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ZEESHAN BAIG[1*], OTHMAN MAMAT[2], MAZLI MUSTAPHA[2], SADAQAT ALI[3]

INVESTIGATING SINTERING BEHAVIOR OF THE GRAPHENE NANOPLATELETS (GNPs) REINFORCED ALUMINUM NANOCOMPOSITES VIA LOW ENERGY SOLUTION BALL MILLING

Owing to the excellent properties, graphene nanoplatelets (GNPs) show great reinforcing ability to improve the mechanical and tribological properties of Al nanocomposites for many automotive applications. In this work, the GNPs dispersion and reinforcing effect in Al nanocomposite was tested. Solvent dispersion via tip sonication and facile low energy ball milling (tumbling milling) using two milling speeds 200 and 300 rpm were employed to develop GNPs/Al powders. Sintering response of the GNPs/Al sintered samples was gauged at two temperatures (550°C and 620°C). The effects of GNPs content, milling rotation speed and sintering temperature on the density, hardness and wear properties of the nanocomposite were examined. The results indicate that relative density % decreases with increasing GNPs content due to possible reagglomeration. The highest hardness of 35.6% and wear rate of 76.68% is achieved in 0.3 wt.% GNPs/Al nanocomposite processed at 300 rpm and 620°C as compared to pure Al due to uniform dispersion, higher diffusion rate at a higher temperature and effective lubrication effect.

Keywords: graphene; aluminum; milling; dispersion; hardness; wear

1. Introduction

Aluminum-based composites have shown great performance in various engineering applications suitable for automotive applications due to their high specific strength/stiffness, high wear resistance and good thermal properties [1]. Newly emerging nanofiller, graphene has attracted tremendous attention owing to its excellent mechanical and tribo properties with unique structure and chemical stability [2]. The graphene structural characteristic combine with extraordinary properties made it most ideal reinforcing agent to bear and transfer load more effectively [3]. Hence, a lot of efforts has been served to develop graphene/Al nanocomposite using various processing techniques. However, researchers faced some serious challenges during incorporation of graphene into the Al matrix are dispersion, strong interface formation and graphene structure retention [4]. These issues resultantly made nanocomposites final properties in the low level of enhancement. To tackle these issues, various techniques were adopted like high energy ball milling, mechanical mixing, sonication followed by powder metallurgy to develop final nanocomposite [5]. Amongst them high energy ball milling have great potential to address these issues and reported by many researchers [6]. On the other hand, during high energy ball milling graphene structure detoriates due to involvement of the aggressive milling condition and high impact stress by the milling balls to produce defects in graphene. These defects are the cause of reactive sites to trigger reaction with metal matrix like Al to produce unwanted reaction product which ultimately reduce final nanocomposite properties [7]. So, there is much need to find such process which effectively incorporate graphene without damaging structure.

Another important aspect of the graphene/Al nanocomposite is a successful consolidation. Many researchers adopted various route to consolidate nanocomposite samples like hot pressing (HIP), spark plasma sintering (SPS) and pressureless sintering under a protective atmosphere [8,9]. Although, the first two techniques are the most successful, they are not industrial scalable due to samples number and size constraints. Whereas, pressureless sintering is a conventional technique widely used to sintered metallic parts and employed in industries for many years. For nanocomposites, many researchers have adopted this technique and reported satisfactory results in terms of mechanical properties improvement of the nanocomposite [10,11]. The Sintering temperature is the most important parameter and needs

[1] DEPARTMENT OF METLLURGICAL & MATERIALS ENGINEERING (MME), UNIVERSITY OF ENGINEERING & TECHNOLOGY (UET), LAHORE 54890, PAKISTAN
[2] UNIVERSITI TEKNOLOGI PETRONAS, MECHANICAL ENGINEERING DEPARTMENT, 32610, BANDAR SERI ISKANDAR, PERAK, MALAYSIA
[3] SCHOOL OF MECHANICAL & MANUFACTURING ENGINEERING, NATIONAL UNIVERSITY OF SCIENCE AND TECHNOLOGY (NUST), H-12, ISLAMABAD 44000, PAKISTAN

