GRAPHENE REINFORCED COPPER MATRIX NANO-COMPOSITE FOR RESISTANCE SEAM WELDING ELECTRODE.

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ABSTRACT

Five samples contain 0, 1.2, 1.4, 1.6, and 1.8 wt. % graphene (GNs) were prepared by the powder metallurgy technique. The 20 (WC-TiC-Co) wt. %, 5wt. %Ni, and the different weight percentages of the graphene (GNs) were coated by copper by the electroless coating process. The effect of the GNs on the density, hardness, compressive strength, and thermal expansion was studied. The microstructure revealed an excellent distribution for the different reinforcements and good adhesion between them. The results indicate that the hardness and the compressive resistance were significantly improved. These improvements were attributed to the remarkable mechanical and physical properties of the GNs. The thermal expansion was decreased by increasing the GNs content.

Keywords

Copper matrix; Hardness; Graphene; Copper electroless; Compression strength.

1. INTRODUCTION

Resistance Seam Welding (RSW) is a kind of resistance welding processes that have advantages of short process time; operator safety because of low voltage, clean, and environmentally friendly. Resistance Seam welding wheel or electrode is subjected to excessive mechanical and thermal stresses during the welding operation. In this process, wheel or electrode achieve three important functions namely; conducting current to work-piece, transmit the proper pressure to the welding zone, and dissipate the heat during the welding process so that it needs to be fabricated from materials with special properties [1, 2].

Copper is one of the most important materials, which is used to fabricate the resistance-welding electrode. It has excellent thermal and electrical conductivity, high corrosion resistance and ease of fabrication, however, it suffers from low tensile strength, low hardness, low coefficient of thermal expansion (CTE) and poor wear resistance. Copper and its alloys are widely used in cables, wires, electrical contacts, and in applications requiring good electrical conductivity [3-5]. To improve the mechanical properties as well as the wear resistance of copper, it must be reinforced with hard particles [6-8]. Mahani Yusoff and Zuhailawati Hussain [10] have investigated the effect of sintering temperature on the density and electrical conductivity of copper- tungsten carbide composite and found that the density increases with increasing the sintering temperature up to 950°C then decreases. Although, reinforcement with WC improves the mechanical properties of copper, physical properties like thermal and electrical conductivities were decreased [9, 11].

Graphene is, basically, a single atomic layer of graphite; it has special properties, which gives it wide range of mechanical, electrical and thermal applications. Adding small amounts of graphene to copper raises the softening temperature substantially, increases strength, and thermal conductivity of copper [13-14]. Generally, wetting and reaction between metals and ceramics is difficult so that the adhesion and the interfacial bonding between them are difficult to obtain. Researchers have tried to overcome these problems by using the electro-less coating process for ceramic powders to produce a wettability layer on the surface of it to facilitate the adhesion between the constituents of the composite [12,15].

In this work, the effect of high graphene content (1.2-1.8 wt. %) on the densification, microstructure, bulk hardness, and compression strength was investigated.

2. RESEARCH PROCEDURES

2.1. Raw Materials

The (WC-TiC-Co) compound that has 0.5µm particle size and 99.97% purity (Hart Metal Company), the nickel powder that has 0.1-0.3µm particle size and 99.9 purity (Aldrich Co.), Graphene layers that have 2-10 nm thickness (Advanced Chemical Supplier ACs materials) are used as reinforcements. Sodium hydroxide and acetone are used to cleaning the surfaces of the (WC-TiC-Co) and GNs from any contamination. Silver nitrate, ammonia, and formaldehyde are used to metalize the surfaces of the reinforcements with Ag. The copper matrix is prepared by the electro-less coating process from a bath contains copper sulfite, potassium sodium tartrate, formaldehyde, sodium hydroxide, and ammonia. All chemicals were purchased from El-Nasr Company, Cairo, Egypt.

2.2. Powder Composites Preparation

In order to coat the reinforcements with the nano-copper layer that is the matrix, they are firstly cleaned and then metalized with the Ag. For cleaning the (WC-TiC-Co) and GNs, they are stirred in 10 wt. % sodium hydroxide and then in acetone for 1 hr., respectively. The powder is washed and dried in an electrical furnace at 80°C for 3 hr. The metallization process for the powder is established from a solution bath contains 2 g/l silver nitrate, 250 ml formaldehyde, and pH=11.5. In this step, the silver nitrate is added to the water, then the powder is stirred for 5 min for suspending it, after that, the pH is adjusted with ammonia to 11, and in the final, the reducing agent (formaldehyde) is added to start the reaction. The solution is filtered, and the powder is washed by distilled water, followed by drying at 70 °C for 3 hr.

