</sub> nanocomposites changed over time for varying particle concentrations; the temperature difference curves that were produced show a linear relationship with time [79].

Figure 6 illustrates that the Debye temperature of pure gold and silver nanoparticles decreases as their grain size



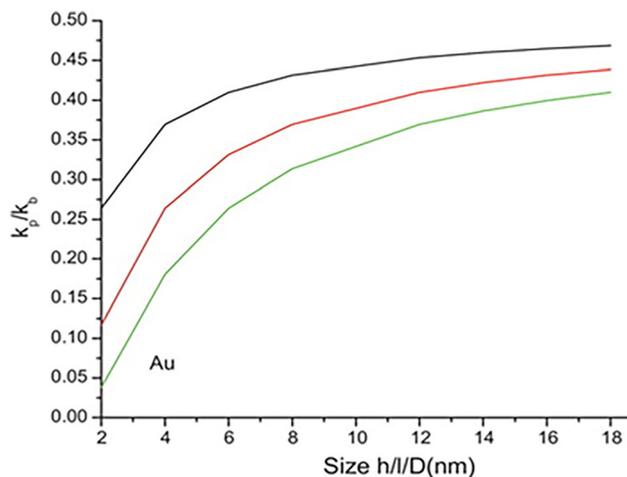
**Figure 6:** Grain size dependence of the  $d$  and  $\theta_D$  (Debye temperature) of gold and silver nanosolid displayed via solid lines. The solid circles represent the experimental values for Au nanosolids [77].

(diameter) decreases. This behavior is attributed to increased surface-to-volume ratio and reduced atomic coordination at smaller sizes, leading to softer lattice vibrations and lower phonon frequencies.

For matrix–nanoparticle composites, the paper explains that surface effects – arising from nanoparticle interfaces with the surrounding matrix – further modify vibrational properties. In a composite, the effective Debye temperature is influenced not just by the nanoparticle size but also by interfacial interactions, bonding strength, and possible lattice strain between the matrix and embedded nanoparticles. Typically, stronger interfacial coupling or stiffer matrix environments may partially counteract the size-induced softening of vibrations, resulting in a Debye temperature for the composite that is between those of the pure constituents and dependent on nanoparticle size, shape, and their dispersion in the matrix [77].

Although experimental data on the size-dependent variation of thermal conductivity in nanomaterials remain limited, Singh et al. calculated that the thermal conductivity of some nanoparticles, including silver and gold nanosolids, was depended on grain size [77]. The shape also acts as a vital function in the thermal conductivity of nanomaterials. For instance, silver nanowires (Ag-NWs) significantly improved the thermal conductivity of composites compared to AgNPs with a spherical shape, achieving a percolation threshold of fewer than 1% volume fraction as a result of improved particle interactions [1].

Singh et al. found that the thermal conductivity of NPs with different shapes but the same size decreases for a spherical nanoparticle, a nanowire, and a nanofilm, becoming 3:2:1, respectively (see Figure 7) [77].



**Figure 7:** Thermal conductivity of Au nanostructure as a basis of grain size for spherical nanosolids, nanowires, and nanofilms, represented by the black, red, and green curves, respectively [77].

According to Singh, et al. [77], the model's prediction that smaller nanoparticles yield lower thermal conductivity due to enhanced surface effects and phonon scattering. The thermal conductivity  $k_p$  of nanomaterials follows the equation of

$$k_p = k_b \left(1 - \frac{N}{2n}\right)^{\frac{3}{2}}$$

where thermal conductivity ( $k$ ) of bulk materials and  $N/n$  related to the diameter of the nanosolid. When spherical nanosolid, nanowire, and nanofilm were compared, the ratio was discovered to be 3:2:1. Therefore, for a spherical nanoparticle, nanowire, and nanofilm, the variation of thermal conductivity becomes 3:2:1 for the same grain size [77].

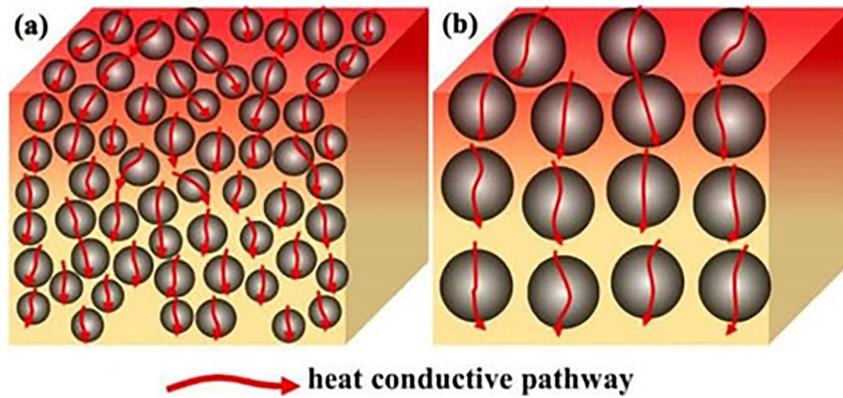
Furthermore, AuNPs with sizes smaller than five nm have proven to be very effective for various significant catalytic reactions. A main factor in their catalytic effectiveness is associated with the NPs' properties, which are totally dependent on the size of the NPs and their chemical composition [44].

The properties of composite materials that contain nanoparticles are significantly influenced by the NPs' size. The addition of NPs to enhance the thermal or electrical conductivity of a material usually influences the mechanical properties of the composite. NPs with the smallest size generally exhibit greater reactivity as a result of their raised surface area in comparison to volume ratio [1].

Additionally, the dimensions and shape of the metal nanoparticle fillers affect how the filler is dispersed within the matrix, determining the continuity of heat conduction channels. The dimensions of the filler particles greatly influenced the heat transfer within the nanocomposite material. For instance, smaller particles facilitate heat transfer more effectively, as they can create additional heat transfer routes within a matrix, as illustrated in Figure 8a. Then, considering interfacial thermal resistance, small-sized filler particles possess a large specific surface area, resulting in the formation of more filler–matrix interfaces. Consequently, larger particle fillers will decrease the area and quantity of heat crossing the filler–matrix interface, thereby lowering the total interfacial thermal resistance of the composites, as illustrated in Figure 8b [29].

The morphology of the filler is also believed to influence its distribution and state of connection within the matrix. The fibrous or lamellar fillers showed a bigger aspect ratio than spherical fillers, resulting in fewer filler–matrix interfaces along the length, which facilitates the establishment of a consistent heat conduction network structure within the nanocomposite [29].

Due to its inexpensive fabrication and its ability to enhance thermal conductivity, metal–epoxy composite has



**Figure 8:** Schematic representation of the heat transfer model of nanocomposites containing (a) small-sized and (b) large-sized particle fillers [29].

**Table 2:** Reported thermal conductivity of nickel–epoxy nanocomposites according to their size [80].

Particle size	10 nm	40 nm	70 nm	1.5 $\mu\text{m}$
Thermal conductivity (W/m K)	36.7	66.0	74.5	90.9

become an excellent choice for thermal interfacial material in various uses. For instance, nickel nanoparticle–epoxy nanocomposites have been developed and were found to exhibit thermal conductivity values exceeding those predicted by effective medium theory, even at low filler concentrations because of their more extensive aggregation structures in epoxy [80]. Likewise, larger enhancements in thermal conductivity are shown at the same concentration of nickel NPs with the smallest size, in contrast to the expectations of traditional effective medium approximation (EMA) models (see Table 2) [80].

### 1.3.2 Effect of concentration of AuNPs-composite on thermal conductivity

While numerous polymer nanocomposites have been reported, the specific impact of colloidal nanoparticles used as fillers on the thermal behavior of the polymer matrix has been addressed in only a limited number of studies [81]. It is well mentioned that the kind of incorporated NPs directly influences the properties of polymer composites. For example, these properties depend on size and shape, concentration of colloidal NPs, as well as their incorporation with the polymer matrix [82]. Few studies have focused on metal particles with a large aspect ratio to improve thermal conductivity. For instance, Zeng et al. [83, 84] used both silver nanowires and copper nanowires to enhance thermal conductivity in organic phase materials. Meanwhile, the thermal properties of cobalt nanowires and copper nanowires incorporated into a polymer matrix were investigated by Chen et al. [85] and Razeeb &

Dalton [86], targeting their application in thermal interface materials. The effect of the aspect ratio of AgNPs on the thermal properties of silver/poly(methyl methacrylate) nanocomposites has also been explored by Rivière et al. [87], where thermal conductivity is further enhanced by the low silver content. As far as we know, randomly dispersed gold nanowires have not been explored to increase thermal conductivity of thermoplastic materials. It has been highlighted that the addition of metallic NPs (such as Au or Ag), even at low percentages (0.8 % and 1.8 %), is enough to considerably raise the thermal conductivity of the polymer composite, which may be linked to the effects on the connections between the polymer chains [81].

Furthermore, the addition of nano-graphene to the composite material led to a promotion in the thermal conductivity of the resulting nanocomposite by about 52 %. Moreover, the addition of carbon nanotubes to the nanomaterial led to an increase in the thermal conductivity of the composite [87].

In addition, one of the common and effective polymers is polyvinylidene fluoride (PVDF), which is a thermoplastic polymer identified for its chemical and thermal stability, consequently being an outstanding material for electronics applications and the chemical industry. It is also eco-friendly and exhibits greater mechanical characteristics, consequently attracting high attention from researchers. However, it showed low thermal conductivity, which made it unsuitable for many different applications. For this weakens the numerous of thermal conductive fillers are added to increase thermal conductivity. The thermal conductivity of the composite materials is significantly increased, partially in the case of modified multiwall carbon nanotubes (s-MWCNTs) and graphene (GE)[s-MWCNTs/GE], and it was 711.1 % [66, 88] (see Table 3).

