

UTILIZATION OF GRAPHENE OXIDE IN BUILDING MATERIALS

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Abstract: Cement-based materials (CBM) is one of the most commonly used building materials. Due to their brittle nature (sudden failure, low tensile strength, low resistant to crack), CBMs were modified through the addition of various traditional fillers/ reinforcement such as fibres and supplementary cementitious materials. Advanced modifiers/fillers such as Graphene-based nanomaterials (GBM) were utilized due to their superior properties in upgrading properties of CBM. GBM have been firstly used to enhance various polymeric matrices for storage energy, sensors and bio-applications. Following that, GBM were utilized in CBM with remarkable improvements in properties. In the current paper, an overview on the synthesis and structure of one type of GBM; namely graphene oxide (GO) was presented. The impact of GO on properties of CBM was addressed. One of the most observations from the highlighted published work was that the enhancements in CBM properties were linked to the uniform dispersion of functionalized GO and template effect of GO in CBM. Although GO with its attractive properties in upgrading properties of CBM, the usage of GO in construction industry is still not at the same demand level as in other applications and further investigations are needed in this regard.

Keywords: *Graphene oxide, Cement-based materials, Building materials, Graphene-based nanomaterials.*

1. Introduction

Cement-based materials (CBM) are the most commonly used building materials to construct various types of structures under different

service conditions. However, some drawbacks are associated with these materials due to the brittle nature of CBM such as weak crack resistance, low tensile strength, sudden failure, high water absorption and low ductility of CBM which led to reduction in the desired properties of CBN. Thus, develop CBM was the target of numerous researchers work that conducted using different techniques and traditional fillers (modifiers) or reinforcement to the CBM such as fibres [1-5] and supplementary cementitious materials [6-9]. The inclusion of nanomaterials into cementitious matrices was one of the methods that used to develop CBM to enhance mechanical and thermal properties of the traditional construction materials that based on cement [10-15]. The k-advantages of utilizing nanomaterials is the size of these nanomaterials when added with very small quantities. The materials at nanoscale have unique properties which different from those from other larger scales which affect the behavior of materials at nanoscale [16].

Polymeric matrices were firstly modified with the addition of different types of nanomaterials. Enhancements in mechanical, thermal and

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electrical properties were achieved by adding small concentrations of nanomaterials into epoxies [17-21], thermoplastics and elastomers [22-24]. Nano-based materials were also incorporated in cementitious materials to upgrade the properties of CBM [25-27]. Graphene-based nanomaterials (GBN), an example of carbon-based nanomaterials, were first employed in polymeric matrices [28-30] to be used in various applications such as energy storage [31], bio-based applications [32] and sensors [33]. Depending on their synthesis method [34], various types of GBN are available such as graphene (GR), graphene oxide (GO) and reduced graphene oxide (rGO). The discovery of GR was later than GO. The GR was discovered on 2004 by two researchers (Geim and Novoselov) at the University of Manchester by isolating GR flakes from graphite. They used a sticky tap to remove some GR flakes from a lump of bulk graphite. It was observed that these flakes were with various thickness. They succeeded in creating flakes with one atom thick by continues separation graphite fragments [35]. Prior to 2004, many efforts were conducted by other researchers to synthesis and characterizing GR using different techniques and routs (Fig.1) [36].

Since 2004, GR became an attractive candidate in reinforcing various types of matrices. GR is a two-dimensional nanomaterials (2D sheets) with sp² of carbon atoms in chicken wire like structure) [37]. Due to this structure, superior properties such as high tensile strength (130 GPa) and high Young modulus (1TPa) [38] are obtained. However, GR is a hydrophobic material which made it not easily dispersed in water and other aqueous matrices [39] in addition to the tenancy to agglomeration in solution and some difficulties that associated the synthesis process of GR. Graphene derivatives were then discovered to be used instead of GR.

