A Novel Alumina-Magnesium oxide- Reduced Graphene Composite: Synthesis and Characterizations

Binod Bihari Palei[*]*, Tapan Dash, Susanta Kumar Biswal. R.Saktivel*

Centurion University of Technology and Management, Odisha, India

CSIR-IMMT, Bhubaneswar-India

Email: bbpalei@immt.res.in

Abstract

A novel new alumina-magnesium oxide with reinforced of reduced graphene oxide (RGO) composite materials have been synthesized by using a horizontal high energy planetary ball milling process. The composite has very high density, improved mechanical properties, retaining strength under higher temperatures, and high thermal conductivity. Al_2O_3-MgO-RGO composites can be used as a novel material for many industrial applications. The effect of RGO in the combination of Al_2O_3 and MgO have a potential scope in several demanding fields, including automobile, aerospace, defenses, sports, electronics, bio-medical, and other industrial purposes. The prepared composites have been studied by X-ray diffraction (XRD), Field emission scanning electron microscopy (FESEM), micro Raman spectroscopy, Transmission electron microscope, Automatic density meter, Particle size analyzer.

Keywords: *Reduced Graphene Oxide, Composite, Microstructural property*

1. Introduction

Needs of human society always insist us for developing smart materials through intelligent way with superior properties. In the process of development, different materials in elemental, compound, composites, and their integrated forms are continuously developed to fulfill the material requirement of a smart and advanced society. In this present context, authors have taken attempt to produce advance composites of alumina-reduced graphene oxide and magnesium oxide because of their superior properties. Alumina (Al_2O_3) is taken as an advanced ceramic material for high-temperature structural applications, and it is currently used in electronics, aerospace, and automotive industries [1-5]. Though Al_2O_3 inspiring mechanical hardness and chemical inertness, moderate thermal conductivity, high corrosion and wear resistance, low density (3.75 to 3.95 g/cm^3), it has the inferior fracture toughness which restricts Al_2O_3 use for advanced high-temperature structural applications [6]. The most common tactics to improve the mechanical properties has been to add a filler agent to formulate a nanostructured composite material. In this context, graphene (reduced graphene oxide (RGO)) was evaluated as the reinforcing agent of Al_2O_3 ceramics using planetary ball milling followed by sonication & sintering in an inert atmosphere. The addition of graphene and Mgo to Al_2O_3 simultaneously improved the density and mechanical strength of Alumina– Magnesium oxide & graphene composite. The graphene has shown exceptionally high mechanical (Young's modulus of 1.0 TPa), high electron mobility (25000 $cm^2 V^{-1}S^{-1}$), 100 times stronger than steel, high electrical (less resistivity than silver). High surface area 2630 m^2/g and thermal properties, which make it one of the most promising reinforcements for ceramic matrix composites (CMCs) [7-10]. Graphene is generally a 2-dimensional (2D) sheet of carbon, having high surface areas as compared to graphite, carbon black and CNTs. Thus a trace loading of less than 0.2 Wt % in a matrix may lead to most significant improvements in the physical properties like density.

In this work attempt has been taken to prepare Al_2O_3- MGO & RGO composite by planetary ball milling followed by Sonication & sintering in an inert atmosphere. The morphology, structure, composition and density of composites were studied using various analytical techniques such as X-ray diffraction (XRD), Field emission scanning electron microscopy (FESEM), Automatic density analyzer, CS analyzer and Micro Raman spectrophotometer.

- The ball milling of the samples was carried out for 12 hr. The wt % of MgO in Al_2O_3 was varied as 5, 10, 12.5, 15 % with constant .04% of RGO in 30 gm total sample. After ball-milled over, the samples were taken for sintering at 1500^0C for time period of 2 hrs in Argon atmosphere.
- Before and after sintering Density is measured by Quantachrome ULTRAPY-1200e Pycnometer.
- XRD patterns of the samples were studied by PANalytical X'Pert Pro diffractometer equipped with Cu & Co-Kα radiation. The patterns were taken in the 2θ range 5–80.

2. Experiment and Observation :

In this work, alumina, MgO, and RGO are taken as the starting material. The four different compositions of Al_2O_3- MGO has taken keeping RGO constant. By varying wt% of MGO & Alumina i.e., 5, 10,12.5 &15% were ball-milled by a horizontal planetary dry ball mill. The planetary ball mill was constructed in such a way to generate both impact and shear forces in the samples. The planetary ball mill has the advantage to overcome the limitation of the gravitational field and continue to supply a strong accelerating field. The planetary ball mill consists of a gyratory shaft (580 mm) and two cylindrical steel jars of diameter 200 mm each; both are rotated simultaneously at a speed of 300 rpm. The rotation of both jars and the shaft makes the balls (inside the chamber) to move very strongly and violently, leading to develop considerable impact energy at balls that improve the grinding kinetics and results of Nanocomposites. The sample to ball charge ratio of 1:10 was maintained in the planetary ball mill. The ball milling of the samples was carried out for 14 hr. The wt % of MGO in Al_2O_3 was varied as 5, 10, 12.5 & 15%. After ball-milled over, the samples were taken for sonication of I hr at 750-watt energy 20 kHz frequencies in ethanol's atmosphere followed by sintering at 1500^0C for a time period of 2 hr in Argon atmosphere.

