



Research article

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Effect of graphene oxide on resistance to action of aggressive environments, tortuosity and economic index of concrete

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Abstract. Cement concrete though has emerged as one of the best construction materials concerning versatility and strength, suffers from durability when subjected to aggressive environments and action of water because of its internal cracks and voids. Further, because of its huge cement demand, it is blamed for concerning carbon emissions. As such when there is one need to strengthen its microstructure by modifying internal defects, there is another need to reduce the consumption of cement by enhancing the efficiency of concrete. To address these issues, this paper explores the impact of incorporating different percentages (0.01–0.1 % of binder content) of graphene oxide (GO) into concrete made with Portland slag cement and its subsequent effects on strength and durability. Apart from compressive strength, durability parameters such as water absorption, sorptivity, and resistance against attack of aggressive environments caused due to chloride (NaCl), sulphate (MgSO₄), and acid (H₂SO₄) have been examined. The results of this study emphasize the beneficial effects of adding GO to concrete regarding compressive strength enhancement 1.35–2.25 times, similarly increased durability against chemical exposure, and improved resistance to transport properties. The performance-related economic analysis in terms of the economic index revealed that the use of GO up to 0.03 % is beneficial from both a technical and an economic point of view. Use of GO more than 0.04 % may be beneficial where strength and time are more important than cost. The findings may contribute to the advancement of construction materials, which can potentially lead to the development of more durable and sustainable infrastructure.

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1. Introduction

Cement concrete has emerged as one of the best construction materials because of its strength and versatility. On the other hand, its durability is challenged due to its poor microstructure containing inherent cracks, voids and weak transition zones. Huge research works have been documented on the use of various kinds of fibers for arresting the micro-cracks and strengthening the microstructure, but the achievement is still far from the intended one. Past studies also reveal the use of finely ground materials as filler materials used for filling the microvoids. In recent years, researchers have explored the use of graphene to enhance the performance of concrete. Experiments and studies have shown that the addition of graphene improves the strength of concrete, making it more resistant to loads. In addition, the incorporation of graphene has led to significant findings indicating improved waterproofing properties.

Concrete is also blamed because of the embodied carbon and energy associated with cement production. As such development of the efficiency of concrete with a reduction of cement consumption is a vital issue.

Graphene and its related compounds are chosen as the most promising candidates for reinforcing material concerning their outstanding elastic modulus strength [1]. Graphene-reinforced materials have been reported in the past, and among these materials, cement-based materials, like concrete, stand out as the most widely consumed human-made material. The reinforcement of concrete and other cementitious materials is essential because they exhibit inherent weaknesses such as susceptibility to crack formation, low tensile strength, and limited strain capabilities [2]. According to Gong et al., the addition of 0.03 wt% graphene oxide (GO) to the cement paste (CPS) resulted in a significant increase, developing more than 40 % of compressive and tensile strengths [3]. In a different study, Pan et al. reported that incorporating 0.05 wt% of GO into the cement mortar led to notable improvements. Specifically, they observed enhancements up to 33 % in compressive and up to 59 % in tensile strength [4]. Lv et al. conducted studies that confirmed the positive impact of incorporating GO into CPS and concrete, leading to improved mechanical properties [5]. Despite numerous studies on the mechanical properties of GO in cement-based materials, information is scarce regarding its impact on durability [6]. Zeng et al. conducted experiments and found that the addition of 0.03 % of GO significantly improved the structural life of nearly 2.3 times cement mortars concerning external sulphate attack. The study informed that the addition of 0.03 % improved compressive strength by nearly 69 % and corrosion resistance by nearly 41 % [7].

Lu et al. investigated the dispersion and stability of GO in neutral water with and without silica fume (SF), as well as in CPS with and without SF. Results indicate that SF contributes to reducing the size of GO in water, enhancing its dispersion [8]. De Souza et al. discussed how graphene-based nanomaterials (GN) improve the properties of cementitious materials like concrete, making them more resistant to various stresses and extending their service life. However, challenges remain in accurately predicting their performance and conducting comprehensive assessments to determine economic and environmental benefits, highlighting the need for further research [9]. Yu et al. reported the impact of GO on the mechanical properties and microstructure of ultra-high-performance concrete. The addition of 0.04 % GO reduces total porosity, especially capillary porosity, enhancing hydration product formation. However, excessive GO content above 0.04 % leads to increased porosity and a slight decrease in mechanical properties due to poor dispersion and fluidity loss [10].

Muthu et al. explained that the characterization of GO synthesized via Hummers' method revealed a highly oxygenated and hydrophilic material, with functional groups including epoxy, hydroxyl, carbonyl, and carboxyl bonded to the graphene sheets. These findings underscore the role of GO content in influencing the hydration kinetics and rheological properties of CPS, with optimal dosage balancing improvements in mechanical strength against potential issues like workability and hydration retardation [11]. Zhao et al. investigated the effects of incorporating modified carbonaceous mesoporous materials (mCMK) and GO on cement mortar properties. Results showed that mCMK and GO improved both flexural and compressive strengths due to pozzolanic reactions, refined pore structure, and enhanced bonding between the matrix and aggregates. Furthermore, mCMK exhibited better dispersion, leading to superior performance compared to CMK, as evidenced by reduced porosity and increased hydration products, ultimately enhancing the mechanical properties of the cement mortar [12]. Dung et al. found that incorporating graphene, whether in pristine (PG) or aggregated (AG) form, enhanced the compressive strength of Portland cement (PC) mortar due to increased hydration and the formation of a denser microstructure. Moreover, the compressive strength improvement was more pronounced at earlier ages and decreased over time, with AG demonstrating better stability and enabling higher hydration levels compared to PG, ultimately resulting in superior compressive strength of AG samples [13]. Du and Dai Pang explained that the introduction of graphene nanoplatelets (GNP) into cement mortar results in a refinement of the pore structure, reducing the critical pore diameter and increasing tortuosity, leading to improved resistance against water penetration and chloride diffusion. However, at higher GNP concentrations, agglomeration becomes more pronounced, compromising the effectiveness of GNP in enhancing barrier properties as reflected in the decrease in water penetration resistance and chloride migration coefficient [14]. According to Yu et al., by incorporating a graphene Nano fluid additive (GNA) into carbon fiber-reinforced cement mortar (CFRCM), a synergistic effect was achieved, resulting in significant enhancements in flexural and compressive strengths by up to 101 and 126 %, respectively, surpassing control specimens. Moreover, the electrical and thermal conductivities of the modified material exhibited remarkable increases of 68 and 36 %, respectively, when compared to controls, indicating its potential for high-efficiency cathodic protection systems with a service life extension of over 200 % [15].

Sui et al. explained the microstructure of GO revealed amorphous multi-layers and wrinkles, with a high purity and oxidation rate as indicated by SEM and EDS analysis. The addition of GO enhanced compactness and mechanical strength while accelerating cement hydration, counteracting the hydration-retarding effect of waste ceramic powder (WCP), thereby enhancing the characteristics of the cement-WCP composite [16]. Jaramillo and Kalfat discuss the impact of different mixing methods on the performance of

concrete, focusing on workability, density, compressive strength, and flexural strength. It highlights that the addition of non-sonicated and sonicated GNP dispersions slightly decreased workability but increased compressive strength, with sonicated dispersions showing a more significant improvement. Moreover, increasing mixing time enhanced the compressive strength, indicating better distribution of GNPs within the concrete matrix [17].