* Corresponding author: meteng81@gmail.com

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to be optimizing to obtain the highest densification and ultimately higher properties. Saboori et al. developed GNPs/Al nanocomposite using pressureless sintering and studied the sinterability of nanocomposite samples at three sintering temperature. They observed improved sinterability with increase in temperature but decreases with increase in GNPs content. Furthermore, 50% enhancement in hardness achieved at highest sintering temperature and GNPs content of 2 wt.% [12]. This kind of behaviour was also reported by Latief et al. and found improvement in the hardness with increase in sintering temperature [13].

In this research, we have adopted a method to disperse various fractions of graphene in the Al matrix using facile low energy ball milling under solution condition at two milling speeds (200 and 300 rpm). While two sintering temperatures (550°C and 620°C) were used to investigate the reinforcing effect of graphene contents in the Al matrix after consolidation. The effect of graphene content at two rotation speeds and sintering temperature were gauged on the hardness and wear properties investigations.

2. Materials and Experimental details

Gas atomized aluminium (Al) powder with >99% purity and <15 µm size was used as matrix whereas graphene nanoplatelets (GNPs) as nanofiller having carbon >99.5 wt.%, average thickness of 5-10 nm and <20 graphene sheets with size equals to 6 µm-26 µm as shown in Fig. 1b.

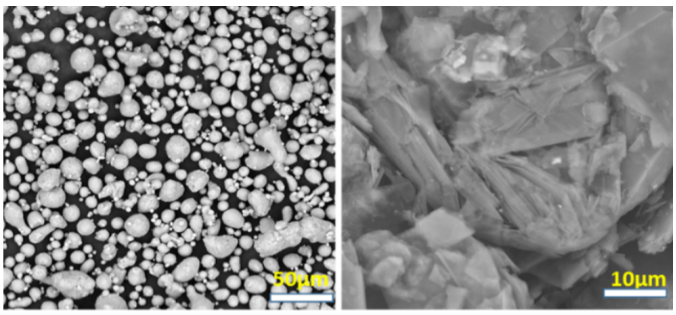

Fig. 1. a) Spherical Al powder b) Graphene nanoplatelets (GNPs)

A simple low energy ball milling (tumbling ball milling) processing was adopted to incorporate graphene nanoplatelets (GNPs) into the Al powder. The blending of the GNPs and Al powder was conducted under solvent also named as solution ball milling. Firstly, GNPs solvent dispersion in ethanol using optimized tip sonication to avoid GNPs structure less defective i.e. sonication time of 30 min at 60% Amplitude (power) was performed [14]. Various fractions of GNPs (0.1, 0.3, 0.5 and 1 wt.%.) solvent dispersions solutions were prepared. All the GNPs sonicated solutions underwent for solution ball milling using tumbling milling for 10 min at 250 rpm for further GNPs exfoliation to receive few layers graphene. The low energy milling was carried out at US Stoneware ball milling machine. Al powder weighing 50 g was added into each ground GNPs solution jars and start milling by maintaining the ball-to-powder ratio of 10:1 for 2 hours at two rotation speeds i.e. 200 and 300 rpm respectively. An interim period of 5 min was given after every 30 min milling to avoid heating. Upon completion of milling, GNPs/Al solutions were vacuum filtered, and vacuum dried at 90°C for 10 hours to acquire GNPs/Al nanocomposite powders. Finally, GNPs/Al nanocomposite powders subjected to uniaxial cold compaction using steel die at 500 Mpa for 1 min hold time. Next, to that, pressureless sintering of the green samples was conducted using box furnace under N_2 atmosphere at two sintering temperature i.e. 550°C and 620°C for 2 hours as dwell time. Samples were left in the furnace for furnace cooling upon completion of the sintering process. The developed samples have dimension equals to 30 mm diameters and 4-5 mm thickness. Pure Al was also subjected to the same processing for comparison with nanocomposite samples.