XRD patterns of the samples were studied by PANalytical X'Pert Pro diffractometer equipped with Cu & CoKα radiation. The patterns were taken in the 2θ range 6–80 at a scan speed of 0.019 min^{-1}. The microstructures of samples were observed by FESEM (model: ZEISS SUPRA 55). Automatic density analyzer of the samples was determined by Quantachrome ULTRAPY 1200e.

3. Results and Discussion:

The X-ray diffraction was carried out to understand crystalline nature, phase purity, and interlayer spacing of typical Al_2O_3-RGO & Mgo (5 wt%) composite sample. The result was carried out and presented in Fig. 1. The composites having phases and planes are identified by comparing the observed *d*-values (experimentally determined) with the *d*-values of standard powder diffraction data file, C (graphite-2H): 00-041-1487 and Al_2O_3: 01-075-1862 supplied in JCPDS-ICDD PDF-2 (2004). The XRD results show patterns in 2θ range of 6^0-80^0. The composite nature of the sample is confirmed from the XRD analysis. The XRD analysis shows the peaks due to C and Al_2O_3. Al_2O_3 phase shows diffraction peaks of (104), (110), (113), (024), (116), (122), and (214). Al_2O_3 shows two major peaks at around $2\theta = 41.0^0$ and $2\theta = 51.0^0$. Similar kind of result was observed by literature [11]. A characteristic peak of C (002) strongly appears at $2\theta = 30.0^0$ is attributed due to the presence of RGO in the composite. The sharpness of the peak and lowest value of FWHM (0.1089) of the peaks indicates that resolution and degree of crystallinity were better, which indicates that there will be an increase in mechanical properties. Different phases were compared to their standard values which indicate the well ordered and better crystalline nature of the composite. FESEM analysis of the typical composites of Al_2O_3-MgO with different content of MgO keeping RGO constant 5, 10, 12.5 &15% are presented in Fig. 2 and 3 respectively. Uniform distribution of MgO & RGO within the Al_2O_3 grains is very important to prepare superior quality composite for various advanced applications. In Al_2O_3 with MgO 5 wt% shows homogeneous distribution with graphene in Al_2O_3 matrix with respect to others wt%ge. The composites were also found to free from microporosity and any kind of surface defects. There is an increase in the percentage of 10.

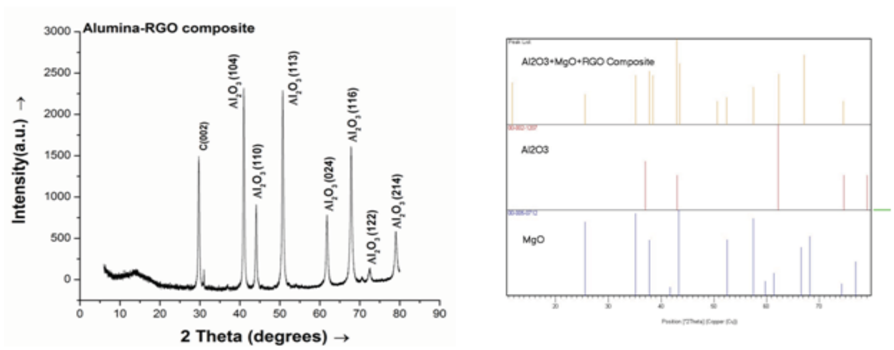
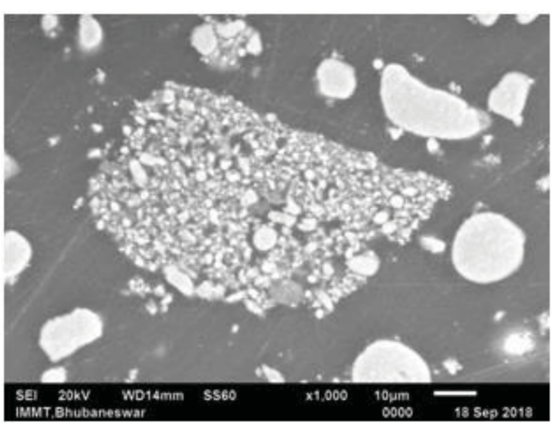


