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Full Length Article

Using cellulose nanocrystals to improve the mechanical properties of fly ash-based geopolymer construction materials

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ABSTRACT

Ordinary Portland cement production is one of the biggest emitters of carbon dioxide. Consequently, there is a strong need for construction materials with lower environmental footprints. However, the development of alternative green construction materials requires a standardized framework. Although cellulose nanocrystals have shown considerable reinforcement potential in conventional construction materials, its effect on the mechanical properties of fly ash-based geopolymers as green construction materials is not known. Consequently, a detailed database outlining the cellulose nanocrystals interactions on the compressive strength, density, and corrosion resistance properties of geopolymers can optimize and guide further research efforts. The aims of this study were to firstly determine the effect of cellulose nanocrystals on the mechanical properties of fly ash-based geopolymers. Secondly, to produce a database of the effects of cellulose nanocrystals concentration and activator concentration on the mechanical properties of the formed geopolymers. Finally, to formulate an empirical framework to develop green construction materials. An empirical framework was developed alongside the cellulose nanocrystals-reinforced geopolymers, which were optimized using a statistical experimental design. The experimental results yielded the geopolymer property database. It was found that low cellulose nanocrystals concentrations (less than 0.5%) favoured the geopolymer mechanical properties. Using industrial wastes to produce green construction materials can divert industrial wastes from landfills and minimize the widespread use of environmentally degrading conventional construction materials. The framework developed in this study can facilitate the commercialization of green construction materials in industry.

1. Introduction

Environmental concerns remain one of the main challenges faced by the cement industry [1]. The production of ordinary Portland cement is energy-intensive and a major contributor to industrial greenhouse gas emissions [2]. Considering the growing global population, novel green alternatives to ordinary Portland cement are required to improve the sustainability of the construction industry.

Fly ash-based geopolymers are a viable alternative to ordinary Portland cement in the building industry [1,2]. However, more research is required to determine methods by which fly ash-based geopolymers can be enhanced to become commercially viable and reliable to completely replace ordinary Portland cement. One option considered in this study was the use of cellulose nanocrystals as a reinforcing component in fly-ash based geopolymers. Cellulose nanocrystals are nanoparticles derived from cellulose, a renewable resource. Due to its unique optical, rheological, and mechanical properties, cellulose nanocrystals are finding widespread applications in the construction, automotive, paper, medical and food sectors. It has been shown to be a high strength polymer material whose mechanical properties compare favorably, and often exceed those of traditional reinforcement materials such as stainless steel and Kevlar [3].

As part of a larger study, a systematic *meta*-analysis concluded that cellulose nanocrystals positively impacted the mechanical properties of construction materials. The aggregate effect of the

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studies included in the *meta*-analysis revealed that cellulose nanocrystals reinforcement exhibited a positive effect on the compressive strength improvement in ordinary Portland cement construction materials. This effect indicated that it would be beneficial to perform further experimental studies to investigate the reinforcement capabilities of cellulose nanocrystals in the development of a novel, green construction material using industrial waste streams such as fly ash and sawdust used as source of cellulose to produce cellulose nanocrystals. Both these waste streams are disposed of by landfilling or stockpiling on site, and in the case of sawdust, is also burned.

The aims of this study were threefold. The first aim was to evaluate the effects of the alkaline activator concentration and cellulose nanocrystals concentration on the mechanical properties of fly ash-based geopolymer construction materials. The second aim was to produce a database of the effects of cellulose nanocrystals concentration and activator concentration on the mechanical properties of the formed geopolymers in the form of three-dimensional surface response plots. Finally, the third aim entailed the development of a globally applicable framework to produce green construction materials based on the work involved to meet the first two aims. Therefore, cellulose nanocrystals-enhanced geopolymer construction materials as a green alternative to ordinary Portland cement, upon which the empirical green construction material development framework was based, could contribute to achieving long-term sustainability of the construction industry.

2. Literature review

2.1. Overview of the current state of the construction industry

The construction industry is challenged by depleting fossil fuel reserves, scarce raw materials, increasing demand, growing environmental concerns, and a stagnant world economy [1]. The production of ordinary Portland cement contributes to approximately 6% of all industrial carbon emissions [1] and 5 to 7% of the emissions directly responsible for global warming [1]. The calcination phase of the ordinary Portland cement production process results in the greatest proportion of carbon dioxide emissions [1]. Two methods were recommended to decrease the carbon dioxide emissions: changing the raw materials in the calcination process and/ or changing the manufacturing process [1]. Considering that these recommendations cannot be easily implemented, greener alternatives are required. Green materials are environmentally friendly, durable, bio-based, recycled and exhibit low toxicity and emissions [1]. There exists a rapidly expanding market for green building materials [1]. Fly ash, blast furnace slag and silica fumes are typical cement replacement materials that have been documented and validated [1].

Geopolymers are cementitious materials with three dimensional structures that are formed by the activation of aluminosilicate materials at relatively low temperatures [2]. Geopolymer cements are produced from secondary raw materials such as fly ash and metakaolin by the activation of alkali and alkali silicate solutions. It is noteworthy that several waste materials can be utilized to produce geopolymer construction materials. This finding enables energy and carbon dioxide savings in the construction sector, substantiating the classification of geopolymers as green construction materials.

Despite the advantages of geopolymers as green construction materials, further work is required to improve the technology and strengthen the potential for commercial applications [2]. To realize the implementation of green construction materials, price stability is mandatory for investment in low carbon cement technologies [1]. Considering that building practitioners identified high initial cost premiums as barriers to investing in green practices [2], government subsidies must be made available to green cement manufacturers. Furthermore, the knowledge gap between researchers and practitioners should be closed. To enable optimized and uniform geopolymerization processes that can be adopted by the construction industry, the implementation of regulatory standards is required [4].

2.2. Overview of green construction materials

Green construction materials are defined as being comprised of at least one waste material, or its production is environmentally friendly, or it has high performance and life cycle sustainability [5]. Considering that cement manufacture accounts for 8 to 10% of the total global carbon dioxide emissions [5], green construction materials are coveted as they reduce the demand for natural resources, the associated energy consumption, and greenhouse gas emissions [6,7]. The use of green construction materials prevents the mining of exorbitant quantities of naturally occurring materials for concrete production [8]. Green construction materials are advantageous in that they reduce environmental pollution, are comparatively economical, and exhibit good thermal and acid resistance [8–10]. The use of waste materials results in better compressive and tensile strength, improved sulphate resistance, decreased permeability and improved workability [8]. Green concrete shows greater durability, strength development, thermal resistance, and lower shrinkage than Portland cement [11]. The use of fly ash in green construction materials saves valuable landfill space and reduces the energy consumption of the production process [7] by eliminating the combustion of fuel and the decomposition of limestone [10].

2.3. Fly ash-based geopolymers as green construction materials

Substantial quantities of fly ash are generated globally, thus posing a serious threat to the environment [12]. Therefore, the use of fly ash as precursor materials in geopolymer production is recommended [12]. The silicone to aluminium ratios, the type and amount of the alkali solution, temperature, curing conditions, and additives are critical factors in the geopolymerization process [13]. The mechanical properties of fly ash-based geopolymers depend on the chemical composition, chemical bonding, and porosity of the geopolymer [13]. The mechanical properties can be improved by adjusting the silicone to aluminium ratios, alkali solutions, curing conditions and reinforcing agents [13]. Although fly ash-based geopolymers can be used as novel green cement, further studies on fly ash-based geopolymers are recommended to enhance the mechanical performance, scale up production, and explore new applications [13].

2.4. Cellulose nanocrystals to enhance geopolymer construction materials

Combining the endorsement of biomaterial applications in construction materials [14] with the global emergence of geopolymers as novel green construction materials [1,2,4], the application of cellulose nanocrystals in the fortification of fly ash-based geopolymers should be considered. Cellulose nanocrystals are a relatively new class of biomaterials known for their high-strength applications. The production of cellulose nanocrystals is environmentally friendly as it can be produced from waste biomass materials.