Five different reinforcements that contain 20WC compound, 5Ni, and (0, 1.2, 1.4, 1.6, and 1.8) GNs are prepared by the mixing for 4 hr. with 2:10 ball to powder ratio. The diameter of the used ball is 5mm.

In the third step that is the coating of the different reinforcements with the predetermined copper weight percentage, the solution that composed of 200mm CH_2O, 170 g ($KNaC_4H_4O_6 \cdot 4H_2O$), 35 g $CuSO_4$, 50 g of NaOH, and 1000mm distilled water is used. The ammonia solution 33% is used to adjust the pH of the solution to be 12.5. For reducing the time of the reaction during the precipitation process, the solution is heated

at 60°C. Each reinforcement for each composite is added before adding the formaldehyde and stirred for 10 min to ensure the complete suspension of it in the solution.

Because of the percentages of the GNs are high, the reinforcements coated copper have been stirred in acetone for 2hr that for improving the distribution of GNs in the matrix. In the final step, the stirred composites powders have been dried inside an electrical furnace at 70°C for 3 hr.

2.3. Powder Composites Fabrication

The precipitated copper is produced in the form of CuO and to reduce it to the copper element; each composite powder is heated at 500°C for 1 hr. in a hydrogen atmosphere.

After the powder composites have been prepared, the green compacts on a hydraulic press are formed. All the composites have been pressed at 900 MPa in a die that has a 7x15mm^2 cross-section area. The compacts are produced with 10mm thickness.

In order to consolidate the powder nanocomposites, they have been placed inside a tube electrical furnace that has a hydrogen atmosphere at 1050°C for 120min. Figure 1 shows the heating cycle of the sintering process. As the figure shows, two soaking of heating is used. The first is at 500°C, to allow gases to escape, and the second is to allow grain diffused in each other.

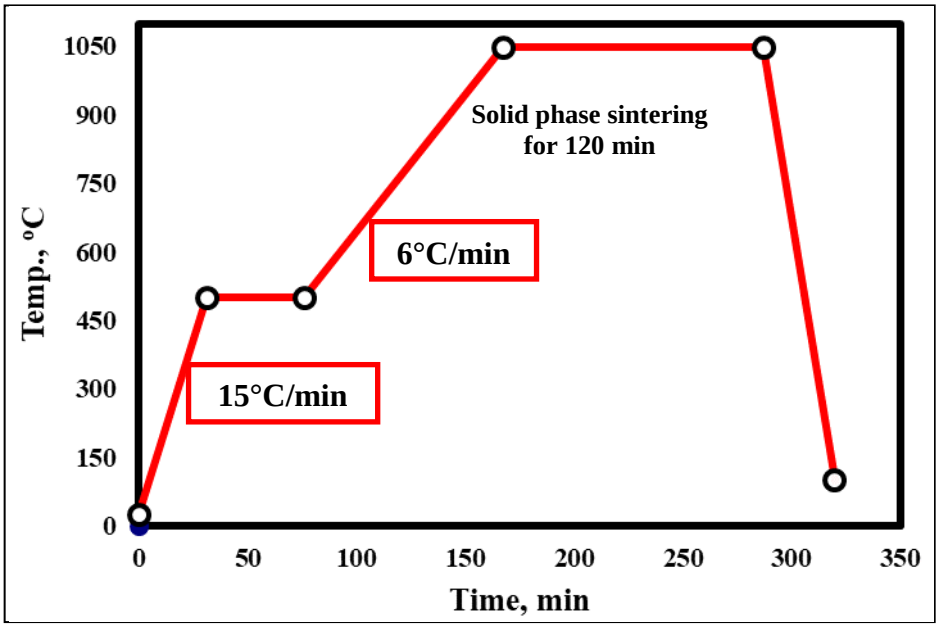

Fig. 1: Fabrication heating cycle.