Zeng et al. explore the impact of GO nanosheets on sulphate resistance in cement-based materials, revealing significant improvements in transport performance and durability. Through numerical simulations and experimental validation, the proposed chemo-mechanical model demonstrates that adding 0.03 wt. % GO extends structural life expectancy by 2.3 times, offering a cost-effective and environmentally friendly approach to enhance sulphate resistance without contributing to additional carbon dioxide emissions [7]. Salami et al. explained that the incorporation of graphene-based materials in cement alters the pore structure, promoting the formation of compact hydrated crystals, which enhances mechanical strength by minimizing large pores and cracks. Optimal quantities of graphene ensure the formation of regular-shaped crystals, improving microstructure densification and minimizing voids in the cement-graphene composite [18]. Sheikh et al. characterize three types of GO as lowly oxidized GO (LOGO), moderately oxidized GO (MOGO), and highly oxidized GO (HOGO) and investigate their hydronium activity and influence on cement performance. It suggests that the presence of a dense hydronium layer, particularly in HOGO, accelerates cement hydration, leading to denser microstructures and improved mechanical properties in GO-incorporated concretes [19].

Cui and Chang explained that the addition of multi-layer graphene (MLG) to ultra-high-performance concrete (UHPC) accelerated the hydration process, evidenced by earlier exothermic peaks, and increased ion concentrations, particularly calcium ions, promoting hydration product formation. Moreover, MLG refinement of the pore structure, evidenced by decreased pore volume and improved pore size distribution, contributed to enhanced mechanical properties, such as increased flexural and compressive strengths, indicating improved performance of UHPC [20]. Du et al. explained that the addition of GNPs to concrete refined the pore structure, reducing pore sizes and enhancing resistance to water and chloride ion penetration. However, beyond a certain threshold (1.5 % GNP), further addition led to diminished dispersion efficiency, resulting in clustered GNPs that negated the benefits of pore refinement [21]. Fonseka et al. reported that the relation between the compressive strength and abrasion coefficient of concrete containing GO is strongly correlated as indicated by the high coefficient of determination values. Essentially, as the compressive strength of the GO-incorporated concrete increases, the abrasion coefficient tends to decrease. This relationship suggests that enhancing the compressive strength of concrete through GO incorporation can lead to improved resistance against surface abrasion, thereby enhancing the durability of the concrete structure [22]. Suh et al. reported that the incorporation of well-dispersed Carbon Nanotubes (CNTs) in CPS generates interconnected and dense structures, enhancing sample durability by reducing pore scattering and forming three-dimensional networks. This highly connected structure leads to increased durability compared to GO-incorporated samples and ordinary CPS, highlighting the critical role of connectivity in determining material strength [23]. Wang and Zhong reported the characterization of synthesized GO revealed a thickness of about 2 nm and identified chemical functional groups through Atomic Force Microscope (AFM) and Fourier Transformed Infrared Spectra (FTIR) analysis. Three dispersion states of GO in Tricalcium Silicate (C_3S) pore solution were observed: well-dispersed (WDS), self-agglomerated (SAS) and grain-adsorbed (GAS), each affecting hydration differently. GO in GAS delayed hydration peak and promoted poison effects due to its conformal coating on C_3S grains, while GO in SAS exhibited delaying effects possibly due to its absorption capability for Ca^{2+} ions, as evidenced by isothermal calorimetry and ion absorption experiments [24].

Ghazizadeh et al. reported the role of GO in PC paste was investigated, revealing that GO aggregates in the pore solution rather than interacting with the surface of hydrating PC particles. While low concentrations of GO slightly increased compressive strength, the extent of improvement was minor and correlated with changes in hydration heat, suggesting that GO primarily enhances strength by increasing hydration degree rather than reinforcing the paste microstructure. Additionally, the presence of polycarboxylate ether (PCE) copolymer did not prevent GO aggregation in PC pastes, indicating that reported strength improvements in PC-GO-PCE systems are likely not due to well-dispersed GO but rather to changes in hydration degree [25]. Lu et al. introduced a novel method for enhancing the mechanical properties of strain-hardening cementitious composites (SHCC) by coating polyethylene (PE) fibers with GO, which improves chemical reactivity and surface wettability. The resulting GO/PE fibers significantly strengthen the bond between fibers and the matrix, leading to a notable increase in strain capacity and tensile strength of SHCC without compromising compressive strength [26].

The literature survey indicated that a good number of papers are documented concerning the use of various forms of graphene in concrete. Most of the papers have focused on mechanical properties. Though some papers are available concerning durability describing a few aspects, there's still a gap in understanding the performance of GO-modified cementitious materials under real-world conditions, like

aggressive environments of chemical exposure and tortuosity under the action of water. Since graphene is a very costly material in comparison to the ingredients of concrete, there exists a need to understand the economic benefits of GO-modified concrete. Further, it is understood that most of the available literature is based on the use of ordinary Portland cement (OPC). Concerning the high embodied energy and carbon of OPC in comparison to slag-based cement like Portland slag cement (PSC) and fly ash-based cement like Portland pozzolana cement (PPC), the use and production of OPC is discouraged. As such, there is a need for knowledge of GO-modified concrete using PSC and PPC to encourage the production and utilization of waste-based cement. The present work may help bridge the above gaps.

By incorporating oxide into the concrete mix, this study aims to enhance the material's mechanical properties, such as compressive strength, and improve its resistance to chemical attack. This advancement could lead to the construction of more durable and sustainable infrastructure, resulting in longer-lasting buildings and structures that require less frequent maintenance and repair. Additionally, the reduced water absorption and improved waterproofing properties of graphene-modified concrete could prevent moisture-related issues, extending the service life of constructions in various environments. The performance-related economic analysis made in terms of the economic index may be a deciding factor in issues related to time, cost and quality in construction management. Ultimately, this research has the potential to contribute to the development of innovative and resilient construction materials, advancing the field of civil engineering and enhancing the sustainability of infrastructure worldwide.

2. Methods

2.1. Materials

In this research, PSC was utilized, adhering to the specifications outlined in IS: 455-2015 (Reaffirmed 2020) [27]. Table 1 displays the various properties of the cement. As for the aggregates, natural granite stone and natural riverbed sand are employed, meeting the requirements of IS: 383-2016 [28] for coarse and fine aggregates. Table 2 provides a presentation of their properties. For the incorporation of GO, it was procured from International PranaGraf Mintech Research Centre (IGMRC), Bhubaneswar, Odisha, India.

Table 1. Properties of PSC.

