Journal of Materials and Environmental Science
ISSN : 2028-2508
CODEN : JMESCN

J. Mater. Environ. Sci., 2020, Volume 11, Issue 6, Page 844-855

http://www.jmaterenvironsci.com

Effect of reinforcement of reduced graphene oxide on Mechanical Properties of Concrete nanocomposite

Bayisa Meka Chufa[1], H. C. Ananda Murthy[1*]

[1*]*Department of Applied Chemistry, School of Applied Natural Science, Adama Science and Technology University, P.O. Box 1888, Adama, Ethiopia.*

Received 28 Dec 2019
Revised 09 May 2020
Accepted 10 May 2020

Keywords
- ✓ Reduced graphene oxide
- ✓ Concrete
- ✓ Nanocomposite
- ✓ Mechanical properties
- ✓ Micro-cracks

anandkps350@gmail.com ;
Phone: +251988683640;
Fax: +251-221-100038

Abstract
Currently, a new dimension in the construction world is being explored to apply nanotechnology and characterize the properties of concrete cementitious material at nanoscale level. This study presents experimental investigation of mechanical properties of reduced graphene oxide (rGO) reinforced structural concrete nanocomposite. rGO was synthesized using modified Hammer`s method and characterized by SEM and XRD. 0.1 %, 0.5 %, 1 %, 2 %, and 5 % of cement, rGO reinforcement were design mixed with concrete and casted into cubic, cylinder and rectangular test specimens. 0.08% polycarboxylate ether disperser was used to uniformly disperse rGO in paste. Three sets of concrete strength tests: compressive, flexural and tensile tests were performed to characterize mechanical properties of concrete. The results indicate as compared to control specimen, 0.5% of rGO flake specimens exhibited gains in concrete compressive strength by 44.3% and 38.8% for cube and cylinder specimens, respectively. Similarly, observed increments for flexural and tensile strength tests were 51.6% and 73%, respectively. rGO reinforcement has proved to increase concrete strength by limiting formation of micro-cracks however as percentage of rGO increases beyond 0.5%, concrete strength decreases due to rGO agglomeration and dissociation of bond between mortar paste and aggregates.

1. Introduction
All elements of nanotechnology have revolutionized the world. Another innovative revolution is the use of nano particles in building products. This is based on physical, chemical, electrical, optical and mechanical characteristics of nanoparticles. The variables influencing the mechanical characteristics of concrete between reactive paste and inert aggregates require a microscopic level or nano level of research. Inertness is permeable to characteristics of air and water that have a higher effect on concrete structures' strength and durability. The presence of nanoparticles in concrete could decrease the porous nature of mortar, boost the binding force between pastes and aggregate to avoid early cracking, enhance the compressive strength, flexural strength and tensile strength and make it more durable. Reduced graphene oxide nanoparticle is a better option to reinforce concrete. The chemical composition of reduced graphene oxide is carbon, oxygen and hydrogen [1].

Concrete is one of the building industry's most prevalent manmade materials. Due to fine and coarse aggregates such as sand and natural gravel which effectively bear the compressive load so that conventional concrete is comparatively powerful against compression. Traditional concrete, however, has heterogeneous micro and mesoporous structures through the random packing of concrete aggregate mixture, cement, and water where the micro cracks begin. Because of the complicated inner pore composition of the concrete between mortar and aggregates, it is therefore soft in tension and flexure.

Carbonaceous nanomaterials are therefore the perfect materials for improving soft tensile strength and stopping concrete structure crack formation at its early stage. There are several possible reasons for improvement of mechanical characteristics of concrete. First, materials of nano size fill the pores of cement and serve as concrete packing materials. Secondly, by binding tightly to the cement hydrate, nanoparticles (NPs) encourage cement hydration. Lastly, nanoparticles prevent the development of big crystals like $Ca(OH)_2$ [2] effectively.

Graphene is produced from powdered graphite. To generate graphene, graphite is exfoliated in layers. Graphene has demonstrated to be even tougher than diamond as the strongest material. Graphene's elastic module is about 1000 MPa [3] and young`s modulus of 1 TPa [4]. This study is therefore attempting to impart these powers to building materials. Graphene is not easily available in Ethiopia however the method for preparing the graphene and its family materials is available. GO and rGO are the most common graphene families with comparable strength [5]. Ordinary Portland cement (OPC) is the single most commonly used building material in the globe as the main ingredient in concrete manufacturing process. The main drawbacks of OPC are its intrinsic brittleness, which is liable for low cracking resistance, low tensile strength owing to inner faults and low strain ability. Such constraints can be overcome by embedding in bulk material either macro-dimensional stages such as steel bars or micro-dimensional stages such as fibres, which can give the material tensile strength and enhance its cracking resistance and damage tolerance by distributing the load applied to ductile reinforcements by developing energy dissipation.

Recent developments in nanotechnology have identified nano-scale materials as promising candidate reinforcements for cement composites of the next generation with enhanced mechanical efficiency as well as multi-functional characteristics such as heat and electrical transport capacity [6]. Therefore, for this study, flexural, compressive and tensile tests were carried out on specimens fabricated by introduction of the reduced graphene oxide mixing in water by aid of a dispersing agent. The mechanical response of the casted concrete was discussed in view of the particularities in rGO percentage. Reduced graphene oxide is regarded in terms of tensile strength and elastic module as the strongest and most rigid materials. Therefore, they would improve the mechanical properties of the construction materials. Also, the small size of reduced graphene oxide should make them fill the cement pores and interrupt crack formation and growth at very early stages. On the other side, the high aspect ratio of reduced graphene oxide would play a part in stopping crack propagation, as the crack around these carbonaceous nanomaterials would require greater energy [7]. In addition, reduced graphene oxide can be chemically functionalized to interact with adjacent cement component. Therefore, this research work is focused on prevention of crack and modification of the weak tensile and flexural nature of concrete by the incorporation of reduced graphene oxide. Hence, the effect of this emerging carbon nanostructure, reduced graphene oxide, on the mechanical properties of concrete was investigated.