2.4. Fabricated Nano-Composites Characterization

The morphology of the powders and the consolidated samples is evaluated by the Field Emission Scanning Electron Microscope (FE-SEM; QUANTAFEG250) that equipped with energy dispersive (EDX) spectrometers. According to MPIF standards 42, 1998, and using distilled water, the Archimedes method is used to evaluate the densities of the

produced samples. The bulk hardness of the sintered composites was measured using hardness Vickers tester 5030 SKV England. The load of the measurements was 5 kg for 15 sec loading time. The average of five readings for each hardness value was calculated. Cold compression test was performed to evaluate the mechanical properties and workability of the composites. The dimensions of the composites were L/D ratio > 0.8. Compression test was carried out at 70% from the length of the sample. Universal testing machine UH-F500KN was used to establish the test. The applied crosshead speed of the machine used in this study was 2 mm/min. The maximum compressive strength and compression strain were measured.

3. RESULTS DISCUSSION

3.1. Powder Characterization

Figure 2 illustrates the morphology of the graphene nanosheets (GNs), the nickel powder (Ni), the tungsten carbide compound (WC-TiC-Co) powder, and the Cu coated (WC-TiC-Co/Ni/1.4 wt. GNs). As shown in the figure, the microstructures of the as-received powders GNs, Ni, and WC are in the form of layers, dendritic, and irregular, respectively. An image (d) shows a completely coating for the (WC-TiC-Co/Ni/1.4 wt. GNs) with Cu.

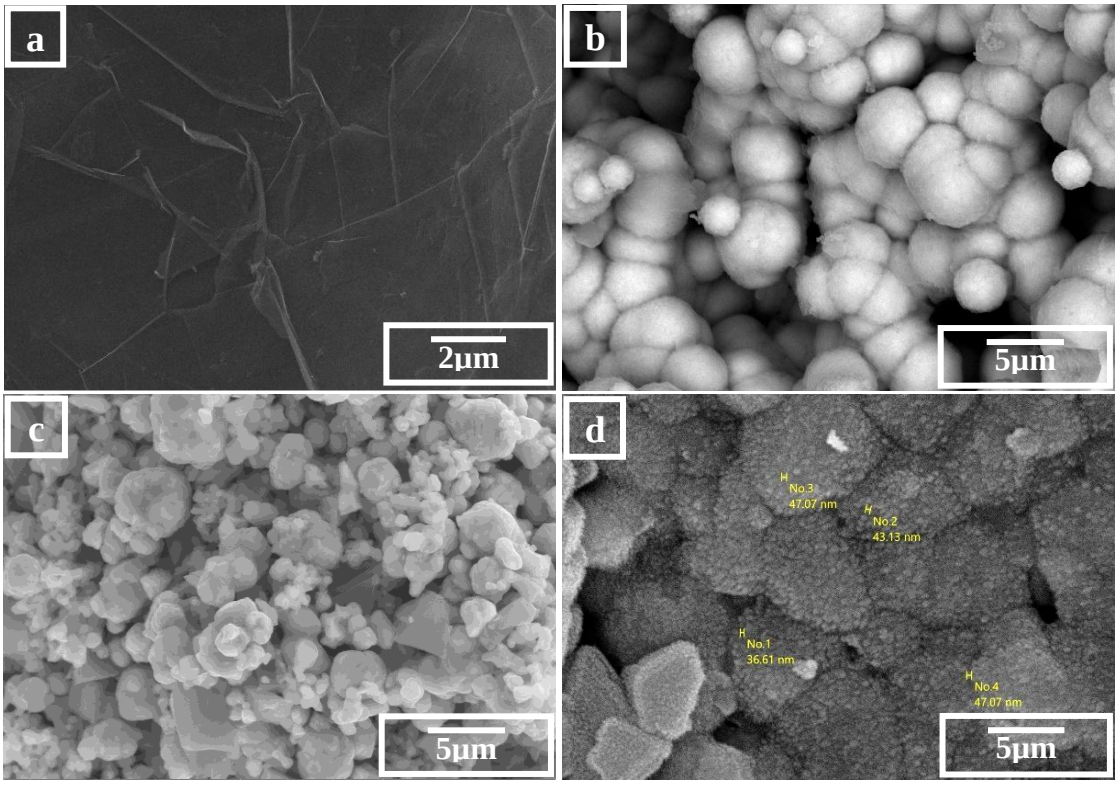

Fig. 2: SEM morphology of the (a) GNs powder, (b) Ni powder, (c) WC compound powder, (d) Ag coated GNs, and (e, f and g) Cu coated (WC-Ni/1.4 GNs) powders.

