

Investigation of mechanical properties of Copper-Graphene composites in terms of production methods and additive ratios: A review

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ABSTRACT

Copper (Cu) is a ductile material with excellent electrical and thermal conductivity. It is widely used in many industries including automotive, electronics and electricity. However, the mechanical properties of copper are relatively poor. Graphene or graphene nanoplatelets (GNPs) have outstanding properties such as high strength, high young modulus, and large surface area. In this way, they significantly change the mechanical properties when used as reinforcement in metal matrix composites. In particular, in the field of powder metallurgy (PM), the properties of metallic matrix composites produced with these two materials are still under study. In the production of powdered metal components, the type of additive is important in terms of production cost. As the proportion of additives in the manufactured part increases, the production cost will increase accordingly. This study aims to determine which fabrication methods are used to obtain the highest mechanical properties values with the lowest amount of graphene contribution for Cu-GNP composites. The percentages of additives used in the studies are indicated together with the consolidation and mixing methods to prove the above-mentioned purpose. Thus, it has been determined by which production methods the studies with the highest percentage increase in mechanical properties were produced by using the optimum additive ratio for Cu-GNP metal matrix composites. In this regard, the highest hardness value was obtained with 118% increase percentage, by High pressure torsion method. In another study, Electro-co-deposition method were applied. As a result, the highest tensile strength value increased by 110%. The highest increase in yield strength value was obtained by Spark plasma sintering method with 239%. In addition, the effects of different additives were also examined. Other inferences from the studies are given in the result and discussion section.

Keywords: Copper, GNP, Composite, Sintering, Powder metallurgy, Mechanical properties.

INTRODUCTION

Composite materials are formed by combining more than one type of material. Thus, products with superior properties can be formed from the components that make up them [1]. Most of the equipment used today is the product of a process that covers many different areas of composite material production [2,3]. Thus, they have caused significant changes in human life by bringing about developments in many fields of technology such as microelectronics [4,5], batteries [6–8] and energy [9,10].

Cu-GNP-based nanocomposites have had significant impacts in recent years as they perform relatively well in terms of both physical and mechanical properties [11,12]. They can be used in mechanical switching materials to extend contact life or against the corrosive effects of sea water [13,14]. The components of these nanocomposites also have superior properties individually. Copper, one of the most basic components of the electrical-electronics industry, is a material with very good heat and electrical conductivity. Additionally, its high resistance to corrosion and high alloying feature makes it stand out among metals. Since it is a relatively soft and easily shaped material, it has a wide place in many areas of the production sector [15]. There are also various studies on copper in other scientific research areas [16–19]. The atomic number of copper, which is a transition metal, is 29 and its standard atomic weight is 63.54 [20]. Detailed information about copper is given in Table 1.

Table 1. Approximate value of some properties of copper (Error Range=E.R.=±3) [20–25]

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No	Properties of Cu	Values
1	Purity Degree	99
2	Mass (bulk) density (g/cm ³)	8.94
3	Real Density (g/cm ³)	8.96
4	Resistance ρ ($\Omega.m$)	1.72×10^{-8}
5	Thermal Conductivity ($W.m^{-1}.K^{-1}$)	401
6	Hardness	50 HV
7	Ultimate Tensile Strength	210 MPa
8	Yield Strength	33.3 MPa
9	Modulus of Elasticity	110 GPa
10	Bulk Modulus	140 GPa
11	Poisson Ratio	0.343
12	Shear Modulus	46.0 GPa
13	Melting Point	1083.2- 1083.6 °C
14	Boiling Point	2562 °C
15	Electron configuration	$3d^{10} 4s^1$

Graphene is a single-layer, 2-dimensional material that is formed by the arrangement of carbon atoms in the form of a honeycomb lattice with sp² bonds, and at the same time forms sp² hybridization by establishing sigma bonds with the 3 closest atoms [26–28]. The carbon atoms that make up the structure of graphene are connected by covalent bonds and π bonds. Covalent bonds increase the mechanical properties of graphene, while π bonds contribute to electron conduction [29]. Graphene, like copper, has very good thermal and electrical properties. An important advantage of graphene is its large surface area and its contribution to mechanical features [30–32].

Particles up to 100 nm in size in all dimensions are called nanomaterials. Nanomaterials show different properties compared to other combinations formed by the basic component. Therefore, it draws a lot of attention in terms of production and use. On the other hand, nanocomposites are composites where one of their components is at the nanoscale and can have different dimensions [33–35].

Powder metallurgy (PM) is the technique of producing new and superior materials using powdered metals through various processes. Metal powders of different sizes produced by various techniques in classical powder metallurgy are subjected to a mixing process. The powder mixture, which is made as homogeneous as possible, is compressed by applying high pressure in a prepared mold. Finally, the material is obtained by applying sintering. All stages of this process can change depending on many different variables of the powders used which are particle shape, size, and amount of combination [36].

Graphene shows weak proximity to copper. This prevents an effective interface bonding to be established. Moreover, graphene agglomerates due to Van der Waals forces and powerful π - π bonds. As a result, it is difficult to distribute graphene uniformly in metal matrix composites [37]. Therefore, choosing the mixing method according to the sintering type is of great importance. Magnetic stirring and sonication mixing methods are chosen in most of the studies in which graphene is subjected to various chemical treatments [38–43]. The ball mill method is used in studies where copper and graphene powders are generally formed with flakes and formed with each other [44–54]. Furthermore, mechanical alloying methods also take place in the literature [55,56]. While applying these mixing methods, some chemicals such as stearic acid or ether can be used to prevent agglomeration [30,57]. In some studies, mixing can be carried out in an atmosphere of hydrogen or argon gases to prevent the oxidation of copper [30].

