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INTENSIFICATION OF THERMAL CONDUCTIVITY AND CONDUCTION HEAT TRANSFER PROCESS IN COMPOSITES WITH THE ADDITION OF FILLER MATERIALS: STATE OF THE ART

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Abstract. *The main characteristics that influence the increase in the thermal conductivity of the composites from the addition of the filling materials were evaluated from the already existing publications. As well as the increase in the heat transfer process from the application of three different composites: silicone, epoxy and gypsum. The factors that increase the thermal conductivity and the heat transfer process of the composites are thermal stability, the distribution of the filler particles in the composite structure for the formation of an improved heat transport chain, the quantity, the size and the shape of the filler particles. In addition, the high aspect ratio is beneficial for increasing thermal conductivity and for the heat conduction process due to the greater contact surface. Compared to gypsum composites, silicone and epoxy matrices enable more significant increases in thermal conductivity without the need of a high filling load. However, the use of silicone and epoxy composites requires greater complexity and cost in production. About the conduction heat transfer analisy, silicone composites was proved to be a promising potential for thermal management applications, whit great heat dissipation and heat absorption performance. The efficiency of removing heat accumulation in electronic devices using epoxy composites was observed as well. Lastly, the gypsum composites could be applied as a building material with high heat storage, in order to increase the thermal efficiency of the structure.*

Keywords: Thermal conductivity, Conduction heat transfer, Composite, Filler particles, Silicone, Epoxy, Gypsum

1. INTRODUCTION

Polymer composites are already widely used in the electronics industry due to their high resistance to corrosion, light weight, ease of processing and low cost (Yanjie Liu et al., 2020). With the addition of suitable filler materials, polymer composites with high thermal conductivity can enable efficient thermal management in many applications, such as electronics, batteries, aerospace devices and LED lighting (Depaifve et al., 2020; Ren et al., 2020; Yetgin et al., 2020).

One of the factors that have more influence on the thermal conductivity of the composites are the filling materials applied to the matrices, the addition of thermally conductive filling particles can significantly improve this thermo-physical property of the composite materials. There are three common types of thermally conductive charges: metals, ceramics and carbon materials. Thermally conductive polymer composites with the addition of metals or carbon materials are used mainly in areas of heat transfer and dissipation where electrical insulation is not necessary, such as heat exchangers (Guo et al., 2020). Expanded graphite (EG) and graphene nanoplatforms (GNP) are the most promising fillers for increasing thermal conductivity in the polymeric composites industry due to their commercial availability, low cost and excellent thermal conductivity (Depaifve et al., 2020).

In these conditions, the present work shows the main characteristics that influence the increase in the thermal conductivity of the composites from the addition of the filling materials. As well as the increase in the heat transfer process from the application of three different composites: silicone, epoxy and gypsum.

Silicone rubbers are stable, flexible, and have a great weather and chemical resistances. They are commonly used in thermal pads and thermal grease. Pure silicone has low thermal conductivity, generally below 0.3 W/mK. Although, their thermal conductivities are improved significantly when filled with high thermal conductive particles (Gao et al., 2015). Epoxy resins are an important class of thermosetting polymers, they are used in different fields of electronic packaging and substrate materials.

Epoxy-based adhesives are used to join dissimilar materials in electronics and aerospace industries. Unfortunately, its low thermal conductivity, around 0.1 W/mK, and high coefficient of thermal expansion limit the direct use in practical applications. However, with the addition of suitable fillers particles it is possible to increase its thermal conductivity and reduce the coefficient of thermal expansion (Yetgin et al., 2020).

Gypsum-based composites are widely used in civil construction because they are not harmful to the environment and have good thermal insulation and fire protection qualities. Gypsum composites are also wide availability and easy manufacture (Du et al., 2020). They are commonly treated with waterproofing materials, cement or slags, or even water reducers and inorganic fillers are used to increase the matrix density, improving their mechanical strength. Nevertheless, the ad of filler particles to improve the thermal conductivity of gypsum-base composites is unusual (Flores Medina and Barbero-Barrera, 2017).

In that way, the objective is to compare these composite materials, gathering information to conclude which of them has the maximum thermal conductivity, which one increases its conductivity the most with the addition of filling materials and which application is most suitable for each. The grouping of this information facilitates the development of new composite materials that can be applied in various thermal management applications from electronic devices, from electronics to building materials.

2. COMPOSITES CHARACTERISTICS

The database used to develop this work was the Science Direct (http://www.sciencedirect.com/). The selection criteria for the papers presented was that the thermal conductivity or the conduction heat transfer in these three materials composites (silicone, epoxy, gypsum) needed to be approached. And it was search for works that used different types of filler particles. According to the number of papers that were found and fitted the addressed topic, epoxy and silicone composites were the ones with the greatest number of used references, 13 and 10, respectively. While only 7 papers cared in research thermo-physical properties of gypsum composites. Which indicates that there is a lot of space in this field of research that must be studied.

The characteristics of the composites that have the greater influence on the thermal conductivity, and also on the heat transfer by conduction of these materials, are the thermal stability of the formed compounds, the treatment or not of the filler particles that are added to the matrix, as well as the quantity, the size and shape of these particles. In this way, Figure 1 published in the work by Fu et al. (2014), indicates the visual difference that epoxy composites can present when adding different types of filling materials. As item (a) is pure epoxy resin, the particles added to the other epoxy matrix from (b) to (i), respectively, are: ZnO (zinc oxide), BN (boronitride), Al_2O_3 (aluminum oxide), graphite, Al (aluminum), Cu (copper), diamond and Ag (silver).