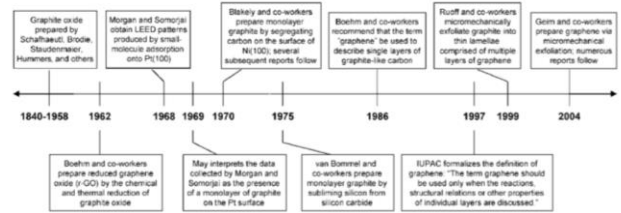


Figure 1. Background about synthesis GBN [36]

Graphene oxide (GO) and reduced graphene oxide (rGO) are alternatives of GR. GO was first used to reinforce various polymeric matrices for various applications such as drug delivery [40] and sensors [41]. It was reported that the inclusion of few percentages of GO either as sole material or hybrid graphene materials into polymeric matrices for industrial applications showed enhancements in mechanical, thermal and electrical properties of the polymer matrices [42-44]. In terms of construction sector, excessive work by researchers was to study the properties of GO-modified cementitious materials and remarkable enhancements in various properties of CBM were recorded [45-47]. Regardless of the attractive properties of GO in modifying CBM, the application of GO in construction industry on large scale is still not in the same demand level like in other applications. In the current paper, the most important features and attractive properties/ structure of GO were addressed. The effect GO on properties of CBM that most commonly used in construction was presented which was based on the conducted work by other researchers and how the dispersion technique of GO affect the required improvements in the properties. The mechanism of the template effect of GO on hydration products is also highlighted.

2. Structure and Synthesis of Graphene Oxide

Graphite is a form of crystalline carbon that occur naturally and stable in this form under

standard conditions. Its structure comprises of hexagonal carbon crystal sheets stacked on top of each other. Since graphite is naturally available, its structure contains numerous localized defects which affect negatively on the chemical reactions [48]. Graphite was used for many different purposes and it led the discovery of graphene (GR). The latter is a single-layered graphite (Fig.2 left) [49] of two-dimensional nanomaterials (2D sheets) of sp^2 carbon atoms in honeycomb pattern with high tensile strength (130 GPa), modulus of elasticity of 1TPa [38] and large specific surface areas ($\sim 2600 \text{ m}^2/\text{g}$) [50]. However, the large scale production of synthesis GR is not easily accomplished. Also, GR has a hydrophobic nature which make it not dispersed easily in certain matrices in addition to the strong interlayer van der Waal forces [51]. Thus, the discovery of derivatives of GR such as graphene oxide (GO) (Fig.2 middle) and reduced graphene oxide (rGO) (Fig.2 right) became an essential demand to overcome the obstacles that associated with GR.

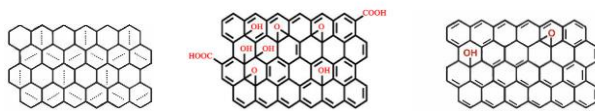


Figure 2. Structure of GR and its derivatives: GR (left), GO (middle) and rGO (right) [49]

Two approaches were utilized to synthesis GR and these are top-down method and bottom-up method (Fig.3) [52]. In the top-down approach, graphite is the main source and different methods are employed to produce GR like material; namely GO.

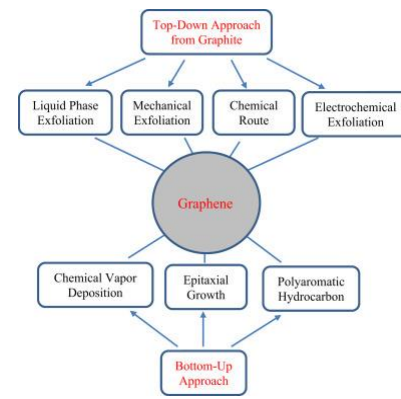


Figure 3. Methods used for the production of GR [52]

GO is one of the derivatives of GR with a similar hexagonal carbon structure to GR, however, groups of hydroxyl ($-\text{OH}$), alkoxy ($\text{C}-\text{O}-\text{C}$), carbonyl ($\text{C}=\text{O}$), carboxylic acid ($-\text{COOH}$) are available in the structure of GO with other oxygen-based functional groups (Fig.4 middle). The oxidation of graphite leads to the formation of graphite oxide. The latter when exfoliated, then GO is obtained. The number of layers is the main difference between graphite oxide (multilayer system) and GO (few layers flakes and monolayer flakes system) (Fig. 4) [53].

