Synergetic Effect Of Graphene Oxide And Fly Ash On Workability, Mechanical And Microstructural Properties Of High-strength Concrete

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ABSTRACT

Graphene oxide (GO) emerged as a new carbon-based nanomaterial known for its complex carbon structure and strength properties. In the current investigation, an attempt is made to improve the workability of GO-concrete; fly ash is introduced to form a cost-effective mineral admixture. The effects of GO addition at 0.15% and replacement of fly ash at 10%, 20% and 30% on fluidity, mechanical and microstructure properties of high-strength concrete were investigated. The experimental results showed that fly ash plays a significant role to enhance fluidity and hence workability, owing to the ball effect, size gradation and less water requirement. Therefore, the influence of GO on the decrease in fluidity was counterbalanced by fly ash. Also, GO counterbalanced the fly ash influence on delaying early-age strength progress. The replacement of fly ash at 20% and GO addition at 0.15% showed an enhancement of compressive, split tensile and flexural strengths by 6%, 7% and 12% at 7 days and by 10%, 12% and 16% at 28 days, respectively, in comparison with control concrete. The microstructure studies of SEM, EDX, FTIR and XRD indicated that the formation of better hydration phases leads to the densification of microstructure in FA-GO-concrete. The present investigation shows an advantage of combining GO and fly ash which play a synergistic role and counterbalance the drawbacks of one another.

KEYWORDS: Concrete, Graphene oxide, Fly ash, Workability, Microstructure, Mechanical properties.

INTRODUCTION

Cement concrete is a commonly used building material in all types of construction, but conventional concrete has less resistance to crack propagation or fracture. In addition to the strength characteristics, the performance of the concrete under the service environment is very much essential for all important structures. Different types of fiber have been incorporated to modify the properties of concrete, although the benefits of adding fibres have been shown at the macro-scale level and they are not yet proven to be effective in reducing porosity or preventing microcracks (Sally and Ali, 2021; Sancheti et al., 2020; Stephen and Mahachi, 2020).

Nanomaterials are currently being used as additives to cement composites as a number of different nanomaterials are available; for instance, nano-silica, nano-fiber, carbon nanotubes, graphene and its derivatives (Hou et al., 2017; Silvestre et al., 2016; Singh et al., 2013; Stephens et al., 2016). Graphene oxide (GO) possesses similar characteristics as graphene, because it is a derivative of graphene (Dong et al., 2016; Horszczaruk et al., 2015; Kuilla et al., 2010). Furthermore, the edges and basal planes of GO are rich in oxygenated functional groups, such as epoxides, carbonyls, hydroxyls and carboxyls. It has been shown that functional groups improve the dispersion of GO in water (Geim and Novoselov, 2007; Zhu et al., 2010). One of the most significant roles of oxygen functionalities on the surface of GO in cement is

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to offer growth points for cement hydration products (Lin et al., 2016).

Currently, investigations have been carried out to verify the effect of GO on cement composites' properties. GO may be involved in the hydration process of cement, improving the crystal phases and promoting the process of hydration, by accelerating the growth and nucleation of hydrated phases, resulting in an improvement in compressive and flexural strengths of cement paste (Wang et al., 2015). GO addition could greatly enhance the cement concrete durability by refining its pore structure. Also, test results showed that GO can modify cement microstructural properties to provide high freeze-thaw resistance (Mohammed et al., 2016).

GO significantly increases concrete's viscosity and decreases its fluidity, owing to large specific surface area (Horszczaruk et al., 2015). Despite much research on concrete with GO addition, research on improving the fluidity of GO-concrete with consistently achieving the required material properties remains limited. Fly ash is a commonly available supplementary cementitious material which can be utilized to enhance the fluidity of cement composites (Hou et al., 2013). In the current investigation, an attempt is made to enhance the workability of GO-concrete by adding mineral admixture of fly ash. The present study systematically investigated the synergetic role of fly ash and GO on the fluidity and mechanical characteristics of high-strength concrete. To understand the effect of fly ash and GO on mechanical characteristics, microstructural characterization techniques, such as SEM, EDX, FTIR and XRD, are also performed.

EXPERIMENTAL STUDY

Materials

The materials used in the present investigation were: Ordinary Portland Cement (OPC) of grade 53 according to IS:269-2015, where the physical properties of cement are given in Table 1; fly ash (FA) having a specific gravity of 2.10 and class F according to IS:3812 (Part 1)-2013, where the chemical composition of cement and fly ash is given in Table 2; fine aggregate (FA) and coarse aggregate (CA) according to IS:383-2016, where the properties of aggregates are given in Table 3; polycarboxylate (PC) superplasticizer conforming to IS:9103-2018.