3.2. Produced Materials Characterization

3.2.1. Microstructure characterization

The effect of reinforcing copper matrix with WC compound, Ni, and different weight percentages of GNs up to 1.8 on the microstructure is shown in figure 3 (a, b, c, d, and e), respectively. As the microstructure shows, the different reinforcements are uniformly distributed in the matrix, which is attributed to the good suspension of the reinforcements in the copper solution during the electroless coating process. In addition, stirring the powders of the prepared nanocomposites in the acetone for 2 hr. had a great effect, especially on the GNs distribution, where it helps in separating the layer of it. No grain pounders between the copper particles are observed, which means a complete reduction of the oxide and good sintering conditions that are time, temperature, and atmosphere. Agglomeration for the GNs layers starts to be formed, especially at 1.6 and 1.8 wt. % GNs.

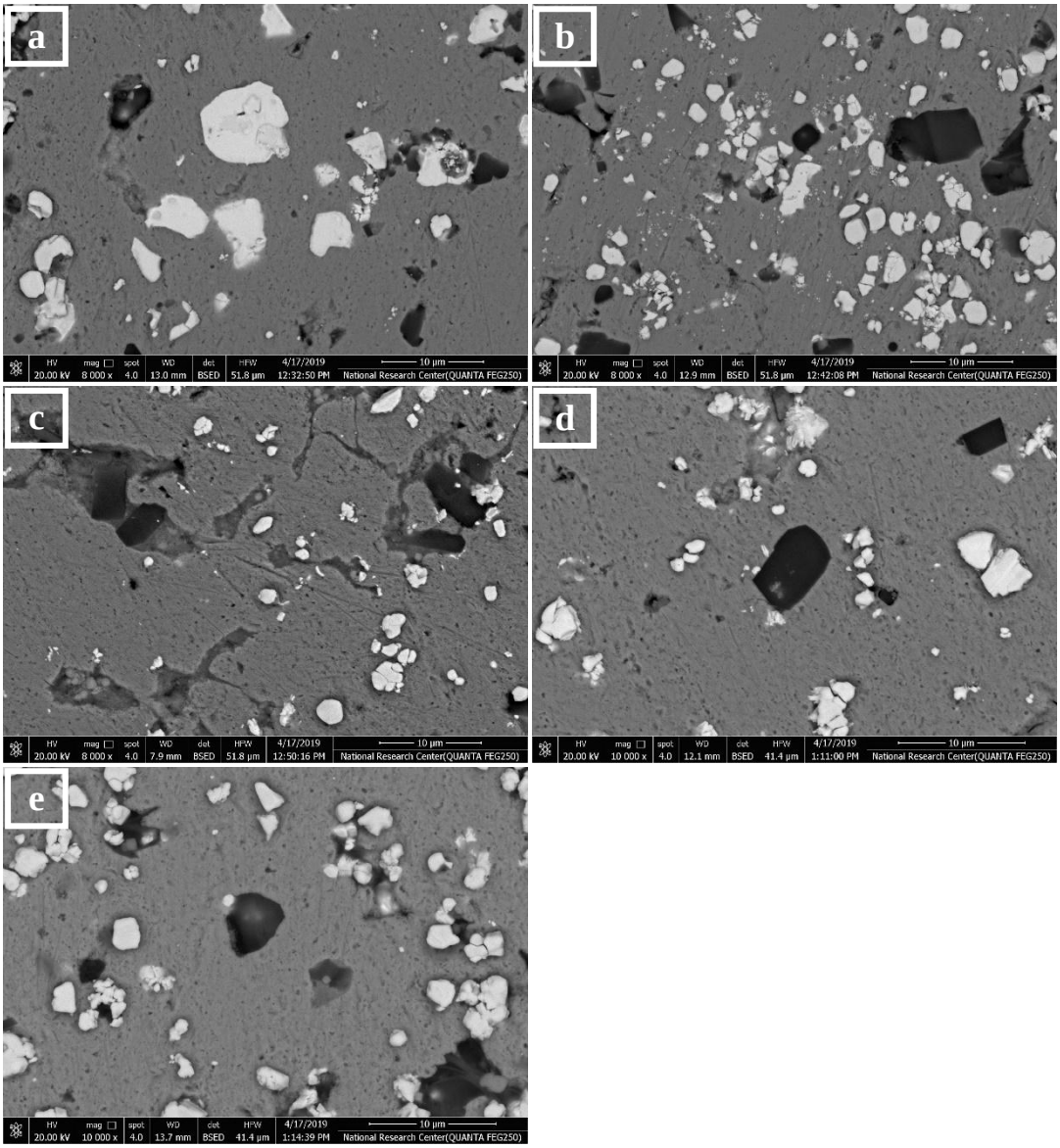

Fig. 3: SEM microstructure of fabricated nanocomposites.