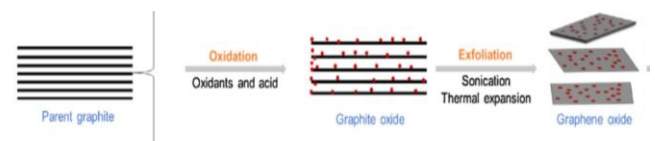


Figure 4. Synthesis of GO from graphite [53]

Different methods are available to synthesize GO from graphite and these are Staudenmaier, Hofmann, Brodie and Hummers (Fig.5) [54]. The ratio of carbon/oxygen (C/O ratio) is the indication on the effectiveness of the oxidation process. The GO sheets would not be stacked with another sheet like in the case of graphite oxide (Fig.4). Different methods are available to obtain chemically modified GO and these methods are varies depending on the applications at which GO is to be utilized.

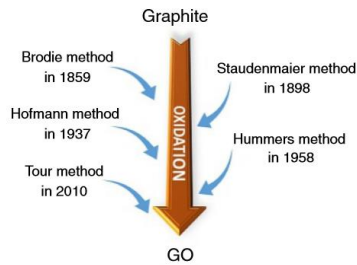


Figure 5. Different synthesis methods of GO from graphite [54]

When GO is chemically and/or thermally reduced graphene, the reduced GO (rGO) is formed (Fig.3 right). The structure of rGO, even after many severe reduction, it cannot reach the pristine GR structure as rGO still contains residual oxygen and structural defects originated in the chemical oxidation synthesis of GO. Many chemical reduction techniques are used to obtain rGO as shown in Fig. 6 [54].

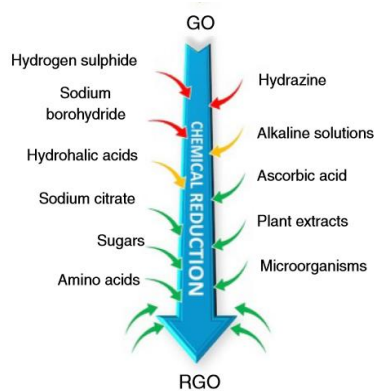


Figure 6. Techniques used to obtain rGO from GO [54]

3. Dispersion Technique of GO

Since GO or rGO is considered one of the nanomaterials, the effectiveness of such materials required well-dispersion into cementitious matrices to obtain the remarkable enhancements in the mechanical properties of nanocementitious. The tendency of GO sheets to agglomerate [55], due to the van der Waals between these sheets and the huge surface area of GO, was the main concern of many researchers to conduct studies to find the suitable dispersion method of GO. Some of

these methods are varied between mechanical and chemical techniques.

Most of the available techniques are based on exfoliating GO nanosheets into water that used in the production of cement-based materials (CBM). Ultrasonication, high shear mixing and magnetic stirring [56] are examples of mechanical methods that adopted to disperse nanomaterials in mixing water (Fig.7) [57]. These methods were couples with the usage of mineral/chemical admixtures to facilitate the dispersion process. In terms of chemicals, surfactants or surface modification aids are utilized which assets the dispersion of GR-based nanoscale materials through different paths (wetting, electrostatic repulsion and/or steric hindrance actions) [58]. Regarding mineral admixtures, such additives help in the dispersion of GR-based nanomaterials due to the influence of gradation, adsorption and separation effects [59].