In this study, the influence of reduced graphene oxide (rGO), on mechanical properties such as compressive, flexural and tensile strength of concrete was determined after curing for 28 days. Further, the synthesized nanomaterials were characterized using X-ray Diffraction technique (XRD), and Scanning Electron Microscopy (SEM) techniques.

2. Material and Methods

2.1. Sample preparation

Cement

The common materials used in this study includes cement, sand, fine and coarse aggregate, reduced graphene oxide, polycarboxylate ether and water. In this experiment, ordinary Portland cement of grade 32.5R, CEM 11 (IS certified number 120003) supplied by Muger cement factory. Portland cement is a finely ground powder chemically formed by combining raw materials containing calcium oxide (CaO), silica (SiO_2), alumina (Al_2O_3), and iron oxide (Fe_2O_3), heating this mixture to a high temperature, and then grinding the resulting material called clinker, with a small quantity of calcium sulfate ($CaSO_4$). The cement was in light grey colour with good chemical and physical characteristics. The cement for entire experiment was procured in a single consignment and stored properly. It contains 95% clinker and 5% gypsum.

Sand

Locally available fresh river sand, free from organic matter was used as per IS 456-2002 [8]. The sand was air dried and free from any foreign material, earlier than mixing. Sand particles mostly range from 0.02 mm to 2.00 mm in diameter. Silica sand is the term used to describe sand that has a very high percentage of silicon dioxide (SiO_2).

Coarse Aggregates

Locally available good quality coarse aggregate from crushed ballast rock were used. The size of coarse aggregate varies from 10mm to 20mm, i.e., the material passed from 20mm IS sieve but retained in 10mm IS sieve. The aggregates were free from adherent coating, injurious amount of disintegrated pieces, alkali, vegetable matter and other deleterious substances. Care was taken that the aggregate does not contain high concentration of flaky, elongated shapes and organic impurities which might affect the strength or durability of concrete. Aggregates are known to be particles of rock or equivalent which, when brought together in a bound or unbound condition, form part or whole of an engineering or building structure. Aggregates, both fine and coarse, take about 65-75% by volume of concrete and are important ingredients in concrete production. The parent materials of aggregates are derived mainly from volcanic activity. The dominant rock for coarse aggregate production in Ethiopia is generally basalt while ignimbrite is most commonly used for masonry stone.

It is an established fact that the compressive strength of concrete is influenced by, among other things, the quality and proportion of fine and coarse aggregate, the cement paste and the paste-aggregate bond characteristics. These, in turn, depend on the macro- and microscopic structural features including total porosity, pore size and shape, pore size distribution and morphology of the hydration products, and the bond between individual solid components. Other qualities of concrete such as durability and abrasion resistance are also highly dependent on the aggregate, which in turn depends on strength of parent rock, purity, surface texture, gradation and so on.

Water

The quality of water is important because contaminants can adversely affect the strength of concrete and cause corrosion of the steel reinforcement. Water used for producing and curing concrete should be reasonably clean and free from deleterious substances such as oil, acid, alkali, salt, sugar, silt, organic matter and other elements which are detrimental to the concrete. Hence potable tap water was used in this study for mixing and curing.

Reduced Graphene Oxide nanoparticles

Reduced graphene oxide (RGO) is the form of GO that is processed by chemical, thermal and other methods in order to reduce the oxygen content, while graphite oxide is a material produced by oxidation of graphite which leads to increased interlayer spacing and functionalization of the basal planes of graphite [9]. Graphene oxide (GO) was prepared from natural graphite powder by a modified Hummers method. As prepared porous reduced graphene oxide (RGO) was synthesized by a plant extract reduction and heat treatment.

2.2. Experimental Design

The currently available and modified test methods used to evaluate the compressive, flexural and tensile strength of rGO reinforced concrete were studied. A trial mix proportions were used for mix of conventional concrete to achieve C25. The rGO nanoparticle was added on the basis of percentage to cement content of conventional concrete. C25 grade of concrete were designed to give compressive strength of 25MPa at the end of 28 days. Cube, cylinder and prism molds of size 150 mm * 150 mm * 150 mm, 150 mm dia. and 300 mm height, and 100 mm * 100 mm * 500 mm respectively were casted. The moulds were oiled properly prior to the casting of the specimens.

The high strength of C25 concrete grade was produced using single mix series besides the control mix. The molds were casted after the incorporation of 0.1% 0.5%, 1.0% 2% and 5% rGO nanoparticles. The volume percentage of rGO is based on the recommendation by earlier researchers [10-13]. As already discussed above, the main factors controlling the mechanical performance of the obtained composite materials are the properties of the reinforcement and the matrix, as well as the bond between them. For this reason, investigation of the properties of the constituent material and preliminary characterization of rGO were made to obtain the necessary data about their geometric and mechanical properties. Tests were conducted on the constituent material to determine the gradation and physical properties of fine and coarse aggregate as well.

The mixing was made in a fixed proportion and the pre synthesized and characterized rGO nanoparticles were added to the mixture replacing cement by five different percentages. Mixing was done by hand as per ASTM C94 standard. The mixed samples were casted followed by the application of pressure and after limited amount of water was added. All the specimens were demoulded after 24hr of casting and water cured for 28 days. At the specified date they were removed from water, surface dried and tested. Each test result represented the mean of two specimens of cube and cylinder each and two specimens of beam. Proportions of these mix series along with the volume of percentage of rGO contents are presented in Table 1.