Table 1. Cement physical properties

Physical properties	OPC
Fineness	3%
Normal consistency	35%
Specific gravity	3.12
Setting time (min)	
Initial	102
Final	195
Bulk density (kg/m^3)	1440
Compressive strength (MPa)	
7 days	47.9
28 days	54.8

Table 2. Chemical composition of cement and fly ash

Material	SiO_2	Al_2O_3	Fe_2O_3	CaO	SO_3	MgO	Na_2O	K_2O
Cement	20.265	4.31	3.315	64.38	3.46	1.355	0.1	0.91
Fly ash	61.97	29.63	3.29	2.57	0.13	0.94	0.33	0.83

Table 3. Properties of aggregates

Physical properties	Sand	CA
Specific gravity	2.68	2.75
Fineness modulus	2.82	6.95
Water absorption (%)	1.25	0.55
Unit weight, kg/m^3 (dry compacted)	1675	1752

Table 4. Technical specifications of GO

Average lateral dimension (X&Y)	Thickness (Z)	Number of layers	Purity	Bulk density	Surface area
5-10 µm	0.8-2 nm	1-3	99%	0.121 g/cm^3	110 - 250 m^2/g

The nanomaterial used in this study was industrial-grade graphene oxide, noting that the technical parameters of GO are given in Table 4. Dispersion of GO in water along with PC was carried out by ultrasonication process. A probe sonicator was operated for 60 min to achieve uniform dispersion. As a result, stable GO dispersion in the form of an aqueous solution at 6 mg/mL concentration was achieved. GO was characterised by SEM, EDX, FTIR and XRD techniques, as presented in Figs. 1-4. SEM image (Fig. 1) reveals that the GO surface has folded and wrinkled morphology. Elemental analysis (Fig. 2) demonstrates that the GO comprises of 73 wt% C, 26.5 wt% O and 0.44 wt% S. FTIR spectrum (Fig. 3) shows the absorption peaks indicating that the surface of GO consists of oxygen functional groups. XRD pattern (Fig. 4) indicates the appearance of a diffraction peak at 2θ=12.61° and a corresponding interlayer distance of 0.704 nm.

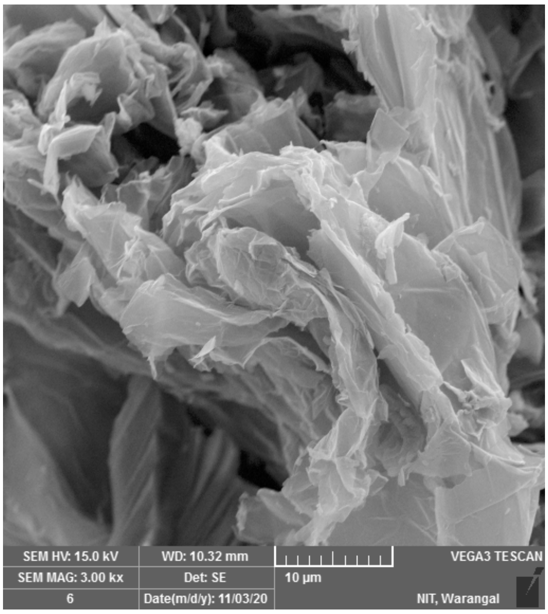


Figure (1): SEM image of GO

Preparation of Concrete Specimens

To investigate the influence of fly ash and GO on the properties of concrete, the GO as addition with a fixed dosage of 0.15% by weight of binder and fly ash as partial replacements of cement of 10%, 20% and 30% were considered in this study. The quantities of ingredients in the concrete mix per cubic meter are: cement 450kg, sand 788kg and coarse aggregate 1150kg with a water content of 135 kg. Concrete mix proportions are presented in Table 5. Concrete mix without fly ash and GO is considered as a control mix (GO-0, FA-0) for later comparison of results. GO-added concrete mix without fly ash replacement is referred to as GO-concrete (GO-0.15, FA-0) and concrete mixes with fly ash replacement and GO addition are referred to as FA-GO-concrete. All the ingredients of concrete mix along with GO dispersion were thoroughly mixed for 4 to 5 minutes in a mixer to achieve a uniform mix. The fresh concrete was poured into moulds of various shapes in accordance with IS: 10086-2008. The samples were removed after one day from the moulds and kept for curing in water.

