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Design and Optimization of Mechanical Properties of Reduced-Graphene Oxide- Loaded Cement

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ABSTRACT

Graphene oxide (GO) is a graphite-based product. GO prepare a new distance to interact with cement matrix. GO due to the high specific surface area, high intrinsic mobility and high Young's modulus leads to a remarkable enhancement in mechanical properties of cementitious material matrix. In the present study, the effect of reduced-graphene oxide (r-GO) on the mechanical properties of Portland cement paste was investigated. Response surface methodology based on central composite design (CCD) was used to predict the interaction effects of curing time (7-21 days) and GO amount (0.02-0.1%) on the compressive strength and flexural strength of the r-GO–cement composite. By optimization of parameters, the compressive strength and flexural strength was increased by 48% and 74% compared with cement without r-GO. The obtained results demonstrated that the r-GO is a promising filler of cement-based composites to enhance the mechanical properties of cement.

Key words: Reduced-graphene oxide, Cement, CCD, Compressive strength, Flexural strength.

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1. INTRODUCTION

Ordinary Portland cement (OPC) is extensively used for building and construction (1). However, the use of OPC due to the poor tensile strength and low strain capacity is limited. Incorporation of fibers into the OPC led to improve the resistance of the OPC structure (2). The mechanical properties of concrete have been reinforced by coconut fiber (3). Furthermore, Nano fillers have been introduced to decrease the cracks of cementitious materials. The flexural and compressive strengths of the cement mortars can be increased by incorporating nano-Fe_2O_3 and nano-SiO_2 particles into cement mortars (4-6). Carbon nanotubes (CNTs) are one-dimensional carbon nanomaterial with hollow tubular structure. CNTs are formed either by single wall (SWCNTs) or multi walls (MWCNTs) of rolled carbon sheets (7, 8). The effect of carbon nanotubes (CNTs) on the compressive strength of cement paste has been investigated. Chen et al. (9) summarized the impact of CNTs on cement paste. However, the main problems of application of CNTs in cementitious materials are a weak bounding between the CNT, s and the matrix of cements is established and the dispersion of the CNT,s in the mixture is very poor, and this incorrect distribution led to decrease the mechanical properties. Also due to the lack of appropriate boundaries between materials, the reduction of concrete performance is also observed (10-12). Although, Collins et al. (13) evaluated the dispersion of CNTs and consistency of fresh CNTs–OPC mixtures by using various chemical admixtures such as air entraining agent, styrene butadiene rubber, calcium naphthalene sulfonate, naphthalene sulfonic acid derivative, lignosulfonate and aliphatic propylene glycol ether including ethoxylated alkyl phenol to improve the dispersion of CNTs in cement matrix; however, the use of them due to the high cost of additives is limited. Graphene oxide (GO) is monolayer of sp^2-hybridized carbon atoms derivatized by a mixture of carboxyl, hydroxyl and epoxy functionalities (14). The oxygen functional groups of GO sheets significantly facilitate the interactions between the GO sheets and cement. Furthermore, the improved mechanical properties of these composites were attributed to the high specific surface area and excellent mechanical properties of GO sheets (15-17). Pan et al. (18) found that the addition of

0.05 wt% GO into the cement can increase the GO–cement composite compressive strength by 15–33% and the flexural strength by 41–59%, respectively. Saafi (19) et al. has been investigated the influence of GO on the cement. The results indicated that the incorporation of 0.35% GO into the cement led to increase in the flexural strength and Young's modulus by 134% and 376%, respectively. However, there is a little study on the interaction effects of GO concentration and curing time and optimization of parameters on the mechanical properties of GO-loaded cement. In recent researches, the factor space Central-Composite Design (CCD) and Box-Behnken Design (BBD) are commonly selected experimental design techniques (20). In the present study, the reduced-GO was added to the cement. The mechanical behavior of the new composite materials was studied by evaluation of compressive strength and flexural strength. CCD was used to predict the simultaneous effects of curing time and GO content on the mechanical properties of cement.