Further studies showed that an increase in the thermal conductivity of polyethylene/AgNPs composites occurs with increasing Ag volume fraction. This increase is to be expected, as the silver nanoparticles possess higher thermal conductivity compared to the polyethylene matrix. A comparable effect was observed, where the addition of 22 vol% of

**Table 3:** Improvement of thermal conductivity in PVDF composites by using MWCNTs and graphene [66, 88].

Composites	Formulation	Output
PVDF composite	PVDF without fillers	Minimal thermal conductivity
p-MWCNTs/GE/ PVDF	Pristine MWCNTs and graphene	Moderate thermal conductivity improvement
a-MWCNTs/GE/ PVDF	Acidified MWCNTs and graphene	Better dispersion and enhanced thermal conductivity
s-MWCNTs/GE/ PVDF	Silanized MWCNTs and graphene	Highest thermal conductivity improvement (711.1 %)

particles made from a polyamide core and a silver shell within a high-density polyethylene (HDPE) matrix resulted in a threefold enhancement in thermal conductivity relative to the unmodified matrix [89].

The thermal conductivity increases at low and medium concentrations of AuNPs ( $1.74 \times 10^2$  and  $4.35 \times 10^2$  wt%) in the waterborne polyurethane (WBPU)/Au nanocomposites. A small quantity of AuNPs could act as a nucleating agent for increased hydrogen bond creation in the WBPU matrix and crystallization. However, the opposite results are observed at higher concentrations (the presence of large quantities of Au,  $6.50 \times 10^2$  wt%), which is attributed to the Au content of the nanocomposite. AuNPs tend to aggregate and increase in size when the Au concentration is increased to  $6.50 \times 10^2$  wt%. The AuNPs with a big size could disrupt the crystallization process and the hydrogen bonding that occurs in the WBPU matrix. For this reason, the decline in thermal conductivity is noted [90].

In addition, the effect of nanoparticle concentration on thermal conductivity would add clarity by stating that several studies have experimentally demonstrated that the thermal conductivity of nanocomposites increases with nanoparticle concentration. For example, a review paper that mentioned that the addition of gold nanoparticles

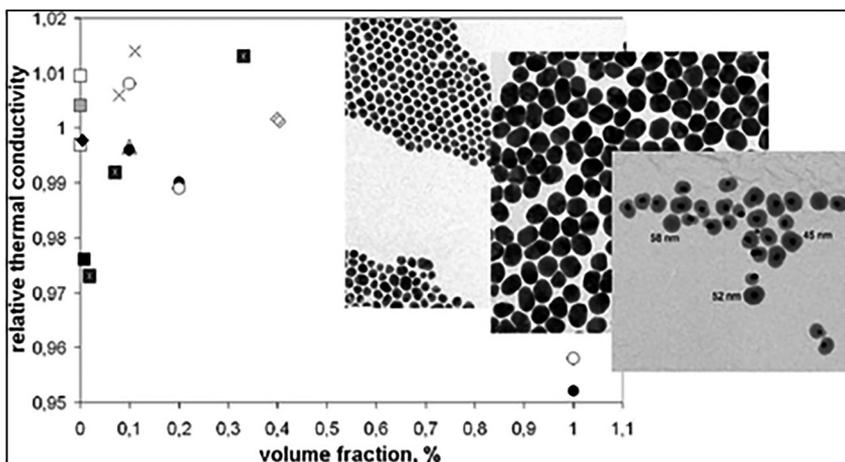
scattered inside poly N-isopropyl amide hydrogels resulted in notable modifications to the hydrogel's mechanical characteristics and thermal response [1].

Figure 9 shows another evidence of how the thermal conductivity of gold nanofluids is affected by the ratio of the volume of the nanoparticles to the total volume of the solution or composite [9].

The larger relative surface area of gold nanoparticles let to improve their stability and heat transfer capabilities. As the particle volume fraction decreases, thermal conductivity improves. Research has focused on elements that improve the thermal conductivity of nanofluids. The first thermal conductivity model for solid–liquid suspensions of spherical particles was created by Maxwell's theory. In order to make the Maxwell model suitable for nonspherical particles, Hamilton extended it. A more precise and dependable model is presented by Bruggeman's model, which was studied for huge volume concentration and takes into account the spherical particle surfaces. The Maxwell model was improved in a different study by Yu and Choi by reflecting a thin nano-layer on the fluid surface of the nanoparticles, which changed thermal conductivity and was shown to be accurate. A thermal conductivity model that examines the effects of variables such as temperature, volume fraction, Brownian motion, and nanoparticle size was provided by Kleinstreuer. The research shows accurate results, and the Brownian motion becomes increasingly significant at high temperatures [91–94].

### 1.3.3 Influence of heating on properties of gold–polymer nanocomposite

The influence of heating on gold–polymer nanocomposite film was studied. For instance, it has been shown that heating a nanocomposite at approximately 90 °C resulted in AuNPs being completely wrapped by the polymer molecules,



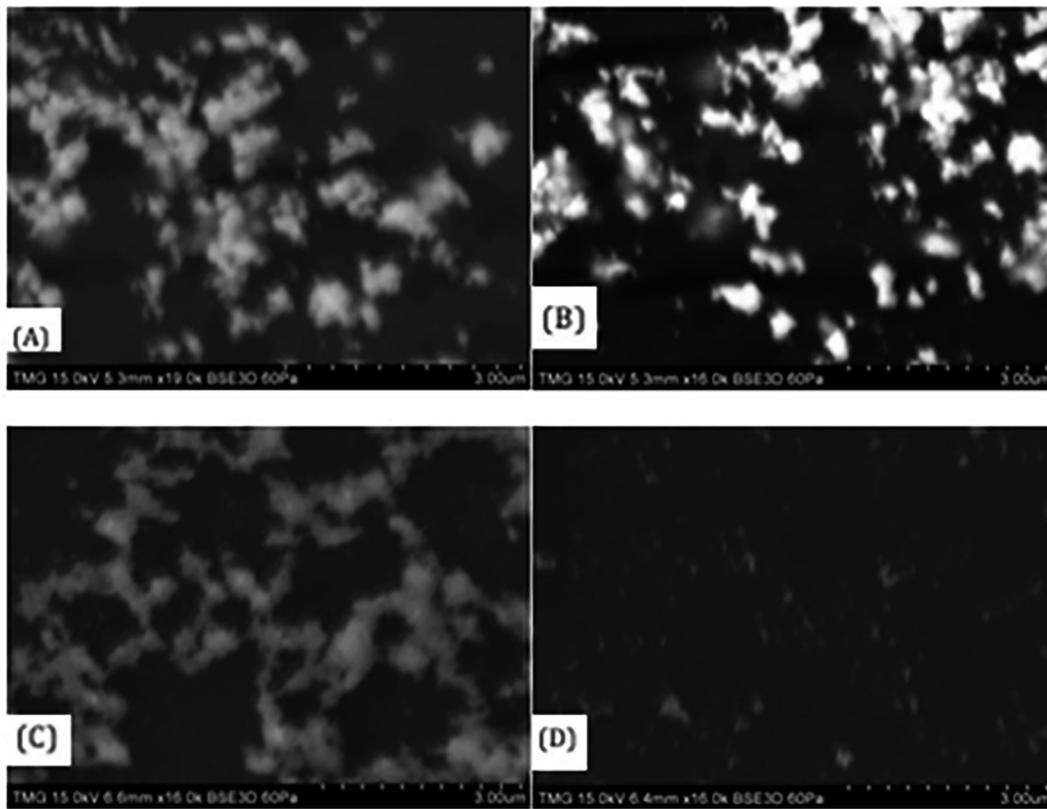
**Figure 9:** The effect of volume fraction of Au NPs on relative thermal conductivity [9].

such as polymethyl methacrylate (PMMA). It was also assumed that the preparation of the nanocomposite is simplified at the glass transition temperature ( $T_g$ ) of the Au-PMMA nanocomposite [35]. It is also worth noting that the novel viscous condition of the polymer adsorbed onto the NPs' surface creates a flow around the NPs, which considerably lowers the surface energy of the nanoparticles. This procedure can improve the dispersion of NPs and subsequently increase their various applications [35]. It is interesting to note that heating gold nanowires to 400 °C–500 °C caused a narrower surface plasmon resonance (LSPR) band and a blue shift; these changes showed a positive impact on sensing capabilities. Nonetheless, the behavior of gold-polymer nanocomposites when gradually heated to lower temperatures remains unreported [95]. Moreover, the nanocomposites undergo controlled heating that allows for the examination of the nanomaterial at various periods of heat treatment, enabling a detailed understanding of the process at both the cross section and on the surface of the nanocomposite. The evaporation of a colloidal gold solution onto the surface of polymer films using a convective technique is considered a novel approach for this kind of research, allowing for the progressive evaluation of gold particle infiltration from the surface to the bottom of the

polymer film, which is a valuable feature for this kind of study [35].