Testing Methods

Fluidity

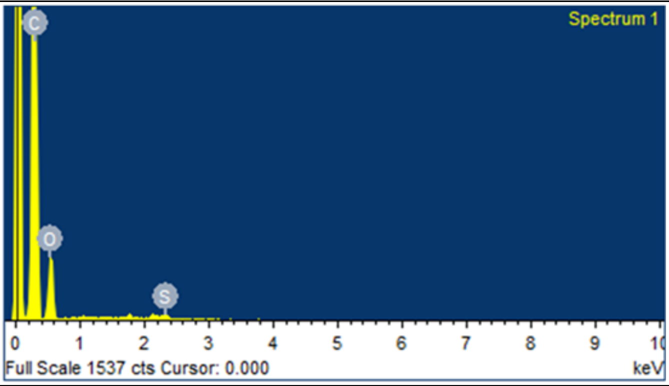
Slump test was conducted to verify the effect of GO and fly ash on the fluidity of FA-GO-concrete. The slump cone test was executed according to IS:1199-2018. The value of slump for various replacements of fly ash in FA-GO-concrete mixes was recorded and compared with those of GO-concrete and control concrete.

Strength Properties

To determine the strength characteristics, the sizes of the specimens used were: 100mm cubes, 100mm dia x 200mm height cylinders and 100x100x500mm prismatic beams for compressive, split tensile and flexural strength tests, respectively. Strength characteristics of the concrete were determined as per IS:516-2021. The specimens were tested at 7 and 28 days. 3 samples of each mix were considered for all tests and averages were determined.

Table 5. Mix proportions of concrete

Material	Unit	MIX ID				
		GO-0, FA-0	GO-0.15, FA-0	GO-0.15, FA-10	GO-0.15, FA-20	GO-0.15, FA-30
GO	kg	0	0.675	0.675	0.675	0.675
Fly ash	kg	0	0	45	90	135
Water	kg	135	135	135	135	135
OPC	kg	450	450	405	360	315
Sand	kg	788	788	788	788	788
CA	kg	1150	1150	1150	1150	1150
PC	(wt.%)	0.8	0.8	0.8	0.8	0.8



Element	Weight %	Atomic %
C K	73.02	78.42
O K	26.54	21.40
S K	0.44	0.18
Totals	100.00	

Figure (2): EDX of GO

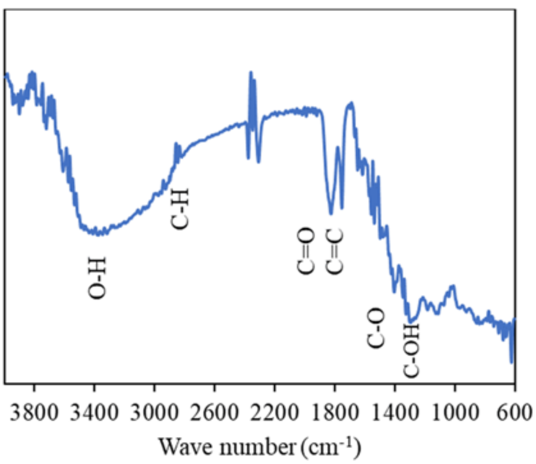


Figure (3): FTIR of GO

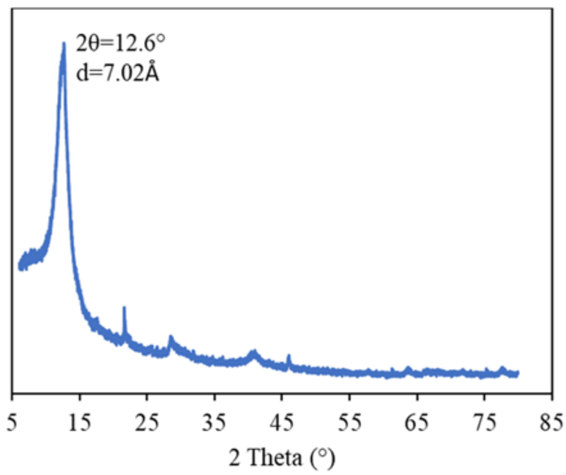


Figure (4): XRD of GO

Microstructure

Microstructure studies of SEM, EDX, FTIR and XRD were used to analyze the morphology, composition of elements and phase identification properties of FA-GO-concrete at 28 days of age to understand the effect of fly ash and GO on strength characteristics. Samples from the core parts of failure surfaces of the specimens were collected for SEM and EDX. Powder samples passing through the 90-micron sieve were taken for XRD and FTIR.