Table 1: Mix proportions for the five mixtures

Cement quantity(kg/m^3)	W/C ratio	Water (liter/m^3)	Fine aggregate (kg/m^3)	Coarse aggregate (kg/m^3)	rGO (%)
108	0.46	50	214	324	0
	0.46				0.1
	0.46				0.5
	0.46				1
	0.46				2
	0.46				5

As we usually use in Ethiopian building industries; the ratio of cement, sand and coarse aggregate were 1: 1.99: 3. The water content to cement ratio used for the mix was 0.46. Then the performances of unreinforced concrete were evaluated in order to better appreciate the improvement gained by the addition of nanoparticles. Thereafter, the compressive strength, tensile strength and flexural strength in the hardened state were evaluated for each mix, varying the volume fraction of nanoparticles. The test results were then analysed and discussed and Conclusions and recommendations were provided for further studies.

2.3. Synthesis of Reduced Graphene Oxide (rGO)

Modified Hummer's Method was partially employed using natural graphite flakes as precursor for the synthesis of reduced graphene oxide [14]. Graphite flakes was added into sulphuric acid and the mixture was sonicated to produce a fine dispersion. This suspension was then heated for 6 hours at 80 °C with continuous stirring. Thus obtained pre-oxidized graphite was further added into sulphuric acid. Potassium permanganate was added slowly to this mixture with constant stirring keeping temperature of the mixture below 100 °C. The mixture was stirred under ice water bath for two hours. The reaction mixture was gradually thickened. This solution was then diluted by distilled water and treated with hydrogen peroxide and was kept undisturbed for 24 hours for precipitation. The upper supernatant was centrifuged and obtained mass was washed with HCl and distilled water repeatedly and finally dried.

The colour of the mixture turned gradually from dark brown to bright yellow. Then, the warm solution was centrifuged and washed with HCl (5 wt.%) and water. It was repeated until the pH value of the filtrate was close to 7 and no deposit appeared in the filtrate with $BaCl_2$ test.

Thus-prepared filtrate was dried in a vacuum oven at 60 °C for 2 days to obtain graphite oxide (Figure 1). The filter cake was re-dispersed in ethanol with mechanical agitation or sonication using an ultrasonic cell disruptor, giving a solution of exfoliated GO. Subsequently, GO solution was put into an autoclave and heated to 100 °C for 4 hr in the presence of plant extract to reduce GO to rGO. The as-synthesized product was isolated by centrifugation, washed with water and ethanol, respectively, and finally dried (as shown in Figure 2) in a vacuum oven at 60 °C for 24 h.