There are many different consolidation methods in powder metallurgy. In the hot-pressing method, heating and compression are carried out simultaneously. Metal powders, which are liquefied by heating, are sintered under a certain pressure applied uniaxially [38,56,58–61]. The spark plasma sintering method is the consolidation of the material in a graphite mold in a certain vacuum environment by applying pulsed DC or AC. In this method, when the current flows directly over the material to be applied, a spark discharge will form and this will cause rapid heating. Therefore grain growth may occur [62–68]. Electrodeposition is based on the principle of depositing the desired metal

on the cathode electrode by applying DC or pulsed DC between two electrodes in an environment containing ions or chemical solution of the desired metal using an electrical source [69].

Although graphene provides many positive contributions to copper composites when used as an additive material, some problems have also been encountered. These problems are usually caused by the difference in some physical and chemical properties between copper and graphene. Copper and graphene are not very compatible in terms of interfacial wettability. In addition, due to the weak affinity between graphene and copper, strong interfacial bonding is very difficult. The density difference between the two materials is also one of these problems. Another issue that may occur during the production of composite is that the graphene may be damaged. For these reasons, it is very difficult for copper and graphene to form covalent bonds and graphene may show an agglomeration tendency during metal composite production. Some articles in the literature have suggested various methods to overcome these problems [28,30,51,70].

Ali et al. [71], have investigated the factors affecting the thermal conductivity of Cu-GNPs composites and the improvement of thermal conductivity by changing these factors. Hidalgo-Manrique et al. [37], have explained the mechanical, electrical, thermal and tribological properties of graphene-reinforced copper composites in detail by revealing the effects of different production methods. Güler et al. [72], have studied the mechanical properties of graphene reinforcement in metal matrix composites and the factors influencing these properties. Zhao et al. [73], have demonstrated the mechanical and functional properties of graphene composites in a comprehensive study. Iqbal et al. [74], have examined graphene synthesis techniques and their advantages. Furthermore, they have demonstrated the effects of graphene reinforcement in the production of nanocomposites of various structures.

Most of the studies have focused on the production of materials with better properties. Additionally, related studies have been presented in detail in the literature review. The objective of this research, considering the results of the studies in the literature, is to determine the production methods of the Cu-GNP composites produced with the lowest additive ratio, which have the highest yield strength, hardness and tensile strength properties. In addition, the effects of different production and mixing methods in terms of these properties are discussed based on the studies examined for each mechanical feature. Thus, the importance of a cost-effective perspective has been emphasized to researchers interested in the subject.

MATERIAL AND METHODS

In this article, studies dealing with these concerned three mechanical properties are examined in detail and given in Table 2. Among such studies, there are also includes studies with the highest percentage increase for the relevant feature in the literature. The studies with the best results will be selected from Table 2 for each mechanical property. Thus, the results of the studies that reached the best values in the literature will be examined.

Table 2. Results of studies on GNP reinforced copper composites.

No	Route	Mixing type	Reinforcement rate of the best result (%)	Results	Reference
1	Hot pressing	Ball milling	Cu-0.5 wt.% 3D-graphene	Yield strength =290 MPa Tensile Strength =308 MPa	[75]
2	Hot pressing + Hot Rolling	In situ growth of graphene on Cu milled powders	Cu-2.5 vol% graphene	Young's modulus = 135 GPa Yield strength =200 MPa Tensile Strength =378 MPa	[76]

3	Melt casting + Hot-rolling	Electromagnetic stirring	Cu- 0.1 wt% GNPs -3 wt% ZrB2	Tensile Strength =426 MPa	[77]
4	Electro-co-deposition + Powder metallurgy	Sonication	Cu-2.57 wt.% GNPs	Hardness =105 HV	[11]
		+		Yield strength =142 MPa	
		Stirring		Tensile Strength=282 MPa	
5	Hot pressing	Ball milling	Cu-8vol % GNPs	Young's modulus = 104 GPa	[78]
				Yield strength =314 MPa	
6	High Pressure Torsion (HPT)	Mechanically mixed	Cu-10 wt.% Gr	Hardness =2.67 GPa (~ 272.3 HV)	[79]
				Young's modulus = 102.03 GPa	
7	Microwave sintering	Pestle and mortar + Cold pressing	Cu-3.6 vol% Gr	Hardness =89 HV	[80]
8	Spark plasma sintering	Molecular level mixing	Cu-0.6 vol% GNPs	Hardness =1.75 GPa (~ 178.4 HV)	[39]
				Young's modulus =135 GPa	
				Yield strength =310 MPa	
9	Pulse reverse electrodeposition	Sonication	Cu-1.3 wt.% GNPs	Hardness =2.3 GPa (~ 237.6 HV)	[81]
				Young's modulus = 127.5 GPa	
10	Spark plasma sintering	Molecular level mixing	Cu-1.3 wt.% GNPs	Young's modulus = 104 GPa	[77]
				Yield strength =363 MPa	
				Tensile Strength =485 MPa	
11	Spark plasma sintering	Molecular level mixing	Cu-0.5 wt.% GNPs-TiC	Tensile Strength =420 MPa	[82]
12	Spark plasma sintering	Molecular level mixing	Cu-0.1 vol% GNPs	Tensile Strength = 315 MPa	[83]
13	Spark plasma sintering	Sonication	Cu/0.8 vol%	Tensile Strength =245 MPa	[84]
14	Cvd + Hot Pressing + Hot Rolling	None	Cu-12 vol% GNPs	Yield strength =256 MPa	[76]
15	Hot pressing	Stirring	Cu-0.3 wt.% GO	Hardness =52 HV	[85]
				Tensile Strength =237 MPa	
16	Hot pressing	In situ growth of graphene on Cu milled powders	Cu-0.4 wt.% Gr	Hardness =131 HV	[86]