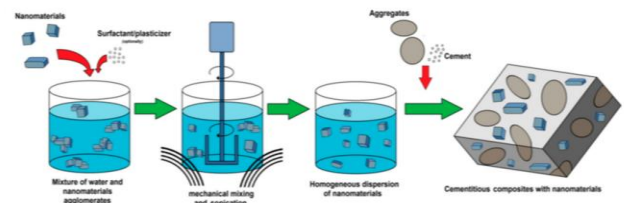


Figure 7. Common dispersion technique of nanosized materials into liquid media [57]

Different periods of mixing GO with water that used for mixing the ingredients of CBM were adopted. Sonication for 3 hrs was adopted [60] to disperse (0-2.5 wt%) of GO into water and then water reducing admixture (Polycarboxylate superplasticizer) was added. Cement and sand were added later to produce GO-modified cement mortars with water-to-cement (w/c) ratio of 0.3. Less sonication time (5 mins) was used when small concentrations (0.02-0.4 wt%) of GO were dispersed to produce cement paste of various water-to-cement ratios (0.4-1). Silica fume was added later with cement and high-

speed mixer was adopted as mixing method [61]. Sometimes, it is required to use more than one technique for dispersion of GO into cement matrix. A combination of two dispersion techniques (ball milling and sonication) [62] were used to disperse different concentrations (0.125-1 wt%) of GO to fabricate samples of cement mortar with w/c ratio of 0.45 using polycarboxylate superplasticizer as well. The ball milling was conducted for 16 hrs followed by sonication for 1 hr.

Regarding the production of CBM with hybrid GBM (GO and graphene nanoplatelets (GNP)), sonication for 30 mins was used to disperse combination of (GNP and 0.1 wt% of GO) [63] as well as (GNP and 1 wt% of GO) [64] to produce cement mortar with w/c ratios of 0.53 and 0.54 respectively with the usage of admixture (water reducer) in the mix. In terms of mix with w/c ratio of 0.53, after dispersion of GO and GPN in water, the ingredients of mortar were mixed for 20 mins using mortar mixer.

4. Role of GO in Cement-Based Materials

The structure of GO contains functional groups on the surface of GO. The role of the functional groups is to make the dispersion of GO into water or other aqueous solution much easier [65]. The k-advantage from inclusion GO in cement-based materials (CBM) is the template function of GO (Fig.8) [66] on the microstructure of cement due to the availability of the functional groups on the surface of GO that facilitate the dispersion of it in aqueous solution. The hydration process of cement is therefore accelerated by GO which assist as well in arranging the products of hydration and the formation of flower-like cement. Denser CBM was obtained since GO works on reducing the pores with the products. Additionally, the functional groups on the surface of GO facilitates the chemical bonds (strong covalent

bond) between GO and products of hydration of cement [67].

It was reported that GO has strong nucleation, connecting the GO monomers to each other through chemical reactions, which causes the formation of 3D network structure in cementitious matrix. Due to the adsorption between GO and hydration products, tight connection between products of hydration and the GO-3D network structure is obtained [68].

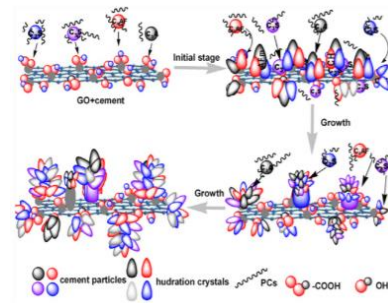


Figure 8. Template function of GO [66]

5. Some Properties of GO-Modified Cement Based Materials

The effect of adding graphene oxide (GO) on properties of cement-based materials (CBM) was addressed by scholars using different types of graphene-based materials (GBM) and various dispersion/mixing techniques. An investigation was conducted to investigate the influence of replacing cement (Portland cement type II 42.5R) with different concentrations (0,0.01, 0.03 and 0.05%) and different types of GO using different water-to-binder (w/b) ratio (0.2, 0.3 and 0.4) [69]. The GO was used with different dimensions (diameter and thickness). The diameter of the three types of GO was 0.5–3 μm , 8–15 μm and >50 μm with thickness of 0.55–1.2nm, and 1.75 nm and 1.8 nm respectively. The procedure of mixing was performed by a uniform mixing of the liquid state materials (GO dispersion solution, water, and superplasticizer) for 30 seconds at low

speed. Sand was then added to the mixtures and stirred for another 30 seconds. The mixture was stirred for 30 seconds at high speed for 30s followed by another mixing at high speed for 60 seconds. The mix of GO-cement mortar was infected into the mold of various tests to obtain the mechanical properties of cement mortar samples.

