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Influence of Graphene Oxide on Cementitious Composites – A Review

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Abstract— ***Graphene is a 2-dimensional monoatomic carbon allotrope with distinguished properties that can prove to be highly beneficial for cementitious composites. Inclusion of derivatives of this material in cementitious composites has proven to be advantageous from the standpoint of mechanical, rheological and thermal properties. This paper provides a comprehensive overview of structure, functionalisation, fabrication along with variation in properties of the cement composites with addition of Graphene Oxide (GO).***

Keywords— ***Graphene oxide, Cement composite, Durability, Nano materials, Functionalization***

I. INTRODUCTION

Ordinary Portland cement is the most popular choice of construction material whose global production stood at 4200 million tons in 2016 and is expected to be around 4.4 billion metric tonnes by 2020 [1]. Although, cement concrete possesses the significant mechanical strength and durability properties, its weakness in terms of tensile load bearing capacity and porous structure restricts its applications in the construction industry. Customary approaches used to overcome these drawbacks of concrete include introduction of different types of materials into the concrete such as bars and fibres of steel and carbon etc. Although the method introduction of these materials proved to be highly efficient, these materials aided the rise of other problems. Reinforcement provided is highly susceptible to the corrosion, whereas the fibers in large amount reduced the workability of the mixture. All these approaches varied the structure and composition of the concrete at a macro level without any variation in the microstructure of the concrete. The strength and durability properties of the concrete are largely dependent on the formation of calcium silicate hydrate gel (CSH gel). Therefore, an approach that improves the microstructure of the concrete to enhance the mechanical properties and durability has to be taken. For the regulation of the microstructure of the concrete, the materials used must have very high strength, great specific surface, good dispersion in water and most importantly the material should be capable of forming strong chemical bonds with calcium silicate hydrate gel. With the advancement in nanotechnology, nanomaterials such as nano silica, nanotitanium oxide, carbon nanotubes (CNT), graphene etc have been newly synthesized. The properties of various materials are listed in table 1. Among the various nanomaterials available, Graphene and Graphene oxide demonstrate properties that prove highly useful for the application of cementitious materials. Additionally, dispersion of Graphene oxide is very easy to form stable dispersions which also have the ability to form the composites because of the presence of functional O2. The biggest advantage of using a nano material is their large surface area compared to the conventional materials. This property can be exploited to ensure complete reaction of with the cement paste. Nevertheless, this is advantageous as seeding sites for hydration products, the problem of agglomeration comes into existence. Consequently, additional requirement of water to maintain same workability becomes necessary. These nano particles also provide filling effect which makes the microstructure dense and compact.

TABLE I
COMPARISON OF VARIOUS OF PROPERTIES OF DIFFERENT NANO MATERIALS USED IN THE CONSTRUCTION INDUSTRY

Type of material	Youngs Modulus(TPa)	Density(kg/m^3)	Particle size/thickness (nm)	Surface Area(m^2/g)	Reference
Graphene	1.033-1.042	2200	0.08	2600	[2] ,[3]
Graphene Oxide	0.023-0.042	1800	0.67	700-1500	[4],[5]
Silica fume	-	2400	6	170-200	[6]
CNT	0.95	1330	15-40	70-400	[7],[8]
Titanium dioxide Nano particle	-	4230	15-30	50	[9]
Fiber (S-glass)	72	2540	5000-10000	0.3	[10]

The properties of the graphene have attracted the researchers all around since 2010 because of its superior properties that enables its application in a wide range of engineering areas. This material is made up of one layer of carbon atoms and surprisingly, it is stronger than steel and is more flexible and lighter. Graphene is the thinnest material, with carbon to carbon bond which makes it gives it unimaginable mechanical strength and makes it the stiffest material known to the humankind. Also, the material is known for its very high surface area. Graphene is relatively a new material for the construction industry which has the potential to be used to reinforce polymers. Recently, significant efforts were made to incorporate graphene as nano filler in polymer matrices. Various studies have shown the significant improvement in modulus of elasticity, tensile strength etc. Large surface area of graphene can be is a very powerful parameter to tailor the composite material based on the requirement of the job at hand. In addition to this, thermal and electrical properties of graphene brings new applications to the construction field especially to the transportation infrastructure where phase change materials which absorb and significant amount of energy without significant change in their temperature. This ability of PCMs is harnessed to de-ice during cold weather conditions. The effectiveness of the PCMs depends on the distance at which they are placed to the surface as the asphalt surfaces are provide great thermal resistance. High quality graphene and graphite could be incorporated with the asphalt mix which improves the thermal conductivity. Also, the improvement in thermal diffusivity and electrical conductivity of the concrete were observed with increasing graphene content.