FIG. 1. XRD pattern observed for typical Al_2O_3-MgO(5 wt%) -RGO (0.04 wt%) composite sample



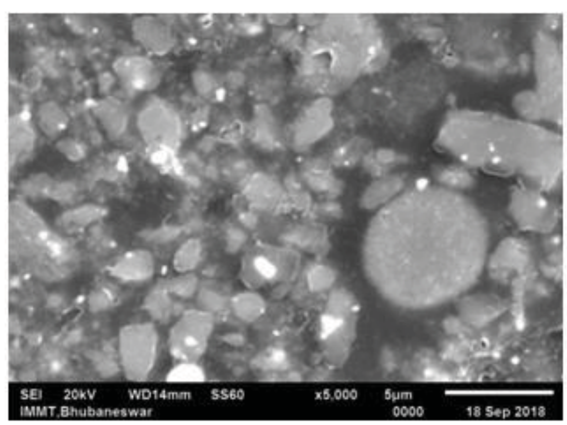


FIG 2. FESEM micrograph of Al_2O_3-MGO(10 wt%) -RGO(0.04 wt%) composite sample

FIG 3. FESEM micrograph of Al_2O_3-MGO(5 wt%)-RGO (0.04 wt%) composite sample

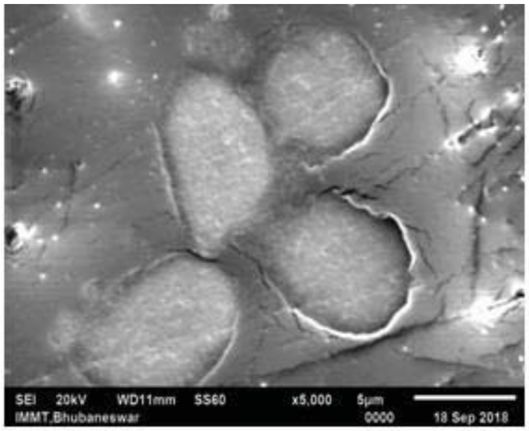


FIG.4. FESEM Micrograph showing MGO & RGO at the grain boundary of Al_2O_3.

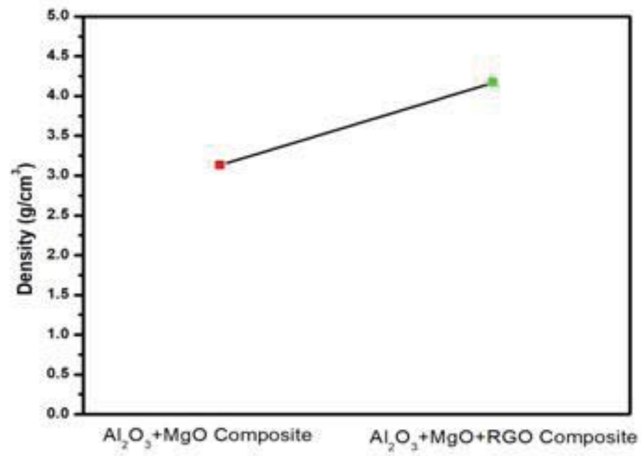


FIG.5. variation of density with of Al_2O_3-MGO(5wt%)-RGO (0.04%) composite sample

Raman data reveals that after grinding for 12 hours, we are getting I_D and I_G ratio 0.9. There is no peak broadening in comparison to 16 hours of data, in which the I_D and I_G ratio is found to be 1.04.