The aim of imaging the AuNPs-polymer systems is to identify the morphological and interdistance variations between the AuNPs on the polymer's surface. Figure 10 displays the images of two examined polymers including poly (dimethyl siloxane) (PDMS) and poly (vinyl alcohol (PVA). Figure 10A displays a PVA film with a gold deposit that has been slowly heated to 250 °C. Where metal particles are seen clustered together, creating filaments, which suggest the direct aggregation of the particles. The subsequent image, Figure 10B, represents PDMS coated with a gold nanoparticle, also heated to 250 °C. The amount of AuNP on PDMS was high, resulting in a branching aggregation, as evident from the image. The final two images, Figure 10C and D, illustrate AuNP on a glass slide, one of them without any polymer and the other one including a PDMS film that has not been heated. The particles in this image appear far sparser and more dispersed, as well as became a weaker attachment to the NPs surface [95].

The LSPR bands associated with 34 nm of AuNP on cyclic olefin copolymer (COC) and poly (methyl methacrylate) (PMMA) show a peak shift during heat treatments ranging from 80 to



**Figure 10:** SEM images of AuNPs on different surfaces: (A) polyvinyl alcohol (PVA) after heating; (B) poly (dimethyl siloxane) (PDMS) after heating; (C) glass slide with no polymer addition and not exposed to heating; (D) PDMS not exposed to heating [95].

250 °C. The LSPR of AuNPs-COC was increased to approximately 575 nm, while AuNPs-PMMA increased to 520 nm, indicating to smaller size. The locations of the AuNPs LSPR bands for heated samples were at longer wavelengths compared to particles located on the polymer's surface, as the bands shift to the right overall with rising temperature. The polymers exhibited an increase in the AuNPs LSPR peak from an average of 7–34 nm within the temperature range of 175–200 °C [95].

This could be due to an early aggregation and dispersal of AuNPs. Subsequently, as AuNPs become more incorporated into the polymer surface, the LSPR shifted toward longer wavelengths as a result of the aggregation process. This means that the LSPR band becomes more red-shifted, indicating variations in the optical properties of the material. The advantages of these observations include enhancing sensing and affinity techniques, as well as choosing the right polymer material for a particular bio-sensing use, based on the analysis involved, a process of gradual preheating [95].

It is important to note that because of the moderate thermal conductivity of solid oxides, high volume fractions (>5 %) are needed to reach notable improvements in thermal conductivity. Elevated particle concentrations cause issues with viscosity and colloidal stability, leading to instability. This makes the suspensions unsuitable for convective heat transfer [9]. The instability is likely due to the aggregation of particles, which can lead to clustering and settling, making it difficult to maintain a stable and uniform suspension. In certain situations, aqueous nanofluids containing very low concentrations of metal nanoparticles, including 50–100 nm CuNPs at 0.001 vol fraction and 10–20 nm AuNPs at 0.00026 vol%, have been noted to produce thermal conductivity improvements of up to 23.8 % and 21 % (at 60 %), respectively [9].

As stated by Ren et al., the spherical AuNPs are capped on the surface of graphene NPs at 25 °C. With the temperature increased, there is no important alteration in the morphology of the AuNPs, suggesting that the AuNPs are deeply attached to the surface of the graphene NPs at R.T. and remain stable with temperature [48].

Uehara et al. developed gold nanoparticles coupled with a thermoresponsive polymer without undergoing chemical reduction. They discovered gold nanoparticles fused to create larger nanoparticles when a solution of gold nanoparticles coated with thermos-responsive polymers was heated because the dehydration caused the chains of the thermos-responsive polymers to break [96].

#### 1.4 Surface functionalization

AuNPs are perfect probes for biological assays because of their many advantages over other NPs, such as their high

molar absorption coefficient, simple surface modification, and controllable manufacturing. Since small biomolecules, such as sulfhydryl compounds and amino acids are attached to the surface of gold nanoparticles more simply and controllably than larger biomolecules. Because of their simpler structure and reduced steric hindrance, AuNPs functionalized by small-molecule are often utilized to design and develop sensors for biochemical analysis [97].

As is known, the surface of AuNPs is usually functionalized with several molecules or ligands, to improve compatibility and dispersion within the composite matrix, thus improving thermal conductivity [26].

Earlier research has mainly concentrated on the AuNPs properties in liquid environments, including ionic liquids and aqueous solutions. This emphasis is largely because unmodified AuNPs often exhibit a tendency to aggregate in aqueous solutions. By functionalizing the surface of AuNPs, scientists can potentially control the equilibrium size of these required nanoparticles, inhibiting additional growth. Moreover, surface modification can also enhance the compatibility between AuNPs and the surrounding medium. Despite the numerous computational studies interactions of gold nanoparticles with liquid, there is a lack of research on incorporating AuNPs into thermoplastic polymers [26].

According to the study's computational modeling, gold nanoparticles and the epoxy matrix form robust interfacial contacts that limit the mobility of polymer chains near to the nanoparticle surfaces. The glass transition temperature ( $T_g$ ) was raised by this confinement, which also enhanced the hybrid nanocomposite's thermal stability. Despite the improved thermo-mechanical behavior at the molecular level, the mechanical characteristics mainly remained the same because of the low concentration of nanoparticles [26].

The study by Milano et al. [98] employed coarse-grained simulations to examine the molecular-scale structuring of AuNPs functionalized by 1-dodecanthiols in a polystyrene matrix environment. Their research revealed that the capping layer surrounding the AuNP affects the orientation of the polymer chains at the AuNP interfaces.

The simulations show that polymer segments near the gold surface tend to adopt more ordered conformations and exhibit slower relaxation times compared to the bulk polymer, indicating a strong interfacial influence. Additionally, the nature of the interaction – whether bare or coated gold nanoparticles – determines the extent of polymer structuring and mobility at the interface [98].

Another method consists of modifying a polymer (poly(vinyl alcohol), PVA) with (3-mercaptopropyl)trimethoxysilane (MPTMS), resulting in the formation of thiol groups on its surface. Subsequently, AuNPs are incorporated

onto the partially dried, modified PVA surface, leading to the chemisorption of AuNPs onto the thiol groups [41]. Theoretically, it was demonstrated that allowing the particle size ratio to rise should result in a notable improvement in specimen conductivity. Comparing the polyethylene matrix to the poly(methyl methacrylate) matrix, the polyethylene matrix displayed an unusual change in volume resistivity with gold content [41].

However, the adhesion of polymer chains to nanoparticle surfaces, whether by adsorption or grafting, plays a critical role in establishing the chemical and physical properties of nanocomposites. In adsorption, polymers adhere to the surface via physical interactions such as van der Waals forces or hydrogen bonding. Grafting involves covalent bonding, leading to more stable attachment and often more controllable interfacial properties [99].

A strong study providing detailed discussion on the role and molecular weight effects of polymer chains adsorbed or grafted onto gold nanoparticles. It discovered that the size and diffusion coefficient of the polymer chains strongly influenced the adsorption rate when using PS ligands with varying molecular weights. It was able to verify the precise adsorption of stabilization of polystyrene (PS) on the AuNP surface by using AFM to view the polymer chains on the particles in their dry state [100].

In the last 10 years, the progression of thermally conductive polymer nanocomposites has attracted notable attention in the thermal management field for electronic devices such as thermal interface materials (TIMs). It is noted that, by integrating the advantages of polymers, such as low cost, varied functionality, lightweight nature, and exceptional chemical resistance, with nanofillers that provide high thermal and electrical conductivity, they become a practical option for thermal management systems in electronic devices. The addition of nanofillers, which exhibit strong thermal conductivity to the polymer matrix, is commonly applied to boost thermal conductivity, since polymers naturally possess lower thermal conductivity, typically, have range from 0.2 to 0.5 W/m.K. Furthermore, the functionalization of the nanofillers' surface is particularly required to prevent the agglomeration of the fillers, which is driven by their higher surface energy [101].

#### 1.4.1 Covalent functionalization

The function of surface modification and physical characteristics of nano-inclusions play a vital role in improving thermal conductivity through the phase of liquid–solid transition in a hexadecane-derived phase change material (PCM). Hexadecane-based PCM incorporates six varieties of nano-inclusions, including copper nanoparticles (CuNPs), carbon black nanopowder (CBNP), silver nanowires (Ag-

NWs), nickel nanoparticles (NiNPs), graphene nanoplatelets (GNPs), and multiwalled carbon nanotubes. The NiNP, CBNP, AgNW, and GNP have undergone surface functionalization using oleic acid as a stabilized ligand. While nano-inclusion-loaded PCM revealed a great improvement in thermal conductivity, this was more pronounced in the solid form [102].