RESULTS AND DISCUSSION

Fluidity of FA-GO-Concrete

A comparison of the fluidity of different concrete mixes with fly ash replacement at 10%, 20% and 30% and a constant GO dosage of 0.15% is given in Fig. 5. The results demonstrate that the fluidity of GO-concrete reduced by 21.5% in comparison with that of control concrete. Similar results were reported on the cement paste, where GO addition resulted in a reduction of fluidity and an improvement in viscosity (Yang et al., 2017), which is due to that more amount of free water is required to wet the surface of GO due to its huge specific surface area. The formation of agglomeration and flocculation is caused due to electrostatic interaction between GO and cement particles. The hydrophilic oxygenated functions of GO absorb the water molecules and remain trapped (Shang et al., 2015). The fluidity of FA-GO-concrete improved efficiently and increased with an increase in the replacement of fly ash in comparison with control concrete. The FA-GO-concrete fluidity improvements with respect to control concrete were -12.3%, -4.6% and +6.2% with the fly ash replacements of 10%, 20% and 30%, respectively. This shows that the formation of flocculation structures in GO-concrete can be decreased by fly ash replacement in cement (Reddy and Ravi, 2022). Henceforth, fly ash counterbalances the GO influence on the decrease in fluidity. This might be ascribed to two reasons. First, because the particle shape of fly ash is a smooth spherical one with less water requirement, the concrete fluidity improves as the fly ash particles act as bouncing balls within the cement particles. Second, fly ash can fill the voids within the cement particles, owing to that the fineness of cement is greater than that of fly ash; so fly ash not only improves the gradation of size, but also the fluidity of concrete (Han et al., 2014).

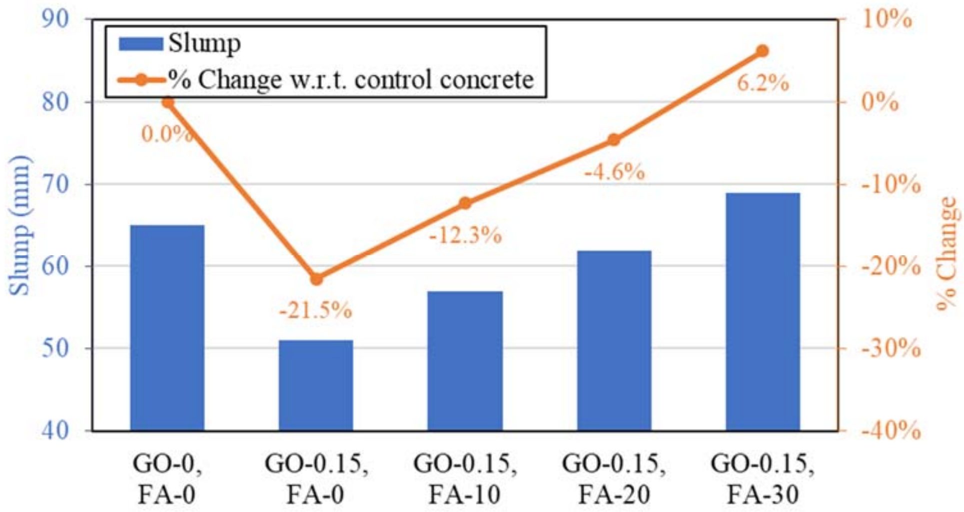


Figure (5): Slump values (mm) of various concrete mixes

Mechanical Properties of FA-GO-Concrete

Compressive Strength

A comparison of the compressive strengths of different concrete mixes is illustrated in Fig. 6. The results demonstrate that the compressive strength of GO-concrete is enhanced with respect to control concrete and the improvement is 53% and 26% at 7 and 28 days, respectively. The percentage growth in compressive strength of GO-concrete is more observed at the early stage. Several studies reported comparable results, where the growth in strength characteristics of GO-concrete is attributed to the influence of GO on the degree of hydration, nanofiller effect and formation of dense microstructure (Reddy and Prasad, 2022; Wang et al., 2015; Yang et al., 2017). FA-GO-concrete with GO content of 0.15% and 20% of fly ash replacement enhanced the compressive strength by 6% and 10% at 7 and 28 days, respectively, compared with control concrete.