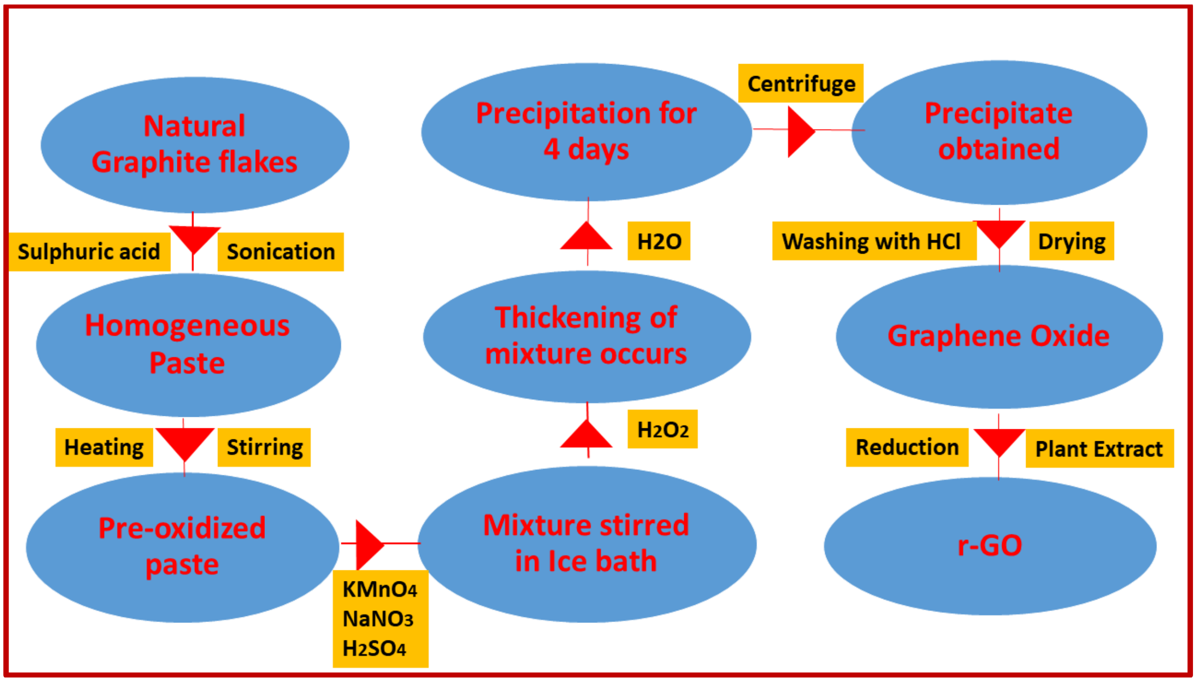


Figure 1: Schematic representation of synthesis of reduced graphene oxide

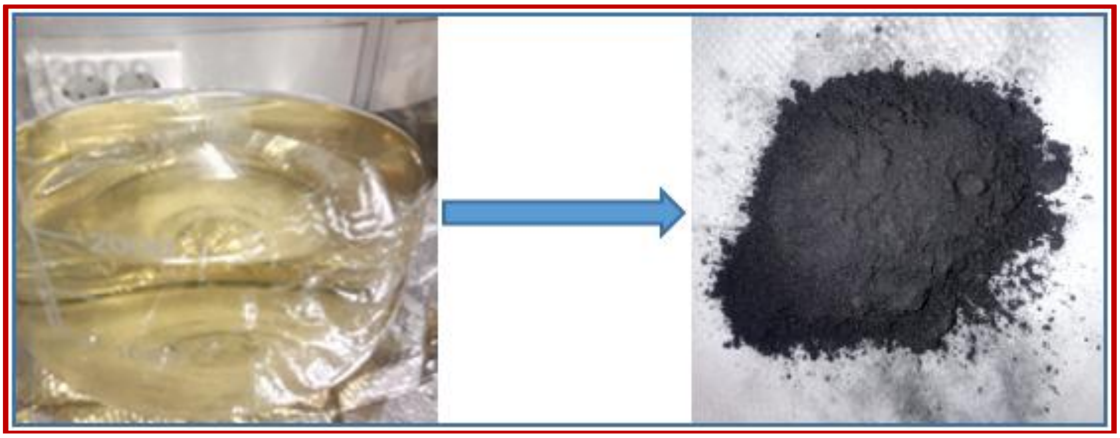


Figure 2: A Scheme of synthesis of reduced graphene oxide

2.4. Synthesis of Reduced Graphene Oxide-Concrete composite

To prepare the rGO - concrete composites, rGO is suspended in distilled water and sonicated for 3 h to obtain a homogeneous solution; cement (such as ordinary Portland cement, OPC) was then added to the mixture while the desired water-to-cement (w/c) ratio is maintained. Sand and aggregate was added to the rGO-cement mix thereafter. RGO–cement-based concrete composite production method [15] is schematically presented in Figure 3.

2.5. Preparation of Concrete Specimens

Forty-eight different specimens of concretes such as cube, cylinder and beam which contain a variety of percentage of rGO such as 0.1 percent, 0.5 percent, 1 percent, 2 percent, and 5 percent was prepared and get ready for tensile, flexural and compressive test. A specimen without rGO was also synthesized to be used as control. The standard size of concrete cube specimen is 150x150x150 mm (150 mm).

2.6. Characterization

As prepared materials were characterized by means of X-ray diffraction (XRD, Rigaku MinFlex, D/max 2550-PC) with Cu Kα radiation (λ=0.15406 nm). The data was collected between scattering angles (2θ) of 10-90° at a scanning rate of 2° min-1. The microstructure of rGO samples was observed by scanning electron microscopy (SEM, JEOL-5600LV). SEM specimens were prepared by drop casting the as prepared sample dispersions onto carbon coated SEM grids.

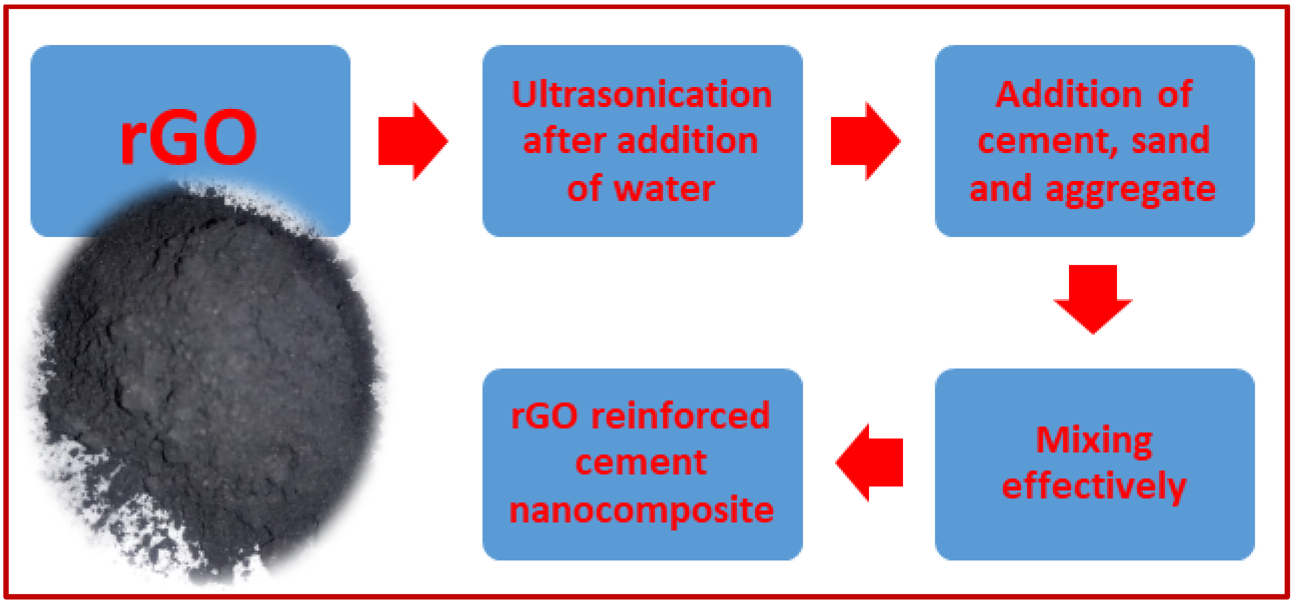


Figure 3: The schematic of Synthesis of Reduced Graphene Oxide-Concrete composite

3. Results and discussion

3.1. Morphology of rGO nanosheets

The morphology of rGO nanosheets were investigated through SEM observation. The figure 4a-d, presents the representative SEM images of free-standing rGO nanosheets, revealing a crumpled and rippled structure which was the result of deformation upon the exfoliation and restacking processes. The rGO nanosheets found to have layered structure with irregular folds [16]. They are entangled with each other. This figure showed the presence of lots of wrinkles in the layered rGO nanosheets. Corrugation and scrolling suggested the intrinsic nature of graphene, because the 2D membrane structure would be thermodynamically stable via blending [13,16]. Above all, this image depicted that rGO nanolayers with 100 nm dimension were formed.