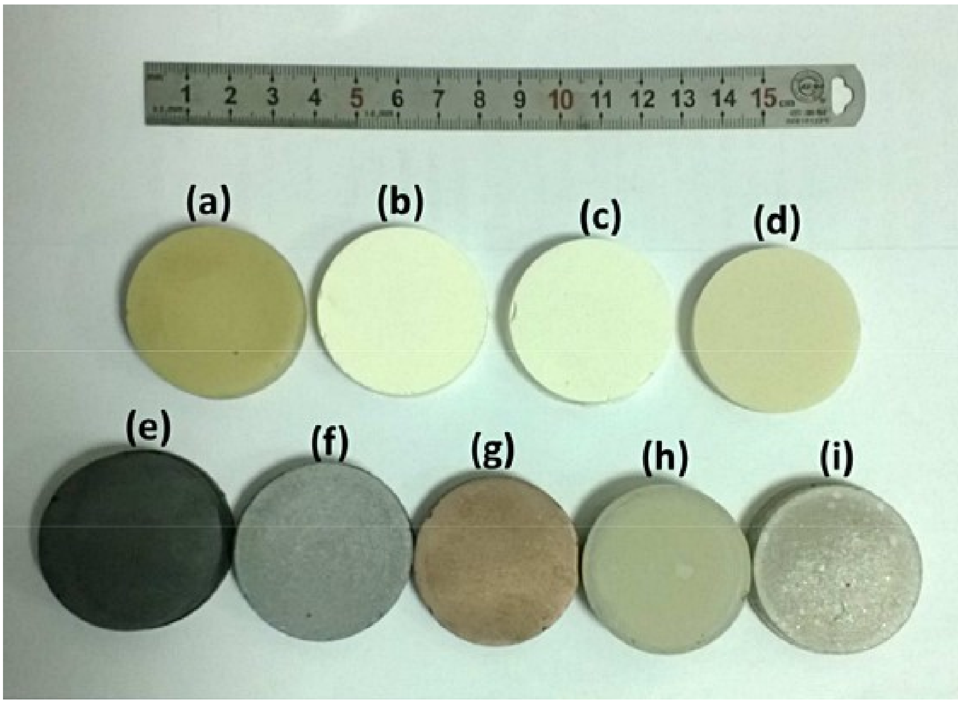

Figure 1. Epoxy composites with different filling materials (Fu et al., 2014).

2.1 Thermal stability and treatment of filler particles

Silicone

The improvement in the thermal stability of silicone composites with the addition of alumina was observed by Liu et al. (2020), the author observed a sample weight loss of less than 2.2 % at high temperatures (400 °C), which indicates good stability in high thermal cycles. In the work of Zhang et al. (2016) and Zhao et al. (2020) an increase in thermal stability was also observed with the addition of filler material, with graphene and boron nitride being added, respectively.

In the work carried out by Chen and Liu (2018), the functionalization of the reduced graphene oxide surface, prepared by graft polymerization, improved the particle distribution in the silicone matrix composite. Some bubbles, cracks and other defects have been described in the composite without functionalization, due to the poor distribution of the filling material, which is harmful to the heat transport. Thus, with the functionalization of the graphene oxide particle, more heat transport chains were formed in the composite. Thus, the heat transport chain benefited the transport of phonons (the main form of energy transport in the composite). Therefore, with the complex network structure of the fill-matrix interface, the formed heat transport chain increased the thermal conductivity of the material.

In another treatment of graphene oxide for silicone composites, the author Ge et al. (2020) showed that the dispersibility of the functionalized particles was excellent and was able to maintain a stable dispersion. This was because the treated graphene oxide surface formed a stable space barrier and a stable dispersion. In this way, the thermal stability of the treated material was significantly better than that of the composite without pre-treated filling material. The thermal coefficient of the modified composite improved and the thermal conductivity increased with increasing temperature.

In the experiments carried out by Song and Zhang (2020) the addition of silicon carbide nanowires (SiCNWs) in the graphene oxide (GO) composite, reduced in silicone, helped to increase thermal conductivity. In addition, the thermal conductivity of the composite with 1.56 vol. % SiCNWs, without graphene oxide, was 1.66 W/mK, while the thermal conductivity reached 2.74 W/mK when the SiCNWs and GO contents were 1.56 vol. % and 0.28 vol. %, respectively. Thus, the authors proved the synergistic effect of both filling materials.

Epoxy

The thermal conductivity stability test of epoxy composites with hybrid carbon fiber particles with Ti_3C_2 was carried out by Guo et al. (2020). In 20 cycles with temperature ranging from 25 to 150 °C, composites with filler material showed better thermal stability compared to pure epoxy. In the work published by Liu et al. (2020) the thermal conductivity of the epoxy resin with addition of graphene and nickel reached 1.6 W/mK when the deposition time of graphene in formation with nickel was 5 min. In the sample in which the graphene deposition time was 45 min, the thermal conductivity of the composite was about 2.7 W/mK, representing an increase compared to pure epoxy greater than 900 %. This significant increase was justified by the fact that, with the prolonged time, more graphene was deposited in the nickel filling, enabling the formation of the 3D graphene network. It has been proven that the 3D filler network was beneficial to improve the thermal conductivity of the polymer. Jing et al. (2020) analyzed that when GO, reduced graphene oxide (rGO) or functionalized SiC particles were used separately as thermally conductive fillers for epoxy resin, the thermal conductivity of the composites was generally low, 0.28, 0.32 and 0.42 W/mK, respectively. However, when using reduced graphene oxide with the addition of SiC as the filling material, the composite reached 1.02 W/mK.

