

Successful Synthesis of Graphene-Aluminum Composite with Improved Microhardness



Binod Bihari Palei, Tapan Dash, Susanta Kumar Biswal

ABSTRACT: In this work we successfully prepared aluminum-graphene composite (0.2 wt%) after 6 hrs of successful ball milling. The ball milling experiments were conducted between 2 and 6 hrs. The microstructural and spectroscopic comparison of aluminum-graphene composites were studied by evaluating properties by characterizing via XRD, FESEM, TEM, Micro Raman and microhardness. The second order disorder peak (2D) was found grow with milling time. In the Raman spectra intensity of 2D addresses about exfoliation take place of graphene in the composite. 0.2 wt% added graphene shows higher hardness than aluminium. Sintering of composites were done at 600 °C. Separation of stacking of carbon layers was confirmed from improvement of D peak in the ball milled samples. Sheet type structure of graphene was shown in TEM study. XRD confirms composite formation.

Keywords: Graphene-Aluminum Composite, Ball milling, Microhardness, Micro Raman

I. INTRODUCTION

Aluminum (Al) has attractive physical and mechanical properties with various common and industrial applications [1]. Generally aluminum uses for different transportation materials, preparing defense components, energy carrier material, material for consumer industries, etc. [1-2]. During synthesis of Al based composites, they face unwanted tribologic behavior. However ductility, thermal and electrical properties make the material unique among other. To develop the application aspects of Al further its wear and mechanical properties have to develop. Many reinforcement components such as Al₂O₃, SiC, BN and B₄C are added with Al to produce composites with enhanced properties [3-11]. These materials also coated on the Al substrate for various coating applications. In the primary battery, Al is used as suitable candidate material. In the new generation lithium ion batteries (LiBs), the uses of this material has significant role. Always attempt has been taken to use Al in composite form to come over its limitations. In this context the future material, graphene has great impact towards improving mechanical, electrical behavior, thermal and tribologic properties of the composite compared with its pure form.

*Correspondence Author Graphene has high strength, high elastic modulus, excellent electrical, thermal and optical properties, and extremely high specific surface area [12-13]. This two dimensional material has flat

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Monolayer of carbon atoms. The best fact about graphene is that it can be reinforced to both polymer and metal matrix composites. Due to such unique properties, it is a candidate material for sensors, energy storage materials, transistors, super capacitor, material solar cell, etc. But it has been an issue to produce graphene with good quality. Graphene with bi-layer or particularly monolayer is the focus of researchers working in this area. This is the demand of current technology.

In this research work, aluminum-graphene composite (0.2 wt%) is successfully prepared by 2-6 hrs of ball milling. Properties of composites were evaluated by employing various characterization techniques such as X-ray diffraction (XRD), field emission scanning electron microscopy (FESEM), micro Raman spectroscopy, energy dispersive spectroscopy (EDS) and Microhardness. Al-graphene composite (0.2 wt%) prepared by 6 hrs of milling shows high microhardness value than pure Al.

II. MATERIAL SYNTHESIS

Composites of aluminum-graphene (0.2 wt%) were prepared by ball milling between 2 and 6 hrs. The ball milling was carried out in dual-drive type of planetary ball mill. In ball milling both impact and shear forces were generated for production of very ultra-fine particles of aluminum-graphene composite. In ball milling, we used one gyratory shaft and two pieces of cylindrical steel jars. Electric motors rotate both shaft and jars. Jars were filed with a sample-to-ball charge ratio of 1:9. Diameter of balls of 12 mm and 8 mm was used for ball milling purpose. Both the shaft and jars are rotated simultaneously and separately at a speed 360 rpm and 140 rpm respectively. For hardness study, samples were pressed at 400 MPa to prepare their pellet forms. The ball milled Al-graphene (0.2 wt%) mixture was sintered at 600 °C 6 hrs.