Split Tensile Strength

Values of split tensile strength for various concrete mixes are illustrated in Fig. 7. The results reveal that the split tensile strength values of GO-concrete are enhanced with respect to control concrete and the improvements are 29% and 21% at 7 and 28 days, respectively. The early-age growth percentage in split tensile strength of GO-concrete was recorded more. FA-GO-concrete with 0.15% of GO and 20% of fly ash replacement enhanced the split tensile strength by 7% and 12% at 7 and 28 days, respectively, when compared to control concrete.

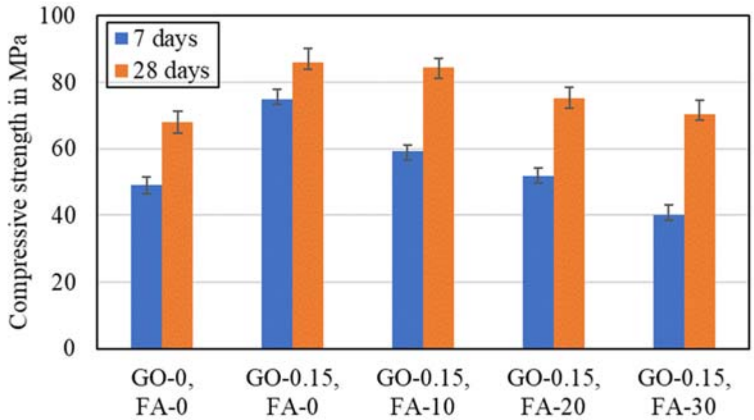


Figure (6): Variation of compressive strength for various concrete mixes

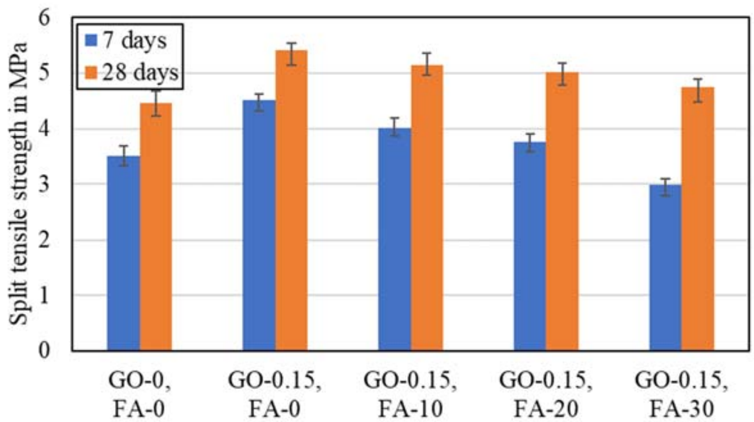


Figure (7): Variation of split tensile strength for various concrete mixes

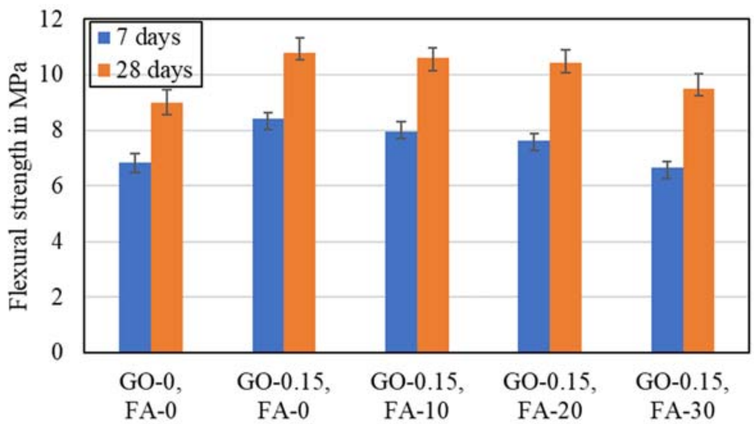


Figure (8): Variation of flexural strength for various concrete mixes

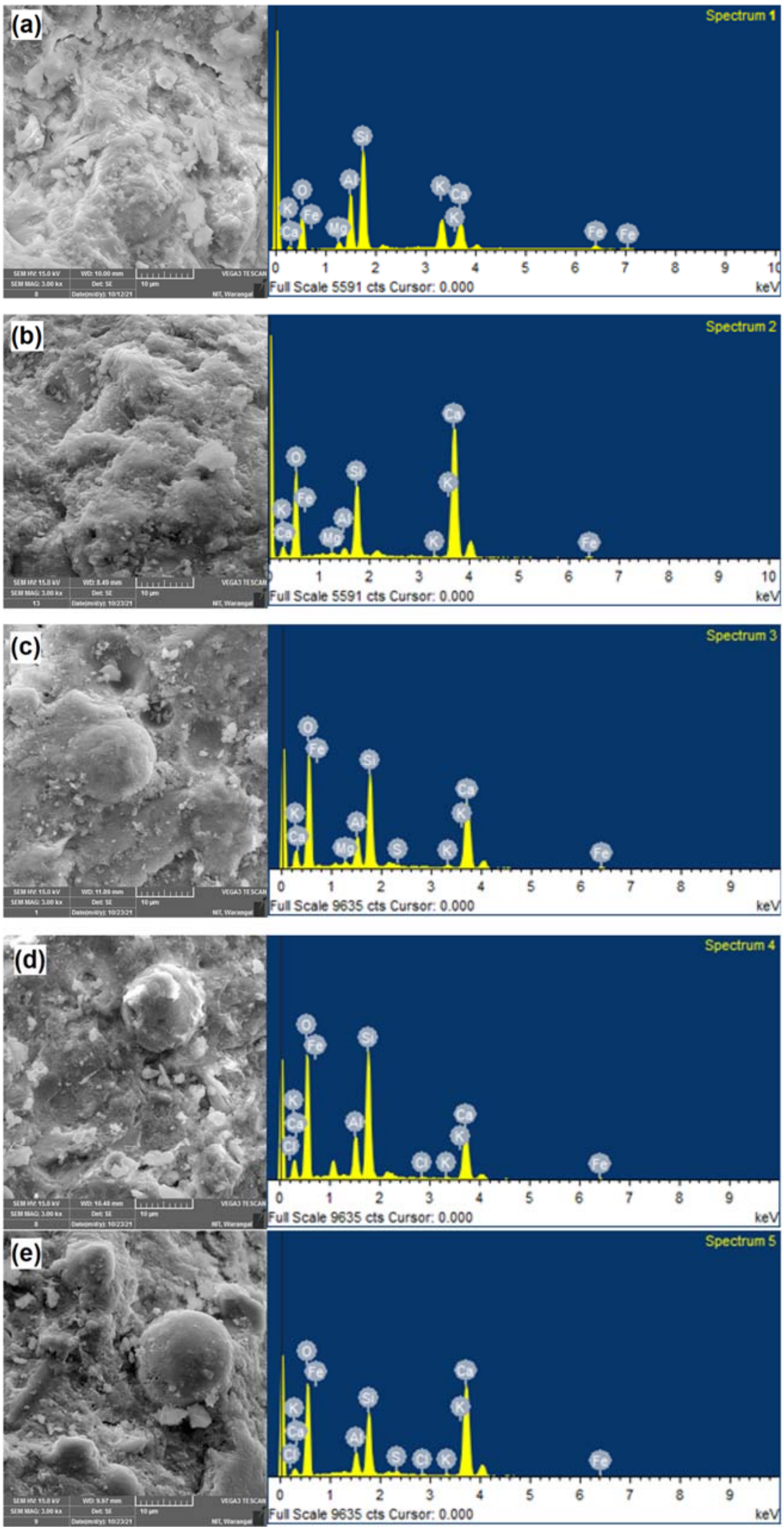


Figure (9): SEM and EDX of various concrete mixes a) GO-0, FA-0, b) GO-0.15, FA-0, c) GO-0.15, FA-10, d) GO-0.15, FA-20, e) GO-0.15, FA-30

Flexural Strength

Values of flexural strength for various concrete mixes are illustrated in Fig. 8. The results indicate that flexural strength of GO-concrete was enhanced with respect to control concrete and the enhancements were 24% and 20% at 7 and 28 days, respectively. The growth rate of flexural strength in GO-concrete is more observed at an early age. FA-GO-concrete with 0.15% of GO and 20% replacement of fly ash improved flexural strength by 12% and 16% at 7 and 28 days, respectively, compared to control concrete.

The growth rate of FA-GO-concrete is lesser at the early stages; however, the strength properties of the samples at 7 days with replacement of fly ash up to 20% are more compared to those of control concrete. The drawback of fly ash on delaying the early-age progress of strength was counterbalanced by GO (Reddy and Ravi, 2022). The influence of GO on the decrease in fluidity was counterbalanced by fly ash. The results illustrated that suitable amounts of fly ash and GO improve fluidity and increase the mechanical properties of the concrete.