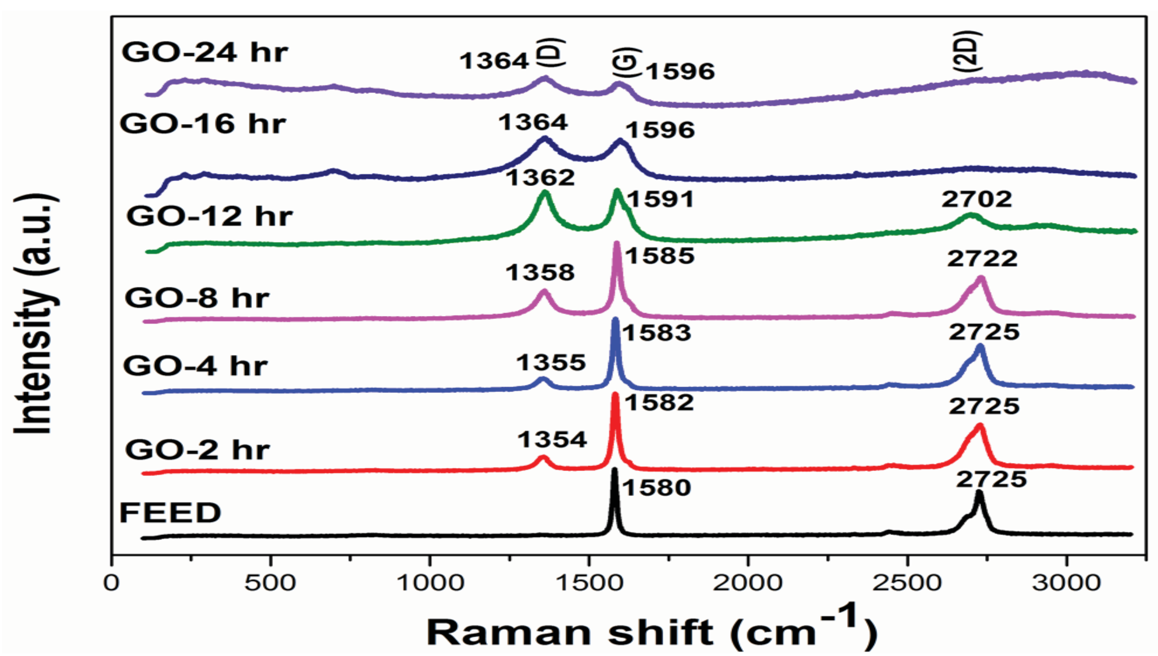


FIG.6. Micro- Raman data

Table 1. Micro -Raman Data analysis

Sample ID	D peak, (cm^{-1})	G peak (cm^{-1})	2D peak (cm^{-1})	I_D	I_G	I_D / I_G
FEED	-	1580	2725	0	4100	0
GO-2 hr	1354	1582	2725	1000	4800	0.2
GO-4 hr	1355	1583	2725	900	4450	0.2
GO-8 hr	1358	1585	2722	1900	4710	0.4
GO-12hr	1362	1591	2700	3558	3599	0.9
GO-16 hr	1364	1596	broad	2400	2300	1.04
GO-24 hr	1364	1596	broad	1520	1642	0.9