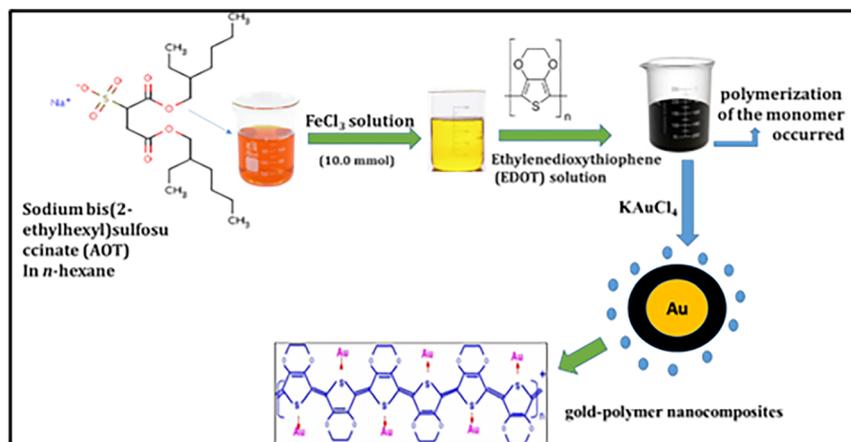
Recent research has shown individually the advantages and disadvantages of filler modification on thermal conductivity, resulting in enhanced thermal conductance at the interface between the matrix and filler; however, a decrease in the intrinsic thermal conductivity of the filler was observed. In addition to the difficulty in this situation, it was figuring out how to benefit from functionalization at the filler/matrix interface, despite keeping the inherent thermal conductivity of the filler unchanged. It is illustrated that the influence of filler functionalization of the bulk composite on the thermal conductivity remains insufficiently clarified. Some findings indicate that functionalization has a positive effect on the thermal conductivity of the final resulting material, while the improvements are usually slight. Nonetheless, functionalization may also contribute to improvements in other properties, including mechanical or electrical properties. The literature reveals enhancements in thermal conductivity and electrical conductivity in polyimide nanocomposites that incorporate one-dimensional silicon carbide (SiC) semiconductor nanowires (SiC-NWs) capped on graphene sheets. Advancements are achieved in utilizing SiC-NWs as a functionalized material to increase thermal conductivity in composites, because SiC nanowires have excellent thermal conductivity [6].

## 1.5 Preparation methods of gold nanocomposites

### 1.5.1 Synthesis of gold–polymer nanocomposites

Multiple approaches for preparing gold nanocomposites that combine polymers have been outlined, such as the electrochemical deposition of nanoparticles onto electrodes capped with conducting polymers (CPs), the photochemical approach, reducing metal precursors in a polymer matrix, blending of nanoparticles into a polymer matrix, and polymerization of CPs around NPs. Among the main CPs, poly(ethylenedioxythiophene) (PEDOT) has been confirmed to be largely interesting because of its optical transparency when it is in a conducting state [103].

Nanocomposite materials containing gold and polymers (polyethylene oxide, polyvinyl pyrrolidone, and polyvinyl alcohol) were synthesized through a chemical reduction method [104]. For instance, gold–polymer nanocomposites



**Scheme 1:** Preparation of gold–polymer nanocomposite, and how AuNPs are incorporated into the polymer structure, the process is taken from Ref [103].

of 50 nm in size have been chemically synthesized using the reverse emulsion polymerization technique, in which the polymer backbone was combined with Au nanoparticles via potential interactions of Au–sulfur bound in the thiophene compound, as illustrated in Scheme 1 [103].

According to Pham et al., thermal treatment technique is used to prepare the polymeric–metallic nanocomposite [105].

It is acknowledged that to produce gold NPs within a polymer matrix, the typical synthesis method involves introducing presynthesized AuNPs into polymer aqueous solutions through various techniques. It was demonstrated that NPs typically aggregated and lost their SPR characteristics. Consequently, stabilizing agents are necessary to achieve uniformly distributed NPs [105]. However, attaining a uniform spatial arrangement at larger scales within the polymer matrix remains challenging [106].

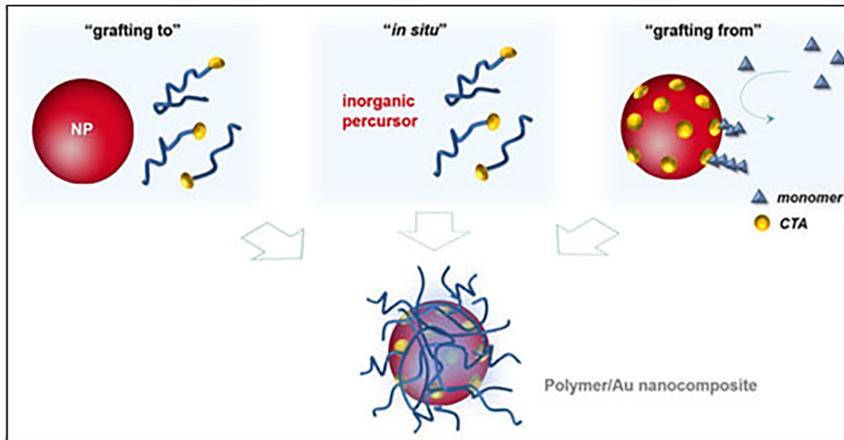
To address these difficulties, AuNPs–polymer composites are prepared by doping polymer films with HAuCl<sub>4</sub> as gold precursor using the spin-coating method. The progress of nanoparticles within the polymer matrix is stimulated by exposing the film to solar radiation [105]. It has been observed that incorporating photo-sensitive chemical ligands into the metal/polymer nanocomposite and exposing them to the light of UV allows for partial control of the shape of the nanoparticles [106].

Among various polymers, SU-8 resist is a preferred polymer used in AuNPs composites due to its affordability and frequent application in diverse areas like photonic devices and microfluidics, attributed to its superior mechanical characteristics, chemical stability, promising biocompatibility, and potential for functionalization. Furthermore, earlier research indicates that SU-8 demonstrates considerable promise in photoreduction as a result of the creation of free radicals and strong chemical functionality through the photochemical reactions [105]. Shukla et al. proposed a technique for preparing Au nanostructures that combines a

polymeric matrix by using the two-photon lithography technique [43]. In this technique, a simultaneous reduction of HAuCl<sub>4</sub> takes place along with the polymerization of SU-8 resist, consequential in polymeric lines saturated with AuNPs [107].

Moreover, polypyrrole (PPy) is important due to its excellent conductivity and durability in the atmosphere. Composites made of PPy and AuNPs are currently a significant focus of research because of the unique optical, charge retention, and catalytic characteristics of MNPs [108]. For instance, gold–PPy core–shell nanoparticles are synthesized in diblock copolymer micelles, and it has been found that significant segregation can be effectively avoided when the composite particles are generated inside the cores of the diblock copolymer micelles. Furthermore, when pyrrole and HAuCl<sub>4</sub> are mixed, the redox reaction that occurs between HAuCl<sub>4</sub> and pyrrole results in the creation of PPy/Au nanocomposites, exhibiting a conductivity of  $12.6 \pm 0.06$  S/cm, which is almost four times that of pure polypyrrole pellets [108].

Additionally, the reversible addition–fragmentation chain transfer (RAFT) polymerization shows control over the composition and polymer shell structure that coats AuNPs. RAFT polymerization enables the creation of a polymer shell featuring specific functional groups designed for Au-based nanocomposites that are appropriate for biomolecular recognition and targeting. Moreover, synthesizing polymer/Au nanocomposites through RAFT polymerization provides a benefit associated with employing chain transfer agents (CTAs). CTAs typically consist of thiocarbonylthio compounds, and they exhibit a strong attraction surface of AuNPs due to the presence of sulfur atoms. Consequently, polymers utilizing di- and tri-thio-CTA agents are used in order to modify the surfaces of gold nanoparticles, as they can create a robust bond between the polymer and the nanoparticle surface (see Figure 11) [109].



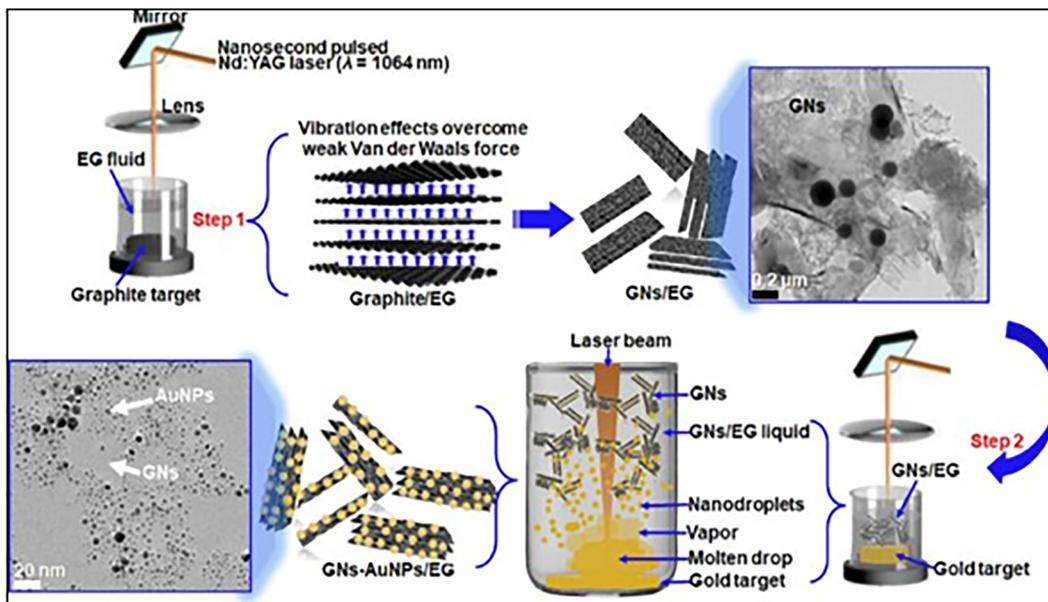
**Figure 11:** Common methods to prepare polymer/Au nanocomposites [109].