PANalytical X'Pert Pro diffractometer (CuK α source) system was used for X-ray diffraction (XRD) study. Micro Raman analysis was done by Renishaw inVia Reflex (UK) spectrometer. The microstructures were taken by field emission scanning electron microscope (FESEM) (model: ZEISS SUPRA 55). Energy dispersive spectra (EDS) taken by Oxford, X-Max system. Microhardness of sample were done by LECO (load 0.4 kg, 15 s dwell time).

III. RESULTS AND DISCUSSION

A. XRD Analysis

XRD analysis of pure aluminium and its typical composites with graphene (0.2 wt%) was carried out and presented in Fig.1. Pure Al shows diffracted peaks of Al.



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The polycrystalline Al has intense of (111), (200) and (311). But when graphene is added and prepared composites with Al by fusing by mechanical milling, it is observed that C (002) peak is strongly appeared. XRD infers formation of the composite of Al-graphene. The peaks corresponding to Al were found to be affected during composite formation. The peaks were found with less intense in comparison to pure aluminum. In case of Al-graphene (0.2 wt%) composites, the peaks of C and Al were found to appeared with relatively more FWHM. The increase in FWHM indicated the reduction of grain or particle size. The above changed in crystalline behavior was attributed to the effect of ball milling. The crystallite dimension was determined for major phases of Al and C by applying Scherrer formula [14]:

$$\delta = 0.9\lambda / \beta \cos\theta \quad (\text{Eq. 1})$$

where, λ =wavelength of the X-ray and β = full width at half maximum (FWHM) in radian. The average crystallite for Al and C (graphene) were determined in the range of ~ 78-85 nm, ~38-52 nm respectively.

B. Micro Raman Spectroscopic Analysis

Micro Raman spectrum for typical pure aluminum and ball milled Al-graphene (0.2 wt%) composites obtained by 2-6 hrs of ball milling are shown in Fig. 2. Raman spectra of pure aluminum were found with no peak. Also Al peak is absent in ball milled composites. Al is found absent in spectra because of metallic nature. The Al-graphene (0.2 wt%) composite samples prepared by 4 and 6 hrs of milling show peaks of G (corresponds to lattice of graphite), D (corresponds to 1st order disorder peak in graphite structure) and 2D (2nd order disorder peak in graphite structure) [15-16]. The 2D position is almost twice of Raman shift of D band. The peak intensity ratio of D w.r.t. to G i.e. I_D/I_G determined from Fig. 2. I_G/I_{2D} ratio increases with increasing ball milling time from 2 to 6 hrs. It directs about the structural modification (structural imperfections and size reduction because of milling. Al-graphene (0.2 wt%) sample prepared by 6 hrs milling shows I_G/I_{2D} ratio around 1, indicating formation of bi-layer graphene. 2D peak is found down-shifting with increasing milling time from 4 to 6 hr. Almost G, D and 2D peaks for 6 hrs balling sample were found with relatively more FWHM. The above results indicate that though graphene is formed at nano level but proper Bernal stacking of the graphene is maintained. The above result may be optimized mechanical and thermal energy supplied at the time of ball milling. The average crystallite size of the sp^2 domains (L_a (nm)) determined by following Lucchese et al.[17]. The average crystallite size values were found to be 70.3 and 58.09 nm for 4 hrs and 6 hrs ball milled samples respectively.