2. EXPERIMENTAL

2.1. Materials

The Portland cement type I (42.5 R) of cement manufacturing company (Darab, Iran) was used in this Study. The chemical properties of the cement are presented in Table 1.

Table 1. Chemical and mechanical properties of used Portland cement

SiO_2 (%)	Al_2O_3 (%)	Fe_2O_3 (%)	CaO (%)	MgO (%)	SO_3 (%)	Cl (%)	InR (%)	L.O.I (%)	Alkali (%)	Compressive (MPa)	Flexural (MPa)
20.40	5.25	3.95	65.5	1.2	1.05	0.03	0.62	1.40	0.60	28.6±0.5	4.3±0.2

2.2. Synthesis of r-GO and incorporation of r-GO into the cement

In a normal procedure, the compounds were formulated according to the report. to 50 mL concentrated sulfuric acid (98%, Merck) 1 g of graphite flakes (99%, Alfa Aesar) was added while shaking in an ice-water won. 3 g potassium permanganate (>99%, Sigma Aldrich) was inchmeal added by retaining the temperature under 10 °C. Then, the pendency was stirred at Normal temperature for 25 min followed by 5 minute sonication in an ultrasonic won. The stirring-sonication process repeating for 12 times, and the reflex was quenched by the addition of 200 mL distilled water. An additional 2h ultrasonic therapy was carried out before distributing the suspension into two equal parts; one washed to obtain GO (described later) and the other was further processed for preparation of R-GO. 1M sodium hydroxide (>98%, Sigma Aldrich) added to adjusting the pH at ~6, then suspension was further sonicated for 1 h. In the 100 mL distilled water 10 g L-ascorbic acid (99%, Sigma Aldrich) was dissolved and then was stilly added to the exfoliated graphite oxide suspension at normal temperature. The reduction was performed at 95 °C for 1 h. cellulose filter paper filtered simply. The yield black precipitates and further were washed with a 1M hydrochloric acid solution (37%, Merck) and distilled water to neutral pH. The filtrate finally freeze-dried to obtain R-GO powder. Different concentrations of GO (0. 2, 0. 6 and 1% by weight of cement) were added into the cement. The predetermined amounts of GO were added into the flasks containing 150 mL of water. Then the prepared suspensions were sonicated for 20 min to obtain the homogenous solutions. The prepared suspensions were added into the cement solutions (the water to mixture ratio used was 4 v/w).

2.3. Mechanical property tests

The flexural strength was measured following the procedure prescribed by ASTM C78/C78 M-10. Flexural strength tests were conducted on 15 mm ×15 mm ×80 mm prisms. To achieve the maximum load for any specimen within the first 50–90 s, the displacement control rate was 0.1 mm/min. The compressive strength was measured following the procedure prescribed by ASTM C109/C109 M-11b. Compressive strength tests were conducted on 15 mm × 15 mm × 15 mm cubes.

2.4. Design of experiments

The Central Composite design (CCD) was used to analyze the simultaneous effects of curing time, and GO content on the flexural strength and compressive strength of cementitious materials. The polynomial models for the obtained responses with respect to the parameters were expressed as follows:

$$Y = \beta_0 + \sum_{i=1}^{2} \beta_i x_i^2 + \sum_{i=1}^{2} \beta_{ii} x_i^2 + \sum_{i=1}^{2} \sum_{j=1}^{2} \beta_{ij} x_i x_j \quad (1)$$

Where Y is the predict response by the model (flexural strength and compressive strength of cementitious materials) and β_0, β_i, β_{ii}, β_{ij} are the constant regression coefficients of the model. X_i, X_{ii} and X_{ij} represent the linear, quadratic and interactive terms of the un coded independent variables, respectively. The factor of designation (R2) was handled to aims the accuracy of the full quadratic equation. The experimental design and results are presented in Table 2.