Gypsum

Graphene oxide (GO) was used as an emulsifier to prepare microencapsulated phase change materials (MePCMs) added to a gypsum matrix, in the work published by Zhang et al. (2019). The result of the application of the filling materials was to guarantee high thermal stability and impermeability to the composites. The performance improvement of gypsum composites through the synergistic work of polypropylene fibers (PPF) and additions of isostatic graphite (IG) was studied by (Flores Medina and Barbero-Barrera, 2017). IG is a waste by-product that can increase the mechanical and thermal properties of both simple gypsum and reinforced gypsum-based composites. The volume of polypropylene fibers varied from 0 to 0.6 % in comparison to the volume of the composite and the mass concentrations of graphite tested were 0, 5, 10, 15, 20, 25 %. Thermal conductivity grew with IG in a linear function. The materials without PPF varied the thermal conductivity value from 0.18 W/mK (without adding graphite) to 0.33 W/mK (with the highest mass concentration of graphite). Therefore, with an addition of 25 % IG, an increase of 79 % was achieved. While, in the composites studied with PPF, the increase in IG also produced an increase in thermal conductivity, ranging from 0.17 W/mK to 0.36 W/mK, providing 115 % increase with an addition of 25 % IG.

2.2 Amount of filling material

Silicone

The observation of scanning electron microscopes confirmed that the noticeable increase in the thermal conductivity of the silicone composite can be attributed to a higher surface-volume ratio of expanded graphite (Mu and Feng, 2007). In the analyzes carried out by Zhang et al. (2016) using graphene as the filling material, the rate of improvement of thermal conductivity varied approximately linearly with the mass concentration of graphene. However, when the fill exceeded 0.5 % by mass, the thermal conductivity increased more significantly, which suggests that a heat transport chain was formed. This heat transport chain benefits the transport of phonons, which is the main way of transmitting energy in the composite. Therefore, the authors concluded that with increasing loads, the formed heat transport chain can suddenly increase thermal conductivity.

Tian et al. (2017) analyzed a silicone composite using graphene as a thermally conductive filler in mass fractions of 0.18 %, 0.36 % and 0.72 %. The thermal conductivity of the three compounds increased by 20 %, 40 % and 50 %, respectively, compared to pure silicone. The graphene fraction of 0.36 % by mass was established as the limit value at which the coefficient of thermal conductivity hardly changed when exceeding this quantity, being the value closer to the ideal proportion. It was concluded that the performance of the heat conduction depends on the dispersion of the graphene and the density of the chain of the heat conduction network inside the silicone.

Both thermal conductivity and tensile strength can be greatly improved by increasing the amount of filler material (Zhao et al., 2020). As the load of thermally conductive fillers increases, thermally conductive fillers begin to come into contact with each other and form thermal conduction paths or networks (Guo et al., 2020).

Liu et al. (2020) observed that with the increase in the load of hybrid Al_2O_3 from 60 vol. % to 80 vol. %, the thermal conductivity of the silicone/Al_2O_3 composites improved from 1.67 W/mK to 2.92 W/mK, being 15 times greater than that of pure silicone resin (0.19 W/mK). However, as the filling load was increased to 85 vol. %, the thermal conductivity dropped to 2.43 W/mK, which can be attributed to the increase in the number of voids.

Epoxy

According to Chen et al. (2019) the increase in thermal conductivity of a composite material with an epoxy matrix is approximately proportional to the content of fillers in the entire composite. The aggregation of charges increases the thermal conductivity of epoxy composites, as long as it does not lead to the encapsulation of voids (Depaifve et al., 2020). Thus, Yanjie Liu et al. (2020) observed that the addition of Ni foam can prevent the agglomeration of graphene in epoxy resin, ensuring the formation of a thermally conductive network. He et al. (2020) also concluded that oriented graphene can build an effective heat transfer path in composites with a low filler load.

Gypsum

Flores Medina et al. (2016) studied the influence of the increased load of isostatic graphite powder in milled molds for electrical discharge machining (EDM). They observed that the filling material significantly increased the mechanical properties of gypsum pastes, due to the great compatibility of the gypsum microstructure with the graphite microgranules, which fill the microstructure of the pastes and increase the density of the hardened paste. They concluded that the progressive increase in the amount of graphite influenced the properties of the pastes, increasing their density and mechanical strength and reducing their porosity and water absorption. Later, Flores Medina and Barbero-Barrera (2017) also observed that the greater addition of graphite intensified the thermal conductivity of gypsum composites. In another study, Barbero-Barrera et al. (2017) observed that the addition of graphite in gypsum matrix significantly increased the bulk density, thermal conductivity and emissivity of the samples up to 19 %, 97 % and 10 %, respectively. In addition, a 54 % reduction in thermal flows to the outside has been reported with the application of graphite/gypsum board replacing pure gypsum.

Jeong et al. (2017) carried out experiments on gypsum composites with hybrid phase change materials that were submitted to a stabilization process (SSPCM). The analysis indicated that the thermal conductivities of the pure gypsum board and with 10, 20 and 30 wt. % of the filling material were 0.35, 0.43, 0.49 and 0.60 W/mK, respectively. They concluded that the gypsum board with the addition of this phase change material can be used as a heat storage structure to increase thermal efficiency. Table 1 groups the increments in thermal conductivity of the composites, with the variation of the amount of filling particle.

Table 1. Increment in thermal conductivity of the composites with the amount of filling particle.