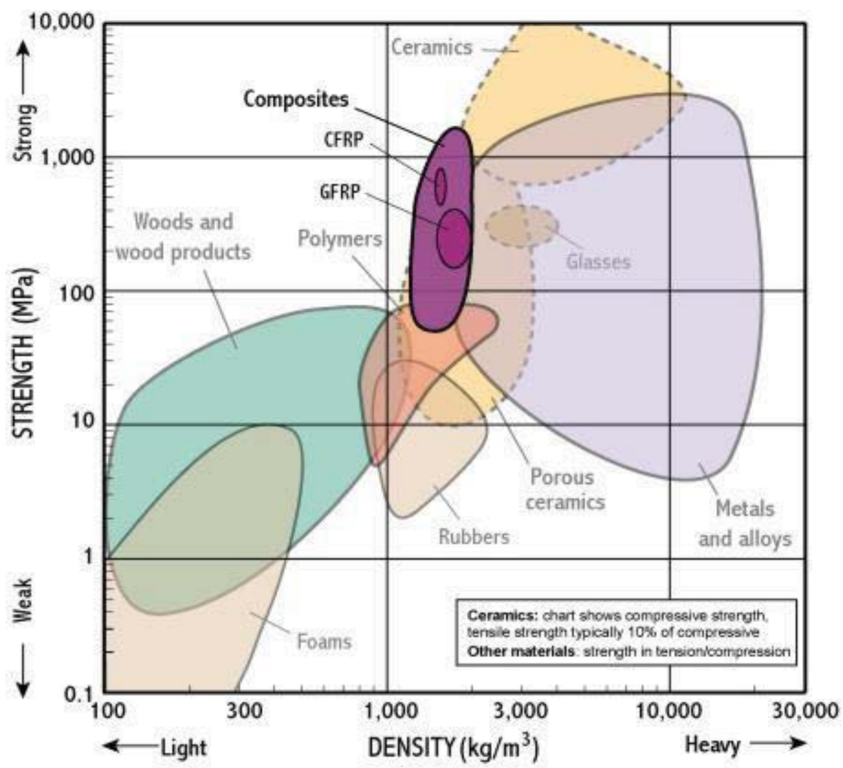


FIG 7. Density versus Mechanical strength

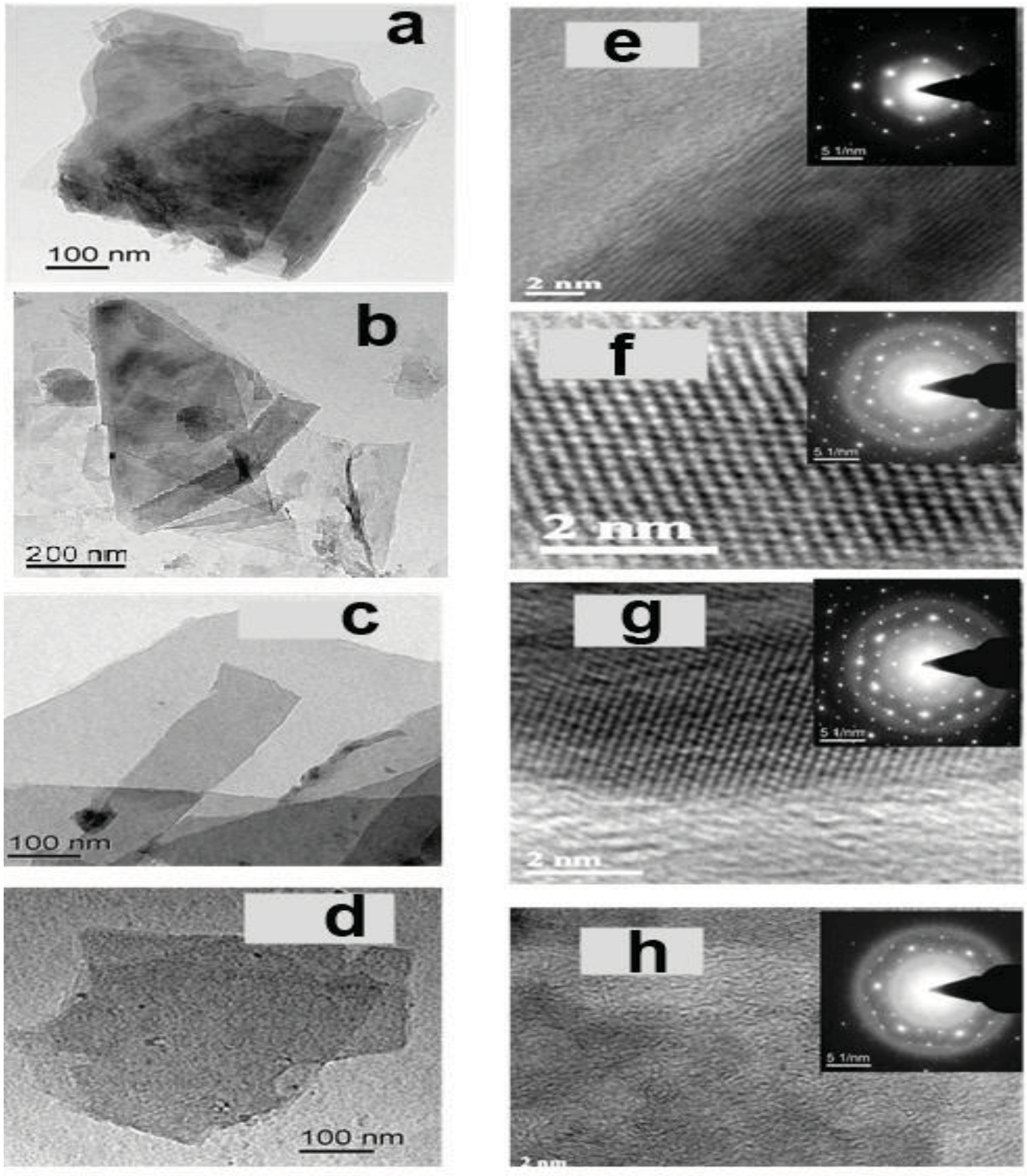


FIG 8. TEM, HRTEM and SAED patterns of ball-milled samples: a, b, c & d are TEM images of 8 hr, 12 hr, 16 hr, and 24 hr ball-milled GO samples; e, f, g & h are HRTEM images with their corresponding selected area electron diffraction pattern (SAED) of 8 hr, 12 hr, 16 hr, and 24 hr ball-milled GO samples

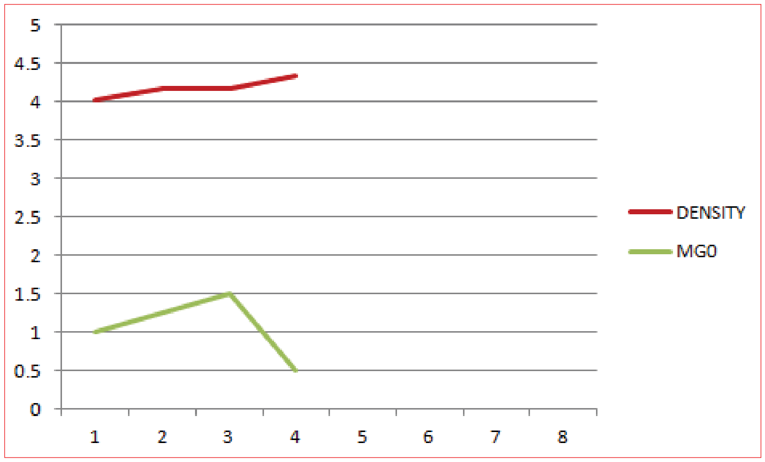
Table 2. Density Calculation of Al_2O_3, MgO and RGO composites