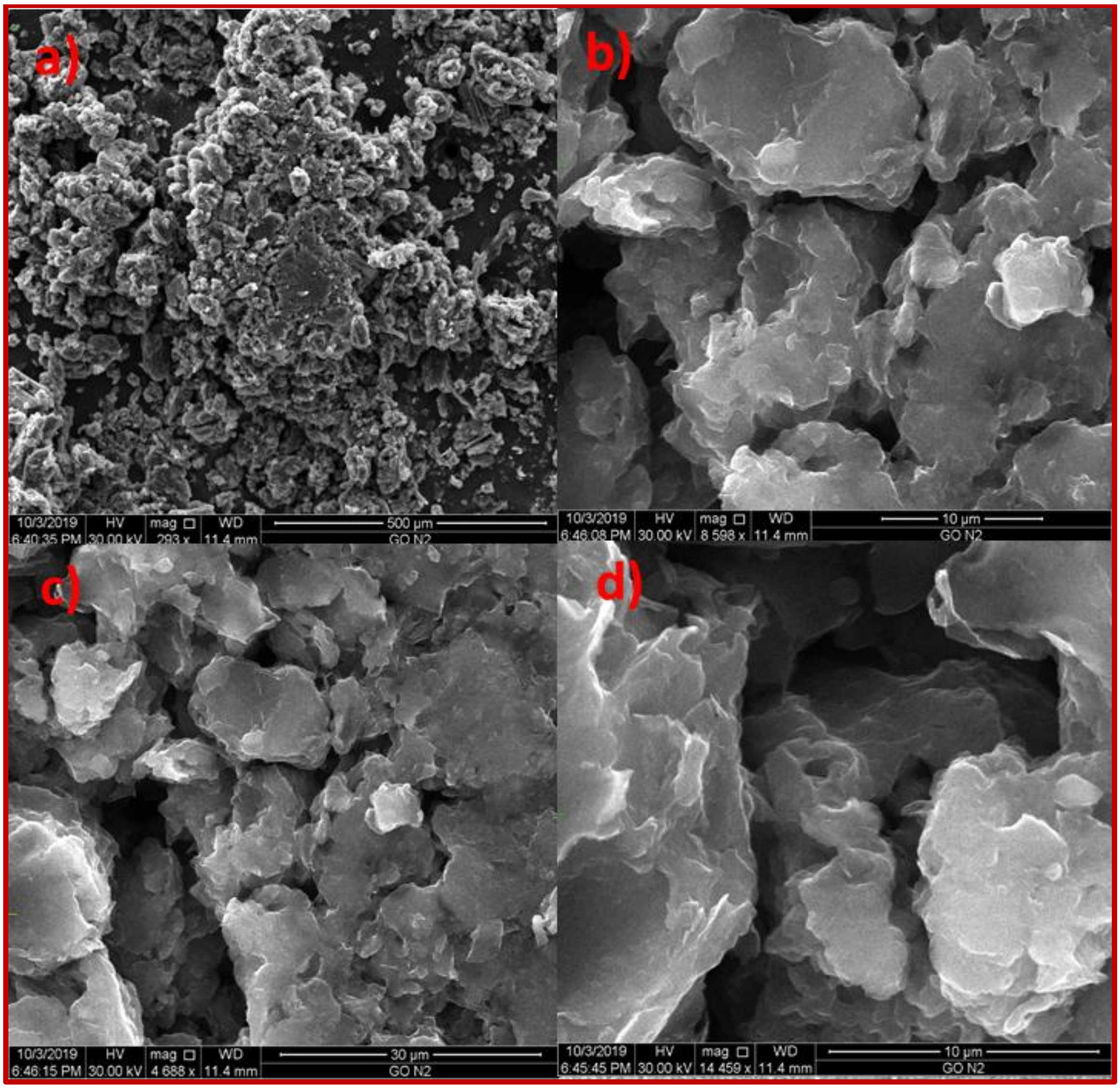


Figure 4: SEM micrographs (a-d) of rGO exhibiting layered structure

3.2. Crystal structure of rGO nanosheets

X-ray diffraction (XRD) analysis was carried out to observe rGO flakes dispersed in water, determine particle size and quantify the visual nature of the material in its raw form. XRD pattern of rGO nanosheets is presented in Figure 4. A very broad characteristic peak of rGO appeared at around 2θ= 25.2^{o} with an interlayer d-spacing of 0.337 nm confirms the conversion of GO to rGO [17,18].

3.3. Determination of Compressive Strength

Generally, three sets of strength testing were carried out: compressive, flexural and tensile testing's. Each set had at least six representative samples with/without rGO flakes. The compressive strength test was carried out in accordance with EBCS 3. A total of 24 test specimens (five cubes of the same size with 0.1 percent, 0.5 percent, 1 percent, 2 percent, and 5 percent rGO, two control specimen and five cylinders of the same size with 0.1 percent, 0.5 percent, 1 percent, 2 percent, and 5 percent rGO and two control specimens) were prepared for compressive strength test. The cubes and cylinders were casted according to Ethiopian Building Code Standard specification and tested at 28 days after casting. Table 2 shows the visual representation of the specimens and their corresponding compressive strength. The compressive strength test was carried out twice and the average value was taken for the comparison.