3.2.2. Density measurements

Figure 4 shows the relative density of the Cu-20WC-TiC-Co-5Ni/GNs as a function of GNs content after the fabrication process. As the figure shows, the relative density of the (Cu-WC-Ni) matrix is decreased - by increasing the content of the GNs - from 98.9 to 94.3 for composites containing 0% and 1.8% GNPs, respectively. As shown, the negative effect of the GNs on the density of the matrix increases as its percentage increases, where it is 97.8 and 97.3 for 1.2 and 1.4 samples, while for 1.6 and 1.8 samples it is 95.4 and 94.3, respectively. It is well known that reinforcing the matrix with light materials leads to a decrease in the density of the matrix [15, 16]. Increasing the ceramic reinforcement percentage, especially that in the nano-size, increases the chance of producing agglomeration that in turn, leads to a decrease in the density of the matrix. As shown from the microstructure of the 1.8 wt.% sample (figure 3), the high GNs content in the composite attributed in precipitating the graphene layers on the copper and tungsten carbide grains boundaries in the form of a carbon tube, which causes in producing some micro-pores at the grain boundaries and between the layers itself, and consequently reducing the density.

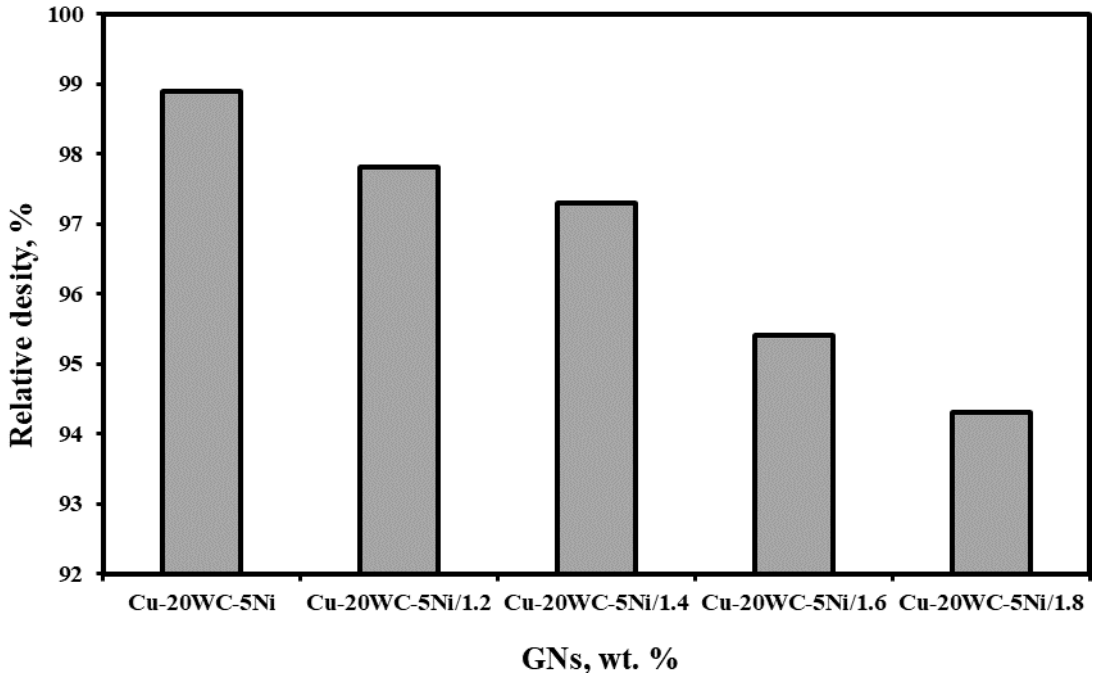

Fig. 4: Variation of copper composite density with GNs wt. %.