Author	Matrix	Filling particle	Concentration	Increment
(Tian et al., 2017)	Silicone	Graphene	18 wt.%	20 %
			36 wt.%	40 %
			72 wt.%	50 %
(Liu et al., 2020)	Silicone	Al_2O_3	60 vol.%	780 %
			80 vol.%	1440 %
			85 vol.%	1180 %
(Jeong et al., 2017)	Gypsum	Hybrid SSPCM	10 wt.%	37 %
			20 wt.%	40 %
			30 wt.%	71 %

2.3 Size and shape of the filler particles

Silicone

With respect to a fixed volume fraction, the effective thermal conductivity of the composites increases with the increase in the average particle diameter (Gao et al., 2015). The results obtained by Zhang et al. (2016) indicated that in the silicone composites the graphene particles formed a network structure, serving as a structure that restricted the movement of the silicone molecule. The high specific surface area of graphene resulted in an increase in the contact areas between graphene and silicone, increasing the surface bond strength of these two materials. They deduced that graphenes with a high aspect ratio can more easily form a heat transport chain compared to other carbon materials.

Epoxy

According to Depaifve et al. (2020), the size of the filler particles has a great influence on the thermal conductivity of the composite. It was reported by them that the increase in thermal conductivity follows a saturation trend around an aspect ratio between 500-700. In the experiment carried out by Jing et al. (2020), when analyzing the influence of the size of the filling materials in an epoxy composite with the addition of 30 % mass fraction of reduced particles of reduced graphene oxide (rGO) with silicon carbide, slightly higher thermal conductivity was observed for the composite with larger filler particles, from the nanometer to the millimeter scale.

Wang et al. (2020) obtained increases in thermal conductivity of about 240 % with the addition of 0.5 % by mass of multi-walled carbon nanotubes (MWCNTs) and 65 % with the incorporation of boron nitride nanofoils (BNNSs) with mass fraction of 2 %. They justified the biggest increase with the addition of MWCNTs due to their greater thermal conductivity, and also to the one-dimensional structure of the MWCNTs, which makes their specific surface area much larger than the BNNSs. According to the authors, a high surface area means a high interfacial volume, which increases the chances that the phonons pass through the filling particles in the transmission process, greatly improving the efficiency of thermal transfer. However, greater interfacial volume also leads to more interfacial scattering of phonons, which can reduce thermal transfer efficiency. They concluded that improving the interface between MWCNTs and epoxy resin matrix to reduce the interfacial thermal resistance may allow the nanocomposites to acquire greater thermal conductivity with the same number of nanoparticles.

When studying epoxy composites with carbon nanotubes, Vahedi et al. (2018) analyzed the influence of interfacial thermal resistance between particles of carbon nanotubes (CNT) and epoxy on the effective thermal conductivity. By decreasing the size of the CNTs, the contact surfaces increased, resulting in greater thermal resistance of the interface and leading to less thermal conductivity. Meanwhile, the increase in the diameter of the filler material charges caused the thermal conductivity to increase due to the suppression of the effect of interfacial thermal resistance. They observed that the effect of the aspect ratio was higher for samples with CNTs of larger diameters and for nanocomposites with higher volumetric concentration of filler particles.

Chen et al. (2019) also observed that the aspect ratio of carbon nanofills had a great influence on the thermal performance of polymeric matrix composite materials. By analyzing different carbon particles as fillers, they concluded that graphene in any form can improve the thermal conductivity of the compound more effectively than conventional carbon nanotubes and carbon-based nanofills. Because graphene has higher aspect ratios and lower interfacial thermal resistance. Among the studied particles, black carbon had the least potential for increasing the thermal conductivity of the composite, since it has thermal conductivity and significantly lower aspect ratios.

Contrary to the diameters and aspect ratios of the filling materials, Ren et al. (2020) observed that the thermal conductivities of epoxy composites with graphene and BNNSs increased with decreasing the thickness of the filler particles.

Gypsum

The thermal conductivity is strongly influenced by the mineral composition, the degree of crystallinity, the average grain size, the porosity and the pore shape of the composites. In porous materials, heat transfer occurs in three modes, conduction through the solid skeleton and gas in the pores, convection and radiation through the pores. For the convection heat transfer mode, the pore size must be large. However, as the pore size in gypsum composites is very small, the convection in the pores can be neglected. Therefore, the increase in thermal conductivity of gypsum composites, at high temperatures, must be related mainly to the increase in the quantity and crystallinity of the anhydrite (Du et al., 2020).

3. THERMAL CONDUCTIVITY OF COMPOSITES

Tables 2, 3 and 4 show the thermal conductivity of the pure matrices of silicone, epoxy and gypsum, respectively. In addition to the value reached when adding filler particles and the consequent increase in conductivity. The tables also contain information on the method of measuring thermal conductivity and the concentration of filling material used.

Table 2. Thermal conductivity of silicone composites.

Author	Matrix	Filling particle	Measurement method	Concentration	K_m [W/m.K]	K_c [W/m.K]	Increment
(Mu and Feng, 2007)	Silicone	Expanded graphite	Thermally conductive probe	9 phr	0.24	0.32	33 %
(Gao et al., 2015)	Silicone	Al_2O_3	Thermally conductive probe	0.62 vol%	0.15	2.25	1400 %
(Zhang et al., 2016)	Silicone	Graphene	Flash laser	1.5 wt.%	0.22	2.75	1150 %
(Tian et al., 2017)	Silicone	Graphene	Flash laser	0.72 wt.%	0.20	0.30	50 %
(Chen and Liu, 2018)	Silicone	Functionalized reduced graphene oxide	Transient hot disk	2 wt.%	0.21	1.31	524 %
(Song et al., 2018)	Silicone	Graphene	Thermogravimetric analysis (TGA)	2.53 wt.%	0.229	2.03	786 %
(Zhao et al., 2020)	Silicone	Boron nitride	Flash laser	50 wt.%	0.16	0.88	450 %
(Ge et al., 2020)	Silicone	Functionalized graphene oxide	Analyzer DTC300	20 wt.%	0.150	1.197	698 %
(Song and Zhang, 2020)	Silicone	Silicon carbide and graphene oxide	Flash laser	1.84 vol%	0.17	2.74	1500 %
(Yi Liu et al., 2020)	Silicone	Hybrid alumina	Thermogravimetric analysis (TGA)	80 vol%	0.19	2.92	1437 %