The research of Cao et al. (a) [15] investigated the influence of raw and sonicated cellulose nanocrystals on the microstructure of cement paste. Considering that the dispersion of cellulose nanocrystals within the geopolymer matrix is a known challenge, a novel method to measure the concentrations of the adsorbed cel-

lulose nanocrystals (on the cement surface) and the free cellulose nanocrystals (mobile in water) was developed. Most of the cellulose nanocrystals (in excess of 94%) actively comprised the cement matrix. Furthermore, the total porosities of cement pastes with raw and sonicated cellulose nanocrystals were determined. The results indicated total porosities of 14.8% and 14.4%, respectively-a reduction from 16%. It was expected that the decreased sample porosity would yield greater strength properties. The sonicated cellulose nanocrystals were well dispersed; thereby preventing the formation of agglomerates that can lead to pores, voids, and air entrapment in cement pastes.

Furthering the research of Cao et al. (a) [15], the study of Cao et al. (b) [16] determined the influence of cellulose nanocrystals dispersion within the cement matrix on its strength by investigating the agglomeration of cellulose nanocrystals. Agglomeration is undesirable, as it inhibits the dispersion of cellulose nanocrystals within the cement matrix. Ultrasonication was used to disperse the cellulose nanocrystals and rheological measurements quantified the agglomeration extent. Agglomerates began to develop when the cellulose nanocrystals concentration exceeded 1.35% by volume. A geometrical percolation model verified this experimental finding. According to the model, the threshold cellulose nanocrystals concentration resulting in agglomeration was 1.38%. This was significant as it indicated that the 1.38% cellulose nanocrystals concentration should not be exceeded to prevent agglomeration. Furthermore, it was found that the optimal cellulose nanocrystals concentration for maximum strength development was 0.18%. It is noteworthy that a relatively minute quantity of cellulose nanocrystals is required to achieve maximum strength development. A similar result was found in the study of [17] where a 0.2% concentration of cellulose nanocrystals improved the compressive strength and fracture properties of the geopolymer. The ultrasonication effectively dispersed the cellulose nanocrystals within the cement matrix, resulting in strength improvements of up to 50%, which was greater than the strength improvement of raw cellulose nanocrystals alone (20 to 30%). This indicated that the dispersion of cellulose nanocrystals significantly improves the flexural strength of cement pastes. Although this study was undertaken using cement paste, the novel dispersion method can be applied to geopolymers to enhance green construction materials.

Geopolymers are not being fully commercialized due to two uncertainties: financial [1] and technical [4]. The research below demonstrates the technical benefits achieved by the application of cellulose nanocrystals in construction materials. Although some research has focused on cellulose nanocrystals addition to ordinary Portland cement, the paradigm can be applied to geopolymers to produce fortified green construction materials.

The study of Barnat-Hunek et al. [18] determined the effect of cellulose nanocrystals on the physical properties of concrete. The study found that cellulose nanocrystals improved the freezing-thawing resistance>14-fold. This result is exceptional for construction applications that are required to withstand extreme temperature conditions and was attributed to the efficiency of nanocellulose in concrete hydrophilization. This result was consistent with the study of Cao et al. (a) [15]. Overall, the cellulose nanocrystals promoted the formation of calcium-silicate-hydrate in the concrete, which improved the density and changed the pore structure and interface characteristics. The improved density resulted in improved concrete strength. A similar finding was observed in the study of Dousti et al. [19] in which the porosity was reduced by 33% and the surface area was reduced by 66% by the addition of cellulose nanocrystals. The cellulose nanocrystals addition increased the compressive and tensile strength by 60% in the first 24 h. The study of Dousti et al. [19] was novel as it investigated the effect of cellulose nanocrystals on the hydration of cement paste. The cellulose nanocrystals promoted the formation of hydration products, resulting in greater compressive and tensile strengths.

The study of Liu et al. [20] investigated the effect of cellulose nanocrystals on the compressive strength of cement pastes. X-ray computed tomography and nitrogen adsorption analyses revealed that the cellulose nanocrystals refined the pore structure in the cement matrix. Almost no hydration products were formed in the cement matrix without cellulose nanocrystals addition. Like the research of Liu et al. [20], the study of Lee et al. (a) [21] investigated the effect of cellulose nanocrystals on the mechanical performance of construction materials. The cellulose nanocrystals solutions were prepared in the following concentrations by volume: 0.4, 0.8, and 1.2. As recommended in the research of Cao et al. (a) [15], Cao et al. (b) [16] and Lee et al. (b) [22], ultrasonication was used to disperse the cellulose nanocrystals in the aqueous solutions. The optimal cellulose nanocrystals solution concentration was found to be 0.8% by volume as it improved the shrinkage rate and mechanical performance. This finding reinforced previous research which found that only a small quantity of cellulose nanocrystals is required for maximum strength development in construction materials [1].

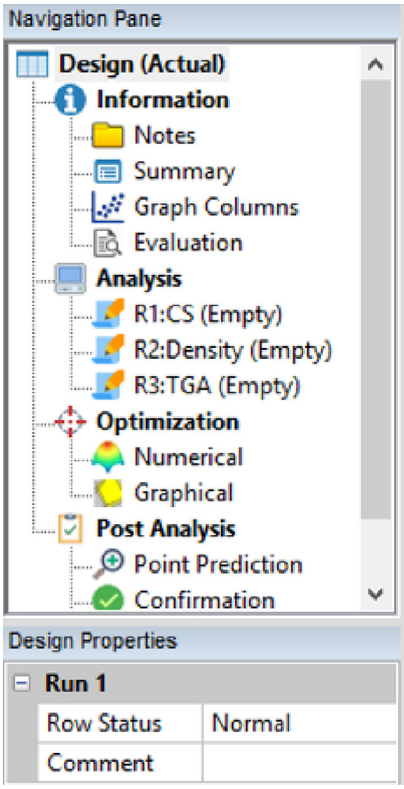
According to the study of Flores et al. [23], the addition of cellulose nanocrystals to the geopolymer mixture initially delays the hydration but improves the hydration at later stages. It was also revealed that cellulose nanocrystals increased the non-evaporable water content with respect to the control mixture. Two cellulose nanocrystals concentrations were investigated: 0.4 and 0.8%. At both concentrations, the non-evaporable water content was improved. This supported the idea that at higher cellulose nanocrystals concentrations, the alkaline activator was slightly diluted due to the additional water content from the solution of cellulose nanocrystals. This finding decreased the effectiveness of the alkaline activator. Furthermore, the cellulose nanocrystals particles fill the smallest gaps in the mixture paste, thus decreasing the porosity of the sample [24].

Table 1
Literary correlations between the electrical resistivity and corrosion resistance of construction materials.

Resistivity Values (Ω.m)		
Corrosion Risk	Hornbostel et al. [29]	Velu [32]
High	<100	<50
Moderate	100 to 500	50 to 100
Low	500 to 1000	100 to 200
Negligible	>1000	>200

2.5. The effect of alkaline activators in geopolymerization

In the study of Hadi et al. [25], fly ash with a high aluminosilicate content were activated at lower dosages of sodium silicate and sodium hydroxide. The opposite effect was observed for fly ash with lower aluminosilicate content. This implied that greater activator concentrations are required for the activation of geopolymers. In the study of Roopchund [26], the alkaline activator concentration was varied between 6 and 16 M and was found to be directly proportional to the compressive strength of the resulting geopolymers. In the study of Hamidi et al. [27], the effect of the alkaline activator concentration within the range of 4 to 18 M on the flexural strength of the geopolymer was investigated. The activator concentration was found to be directly proportional to the flexural strength until the activator concentration of 12 M was reached. Thereafter, a decline in the flexural strength was observed, implying that the 12 M concentration was optimal for the development of flexural strength.



Run	Factor 1 A:CNC concentr... %	Factor 2 B:Activator conc... M
1	0.86	9.7
2	1.28	12
3	0	11.4
4	2	10.52
5	0.86	8.02
6	0	8
7	2	10.52
8	0.71	11.12
9	2	12
10	0	11.4
11	0	9.72
12	1.56	9.4
13	0.86	9.7
14	0.86	9.7
15	1.7	8
16	1.7	8

Fig. 1. Summary of the statistical experimental design.