Property	Results	Requirements
Setting time-initial (min)	89	≥30
Setting time-final (min)	198	≤600
Standard Consistency (%)	32	–
Soundness (mm)	1	≤10
Sulphur trioxide (%)	2.49	≤3
Chloride content (%)	0.01	≤0.1
Magnesium oxide (%)	4.37	≤6
Sulphide sulphur (%)	0.16	≤1.5
Insoluble residue (%)	2.28	–
Loss in ignition (%)	1.51	≤5

Table 2. Properties of the aggregates.

Property	Coarse Aggregate	Fine Aggregate
Flakiness index (%)	17.5	–
Elongation Index (%)	12.5	–
Specific Gravity	2.83	2.7
Impact Value (%)	14.7	–
Abrasion Resistance	18.4	–
Water Absorption (%)	0.3	0.76
Crushing Value (%)	19.7	–

2.1.1. Characterizations of graphene oxide

The GO used in this work has a purity of about 99.99 % carbon and the Brunauer–Emmett–Teller (BET) specific surface area value of 384.180 m²/g. The micro Raman spectroscopy of GO is shown in Fig. 1. It shows peaks of G (corresponds to the lattice of graphite), D (corresponds to 1st order-disorder peak in the graphite lattice structure), and 2D (2nd order-disorder peak in the graphite lattice structure). The D, G, and 2D peaks were observed at 1344, 1573, and 2686 cm⁻¹, respectively. The intensity ratio of

peaks of 2D to G (I_{2D}/I_G) is 0.4, indicating a few layers (4–5 layers) of graphene sheets present in the GO. The band gap energy value of GO was determined by ultraviolet–visible diffuse reflectance spectroscopy (UV–Vis DRS) using the Kubelka–Munk function vs. the energy of light. The band gap energy was found to be 3.59 eV (Fig. 2).

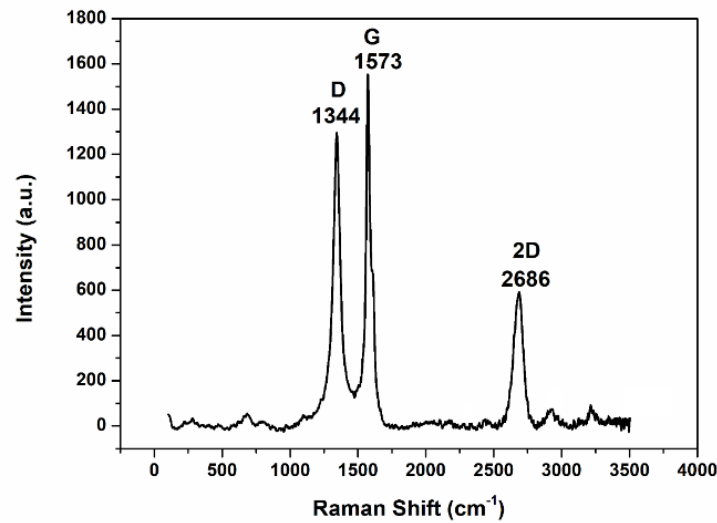


Figure 1. Micro Raman spectrum of graphene oxide.

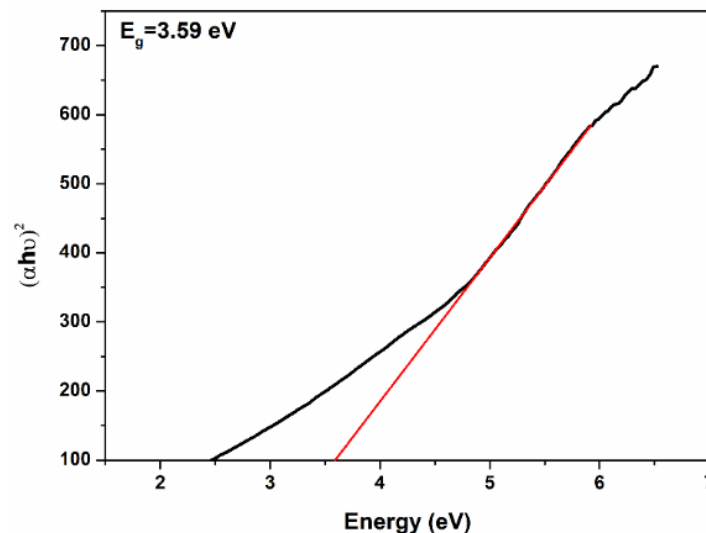


Figure 2. Determination of band gap energy (E_g) of graphene oxide from the UV-Vis DRS spectrum.

2.2. Methodology

2.2.1. Mix design and ingredients

Following the guidelines of IS: 10262-2019 [29], a mix design of M20 grade concrete was prepared using PSC, 12–20 mm size black hard granite aggregates, river sand, portable tap water, and a carboxylate-based superplasticizer. The mix design suggested a proportion of 1:2:4, wherein the fine aggregate quantity was 2 times and that of coarse aggregate was 4 times of cement used. A water-cement ratio of 0.45 and a superplasticizer dosage of 0.5 % of cement were used. As such, for making 1 cum of concrete, 330 kg of PSC, 660 kg of fine aggregates, 1320 kg of coarse aggregates, 149 kg of water, and 1.65 kg of superplasticizer were used. The GO was added to such concrete in 0, 0.01, 0.02, 0.03, 0.04, 0.05, and 0.1 %. These mixes were named as PSC-0, PSC-1, PSC-2, PSC-3, PSC-4, PSC-5, and PSC-10 respectively.

2.2.2. Preparation of concrete and samples

For the preparation of concrete, the GO was mixed with cement thoroughly in dry conditions in a laboratory-type pan mixture for 5 minutes. This process was carried out under controlled laboratory

conditions to avoid agglomeration and achieve better dispersion. Then the dry mixture of GO and PSC was further mixed with fine aggregate for 5 minutes and then with coarse aggregate for 5 minutes in two different stages, one after another. The required amount of superplasticizer was added to water and mixed with the dry mixture of all other materials. Mixing continued for nearly five minutes to get a uniform homogeneous concrete mix. The mixing time adopted was more than twice of normal concrete for good dispersion of GO. After measuring the slump of concrete, the required number of cubes of 100 mm size were prepared for the test of compressive strength, acid resistance, sulphate resistance, chloride resistance, water absorption and sorptivity. The prepared samples covered with PE were allowed to be cured at room temperature for 24 hours, after which they were put under water curing till attaining the required test age.

2.2.3. Test ages

When the strength test was conducted at the ages of 7, 28, and 91 days, the durability parameters concerning acid resistance, sulphate resistance, chloride resistance, water absorption, and sorptivity were examined at the ages of 28 and 91 days. The average results of three samples were considered as the result of a particular mix.

2.2.4. Test procedure

The workability of concrete mixes was examined as per the guidelines of IS: 1199-1959 (Reaffirmed 2004) [30]. For the compressive strength test, IS: 516-2021 [31], for the test of acid and sulphate resistance, the procedure, as reported in past publications [32], was followed. For this purpose, 28 days of water-cured cubes were further cured for 28 days in 1 % acid and sulphate solutions. The results of acid and sulfate-cured samples were studied, considering strength and weight loss and compared with the results of water-cured samples of the same age. The test of sorptivity, the procedure published by Acharya and Patro [33] was followed, wherein the samples were wax-painted from all sides. The unpainted bottom surface was kept over the stands in such a position that only 5mm of the bottom was immersed in the water. The ingress of water was noted at the end of 9, 49, 100, 169, 256, and 361 minutes by taking the weight before and after water immersion. The water absorption test was conducted as per IS 1124-1974 (Reaffirmed 2003) [34]. The chloride resistance test was conducted by immersing the samples in a solution containing 20 % sodium chloride. The effect of sodium chloride was checked in terms of strength change and mass change. The results of GO-modified concrete were compared with normal concrete without GO.