### 1.5.2 Synthesis of gold–graphene nanocomposites

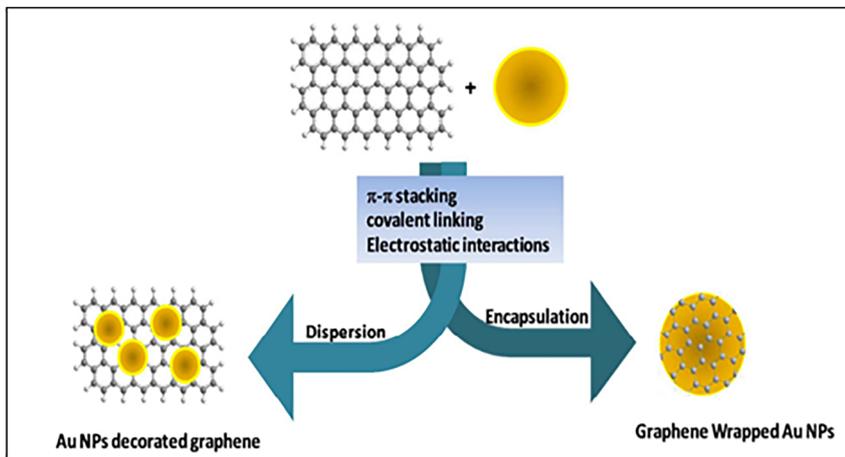
Most of the published research on the fabrication of graphene–metal nanocomposites utilizes organic ligands, such as octadecylamine, which is used to attach MNPs to the graphene surface, or uses some organic solvents, including ethylene glycol, tetrahydrofuran, and methanol. Recently, graphene/gold nanocomposites were successfully created using a straightforward preparation technique in an aqueous medium [51].  $\text{HAuCl}_4$  serves as the frequently used metal precursor for the creation of gold nanoparticles (AuNPs) [69]. Moreover, the graphene–AuNPs nanocomposites are created via a nanosecond pulsed Nd: YAG laser technique to remove the graphite target, subsequently introducing gold into

ethylene glycol (EG) fluid to generate GNs–AuNPs/EG hybrid nanofluid, as illustrated in Figure 12 [51].

Furthermore, graphene possesses various properties that render it a more advantageous material in comparison to other types of carbon-based substances like carbon nanotubes (CNTs). Currently, there are two synthetic methods for creating graphene–AuNPs composite materials. The distribution of AuNPs on graphene oxide (GO) or reduced graphene oxide (rGO) films offers a direct technique to embellish graphene sheets with Au nanostructures. The other method involves encapsulating Au clusters within graphene-based cages. For example, both GO and rGO are suitable for these techniques, due to the presence of oxygen functional groups in their structures, which are crucial for the following chemical



**Figure 12:** A diagrammatic representation of the preparation method for GNs–AuNPs/EG hybrid nanofluid produced by pulsed Nd: YAG laser ablation of graphite targets and gold targets in EG and GNs/EG fluid [51].



**Figure 13:** Decoration of reduced graphene oxide and graphene oxide with Au-NPs using different interaction forces [110].

reactions as well as in the combination of gold nanostructures (Au-NSs) via covalent bonding,  $\pi$ - $\pi$  interaction, and electrostatic interactions (see Figure 13) [110].

The attachment of AuNPs on GO layers inhibits the graphene sheets from restacking. Recently, the Au-NSs have served as spacers, enhancing the gap between the sheets of graphene, thus allowing access to all sides of the graphene. The synthesis of Au-NSs loaded on graphene nanocomposites is generally divided into two groups, namely *ex situ* and *in situ* growth. In the *ex situ* approach, the synthesis of AuNPs of specific shape and size was achieved, followed by their attachment onto a GO, graphene, or rGO matrix. Whereas in the *in situ* approach, GO and Au salt undergo simultaneous chemical reduction to synthesize AuNPs-rGO nanocomposite materials. Additionally, the AuNPs are prepared individually from GO or rGO nanosheets in the *ex situ* method and are combined through using  $\pi$ - $\pi$  bond interactions, electrostatic, or van der Waals interactions. In this regard, GO nanosheets are connected to AuNPs functionalized by 2-mercaptopyridine *via*  $\pi$ - $\pi$  stacking (see Figure 13). Cysteine is used as a linker molecule, which is particularly desirable due to the presence of amine ( $\text{NH}_2$ ) and thiol ( $\text{SH}$ ) as active functional groups in its structure. Additionally, the cysteine uses its  $\text{NH}_2$  group to link with the carboxylic acid groups of GO, while its  $\text{SH}$  group is used to link with AuNRs. The integrated AuNRs were observed to disperse the GO sheets within a 3D layered structure. A comprehensive 3D tomography study investigated that graphene sheets were organized in a parallel configuration rather than randomly [110].

Similarly, various approaches have been described for creating different metal nanoparticles/reduced graphene oxide (MNPs/rGO) nanocomposites, including electrochemical deposition and chemical reduction [111], thermal reduction [112], photochemical reduction [113], microwave

irradiation [114], and sono-chemical reduction [115], among others. Many of these techniques involve combining a metal precursor with a water or ethanol-based GO suspension, followed by the addition of hydrazine hydrate, glucose, ascorbic acid, or sodium citrate as reducing agents in order to reduce both GO and metal ions, leading to the production of MNPs/rGO nanocomposites [69].

Although there is limited research on metal/graphene/conducting polymer composites. However, nanocomposites (NSPANI/AuNP/GR) consisting of nanostructured polyaniline (NSPANI), gold nanoparticles (AuNPs), and graphene nanosheets (GR) are prepared by using *in situ* polymerization, demonstrating excellent conductivity ( $4.31 \times 10^{-5}$ ) and strong stability. Nanostructured polyaniline (NSPANI) has unique properties compared to other polymers, due to its ease of preparation, stability in various environments, adjustable electronic conductivity, flexible electrochemical switching behavior, reversible doping/dedoping chemistry, strong mechanical properties, and suitability for making composites with different kinds of binders [116].

Another study demonstrated the synthesis of AuNPs/graphene composites functionalized by poly (diallyldimethylammonium chloride) (PDDA) using a one-pot method, which is fast, easy, and moderate conditions. The produced nanocomposites exhibited the superior conductivity and electro-catalytic abilities of graphene and gold nanoparticles [117].

## 1.6 Characteristic methods in thermal conductivity of gold nanocomposite

Accurate evaluation of thermal transport properties is important in nanoparticle-enhanced materials; the thermal conductivity has been evaluated by using different methods including.

### 1.6.1 Transient hot-wire method (THW)

Numerous methods are applied in order to measure the thermal conductivity of materials. Nevertheless, the transient hot-wire method has several remarkable advantages compared to other methods, because of its uncomplicatedness and suitability for many materials. The hot-wire method can provide rapid and optimum measurements of liquids, gases, and some solids over a relatively wide range of thermal conductivity. Furthermore, it also can apply to measure the thermal conductivity under high pressure and high temperature with careful calibration of the hot-wire instrument, which is important for several industrial applications. This mention advantage made the THW method one of the most commonly used [118].

In addition, the THW method is a standard method compared to the consequences achieved with any newly advanced method used to measure the evolution of special heat transfer fluids such as colloidal suspensions of nanometer-sized particles. Transient methods rely on transferring heat from a controlled source to a material and measuring the temperature variations resulting from the heat dissipation to reveal thermal properties over time. The hot-wire technique, in particular, is a worthy method for determining the thermal conductivity of nanomaterials [119].

The transient hot wire (THW) method normally involves immersing a metal wire in a fluid. The fluid and metal wire are initially in thermal equilibrium. The wire is subjected to a potential gradient, producing constant heat along its length. The transient rise in the wire's temperature depends on the thermal conductivity of the surrounding fluid, while the wire's temperature is typically evaluated by the alteration in electrical resistance. When the wire is away from its ends, it acts as an infinite linear heat source surrounded by an infinite volume of fluid.

The thermal conductivity of the fluid,  $\lambda$ , can be detected by the following equation

$$\lambda = \dot{q} / 4\pi (T_2 - T_1) \ln(t_2/t_1).$$

$\lambda$  is the thermal conductivity of the fluid,  $\dot{q}$  is heat generation per unit length of the wire,  $T_2 - T_1$  is the temperature increase of the wire between two times, and  $t_1, t_2$  are two different heating times during the transient process [9].

The improved mathematical model of the hot wire technique considers an infinitely long, thin, linear heat source (wire) with uniform temperature distribution, immersed in a homogeneous, infinite experimental sample [119].

It is illustrated that the wire serves as a source of heat with an approximately constant heat flux per unit length,

generating a time-dependent thermal field within the wire and the test material. The hot wire transient kind can generate thermal conductivity measurements with the lowest uncertainty achieved to date ( $\pm 0.2\%$ ). In reality, the transient hot wire technique uses a thin metal wire of limited length to provide a heat source when current flows through it, and the change in its resistance over time is used to monitor the temperature rise within it. The temperature evolution of a wire depends partly on the thermal conductivity of the surrounding material [120].

### 1.6.2 Laser flash analysis (LFA)

Laser flash analysis (LFA) is widely employed to determine the thermal conductivity of solids at different temperature ranges. Where a sample receives a short laser pulse during testing from its bottom surface, and the corresponding temperature increase is measured on its top surface, which is used to measure the thermal conductivity of the solid sample. Liquid samples, on the other hand, can be encapsulated in a solid vessel with much higher thermal conductivity, so the resulting temperature increase is highly sensitive to the thermal conductivity of the sample of liquid sample within the vessel. Among all the aforementioned investigations, LFA was the most widely used technique, and the design of the apparatus and associated testing system was relatively more advanced [121].