2.6. Corrosion resistance in construction materials

The corrosion process in concrete is partially controlled by the transport of ions through the concrete microstructure [28,29]. Considering that ions are charged, the ability of a material to resist the transfer of charge depends on its electrical resistivity [29]. Thus, a relationship is expected between the concrete corrosion and the corresponding electrical resistivity. Although the studies of Alonso et al. [30], Farhana et al. [31] and Velu [32] found clearly defined relationships of inverse proportionality between the electrical resistivity and corrosion resistance of construction materials, specific experimental requirements are outlined to study the relationship between concrete corrosion and electrical resistivity [29]:

1. A working electrode, preferably construction steel embedded in a mortar or concrete sample to reflect practice-related conditions.
2. A technique to measure corrosion rate, either from the surface or as an embedded device.
3. A technique to measure concrete resistivity.
4. A method to initiate corrosion.

The research of Velu [32] presented an experimental study of the electrical resistivity of geopolymer paste using the Wenner four probe method. The range of electrical resistivity values obtained varied between 537 Ω.m and 61575 Ω.m. As with previous studies, a relationship of inverse proportionality between the electrical resistivity and corrosion rate was exhibited [28,30,31]. A guideline to determine the potential corrosion resistance of a construction material based on its electrical resistivity is indicated in Table 1.

The electrical resistivity of concrete depends on two broad categories: intrinsic factors and factors affecting the resistivity measurements [28]. The intrinsic factors include the water to cement ratio, aging, and pore structure, while the factors affecting the resistivity measurements include the specimen geometry, moisture content, temperature, electrode spacing, and presence of a rebar. Furthermore, the addition of reactive supplementary cementitious materials such as blast furnace slag and fly ash leads to higher electrical resistivity due to reduction in capillary porosity and hydroxyl ions.

Electrical resistivity measurements can be undertaken using electrodes positioned on a specimen surface or placing an electrode-disc or linear array or a four-probe square array on the concrete's surface [31,33]. There are four principle device techniques that are used to measure resistivity: bulk electrical resistivity test, surface disc test, Wenner four-point line array test, and four-probe square array test.

3. Materials and methods

3.1. Materials

3.1.1. Fly ash

The fly ash was sourced from Matla Power Station (Eskom) in South Africa. Considering that it was classified as Class F, with a silicon to aluminium ratio of unity and a low organic content (characterized by a loss on ignition value less than 5%) it was identified as a suitable precursor fly ash material to be used in the production of the geopolymers.

3.1.2. Sawdust-based cellulose nanocrystals

The sawdust was obtained from Sappi sawmills in South Africa. Considering that the overall yield of the South African Forestry Sector is only 47% [34], it is deemed wasteful. To improve the sustainability of the industry, sawdust was identified as a viable by-product that could be used to produce cellulose nanocrystals, a high-value organic material with a wide variety of applications. The cellulose nanocrystals were produced using a proprietary tech-

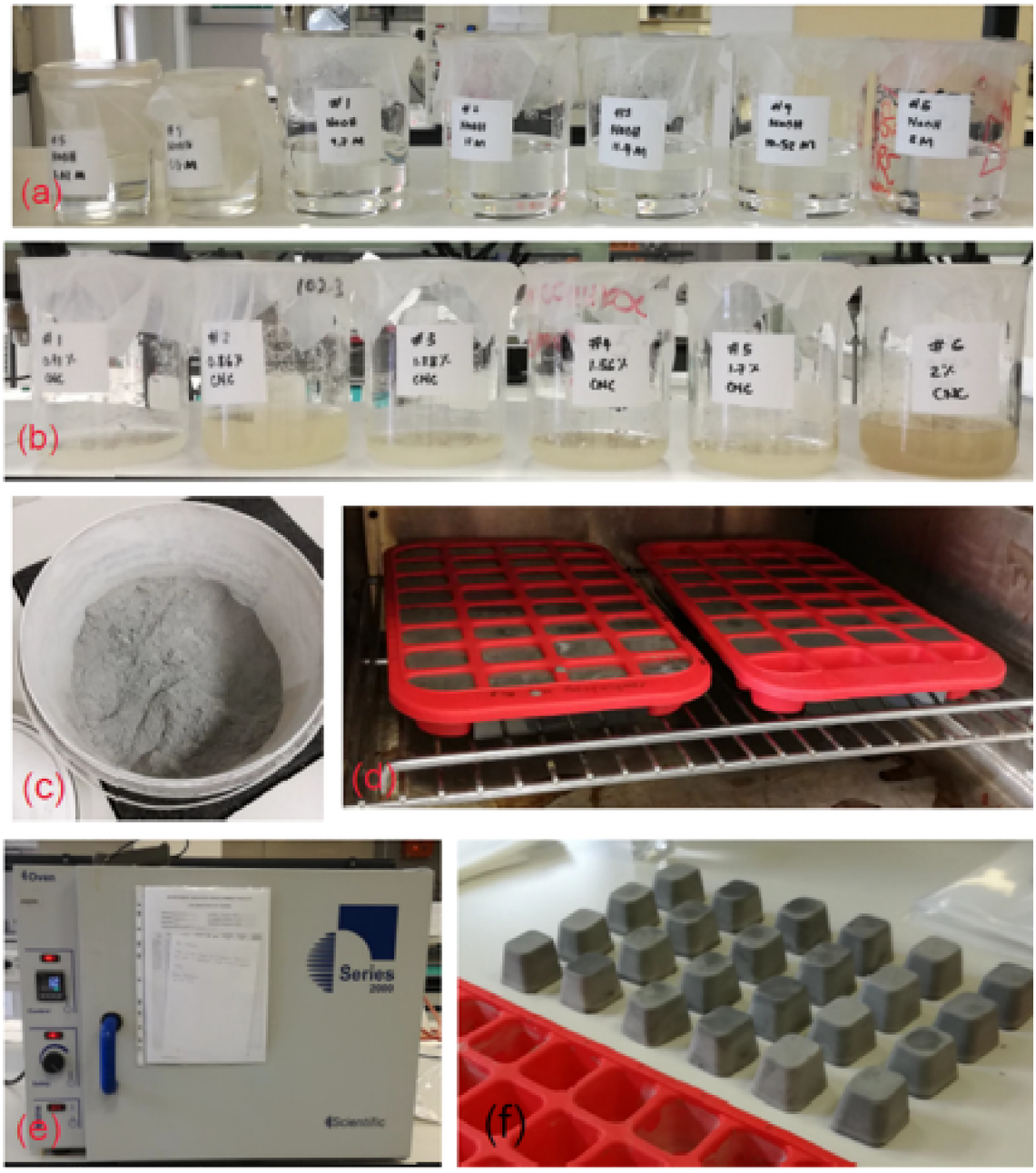


Fig. 2. (a) Alkaline activator solutions (b) Cellulose nanocrystals solutions (c) Fly ash (d) Moulded geopolymer pastes prior to curing (e) Oven used for curing (f) De-moulded geopolymers post-curing.

nology and was subsequently used as a reinforcing agent for the geopolymer construction materials in this study.

3.2. Geopolymer mixture design

A trial-and-error procedure was used to design the optimal geopolymer mixture ratios of the comprising components: fly ash, water, alkaline activator, and cellulose nanocrystals suspension. The optimal ratio resulted in mixture consistency and workability to allow effective moulding. This optimal ratio was 60.8 ml of the liquid component (comprising of the alkaline activator and cellulose nanocrystals solution) to 100 g of the fly ash. The liquid component was comprised of 75% alkaline activator and 25% cellulose nanocrystals suspension. For the trials that did not require cellulose nanocrystals solution, deionized water was used instead. Considering that the geopolymers were required to undergo multiple tests, the quantities used in this mixture design allowed the production of four geopolymer cubes per trial.

3.3. Statistical experimental design

The experimental trials entailed variation of the alkaline activator concentration and the cellulose nanocrystals concentration. The Design Expert software was used to statistically design the experimental trials to optimize the number of trial runs. An optimization purpose was selected, and a response surface model was chosen. The response surface model enabled the effects of the parameters (alkaline activator concentration and cellulose nanocrystals solution concentration) to be visually observed on a single three-dimensional surface curve.