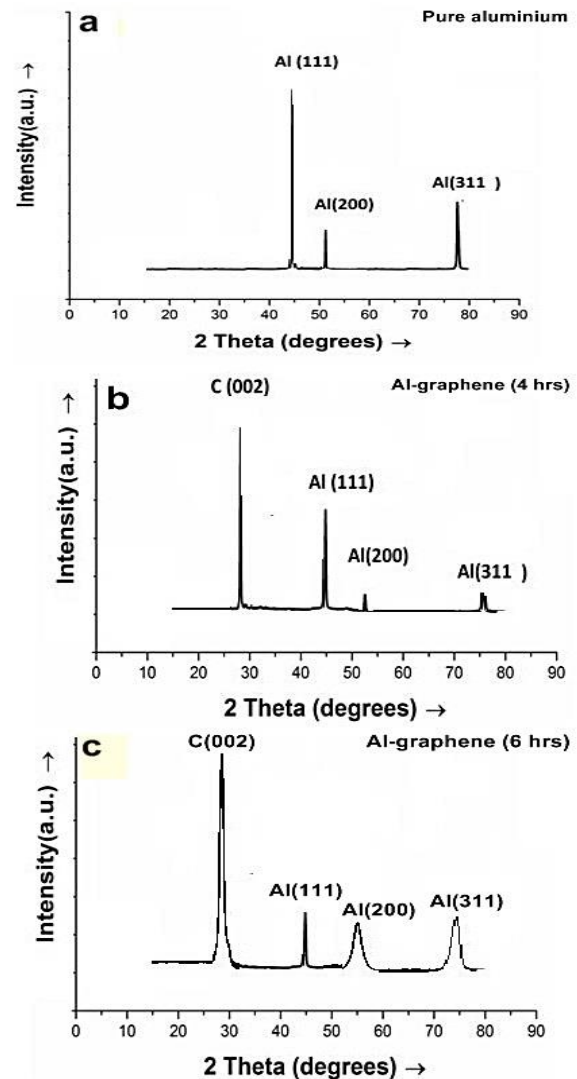


Fig.1. XRD spectra: a-aluminum sample; b-aluminum-graphene (0.2 wt%) (4hrs milling) ; b-aluminum-graphene (0.2 wt%) (6 hrs milling)

C. Micro Raman Spectroscopic Analysis

Micro Raman spectrum for typical pure aluminum and ball milled Al-graphene (0.2 wt%) composites obtained by 2-6 hrs of ball milling are shown in Fig. 2. Raman spectra of pure aluminum were found with no peak. Also Al peak is absent in ball milled composites. Al is found absent in spectra because of metallic nature. The Al-graphene (0.2 wt%) composite samples prepared by 4 and 6 hrs of milling show peaks of G (corresponds to lattice of graphite), D (corresponds to 1st order disorder peak in graphite structure) and 2D (2nd order disorder peak in graphite structure) [15-16]. The 2D position is almost twice of Raman shift of D band. The peak intensity ratio of D w.r.t. to G i.e. I_D/I_G determined from Fig. 2. I_G/I_{2D} ratio increases with increasing ball milling time from 2 to 6 hrs. It directs about the structural modification (structural imperfections and size reduction because of milling. Al-graphene (0.2 wt%) sample prepared by 6 hrs milling shows I_G/I_{2D} ratio around 1, indicating formation of bi-layer graphene.

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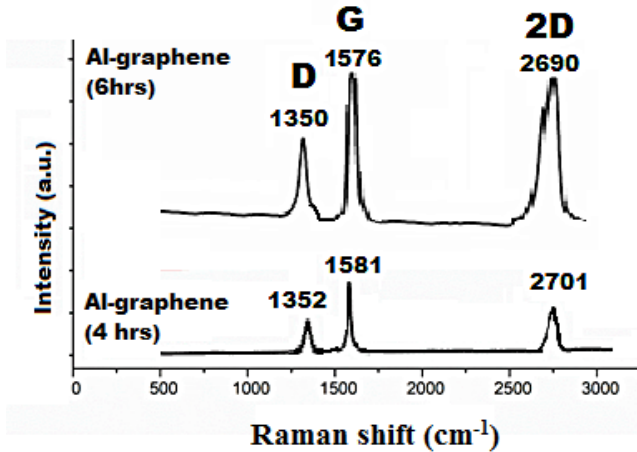


Fig.2. Micro Raman spectra of aluminum-graphene (0.2 wt%) composite prepared at different ball milling time.