Cement composite is the most widely used construction material which has the ability to resist high compressive stress. However, intrinsic brittleness that is associated with the concrete that makes it vulnerable to the tension and flexure. So far, various methods have been tested to make up for the brittleness concrete which includes incorporation of reinforcing bars, polymer fibres, mineral fibres etc. Although all these methods were successful in improving the tensile strength of concrete, the structures are susceptible to the formation of cracks when tensile stresses are generated. This drawback is mainly because of over reliance on toughening methods without due consideration to the microstructure. Graphene has great surface to volume ratio with very high mechanical strength which improves the microstructure of the cement. A study conducted at Monash University showed that incorporation of graphene oxide tremendously improved the mechanical and physical properties of concrete. With the incorporation of 0.05 % of graphene oxide in OPC, the bending strength of concrete was improved by approximately 50 % and compressive strength improved by 14-15% [11]. The improvement in thermal diffusivity was because of the heat sink capacity of the graphene. This is particularly important during the initial hydration of cement which releases significant heat thus reducing the risk of thermal cracking. Consequently, the durability of the concrete structure improves. The enhancement of the electrical conductivity of the concrete is desirable where dissipation of charge is necessary.

II. STRUCTURE AND ORBITAL INFORMATION OF GRAPHENE

The material graphene is derived using a monolayer of Carbon atoms which are closely packed in a matrix which forms a two dimensional honeycomb structure. This material is the basic building block of all the graphite based materials such as fullerenes, carbon nanotubes etc. [12]

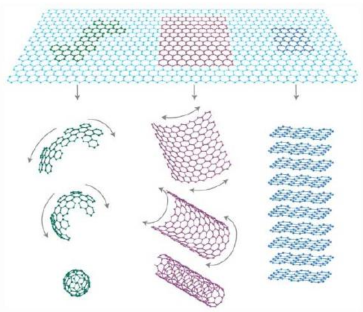

Fig. 1. Basic structure of Graphene showing zero-D buckyballs, one-D nanotubes; stacking.[12]

The strength and modulus of elasticity were found to be in the ranges of 100-130 GPa and 1-1.2 TPa respectively [13]. The configuration of graphene has been widely reported to be planar which is highly advantageous for the formation of proper bond between the surfaces of the host. From table 1, it can be observed that the specific surface of this material is very large which makes increases the interactions between the graphene and other constituent materials forming the composite. Consequently, the bond formation of graphene and the constituent materials grows stronger. The only limitation associated with graphene is the issue of economical mass production to be used in the construction industry [14]. Therefore, researchers started examining the Graphene derivatives that can be used in the construction industry. One strong contender that emerged that was deemed to be fit was for the construction industry was Graphene Oxide which too is a monolayer of carbon atoms that is densely packed and has the similar hybridization of Graphene. The additional groups associated with this material is hydroxyl (OH-) and epoxy(-O-) functional groups and carboxylic acid groups at the edge [15].

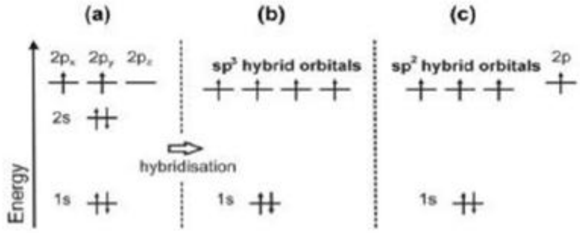

Fig.2: Hybridization of Carbon [16]

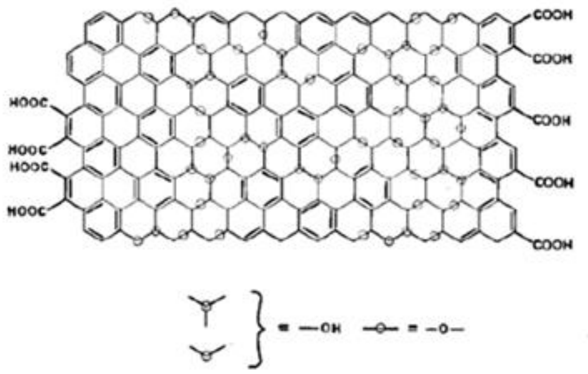

Fig.3. Basic structure of Graphene oxide[16]