Table 2. The experimental design and responses

Predicted value (MPa)	flexural strength (MPa)	Predicted value (MPa)	compressive strength (MPa)	Curing time (day)	r-GO content (%)	Run Order
4.77222	4.80	41.2253	41.25	7	0.2	1
7.64222	7.63	60.4203	60.32	7	1.0	2
5.28889	5.29	47.4303	47.52	21	0.2	3
8.71889	8.68	69.6953	69.66	21	1.0	4
5.08889	5.06	44.7544	44.64	14	0.2	5
8.23889	8.29	65.4844	65.62	14	1.0	6
5.91556	5.90	50.2244	50.30	7	0.6	7
6.71222	6.75	57.9644	57.91	21	0.6	8
6.37222	6.35	54.5211	54.50	14	0.6	9

3. RESULTS AND DISCUSSION

3.1. Statistical model for compressive strength and flexural strength

Analysis of variance (ANOVA) is a statistical technique that subdivides the total variation in a set of data into component parts associated with specific sources of variation for the purpose of testing hypotheses on the parameters of the model. The P- value lower than 0.05, indicates the significant of term in surface response analysis. ANOVA results obtained from Table 3 indicated that the linear, square and interaction variables have a significant effect on the both responses. Finally, the polynomial equations of compressive strength and flexural strength are obtained as follows:

$$Y_1(MPa) = 54.521 + 10.365x_1 + 3.870x_2 + 0.598x_1^2 - 0.427x_2^2 + 0.767x_1x_2 \quad (1)$$

$$Y_2(MPa) = 6.372 + 1.575x_1 + 0.398x_2 + 0.292x_1^2 - 0.058x_1^2 + 0.140x_1x_2 \quad (2)$$

Where Y_1 and Y_2 are the compressive strength and flexural strength; x_1 and x_2 are the r-GO content (%) and curing time (day), respectively. As shown in Table 1, the experimental values of compressive strength and flexural strength were in close agreement with the predicted values of model.

Table 3. ANOVA results for the compressive strength at different levels

Source	DF	Seq SS	Adj SS	Adj MS	F	P
Regression	5	737.897	737.897	147.579	7311.61	0.000
Linear	2	734.461	734.461	367.230	18193.90	0.000
r-GO content (%)	1	644.599	644.599	644.599	31935.74	0.000
Curing time (day)	1	89.861	89.861	89.861	4452.05	0.000
Square	2	1.080	1.080	0.540	26.76	0.012
r-GO content (%)*r-GO content (%)	1	0.716	0.716	0.716	35.47	0.009
Curing time (day)*Curing time (day)	1	0.364	0.364	0.364	18.04	0.024
Interaction	1	2.356	2.356	2.356	116.74	0.002
r-GO content (%)*Curing time (day)	1	2.356	2.356	2.356	116.74	0.002
Residual Error	3	0.061	0.061	0.020		
Total	8	737.958				

DF: degree of freedom; Seq SS: sequential sum of square; Adj MS: adjusted mean of square.

3.2. Validation of the experimental and predicted model data

The probability distribution plot of residuals (difference between the model predicted compressive strength and flexural strength values and those derived experimentally) is presented in Figure 1. As shown, the errors were normally distributed. Furthermore, it was observed that the established model was sufficient to estimate the compressive strength and flexural strength values, as all the residuals were smaller than 5 %.

3.3. Surface plots

Based on ANOVA results, the interaction effect of GO content and curing time on the both compressive strength and flexural strength values was significant. For this, the simultaneous relation of GO content and curing time are illustrated in Figure 2. As shown, increase in GO content and curing time led to increase in the compressive strength and flexural strength values of cementitious materials. Furthermore, the effect of GO content on the compressive and flexural strength enhancement was more than curing time.