Fig. 3. Compressive strength equipment; A- Tensile testing machine, B- Load indicator, C- base plate, D- Load exertion controls.

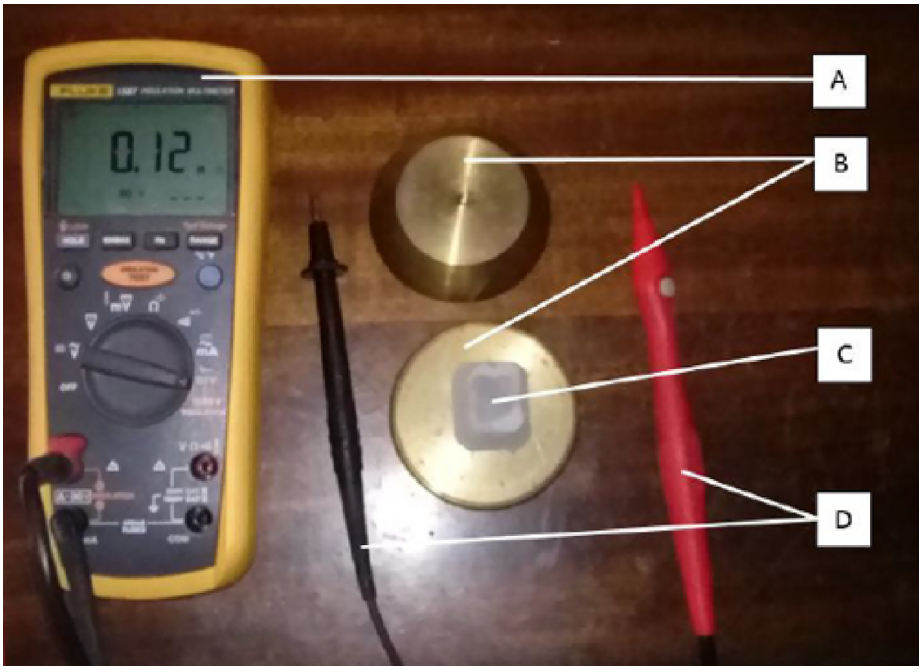


Fig. 4. Electrical resistivity experimental setup; A- Fluke 1577 insulation multimeter, B- Conductive plates, C- Geopolymer sample, D- Current-measuring electrodes.

Based on the design requirements, the experimental plan was calculated and output by the software as shown in Fig. 1. The combination of the variation factors resulting in the 16 experimental runs can be clearly observed.

3.4. Geopolymer synthesis

3.4.1. Preparation of solutions

Following the literature recommendation to improve the mechanical properties of the geopolymer construction materials [13], freeze-dried cellulose nanocrystals were mixed into the required quantities of deionized water to produce the required solution concentrations shown in Fig. 1. The solutions were left mixing overnight to ensure adequate hydration and dispersion. Novel cellulose nanocrystals dispersion techniques were not considered as a large proportion (exceeding 90%) of the cellulose nanocrystals were found to be adsorbed into the geopolymer matrix [15]. The cellulose nanocrystals concentration range was based on the range reported in the literature [15,16,21]. The required masses of the sodium hydroxide pellets were weighed to prepare the activator solutions, according to the activator concentrations shown in Fig. 1.

3.4.2. Geopolymer moulding

For each of the experimental runs dictated by the experimental design software (Fig. 1), 100 g of fly ash was weighed, placed into a glass beaker, and contacted with the liquid components (alkaline activator solution and cellulose nanocrystals solution). The con-

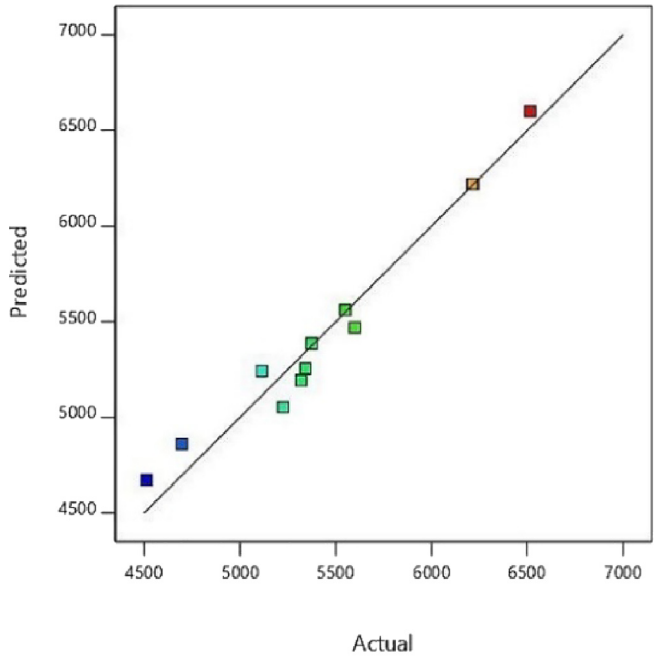


Fig. 5. Compressive strength predicted versus actual.

tents were mixed until a uniform paste was formed. The paste was then decanted into the silicone mould.

3.4.3. Geopolymer oven curing

The silicone moulds containing the geopolymer pastes were placed into an oven which was preset at 40 ℃. Two different oven curing techniques were investigated: curing at 40 ℃ for two hours and then at 60 ℃ for an additional 24 h. Thereafter, the samples were rotated and cured at 60 ℃ for a further 24 h to ensure spatial uniformity with respect to the curing. Secondly, the samples were cured at 40 ℃ for two hours and then at 60 ℃ for another 24 h. In the second case, the samples were not rotated and there was no further curing. A pictorial representation of the steps encompassing the geopolymer synthesis can be observed in Fig. 2.

3.5. Geopolymer testing

3.5.1. Compressive strength

Each sample was placed between two flat plates on a Rohloff Universal Tensile Tester. A force was applied causing the plates to move closer, resulting in the crushing of the sample. The force at which the sample was crushed was recorded and displayed by the machine. This force was the compressive strength of the sample. The compressive strength tests were done in duplicate. The apparatus used to measure the compressive strength of the geopolymer samples is shown in Fig. 3.

3.5.2. Density

The mass of each sample was measured using a Mettler Toledo laboratory scale. The dimensions of each sample were determined using a digital Vernia caliper. The volume of the sample was calculated using the sample dimensions and the density calculated using equation (1):

$$Density = \frac{Mass}{Volume} \quad (1)$$

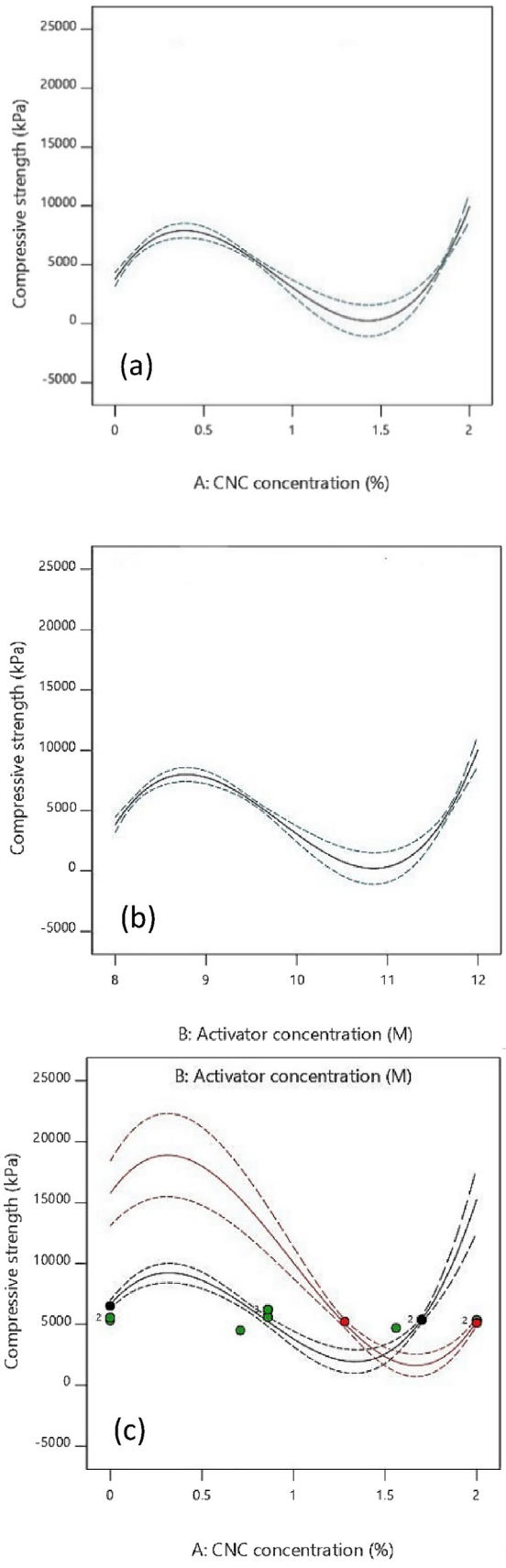


Fig. 6. Panel plot of compressive strength interactions.