III. STUDY OF SHAPE AND SIZE OF GRAPHENE FOR ITS USE IN CONCRETE INDUSTRY

Dispersion of graphene is a critical parameter for the formation of good bond between the graphene and the host material as it significantly influences the interaction of the constituent materials involved. Good dispersion of graphene is not easy as the individual particles of the Portland cement are much larger than the individual particles of the graphene. In addition, when the paste hardens, the pores formed will be much bigger than the individual particles of graphene which reduces the area of interaction of graphene with the products of hydration. Furthermore, these materials at nano scale may occupy the interparticle void. Therefore, to ensure that particles are well dispersed, a substitution of a portion of the Portland cement by supplementary cementitious material of equivalent individual size is necessary. The quantity of water added to the paste significantly influences the pore formation in the hardened cement paste. Furthermore, the specific surface of calcium hydroxide formed post hydration is very small which limits its bonding ability with the monolayer. Therefore, the larger porous structure (with respect to graphene particles) along with the hydrated products affects the bonding of the graphene with the host material. All this suggest that functionalization of graphene is important for its wide spread use as a material in the construction industry [16]. This process results in a finished product that has better compatibility with the products of hydration, and as a result, it improves the overall structure of the hardened paste along with the decrease in the size of the pores formed. Although, this process is an essential part for the proper results, care should be exercised that the basic configuration and structure is not altered.

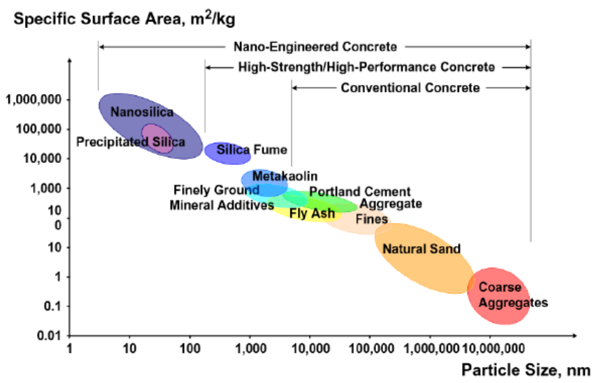

Fig.4. variation in specific surface area with the diameter of the particles [17]

IV. FUNCTIONALIZATION OF GRAPHENE

There exist various options to fabricate high quality graphene. Usually this material is derived using Graphene oxide for the various advantages it provides over other methods of extraction as mentioned previous sections. When the base material for extraction is Graphene oxide, the product obtained is already functionalized. However, this functionalization of graphene does not impart enough O_2 rich groups. As a result, it gets difficult to disperse the material in the aqueous medium. Therefore, there is a pressing demand to properly functionalise the graphene so that the graphene is properly soluble which ensures that this monolayer interacts properly the products of hydration and provides a very good bond. The techniques that are proven to be useful for functionalization of graphene for better dispersion and compatibility with the hydration products of cement are:

A. Noncovalent attachment

B. Covalent attachment

A. *Noncovalent attachment*

Graphite is a 3D, naturally occurring economical source material for the bulk manufacturing of graphene. The π–π stacking between individual layers results in a multilayer where not all adsorbed molecules are in contact with the surface layer of the adsorbent. Water provides cohesive and adhesive properties to the cement which binds all the constituent materials of concrete. Water being a polar liquid, and graphene by itself is water repelling, the dissolution becomes a major problem for its use. Therefore, the issue of solubility can be solved by functionalization of the monolayer by using organic compounds. The functionalization method utilized must not disturb the basic form of structure of graphene and one such method is by π-interactions [18].

B. *Covalent attachment*

A couple of research studies concluded that production of graphene without the presence of O2 functional group is not possible. These groups can be beneficial for the covalent functionalization. The functional groups such as –OOH, -O- etc., can interact with the functional groups of the host material thereby improving the dispersion. As a result, the basic form and structure of the graphene is not altered by the process of functionalization [19].