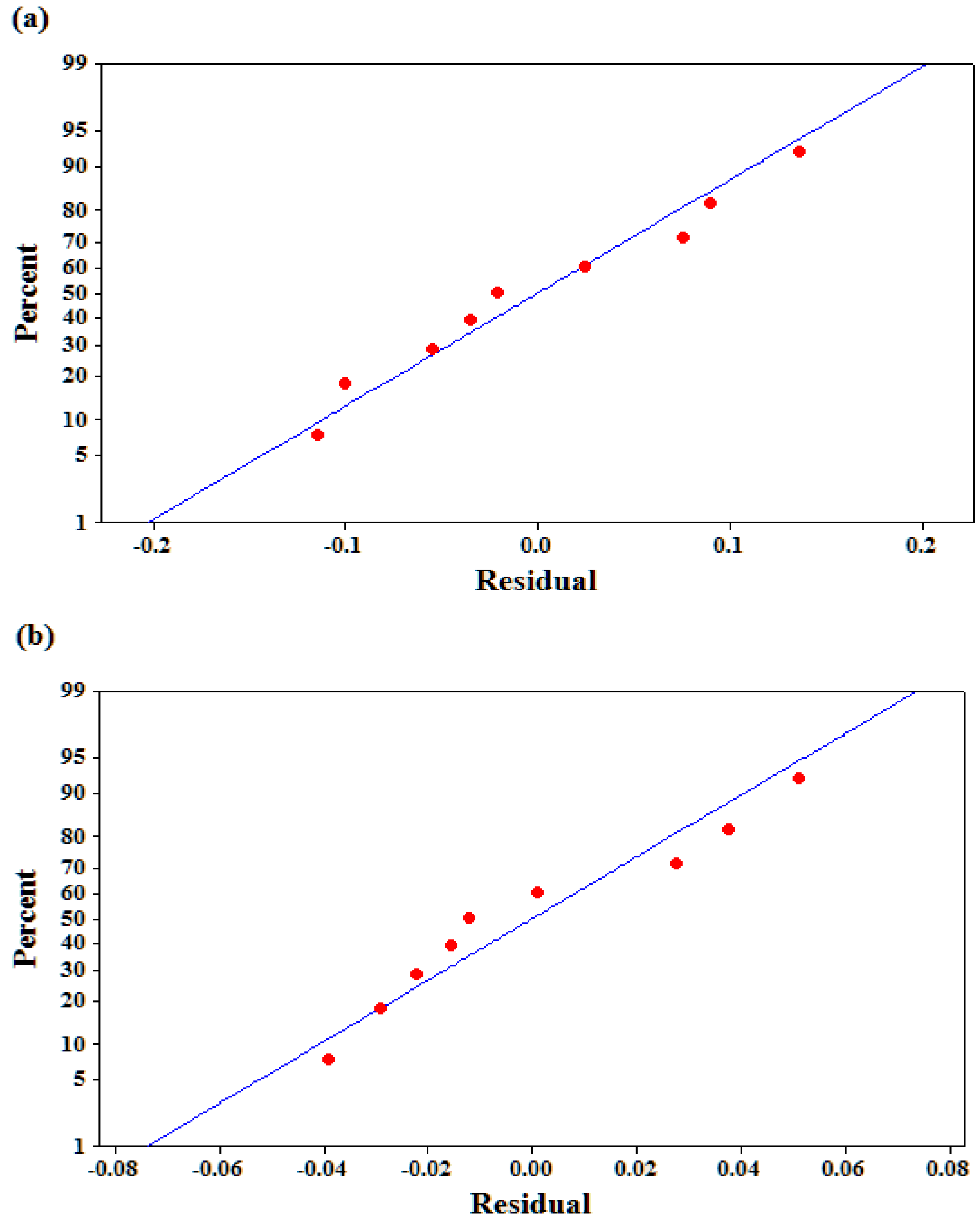
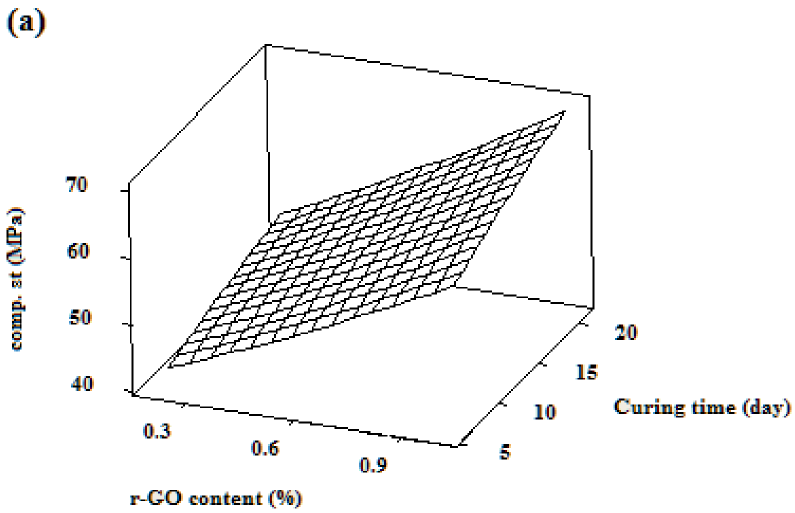