3. Results and Discussions

3.1. Workability

The workability of all concrete mixtures was checked after 15 minutes of adding water. The slump of normal concrete (PSC-0) was measured as 75 mm. The same for mixes (PSC-1, PSC-2, PSC-3, PSC-4, PSC-5, and PSC-10) having different dosages of GO were recorded as 68, 67, 65, 64, 63, and 60 mm. Thus, it can be said that there is not much influence of GO on workability. With the highest dosage application of GO (0.1 %), the slump value reduction is only 20 %.

3.2. Compressive Strength

The first set of data concerning (PSC-0) shows the compressive strength values of the material without adding any graphene. At 7 days, the compressive strength is 15.96 MPa, which increases to 20.93 MPa at 28 days, and further increases to 26.94 MPa at 91 days. These values represent the baseline compressive strength of the material, which is used for comparison with the values obtained after adding different amounts of graphene. The results are presented in Fig. 3.

The subsequent data shows the compressive strength values after adding different amounts of graphene to the material concerning mixes PSC-1, PSC-2, PSC-3, PSC-4, PSC-5, and PSC-10 that contained GO 0.01, 0.02, 0.03, 0.04, 0.05, and 0.10 %. Adding 0.01 % graphene results in a significant increase in compressive strength by nearly 27 % at 7 days, 35 % at 28 days, and nearly 38 % at 91 days. As the amount of graphene added increases, so does the compressive strength of the material. Adding 0.02 % graphene increases the compressive strength by nearly 62 % at 7 days, 70 % at 28 days, and a similar amount at 91 days. A dosage of 0.03 % graphene results in compressive strength development of 95 % at 7 days, 1.04 times at 28 days, and 1.05 times at 91 days in comparison to PSC-0, which was made without GO. Adding 0.04 %, the strength development is nearly 1.05, 1.16, and 1.08 times of normal concrete at 7, 28, and 91 days. The same inclusion of 0.05 % grapheme oxide content was found to be 1.08, 1.21, and 1.21 times. The results on a higher dosage of grapheme oxide amounting to 0.1 % were found to be 1.08–1.24 times of normal concrete made of PSC.

On reviewing the percentage of strength increase it is seen that at the end of 7 days, the strength development varied 0.27–1.08 times of normal concrete on application of 0.01–0.1 % of GO. The same at 28 days was 0.35–1.25 times and at 91 days it was 0.38–1.24 times. The result indicated very good strength

development at an early age, which may bring huge technical and economic advantages due to less requirement of stripping time.

Concerning the 28-day results, it is seen that the normal concrete (PSC-0) attained a strength of 20.93 MPa. At a dosage of 0.01 % GO, the achieved strength is related to the strength of M25-grade concrete. Similarly, with the addition of 0.02 % of GO, the achieved strength relates to the M35 grade of concrete. The trend of results on the application of 0.03 and 0.04 % is related to the strength of M40 and M45 grades. Further increases in the dosage of GO, like 0.05 and 0.1 % enhanced the strength but not significantly, as was noticed in the case of the application of GO up to 0.04 %. The results may be very useful in structural applications by enhancing the efficiency of concrete by applying small dosages of GO based on the requirement for achieving different grades of concrete.

Visiting the results of 91 days, it is seen that there is a continuous development of strength, which is very encouraging. The results showed that after 13 weeks of water curing, M20 grade concrete is converted to M35–M60 grades of concrete on application of 0.01–0.1 % of GO. The results revealed that upon the application of GO, 0.03 % more than 55 MPa strength was achieved. After that, the strength development slows down and with the application of GO 0.1 %, 60 MPa is achieved. This data may be very useful in construction activities where curing can be continued for 13 weeks.

The reason behind the development of strength is attributed to the physical properties of GO. It affects concrete health as a filler as well as a reinforcing material. As a filler, at an early age, it modified the transition zone by increasing bonding between the interface of coarse aggregate and paste. The addition of graphene improves the dispersion and bonding within the concrete matrix. This enhances load transfer and increases compressive strength. Findings are found in line with the views of the authors of the past [36]. At a later age, it stitched the micro-cracks and microvoids. It also helped in arresting the development and propagation of internal cracks. The GO also contributed to the refinement of the pore structure and reduced the pore size. Graphene acts as a nucleation site for hydration products, accelerating the hydration process and promoting the formation of a denser microstructure. GO fine particles help in promoting the formation of regularly shaped compact crystals of hydration products that enhance the internal structure of GO-modified concrete. The studies of the past by Bai et al. and Wang et al. validate the results of the present study [35, 36]. Comparing the results of all ages, the rate of strength development was best up to 0.04 % application of GO. Further application of GO, though it enhanced the strength, the rate of strength development was slower, which may be due to improper dispersion and loss of fluid.

The observed strength enhancement can be attributed to the optimized dispersion of GO, achieved through thorough dry blending with cement, followed by separate blending with fine and coarse aggregates for five minutes each. This stepwise mixing process ensured effective utilization of GO's high specific surface area and abundant functional groups. Such uniform dispersion likely facilitated accelerated hydration kinetics and the development of a refined, denser matrix, thereby contributing to improved mechanical performance.

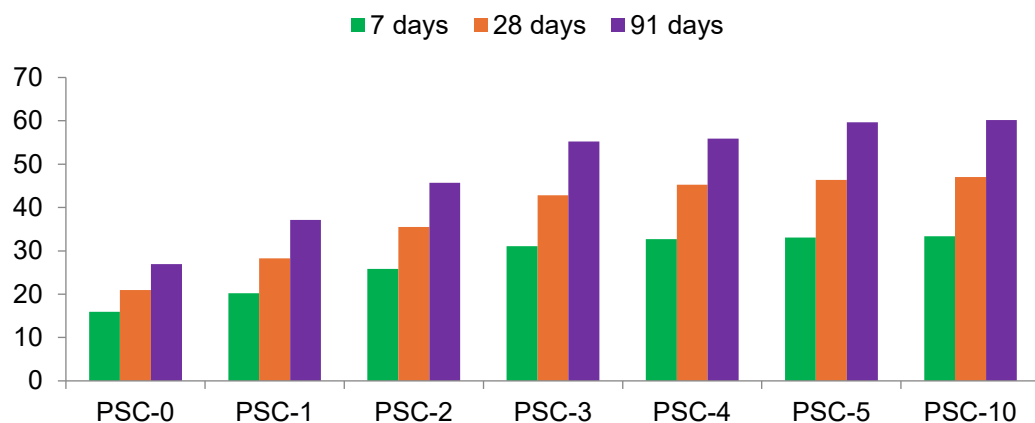


Figure 3. Compressive strength of concrete with and without GO.

