



Content list available at [ICONSMAT](https://www.iconsmat.com.au)

Journal of Construction Materials

Journal homepage: www.iconsmat.com.au/publication

Article history:
Received 14 June 2019
Received in revised form
20 July 2019
Accepted 12 October 2019
Available online
22 October 2019

A review on graphene reinforced cement composite: technical approach for ecofriendly construction

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Abstract

Graphene as an emerging material comprising outstanding properties had shown a great potential for commercial application in many industries including construction. The current paper seeks to review the advancements in the area of building and construction by focusing on the composition of graphene reinforced cement paste. The potential areas enhancing the performance of conventional cement mortar by graphene had been identified and investigated with a practical perspective based on the existing advancements. Mechanical properties, electrical conductivity and storage capacity of graphene for the purpose of producing an exposed concrete sample with the energy generation capacity had been briefly also studied. The results show that if a proper commercial synthesis method emerges for reducing the unit price of graphene, a graphene reinforced cement paste is a suitable material for ecofriendly high performance construction purposes. Following this result, the Australian market capital undergone a financial research where the graphene was subject as the commodity. A comparison had also been made between the environmental impact of graphene and cement.

Keywords

Graphene; Cement; Sustainability

Introduction

Graphene has recently attracted significant attention from the scientific community because of its extraordinary mechanical and electrical properties. In this paper, properties of graphene are being first explained followed by the barriers encountered with the commercial usage of graphene in construction practices. The state of ecofriendly construction approach utilizing graphene had also been mentioned in the following by promoting the production method of carbon nano particles from CO₂ gas. Another fact that wears the graphene reinforced concrete an environmental dress is the results from the compressive tests which shows that the use of cement can be limited by a drastic rate, keeping the compressive responses constant.

Recent mechanical experiments have shown that graphene is the strongest material measured so far. This opens up opportunities for graphene as a nanomechanical material to be used in variety of industries including construction. The mechanical properties of graphene under tension have been investigated extensively using both experiments and atomistic simulation methods [1]. Carbon nanotubes and nanofibers were explored as promising promoters for reinforcements in cement-based materials at nanoscale [2, 3]. Graphene is consist of a one-atom-thick planer sheet comprising a sp² –bonded carbon structure with exceptionally high crystal and electronic quality [4-7]. Graphene intrinsically has a high thermal conductivity of 3000 W/mK [8] which can be seen as a solution for the high heat generation upon the development of the hydration reaction. The unique mechanical properties of graphene attract attentions for the potential applications in construction. Theoretical and experimental results on individual graphene nanosheets exhibit extremely high values of Young’s modulus of about 1000 GPa and fracture strength of about 125 GPa[9].

Agglomeration of graphene particles caused by the process involved in the mass synthesis and the van der Waals forces between the nano platelets brings issues for a wider mechanical application. It compromises a hydrophobic characteristic which brings difficulties for its concrete application in terms of homogenous dispersibility by considering its low solubility in water[10]. On the other hand, the same hydrophobic characteristic of graphene may result in a more workable concrete as it would have almost the same functionality as superplasticizers by accelerating the development of the hydration reaction. This roots from a more exposure of cement particles to water. However, the graphene oxide (GO) had shown a good dispersibility in aqueous solvents. It has also to be mentioned that GO nano sheets are performing electrical insulation. GO sheets bear various oxygen-containing groups, mainly epoxides and hydroxyls on their basal planes and carboxyls on the edges, which facilitates the dispersion of GO in water[3, 11]. Dan Li et. Al [12] show that the surface charge (zeta-potential) of GO sheets are highly negative when dispersed in water. Since most of the extraordinary characteristics of graphene is associated with the individual sheets, the homogeneous dispersion of graphene nanoparticles becomes significant[12]. Surface modification using effective dispersion and exfoliation methods are some of the solutions for these types of barriers. Another method to prevent agglomeration is to attach other molecules or polymers onto the sheets which also increases the impurity of the product to be seen as an adverse effect. As reported by Tang et al [13], due to the lack of appropriate industrial dispersion method, graphene once it’s used in a composite forms an instable cluster which significantly lowers the efficiency as reinforcement. Kuilla et al[14], suggests the advancement of graphene

dispersion in polymer composites in another literature. A well-structured graphene reinforced composite must perform a modified interfacial area by producing a co-continues network[15].