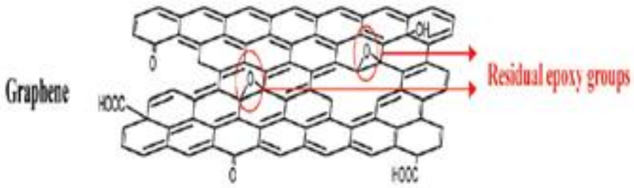

Fig. 5. Graphene with residual epoxy groups [20]

A study was conducted to evaluate the influence of graphene functionalization on the interfacial strength between grpahene composite and C-S-H gel. The bond strength is evaluated using the equation of energy:

$\delta E = E_{total} - (E_f + E_m)$

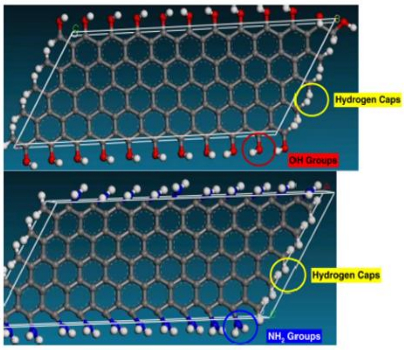

Fig.6. Structure of Graphene functionalized with (OH) and NH2 [20]

It was observed that the net difference was always found to be negative which directly implies that the material is energetic. The results of the test analysis is provided in Table III.

TABLE III

VARIATION IN INTERFACIAL STRENGTH WITH DIFFERENT FUNCTIONALIZATION [20]

Composite	**Interfacial strength (N/mm2)**
C-S-H-G()-C-S-H	1200
C-S-H-G(NH2)-C-S-H	6100
C-S-H-G(COOH)-C-S-H	11800
C-S-H-G(OH)-C-S-H	13500

V. **Production of Graphene based cement composite**

Production of GO with least impact on the environment for economies of scale is achieved by chemical reduction or by using ultra sonication. Synthesis of GO is carried out by using Modified Hummers Method [21]. The process is provided in fig. 7.

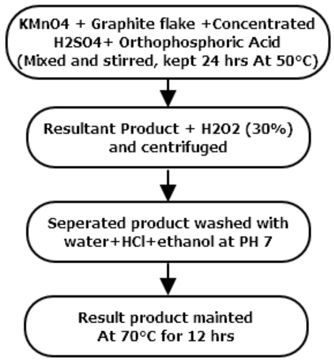

Fig. 7 Production of Graphene Oxide

To synthesise GO cement composite, GO is dispersed in water (quantity depends on the required water cement ratio) and sonicated to obtain a homogenous solution. Cement is then added and mixed thoroughly. Subsequently, aggregates are added to the mix. This process is briefly explained in fig.8.

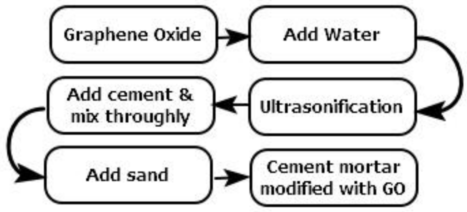

Fig. 8 Production of Graphene based cement composite

VI. RHEOLOGICAL PROPERTIES

Shang et al. studied the influence of graphene oxide, graphene oxide encapsulated silica fume on the rheology of the cement pastes. The rheological behaviour of the cement paste was found to be similar to that of other materials at a nono level. When graphene oxide was used, the parameters that affect the rheology of the cement paste amplified [22-24]. The workability of the cement paste decreased beyond 40% when the graphene oxide content was 0.05%. In addition to this, even when minute portion of graphene oxide is incorporated, there is a significant increase in the viscosity and yield stress. Similarly, Gong et al. observed a decrease in workability of 41.7 % by integration of 0.05 wt% of GO (22). According to Shang et al., shear stress and apparent viscosity of the mix rises with increase in concentration of GO [24]. The reason for this reduced workability is interparticle friction. Furthermore, the reduction in viscosity is directly proportional to the size of the graphene oxide. The intrinsic disadvantage of using a nano material is their large water demand and reduction in the workability of the mix.

Fig. 9. Flocculation of particles when graphene oxide is incorporated into cement [25]

The reduction in workability and improvement in parameters of the rheology of the mix was also observed by a few other researchers when graphene oxide was incorporated in the mix. Therefore, the graphene oxide was mounted on the silica fume to produce a new additive that is graphene oxide encapsulated silica fume. It was observed that the pastes that incorporated this modified form of graphene flows easily and it is easier to work with, with respect to the ordinary cement paste and the paste that incorporated silica fume at the same water content. The basic reason behind this was thought to be the combined effect of silica fume and graphene oxide. The improvement in the workability can be attributed to the surface effect of Graphene oxide and form effect of silica fume. The large surface area of the graphene oxide and its affinity towards water is not the only reason for the reduction in workability. The particles of the graphene oxide are negatively charged which upon interaction with the cement particles, causes agglomeration of the particles. This agglomeration of particles entraps a free water that was originally available to fluidify the mix. Fig 10 shows the flocculated structure of the particles of the cement when graphene oxide alone is incorporated. However, in the case of graphene oxide encapsulated silica fume, the fluidity is better than the ordinary graphene oxide incorporation. The reason for this behaviour can be attributed to the adsorption of water molecules to the surface which enhances the ball bearing action of the spherical particles of silica fume. Therefore, better workability and lower rheological parameters are observed in the case of graphene oxide encapsulated silica fume.