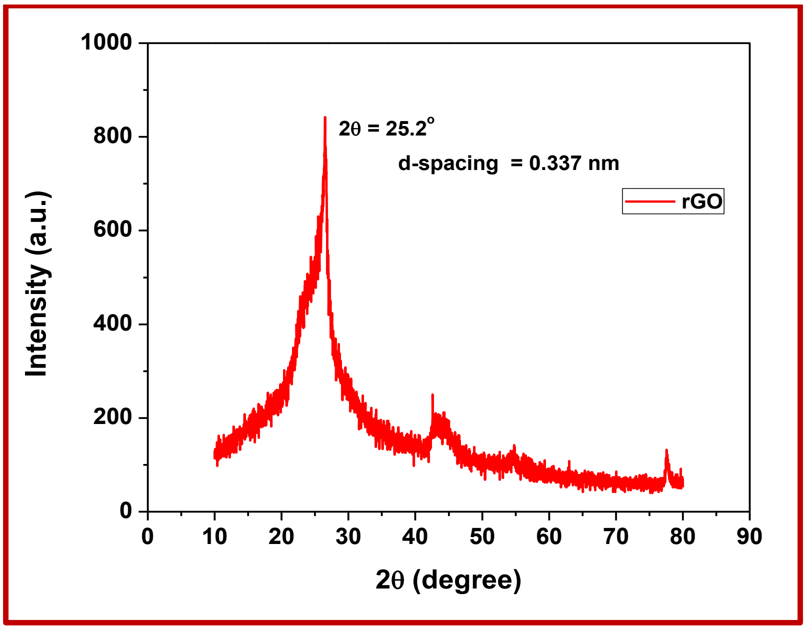
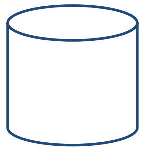
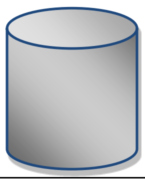
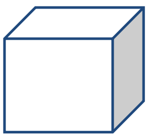
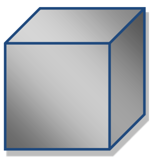


Figure 5: XRD pattern of rGO

Table 1: Visual representation of test cylinders and cube specimens for compression test

Specimen	rGO (%)	Average Compressive strength (MPa)	Dimension (mm)
	0	24.01	150 *300
	0.1	39.03	150 *300
	0.5	33.40	150 *300
	1	34.31	150 *300
	2	38.08	150 *300
	5	23.03	150 *300
	0	28.33	150*150*150
	0.1	40.34	150*150*150
	0.5	40.91	150*150*150
	1	38.81	150*150*150
	2	28.01	150*150*150
	5	31.1	150*150*150

As shown by Figure 5, the concrete mould failure was observed as a function of load per unit area of the cube. It was found that the average cube and cylinder strengths were increased by 44.3% and 38.8% respectively upon 0.5% rGO flakes reinforcement. This result was the maximum strength recorded for the rGO reinforcement used [19]. Beyond 0.5 %, the strength was found to decrease. The decrease could be due to thermal crack induced by the aggregation of rGO during the hydration process as the percentage rises with fixed disperser.

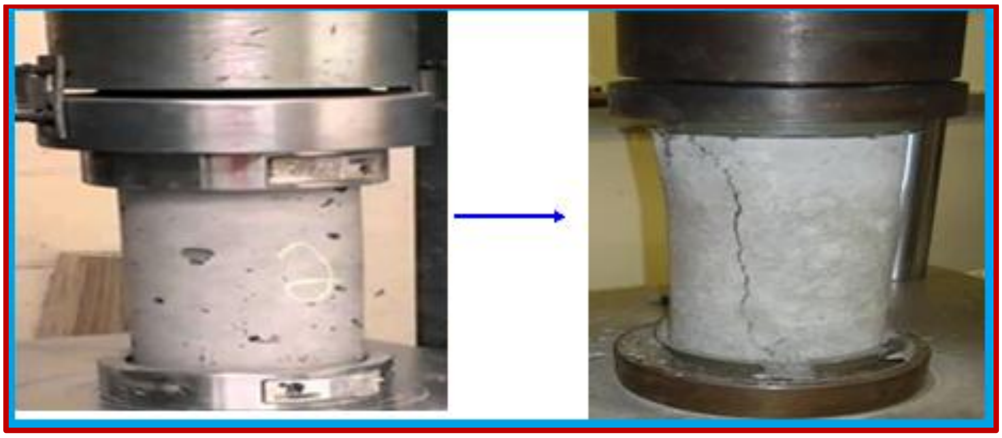


Figure 5: Test specimens during and after compressive test

3.4. Determination of Flexural strength

As presented in Table 3, different percentages of rGO were used to cast the rectangular beam and the test was done at the end of 28 days. Above all, the 0.5% rGO flakes enhanced the flexural strengths of the beams by 51.6% which is the optimum reduced graphene oxide percentage reinforcement to be used in the concrete reinforcement. Beyond 0.5% rGO reinforcement, the flexural strength decreases due to the aggregation of the nanoparticles and dissociation of bond between mortar and aggregates [20, 21].

Table 2: Visual representation of test rectangular specimens for flexural test

Specimen	RGO (%)	Average flexural strength (MPa)	Dimension (mm)
	0	4.17	100 * 100 * 500
	0.1	5.26	100 * 100 * 500
	0.5	6.32	100 * 100 * 500
	1	5.00	100 * 100 * 500
	2	4.44	100 * 100 * 500
	5	4.41	100 * 100 * 500

Figure 6: shows the setup for a three-point bending test and the failures of the beams.

Tensile strength

Table 4 shows the visual representation of the specimens and their corresponding tensile test. The average tensile strength was found to decrease as value of rGO is increased beyond 0.5% of cement used. The decrease can be attributed to thermal crack induced by the temperature rise during the hydration process. However, the tensile strength was increased by 73% at 28 days, compared with the specimens without rGO flakes at 0.5% rGO load. The 73% increase at 28 days after casting by rGO is significantly greater than the 18% as reported [22-25]. The difference could be due to application of different exfoliating techniques. Hummers method to exfoliate GO flakes, while the rGO flakes used in this research were exfoliated using environmentally friendly techniques and hence no harmful chemicals were used. The difference may also come from the use of disperser used. Hence, it is also possible to include that the intrinsic tensile strength performance of rGO reinforced concrete is greater than that of GO. The observed tensile strength failure is presented in Figure 7.

