RSM Analysis for Optimum Content of Graphene Nanoplatelets for 3D-Printed Clay Strength

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Abstract

This study applies Response Surface Methodologies (RSM) methods to maximize 3D-Printed clay mechanical properties. Mixes containing different Graphene Nanoplatelets (GNPs) contents were printed and tested in compression and flexure. The Central Composite Design method was used by coding the mixes fabrication method, i.e. moulding and printing, and GNPs content as variables. The analysis showed that the mixes containing low GNPs content of 0.1 wt.% attained higher compressive and flexural strengths than those containing a higher content of 0.2 and 0.3 wt.%. The results also highlighted that GNPs' efficiency was better observed in the printed samples other than the moulded ones, indicating that the printing process contributed to a better and uniform dispersion of GNPs in the clay matrix. RSM analysis confirmed that the maximum flexural strength response could be obtained using a GNPs content of 0.1 wt.%. Furthermore, the desirability analysis showed that a maximum predicted flexural and compressive strength improvements of 21% and 36 % compared to the control mixes could be obtained, respectively. In summary, this study proposed the importance of using Nanofilaments in 3D printing activities to achieve the desired elements' mechanical properties.

Keywords: 3D-printed clay; Graphene Nanoplatelets; Mechanical strength; RSM

1 Introduction

Three-dimensional printing (3DP), also known as rapid prototyping, is an additive manufacturing (AM) technique in which a variety of structures and complicated geometries are fabricated using computer-generated model data (Ngo et al., 2018). The printing process comprises printing a material layer-by-layer on top of each other. Over the years, this technology gained wide attention and tremendous development in different industrial sectors such as construction, medicine, and biomechanical. Despite the numerous advantages of 3DP, e.g. design freedom, time efficiency, labour safety, and environmental hazard reduction, imparting this technology to the construction industry is still relatively limited and very slow. However, the limited selection of printable materials is the key challenge. Currently, conventional ceramics engineering, involving clay and concrete sectors, is the largest materials industry in the world in terms of materials produced (Revelo & Colorado, 2018).

Clay is one of the oldest human-made materials, still widely utilized in construction and infrastructure today (Moropoulou et al., 2005). Building with clay has many features in the 21st century since it has superior thermal mass characteristics to any other material, acting as a buffer to exterior temperature fluctuations by delaying the release of immersed solar energy, resulting in a stable interior (Rael & San, 2017). Furthermore, it is easily available, affordable, fire-resistant and arguably the most earth-friendly material. These advantages are the main reasons the ceramic industry is of great interest to additive manufacturing technologies that use clay as raw materials. Although several additive processing techniques for ceramic components have been studied, research on ceramic and clay 3D printing for construction applications is still in its early stages (Wolf et al., 2018). To date, few studies in this area dealt with the performance of 3D-printed clays under different conditions and printing parameters, while research on the impact of various additives such as Nanomaterials was absent (Manikandan et al., 2020; Sangiorgio et al., 2022). A major challenge hindering the expansion of the cementitious materials 3D-printing industry is the reduced mechanical properties due to the layering process.

Nano-inclusions such as Graphene Nanoplatelets (GNPs) have gained interest in building technologies due to their exceptional properties at the nanoscale (Balaguru & Chong, 2006; Shen et al., 2013). Past work in this field showed that small additions of GNPs could effectively enhance cementitious composites' rheological, mechanical, and microstructural characteristics (Wang et al., 2016; Baomin & Shuang, 2019; Tao et al., 2019). However, using this technology to advance the production of 3D-printed clay elements is still missing. This research used the RSM methods to study the optimum GNPs content needed to maximize 3D-printed clay compressive and flexural strengths.

2 Research Methodology

Three different GNPs-to-clay dosages of 0.1, 0.2 and 0.3 wt.% were employed to evaluate the moulded and printed clay mechanical properties. Study samples were divided into two main groups: printed and moulded, with and without Nano-inclusions. Table 1 shows the tested batches. The experimental methodology consisted of samples preparation, first. Then, sample testing for flexure and compression. After that, test results were analysed using RSM techniques and reported accordingly.