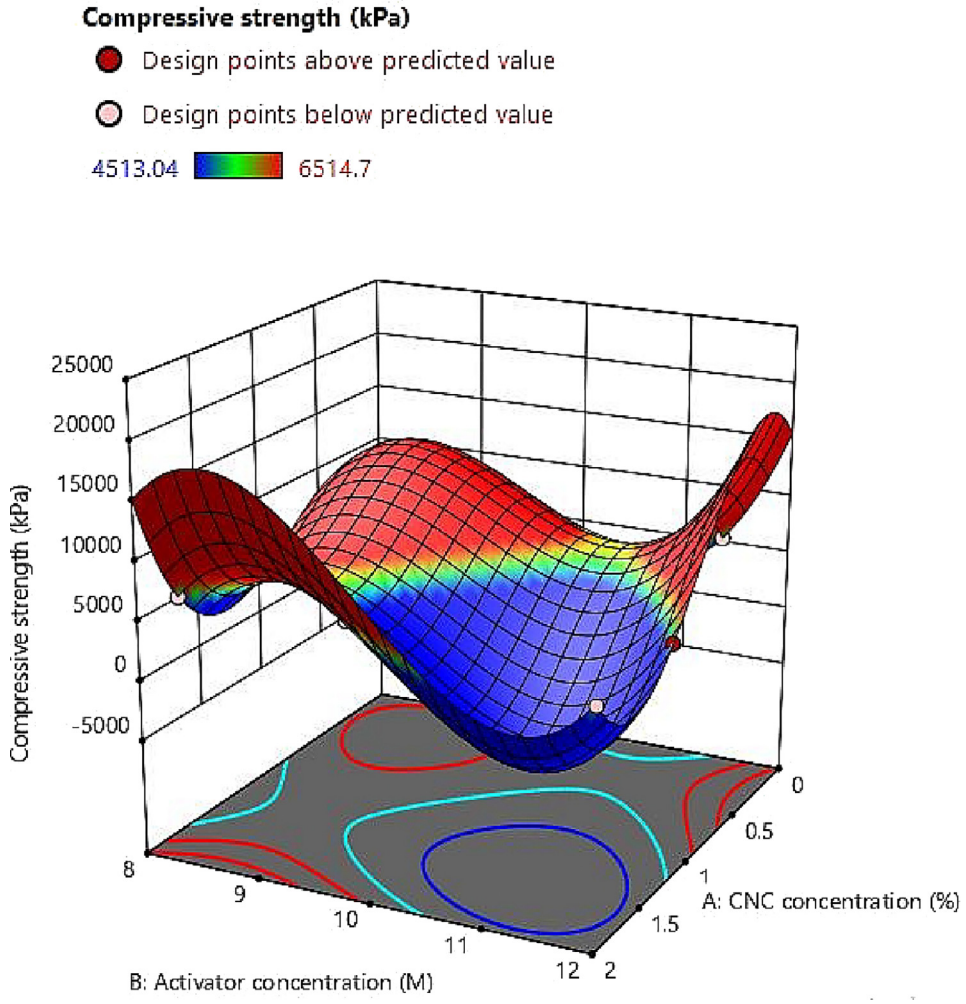


Fig. 7. Compressive strength surface response model.

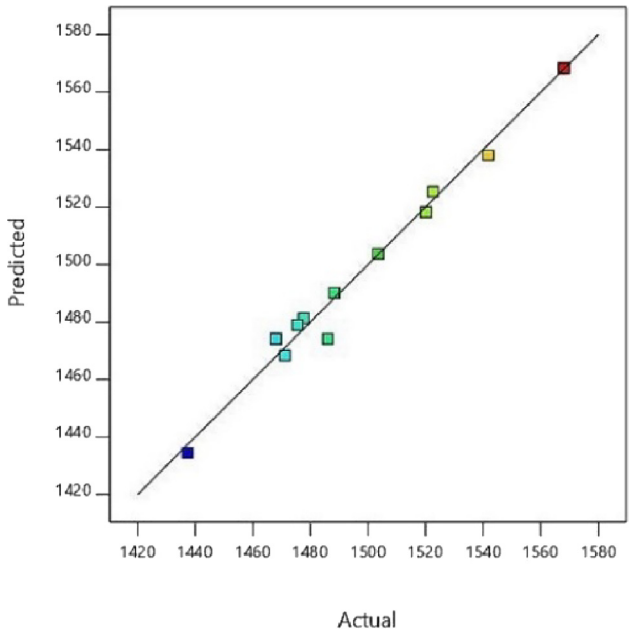


Fig. 8. Density actual versus predicted values.

3.5.3. Electrical resistivity

The following equipment was used for the testing: Fluke 1577 Insulation Multimeter, conductive plates and measuring electrodes. The geopolymer samples were placed between the top and bottom conductive plates. The Insulation Multimeter included a positive, negative and a guard terminal. The guard terminal is at the same potential as the negative terminal but not in the measurement path. The guard terminal is used to improve the accuracy of the measurements by eliminating stray measurements. The volume resistivity (ρ) relationship with respect to the measured resistance (R), area of the top electrode (A) and height of the construction material sample (h) is mathematically represented by equation (2) [33]:

$$\rho = \frac{RA}{h} \tag{2}$$

The experimental setup used to measure the electrical resistivity of the geopolymer samples is shown in Fig. 4.

4. Results and discussion

4.1. Geopolymer synthesis

The first curing procedure (48-hour curing with rotation) resulted in the formation of cracks on all the geopolymer samples,

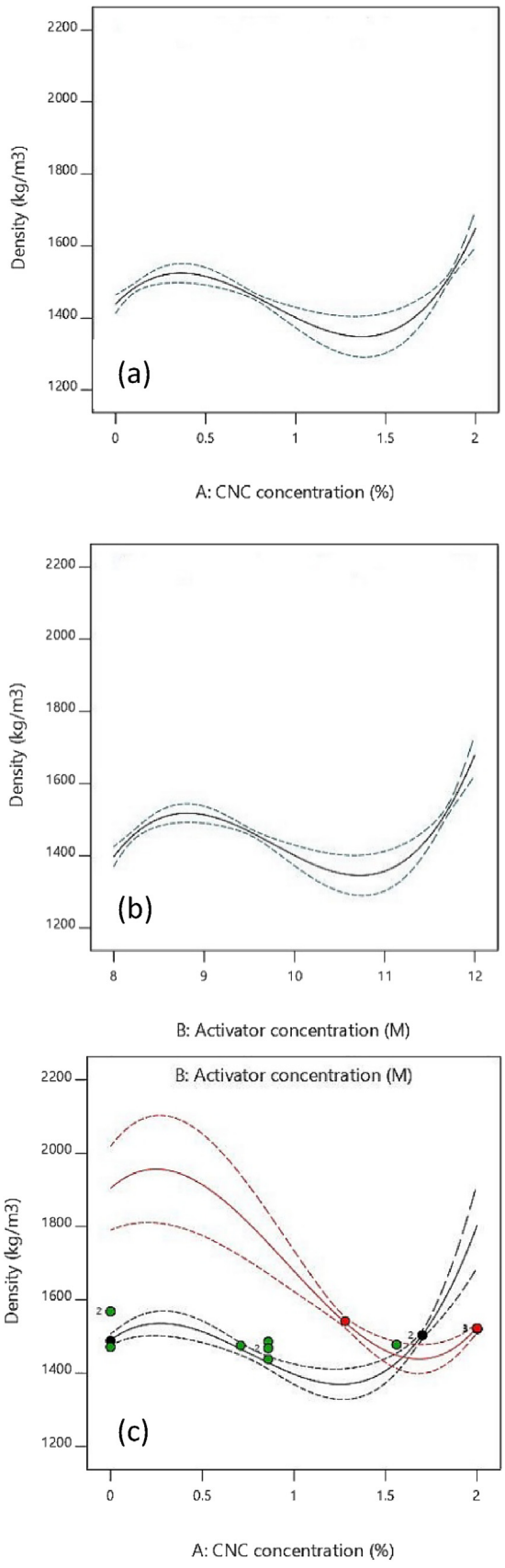


Fig. 9. Panel plot of density interactions.