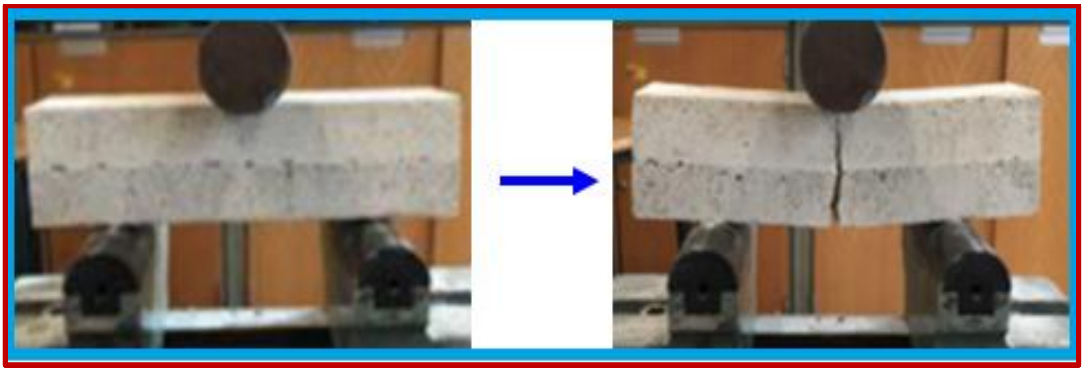


Figure 6: Test specimens during and after flexural test

Table 3: Visual representation of test cylinder specimen for tension test

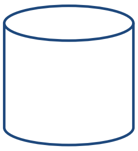
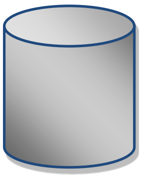
Specimen	RGO (%)	Average tensile strength (MPa)	Dimension (mm)
	0	2.16	150 *300
	0.1	2.22	150 *300
	0.5	3.74	150 *300
	1	2.08	150 *300
	2	2. 61	150 *300
	5	1.91	150 *300

Figure 7: Test specimens during and after tensile test

Conclusion

The concrete was successfully reinforced with the incorporation of 0.1%, 0.5%, 1%, 2% and 5% of reduced graphene oxide to investigate the mechanical properties of produced nanocomposite. The result showed that addition of 0.5% rGO enhanced the mechanical properties of concrete. Images from SEM and XRD analyses indicated formation of rGO nanosheets. The addition of rGO proved to enhance compressive, flexural and tensile strength of concrete nanocomposite. The increase was up to 51.6%, 38.8 & 44.3%, and 73% for the flexural, compressive (cylinder and cube) and tensile strength, respectively. One of the investigations of the variability of compressive strength in shape has revealed that rGO accelerates the cement hydration, in particular the early age hydration and also contributes to shape factor. Notably, the significant improvement of mechanical properties has been achieved with simultaneous low material consumption. Such findings indicate excellent potential of reinforcing cementitious composites with rGO and represent a step forward towards practical applications of nanomaterials in civil engineering. GO is better in dispersion capacity in water than rGO but it is less in strength. GO dehydrates water and facilitate the formation of $Ca(OH)_2$ crystals which is the cause for the micro crack formation. The inclusion of the rGO flakes in general led to positive performance gains in concrete's mechanical property as compared to control specimen and favours the formation of strong C–S–H bond. The incorporation of 0.08% superplasticizer was necessary to facilitate the dispersion of the GO flakes.

Acknowledgement-Authors are grateful to Adama Science and Technology University, Ethiopia for support towards carrying out this work and publication of this work.

References

1. V. R. J. Antonio, C. S. German, Optimizing content graphene oxide in high strength concrete, *International Journal of Scientific Research and Management*, 4 (2016) 4324–4332. https://doi.org/10.18535/ijsrm/v4i6.14A.
2. J. Kang, K. Kim, Y.M. Lim, J. E. Bolander, Modeling of fiber-reinforced cement composites: Discrete representation of fiber pullout, *International Journal of Solids and Structures*, 51(10) (2014) 1970–1979. https://doi.org/10.1016/j.ijsolstr.2014.02.006 .
3. A. Romero, M. P. Lavin-lopez, L. Sanchez-silva, J. L. Valverde, A. Paton-carrero, Comparative study of different scalable routes to synthesize graphene oxide and reduced graphene oxide. *Materials Chemistry and Physics*, 203 (2018) 284–292. https://doi.org/10.1016/j.matchemphys .
4. N. I. Zaaba, K. L. Foo, U. Hashim, S.J. Tan, W.W. Liu, C.H. Voon, Synthesis of Graphene Oxide using Modified Hummers Method: Solvent Influence, *Procedia Engineering*, 184 (2017) 469–477. https://doi.org/10.1016/j.proeng.2017.04 .
5. N. Hashim, Z. Muda, M. Z. Hussein, I. M. Isa, Mohamed, A. Kamari, S.A. Bakar, M. Mamat, A.M. Jaafar, A brief review on recent graphene oxide-based material nanocomposites: Synthesis and applications, *Journal of Materials and Environmental Science*, 7(9) (2016) 3225–3243.
6. Yoo, Doo-Yeol, Ilhwan Lee, Seung-Jung, Electrical Properties of Cement-Based Composites with Carbon Nanotubes, Graphene, and Graphite Nanofibers. *Sensors*, 17 (2017) 1064-1076. https://doi.org/10.3390/s17051064.
7. L. Lu, D. Ouyang, Properties of Cement Mortar and Ultra-High Strength Concrete Incorporating Graphene Oxide Nanosheets. *Nanomaterials*, 7(7) (2017) 187-198. https://doi.org/10.3390/nano7070187
8. FJ. An, M. McInnis, W. Chung, B.H. Nam, Feasibility of using graphene oxide nanoflake (GONF) as additive of cement composite. *Applied Sciences* (Switzerland), 8(3) (2018) 419. https://doi.org/10.3390/app8030419 .
9. P. Solís-Fernández, M. Bissett, H. Ago, Synthesis, structure and applications of graphene-based 2D heterostructures. *Chemical Society Reviews*, 46 (2017) 4572–4613. https://doi.org/10.1039/C7CS00160
10. S. Imani Yengejeh, S.A. Kazemi, A. Öchsner, Carbon nanotubes as reinforcement in composites: A review of the analytical, numerical and experimental approaches. *Computational Materials Science*, 136 (2017) 85–101. https://doi.org/10.1016/j.commatsci.2017.04.023 .
11. G. Jing, Z. YX. Lu, P. Hou, Effect of graphene Nano platelets on hydration behavior of Portland cement by thermal analysis. *Advances in Cement Research*, 29(2) (2017) 63–70. https://doi.org/10.1680/jadcr.16.00087 .