Table 1: Test Batches

Test Group	Production Technique	Batch #	Batch Code	GNPs/clay Wt.%
Moulded Group	Moulding	1	MC	0.0
		2	M-0.1%GNPs	0.1
		3	M-0.2%GNPs	0.2
		4	M-0.3%GNPs	0.3
Printed Group	3D printing	5	PC	0.0
		6	P-0.1%GNPS	0.1
		7	P-0.2%GNPS	0.2
		8	P-0.3%GNPS	0.3

2.1 Materials and Equipment

The clay used was a high-quality Italian clay paste with a solid appearance. The GNPs used were industrial-grade 4 COOH Graphene Nanoplatelets of 4 nm thickness and 700 m^2/g surface area. The equipment included a clay 3D printer, an ultrasonic wave mixer, an electric ceramic kiln, a strength testing machine, and a scanning electron microscope (SEM).

2.2 Experimental Procedure and Testing

First, an aqueous solution of GNPs and water was prepared using an ultrasonic wave mixer for 30 minutes at a power of 400W to achieve a good dispersion of GNPs inside the clay matrix. The hard clay was cut into small pieces, and the aqueous solution was added gradually. After that, the clay was kneaded manually until a homogeneous paste free of cracks and not sticking to the hand was obtained; hence, the clay was ready to print. The same type of clay was used for all batches with a fixed water-to-clay ratio of 0.05. Printing of clay samples was performed using a Delta WASP 2040 3D printer along with an LDM WASP Extruder having a nozzle 3 mm in diameter. The printed specimens were sketched using the computer-aided design software Onshape, and a 3D printing slicing software simplifying 3D was utilized to generate g-code print paths. The compression and flexure sample configuration and the layered printing are presented in Figures 1a and 1b, respectively. After preparing the clay mixture, it was directly loaded into the 3D-printing system storage container. Then, a pressure of 6 bar was used on the storage container piston and kept constant during printing. Figure 2a shows the 3D-printing process for a cube sample. The best results were obtained using a printing speed of 90 mm/s and 300% flow. The same mix of proportions of each printed batch was used to make the same size moulded specimens using steel moulds. Finally, high-temperature heating was employed to turn raw clay into ceramic inside an electric ceramic kiln, as illustrated in Figure 2b. After the burning process, flexural and compressive strength tests were performed according to ASTM C348 and ASTM C109 standards.

(a) (b)

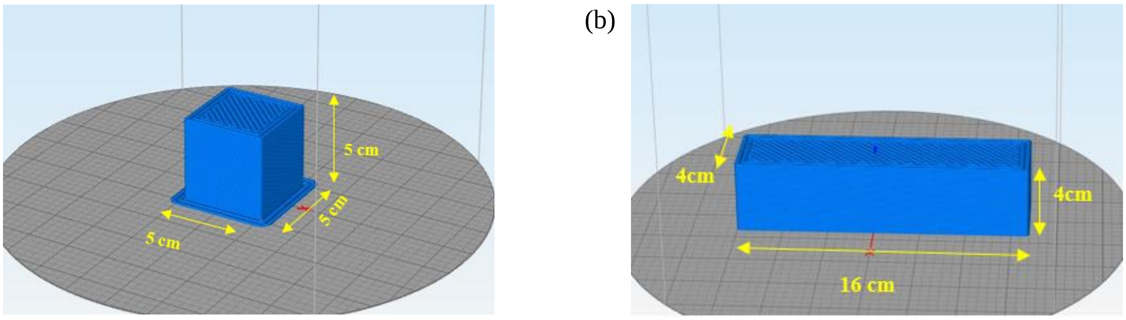

Fig. 1: (a) Details of ready-to-print 3D models of a) cube for compressive strength and b) prism for flexural strength

(a) (b)