				Tensile Strength =251 MPa	
				Yield strength =103 MPa	
17	Spark plasma sintering	Ball milling + Electrostatic self-assembly + Electroless copper plating	Cu-0.2 wt.% GNPs	Yield strength =195 MPa	[87]
				Tensile Strength=274 MPa	
18	Hot pressing	Ball milling	Cu-0.2GNPs-0.5Co	Microhardness=72.0 HV	[52]
19	Hot pressing	High-speed mixer		Yield strength =145 MPa	[88]
				Tensile Strength =253 MPa	
20	Hot isostatic pressing (HIP)	Wet mixing method	Cu-8.0 vol % Percent GNPs	Hardness =62.3 HV	[89]
				Tensile Strength=251 MPa	
21	Spark plasma sintering	Ball milling	0.8wt%MWCNTs+ 0.2 wt% GNPs	Tensile Strength =103.63 MPa	[90]
22	Powder injection molding	Ultrasonication	0.1 wt% Gr	Hardness =46.9 HV	[91]
23	Spark plasma sintering	Ball milling	1.0 vol% GN	Yield strength =128.6 MPa	[92]
				Tensile strength=288.6 MPa	
24	Direct current electro-deposition	Magnetic stirring	0.4 g/L GO	Hardness =3.32 GPa (~338.5HV)	[41]
				Young's modulus=201.57 GPa	
				Ultimate tensile strength=280 MPa	
25	Spark plasma sintering	Wet mixing	0.70 wt.% Gr	Yield strength =132 MPa	[93]
				Tensile strength=252 MPa	
				Hardness =95 HV	
26	Electric field-activated pressure-assisted synthesis (FAPAS)	Ball milling	0.5 wt.% Gr	Hardness =85 HV	[47]
				Yield strength =122 MPa	
				Tensile strength=270 MPa	
27	Spark plasma sintering	Vibration mixing	0.3 wt.% GNPs+2.0 wt.% Ti	Yield strength =295.6 MPa	[94]
				Tensile strength=415.8 MPa	
				Hardness =81 HV	

According to many studies examined, it can be concluded that the factors that have the greatest impact on the results are the difference in material selection and production method. Therefore, it will be useful to mention the production methods in some studies.

Chu et al. [78], investigated the mechanical properties of Cu-GNPs composites and the details of the production method. They obtained the graphene which they used in their studies according to the modified Brodie's method [95]. They used the ball milling mixing method for the homogeneous distribution of graphene. Samples were produced in various proportions by volume. The weight ratio of the produced samples to the balls is 1:5. The powders were mixed in an argon atmosphere for 3 hours at a rotational speed of 1200 rpm and ether was used for process control. Bulk composite powders were sintered at 800 °C for 15 minutes. This process was carried out by applying a pressure of 40 MPa with a heating rate of 500 °C min⁻¹. Chen et al. [86], presented a new method in which they obtained graphene on copper powders and used PMMA (Polymethyl methacrylate) as the graphene source. In their work, they focused especially on energy efficiency and structural durability. Three separate samples with copper and PMMA mass ratios of 10:0.1, 10:0.2 and 10:0.3 were produced. First, copper powder and PMAA powder were mixed in a 150 g stainless steel ball mill with Argon (Ar) gas at 400 rpm for 2 hours. PMMA/Cu composite powders were calcined for 10 minutes in a quartz tube furnace containing Ar (200 ml/min) and H₂ (100 ml/min) gases at 800 °C and cooled to room temperature rapidly. Composite powders were produced by the hot pressing sintering method for one hour by applying 50 MPa pressure at 800 °C under a vacuum (10⁻⁴ MPa) in a graphite mold. Inspired by the natural mother-of-pearl structure, Cao et al. [76], developed a new method to improve the decreasing ductility and electrical conductivity properties after the hardening process. In their studies, they emphasized the importance of architectural design in improving the structural properties of metal composites. There is a 1:20 mass ratio between copper powders and stainless-steel balls. Copper powders were mixed in ethanol for 5 hours by ball milling method. Copper powders flaked in this way were mixed with 0.05-0.5 wt.% PMMA anisole solution and turned into a slurry. This slurry was stirred for 12 hours and then centrifuged at 4000 rpm for 10 minutes. The PMMA/Cu flakes obtained by this method were dried in the oven at 85 °C for 2 hours and removed from the solvent. The produced composite powders were obtained by heating for one hour in a quartz tube furnace containing Ar (400 sccm) and H₂ (100 sccm) gases at 900 °C and were cooled to room temperature rapidly. Composite powders were produced by the hot pressing sintering method for 20 minutes at a heating rate of 15 °C/min by applying 50 MPa pressure at 900 °C in a graphite mold under Ar atmosphere. Hot pressed composites were hot rolled at 850 °C. Shengcheng et al. [88], recommended using a nanocellulose gel (NCG) assisted production method for the homogeneous dispersion of graphene on copper. Because this method is simple, inexpensive, and efficient, it can be used in the mass production of copper-graphene composite material. 20 g of copper powder and 2 g of 0.65% NCG were mixed with a high-speed mixer at a rotational speed of 3500 r/min. Thus, copper powders were homogeneously coated with gel. 0.5 g of GNPs was also added to the mixture by a similar method. Thanks to NCG, GNPs were able to adhere to copper particles. After the water at 60 °C was evaporated in 2 hours by applying vacuum, the produced composite powders were heated at a heating rate of 15 °C min⁻¹ in a quartz tube furnace containing Ar (100 sccm) and H₂ (30 sccm) gases and the temperature of which was at 800 °C. Then, the annealing process was carried out under similar atmospheric conditions at 850 °C for 2 hours by keeping the temperature constant. After that, the prepared particles were cooled at a cooling rate of 35 °C min⁻¹. Composite powders were produced by the hot pressing sintering method for 20 minutes at a heating rate of 25 °C min⁻¹ by applying 40 MPa pressure at 850 °C in a graphite mold under an Ar atmosphere. Fanyan et al. [39], formed composites using molecular level mixing process and spark plasma sintering methods by preventing agglomeration of graphene. They also examined the effect of graphene content on various material properties. To suspend graphene in the solution containing copper ions, it was stirred in the alcohol solution. Then, sonication was continued by adding liquid C₆H₁₂O₆ and liquid NaOH, respectively. The resulting mixture was kept in the oven for 4 hours. Thus, precipitate powders were obtained. These were washed with 50 vol.% ethanol solution, filtered and then dried. Finally, the obtained powders were reduced to 300 °C for 3 hours. Composite powders containing graphene in different proportions were put into molds and sintered at 700 °C in a vacuum environment by spark plasma sintering (SPS) method. The consolidation pressure was initially applied at 40 MPa and then at 50 MPa in the cooling section. Ke et al. [87], used electrostatic self-assembly and electroless plating methods together in this study. Thanks to the electrostatic self-assembly method, GNPs were homogeneously absorbed by the copper powders. Thus, the interfacial strength was increased by homogeneously dispersing the graphene in the prepared composite. Modified Hummer's method was used to obtain GO liquid solution. The liquid GO solution was mixed first with SnCl₂ and then with PdCl₂, and the pre-treated GO solution was prepared. After the copper powders were ball milled at 300 rpm for 4 hours, they were placed