From Table 2, it can be inferred that in most of the studies carried out, small amounts of filling material were added to the silicone matrix. Below 2 % volumetric concentration (Gao et al., 2015; Song and Zhang, 2020), or with mass fractions below 3 % (Zhang et al., 2016; Tian et al., 2017; Chen and Liu, 2018; Song et al., 2018). Even so, very high increments were found, which in some cases made it possible for the thermal conductivity of the composite to be more than 10 times higher than that of the pure matrix, as in the case of the experiments carried out by Zhang et al. (2016), which added a mass fraction of only 1.5 % graphene.

Table 3. Thermal conductivity of epoxy composites.

Author	Matrix	Filling particle	Measurement method	Concentration	K_m [W/m.K]	K_c [W/m.K]	Increment
(Fu et al., 2014)	Epoxy	Graphite	Hot Disk TPS 2500S	44.3 wt.%	0.17	1.68	888 %
(Chen et al., 2019)	Epoxy	Graphene oxide	Hot Disk TPS 2500S	6 wt.%	0.22	1.54	700 %
(Depaifve et al., 2020)	Epoxy	Graphene Nanoplates	Hot Disk TPS 2500S	4 wt.%	0.24	0.77	220 %
(Yanjie Liu et al., 2020)	Epoxy	Graphene-nickel	Transient hot disk thermal analyzer	10.1 wt.%	0.2617	2.6549	915 %
(Jing et al., 2020)	Epoxy	Graphene oxide/SiC	Hot Disk TPS 2200	30 wt.%	0.22	1.02	364 %
(Wang et al., 2020)	Epoxy	BNNS / MWCNT	Flash laser	5 wt.%	0.2	1.8	819 %
(He et al., 2020)	Epoxy	Graphene/SiC	Hot Disk TPS 2200	3.36 wt.%	0.172	0.708	311.6 %
(Guo et al., 2020)	Epoxy	Carbon fiber/MXenes	Flash laser	30.2 wt.%	0.21	9.68	4509 %
(Kim et al., 2020)	Epoxy	Ozone-functionalized nanodiamonds	-	1 wt.%	0.2	0.4	100 %
(Yeom et al., 2020)	Epoxy	Graphene	Flash laser	13.6 vol.%	0.924	44.9	5800 %
(Ren et al., 2020)	Epoxy	Boron nitride graphene/nanoparticles	Flash laser	5 wt.%	0.18	0.43	140 %
(Yetgin et al., 2020)	Epoxy	Hexagonal boron nitride/Al_2O_3	Heat flow meter	30 wt.%	0.19	0.35	84 %

When analyzing Table 3, it is noted that most of the studies carried out with epoxy composites used a small portion of filler particles, in a range between 1 % and about 10 % of mass fraction (Chen et al., 2019; Depaifve et al., 2020; Yanjie Liu et al., 2020; Wang et al., 2020; He et al., 2020; Kim et al., 2020; Ren et al., 2020). Within this range, the increments in thermal conductivity were 2 to 10 times the values obtained with the matrix without the filling material.

The studies carried out with treated particles showed the greatest results, showing increment in a very different order from the others. As in the case of Guo et al. (2020) which obtained an increase close to 50 times of the thermal conductivity of the pure epoxy matrix, with a mass fraction of about 30 % of carbon fiber. And in the study published by Yeom et al. (2020), in which, starting from a matrix with thermal conductivity almost 5 times higher than in the other studies, it reached a conductivity value approximately 60 times higher for the composite with the addition of graphene in a volumetric concentration of about 14 %, resulting in around 45 W/mK.

As can be seen in Table 4, the mass fractions applied to gypsum composites were higher compare to the other matrices (25 or 30%). As well as the increments in the values of thermal conductivity were less significant than with the other matrices. However, an increase of more than 100 % in the value of the pure matrix was observed (Flores Medina and Barbero-Barrera, 2017). In addition, the results were similar for pure gypsum (Jeong et al., 2017; Barbero-Barrera et al., 2017), in the same way that studies in which graphite was added as filling material obtained similar increments (Barbero-Barrera et al., 2017; Barrera et al., 2017; Flores Medina and Barbero-Barrera, 2017). It is worth mentioning that the thermal conductivity in the wet curing condition is superior compared to the dry curing condition. This happens because the samples in the wet curing condition contain water, so that the heat can be transmitted by it (Liu et al., 2017).

Table 4. Thermal conductivity of gypsum composites.

Author	Matrix	Filling particle	Measurement method	Concentration	K_m [W/m.K]	K_c [W/m.K]	Increment
(Jeong et al., 2017)	Gypsum	Hybrid SSPCM	Thermal conductivity analyzer	30 wt.%	0.35	0.60	71 %
(Barbero-Barrera et al., 2017)	Gypsum	Graphite	Relationship between thermal flow	25 wt.%	0.37	0.73	97 %
(Flores Medina and Barbero-Barrera, 2017)	Gypsum	Graphite/polypropylene fibers	Relationship between thermal flow	25 wt.%	0.167	0.360	115 %

Figure 2 composes some of the main thermal conductivity increments from the addition of particles based on graphite or graphene in composites with matrices of silicone, epoxy and gypsum. For all studies presented in the graphic, the material was added according to the mass fraction in relation to the matrix.