Archimedes' Principle was applied to calculate the densities of the pure Al and nanocomposite sintered samples. Relative sintered densities (%) were measured using theoretical densities of the pure Al and nanocomposite powders using the rule of mixture. For theoretical density calculations, the density of Al (2.7 g/cm^3) and GNPs (2.17 g/cm^3) were used. Vickers hardness testing on polished sintered pure Al and nanocomposite samples were carried out at 200 gf for 15 minutes holding time to measure micro hardness (HV). For each sample, five readings were recorded, and the averaged value was reported. Scanning electron microscope (SEM) (Phenom-Pro X) equipped with an energy-dispersive X-ray spectrometer (EDS) was used for raw and nanocomposite morphology along with graphene presence in the Al nanocomposite powders. To investigate the wear behaviour, wear testing on the polished samples were executed using the pin on disc testing under dry and ambient conditions using Taber Linear Abraser 5750. The adopted process detail and parameters to perform wear test can be found elsewhere [11]. Weight losses (mg) of the tested samples were measured before and after wear testing using Mettler Toledo weighing machine with 0.0001 g accuracy. After that, wear volume loss calculated by using Eq. (1) to further find out the wear rate following Archard's equation as shown in Eq. (2)

$$\text{Wear Vol Loss (mm}^3\text{)} = \text{mass loss (g)} / \text{density} \quad (1)$$

$$W = \frac{V}{PS} \quad (2)$$

where V is the wear volume (mm^3), P was the applied load (N) and S was the total sliding distance (meters).

3. Results and discussion

Fig. 2(a-d) represents the dispersion and presence of the GNPs on the Al powder particles after ball milling at two rotational speeds i.e. 200 and 300 rpm. Uniform dispersion of the GNPs within Al powder is the assurance of the improvement

in the final nanocomposite properties as also earlier reported. Fig. 2(a and b) illustrates the GNPs dispersion in Al powder at 200 rpm along with EDX results to confirm the GNPs presence. At low content (0.1 wt.%) of GNPs cannot be visibly seen on the Al surface. Whereas, with an increase in content (1 wt.%), GNPs can be seen on the Al powder surfaces as also marked by arrows. EDX analysis was also carried out to confirm the GNPs presence while detecting the carbon element on powder particle. So, analyzing the EDX results, presence of carbon on each GNPs fraction confirm the GNPs availability on the Al surface. On the other hand, the same results were also produced by the GNPs/Al nanocomposite powder ball milled at 300 rpm (Fig. 2(c and d)) depicting results of 0.1 and 1 wt.% GNPs fraction incorporation into the Al powder. One aspect should also be noted that at both rotation speeds, GNPs/Al powder morphology has not changed after milling which can be ascribed to low energy transmitted by the ball's impact during tumbling milling processing which is also beneficial to protect GNPs structure while achieving dispersion.