in 0.5 wt.% cetyltrimethyl ammonium bromide (CTAB) solution for positive electrical charge. Thus, a pre-treated Cu slurry was obtained. The pre-treated GO and Cu slurry obtained were mixed. This mixture $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ was added to the solution and mixed in a water bath at 80 °C for 30 minutes. Finally, composite powders were obtained by heating the obtained powders at 500 °C for 2 hours under H_2 and Ar atmosphere. Composite powders were sintered by spark plasma sintering method by applying at 35 MPa pressure at 600 °C. Ajay et al. [11], used the electro-co-deposition method to obtain Cu-GNPs composite powders in this study. Classical powder metallurgy methods were used for bulk material production. Apart from the main components of the study Cu and GNPs, there are also copper sulfate pentahydrate ($\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$) and sulfuric acid (H_2SO_4). First, GNPs and $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ aqueous solution were prepared, then H_2SO_4 was added to the solution for pH balancing. The prepared solution was mixed both ultrasonically and magnetically to prevent aggregation and to distribute GNPs homogeneously. As a result of the process, the composite powders accumulated at the end of the cathode electrode. The powders were filtered and dried in an inert environment. The obtained composition was compressed in the mold using a hydraulic press. The compacted samples obtained were sintered in the Ar environment in the furnace at 950 °C for 2 hours and cooled at room temperature.

The structural properties of the powders preferred in the above-mentioned studies and the chemicals used in the production process are shown in Table 3.

Table 3. Properties of powders and chemicals

No	Structure of Powders	Process Chemicals	Reference
1	Cu powders are 400 mesh and PMMA powders are about 80 μm in diameter and both are 99.9% pure.	PMMA (Polymethyl methacrylate)	[86]
2	The average particle size of Cu powders is 40 μm and they have a purity of 99.9%. PMMA (Polymethyl methacrylate) was used as carbon source.	Solvent ($\text{C}_6\text{H}_{14}\text{O}_2$), Ferric chloride (FeCl_3), Ethanol ($\text{C}_2\text{H}_6\text{O}$)	[76]
3	Copper particles are 20 μm in diameter. 0.65% nanocellulose gel (NCG) was used.	None	[88]
4	GNPs are on average 2.4 mm thickness.	The alcohol solution of Cupric nitrate trihydrate ($\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$) and $\text{C}_6\text{H}_{12}\text{O}_6$ aqueous liquid solution.	[39]
5	GO aqueous solution was obtained by Hummer method.	Graphene oxide (GO) aqueous solution, Tin (II) chloride (SnCl_2) and Copper (II) sulfate pentahydrate ($\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$)	[87]
6	Surface area 500 m^2/g and 90% purity GNPs were used.	Sulfuric acid (H_2SO_4) and Copper (II) sulfate pentahydrate ($\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$)	[11]

RESULTS

In this section, five studies with the best change rates according to hardness, tensile strength and yield strength properties among the articles given in Table 2 have been examined separately and the results are discussed.