The Microstructure of FA-GO-Concrete

SEM

The SEM images of the different concrete mixes with GO content of 0.15% and different fly ash percentages of 10%, 20% and 30% are illustrated in Fig. 9. The surface morphology of the control concrete sample is illustrated in Fig. 9(a). From the SEM image, it is observed that the development of CH, C-S-H, Afm and Aft with pores and micro-cracks leads to a loose and non-uniform structure. SEM image of GO-concrete is illustrated in Fig. 9(b), clearly indicating that for concrete with GO addition, the crystallinity of hydration phases is closely interwoven with one another having less micro-cracks and pores. The surface of GO-concrete shows the formation of a compact, uniform and densified structure at micro-level compared with control concrete, which is responsible for the development of mechanical properties. FA-GO-concrete SEM images are illustrated in Figs. 9(c-e), where the structure of hydrated products of cement with the combination of fly ash and GO was improved compared to control concrete. Unreacted smooth spherical-shape fly ash particles can also be noticed, which demonstrates that the hydration of fly ash is in progress, for the reason that the growth rate of FA-GO- concrete strength properties improved.

EDX

EDX analysis was carried out to examine the elemental composition of hydration products from the SEM images. Spectrum 1 to spectrum 5 are given in Fig. 9 and the composition of elements' percentages is given in Table 6. From the composition of elemental percentages, it is noticed that there was an increase in the percentages of elements of Ca, C and Si in comparison with the control concrete and also a substantial reduction in the elemental percentage of O was noticed. For control concrete, the Ca/Si elemental ratio was high and the addition of GO decreased the Ca/Si ratio. This shows that the process of hydration in the presence of GO leads to densified C-S-H formation and other hydration products interwoven with one another and improves the mechanical characteristics (Kunther et al., 2017).

Table 6. EDX analysis of different concrete mixes from Fig. 9

Spectrum	Weight percentage (wt.%)								
	O	Ca	Si	C	Al	S	K	Mg	Fe
1	56.95	23.92	4.44	4.31	3.88	2.23	1.55	1.29	1.43
2	47.39	27.61	8.79	10.61	1.75	0.64	0.88	0.73	1.6
3	45.28	29.63	8.58	10.14	2.14	0.55	0.68	0.57	2.43
4	56.12	24.29	5.64	6.35	1.82	0.68	1.46	1.22	2.42
5	58.54	23.89	5.24	5.16	2.73	1.05	1.27	1.06	1.06

XRD

The XRD patterns of different concrete mixes are illustrated in Fig. 10. The patterns demonstrate no major difference in the positions of diffraction peaks (2θ = 18.12°, 20.83°, 34.07°, 39.73°, 42.50°, 50.76°, 54.86°, 60.25°) among the various concrete mixes, indicating that combination of fly ash and GO shows the identical crystals of hydration phases of CH, Aft and Afm.

Moreover, the addition of GO and replacement of fly ash enhance the peaks' intensity compared to control concrete, showing the increase in crystallinity of hydration phases. Additionally, the reaction of cement and the oxygen functionalities of GO will help increase the crystalline phase formation, where the new crystalline phase formation contributed to enhancement of mechanical characteristics of FA-GO-concrete.