except for those containing high amounts of cellulose nanocrystals (Runs 4, 7 and 9). The cracking was attributed to the rapid exposure of the sample to higher temperature when rotated and cured for an additional 24 h. The rapid temperature exposure caused rapid moisture evaporation from the microstructure of the sample, culminating in the formation of the cracks [26]. For the samples containing the high cellulose nanocrystals concentrations, the cellulose nanocrystals within the geopolymer matrix prevented the rapid moisture evaporation [23]; thereby preventing cracking. This result was also observed in the research of Lee et al. (a) [21]. Hence it is recommended that higher cellulose nanocrystals concentrations be used for applications that require thermally stable geopolymers as opposed to high strength geopolymers.

That the alternative curing method (without de-moulding and rotating the samples after the initial 24 h of curing at 60 °C) did not result in any cracks was a good finding as cracking is undesirable as it compromises the structural integrity of the geopolymer samples. Hence, de-moulding and sample rotation is not recommended in the curing phase.

4.2. Compressive strength

4.2.1. Statistical model

The software recommended the 2FI statistical model to represent the combined effects of the cellulose nanocrystal concentration and alkaline activator concentration on the compressive strength. The adequate precision indicated by the software combined with the insignificant lack of fit, implied that the model could be deemed accurate. The model accuracy is validated by the graphical representation of the predicted versus actual values in Fig. 5.

4.2.2. Two-dimensional interactions

The panel plot in Fig. 6 is a visual representation of the interactions between the individual and combined variation parameters on the compressive strength. It is noteworthy that lower concentrations of cellulose nanocrystals favoured the compressive strength in Fig. 6 (a), as affirmed by the findings of Cao et al. (b) [16], Lee et al. (a) [21], Imbabi et al. [1] and Ghahari et al. [17]. At smaller concentrations, cellulose nanocrystals increase the bonding between particles in a composite material due to its adhesive property [24]. The increased bonds result in the formation of a network, which improves the strength of the material. Based on the results, the cellulose nanocrystals concentration should not exceed 0.5% to avoid a reduction in the compressive strength. It is likely that the cellulose nanocrystals may have agglomerated at concentrations exceeding 0.5% [16]; thereby resulting in decreased compressive strength results.

It is evident from Fig. 6(b), that the lower end of the alkaline activator concentration favoured the compressive strength of the geopolymer samples. This finding contradicts the study of Hamidi et al. [27] in which the best mechanical properties of the geopolymer were obtained at a high alkaline activator concentration of 12 M. However, the result agrees with the study of Hadi et al. [25] which indicated that fly ash containing a large quantity of fine particles and amorphous content would require a low dosage of alkaline activator to achieve good mechanical results. Considering that lower concentrations of the cellulose nanocrystals solutions and alkaline activators are required for greater strength development; the production costs can be reduced. This reduced production cost would effectively aid the long-term sustainability of the green material being proposed and directly addresses the financial concerns when developing green construction materials [1].

The intersection of the combined interactions shown in Fig. 6(c) is noteworthy as the intersection between the fitted curves would offer a trade-off between the optimal cellulose nanocrystal concen-

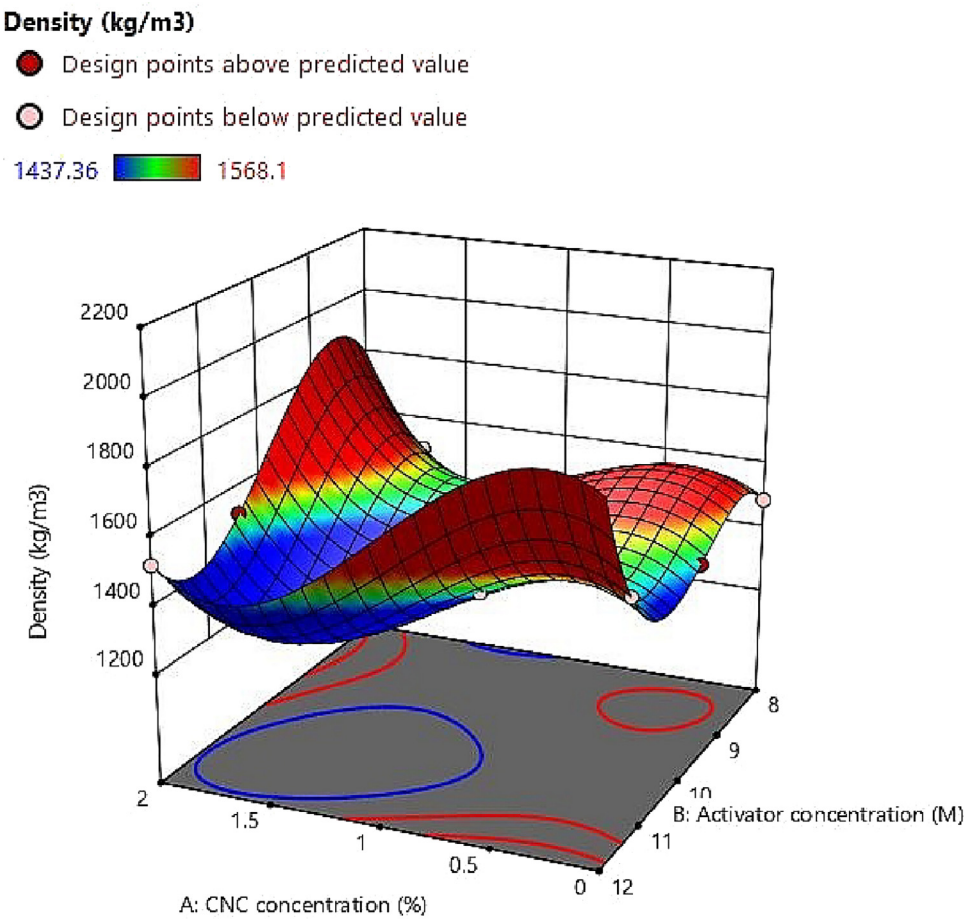


Fig. 10. Three-dimensional density response curve.

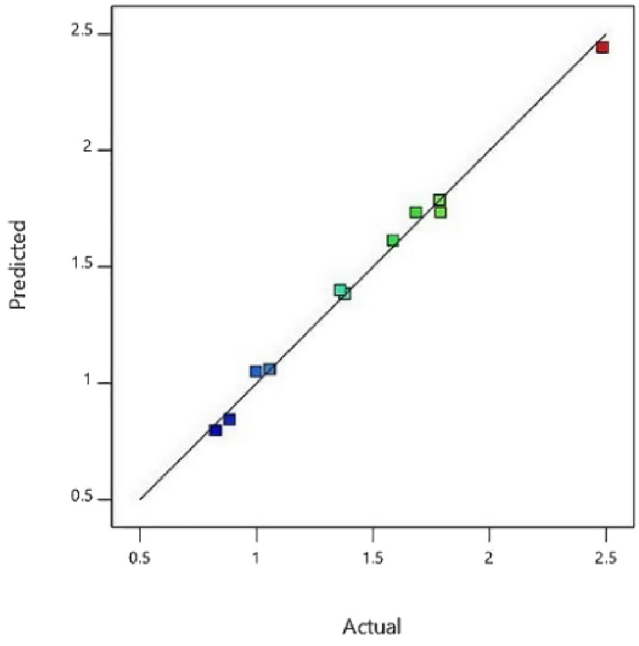


Fig. 11. Electrical resistivity actual versus predicted model (24-hour curing condition).

tration and alkaline activator concentration on the compressive strength. It can be observed that a combination of "middle point" concentrations offer the best trade-off, which would positively impact the economic sustainability of the production process.