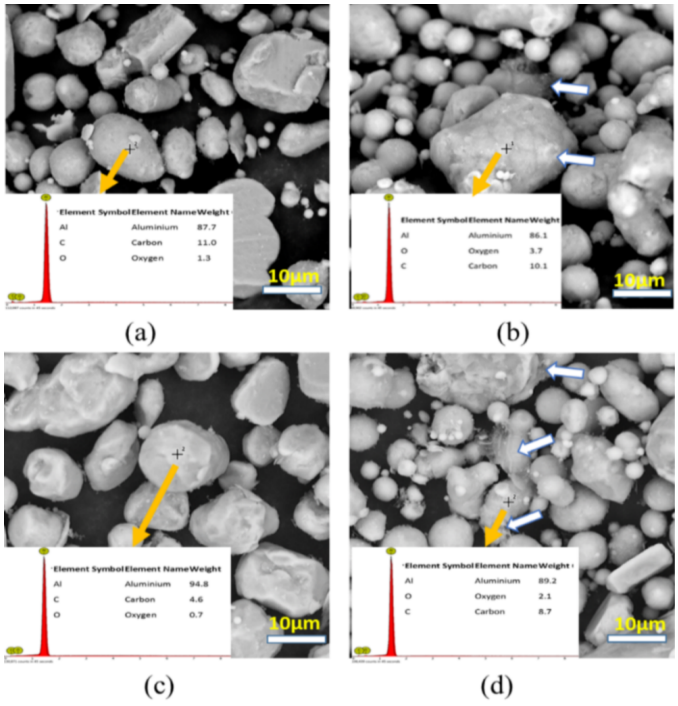

Fig. 2. SEM images of the GNPs/Al nanocomposite powders ball milled at 200 rpm (a and b) and at 300 rpm (c and d) showing GNPs of 0.1 and 1 wt.% along with respective EDX results

Fig. 3 demonstrated the relative densities behaviour of the sintered samples processed at two rotational speeds and two sintering temperatures (550°C and 620°C). Interestingly, Fig. 3(a and b) shows the same behaviour of the relative densities exhibited by the GNPs nanocomposites at both rpm and sintering temperature. As can be seen that with the addition of the GNPs at lower content i.e. 0.1 wt.%, relative densities curtly increase as compared to pure Al and then sharply decrease with increase in GNPs content up to 1 wt.%. This could be due to GNPs ability to uniformly dispersed at low content and filled the gaps between the Al particles during compaction and sintering irrespective of the sintering temperature. Whereas, the decrease in the relative densities at both temperatures can be ascribed to two reasons i.e. due to the formation of GNPs agglomerates after reagglomeration at higher content and the other reason might be the presence of the low-density GNPs which ultimately reduce the density of the nanocomposites at a higher content. This kind of behaviour has been highlighted in previous reports [15]. Importantly, researchers reported the decrease in density of graphene/Al nanocomposite developed by other processing techniques like mechanical mixing and high energy ball milling. Therefore, it can be concluded that the processing technique has not much influenced on the density, but the presence of GNPs played a decisive role showing such kind of density behaviour.

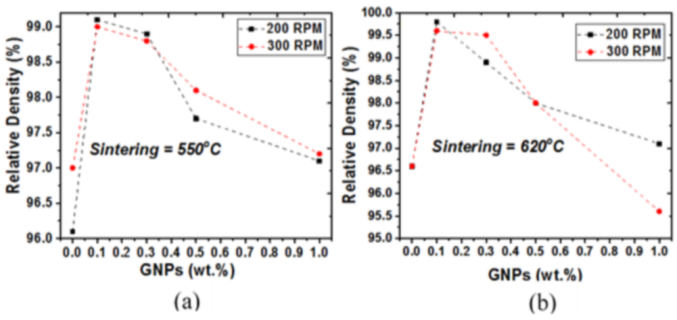

Fig. 3. The trend of relative densities (%) of the GNPs/Al nanocomposite samples at two sintering temperatures a) 550°C and b) 620°C

Fig. 4 exhibited the micro hardness of the GNPs/Al nanocomposite managed at two rotational speeds and two sintering temperatures (550°C and 620°C). GNPs/Al nanocomposite presents the different evolution of hardness values at different GNPs content and sintering temperatures along with rotational speeds (200 and 300 rpm). At sintering temperature of 550°C, the maximum hardness of 42 Hv exhibited by the 0.1 wt.% GNPs/Al nanocomposite processed at 300 rpm equals to 20% than pure Al hardness value (35 Hv). Whereas at 200 rpm processed GNPs/Al samples exhibited a maximum hardness of 39.5 Hv showed a 13% increase than pure Al (Fig. 4a). Interestingly, at both rpm and 550°C, the GNPs/Al showed maximum hardness at 0.1 wt.% of GNPs content whereas after that content hardness decreases with rising in GNPs content which could be due to the graphene possible reagglomeration. On the other hand, GNPs/Al nanocomposite sintered at 620°C showed a little different aspect of hardness evolution (Fig. 4b). As can be seen that highest hardness of 48.8 Hv shown by the GNPs/Al samples at 300 rpm with 0.3 wt.% GNPs content equals to 35.6% than pure Al whereas at 200 rpm depicts maximum hardness of 45 Hv at 0.1 wt.% GNPs content equals to 25% than pure Al. Overall, it can be analyzed that GNPs/Al nanocomposite samples ball milled at 300 rpm and sintered at 620°C showed the highest increase in hardness than other samples and pure Al. The increase in hardness can be indorsed to GNPs reinforcing effect by blocking the dislocations movement results in pinning effect and results in an increase in hardness [16,17].