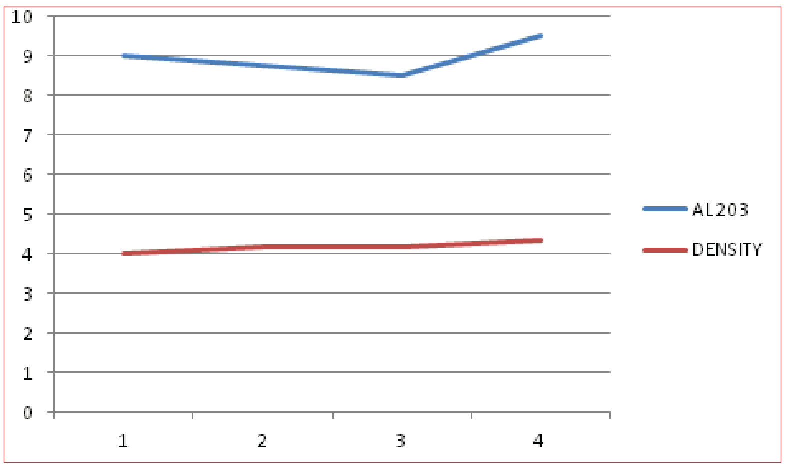
Al_2O_3(9gm)+ MgO(1 gm)+RGO(0.04gm)								
Reading No	Volume	Density	Avg. Volume	Avg. Density	The. Density	Green Density	% increment from TD	% increment from GD
1	0.159	4.077	0.1611	4.0274	3.913	3.1304	2.92358804	28.65448505
2	0.1624	3.996						
3	0.1601	4.0547						
4	0.1636	3.968						
5	0.1605	4.0427						

Al_2O_3(8.75)+ MgO(1.25 gm)+RGO(0.04gm)								
Reading No	Volume	Density	Avg. Volume	Avg. Density	The. Density	Green Density	% increment from TD	% increment from GD
1	0.1792	4.1579	0.179	4.1632	3.90375	3.123	6.646173551	33.30771694
2	0.1787	4.1702						
3	0.1785	4.1728						
4	0.1785	4.1737						
5	0.1801	4.1373						

Al_2O_3(8.5)+ MgO(1.5 gm)+RGO(0.04gm)								
Reading No	Volume	Density	Avg. Volume	Avg. Density	The. Density	Green Density	% increment from TD	% increment from GD
1	0.1588	4.2261	0.1609	4.1699	3.8945	3.1156	7.071511105	33.83938888
2	0.1619	4.1437						
3	0.162	4.1416						
4	0.1619	4.1456						
5	0.16	4.194						

Al_2O_3(9.5)+ MgO(0.5 gm)+RGO(0.04gm)								
Reading No	Volume	Density	Avg. Volume	Avg. Density	The. Density	Green Density	% increment from TD	% increment from GD
1	0.1582	4.3939	0.16	4.3442	3.9315	3.1452	10.49726567	38.12158209
2	0.1613	4.3081						
3	0.162	4.2913						
4	0.1605	4.3311						
5	0.158	4.3989						