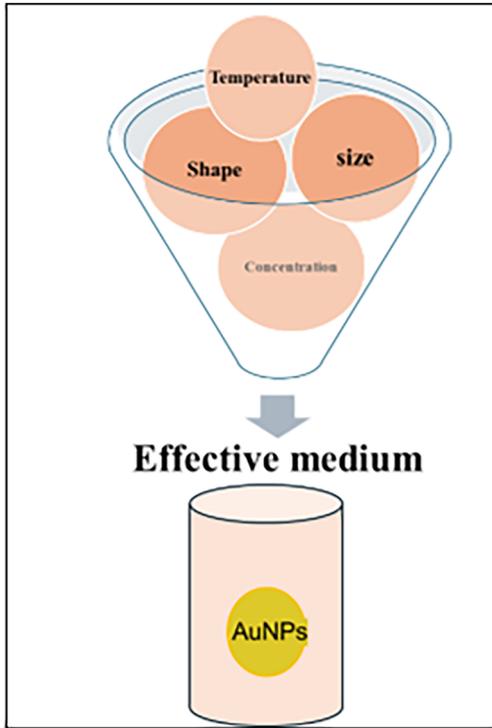
### 1.6.3 Transient plane source (TPS) or hot-disk technique

The hot-disk transient plane source (TPS) method is an extensively used standard technique for evaluating the thermal conductivity of materials [122].

By using this technique, it is potential to measure thermal and diffusivity conductivity over nearly six orders of magnitude under well-controlled conditions at temperatures ranging from 25 K up to 1,500 K. Another benefit of using the transient method is the probability of eliminating the effect of contact resistances that are continuously present between the heating element, which acts as a temperature recorder as well, and the substrate's surface under investigation [123].

## 1.7 The fundamental thermal conductivity mechanisms in gold nanocomposites

The fundamental mechanisms and key characteristics that influence heat transfer in nanocomposites, including different factors: Brownian motion of NPs, the nanoscale layer effect, nanoparticle aggregation, and the nature of heat



**Scheme 2:** The common factors that affect the thermal conductivity of nanomaterials.

transfer within the NPs. The main elements affecting the thermal conductivity modeling of nanofluids are the nanoparticle concentration, temperature, particle size, shape, pH, and properties of the nanoparticle layer (see Scheme 2) [124].

Although several mechanisms, such as Brownian motion, aggregation, ballistic transport, and internal nanoparticle potential, have been speculated, experimental proof of any of these mechanisms is not easy [125].

The mechanisms of thermal conductivity enhancement in nanocomposites remain unclear, and new experiments are required in order to understand the real mechanisms. Nanocomposite is a hopeful material to control the thermal conductivity of organic compounds via the incorporation of fillers with a high thermal conductivity. Furthermore, electrically insulating fillers, including aluminum oxide, boron nitride, and boron carbide, are usually applied when insulator nanocomposites are needed in some desired applications. Applications that do not require insulation use electrically conductive fillers like carbon nanotubes (CNTs), metal nanowires, graphene, and metal nanoparticles, where heat will flow through both the organic matrix and inorganic fillers. Thus, the loading ratio, shape, and size of filler, filler dispersion, and ITR, all determine how thermal conductivity affects the composite material. For example, in the case of polymer-based nanocomposites embedded with randomly dispersed CNTs or graphite, a point contact with a small

contact area will be formed between two cross CNTs or rigid graphite nanoplatelets. The high contact thermal resistance can be observed as a result of the lack of interaction between the contacted fillers [126].

Thermal conductivity (TC) is considered to be one of the significant properties of materials, and its exact value is very valuable for the thermal design of individual components as well as the all system. The thermal conductivity of a material depends on many parameters such as composition, impurities, and temperature of the material. There are many composite materials with the progress of technology; these composites are established with known thermal conductivity values for practical applications [127].

Moreover, nanofluids, as a new class of solid/liquid suspensions, show scientific challenges due to their thermal conductivity being one higher than expected. It has been commonly known that nano-layered solid-like structures are formed due to the closeness of liquid molecules to a solid surface, while the connection between thermal properties of the suspensions and these nano-layers is still not well known [4].

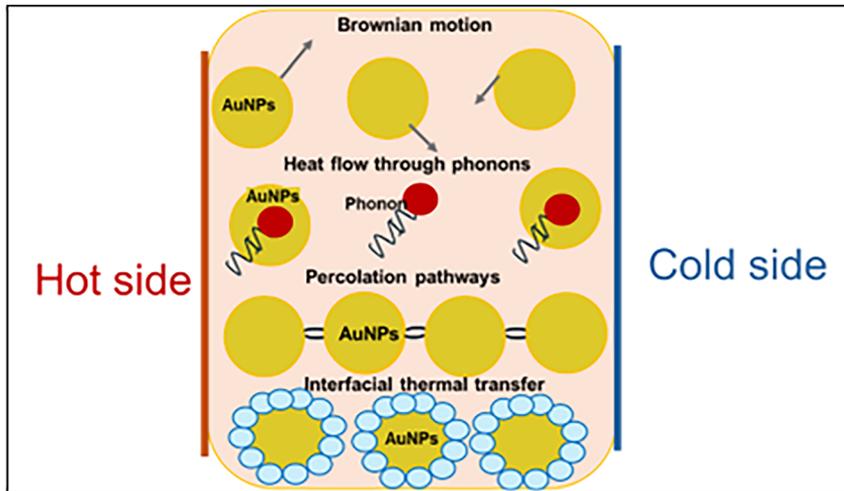
The effective medium theory is a theoretical framework that is usually used to predict the properties. The individual components in nanofluids are the fluid and the NPs suspended within it. The effective medium theory is used to evaluate the viscosity and thermal conductivity of nanofluids according to the properties of both NPs and the fluid [128, 129].

There are many base oils and nanoparticles involved in the engineering application. Heat transfer fluids, such as engine oil and transformer oil, are among the most common base fluids. Also, different metallic particles, such as copper, silver, and gold, and metal oxide particles, such as copper oxide, zinc oxide, aluminum oxide, and titanium oxide, are mainly used for enhancing the thermal properties of base fluids [93].

The fundamental mechanisms and key characteristics that influence heat transfer in nanocomposites include four main factors: the nanoscale layer effect, Brownian motion of NPs, phonon transport, and percolation pathways [124] (see Scheme 3).

#### **First: Interfacial Layering**

The investigation of the creation of an interfacial layer around the NPs that are dispersed in a fluid has been conducted. The formation of liquid molecules around the NP creates an interfacial layer; this interfacial layer can be known as one of the important mechanisms in order to increase the thermal conductivity of nanofluids. This layer around the particles displays a higher thermal conductivity compared to the base fluid, and it serves as a thermal bridge between the base fluid and nanoparticles [130–132].



**Scheme 3:** The fundamental thermal conductivity mechanisms in gold nanocomposites.

### Second: Brownian Motion

The Brownian motion is the random motion of the NPs in the fluid, which assists in transferring heat more efficiently. The main factor of this motion is the temperature increase, which decreases the fluid's viscosity and reduces the agglomeration while increasing the Brownian motion [133].

### Third: Phonon Transport

Phonons are responsible for heat transport through thermal vibration motion, which are more significant in nanocomposites. It implies that enhanced thermal conductivity occurs due to the increased electron–phonon interactions between the fluid and the nanoparticles. In addition to promoting Brownian motion, interfacial layers enhance heat transfer via phonons [134].

### Fourth: Percolation Pathways

This theory indicates that the enhanced thermal conductivity is formed due to the formation of a percolation network of NPs within the fluid. NPs are dispersed randomly in the fluid at low concentration and do not contribute to thermal conductivity. However, when their concentration increased, they began to aggregate to form a network that enhances thermal conductivity. However, these aggregated clusters lead to reduced Brownian particle velocity within the nanofluid [28, 127].

This work deals with the prediction of thermal conductivities (TCs) of a composite material wherein the filler particles of high TC are randomly distributed in a base matrix of low TC. It has been an important issue to predict the effective TC of composites with different volume fractions of filler particles of high thermal conductivity. When the filler volume fraction rises beyond a specific value, the filler particles come into contact with each other and generate

heat transfer paths, the latter is known as percolation. In the latter case, a sudden increase in the thermal conductivity of the composite materials is expected [127].

## 1.8 Comparative evaluation of gold nanocomposite with other nanoparticles in enhancing conductivity

The thermal conductivity of for copper oxide (CuO, 36 nm) and alumina ( $\text{Al}_2\text{O}_3$ , 33 nm) nanoparticles dispersed in water at R.T was calculated, where these NPs were dispersed in deionized water. The high thermal conductivities of CuO and  $\text{Al}_2\text{O}_3$  nanoparticles were 7.8 W/mK and 4 W/mK, respectively, confirming excellent thermal conductivity values. Furthermore, the thermal conductivity of  $\text{SiO}_2$  with a size of 20 nm showed a low value of 1.39 W/mK compared to CuO and  $\text{Al}_2\text{O}_3$  NPs, while copper oxide nanoparticles exhibited moderate thermal conductivity with a value of 1.618 W/mK. The authors showed that the increase in the thermal conductivity of nanofluids was caused by an increase in the density of NPs instead of their composition. Where the density of a nanosized material is a size-dependent parameter, the density of nanomaterials declines as their size declines, especially at a size of less than 20 nm. Therefore, the decrease in the density of CuO and  $\text{Al}_2\text{O}_3$  nanoparticles with diameters of 36 nm and 33 nm, respectively, was not statistically significant because their sizes were greater than 20 nm (see Table 4). The thermal conductivity enhancement of the CuO nanofluid was 28 % higher than that of aluminum oxide ( $\text{Al}_2\text{O}_3$ ) at 5 % volumetric concentration. This augmentation resulted in a higher density of the CuO (bulk density = 6.3 gm/cm<sup>3</sup>) compared to the  $\text{Al}_2\text{O}_3$  (bulk density = 3.9 gm/cm<sup>3</sup>) [76].