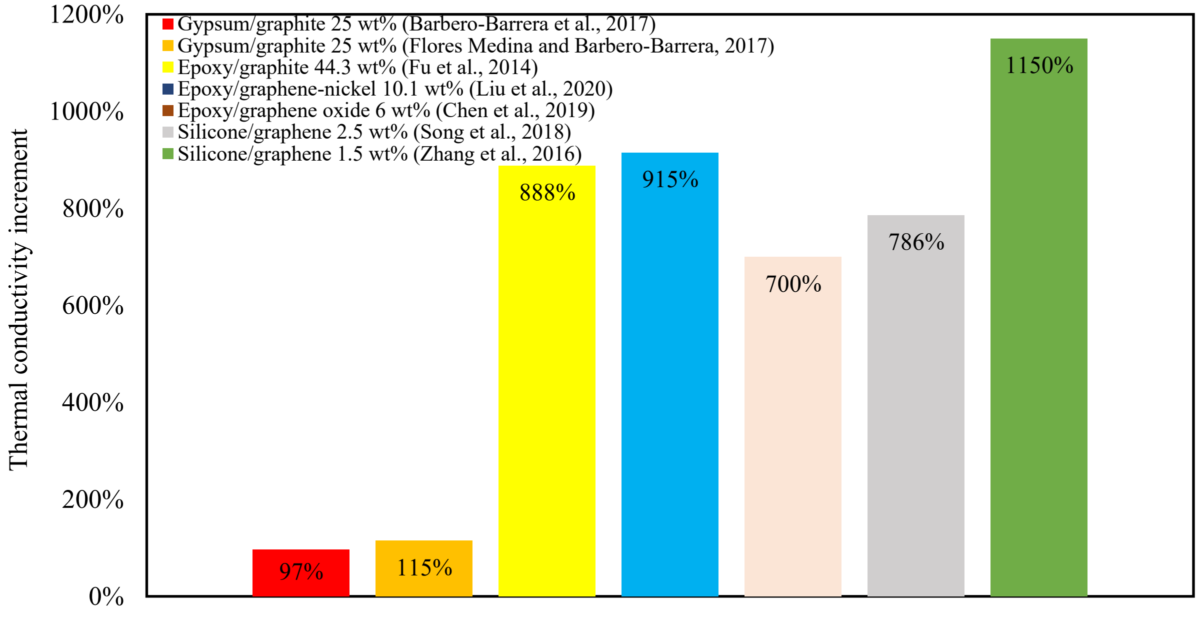

Figure 2. Thermal conductivity increments in silicone, epoxy and gypsum composites.

It can be seen from Figure 2 that the silicone and epoxy composites reach very significant thermal conductivity increments, in the range of 7 to 12 times the value of the pure matrix, when correctly applying the appropriate filler material loads, which do not need necessarily be excessive. Gypsum-based composites, on the other hand, allow lower increments, of about 100 %, even with mass fractions of superior filling material. Thus, it is necessary to know the application in which the composite will be used to analyze the viability of these matrices, if the silicone and epoxy materials will provide benefits that outweigh the greater complexity and cost, or if it is more viable to use gypsum as a matrix, being a much more accessible material, its composites being easy to manufacture.

4. CONDUCTION HEAT TRANSFER IN COMPOSITES

As shown in the previous item, several studies have observed significant increases in the thermal conductivity of composites when adding filler particles. In this way, the analyzes and results regarding the conduction heat transfer process will be presented below, since this property is directly linked to thermal conductivity. As in the recent study published by Wei et al. (2021), in which a composite was manufactured with expanded graphite, stearic acid and polyethylene wax, the impact of adding low density polyethylene particles to the mixture was analyzed. It was found that the maximum thermal conductivity of composites with about 25 % volumetric concentration of EG was 8.6 W/mK. This value was obtained for the common fabrication which resulted in a random distribution of the graphite particles. While, in the case of the composite that was manufactured from a thermal molding to form a 3D matrix network with graphite, the conductivity value reached was 19.6 W/mK. The equivalent of an increase of approximately 6000 % in relation to the pure matrix. In addition, the heat dissipation experiment revealed the high cooling efficiency of the composite and the finite element simulation visually confirmed the excellent heat transfer capabilities.

The improvement of the heat transfer process makes it possible to use these composites in several applications. Thus, the analyzes made for the composites of silicone, epoxy and gypsum, respectively, will be presented.

Silicone

The graphene silicone composite applied to the LEDs reduced the temperature difference between the substrate and the housing. It also improved the heat transfer efficiency of the system. So that with a 1.5 % mass loading of graphene, reaching a thermal conductivity of 1.5 W/mK, the temperature difference between the heat block and the heat sink was less than 2 °C and between the LED module and heat sink was 5 °C (Zhang et al., 2016).

The silicone composite with the addition of boron nitride (BN) was subjected to a heat dissipation test, the specimens were heated at 80 °C for 2 h in an air oven and then transferred to the plate cold steel and thermal insulating foam board, respectively. The rate of heat dissipation increased significantly with the increase in the amount of filler material, due to the proportional increase in the thermal conductivity of the composites. The temperature of the composite surface with a mass fraction of 50 % NB was approximately 30 °C after cooling for 25 seconds, a value significantly lower than that of the silicone elastomer without the addition of filler material, which with the same cooling time reached 65 °C. The faster heat dissipation and the excellent heat absorption performance of the composite demonstrated that the composites have promising potential for thermal management applications (Zhao et al., 2020).