Despite the fact that many studies in the field of nanoscience show the high dispersibility of GO sheets in aqueous solutions, S. Ghazizadeh et al. [16] argues that GO nanosheets aggregate in the Portland cement paste as it reacts with divalent and trivalent cations as well as hydroxide ions present in the paste solution causing the loss of oxygen groups. The reduction of GO in alkaline solutions had also been reported in other studies (apart from the construction related articles). This switches the repulsion between GO sheets to attraction and thus, the agglomeration of particles. In another research done by A. Hassani et al. [17], it's been claimed that based on the FE-SEM test results there were no aggregation of GO flakes had been found indicating a proper dispersion of nano particles in the cement paste. The controversy, thus, still remains for further investigations.

Many research studies reported an average enhancement in the compressive strength of graphene reinforced cement paste. The major effect of nanoparticles on the cement paste is the acceleration of the hydration reaction whereas 1D and 2D nanosheets are not only accelerate hydration but also reinforce the cement matrix due to their high aspect ratio[18-20]. It's been reported that the introduction of 0.05%wt GO can increase the compressive strength of cement paste by 15-50% and the associated flexural strength by 41-60%. The stress-strain curve derived from the experimental test on GO reinforced cement paste illustrate a longer portion of the plastic zone indicating the higher flexibility and a lower brittleness compared to the samples with no GO content [21, 22]. The main reason suggested and agreed across the scholars roots from the nucleation of calcium silicate hydrates as XRD diffraction data shows growth of C-S-H gel in GO cement mortar.

Commercial production of graphene

Graphite, consisting of stack of flat graphene sheets, inexpensive and available in abundance from both natural and synthetic sources, is the main resource for the mass production of graphene[12, 23-27]. The suitability of the application of graphene in various industries arises the necessity of a commercially scaled production method. Currently there are three major methods for a mass production of graphene compared in table 1.

Table 1 Graphene synthesis methods

Method	Cost of production	Quality of the product
Electrochemical exfoliation	Low – fast production	Low – damage to the honeycomb lattice caused by the oxidation step
Chemical vapour deposition	High – high temperature and expensive substrate	High – Large area graphene platelets gain

Electrochemical exfoliation

This method involves the oxidation of graphite sheets in an electrolyte followed by ultrasonic dispersion. Studies show that amongst the range of electrolytes including HBr, HCl, HNO₃, and H₂SO₄ only the electrolyte containing H₂SO₄ exhibit ideal exfoliation efficiency. Once H₂SO₄ solution (4.8 g of 98% H₂SO₄ diluted in 100ml of deionized water) was used as an electrolyte, the static voltage of +1 V was first applied to the graphite for 5-10 min, followed by a ramped up +10 V voltage for 1 minute. The low voltage application at the beginning helps the graphite become wet and prepared for a proper exfoliation. The electrochemical exfoliation of graphite sheets usually yields thick layers of graphene with small surface area. However, the modification of the acidity of the electrolyte and the voltage applied can enhance the quality of the final product[28-32].

Chemical vapour deposition

Among all the strategies to produce graphene, chemical vapour deposition (CVD) secured the place as a high efficient method producing single-layer large-area graphene sheets. The process involves the introduction of gas species in the reactor while heat applies. This is where the hydrocarbon precursors decompose to carbon radicals on the metal substrate surface forming the graphene nanosheets. The metal substrate acts as a catalyst and also provides the base for the deposition mechanism. Ni and Cu are the two major widely used metal substrates in the graphene CVD process[33-35].