It was found that compressive and flexural strengths of mortar were increased with the increment in GO and up to 0.03%. The highest increment in compression was 21.37% and it was 39.62% for flexural strength when GO was 0.03% and w/b ratio is 0.2 when compared with control samples. In terms of tension strength, the increment was 53.77% for GO -0.03%. The reduction in the mechanical properties with the increase in the percentages of GO to 0.05% was attributed to difficulties in obtaining uniform dispersion of GO into the cement pastes as GO sheets will tends to agglomerate.

Regarding the diameter of GO, higher compressive and flexural strengths for 28 days of curing were obtained for samples with high diameter of GO. The highest increment in compressive (20.58%) and flexural (11.59%) strength was reported for GO samples of diameter greater than 50 μ m. Such improvements in the strengths was attributed to the effect of large diameter of GO in blocking and bridging of microcarcks. Regarding tensile strength, an increment of 53.77% was reported for GO samples with 0.03% and for diameter greater than 50 μ m for GO when compared with control specimens. In terms of the studying the microstructure of the GO-cement mortar and based on TAG results, it was founded that the addition of GO has no influence on the hydration of cement. Porosity was found to be reduced with the addition of GO and the highest reduction (0.78%) in the porosity was reported for 0.03% samples. Regarding SEM images,

same surface fracture was found for samples with and without GO.

When non-functionalized GO was used in another investigation [70], reduction in the compressive strength with no effect on density of cement mortar was noted when GO (0, 0.1, 0.3, 0.5, 1, 1.5 and 2%) was used. However, the addition of GO was found to increase the tensile strength of mortar when compared with control mix. As the percentages of GO was increased, the enhancement in the strength was achieved except mixes with 2wt%. Highest increment in the strength (48%) was recorded for mortar samples with 1.5% of GO. This was attributed to the better dispersion of GO nanoflaks into the cement matrix. This was confirmed by the SEM images as no agglomeration of the flaks was observed and good dispersion of GO was achieved with the usage of ultrasonic method. The data from XRD indicated that calcium silicate hydrates (C-S-H) gels was growing in GO mortar as compared with control mortar. This was attributed to the nucleation of C-S-H by the GO flakes due to the higher surface energy and the presence of hydrophilic groups on the GO surfaces acted as a nucleation site.

The effect of GO on dry shrinkage of C40 concrete was studied by other scholars [71]. Small concentrations of GO powder (0, 0.02, 0.05 and 0.08 wt. %) were utilized with w/c ratio was 0.49. The GO was first dissolved in water and then superplasticizer was added. The solution was then mixed with concrete ingredients. Samples was 100 \times 100 \times 515 mm. Dial indicator was utilized to get the data of drying shrinkage at different ages (1, 3, 7, 14, 28, 45 and 60 days). An increment in the shrinkage strain of concrete was reported for GO samples. The increment was found to be increased with the increase of GO as in case of samples at 60 days. The reported increment was

1.99%, 5.79% and 7.45% for samples with 0.02 wt%, 0.05 wt% and 0.08 wt% GO respectively.