**Table 4:** Thermal conductivity values of metal oxide nanofluids [76].

Type of nanoparticle	ZnO (20 nm)	SiO <sub>2</sub> (20 nm)	Al <sub>2</sub> O <sub>3</sub> (33 nm)	CuO (36 nm)
Thermal conductivity (W/m·K)	1.61	1.39	4	7.8

Moreover, there were variations in the effective thermal conductivity of ZrO<sub>2</sub>, Ni, Cu, Pb, In, and Sn nanoparticles with their shape and size. As is known, when the size of NPs decreases, it is expected to increase. For instance, the thermal conductivity decreased sharply when the size of the nanoparticle was below 30 nm [135]. Additionally, the thermal conductivity of titanium dioxide (TiO<sub>2</sub>) nanofluid is quite low, resulting in a low liquid temperature [136], (see Scheme 4).

It is known that the thermal conductivities of nanofluids including 250 nm gold nanoplate were found to be higher than 15 nm spherical gold particles [137], confirming the size- and shape-dependent thermal conductivity of the nanoparticles.

As reviewed recently, different shapes of carbon nanotubes significantly affect the effective thermal conductivity, increasing the dispersion of multiwalled carbon nanotubes by 37 % [137].

High thermal conductivity of nanotubes (CNTs) was described as 6,600 W m<sup>-1</sup>K<sup>-1</sup> in the literature, where the thermal conductivity of CNTs depends mainly on their sizes. For example, thermal conductivity increases with decreasing sizes due to the lower incidence of Umklapp processes in smaller carbon nanotubes. For example, the thermal conductivity of CNTs with a diameter of 3.46 nm was 1,512.4 W/m K and decreased to 738.8 W/m K when the size decreased to 2.59 nm. In another study, the thermal conductivity increases

with increasing sizes of CNTs as a result of longer phonon relaxation time [138].

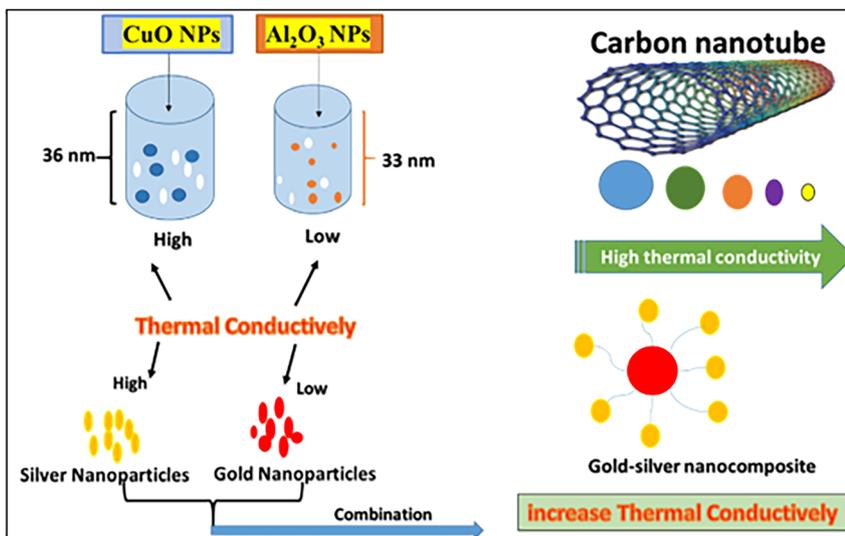
According to the literature, CNTs with tube lengths of 2.5 nm, 20 nm, 40 nm, and 1,000 nm showed thermal conductivities of 6,600, 7,000, 2,980, and 355 W/m K, respectively [139].

It is indicated that multiwalled carbon nanotubes (MWCNTs) perform better than single-walled carbon nanotubes (SWCNTs) in terms of thermal conductivity. This is because the larger diameter of the nanotubes is accompanied by more optical phonon modes, which can be susceptible to excitation and contribute to thermal conductivity. If a defect occurs in the SWCNT structure, its effect on the conductivity properties is much stronger than in MWCNTs. This consequence of the fact that the adjacent shell in MWCNTs may create additional new phonon channels, which is impossible in SWCNTs [140].

According to Balogun et al., AgNPs in ethylene glycol have higher thermal conductivity (429 W m<sup>-1</sup>K<sup>-1</sup>) than gold nanoparticles (318 W m<sup>-1</sup>K<sup>-1</sup>) [141]. However, one common disadvantage of AgNPs is that it is very difficult to prepare silver nanoparticles with a well-defined size, as this requires an additional step to prevent particle aggregation, and silver nanoparticles are more easily oxidized than gold nanoparticles, which reduces their stability [23].

When gold nanoparticles are combined with silver nanoparticles or with other highly conductive materials, such as graphene have shown higher thermal conductivity than the individual materials alone [141].

Therefore, improvements of 0.723 W m<sup>-1</sup>K<sup>-1</sup> and 0.698 W m<sup>-1</sup>K<sup>-1</sup> in the thermal conductivity were shown for Ag-nanofluids with a size of 15 nm and Au-nanofluids (70 nm), respectively, compared with pure water.

**Scheme 4:** Influence that influences size, shape, and composition of nanoparticles on their thermal conductivity.

Adding AuNPs to water increased the thermal diffusivity by 15.9 %, while the increase with silver nanoparticles (AgNPs) was 20.1 %. The thermal conductivity of aluminum nanofluids decreased with increasing particle size, practically for sizes less than 50 nm.

The increase in thermal conductivity was observed for the ethylene glycol nanofluid base and titanium particles with a diameter of 10 nm, found to be double that of 70 nm particles at the same volume fraction [142].

## 1.9 Current and emerging applications in thermal conductivity of AuNPs-nanocomposites

Thermal management technology is widely used in an extensive range of applications, from small electronic devices for daily use (such as mobile phones and laptops) to large-scale heat exchange systems in the energy, chemical, automotive, aerospace, and other industries. The combination of small-sized nanoparticles with high dispersibility in fluids offers significant potential for achieving high efficiency, energy savings, and cost reduction in thermal management systems [143].

In 1995, researchers developed nanofluids and studied their thermal properties. Their results showed that adding copper nanoparticles to water increased its thermal conductivity by 60 % [144].

Furthermore, the distribution of these nanoparticles in the fluid not only enhances thermal conductivity in the two-phase (solid/liquid) medium but also prevents issues such as particle sedimentation and clogging of pipes, which can occur in fluids containing micron-sized particles [143].

In recent years, numerous gold-based nanomaterials have been developed, such as gold-carbon nanotube composites, gold-polymer composites, gold-graphene composites, gold-biomolecule composites, and gold-metal oxide composites. These various types of gold-containing nanomaterials have been used in a wide range of applications, including sensors, biomedical devices, optical technologies, and the medical field. For example, a novel biosensor for detecting acetylcholinesterase enzyme has been developed using a composite nanostructure of 3-carboxyphenylboronic acid, reduced graphene oxide, and gold nanoparticles (CPBA/rGO-AuNP) on the electrode surface. This biosensor enables highly accurate detection of organophosphorus (chlorpyrifos and malathion) and carbamate (carbofuran and isoprocarb) pesticides. The superior sensitivity of this biosensor is attributed to the excellent properties of the gold nanoparticles and reduced graphene oxide [145].

Metal matrix composites show a crucial role in thermal management for microelectronic devices, such as high-power processors, semiconductors, and microcommunications devices. In this field, common metal matrix composites are typically made of aluminum or copper, reinforced with materials such as silicon carbide (SiC), tungsten (W), beryllium (Be), molybdenum (Mo), aluminum oxide ( $\text{Al}_2\text{O}_3$ ), carbon, or diamond. The primary objective of these composites is to achieve high thermal conductivity with low weight, while also controlling the coefficient of thermal expansion to minimize thermal stress. Some commercially available metal matrix composites are made of indium, reinforced with a mesh of copper, stainless steel, or random glass fibers, and are specifically designed for soldering applications. The reinforcing materials in these composites have a higher modulus of elasticity than the indium matrix, thus enhancing the overall strength [146].

Thermally responsive metal-polymer nanocomposite systems combine the ability of some metal nanoparticles to convert external stimuli into heat with the properties of polymers that exhibit significant changes in their characteristics in response to temperature changes, thus permitting external control of the polymer properties. These systems have been studied for a wide range of biomedical applications, such as drug delivery, cancer treatment, and microfluidic valve control [147].

Gold is known for its high thermal conductivity, which allows for efficient heat dissipation even at low concentrations of AuNPs. The thermal conductivity and thermal expansion coefficients of nanocomposites containing gold particles of various shapes have been studied. Au nanoparticles played an active function in the photo-thermal effect of the nanocomposites, for this reason, gold nanoparticles with different surface morphologies (spherical, aggregated, and rod-shaped) were selected as model additives. To prevent aggregation of the nanoparticles in the solvent (N-methyl-2-pyrrolidone), the thermal conductivity of the polyimide (PI) nanocomposite films increased with increasing aspect ratio (as in the case of rod-shaped particles) or with increased aggregation. The addition of gold nanoparticles also reduced the coefficient of thermal expansion of all types of polyimide films, with the greatest reduction observed for the smallest particle sizes [148].