Epoxy

The results obtained by Vahedi et al. (2018) indicated that the interfacial thermal conductance between the carbon nanotube filler particles and the epoxy polymer dominated the heat transfer mechanism on the internal scale of the composite. In another publication, Yeom et al. (2020) proved the efficiency of removing heat accumulation in electronic devices from the application of epoxy composites. Similarly, Ren et al. (2020) observed that epoxy composites with the addition of hybrid nanoparticles of graphene and boron nitride showed improved thermal conductivity and desirable dielectric properties, suggesting their potential for thermal management applications in order to overcome the increasing problem of heat dissipation in devices electronics.

Gypsum

Regarding to gypsum composites, Jeong et al. (2017) proved the high thermal properties of gypsum board with the addition of phase change materials with hybrid composition and stabilized structure. As well as, the feasibility of using these composites in heat storage structures. In addition, they concluded that the plates could be applied as a building material with high heat storage for the formation of structures with similar properties, in order to increase the thermal efficiency of the structure.

5. CONCLUSIONS

The main characteristics that influence the increase in thermal conductivity and the heat transfer process of the composites are thermal stability, the distribution of the filler particles in the composite structure through the formation of an improved heat transport chain, as well as the quantity, the size and shape of these filling materials, the high proportion of aspect being beneficial for increasing thermal conductivity and the conduction process due to the greater contact surface. Among the matrices of silicone, epoxy and gypsum, the two initials allow very significant increases in thermal conductivity without the need for a high filling load. Gypsum-based composites, on the other hand, result in less expressive increases in thermal conductivity. According to the conduction heat transfer in these materials, silicone composites was proved to be a promising potential for thermal management applications, due to its fast heat dissipation and heat absorption performance. Similarly, it was observed the efficiency of removing heat accumulation in electronic devices using epoxy composites. As to the gypsum composites, they could be applied as a building material with high heat storage, in order to increase the thermal efficiency of the structure.

Therefore, to choose which matrix to use it is necessary to analyze whether the use of silicone and epoxy materials pays off, even with the greater complexity and cost of composites formed from these materials, or whether it is more viable to use gypsum as a matrix, being a much more accessible material, its composites being easy to manufacture and which also allows for increases in thermal conductivity compared to pure gypsum. And above all, it is necessary to know which thermal conductivity is most suitable for the application, since from the variation of the matrices and the filling materials it is possible to adjust the properties of the produced material according to what is necessary.

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7. REFERENCES

Barbero-Barrera, M.M., Flores-Medina, N., Pérez-Villar, V., 2017. “Assessment of thermal performance of gypsum-based composites with revalorized graphite filler”. *Constr. Build. Mater*. 142, 83–91.

Chen, J., Gao, X., Song, W., 2019. “Effect of various carbon nanofillers and different filler aspect ratios on the thermal conductivity of epoxy matrix nanocomposites*”. Results Phys*. 15.

Chen, T., Liu, B., 2018. “Improvement of thermal conductivities for silicone nanocomposite via incorporating poly(Γ-methacryloxypropyltrimethoxy silane) grafted graphene fillers”. *Chem. Phys. Lett*. 693, 121–126.

Depaifve, S., Hermans, S., Ruch, D., Laachachi, A., 2020. “Combination of micro-computed X-ray tomography and electronic microscopy to understand the influence of graphene nanoplatelets on the thermal conductivity of epoxy composites”. *Thermochim.* Acta 691, 178712.

Du, Z., She, W., Zuo, W., Hong, J., Zhang, Y., Miao, C., 2020. “Foamed gypsum composite with heat-resistant admixture under high temperature: Mechanical, thermal and deformation performances”. *Cem. Concr. Compos*. 108, 103549.

Flores Medina, N., Barbero-Barrera, M.M., 2017. “Mechanical and physical enhancement of gypsum composites through a synergic work of polypropylene fiber and recycled isostatic graphite filler”. *Constr. Build. Mater*. 131, 165–177.

Flores Medina, N., Barbero-Barrera, M.M., Bustamante, R., 2016. “Improvement of the properties of gypsum-based composites with recycled isostatic graphite powder from the milling production of molds for Electrical Discharge Machining (EDM) used as a new filler”. *Constr. Build. Mater*. 107, 17–27.

Fu, Y.X., He, Z.X., Mo, D.C., Lu, S.S., 2014. “Thermal conductivity enhancement with different fillers for epoxy resin adhesives”. *Appl. Therm. Eng*. 66, 493–498.

Gao, B.Z., Xu, J.Z., Peng, J.J., Kang, F.Y., Du, H.D., Li, J., Chiang, S.W., Xu, C.J., Hu, N., Ning, X.S., 2015. “Experimental and theoretical studies of effective thermal conductivity of composites made of silicone rubber and Al2O3 particles”. *Thermochim.* Acta 614, 1–8.

Ge, T., Zhang, M., Tang, K., Tang, H., 2020. “Diisocyanate-modified graphene oxide/hydroxyl-terminated silicone rubber composites for improved thermal conductivity”. *Mater. Chem. Phys*. 252, 123250.

Guo, L., Zhang, Z., Li, M., Kang, R., Chen, Y., Song, G., Han, S.T., Lin, C. Te, Jiang, N., Yu, J., 2020. “Extremely high thermal conductivity of carbon fiber/epoxy with synergistic effect of MXenes by freeze-drying”. *Compos. Commun.* 19, 134–141.