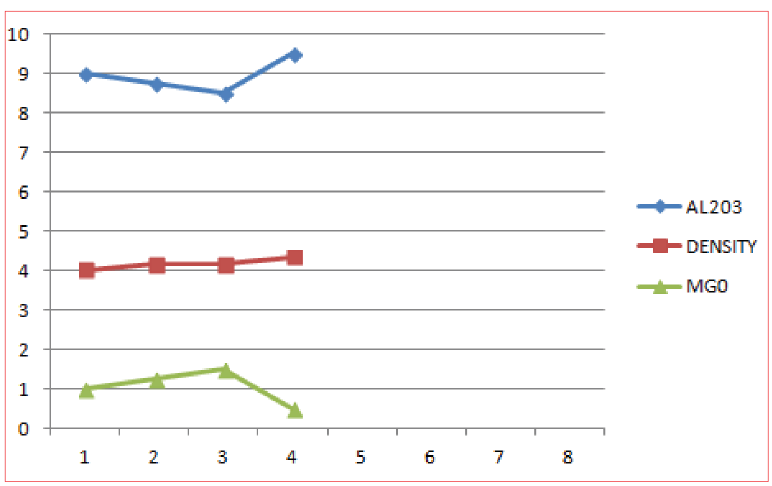
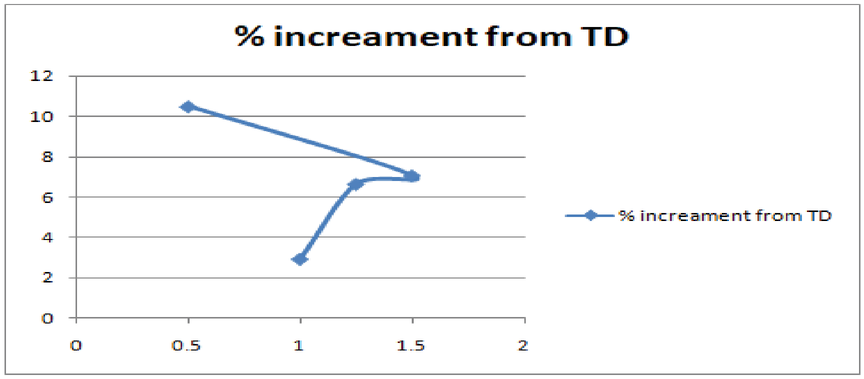


FIG 9. Graphs showing density variation with respect to Al_2O_3 and MgO with constant RgO