12. M. Krystek, Mechanical Properties of Cement Mortar with Graphene Oxide. Architecture, Civil Engineering, *Environment*, 12(1) (2019) 91–96. https://doi.org/10.21307/acee-2019-008
13. C. Phrompet, C. Sriwong, C. Ruttanapun, Mechanical, dielectric, thermal and antibacterial properties of reduced graphene oxide (rGO)-nanosized C3AH6 cement nanocomposites for smart cement-based materials, *Composites Part B* 175 (2019) 107128. https://doi.org/10.1016/j.compositesb.2019.107128
14. D. Dimov, I. Amit, O. Gorrie, M. D. Barnes, N. J. Townsend, Ana I. S. Neves, F. Withers, S. Russo, M. F. Craciun. Ultrahigh Performance Nano engineered Graphene–Concrete Composites for Multifunctional Applications, *Advanced Functional Materials*, 2018, 1705183. https://doi.org/10.1002/adfm.201705183 .
15. Gholampour, Ali A , Meisam Tran, Diana, Togay,. From Graphene Oxide to Reduced Graphene Oxide: Impact on Physiochemical and Mechanical Properties of Graphene–Cement Composites. *ACS Applied Materials & Interfaces*, 9 (2017) 43275-43286. https://doi.org/10.1021/acsami.7b16736.
16. A. Hassani, B. Fakhim, A. Rashidi, P. Ghoddousi, Preparation and Mechanical Properties of Graphene Oxide: Cement Nanocomposites, *Scientific World Journal*, Volume 2014, Article ID 276323, 10 pages. http://dx.doi.org/10.1155/2014/276323.
17. S. Ahmadreza, Manoj K, Ram, A Zayed, Kamal Rajeev, Shanahan Natallia. Investigation of physical properties of graphene-cement composite for Structural Applications. J*ournal of Composite Materials*, 4 (2014) 12–21. DOI: 10.4236/ojcm.2014.41002.
18. A. Gholampour, M. Valizadeh Kiamahalleh, D. N. H. Tran, T. Ozbakkaloglu, D. Losic, Revealing the dependence of the physiochemical and mechanical properties of cement composites on graphene oxide concentration. *RSC Advances*, 7 (2017) 55148-55156 DOI:10.1039/c7ra10066c.
19. C. Lu, Z. Lu, Z. Li, C.K.Y. Leung, Effect of graphene oxideon the mechanical behavior of strain hardening cementitious composites. Construction Building. Materrials 120 (1) (2016) 457−464. DOI: 10.3390/ma12223753.
20. M. Saafi, L. Tang, J. Fung, M. Rahman, J. Liggat, Enhanced properties of graphene/fly ash geopolymeric composite cement. *Cement Concrete Research*. 67, (2015) 292−299. DOI: 10.1016/j.cemconres.2014.08.011.
21. M. Cao, H. Zhang, C. Zhang, Effect of graphene on mechanical properties of cement mortars. *Journal of Central South University*, 23 (2016) 919−925. https://doi.org/10.1007/s11771-016-3139-4.
22. Rafiee M., Nitzsche F., Laliberte J., Thibault J., Labrosse M.R. Simultaneous Reinforcement of Matrix and Fibers for Enhancement of Mechanical Properties of Graphene-Modified Laminated Composites. *Polymer Composites*, 40 (2019) E1732–E1745. DOI: 10.1002/pc.25137.
23. Qureshi T.S., Panesar D.K. Impact of graphene oxide and highly reduced graphene oxide on cement based composites. *Construction and Building Materials*, 206 (2019) 71–83. doi: 10.1016/j.conbuildmat.2019.01.176.
24. Li X., Liu Y.M., Li W.G., Li C.Y., Sanjayan J.G., Duan W.H., Li Z. Effects of graphene oxide agglomerates on workability, hydration, microstructure and compressive strength of cement paste. *Construction and Building Materials*, 145 (2017) 402–410. DOI: 10.1016/j.conbuildmat.2017.04.058.
25. Zhao L., Guo X., Liu Y., Zhao Y., Chen Z., Zhang Y., Guo L., Shu X., Liu J. Hydration kinetics, pore structure, 3D network calcium silicate hydrate, and mechanical behavior of graphene oxide reinforced cement composites. *Construction and Building Materials*, 190 (2018) 150–163. DOI: 10.1016/j.conbuildmat.2018.09.10.

(2020) ; http://www.jmaterenvironsci.com