3.2.3. Hardness evaluation

The hardness of the Cu-20(WC-TiC-Co)-5Ni/GNPs nanocomposites with the variation of GNPs content is shown in figure 5. The figure shows that the hardness increases with increasing the GNs content up to 1.6 then decreases. It reaches 322.7 HV for a sample that contains 1.6% GNPs compared to 226.4 for pure Cu-20(WC-TiC-Co)-5Ni. Regardless of reduction of hardness of 1.8 wt. %GNs sample that is 318.9 HV, it is still greater than the hardness of the Cu-20(WC-TiC-Co)-5Ni matrix that is 226.4 HV. Improving the hardness of the matrix as a result of reinforcing it with GNS up to 1.6 wt. % attributes to the extremely high mechanical properties of the GNs, the homogenous

distribution of the reinforcements in the matrix, and the good adhesion of the GNs with the Cu matrix. The microstructure of the fabricated composites shows that a lot of graphene layers take the horizontal position, which in turn prevents the indenter of the hardness tool to penetrate in the matrix, and consequently increases the hardness. Decreasing the hardness of the 1.8 wt. % GNs sample attributes to the producing of some agglomerations of the GNs at different regions of the matrix. Agglomerations facilitate the penetration of the indenter, and consequently, the hardness values of the matrix decrease.

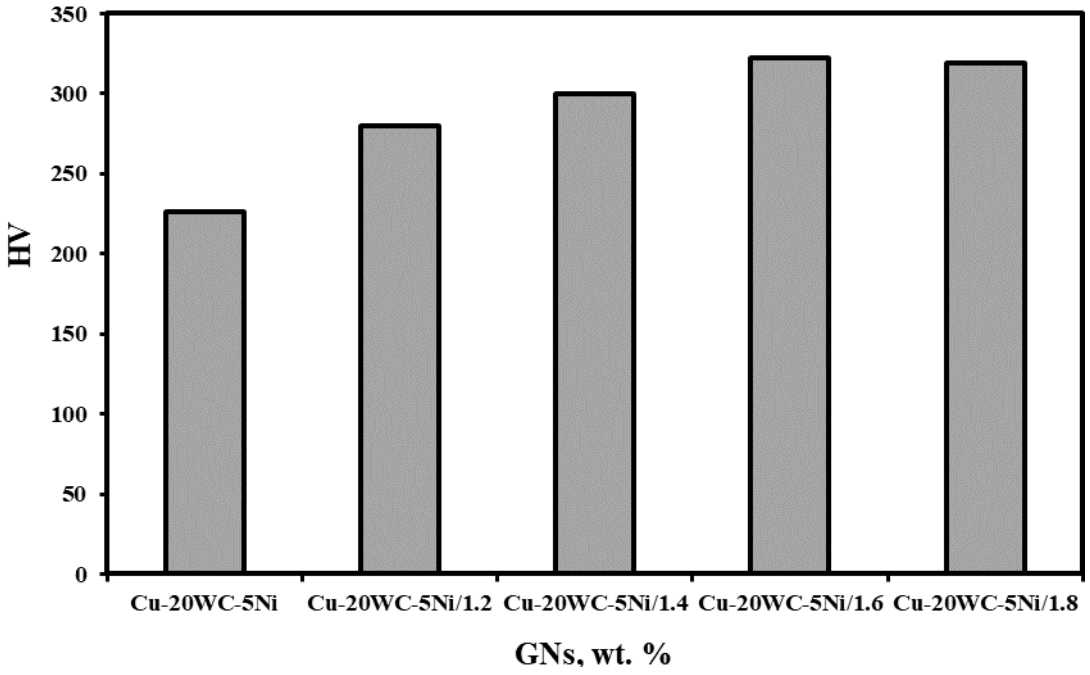

Fig. 5: Variation of the Cu-20(WC-TiC-Co)-5Ni/GNPs nanocomposites hardness with the GNs wt. %.

3.2.4. Compression measurements

The behavior of the axial compressive strength as a function of graphene content for (Cu-WC-TiC-Co-Ni) composites is shown in the figures (6) and (7). The results show that the compressive strength gradually increases by increasing the graphene weight percent from 1.2 up to 1.8 wt. %. These results are in agreement with the hardness results. This is may be due to the good properties of graphene and the good distribution and adhesion between the different constituents of the composites as shown in the microstructure. The effect of graphene addition on the mechanical properties of polymer matrix nano-composites has investigated by Rafiee et al [16]. Their results demonstrated that a significant improvement in tensile strength, Young's modulus and fracture toughness of the fabricated composites had achieved.