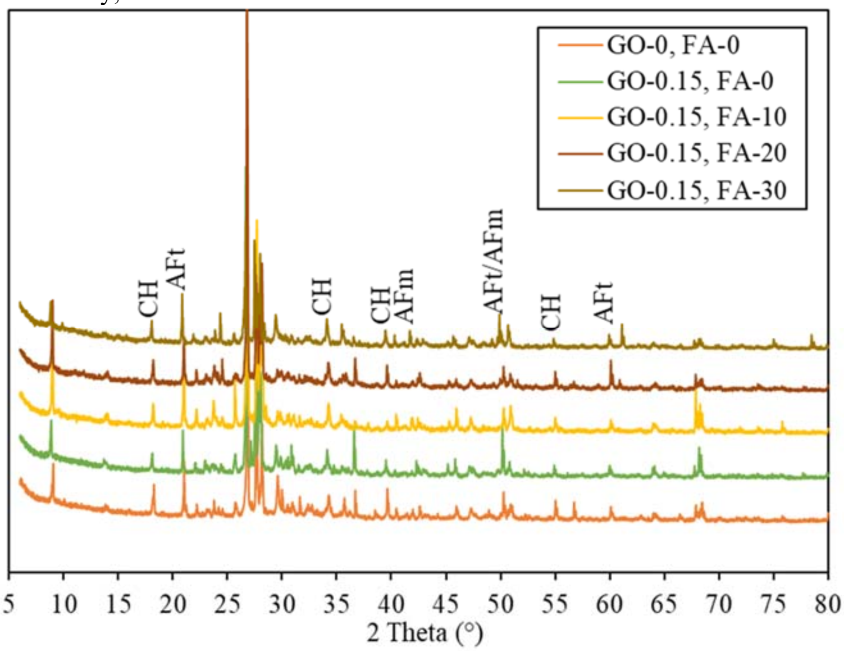


Figure (10): XRD patterns of various concrete mixes

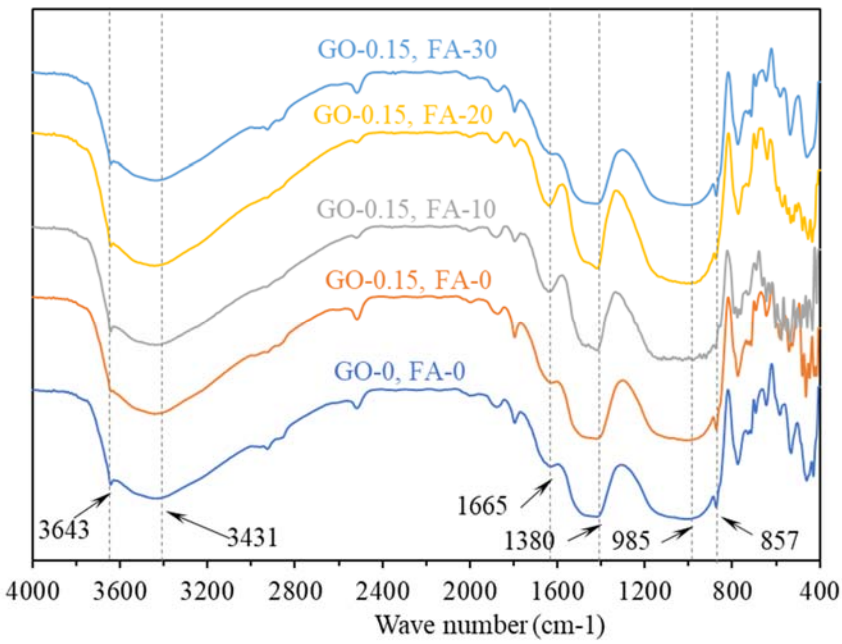


Figure (11): FTIR spectra of various concrete mixes

FTIR

The FTIR spectra for various mixes of concrete with GO and different replacements of fly ash are shown in Fig. 11. The presence of calcium silicate hydrate in the band region of 950-1000 cm^{-1} can be observed and GO addition increased the peaks' intensity. The absorption peak of calcium hydroxide can be noticed at 3643 cm^{-1}. Mono-sulfate aluminate peaks are observed at 1380 and 1665 cm^{-1} and peaks of strong trisulfate aluminate are observed at 857, 1640-1680 and 3431 cm^{-1} (Horgnies et al., 2013). Because hydrated products are identical in all concrete mixes, the FTIR spectra are similar, except for differences in peaks between them. Additionally, when GO is added, the absorption peak of calcium silicate hydrate shifts to a higher wavenumber than that of the control concrete. This demonstrates that the addition of GO changes the Ca/Si ratio and leads to the formation of densified calcium silicate hydrate gels. Because of this, the strength characteristics of FA-GO-concrete increased.

CONCLUSIONS

The synergetic effects of addition of GO and replacement of fly ash on the fluidity, mechanical and microstructure properties of high-strength concrete were studied. The results demonstrated that:

- The fluidity of GO-concrete improved with fly ash as a partial replacement; it enhances with an increase in the replacement of fly ash. The improvement in fluidity properties of FA-GO-concrete could be attributed to the size gradation, ball effect and less water demand.
- The strength characteristics of FA-GO-concrete have higher values at 7 and 28 days up to 20% replacement of fly ash. The growth of strength characteristics of FA-GO-concrete is attributed to the GO influence on the hydration degree, formation of dense microstructure and effect of nanofiller. The results demonstrate that replacement of fly ash in GO-concrete is an economical and effective technique to achieve the desired characteristics.
- Microstructural results of concrete mixes using SEM, EDX, XRD and FTIR show that GO addition in concrete affects crystalline phases and surface morphology. This is due to the GO consisting of oxygenated functionalities having a huge specific surface area, which improved the degree of hydration, the nucleation of hydration phases and the formation of more uniform and densified structure at the micro-level.

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