3.3. Chloride Resistance

The chloride resistance of concrete samples with and without GO was examined through the calculation of weight change and strength change after 28 days of chloride solution curing. The mass and strength change were compared with the 28-day results of the water-cured samples. The results of mass change and strength change are reported in Figs. 4 and 5.

After 28 days of chloride curing, there is a minor decrease in weight up to 1 % in concrete samples without GO (PSC-0). On the other hand, there is an increase in weight up to 1 % in samples with GO (PSC-1 to PSC-10). A much smaller amount of loss in weight of normal concrete is due to its slag cement, made of ground granulated blast furnace slag. The elemental composition of slag cement makes the cement composite resistant to aggressive environments. The increase in weight of the GO-modified samples is due to the robustness of their internal structure and surface hardness. From visual inspection, it was seen that the surface of the GO-modified samples was very hard, which repelled the ingress of chloride and external moisture thereof. Due to the presence of moisture outside the sample, concerning loss of moisture from the internal part of the sample. As such, the hydration continued with the help of internal moisture. Because of the hydrated products, the weight of the samples increased. In the case of normal concrete, it was seen that the surface was a little affected, for which there was a little mass loss.

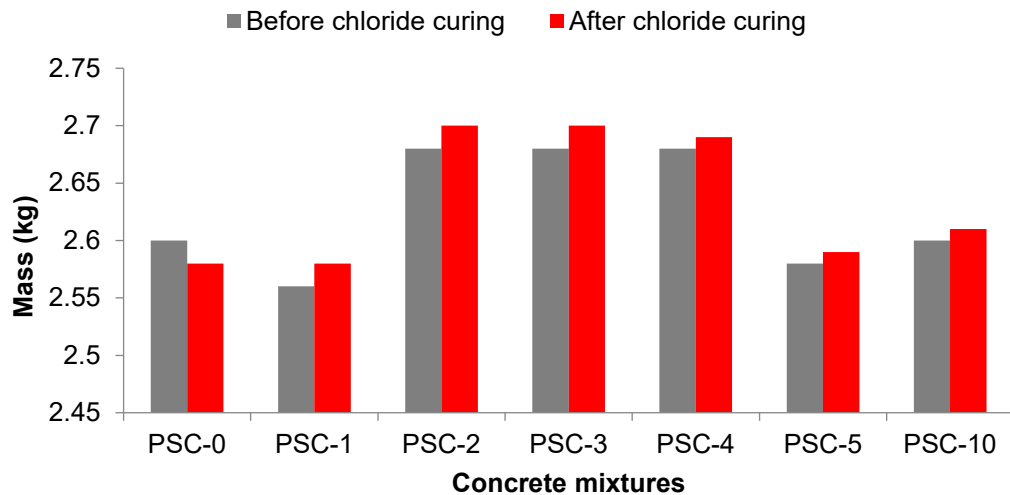


Figure 4. Change in mass of concrete with and without GO under chloride curing.

After 28 days of chloride curing, the samples with GO and without GO were subjected to a compressive load test to estimate the effect of chloride on concrete health. It is seen that chloride could not affect negatively. When the concrete made of PSC suffered a minor loss of strength up to less than 1 %, there was an improvement in the strength of 4–19 % in GO-modified concrete. PSC, which is made of blast furnace slag, is said to have better resistance to aggressive environments. In the case of GO-modified concrete, there was no moisture movement due to the dense internal structure on modification of transition zones, filling of microvoids and arresting the microcracks. In the absence of moisture movement, there was also no loss of internal moisture. The bound water and capillary water helped in continuing the hydration process, for which the strength improvement is noticed. Further to state, GO has a high surface area and can chemically adsorb chloride ions onto its surface, reducing their concentration in the concrete pore solution and mitigating the chemical degradation of concrete due to chloride attack. Functionalized graphene nanosheets can undergo ion exchange reactions with chloride ions, leading to the formation of less soluble compounds or complexes that are less harmful to concrete. The results and the findings thereof are validated by the previous studies [37, 38].

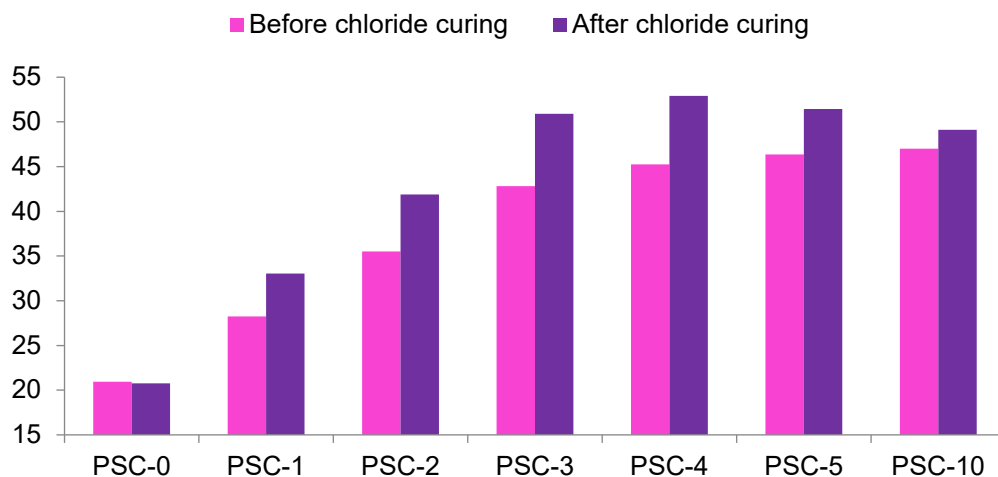


Figure 5. Change in strength of concrete with and without GO under chloride curing.

3.4. Sulphate Resistance

The resistance of GO-modified samples against sulphate attack was determined in terms of weight and strength variation after immersion of samples in sulphate solution for 28 days. The results were compared with concrete samples made without GO. For this purpose, the weight and strength of samples were measured before and after immersion and the variation was computed. Such results are presented in Figs. 6 and 7.

The effect of magnesium sulphate on normal concrete was noticed, wherein the weight loss was more than 1 %. However, this loss is much less because of the positive effect of the slag, from which the cement was made. The action of sulphate on GO-modified concrete was defended by the hard surface structure and dense core structure, for which there was no negative impact. Rather, a little growth in weight was noticed. This growth may be due to the production of hydrated products. From visual inspection, it was seen that the surface of the GO-modified samples was very hard, which repelled the ingress of sulphate and moisture thereof. In the case of normal concrete, it was seen that the surface was a little affected, for which there was a little mass loss.