In the medical field, graphene-coated gold nanoparticles (GGNPs) have garnered significant attention recently due to their distinctive optical properties and high thermal stability. Studies on light absorption efficiency and temperature distribution in cancerous tissues suggest that graphene-coated gold nanoparticles with different shapes (spherical, nano-disc, and nanorods), with their ability to generate substantial heat upon laser irradiation, represent a more

promising option for photo-thermal therapy applications. The researchers studied the effect of the number of graphene layers on tumor temperature and found that the temperature increase increased with the number of graphene layers, the temperature rise for nanorod is greater than others AuNPs (spherical and nano-disc). Gold nanorods have a significantly higher light absorption efficiency than other forms of gold, as all the light energy absorbed by the nanoparticles is converted into heat [149].

The three-dimensional nanocomposite of MoS<sub>2</sub> and gold demonstrated a greater ability to provide active sites on the material surface, as well as enhanced optical and electrical properties, ultimately leading to superior performance in photoelectrochemical applications [150].

The graphene–gold nanocomposite was prepared using an environmentally friendly method, employing ascorbic acid as a reducing agent. This nanocomposite exhibits high efficiency in removing mercury ions (Hg<sup>2+</sup>) from water samples, with an adsorption capacity of approximately 94 %. Furthermore, the nanocomposite can be easily separated from the water sample by simple filtration. The addition of mercury ions in the presence of ascorbic acid causes a noticeable color change in the nanocomposite solution, from purple-red to light brown, enabling a colorimetric detection method for mercury ions [151].

Gold nanocomposites are used as an electrochemical biosensing for food safety, where Islands of the chemisorption material (gold nanoparticles) are fabricated and then combined with a graphite–epoxy composite [152].

Gold nanoparticles have certain limitations in their fluorescent properties, which restricts their use alone in fluorescence detection applications. However, their fluorescent properties can be significantly improved by combining them with other materials to form composite nanomaterials. For example, carbon dots (CDs) and metal–organic frameworks with gold nanoclusters to create composite nanomaterials have attracted considerable attention from researchers. These composite nanomaterials displayed a variety of applications in food safety detection, disease diagnosis, and environmental monitoring [153].

In addition, the nanocomposite containing carbon dots and gold nanoparticles (CD–AuNP) was used to degrade hazardous organic dyes, such as Congo red (CR), methyl orange (MO), and Evans blue (EB), within a time of 10–12 min [154].

It is well mentioned that gold-doped lanthanide NPs (Au–Ln NPs) represent a significant advancement in the field of photodynamic therapy, because of their unique combination of the surface plasmon resonance properties of gold nanoparticles and the upconversion luminescence (UC) properties of Ln NPs. By utilizing near-infrared (NIR) light, these nanocomposite can penetrate deeply into tissues,

making them particularly effective for treating tumors located deep within the body [155].

Gold nanoparticles, in combination with organic Raman molecules and graphene materials, are used to produce a variety of nano-devices that enhance the Raman signal, thus improving detection sensitivity. Graphene–gold nanocomposites are particularly effective in enhancing the Raman signal.

The enhanced surface-enhanced Raman spectroscopy (SERS) signal in nanocomposites composed of graphene and gold nanoparticles is mainly attributed to the synergistic effect of the electromagnetic field enhancement by the gold nanoparticles and the chemical effect of graphene oxide. This synergistic effect, coupled with the unique properties of graphene, provides these nanocomposites with high long-term stability. Compared with individual materials, these nanocomposites can be considered a promising alternative for developing substrates used in surface-enhanced Raman spectroscopy (SERS) applications. As an example, graphene–gold nanocomposite-SERS substrate coated with a vitamin D<sub>3</sub> antibody were used to detect vitamin D<sub>3</sub> by analyzing the characteristic Raman peaks at 1,223, 1,253, and 1,527 cm<sup>-1</sup>. In this regard, the level of vitamin D<sub>3</sub> was detected through its characteristic Raman spectral peak, utilizing the selective binding between vitamin D<sub>3</sub> and a specific antivitamin D<sub>3</sub> molecule. This approach reduces analysis time and enhances the ability to distinguish between different forms of vitamin D [156].

Despite all the successful applications mentioned for nanocomposite materials containing gold nanoparticles, further innovative strategies are needed to broaden the scope of their use, where the high cost of gold remains a major obstacle to large-scale production.

## 1.10 Challenges and future directions

Gold nanoparticles (AuNPs) have recently been used in medicine, electronics, and the environmental field; however, several critical challenges prevent their ideal use.

Inconsistencies in composition lead to differences in the size, morphology, and stability of NPs, which in turn affects reproducibility and scalability. The laboratory methods are considered the main limitation, which often do not meet industrial requirements. Several new approaches such as artificial intelligence-based optimization, microfluidic synthesis, and hybrid nanomaterials showing enhanced control and scalability in production. In healthcare, AuNPs are revolutionizing diagnostics, cancer treatment, and drug delivery; however, cost is still a major concern [157].

Hybrid nanomaterials are a promising and exciting research area in nanoscience and technology. These

interesting materials can be defined as a combination of two or more organic and/or inorganic components [158].

The hybrid materials consisting of metal nanoclusters and carbon nanomaterials are expected to consider a wider range of hybrid materials in the future studies with tunable properties, achieved through careful management of synthetic parameters. For example, one- and two-dimensional carbon nanomaterials are important for electrochemical catalysis and sensing applications when combined with gold nanoclusters [158].

The incorporation of several functional materials into a single composite can help overcome limitations and improve system performance [159].

The challenges in creating hybrids are closely related to the generation of the building blocks themselves and discovering ways to manufacture suitable mixtures from them.

Scalability of Au nanomaterial synthesis is a challenge; however, this challenge being addressed through applying some approaches such as biosynthesis, semi-automated systems, and large-scale chemical reduction to lower costs and minimize environmental effects. Although traditional laboratory techniques are expensive and difficult to scale up, innovations in these alternative approaches require reducing production costs via improving efficiency and reducing waste [160, 161].

In addition, harnessing the numerous fascinating electronic and spectral properties of metal nanoparticles within a stable nanocomposite polymer matrix is considered to be a major problem. On the other hand, despite the importance of nanocomposites, the procedures for dispersing NPs within a polymer matrix to keep them isolated for long time have demonstrated great challenges.

The interaction is the main factor that determines the dispersion of the NPs. It is better for the NPs and polymers to be highly monodisperse, which can be achieved if the polymer is prepared by a living polymerization technique, such as anionic polymerization, and if postsynthesis fractionation of the NPs is performed. The dispersion of polymer-covered AuNPs with high molecular weight (MW) (up to 80,000 g/mol) polymer matrices showed partial dispersion. However, polymer–Au in the polystyrene (PS) matrices in a low-MW PS matrix ( $M_n = 2,000$  g/mol) exhibited complete particle dispersion [162, 163].

When poly (N, N-dimethylaminoethyl methacrylate) was applied as a stabilizing and reducing agent for the synthesis of AuNPs with diverse dispersibility in water and organic solvents, the increase in dispersibility of AuNPs is shown [161].

The thermal conductivity percolation model considers interactions between particles, distinguishes particles and matrices, and utilizes the percolation threshold and

percolation exponent as fitting parameters. In metal/polymer composites, thermal conductivity has not improved as dramatically as electrical conductivity. Consequently, the fitting parameters may be influenced by the experimental data [80].

The influence of aggregation is commonly noted in nanocomposites, and it is an essential element in enhancing thermal conductivity. On the other hand, its complex shape presents a major issue for theoretical modeling and nanocomposite design. Numerous modifications, such as the Hamilton model and modified effective medium approximations, including interfacial resistance, a geometry factor of fillers, and size, have been influenced by the matrix material and filler. Recently, biosynthesis is often used, as it employs advanced models that consider filler aspect ratio, interfacial resistance, and orientation. In contrast, these advanced models focus exclusively on individual particles, ignoring the influence of aggregation [80].

Preventing aggregation is critical, as it can disrupt thermal pathways and reduce conductivity. Advanced surface functionalization techniques are essential, as is manufacturing composites with controlled nanoparticle size and shape at industrial scales, which remains a main challenge. Finally, combining thermal regulation with additional functionalities (such as electrical conduction, physical durability) necessitates precise planning of nanoparticle characteristics.

In the future, highlighting eco-friendly and sustainable synthesis methods for AuNP composites could reduce environmental impact. Using biological or waste materials for the synthesis of AuNPs can create a more eco-friendly and cost-effective process. Extensive studies should be conducted on methods to disperse and concentrate AuNPs in various matrices, as this can enhance thermal management systems. This is particularly valuable in electronics and high-power device applications of the energy, where efficient heat dissipation is critical.

## 2 Conclusions

Gold nanoparticles mixed with other materials, such as nanocomposites, have shown potential to improve thermal conductivity in various applications. Although progress has been achieved, continued research is crucial to address issues regarding dispersion, cost, and scalability and to deepen our understanding of the underlying mechanisms involved. Further advancements in this area may result in significant developments in thermal control technologies. Incorporating gold nanoparticles into a composite allows for the formation of a network that enhances heat transfer efficiency. This is due to AuNPs having high thermal

conductivity, which assisting in more efficient heat dissipation in the composite material. For instance, incorporating gold nanoparticles into a polymer matrix can significantly enhance its thermal conductivity, making it useful for various applications. Additionally, AuNPs and GO/rGO mediums suggest the potential option of functionalizing their surfaces with several suitable ligands, allowing for the creation of multifunctional nanocomposites that have applications in several research fields.

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