4.2.3. Three-dimensional response surface model

The three-dimensional model depicted in Fig. 7 was able to depict a surface based on the interaction observed in the previous section. It was beneficial that the response curve was color coded, highlighting the areas of maximum and minimum compressive strength values. It was noteworthy that the lower to middle compressive strength range was more prominent than the higher end. However, if lower concentrations of the cellulose nanocrystals [1,16,17,21] and alkaline activator concentrations [25] were used, greater compressive strengths could have been achieved.

4.3. Density

4.3.1. Statistical model

The software recommended a cubic statistical model to represent the effects of the alkaline activator and cellulose nanocrystals concentrations on the geopolymer density. The statistical F and P values indicated that the model was significant by predicting a maximum 0.01% likelihood of noise data. In addition, the lack of fit was found to be insignificant, implying the validity of the cubic model representation. From the graphical representation of the actual and predicted values shown in Fig. 8, the experimental data points can be observed to follow the linear trend recommended by the software.

4.3.2. Two-dimensional Interactions

Fig. 9(a) clearly shows a cubic relationship between the concentration of cellulose nanocrystals and the geopolymer density. To achieve a high density geopolymer, the concentration of cellulose

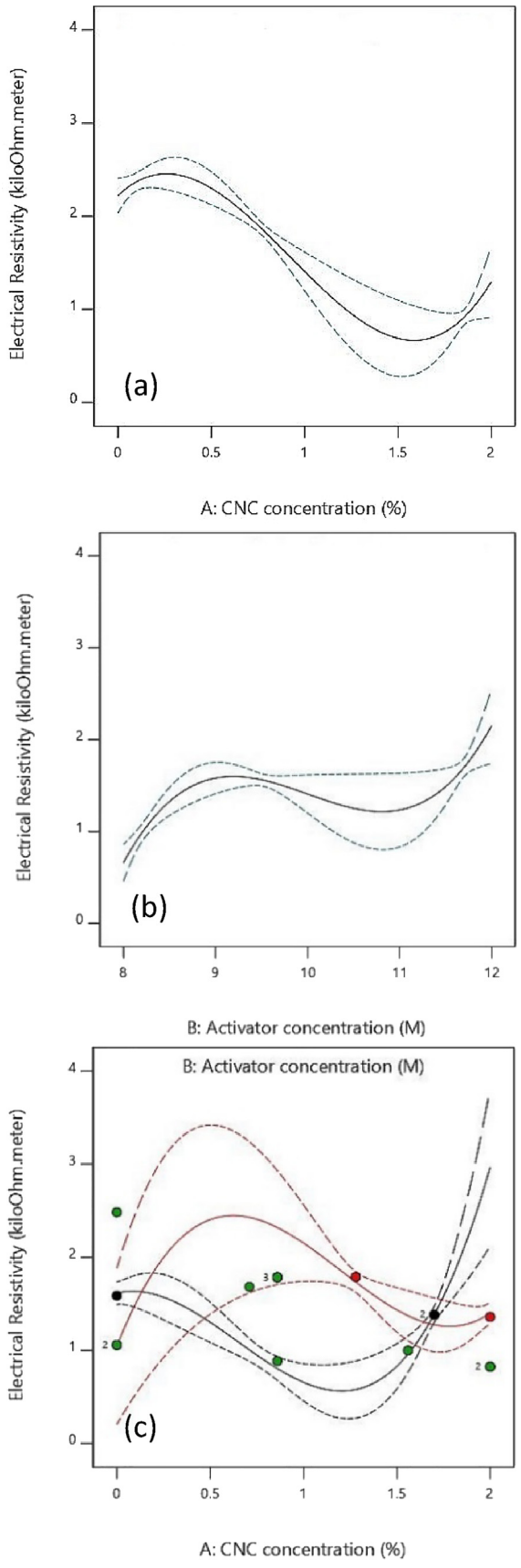


Fig. 12. Electrical resistivity interaction panel plots (24-hour curing condition).

nanocrystals should not exceed 0.5% (the region of the global maximum). Alternatively, a concentration range of cellulose nanocrystals within 1.75% and 2% was also found to favoured high geopolymer densities. The higher densities at the higher concentration range were attributed to the fact that as the cellulose nanocrystals hardens and merges into the geopolymer matrix during the curing process, the additional cellulose nanocrystals mass of the higher concentration of cellulose nanocrystals becomes apparent in the density of the formed geopolymer. This finding agrees with those of Cao et al. (a) [15] and Barnat-Hunek et al. [18] in which the cellulose nanocrystals were found to increase the sample density by reducing the porosity and promoting the formation of calcium silicate hydrate, respectively. Furthermore, this finding was supported by the fact that nanosized cellulose-based particles fill the smallest gaps in the mixture paste, thus increasing the overall density [24].

It was also notable that the concentration range between 0.5% and 1.75% favoured low geopolymer densities and should be applied for construction applications that required low densities.

The alkaline activator concentration also exhibited a cubic relationship with the geopolymer density as shown in Fig. 9(b). Again, this mathematical relationship is valuable when attempting the application of geopolymers in low density construction material applications. The individual interactions are clearly observed in the combined interactions plot of Fig. 9(c). This plot indicated that at concentrations of cellulose nanocrystals exceeding approximately 1.5%, the effect of the alkaline activator concentration also influences the geopolymer density.

4.3.3. Three-dimensional response surface model (24-hour curing)

The cubic functions representing the relationships between the cellulose nanocrystal concentration and alkaline activator concentration on the geopolymer density were clearly exhibited in the three-dimensional response curve in Fig. 10. This is favourable as the response model offers a clear exhibit of information when trying to customize a specific density for a specific construction application. This type of analysis can certainly aid the design and development process and support further research to commercialize the application of geopolymers as novel green construction materials.

4.4. Electrical resistivity

4.4.1. Statistical model (24-hour curing)

A cubic statistical model was suggested by the software to represent the electrical resistivity data. As with previous statistical models, a linear trend was observed between the actual and predicted values (Fig. 11). The F and P statistical values implied the significance of the model with only a 0.01% likelihood of noise in the data. This high degree of accuracy can be attributed to following the literary recommendations to perform accurate measurements [28,29].

4.4.2. Two-dimensional interactions (24-hour curing)

As observed in Fig. 12(a), a decreasing cubic trend was observed between the electrical resistivity and cellulose nanocrystals concentration. This implied that the lower cellulose nanocrystals concentrations favoured the electrical resistivity of the geopolymer construction materials. The electrical resistivity values exhibited a high range (exceeding 800 Ω.m). According to the literature, this implied a low to negligible potential for corrosion to take place [29,31,33]. However, the electrical resistivity range observed for the 24-hour cured samples was substantially lower than that of the 48-hour cured samples. This finding was directly attributed to the relatively higher moisture content present in the geopolymer samples that were cured for only 24 h when compared against