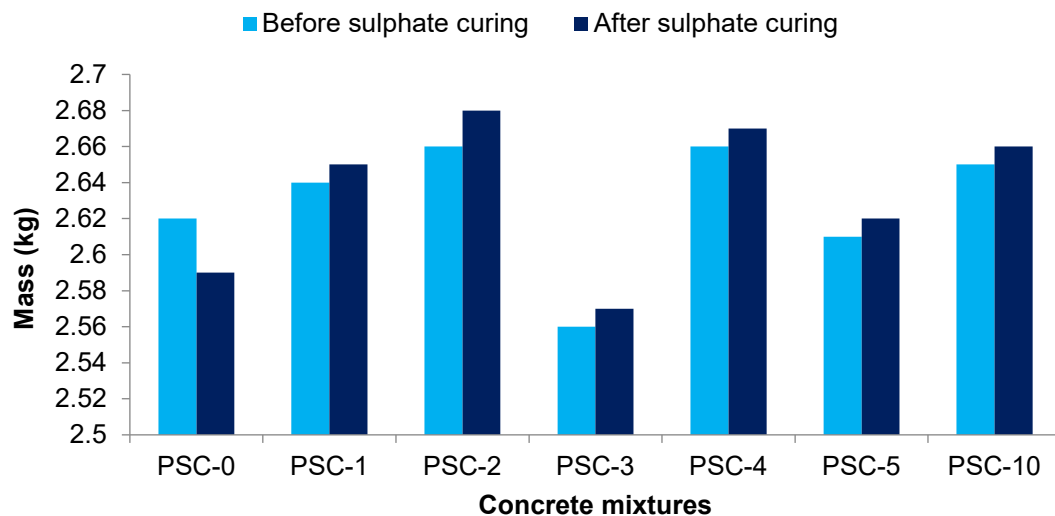


Figure 6. Change in mass of concrete with and without GO under sulphate curing.

On a compressive strength test of normal concrete (PSC-0), it was estimated that there was a loss of strength of 3 %. There was no detrimental effect of magnesium sulphate on GO-modified concrete. There was a growth of strength of 5–13 % after 28 days of sulphate curing. Here, the sulphate solution did not contribute to the health and hydration of concrete but at least helped in arresting the release of internal moisture. The sulphate solution could not enter due to the hard external structure. In the absence of moisture movement from the sample or into the sample, the hydration continued with the internal moisture available in gel pores and capillaries, for which strength development was noticed. GO's high surface area and reactivity can facilitate chemical interactions with sulphate ions, potentially forming barrier layers or stable compounds that inhibit sulphate attack [7]. Through its ability to adsorb and immobilize ions, graphene may reduce the availability of sulphate ions for detrimental reactions, thereby preserving the integrity of the concrete matrix. By incorporating graphene into concrete, both physical and chemical actions during sulphate attack can be influenced positively, resulting in improved resistance and durability. These advancements underscore graphene's potential as a multifunctional additive in concrete materials. The findings of the present study are somewhat in line with past studies [7, 39].

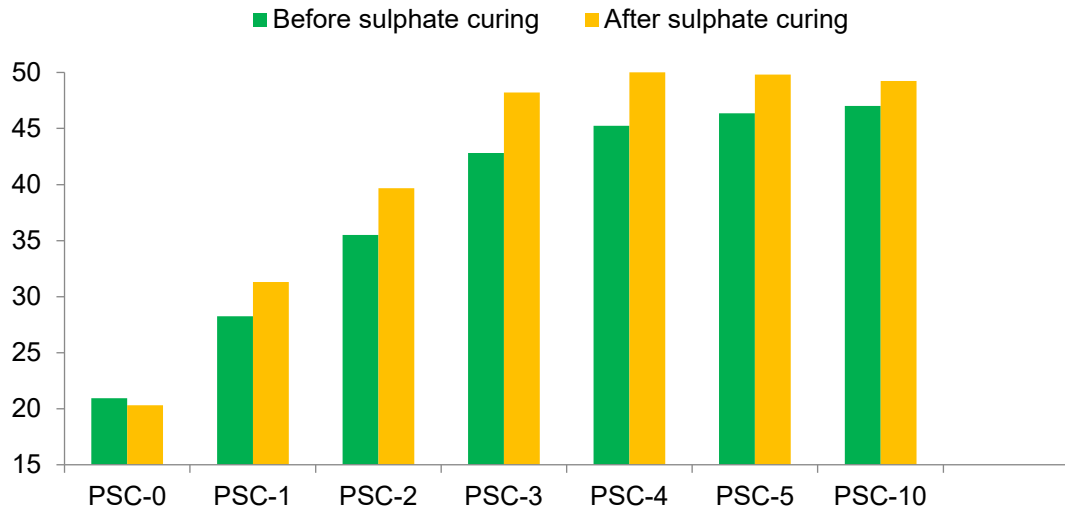


Figure 7. Change in strength of concrete with and without GO under sulphate curing.

3.5. Acid Resistance

The response of GO-modified samples against the action of sulphuric acid was determined by calculating the change in weight and strength after immersion of 28-day-old water-cured samples in the sulphuric acid solution for 28 days. The results were compared with concrete samples made with PSC only. For this purpose, the weight and strength of samples were measured before and after immersion and the variation was computed. The results are presented in Figs. 8 and 9.

The change in weight in normal concrete was negatively calculated as 0.74 %. On the other hand, the weight change was positive in the case of GO-modified concrete varied by 0.39–0.75 %. Thus, it can be said that the effect of acid was not efficient on the Go-modified concrete sample. Graphene, when incorporated into concrete, forms a physical barrier against acid ingress due to its impermeable nature, hindering the penetration of acid ions into the concrete matrix. In the interior of the sample, the hydration of cement continued and because of the hydrated products, the weight increased. The increase in weight during an acid attack can be explained by the absorption of acid into the material matrix. This absorption occurs due to the porous nature of the material, allowing the acid molecules to infiltrate its structure. As a result, the material gains weight.

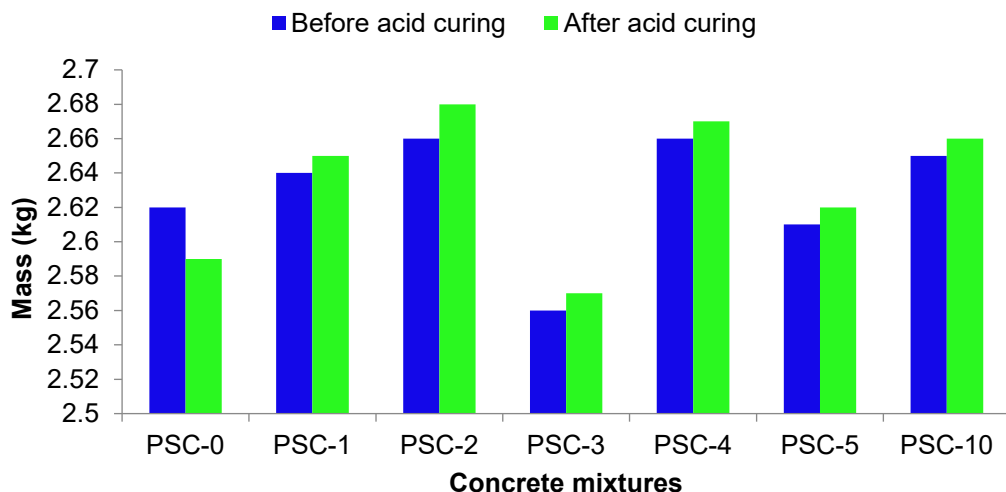


Figure 8. Change in mass of concrete with and without GO under acid curing.