Decomposition of carbon dioxide into oxygen and carbon nano particles

The reduction of greenhouse gas emission had become one of the main concerns of governments and scientists for the past decades. Many absorption and decomposition methods had been suggested since in order to trap and store the CO₂ gas for future consumptions[36-41]. A new area of study is emerging suggesting the conversion of carbon dioxide into carbon nano particles. For instance, one of the methods is to react the CO₂ gas in a saline solution of CaCl₂-NaCl-CaO to obtain carbonate ions and subsequently splitting the carbonate formation into graphene on a stainless steel cathode by conducting electrochemical decomposition[42]. Other studies show the possibility of producing high quality carbon

nanotube from carbon monoxide[43-45]. Carbon monoxide itself can be synthesised easily by thermal decomposition of CO₂ into CO and O₂.

Application areas of graphene reinforced cement mortar

Photovoltaic concrete panel

To have a proper understanding of the electrical conductivity and storage capacity of graphene, it is better to first identify the general relevant terminologies in the area:

Current-voltage (I-V) characteristic: As the name suggests, the I-V characteristics curves show the relationship between the current flowing through an electronic device and the applied voltage across its terminals. The following figure shows the I-V characteristics of an ideal resistor.

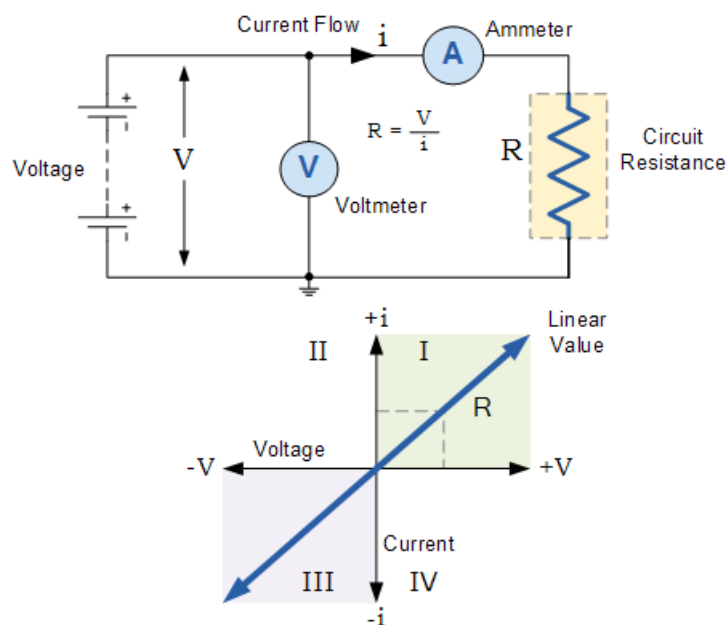


Figure 1 schematic current voltage characteristics of ideal resistors

Short circuit current: Is the associated current where the zero voltage applied.

Open-circuit voltage: Is where the I-V curve intersects with the horizontal axis representing voltage value at zero current.

Fill Factor: Is defined as the area under the I-V curve where the maximum power occurred. The power, essentially is derived from the equation $P = I.V$.

For a typical photovoltaic solar panel as terms defined above the I-V and P-V (Power-voltage) curves and be depicted in the following figure.