The possible reason for such behaviour can be attributed to the following reason connected with dispersion processing and consolidation temperature. Firstly, at higher rotational speed (300 rpm), GNPs evenly distributed within the Al powder due to higher impact forces delivered by the milling balls to facilitate GNPs dispersion at a higher content. Secondly, at the higher sintering temperature, the diffusion of Al atoms is easier to thus higher diffusion rates facilitate good bonding between the Al particles [13]. As known, sintering temperature has a pronounced effect and the controlling factor of sintering which is govern by the following Eq. (3);

$$D = D_o \exp\left(\frac{-Q}{RT}\right) \tag{3}$$

where D denotes diffusion coefficient, D_o (constant), Q (activation energy), R (Boltzmann's constant) and T is absolute temperature.

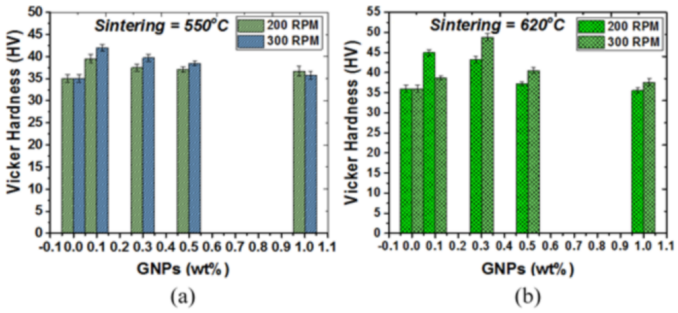

Fig. 4. The trend of micro hardness of the GNPs/Al nanocomposite samples at two sintering temperatures a) 550°C and b) 620°C

Graphene nanoplatelets (GNPs) contains the number of graphene sheets layered to form stacking morphology up to certain thickness. Such kind of morphology is ideal to use them for tribology applications which provide enough lubrication between the mating surfaces. Therefore, investigating GNPs incorporation into the Al matrix to study wear behaviour is also beneficial for many applications like automotive and aerospace. As known, hardness and wear properties are dependent on each other i.e. hardness inversely proportional to wear resistance. In this research, wear loss and wear rate of the nanocomposite samples were calculated and compared with the pure Al results. Fig. 5 reveal the wear loss (mg) and wear rate trends of the tested samples using a pin on disc method applied to samples sintered at two temperatures. Fig. 5(a and b) presents the wear loss (mg) behaviour of the nanocomposite samples after wear testing at both sintering temperatures. As expected, wear loss suddenly reduces after the addition of the GNPs into the Al up to 0.1 wt.% at 550°C due to uniform dispersion at both rotation speeds (Fig. 5a). Similarly, at 620°C nanocomposite showed varied behaviour connected with hardness improvement i.e. at 200 rpm the wear loss drops up to 0.1 wt.% and at 300 rpm wear loss reduction exhibit up to 0.3 wt.% (Fig. 5b). It should be noted that 300 rpm processed samples have much less wear loss as compared to pure Al and nanocomposite samples at 200 rpm. Likewise, samples sintered at 620°C has shown much reduce values of wear loss than 550°C sintered ones.