The negative effect of sulphuric acid on the strength of PSC-made concrete was 2.05 %. It is seen that the effect of acid on samples containing GO was not active because of the hard surface structure and dense microstructure. When the inner part of the samples remained unaffected, the hydration process continued. The cohesive nature of graphene enhances the interfacial bonding between cementitious materials and aggregates, resulting in higher compressive strength, for which the strength developed 7.51–16.02 %. The reason behind the development of the strength of GO-modified concrete may be described as the two-dimensional structure of GO provides an impermeable barrier to acid molecules,

effectively limiting their penetration into the material. This barrier effect prevents direct contact between the acid and the material's reactive components, reducing the rate of chemical degradation. Further, GO is chemically inert and resistant to corrosion, making it highly stable in acidic environments. Unlike traditional materials, graphene does not undergo chemical reactions with acids, thereby preserving its structural integrity and mechanical properties. This inert behavior ensures the long-term durability and performance of graphene-enhanced materials under acid exposure. As such, the remarks of past authors [40, 41] validate the results and findings of the present study.

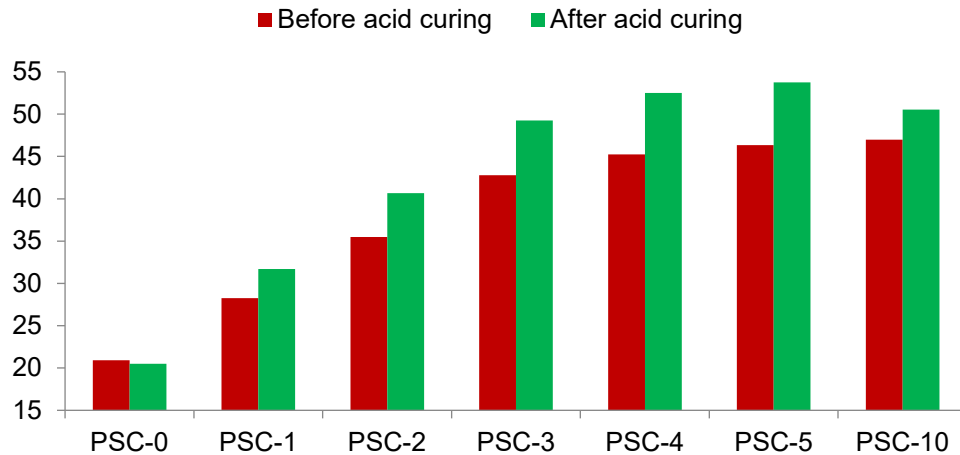


Figure 9. Change in strength of concrete with and without GO under acid curing.

3.6. Water Absorption

Water absorption characteristics of concrete affect its durability and strength. In this study, the water absorption percentage was measured for concrete cubes containing different percentages of graphene. The results show that as the percentage of graphene increases, the water absorption percentage decreases.

Concrete cubes with no graphene had a water absorption percentage of 2.8 %. This value decreased to 2.29 % when 0.01 % graphene was added to the mix. The reduction in water absorption was more significant as the percentage of graphene increased. Concrete cubes with 0.02 % graphene had a water absorption percentage of 2.15 %, while those with 0.03 and 0.04 % graphene had values of 1.94 and 1.89 % respectively. The water absorption percentage for concrete cubes with 0.05 % graphene was 1.62 %. Finally, the addition of 0.1 % graphene resulted in a water absorption percentage of 1.6 %, indicating the highest reduction in water absorption among all percentages tested. Thus, due to the addition of GO (0.01–0.1 %), the water absorption was reduced by 18–43 % in comparison to normal concrete.

GO, being a two-dimensional nanomaterial, can physically block water ingress into concrete pores due to its impermeable nature. This barrier effect reduces the absorption of water by the concrete. The results suggest that the addition of graphene to concrete can significantly reduce its water absorption percentage. This can lead to better durability and strength of the concrete, as lower water absorption means less moisture in the concrete, reducing the likelihood of damage due to freeze-thaw cycles or other forms of weathering. The reduction in water absorption may also result in less corrosion of the steel reinforcement in the concrete, further increasing the durability of the structure. Overall, the addition of graphene to concrete can lead to a more sustainable and long-lasting construction material.

3.7. Sorptivity

The sorptivity (**S**) is a function of water ingress due to the capillary action of water. This transport property of concrete samples with GO and without GO was examined using oven-dried samples, considering weight gain due to sorption of water. The samples were placed in water, with 5 mm immersed from the bottom face. The water ingress was calculated at 9, 16, 25, 36, 49, 64, 91, 100, 121, 144, 169, 196, 225, 289, 324, and 361 minutes by finding the difference in weight after and before immersion. The interval of time (**t**) for measurement of weight gain was so selected that the value of $\sqrt{t}$ would be a whole number for ease of plotting. The **S** was calculated using a linear regression of **i** versus $\sqrt{t}$, where **i** is cumulative water absorption per unit area inflow in g/mm^2 . The results of the **S** test are shown in Fig. 10. When the **S** index of normal concrete was found as 0.9035, it was reduced to 0.7592 on application of 0.01 % of GO, which shows a reduction of sorption due to capillary action of nearly 16 %. Further application of GO reduced the sorption value by up to 23 %. GO acts as a barrier to water penetration due to its impermeable nature. As water molecules attempt to pass through the concrete matrix, the presence of

graphene can hinder their movement, reducing **S**. Incorporation of GO increases the tortuosity of the pore structure within concrete. This means that water molecules have a more convoluted path to travel through the material, which slows down their movement and reduces **S**. The reports of the past [42, 43] validate the findings of the present study.