3.2.5. Coefficient of thermal expansion (CTE)

The thermal expansion measurements of the Cu-20WC/GNs nanocomposites are shown in figure 8. Two phenomena are shown in the figure, the first is the effect of the heating temperature on the coefficient of thermal expansion, and the second is the effect of the GNs content on the coefficient of thermal expansion of the Cu/20WC-5Ni hybrid

nanocomposite. It is obvious that the CTE increases by increasing the heating temperature. This may be due to increasing the kinetic energy of the atoms by increasing the temperature. It is clear that the change in the CTE is not high that means keeping the composites in its shape at high temperature, and consequently the properties. On the other hand, the CTE of the Cu/20WC-5Ni hybrid nanocomposite is dramatically decreased by increasing the GNs percent.

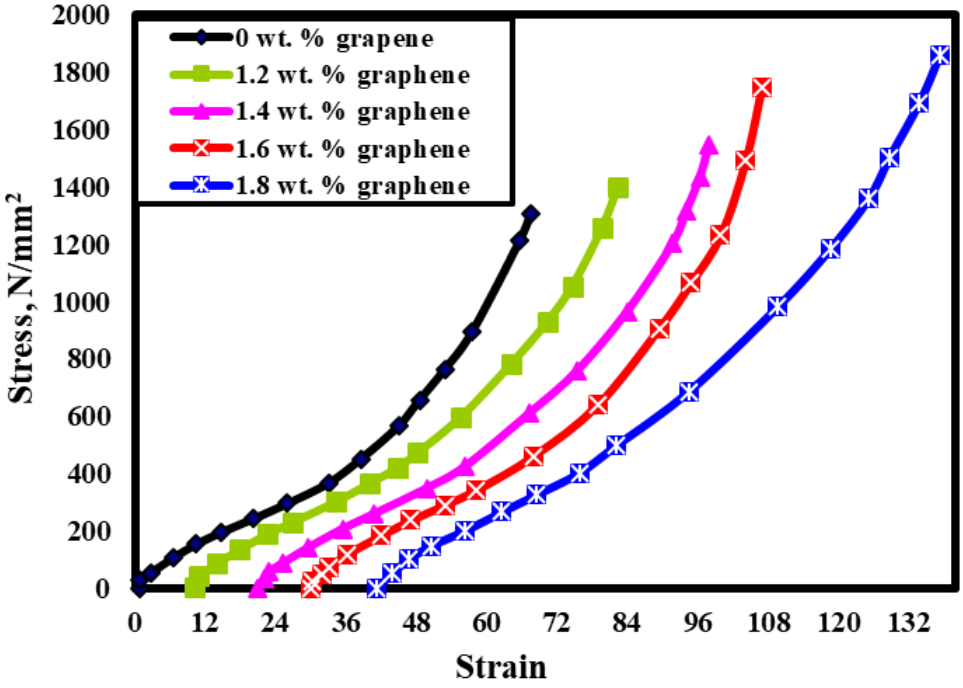

Fig. 6: Stress-strain curves for fabricated nano-composites.