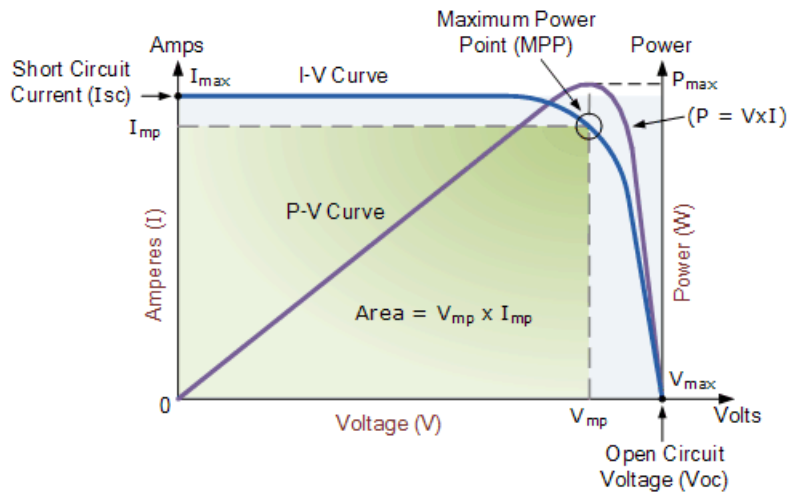


Figure 2 I-V and P-V graphs of photovoltaic cells

The excellent electron-transport properties and extremely high carrier mobility of graphene makes a suitable material for energy storage solutions and solar cells. Based on this, it has a great potential to be used for a low-cost, flexible, and highly efficient photovoltaic devices. The current–voltage (I – V) characteristics of the device showed a short-circuit photocurrent density (I_{sc}) of 1.01 mA cm^{-2} with an open-circuit voltage (V_{oc}) of 0.7 V , a calculated fill factor (FF) of 0.36 , and an overall power-conversion efficiency of 0.26% . The relatively low I_{sc} and efficiency of this cell is related to the series resistance of the device, the relatively low transmittance of the electrode, and the electronic interfacial change. For instance, solar cells, based on a reduced graphene oxide film electrode with a sheet resistance of $40 \text{ k}\Omega \text{ sq}^{-1}$ and a transparency of 64% had a conversion efficiency of 0.1% [46]; while a reduced graphene oxide film of $5 \text{ k}\Omega \text{ sq}^{-1}$ and $\approx 80\%$ transparency led to a device conversion efficiency of 0.4% [47]. Many efforts have thereby been made recently to improve both the conductivity and transparency of such graphene thin films to be used as electrodes in solar cells [48].

From the results of the experimental test on the electrical conductivity and the storage capacity of graphene nanosheets, the opportunity of developing concrete samples being capable of generating electricity arises. The current technology for solar energy generation compromises the use of silicon based material with NP-type poles. Studies show the enhancement of the efficiency of the solar panels made with graphene nanosheets[49-54]. It is reported that the photovoltaic graphene cells obtained the highest power-conversion efficiency recorded as 1.1% [51]. The structure of a typical silicon base solar cell is demonstrated in the following figure:

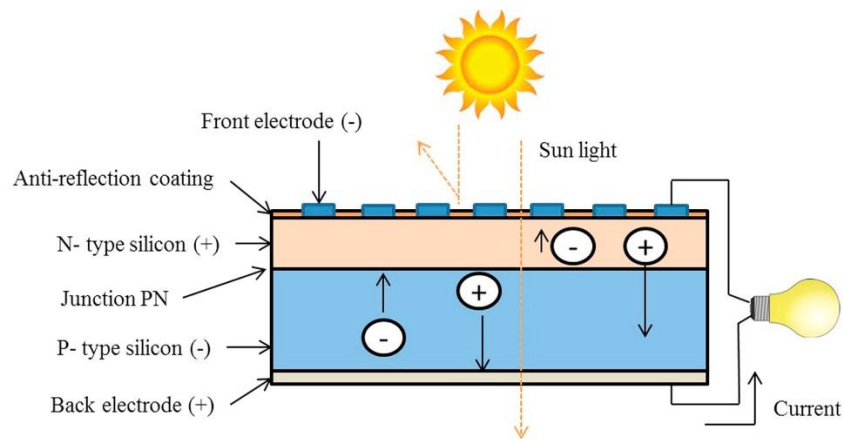


Figure 3 Schematic illustration of silicon based solar cells' functionality

A conceptual photovoltaic graphene reinforced concrete panel is anticipated to be capable of generating power, although in a much lower efficiency rate than a fully optimized solar cell, once it's been used in a high portion of concrete walls and slabs it can achieve a high energy yield.

Mechanical strength

Properly designed high quality graphene fibres can be seen as a tensile component of concrete panels to replace the conventional steel reinforcements. The fact that the graphene nano fibres represents a high tensile strength more than steel reinforcement and the lightweightness of such material brings the opportunity to develop lighter and stronger concrete panels. The reported 130 GPa tensile strength of graphene compared to the 1.5 GPa tensile strength of conventional steel reinforcement introduces the opportunity of reducing the cross sectional area of tensile reinforcement in the concrete composites by a factor of about 100[18]. This means that a 20 mm typical diameter of steel reinforcements can be replaced by a 0.2 mm thick graphene fibre exhibiting the same load carrying capacity.