HARDNESS PROPERTIES

In Table 4, the hardness change rate has increased in almost all the studies given. To obtain the best hardness value, the GNP contribution rates differ in the studies.

Table 4. Best five results of Hardness.

No	Route	Mixing type	Reinforcement rate of the best result (%)	Hardness	% +	Reference
1	High Pressure Torsion (HPT)	Mechanically mixed	Cu-10 wt.% Gr	Hardness =2.67 GPa (~ 272.3 HV)	118%	[79]
2	Microwave sintering	Pestle and mortar + Cold pressing	Cu-3.6 vol% Gr	Hardness =89 HV	93%	[80]
3	Spark plasma sintering	Molecular level mixing	Cu-0.6 vol% GNPs	Hardness =1.75 GPa (~ 178.4 HV)	75%	[39]
4	Electro-co-deposition + Powder metallurgy	Sonication + Stirring	Cu-2.57 wt.% GNPs	Hardness =105 HV	55%	[11]
5	Pulse reverse electrodeposition	Sonication	Cu-1.3 wt.% GNPs	Hardness =2.33 GPa (~ 237.6 HV)	54%	[81]

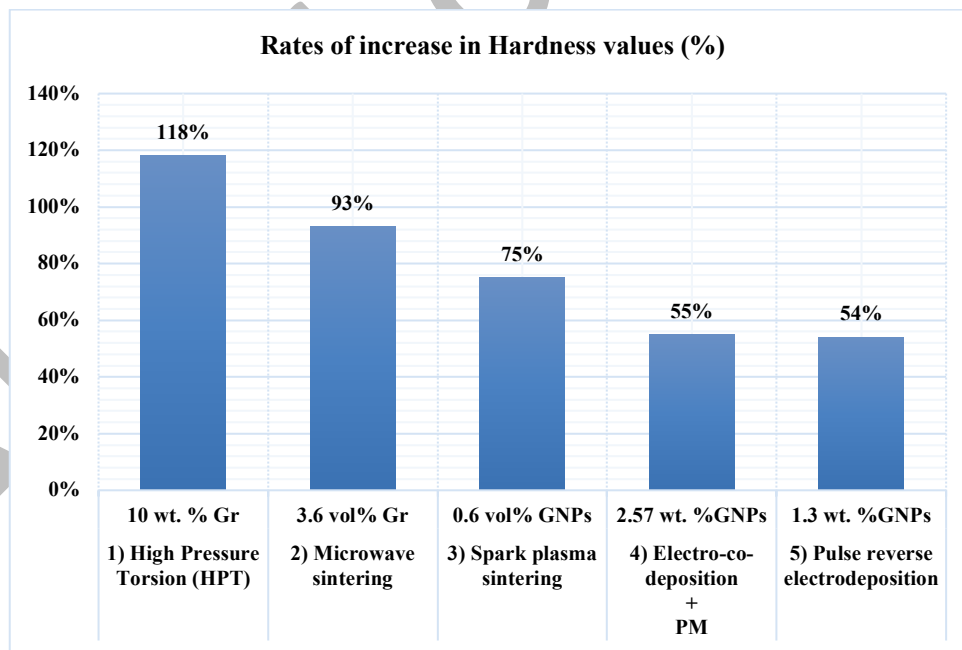


Figure 1. Rates of increase in hardness values

According to the values in Table 4 and Figure 1., Khobragade et al. [79], achieved a 118% increase in the hardness value of the sample sintered by the High-Pressure Torsion (HPT) method, using 10 wt.% graphenes. The contribution of graphene to the microstructure and its mechanical strengthening effect affected the result Graphene

improves material strength by preventing dislocation motion. In addition, the particles bond as a result of the strong axial compression force and torque used during powder consolidation. Ayyapadas et al. [80], obtained a 93% increase in the hardness value of the sample sintered by microwave sintering when they used 3.6 vol%. graphenes. Graphene may aggregate at grain boundaries if it is not uniformly distributed in metal matrix composites. In this case, the hardness value may decrease. However, in this study, the GNP is uniformly distributed. Microwave sintering produces a significantly more homogeneous microstructure because of the extremely quick heating that takes place during the process. This reveals a dispersion enhancement mechanism. This is one of the most important factors affecting the hardness value. The hardness decreases due to graphene aggregation at grain boundaries. To prevent this situation, mixing with pestle was applied and the mortar method was used. Moreover, the main reinforcement mechanism responsible for the increase in hardness values of copper-graphene composites is dispersion strengthening. Twin borders occur in microwave sintered samples. These boundaries change the crystal direction along the interface, and this causes the shear system discontinuity, thus strengthening the material. The unique properties of microwave sintering have played a role in the hardening of the Cu-GNPs composites. Chen et al. [39], have seen a 75% increase in the hardness value of the sample sintered by spark plasma sintering when they used 0.6 vol% GNPs. There is a large difference in thermal expansion rates between copper and graphene. Therefore, a plastic zone formation is observed in Cu-GNPs composites. As the grain size decreases, the grain boundary increases. Thus, a resistance to the dislocation motion is created. The CU-O bonds formed during molecular mixing are useful for increasing load transfer. Pingale et al. [11], achieved a 55% increase in the hardness value of the sample sintered by the electro-co-deposition method, using 2.57 wt.% GNPs. The most significant factors contributing to the increase in mechanical strength of composite materials are the uniform distribution of GNPs and the robust interfacial bonds in Cu-GNPs composite. The study claims that a rise in the grain boundary is seen because grain refinement is brought on by the input of GNPs. This increase might stop the movement of grain dislocation. As a result, the hardness rises. Pavithra et al. [81], using 1.3 wt.% GNPs additive, acquired a 54% increase in hardness value in the sintered sample by pulsed reverse electrodeposition. In this method, if the pulse parameters and current density are well designed, the graphene is well distributed in Cu-GNPs composite and this makes a significant contribution to the hardness value. Adjusting the current density from the electrolysis parameters affected the hardness values. Especially creating a forward pulse or reverse pulse current and optimizing the on-off times increased the hardness values.