D. FESEM, TEM and EDS studies

FESEM, TEM and EDS analysis the typical Al-graphene (0.2 wt%) composite prepared by 6hrs of ball milling were carried out and shown in Fig. 3-5. Morphological characterization is focused on to know behavior of distribution of Al and graphene layers and distribution of Al particles. FESEM shows stacking behaviour of graphene layers in the composites. TEM analysis of composite shows amalgamation of Al particles in the sheets of graphene. Such improvement and dispersion of Al has not been worked out earlier. No crystal defect is observed in the microstructural analysis which indicates the improved quality of composite prepared by ball milling route. EDS of typical Al-graphene (0.2 wt%) composite shows peak of C and Al.

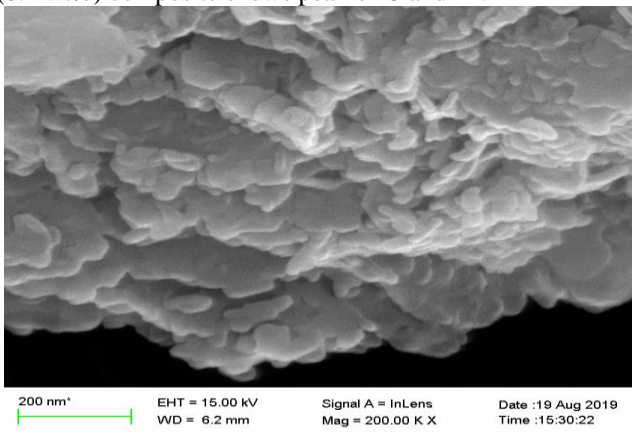


Fig. 3. FESEM analysis of Al-0.2 wt% graphene composite (6 hrs milling)

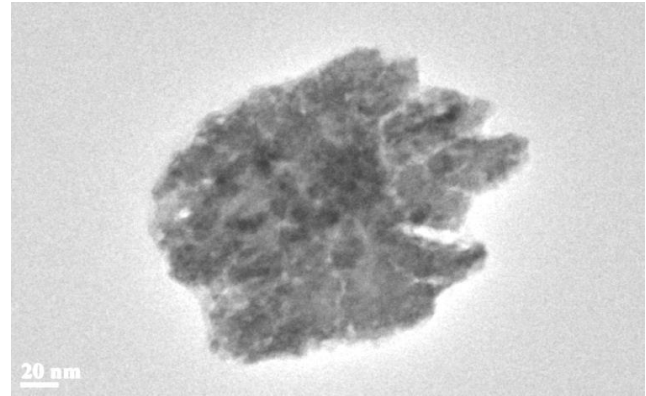


Fig. 4. TEM analysis of Al-0.2 wt% graphene composite (6 hrs milling)

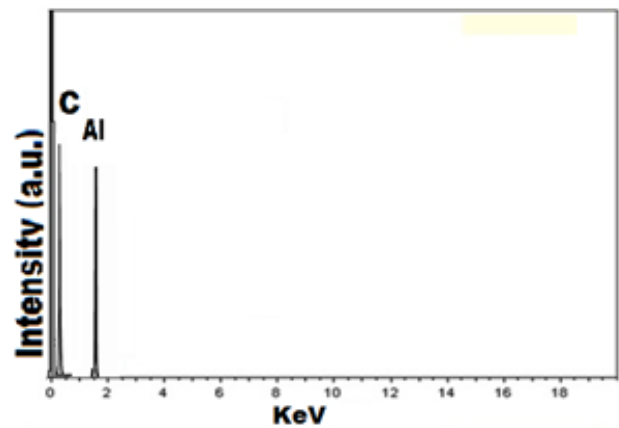


Fig. 5. EDS of Al-graphene (0.2 wt%) composite (6 hrs milling) taken on FESEM study

E. Microhardness Study

Sintered pure Al and Al-graphene composites were taken for microhardness evaluation. Microhardness were calculated and presented in Table 1. It is marked that microhardness increases by graphene addition in the composite. Pure Al shows hardness value of 51 ± 4 VHN. Typical Al-graphene (0.2 wt%) prepared by 6 hrs of ball milling shows higher hardness (85 ± 6 VHN) value than pure Al. As per the microstructural analysis and hardness values are concerned, it is observed that graphene addition improves structural and microhardness properties of composite. Overall work can be attributed to the optimized ball milling condition to successfully prepare Al-graphene composite with good dispersion.