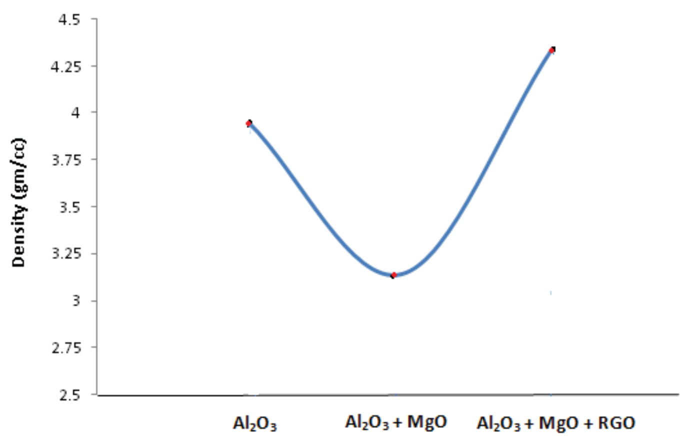


FIG 10. Increase of Densityfrom Theoritical value

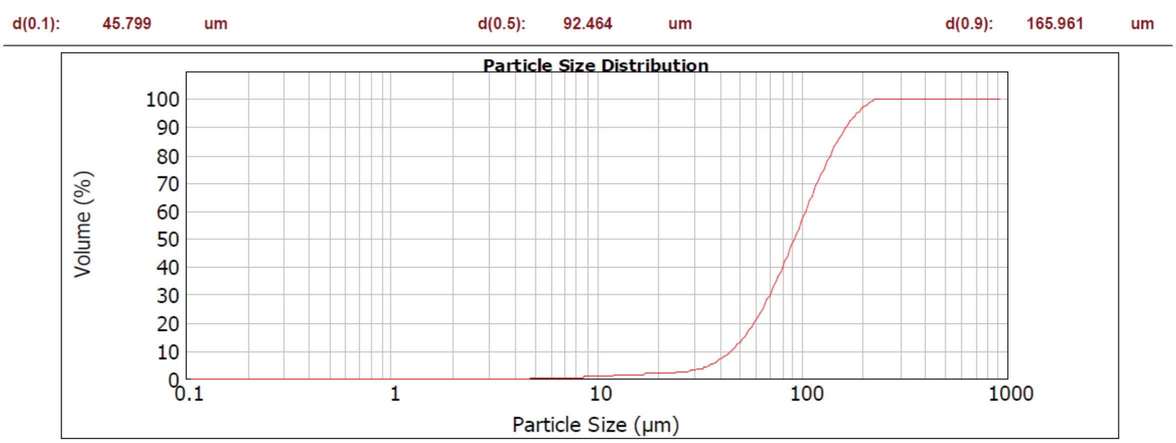


FIG 11. Particle size before grinding

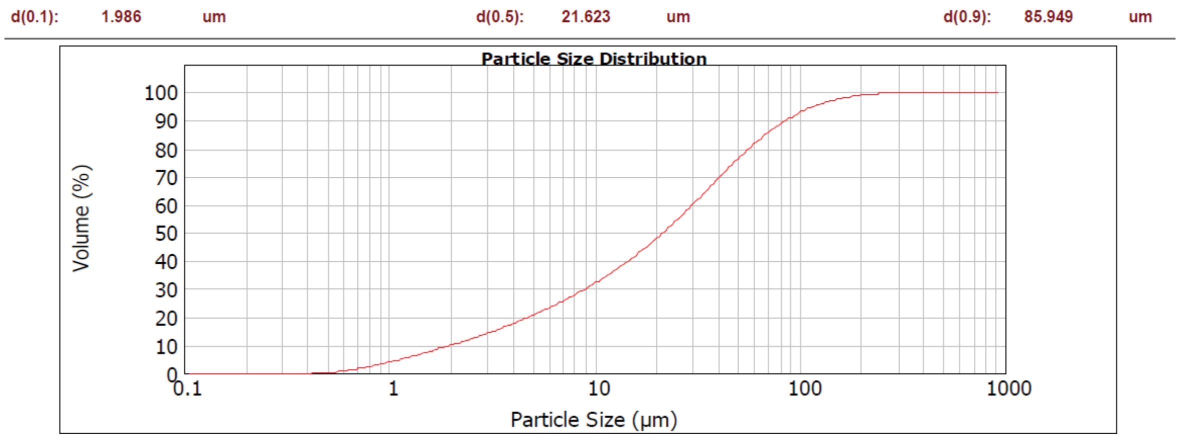


FIG 12. Particle size after 14 hrs of grinding

4. Conclusion

After 14 hours of grinding followed by Sonication in ethanol atmosphere, which induces atomic vibration resulting increase in density (due to RGO penetrating into the interstitial spaces in the material) and found to be 4.3442 gm/cc which is 10.49% higher than the theoretical value i.e. 3.9315 gm/cc. Also we found that after grinding, 50% particle size is less than 21.6 µm and 90% particle size is less than 85 µm whereas without grinding 50% particle size less than 92 µm and 90% particle size is less than 165 µm.

In this work, it is evident that an increase in density from the theoretical value by addition of MgO and RGO is 10.49% when RGO is 0.04% MgO is 5% and the overall attempt has been taken in work to correlate RGO content with various microstructural properties and density. The composite with improving mechanical properties has enormous potential scope for various advanced ceramic, structural applications, and aerospace due to preferred orientation of the atoms .

REFERENCES

[1] K. Niihara, New design concept of structural ceramics-ceramics nanocomposites. J. Ceram. Soc. Japan, 99 (1999), 974-982.

[2] Centeno, V.G. Rocha, B. Alonso, A. Fernández, C.F. Gutierrez-Gonzalez, R. Torrecillas and A. Zurutuza, Graphene for tough and electroconductive alumina ceramics, J. Eur. Ceram. Soc., 33 (2013), 3201–3210.

[3] Y. Wu, X. Zhang and J. Guo, Microstructural development and mechanical properties of self-reinforced alumina with CAS addition, J. Eur. Ceram. Soc., 2 (2001), 581–587.

[4] A. G. Evans, Perspective on the development of high-toughness ceramics, J. Am. Ceram. Soc., 73 (1990), 187-206.

[5] J. R. Martinlli and F. F. Sene, Electrical resistivity of ceramic–metal composite materials: application in crucibles for induction furnaces, Ceram. Int., 26 (2000), 325-335.

[6] L. Osayande and I. Okoli, KIC enhancement for alumina system: a review, Int. J. App. Ceram. Technol., 5 (2008), 313–323.

[7]. K. S. Novoselov, A.K. Geim, A. K, S.V. Morozov, D. Jiang, M.I. Katsnelson, I.V. Grigorieva, S.V. Dubonos and A.A. Firsov, Two-dimensional gas of massless Dirac fermions in graphene, Nature. 438 (2005), 197–200.

[8]. S.V. Morozov, K. Novoselov, M. Katsnelson, F. Schedin, D. Elias, J. Jaszczak, A. Geim, Giant Intrinsic Carrier Mobilities in Graphene and Its Bilayer, Physical Review Letters, 100 (2208), 016602.

[9]. J.H. Chen, Chaun Jang, Shudong Xiao, Masa Ishigami and Michael S. Fuhrer (2008), Intrinsic and Extrinsic Performance Limits of Graphene Devices on SiO, Nature Nanotechnology, 4 (2008), 016602.

[10]. L. S. Walker and E. L. Corral, Toughness in Grapheme Ceramic Composites, ACSNANO, 4 (2011), 3182-3190.

[11]. B. Yazdani, F. Xu, I. Ahmad, X. Hou, Y.Xia and Y. Zhu, Scientific Reports, 5 (2015), 1579: 1-11.