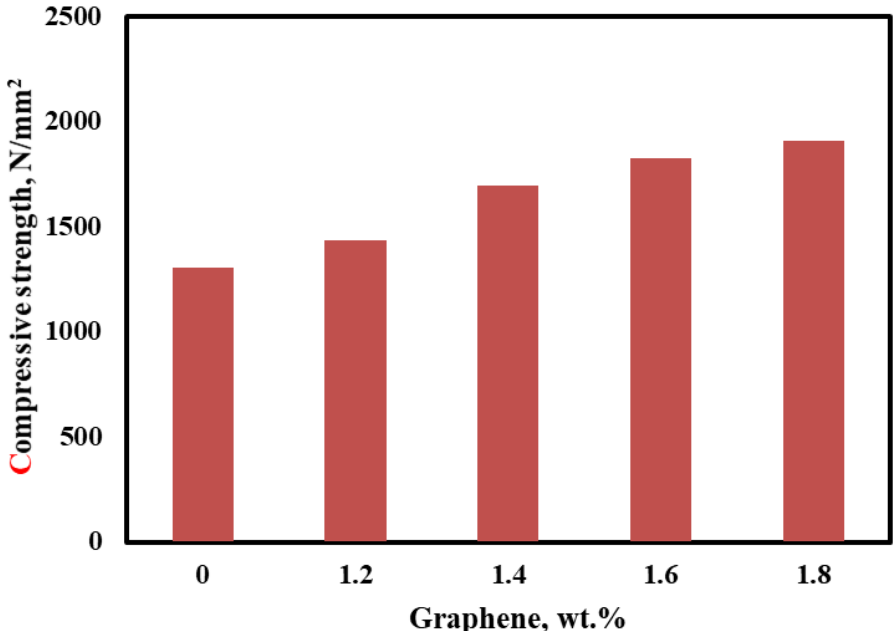

Fig. 7: Compression strength versus graphene content.

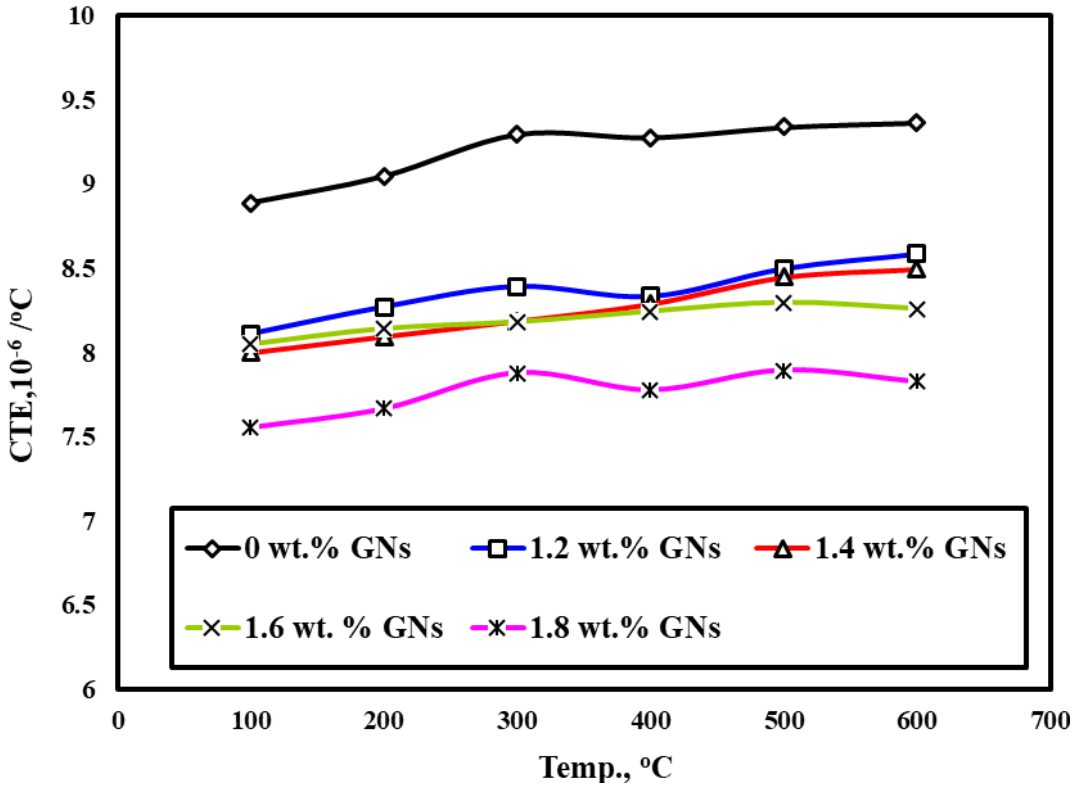

Fig. 8: Thermal expansion measurements.

4. CONCLUSIONS

From the results and discussions, the following conclusions are obtained:

- (Cu-20(WC-TiC-Co)-5Ni) / graphene nano-composites can be prepared by powder metallurgy technique
- Electro-less copper deposition is a suitable technique for preparing a nano copper matrix composite to overcome the non-wettability between Cu with WC-TiC-Co-Ni and graphene nano-sheets mixture.
- The density was decreased by increasing the graphene nano-sheets percent
- The microstructure exhibited homogeneous distribution for all the reinforcements.
- Both hardness and compression strength were improved with graphene nano-sheets additions.
- The coefficient of thermal expansion was increased by increasing the heating temperature and was decreased by increasing the graphene content.

5. REFERENCES

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