Figure 1. Normal probability plots for (a) compressive strength and (b) flexural strength data



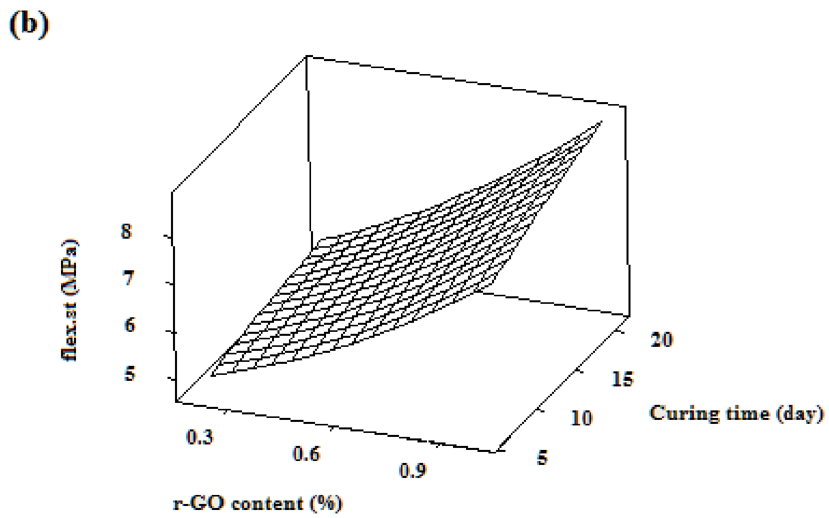


Figure 2. Surface plots of the (a) compressive strength and (b) flexural strength data versus r-GO content and curing time

3.4. Optimization of compressive strength and flexural strength

By solving the statistical models and optimization of variables at the time, the optimal un coded values of GO concentration (X1) and curing time (X2) were estimated to be 1 % and 21 days, for both compressive strength and flexural strength, respectively. The optimum predicted values for compressive strength and flexural strength by the model were estimated to 69.70MPa, and 8.72MPa, respectively. The experimental values for compressive strength and flexural strength in optimum conditions were found to be 69.66MPa, and 8.68MPa.These values were in good agreement with the estimated values by the model in optimum conditions.

4. CONCLUSION

The cementitious materials treated with r-GO were successfully developed to improve their mechanical properties. Central Composite design (BBD) was used to determine the optimal conditions for compressive strength and flexural strength. The analysis of CCD response confirmed that linear, square and interaction terms were found statically significant on the strength and flexural strength values. Surface plots indicated that the effect of GO content on the compressive and flexural strength enhancement was more than curing time. By optimization of parameters including curing time of 21 days, and GO concentration of 1%, the simultaneous maximum values for compressive and flexural strength enhancement were found to be 69.66 MPa, and 8.68 MPa, respectively.

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AUTHORS CONTRIBUTION

This work was carried out in collaboration among all authors.

CONFLICT OF INTEREST

The author (s) declared no potential conflicts of interests with respect to the authorship and/or publication of this paper.

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