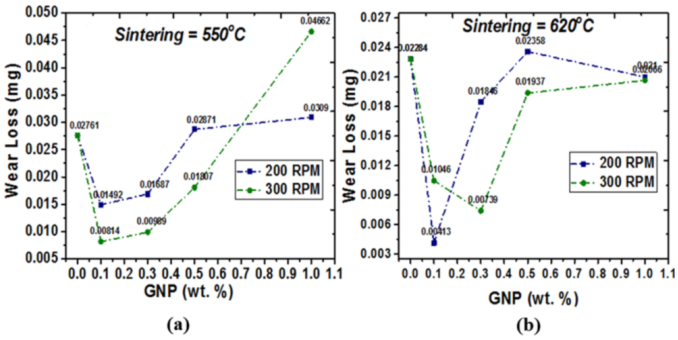

Fig. 5. The trend of wear loss of the GNPs/Al nanocomposite samples at two sintering temperatures: a) and c) 550°C and b) and d) 620°C

Fig. 6(a and b) illustrates the wear rate behaviour at both sintering temperatures after calculating from Archard's equation (2) using wear loss of all the samples. GNPs/Al samples sintered at 550°C showed the reduce wear rate up to 0.1 wt.% GNPs content showing an increase of 39.42% (at 200 rpm) and 33.65% (at 300 rpm) higher than pure Al. Correspondingly, at 620°C, 200 rpm processed samples have **reduced** wear rate up to 0.1 wt.% GNPs and at 300 rpm wear rate up to 0.3 wt.% GNPs showing improvement in wear resistance of 80.92% (200 rpm) and 76.68% (300 rpm) than pure Al respectively. Importantly, all samples exhibited the same behaviour as to wear loss as discussed above. Conclusively, wear rate of samples processed at 300 rpm and sintered at 620°C showed wear resistance up to 0.3 wt.% due to the lubrication effect of the GNPs. At higher content of GNPs, all samples showed higher wear rates due to the possible reagglomeration.

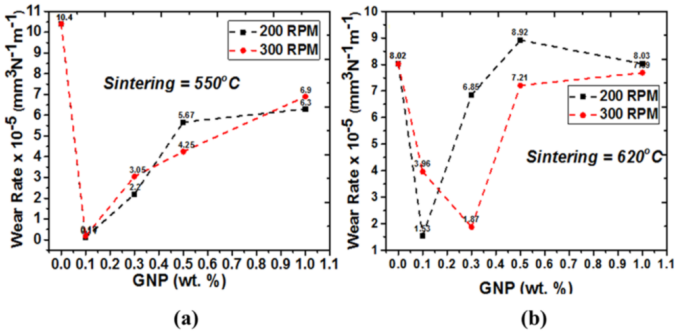

Fig. 6. The trend of wear rate of the GNPs/Al nanocomposite samples at two sintering temperatures: a) and c) 550°C and b) and d) 620°C

4. Conclusion

This paper presents the investigation of the graphene nanoplatelets (GNPs) behaviour at two sintering temperature (550°C and 620°C) in terms of dispersion, hardness and wear rate of the developed GNPs/Al nanocomposite. Different fractions of the GNPs dispersed into the Al powder combining tip sonication

for solvent dispersion and facile solution low energy ball milling and followed by cold compaction and pressureless sintering. The two-rotation speed 200 and 300 rpm) of milling were used to disperse GNPs into the Al powder. The SEM analysis confirms uniform dispersion and EDX validate the graphene presence. Relative density decreases at higher content possible due to graphene reagglomeration. Improvement in hardness and wear rate of GNPs samples at both rotation speed and sintering prove the GNPs reinforcing effect. In conclusion, 0.3 wt.% GNPs/Al samples processed at 300 rpm and 620ºC sintering temperature exhibited maximum hardness wear properties. At higher GNPs content properties reduction observed at all samples due to GNPs reagglomeration at both rpm and sintering temperatures.

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