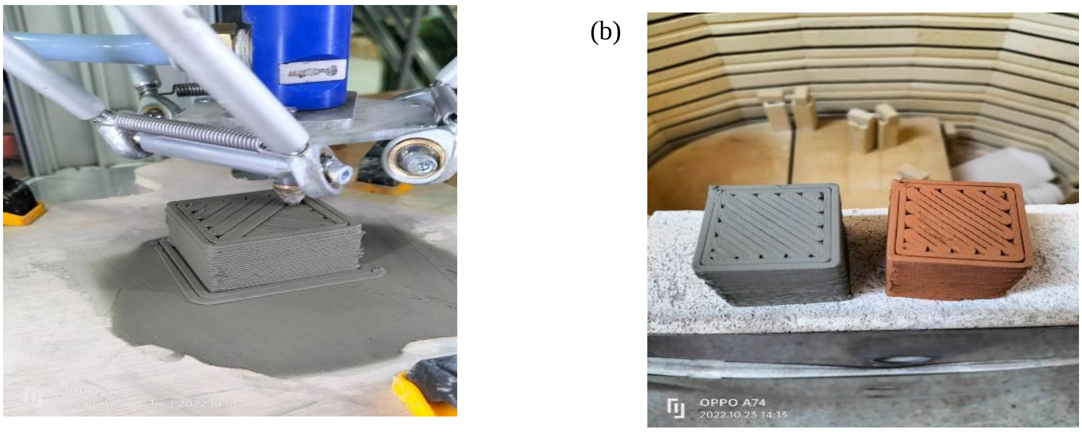

Fig. 2: (a) 3D-printing process, and (b) burnt vs unburnt sample

2.3 Response Surface Methods (RSM) Techniques

Response Surface Methodologies (RSM) techniques were used to study the effect of the fabrication method and GNPs weight fraction variables on the 3D-printed clay flexural and compressive strength factors' response. The following procedure was used to conduct this analysis:

1) Production type and Nanofilaments' weight fractions variables are coded (Table 2).
2) Second-order equations were employed to predict the flexural and the compressive strength functions as:

$$FS = b_0 + b_1x_1 + b_2x_2 + b_{11}{x_1}^2 + b_{22}x^2 + b_{12}x_1x_2$$

$$CS = a_0 + a_1x_1 + a_2x_2 + a_{11}{x_1}^2 + a_{22}x^2 + a_{12}x_1x_2$$

Where, FS is flexural strength, (MPa), CS is compressive strength, (MPa), $x1$ is samples production technique, $x2$ is GNPs' weight fraction, (%), and $a0$, $a1$, $a2$, $a11$, $a22$, $a12$, $b0$, $b1$, $b2$, $b11$, $b22$, and $b12$ are the response surface coefficients.

3) The prediction models and coefficients were measured.
4) The contour lines and desirability functions were plotted by optimizing the desirability function.

Table 2: Coding of production technique and GNPs weight fraction variables for RSM analysis.

Batch Code	Production Technique	Coding Production Technique	GNPs/clay Wt.%	Coding GNPs/clay Wt.%
MC	Moulding	-1	0.0	-1
M-0.1%GNPs	Moulding	-1	0.1	-0.333
M-0.2%GNPs	Moulding	-1	0.2	0.333
M-0.3%GNPs	Moulding	-1	0.3	1
PC	3D-printing	1	0.0	-1
P-0.1%GNPS	3D-printing	1	0.1	-0.333
P-0.2%GNPS	3D-printing	1	0.2	0.333
P-0.3%GNPS	3D-printing	1	0.3	1

3 Results & Discussion

3.1 Flexural and Compressive Strength

Figures 1a and 1b show the flexural and compressive strength results of all tested batches. Among all batches, the moulded mix containing 0.1 wt.% GNPs achieved the highest flexural strength of 18 MPa with an enhancement of about 7.5% compared to the moulded plain control mix. This finding indicates the role of proper Nanofilaments' dispersion on the strength enhancement of clay. The mix containing lower GNPs' content of 0.1 wt.% would probably have better dispersion quality than those containing higher GNPs' content of 0.2 and 0.3 wt.%. On the other hand, GNPs' efficiency was better observed in the printed samples using both low and high concentrations, as the printing process may contribute to a better and uniform dispersion of GNPs in the clay matrix. Compressive strength results revealed that both fabrication methods resulted in approximately equivalent compressive strength regardless of the weight fraction used. The maximum compressive strength improvement of about 42-48 % was obtained using a GNPs' weight fraction of 0.1%. The bridging and pore-filling impact of GNPs at low concentrations maybe responsible for this improvement pattern.