The influence of inclusion GO on properties of concrete that containing rice husk ash was investigated by effect of another group of researchers [72] explored the. In their study, a total of 11 concrete mixes (M1-M11) were prepared. The ingredients of concrete were mixed using (1:2.46:1.36) mixing ratio for (cement:sand:gravel) with water-to-binder (w/b) ratio of 0.42. Ordinary Portland cement (M43) grade was used. Different percentages (0, 5, 10, 15, 20, 25, 30, 35, 40, 45 and 50%) of Rice Husk ASH (RHA) were used as partial replacement by weight of cement. Silica content in RHA was 90.99%. GO solution was prepared in accordance to modified Hummers method using flake graphite which was provided by Tamil Nadu Minerals Limited. As provided by the supplier, the molecule weight of GO was 12.01 g/mole. The addition of GO solution to the mixes was at different ratios (0, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9 and 1wt%). The symbols of M1 was referred to control mix (without GO and RHA). Other symbols from M2 to M11 were referred to concrete mixes with different percentages of GO and RHA. The M2 was concrete mix with lowest percentage of both GO and RHA (0.1% GO+5% RHA) while M11 was for mix with the highest percentages of GO and RHA (1% GO+50% RHA).

All mixes (M2-M11) which containing GO and RHA showed higher workability than control mix (M1). Highest slump value (155 mm) was reported for M11 with the highest percentages of GO and RHA (1% GO+ 50%RHA). Bulk density was reduced with the addition of GO and RHA when compared with control mix. It was suggested to the possibility of using mixes (M2-M7) as light weight concrete. Regarding air content, results showed similar content for all mixes. In terms of heat of hydration, the

obtained images from SEM technique for specific mixes (M1-M6) at 28 days showed that lower traces of CH crystals was observed for M5 (0.4% GO+20% RHA).

Compressive strength of concrete was found to be increased with the addition of GO and RHA when compared with control samples (M1) for all ages of curing except M11 (1% GO+50% RHA) in which reduction in the strength was reported. Among the mixes that containing GO-RHA (M2-M10), all mixes showed increment in the compressive strength but the highest strengths (31.65 MPa, 32.68 MPa and 43.95 MPa) were observed for M6 (0.5% GO+25% RHA) and for age of curing at (7, 14 and 28 days) respectively. Consequently, the modulus of elasticity for M6 was also the highest when compared with other mixes (M2-M10).

Results of other mechanical properties (indirect tension and flexural strength tests) showed a similar trend of the combined effect of GO and RHA. All mixes (M2-M10) were observed to increase splitting tensile and flexural strengths and for all ages of curing when compared with control specimens (M1). Only M11 was found to reduce the strengths. It was reported that, splitting tensile strength (5.59 MPa and 6.64 MPa) of M7 was the highest for 14 and 28 days respectively while M6 showed the highest strength (4.58 MPa) for 7 days. On the other hand, the highest flexural strength (7.15 MPa, 7.68 MPa and 9.67 MPa) was recorded for M7 and for (7, 14 and 28 days) respectively. Results that obtained from testing reinforced beams showed the lower deflection was reported for beams for M6 and M7. A smaller number of cracks and minimum width of crack were noted for samples of M7.

6. Conclusions

The development of building materials especially cement-based materials (CBM)

received great attention by researchers to overcome the brittle nature of CBM by adding fillers/reinforcement. Nanomaterials are advanced fillers to be used in upgrade the properties of CBM. Graphene oxide (GO) is one of nanomaterials in which it was firstly incorporated in polymeric matrices and showed remarkable increments in properties of the host matrix which enable the application of GO-modified polymers into various applications such as sensors, energy storage and bio-applications. Due to such achievements in other industries and the attractive properties of GBM, researchers in the field of construction and building materials adopted the inclusion of different forms of GBM such as GO into water that used in the production of CBM. The key-challenge to obtain GO-modified CBM is the uniform dispersion of GO within the CBM in order to achieve the potential of adding GO to improve the properties of CBM. The utilization of GO into building materials is need to be extend to large scale to be used in the actual construction site after finding the suitable dispersion method of GO in the site. Thus, further investigations are needed in this regard.

Conflict of Interest

The author confirms that the publication of this article causes no conflict of interest.

7. References

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