Guo, Y., Ruan, K., Shi, X., Yang, X., Gu, J., 2020. “Factors affecting thermal conductivities of the polymers and polymer composites: A review”. *Compos. Sci. Technol*. 193, 108134.

He, J., Wang, H., Qu, Q., Su, Z., Qin, T., Tian, X., 2020. “Three-dimensional network constructed by vertically oriented multilayer graphene and SiC nanowires for improving thermal conductivity and operating safety of epoxy composites with ultralow loading”. *Compos. Part A Appl. Sci. Manuf.* 106062.

Jeong, S.G., Chang, S.J., Wi, S., Lee, J., Kim, S., 2017. “Energy performance evaluation of heat-storage gypsum board with hybrid SSPCM composite”. *J. Ind. Eng. Chem*. 51, 237–243.

Jing, H., Hua, W., Qiqi, Q., Zheng, S., Tengfei, Q., Yunsheng, D., Xingyou, T., 2020. "Construction of interconnected SiC particles attached rGO structure in epoxy composites to achieve significant thermal conductivity enhancement". *Mater. Today Commun*. 101584.

Kim, S.H., Rhee, K.Y., Park, S.J., 2020. "Amine-terminated chain-grafted nanodiamond/epoxy nanocomposites as interfacial materials: Thermal conductivity and fracture resistance". *Compos. Part B Eng*. 192, 107983.

Liu, Y., Chen, Z., Shen, Y., Zhou, Y., Wang, D., Lei, Z., Feng, W., Min, Z., 2020. "Silicone-based alumina composites synthesized through in situ polymerization for high thermal conductivity and thermal stability". *Mater. Lett.* 261, 127002.

Liu, Y., Lu, J., Cui, Y., 2020. "Improved thermal conductivity of epoxy resin by graphene–nickel three-dimensional filler". *Carbon Resour. Convers*. 3, 29–35.

Liu, Z., Hu, D., Lv, H., Zhang, Y., Wu, F., Shen, D., Fu, P., 2017. "Mixed mill-heating fabrication and thermal energy storage of diatomite/paraffin phase change composite incorporated gypsum-based materials". *Appl. Therm. Eng*. 118, 703–713.

Mu, Q., Feng, S., 2007. "Thermal conductivity of graphite/silicone rubber prepared by solution intercalation". *Thermochim*. Acta 462, 70–75.

Ren, J., Li, Q., Yan, L., Jia, L., Huang, X., Zhao, L., Ran, Q., Fu, M., 2020. "Enhanced thermal conductivity of epoxy composites by introducing graphene-boron nitride nanosheets hybrid nanoparticles". *Mater. Des*. 191, 108663.

Song, J., Chen, C., Zhang, Y., 2018. "High thermal conductivity and stretchability of layer-by-layer assembled silicone rubber/graphene nanosheets multilayered films". *Compos. Part A Appl. Sci. Manuf.* 105, 1–8.

Song, J., Zhang, Y., 2020. "Vertically aligned silicon carbide nanowires/reduced graphene oxide networks for enhancing the thermal conductivity of silicone rubber composites". *Compos. Part A Appl. Sci. Manuf.* 133, 105873.

Tian, L., Wang, Y., Li, Z., Mei, H., Shang, Y., 2017. "The thermal conductivity-dependant drag reduction mechanism of water droplets controlled by graphene/silicone rubber composites". *Exp. Therm. Fluid Sci*. 85, 363–369.

Vahedi, A., Sadr Lahidjani, M.H., Shakhesi, S., 2018. "Multiscale modeling of thermal conductivity of carbon nanotube epoxy nanocomposites". *Phys. B Condens*. Matter 550, 39–46.

Wang, R., Xie, C., Luo, S., Xu, H., Gou, B., Zeng, L., 2020. "Preparation and properties of MWCNTs-BNNSs/epoxy composites with high thermal conductivity and low dielectric loss". *Mater. Today Commun*. 24, 100985.

Wei, B., zhang, L., Yang, S., 2021. "Polymer composites with expanded graphite network with superior thermal conductivity and electromagnetic interference shielding performance". *Chem. Eng. J*. 404, 126437.

Yeom, Y.S., Cho, K.Y., Seo, H.Y., Lee, J.S., Im, D.H., Nam, C.Y., Yoon, H.G., 2020. "Unprecedentedly high thermal conductivity of carbon/epoxy composites derived from parameter optimization studies". *Compos. Sci. Technol*. 186, 107915.

Yetgin, H., Veziroglu, S., Aktas, O.C., Yalçinkaya, T., 2020. "Enhancing thermal conductivity of epoxy with a binary filler system of h-BN platelets and Al2O3 nanoparticles". *Int. J. Adhes*. 98, 102540.

Zhang, H., Lin, Y., Zhang, D., Wang, W., Xing, Y., Lin, J., Hong, H., Li, C., 2016. "Graphene nanosheet/silicone composite with enhanced thermal conductivity and its application in heat dissipation of high-power light-emitting diodes". *Curr. Appl. Phys*. 16, 1695–1702.

Zhang, Y., Wang, K., Tao, W., Li, D., 2019. "Preparation of microencapsulated phase change materials used graphene oxide to improve thermal stability and its incorporation in gypsum materials". *Constr. Build. Mater*. 224, 48–56.

Zhao, L., Shi, X., Yin, Y., Jiang, B., Huang, Y., 2020. "A self-healing silicone/BN composite with efficient healing property and improved thermal conductivities". *Compos. Sci. Technol*. 186, 107919.

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