Generally, the use of nanomaterials in cement composites results in a higher degree of hydration reaction. Carbon nano particles in particular had been investigated to represent an enhancement in the early age strength gain of cement composites[55, 56]. According to the literature review, an average 30% compressive strength improvement had been recorded once carbon nanotubes were added to the cement composites.

Table 3 Mechanical improvement of cement composite reinforced by carbon nano particles

Methodology	CNT weight fraction (%)	Results (% improvement)	Code
Ultrasonication	1	Compressive: SWCNT : 6% MWCNT: 30%	N.A.
Sonication and carboxylation with sulfuric and nitric acid	0.5	Compressive: 19% Flexural: 25%	N.A.
Sonication with Polyacrylic acid	0.045	Compressive: 50% Flexural: 10%	N.A.
Sonication with acetone and modified acrylic polymer and superplasticisers	0.5	Compressive: 11% Flexural: 34%	UNI-EN 196-1
Sonication and NaDC surfactant	0.2	Compressive: 29.50% Flexural: 35.45%	GB/T1 7671
Sonication and surfactant	0.08	Elastic Modulus: 35% Flexural: 40%	N.A.
Sonication and surfactant	0.08	Elastic Modulus: 45% Flexural: 25%	ASTM C348
Sonication and polyvinylpyrrolidone organic solvent	0.25	Load capacity: 47% Toughness: 25%	N.A.
Naphthalene-sulfonate plasticizer and modified polycarboxylate admixtures	0.3	Modulus: 14% Compressive: 12% Splitting tensile: 34%	NBR 7215, 8522 7222
Sonication, surfactant and swing centrifugation	0.08	Elastic Modulus: 35% Flexural: 35%	ASTM C348
Sonication and water reducing admixture ADVA cast 575	0.2	Flexural : 269% Ductility: 81%	N.A.
Sonication and polycarboxylate admixture	0.5	Compressive: 25%	N.A.
Sonication and surfactant (Brij35 and foam reducing agent)	0.05	Compressive: 40%	N.A.

Sonication and purified from carboxylated carbonaceous fragments	0.03	Compressive: 97.2%	N.A.
Sonication	0.5	Compressive: 15% Splitting tensile: 36%	ASTM C39 C496
Debulking, sonication and surfactant	0.048	Elastic Modulus: 75% Flexural: 50%	N.A.
Sonication and Gum Arabic surfactant with tributyl phosphate defoamer	0.08	Flexural toughness index: 57.5%	ASTM C1018-97

Thermal conductivity

The high thermal conductivity of the graphene provides the base for transferring the internal heat generated caused by the hydration reaction in concrete to the outer layers and thus, reducing the risk of the occurrence of early age cracking. The development process of hydration reaction in ordinary Portland cement is composed of several periods each having their own unique heat flow. These steps are commonly known as i. the initial period, ii. The induction period, iii. The acceleration period, and iv. The retardation period. This behaviour related mainly to three mechanisms, the silicate reaction, the dissolution of C3A and the precipitation of ettringite[57]. The early age cracking of concrete due to lack of sufficient curing treatment perhaps roots from the heat variation across the cross section of concrete element. The cracking becomes more significant in thicker concrete structure where the heat transformation zone becomes longer relatively. It might be of value to be mentioned that cracks generate due to the thermal gradient between layers. In terms of the time required for a proper curing, the use of Graphene in concrete will shorten this period as there would be less heat gradient in the cross section. A shorter curing period basically means less labour cost associated with the concrete projects as there would be less number of labours and hours required to take care of the poured concrete.