Although the highest increase rate and hardness value are in study 1, it has a high contribution rate with 10% graphene contribution.

In Study 3, 1.75 GPa is reached with the lowest GNPs contribution ratio compared to the others.

Although the lowest increase rate is in Study 5, it has the second highest hardness value.

TENSILE STRENGTH PROPERTIES

Among the studies in Table 2, the 5 studies with the best tensile strength values are shown in Table 5.

Table 5. Best five results of Tensile Strength.

No	Route	Mixing type	Reinforcement rate of the best result (%)	Tensile Strength (MPa)	% +	Reference
1	Electro-co-deposition + Powder metallurgy	Sonication + Stirring	Cu-2.57 wt.% GNPs	Tensile Strength=282	110%	[11]
2	Spark plasma sintering + Electroless plating	Molecular level mixing	Cu-1.3 wt.% GNPs	Tensile Strength =485	107%	[77]
3	Spark plasma sintering	Vibration mixing	0.3 wt.% GNPs+2.0 wt.% Ti		100%	[94]

				Tensile strength=415.8		
4	Hot pressing + Hot rolling	In situ growth of graphene on Cu milled powders	Cu-2.5 vol% graphene	Tensile Strength =378	73%	[76]
5	Spark plasma sintering	Molecular level mixing	Cu-0.5 wt.% GNPs-TiC	Tensile Strength =420	56%	[82]

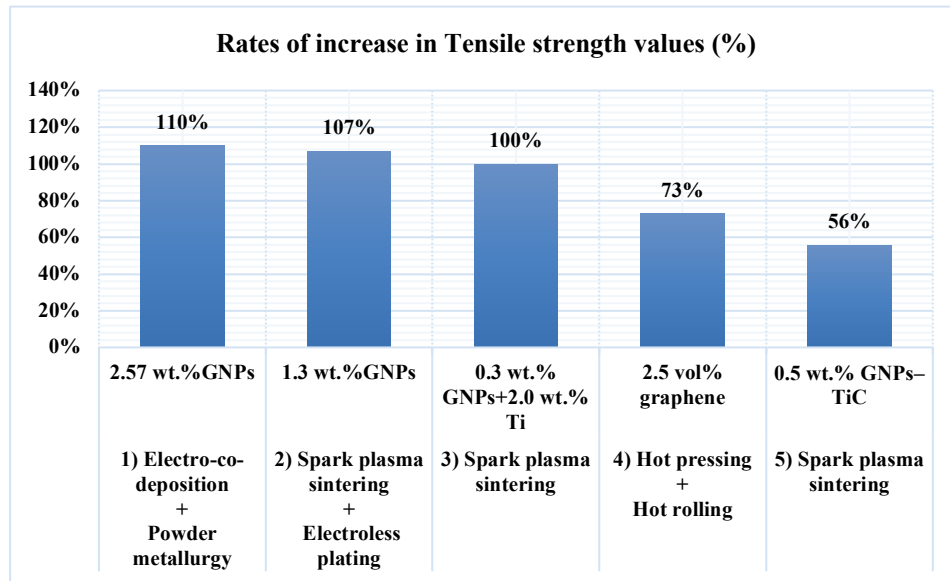


Figure 2. Rates of increase in tensile strength values

According to the values in Table 5 and Figure 2, Pingale et al. [11], achieved an 110% increase in the tensile strength value of the sample sintered by Electro-co-deposition and powder metallurgy, using 2.57 wt.% GNPs. The most important effect on the increase in the tensile strength value in this study is related to the homogeneous distribution of graphene in the composite and the strength of the interface bond in the composite [30]. Enhanced interfacial interaction increases load transfer and tensile strength value. Further, the addition of GNPs to the composite causes grain refinement. Thus, the grain boundaries increase. Grain boundaries can also prevent the movement of dislocations. Zhao et al. [77], achieved a 107% increase in the tensile strength value of the sample sintered by spark plasma sintering (SPS), using 1.3 %wt. GNPs. The ball milling process was not used in this investigation. Graphene can be damaged during ball milling which can impair its mechanical qualities. Furthermore, the link established between the copper and carbon atoms via oxygen increased the interface bonding. As a result, improved load transfer was accomplished. Shi et al. [94], obtained a 100% increase in the tensile strength value of the sample sintered by spark plasma sintering when they used 0.3 wt.% GNPs+2.0 wt.% Ti. The use of Ti-GNPs as an additive in Cu metal matrix composites can be explained by different mechanisms to increase the mechanical properties. There is poor interface affinity between copper and GNPs. The better interface bond of the Ti interface layer naturally strengthens the interfacial shear stress. Thus, the load transfer will be increased. Grain refinement is another factor. The composite will be more refined when the Ti interlayer and GNPs surround the copper grains. Cao et al. [76], accomplished a 73% increase in the tensile strength value of the sample sintered by hot pressing and hot rolling, using 2.5 vol% Gr also they had interfacial bonding between Cu and graphene was increased. A new method has been developed, inspired by the nacreous part of seashells. A building with a brick-mortar architecture greatly increases properties such as

toughness and ductility. The produced copper-graphene flakes were combined in the form of bricks. Instead of the protein layer in seashells, a graphene layer was formed. Si et al. [82], have seen a 56% increase in the tensile strength value of the sample sintered by spark plasma sintering when they used 0.5 wt.% GNPs and TiC. Carbide coating was formed on graphene nanoplatelets (GNPs) using the molten salt treatment method to strengthen the interfacial bond. Thus, the interfacial bonding is strengthened by forming a carbide layer in the gaps between Cu and GNPs. Because early transition metals are very suitable for forming interlayers in terms of bond structures.