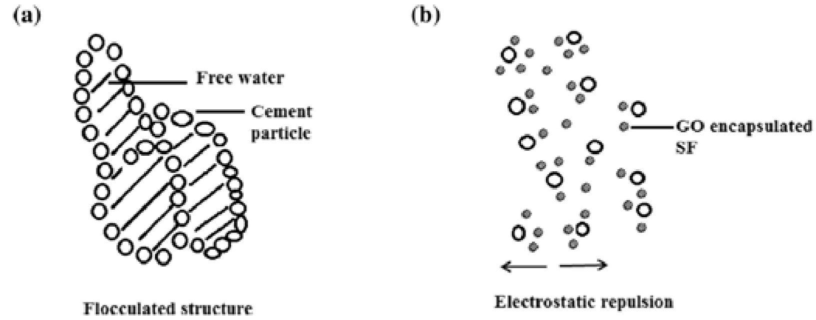

Fig.10.Mechanisms involved in (a) cement paste with graphene oxide (b) cement with graphene oxide encapsulated silica fume [25]

VII. EFFECTS OF GO ON MECHANICAL PROPERTIES AND MICROSTRUCTURE OF CEMENT COMPOSITES

A few research studies were conducted the know the influence of graphene on the mechanical properties of the cement-based materials. And all the research studies suggested that there exists considerable positive influence of incorporation of graphene in the cement-based building materials. Shengua et al. (2013) [26] conducted tests to determine the strength of cement-based composites under compression, tension and bending by varying the dosage of graphene oxide (Table-5). It was observed that the performance of the cement based composite improved drastically under the action of tension and bending (28 days strength) initially up to a dosage of 0.03% and then a mild decrease was observed. The increase in was observed to be 78.6% and 60.7% under tensile and flexure respectively. Furthermore, the compressive strength of cement-based composite which saw an increase of 47.9% at dosage level of 0.05% [26]. Microstructure of the cement-based composite is the primary influencer on the mechanical properties. Pan et al. incorporated GO sheets into the ordinary Portland cement and observed that the strength under compression and flexure saw an increase of 15-33 % and 41 -59% respectively (table 5). Compressive strength at 7 and 28 days are provided in table IV.

TABLE IV
VARIATION IN STRENGTH PROPERTIES [26]

Specimen	Compressive Strength		Observed increase in flexural strength	
	7 days	28 days	7 days	28 days
Ordinary Portland cement	31.3± 0.17	37.15	4.7± 0.25	-
GO-cement based composites	38.8± 0.13	54	7.0± 0.2	-

TABLE V
VARIATION IN STRENGTH OF CEMENT PASTE WITH CHANGE IN GRAPHENE OXIDE CONCENTRATION [27]

Percentage of GO	Percentage increase in tensile strength		Percentage increase in flexural strength		Percentage increase in compressive strength	
	3 day	**28 day**	**3 day**	**28 day**	**3 day**	**28 day**
Control specimen	0	0	0	0	0	0
0.01	28	47	51.9	51.7	12.2	13.4
0.02	27.8	59.5	54.2	32.9	31.5	27.6
0.03	51	78.6	70.7	60.7	45.1	38.9
0.04	24.7	36.6	28.4	30.5	53.6	42.2
0.05	24.2	35.8	28.1	30.2	59	47.9

The mechanical properties are primarily influenced by the variation in microstructure when a different material is incorporated into the existing system. Therefore, microstructural studies form an integral part of the study that involves mechanical strength evaluation. Scanning electron microscopy of graphene-based cement composites is necessary to identify the cause of variation in the properties of the materials. SEM images of Cement composite mixed with graphene oxide indicated clear change in the shape (fig.10). Areas around the fracture surface had deposits of needle and bar like crystals of ettringite, calcium hydroxide and monosulphate which were stacked disorderly. When the graphene oxide was increased from 0.01% to 0.03% formation of dense interwoven flower like crystals was observed. For a dosage of 0.02% of Graphene oxide, crystals of flowers were abundantly observed throughout the composite. However, for a dosage of