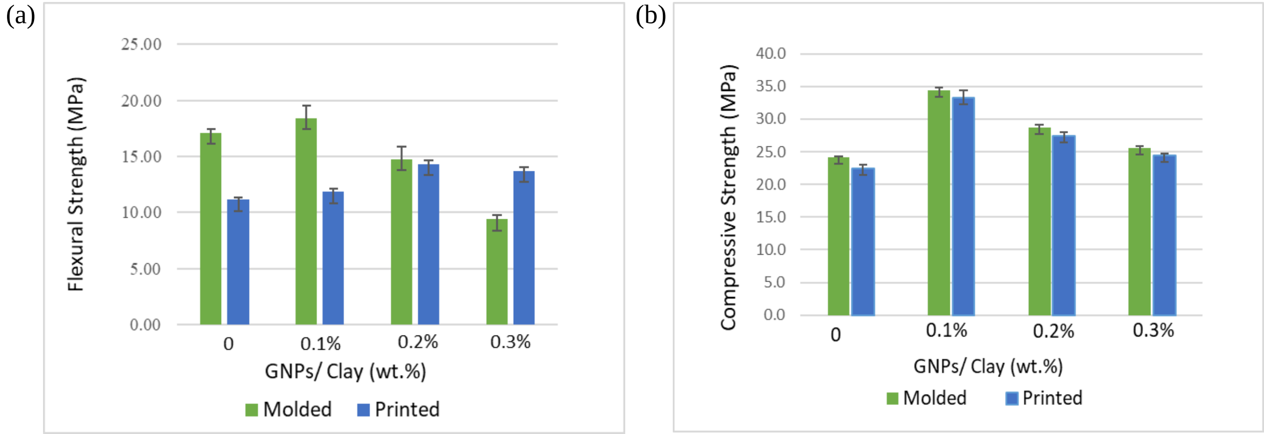

Fig. 3: (a) Flexural strength test results, and (b) Compressive strength test results

3.2 RSM Analysis

Figure 4 shows flexural and compressive strength factors' surface response against the production type and GNPs' weight fraction variables. The figure also shows the quadratic prediction formulas obtained using the RSM analysis techniques. The flexural strength response (Figure 4a) shows peaks at the negative coding boundaries indicating the highest flexural strength response at the moulded production type with 0.1 wt.% GNPs content. Alternatively, the compressive strength model (Figure 4b) shows the peaks at the mid boundaries indicating the success of both 0.1 and 0.2 wt.% GNPs at attain maximum compressive strengths peaks regardless of the production type. Figure 5a shows the production type and GNPs' weight fraction variables combined contour profiles versus the flexural and compressive strength. The coloured areas shown include all strengths falling below the plain clay flexural and compressive strengths. This analysis illustrates the significance of having a GNPs' weight fraction not exceeding 0.2625 wt.% (corresponds to a code of 0.75 on the chart) in case an improvement in both flexural and compressive strengths of printed clay is required.

Figure 5b shows the maximized Desirability Strength Function's behaviour of both production type and GNPs' weight fraction variables. The results showed that maximum flexural and compressive strength behaviours of printed elements could occur at a GNPs' weight fraction of 0.1125 wt.% (corresponds to a code of -0.25 on the chart). The maximum predicted flexural and compressive strengths are 13.45 and 30.70 MPa, respectively. Compared to the plain clay batch, these values represent 21% and 36% improvement in flexural and compressive strengths, respectively.