Table I: Microhardness values determined for Al and Al-graphene composites

Sample ID	Micro hardness in VHN
Al	51 ± 04
Al-Graphene (0.2 wt%): 2 hrs	68 ± 05
Al-Graphene (0.2 wt%): 4 hrs	75 ± 8
Al-Graphene (0.2 wt%): 6hrs	85 ± 6

IV. CONCLUSION

Al-graphene (0.2 wt%) composites prepared by dry ball milling technique between 2 and 6 hrs. The composites were sintered at 600 °C for 6 hrs. Pure aluminum has additionally C peak in XRD spectra in its composite with graphene. The 6 hrs ball milled sample shows broadening of FWHM of peaks indicating successful reduced particle size. Stacking behavior of graphene was clearly observed in FESEM analysis. Purity of sample was checked by EDS study. Micro Raman indicates the bi-layer graphene present in composite. While pure Al exhibited microhardness of 51 ± 04 VHN, Al-graphene (0.2 wt%) composite displayed microhardness value of 85 ± 6 VHN. Thus, effective microstructural and microhardness results directs that graphene addition and optimized ball milling condition for the present work enhances the property of composite.

REFERENCES

1. D.B. Miracle Compos. Sci. Technol., 65, 2526 (2005)
2. M.K. Surappa, Sadhana, 28, 319 (2003)
2. J.C. Williams, E.A. Strake, Acta Mater., 51, 5775 (2003)
3. B. Li, Q.L. Pan, C.P. Chen, H.H. Wu, Z.M. Yin, J. Alloys Compd. 664, 553 (2016)
4. V.G Davydov, T.D Rostova, V.V. Zakharov, Y.A.Filatov, V.I. Yelagin, Mater. Sci. Eng. A 280, 30 (2000)
5. D.B. Miracle, Compos. Sci. Technol 65, 2526 (2005)
6. F. Chen, N. Gupta, R.K. Behera, P. K. Rohatgi, JOM, 70, 1 (2018)
7. R. Casati, M. Vedani, Metals, 4, 65 (2014)
8. K.K. Alaneme, J. Mater. Res. Technol. 2, 188 (2013)
9. A. Devaraju, A. Kumar, B. Kotiveerachari, Trans. Nonferrous. Met. Soc. China, 23, 1275 (2013)
10. A. Devaraju, A. Kumar, B. Kotiveerachari, Mater Des, 45, 576 (2013)
11. P. Dash, T. Dash, T.K. Rout, A.K. Sahu, S. Das, S.K. Biswal, B.K. Mishra, RSC Adv. 6, 12657 (2016)
12. R.K. Gunasekaran, K. Jayasankar, T. Dash, A. Dash, B.K Jena and B.K. Mishra, RSC Adv. 6, 20067 (2016)
13. A.L. Patterson, Phys. Rev., 56, 978 (1939)
14. B. B. Nayak, R. K. Sahu, T. Dash and S. Pradhan, *Applied Physics A*, 124, 1 (2018)
15. R.K. Sahu, T.Dash, V. Mukharjee, S.K. Pradhan, B.B. Nayak, Chemical Physics Letters, 739, 136986 (2020)
16. M. M. Lucchese, F. Stavale, E. H. M. Ferreira, C. Vilani, M. V. O. Moutinho and R. B. Capaz, Carbon, 5, 1592 (2010)

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