Although the highest increase rate of tensile strength is in study 1, the highest tensile strength value is observed in study 2. At the same time, the least graphene contribution is obtained in study 5 by the spark plasma sintering method.

The spark plasma sintering method reaches the highest two tensile strength values.

The contribution of GNPs and TiC or Ti increases the tensile strength value and increases the contribution efficiency.

YIELD STRENGTH PROPERTIES

The best five yield strength values in Table 2 are seen in Table 6.

Table 6. Best five results of Yield Strength.

No	Route	Mixing type	Reinforcement rate of the best result (%)	Yield strength (MPa)	% +	Reference
1	Spark plasma sintering	Vibration mixing	0.3 wt.% GNPs+2.0 wt.% Ti	Yield strength =295.6	239%	[94]
2	Hot pressing	Ball milling	Cu-0.5wt.%3D-graphene	Yield strength =290	233%	[75]
3	Hot pressing + Hot rolling	In situ growth of graphene on Cu milled powders	Cu-2.5 vol% graphene	Yield strength =200	178%	[76]
4	Spark plasma sintering	Molecular level mixing	Cu-1.3 wt.% GNPs	Yield strength =363	133%	[77]
5	Electro-co-deposition + Powder metallurgy	Sonication + Stirring	Cu-2.57 wt.% GNPs	Yield strength =142	129%	[11]

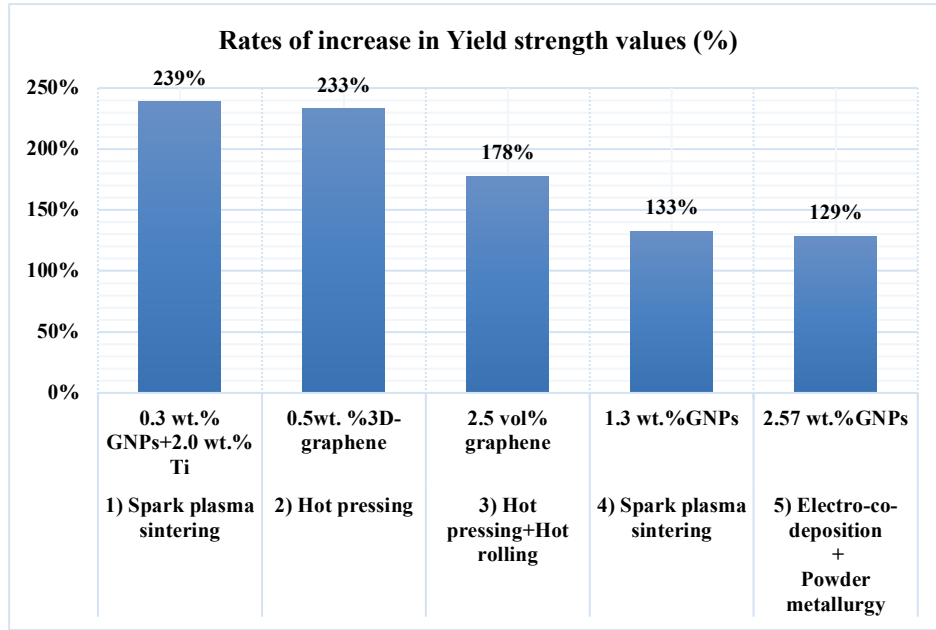


Figure 3. Rates of increase in yield strength values

According to the values in Table 6 and Figure 3, Shi et al. [94], acquired a 239% increase in the yield strength value of the sample sintered by spark plasma sintering, using 0.3 wt.% GNPs+2.0 wt.% Ti. Interaction between dislocations and additives is one of the mechanical strengthening factors. Ti interlayer prevents the dislocations from moving freely and allows them to agglomerate in their borders. In this way, it causes an increase in the densities of dislocations within the grains. Strengthening dislocations is a very important parameter in terms of yield strength. The other important reason for the strengthening effect is that the Ti transition layer creates a strong interface bonding between Cu and GNPs. Thus, a more efficient load transfer reinforcement can be created. Grain refinement has been made possible to the Ti transition layer and GNPs wrapping around the grains. Chen et al. [75], achieved a 233% increase in the yield strength value of the sample sintered by the hot-pressing method, using 0.5 %wt. %3D-graphene. The use of 3D graphene has increased the yield strength property. The number of bonds in the Cu matrix of 3D graphene can improve the toughening property of the composite. In this study, High-ratio Differential Speed Rolling (HRTEM) mixing method was applied to increase the number of these bonds and to distribute the graphene dispersion in the Cu matrix without agglomeration. Another important effect is the interface bonding of the materials that make up the composite. It could be crucial for the interface to be empty and pure to increase this impact. A pure interface was made in this investigation. In this work, 3D graphene served as a barrier to prevent dislocations from spreading. Cao et al. [76], have seen a 178% increase in the yield strength value of the sample sintered by hot pressing and hot rolling when they used 2.5 vol% graphenes. Graphene grown in-situ enhances interface bonding and mechanical properties. The combination of the nanolamination technique and the graphene additive has given a good result in terms of the yield strength property of composite materials. Moreover, the in situ catalytic growth method offers good structural quality and improved interface binding strength. Zhao et al. [77], obtain a 133% increase in the yield strength value of the sample sintered by spark plasma sintering when they used 1.3 wt.% GNPs. According to the study, there is a large difference in thermal expansion rates between copper and graphene. Therefore, a plastic zone formation is observed in Cu-GNPs composites. As the grain size decreases, the grain boundary increases. Thus, a resistance to the dislocation motion is created. Pingale et al. [11], achieved a 129% increase in the yield strength value of the sample sintered by the electro-co-deposition and powder metallurgy, using 2.57 wt.% GNPs.