0.01% very few crystals unevenly distributed through the composite was observed. When the dosage was further increased to 0.04 to 0.05% the shape of the crystals post hydration was found to be polyhedron which were much denser compared to the crystals that were formed for lower dosages. The formation of such crystals at different dosages post hydration suggests that the flowery hydration products at lower dosages are responsible for improved toughness and polyhedron crystals significantly improve the composites performance under compression. It was observed that crystals formed at lower dosages has flowery crystals with interwoven needle like crystals and some voids that allows some movement implying that strength of composite under tension and bending improves significantly. The increase in compressive strength can be attributed to dense polyhedron shaped crystals which offers great resistance to compressive forces.

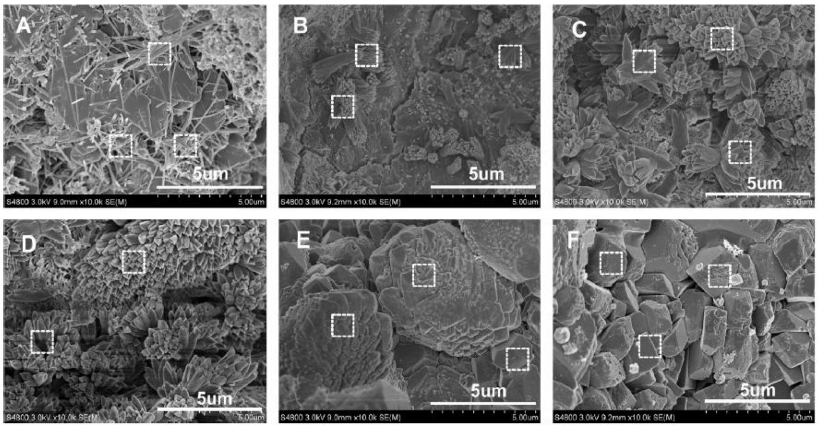

Fig. 11 SEM images of graphene oxide modified compsoite: (A) Without graphene oxide(GO); (B) With GO content of 0.01%; (C) With GO content of 0.02%; (D) With GO content of 0.03% (E) With GO content of 0.04%and (F) With GO content of 0.05%. [27]

Saafi et al. [27] incorporated reduced graphene oxide into geopolymers and observed that the reaction among the Graphene oxide sheet and the alkaline solution resulted in heavily reduced and cross-linked GO sheets. When the content of the reduced Graphene oxide was 0.35% by weight, the strengths under compression, bending and tension had significant improvements. At 0.35% concentration, the bending strength increased by 134% and the observed Youngs modulus increased by 375%. They also concluded graphene oxide based geopolymers are ecofriendly alternatives to the conventional cement because of the reduction in the greenhouse gas emissions. Several other researchers have conducted tests to ascertain the mechanical properties of concrete with addition of GO and the results are as provided in table. A positive variation indicates an improvement in the property and the converse is also applicable. Variation in porosity levels as seen in table further substantiates the SEM images of fig .11.

Property	**% wt. of GO**	**Variation (%)**	**Reference**
Compressive strength	0.01	29	28
	0.02	160.1	29
	0.05	11.05	30
	0.08	28.8	31
	0.5	20	32
	1	78	33
Tensile strength	0.02	197.2	29
	0.03	78.5	34
	0.08	37.7	31
	1.5	48	35
Flexural Strength	0.03	60.7	34
	0.02	184.5	29
	0.08	80.6	36
	0.04	14.2	37
	0.04	16.7	38
	0.05	59	36
	0.022	30.37	39
	0.03	60.7	34
Hardness	30	120	40
Shear Modulus	0.5	37.3	41
Porosity	0.02	-2.2	38
	0.03	-13.5	42
	1	-14.6	43
Flexural toughness	0.08	105	31

VIII. CONCLUSIONS

In the field of civil and construction engineering, the material graphene is a promising newcomer which has wide variety of applications. The inherent properties of this material have to be properly utilized which can impart great strength and durability to the structural member. However, the functionalization of the material is essential to be incorporated into the cement.

Several researchers have demonstrated improvement reinforcement and microstructure because of the active functional groups as discussed in section and very high specific surface area. Consequently, mechanical properties have shown significant improvement for optimum dosages.

As Graphene is still comparatively new to civil construction industry, studies related to its durability characteristics is still a big research void. For its acceptance as a mainstream construction material, comprehensive tests for determining the durability characteristics is highly necessary.

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