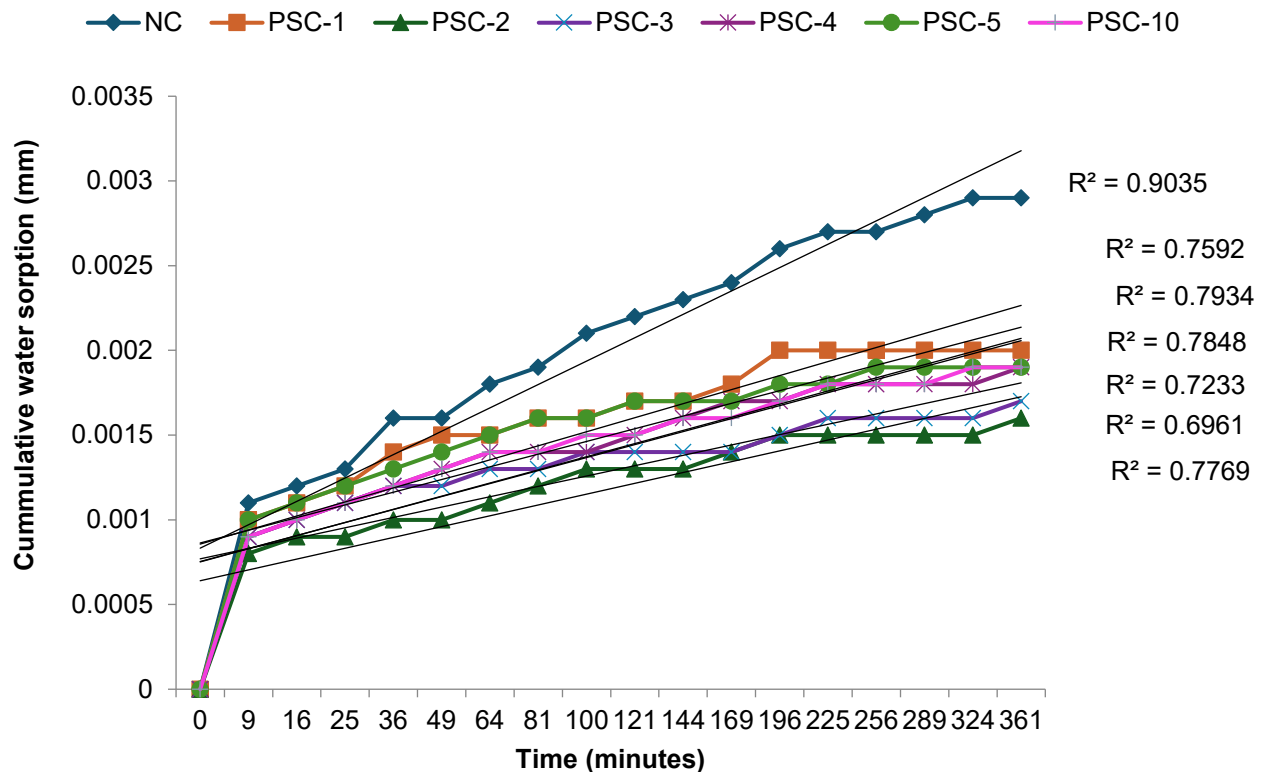


Figure 10. Sorptivity of concrete with and without GO.

3.8. Economic Analysis

The economic analysis of all concrete mixes with GO and without GO was analyzed in terms of an economic index (EI), considering both cost and performance at the end of 28 and 91 days. The EI was computed as a ratio of compressive strength to the cost of the mix. The cost of mixes per cubic meter has been derived based on the market available price of all materials those are mentioned in Table 3. Since the cost of machinery, labour, and tools is equal for all concrete mixes, the same has not been taken into the calculation of cost. Table 3 presents the EI of all concrete mixes.

The results show that though GO is very costly in comparison to other ingredients of concrete, still the concrete mixtures containing 0.01 % (PSC1), 0.02 % (PSC2), and 0.03 % (PSC3) have economic viability with EI values more than the mix without GO (PSC0) at the end of 28 and 91 days. On application of 0.01–0.03 % of GO to concrete, having 0 % GO has changed the concrete strength M20 grade to M40 grade at 28 days and to M55 at 91 days. At the same time, it is also estimated that the EI has increased 1.38–2.54 % at 28 days and 2.75–3.43 % at 91 days. Such concrete mixes (PSC1, PSC2, and PSC3) are found suitable for all types of structural works. On application of further increased doses of GO, like 0.04–0.1 %, the strength has increased up to 47 MPa at 28 days, but the EI decreased 7.01–47.94 %. Similarly, the mixes containing 0.04–1 % of GO enhanced the strength up to 60 MPa but brought the EI value 10.77–48.18 %. Such types of concrete mixes (PSC4, PSC5, and PSC10) may be suitable for restoration and rehabilitation works in case of natural calamities and war situations where cost is not a factor in comparison to time and strength requirements.

Table 3. Economic indices of various concrete mixes per cum.

Mix	CS-28d	CS-91d	Concrete ingredients						Cost	EI-28d	EI-91d
			PSC	GO	CA	FA	W	SP			
Rate (INR)			7	50000	1.25	0.45	0.2	420			
PSC0	20.93	26.94	330	0	1320	660	149	1.65	4979.8	0.004203	0.005410
PSC1	28.25	37.10	330	0.033	1320	660	149	1.65	6629.8	0.004261	0.005596
PSC2	35.50	45.70	330	0.066	1320	660	149	1.65	8279.8	0.004288	0.005519
PSC3	42.80	55.20	330	0.099	1320	660	149	1.65	9929.8	0.004310	0.005559
PSC4	45.25	55.90	330	0.132	1320	660	149	1.65	11579.8	0.003908	0.004827
PSC5	46.35	59.65	330	0.165	1320	660	149	1.65	13229.8	0.003503	0.004509
PSC10	47.00	60.20	330	0.33	1320	660	149	1.65	21479.8	0.002188	0.002803

(CS: Compressive strength, PSC: Portland slag cement, GO: Graphene oxide, CA: Coarse aggregate, FA: Fine aggregate, W: Water, SP: Superplasticizer and EI: Economic index)

4. Conclusions

The experimental work on graphene-reinforced concrete has provided valuable insights into the potential of GO as a reinforcement material for concrete. By varying the dosage of graphene from 0.01–0.1 % and conducting various tests on strength and durability, the present work has yielded the following findings:

1. On application of 0.01 % of GO, a concrete of M20 yields strength more than that of M25 and M35 at the end of 28 and 91 days. The strength of M20-grade concrete achieved the strength of M35 and M45 at 28 and 91 days on application of 0.02 % of GO. At the same period, a dosage of 0.03 % of GO converted M20 grade concrete to M40 and M55 grade. At 28 days, the strength further increased to M45 grade on application of 0.04 % GO. Further application of GO at the rate of 0.05 % and 0.1 % enhanced the strength up to 47 MPa and 60 MPa at 28 and 91 days, but the rate of growth in strength was lower than the GO dosages 0.01–0.04 %.
2. Facing an adverse environment of chloride, when PSC-made normal concrete suffered a loss of weight and strength up to 1 %, GO-modified concretes gained in weight and strength up to 1 % and 19 % respectively.
3. Fighting with an aggressive environment of magnesium sulphate, when concrete without GO lost weight by up to 1 % and strength by up to 3 %, the concrete mixed with GO gained a little weight and strength by 5–13 %.
4. On an attack of sulphuric acid, when normal concrete lost a mass of 0.74 %, GO-modified concretes gained a mass of 0.75 %. Under the same condition, strength loss was 2 % in normal concrete and strength gain was 8–16 % in GO-modified concrete.
5. The water absorption is reduced by 18–43 % on the application of 0.01–0.1 % of GO.
6. The sorption due to capillary action is reduced by 16–23 % when 0.01–0.1 % of GO was added to concrete.
7. The EI concerning to strength and cost of concrete mixes containing 0.01–0.03 % of GO is found to be better than normal concrete.
8. Beyond 0.03 % of the application of GO, the strength increases, but the EI decreases. However, such types of concrete mixes containing 0.04–0.1 % of GO may be suitable for restoration and rehabilitation works in case of natural calamities and wars, where cost is not a factor in comparison to time and strength requirements.
9. The findings of this study may support the transition to a more sustainable and resilient built environment.

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