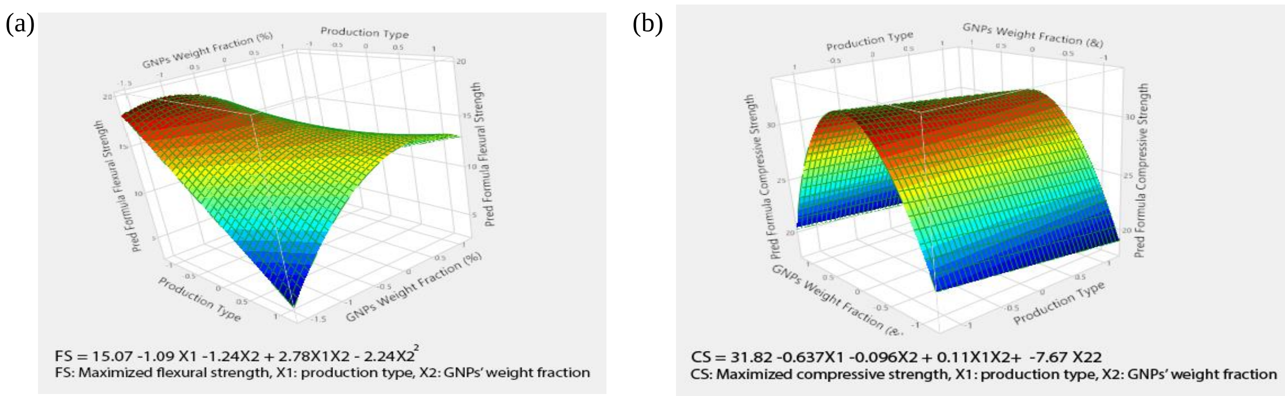

Fig. 4: RSM plots production type and GNPs' weight fraction (a) flexural strength and (b) compressive strength

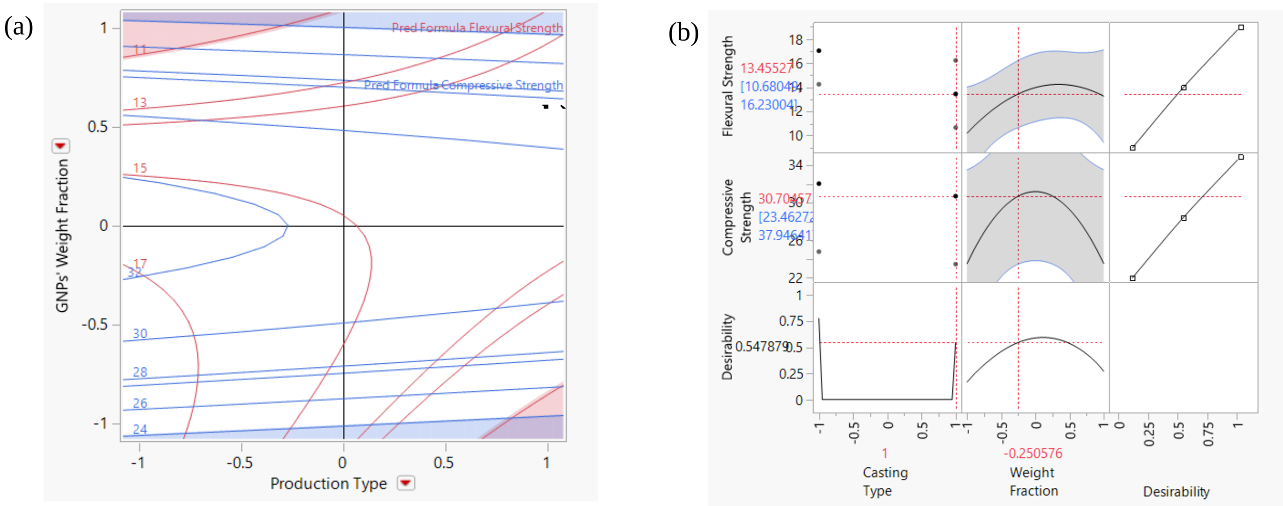

Fig. 5: (a) Production type and GNPs' weight fraction variables contour profiles and (b) maximized desirability functions

4 Conclusions

In this research, the RSM methods were used to investigate the impact of adding different GNPs' concentrations on the mechanical properties of 3D-printed clay. Mixes containing several Nanoparticles weight fractions were tested for flexure and compression, and the results were then analysed. Considering the study's findings, the following conclusions have been drawn:

i. The results showed that adding an appropriate dosage of GNPs to conventional clay could significantly improve the mechanical properties, thus increasing its suitability for construction printing applications without effecting the structural components' capacity, such as clay walls.

ii. Among all mixes, the one fabricated by traditional moulding with 0.1% GNPs had the highest flexural strength, while the mix containing 0.2% GNPs achieved the highest flexural strength between the printed mix, with an increment of about 7.5% and 21.1%, respectively, compared to the plain mix of each series.

iii. The compressive strength results showed a similar trend for the clay samples fabricated by both methods under the different dosages of GNPs.

iv. RSM techniques proposed empirical formulas to determine the compressive and flexural strengths of GNPs-modified 3-D printed clay.

v. RSM analysis showed that the maximum flexural and compressive strength response for 3D-printed clay was obtained at low dosages of GNPs (0.1 wt.%).

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