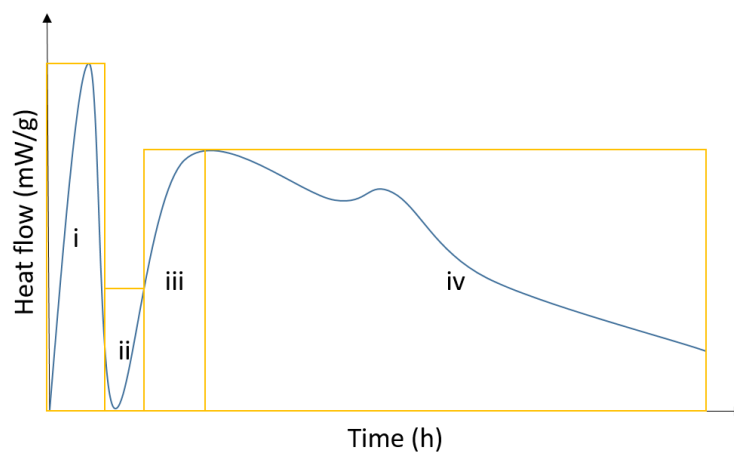


Figure 4 Hydration reaction enthalpy

The use of graphene in cement composite becomes beneficial once again in the context of thermal conductivity of concrete. A consistent heat transformation can be achieved reducing the chance of the development of the cracks.

Due to the fact that graphene presents a hydrophobic characteristic, and considering that the superplasticizers perform the workability solution based on the very hydrophobic characteristic, the use of graphene in cement composites is anticipated to exhibit a highly workable concrete with a right mixing method selected. Graphene, indeed, had been used in other industries as lubricant in order to reduce the friction.

Australian financial market position

As per May 2019, the data from the Australian Securities Exchange (ASX) reports a AUD\$78.8 M market capital for graphene. This is at the same time as the 4 largest builder companies active in the construction industry report a total of AUD\$19 B market capital. The Australian government is encouraging the industry leaning towards a more ecofriendly approach. Cement industry is one of the top ranked industries by carbon emission indices as production of each ton of Portland cement emits the same amount of carbon dioxide in to the atmosphere. The construction industry is leaning towards using Supplementary Cementitious Materials (SCM) in a wider scale in order to reduce the consumption of Portland cement in the benefit of environmental conservation[58]. Having an environmental perspective to the mass production of graphene, this material is more likely to be widely accepted in the Australian construction industry.

Environmental impacts of graphene production

Construction is identified as one of the most environmental hazardous industries across the globe. Enterprises, thus, are encouraged recently to adopt the sustainability and environmental construction approaches in their practices in many countries. Australia as a leader in the environmental conservation practices across the globe, puts restrictions and also motivators to employ the environmental approaches. Therefore, the succession of a project in Australia is closely dependant on the degree of environmental awareness. The financial aspects of graphene commercialization had been discussed in the previous section. However, it is less likely in Australia to achieve a higher market capital without a proper environmental evaluation on the manufacturing process of graphene nano particles.

Rickard Arvidsson et al. [59] reported the energy consumption of producing 1 kilogram of graphene from two separate routes known as ultra-sonication and chemical reduction of graphite as 470 MJ/kg and 1,100 MJ/kg respectively. This is while the green cement production reported to consume only about 3.3 MJ/kg ordinary Portland cement [60, 61].

Acknowledgement

This research could not be accomplished by the facilitation of Western Sydney University technical staff members, supervisors and fellow academics. Special thank goes to the Graduate Research School of Western Sydney University for regulating and providing the necessities required for the accomplishments of this paper. Western Sydney University library staff had fantastic contribution in the data collection procedure. The contribution of Institute of Construction Materials (ICONSMAT) had also shed light on the alignment of this research.

Conclusion

Based on the bibliometric analysis a gap had been identified between the studies conducted in the area of material science and the construction and building from 2008 until 2018. The present paper introduces the potentials of the use of graphene in the modern construction practices by focusing on sustainable construction principles. The photonic and electronic characteristics of graphene opens an area for development of a photovoltaic concrete panel to generate a portion of the electricity demand of buildings. Its outstanding mechanical properties had already showed satisfying results improving the compressive, tensile and flexural performance of cement composites made. The thermal conductivity and the hydrophobic characteristics are providing solutions for early age cracking and workability issues in concrete. However, the current high unit price of graphene is a barrier for a wider use of such material in concrete structures, thus, further detailed studies are required to facilitate the mass production by utilizing efficient processes for a wider use of graphene as a construction material.

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