In all articles in Table 6, the increase rate in the yield strength feature is higher compared to the increase rate in hardness and tensile strength properties.

Study 2 has the lowest contribution rate and the second highest increase rate.

DISCUSSION AND CONCLUSION

This study examined the mechanical effects of graphene contribution according to various production methods among the articles selected from the literature. In this section, many different factors such as temperature, pressure, and powder structure affecting production are ignored and inferences are made only on the results which are obtained. The principle of the work with the least amount of additive material among the studies with the best percentage increase is the most efficient work has been accepted as the basic idea in all inferences. In addition, the studies with the least additive ratio and the highest mechanical property value were also emphasized. The inferences obtained according to the findings are as follows.

- The graphene contribution depends on the production method, the type of mixture, the type of powder used, the temperature and many variables.
- Whatever production method is used, the least graphene contribution for all mechanical properties will be when using GNPs or 3D graphene.
- The amount and cost of additives in graphene-reinforcement composites are very important in terms of production and efficiency. When evaluated for hardness, the best production method is microwave sintering, with an additive rate of 3.6 vol% and an increased rate of 93% in the second study in Table 4. However, it is quite remarkable that the value of 1.75 GPa was reached with the contribution of 0.6 vol% GNPs with the spark plasma sintering method.
- The use of certain tensile stress and pressure during consolidation may be one of the significant factors that improve the hardness characteristic. In addition, the excess amount of graphene additive can affect the rise in hardness.
- Interface bonding and strengthening dislocations are efficient causes for increasing the tensile strength feature.
- When graphene additive and increase rate are evaluated together in Table 5, the spark plasma sintering method was obtained with the best tensile strength value, 1.3 wt.% GNPs contribution and 107% increase rate. However, the lowest contribution rate of 0.5 wt.% GNPs was similarly achieved by the spark plasma sintering method.
- Considering the percentage increase rates in Table 4-6, the highest increase rates were seen in the yield strength feature, although the graphene additive caused an increase in all mechanical properties regardless of the production method.
- When the studies in Table 5 are evaluated, the best percentage increase value was reached by the spark plasma sintering method for the others. Therefore, the spark plasma sintering method can provide more reliable results for a percentage increase rate in tensile strength.
- When the studies in Table 6 are evaluated, the studies showing the best percentage increase value were hot pressing sintering and spark plasma sintering methods. In addition, the lowest additive ratio (0.5% by weight of 3D-graphene) was used in the hot pressing method. The hot pressing method may be considered the most successful technique, and 3D-graphene can be accepted as the most effective additive material because the first research includes extra Ti contribution.
- The use of 3D graphene in Cu-GNP composites reduces the graphene additive rate and also increases the yield strength percentage increase rate. However, the falling graphene amount causes a decrease in the value of the yield strength.
- The highest yield strength (363 MPa) and tensile strength (485 MPa) values were reached by the spark plasma sintering method.
- The study using the electro-co-deposition method [11] is available in the hardness, tensile strength and yield strength tables. Therefore, the electro-co-deposition method is the best method to increase all property values.

- Mixing methods are an important part of the process. There is a mixing method suitable for each production method. The molecular level mixing method is preferred in most studies using the spark plasma sintering method. Ball milling was most used in the hot press sintering method.

NOMENCLATURE

GNPs	Graphene nanoplatelets
GO	Graphene oxide
PM	Powder metallurgy
3D	Three dimensional
DC	Direct current
AC	Alternating current
CTAB	Cetyl trime ammonium bromide
PMMA	Polymethyl methacrylate
NCG	Nanocellulose gel
SCCM	Standard cubic centimeters per minute
HPT	High-Pressure Torsion
SPS	Spark plasma sintering
TiC	Titanium Carbide
Ti	Titanium
C ₆ H ₁₄ O ₂	Solvent
FeCl ₃	Ferric chloride
C ₂ H ₆ O	Ethanol
Cu (NO ₃) ₂ .3H ₂ O	Cupric nitrate trihydrate
SnCl ₂	Tin (II) chloride
CuSO ₄ .5H ₂ O	Copper (II) sulfate pentahydrate
H ₂ SO ₄	Sulfuric acid
CTAB	Cetyltrimethylthyl ammonium bromide
PdCl ₂	Palladium(II) chloride
wt.	by weight

vol	by volume
rpm	Revolutions Per Minute
HV	Hardness Vickers
MPa	Megapascal
GPa	Gigapascal
Ar	Argon
H ₂	hydrogen

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