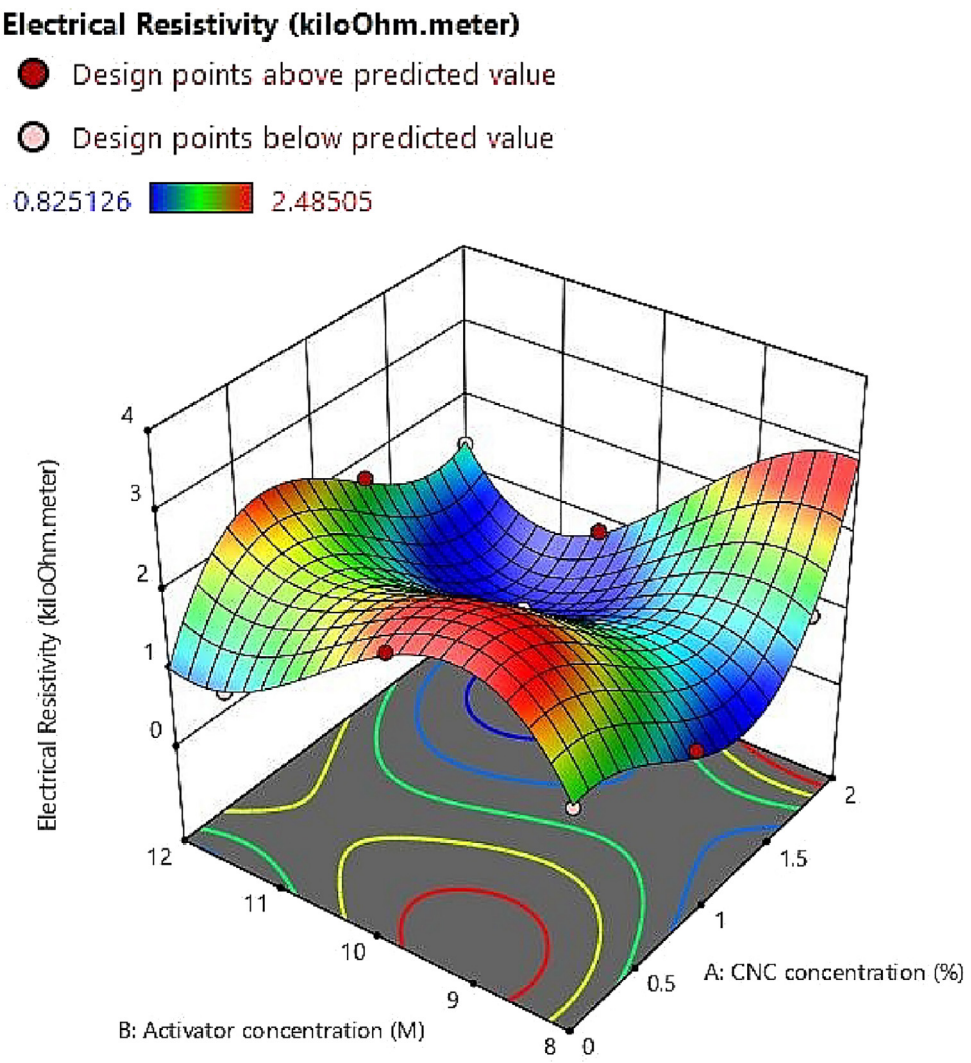


Fig. 13. Electrical resistivity three-dimensional surface response curve (24-hour curing condition).

the samples cured over the 48-hour period with rotation. The higher moisture content enabled the effective transfer of ions, thus decreasing the overall electrical resistivity of the construction material. Furthermore, the decrease in electrical resistivity observed with increasing cellulose nanocrystals concentrations can be directly attributed to the fact that the cellulose nanocrystals solution contains a great moisture content (in excess of 90% by volume). Considering that a higher cellulose nanocrystals concentration yields an inherently higher non-evaporable water content [23], the observed decreased electrical resistivity is expected, as confirmed in Fig. 12(a). Considering that the higher electrical resistivity values are linked to a lower corrosion potential in construction materials [29,31,33], it is recommended that lower cellulose nanocrystals concentrations be used in the development of the geopolymer construction materials.

As observed in Fig. 12(b), the activator concentration was found to rapidly improve the electrical resistivity as it was increased from 8 to 9 M, after which the electrical resistivity was slightly decreased as the activator concentration was increased from 9 to 11 M. Thereafter, a rapid increase in the electrical resistivity was observed as the activator concentration was increased from 11 to 12 M. It is therefore not feasible to use alkaline activator concentrations within the range of 9 to 11 M. Such feasibility considerations are vital in the development of economic green construction materials [1]. Interactions were observed between the cellulose nanocrystals concentration and the alkaline activator concentration at a low cellulose nanocrystals concentration of 0.2% and again at a concentration exceeding 1.5% as confirmed in Fig. 12 (c).

4.4.3. Three-dimensional response surface model (24-hour curing)

It is apparent from the three-dimensional response plot (Fig. 13) that the greatest electrical resistivity values were achieved at the combined conditions of high cellulose nanocrystals concentration and low activator concentration. Depending on the extent to which corrosion resistance is required in the geopolymer construction material being developed, it is recommended that the cellulose nanocrystals concentration and activator concentration be adjusted to achieve the desired degree of corrosion resistance. Considering that high cellulose nanocrystals concentrations are required, the least alkaline activator concentration must be determined to achieve the desired degree of corrosion resistance to ensure economic feasibility.

4.4.4. Statistical model (48-hour curing)

The quadratic and cubic statistical models were suggested to represent the electrical resistivity data for the 48-hour cured samples. As with previous statistical models, a linear trend was observed between the actual and predicted values (Fig. 14). The

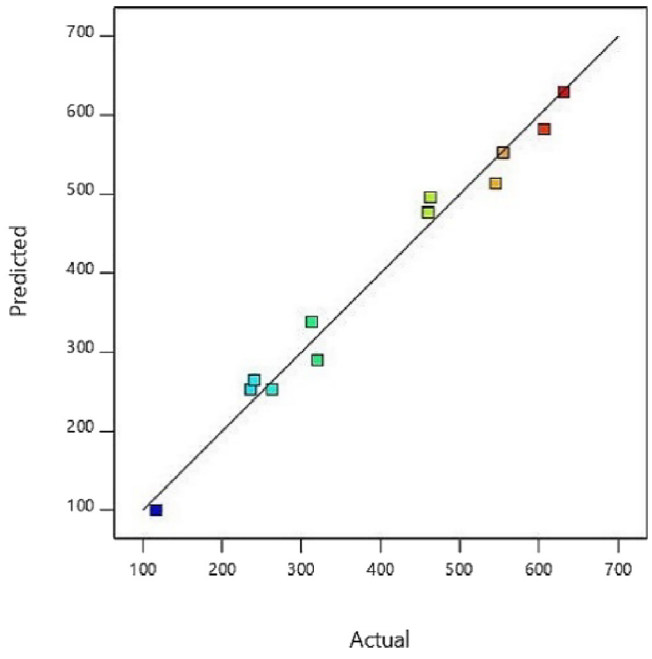


Fig. 14. Electrical resistivity actual versus predicted model (48-hour curing condition).

model F value suggested that the recommended model was significant with only a 0.01% chance of noise.

4.4.5. Two-dimensional interactions (48-hour curing)

The results obtained for the 48-hour cured samples differed greatly in two respects when compared against the results of the 24 h-cured samples. Firstly, the range of electrical resistivity values were substantially greater as compared to the 24-hour cured samples. This was directly attributed to the greater degree of moisture removal experienced by the 48-hour samples during the extended curing procedure. Secondly, as observed in Fig. 15(a), the electrical resistivity was found to increase as the cellulose nanocrystal dosage was increased. This finding implied that the 48 h-cured samples enabled greater moisture removal to the extent that the true reinforcing potential of the cellulose nanocrystals within the geopolymer matrix could be experienced.

As with the 24-hour cured samples, the effect of the alkaline activator on the electrical resistivity was marginal, as observed in Fig. 15(b). This implied that from the perspective of the electrical resistivity, there was no need for higher alkaline activator concentrations. The needless requirement of higher alkaline activator concentrations is a positive finding from the perspective of economic green construction material development [1].

Based on the intersection of the two independent variables in the combined interaction plot in Fig. 15(c), the presence of an interaction between the two independent variables was confirmed. This interaction implied that the effect of the cellulose nanocrystal concentration was dependent on the concentration of the alkaline activator. Hence, the alkaline activator concentration would need to be fixed while manipulating the cellulose nanocrystals concentration to achieve the desired degree of corrosion resistance.

4.4.6. Three-dimensional response surface model (48-hour curing)

From the three-dimensional response curves (Fig. 16), it was apparent that the greatest electrical resistivity values were obtained under conditions of high cellulose nanocrystals concentration and low alkaline activator concentration. The range of electrical resistivity values obtained under the 48-hour curing

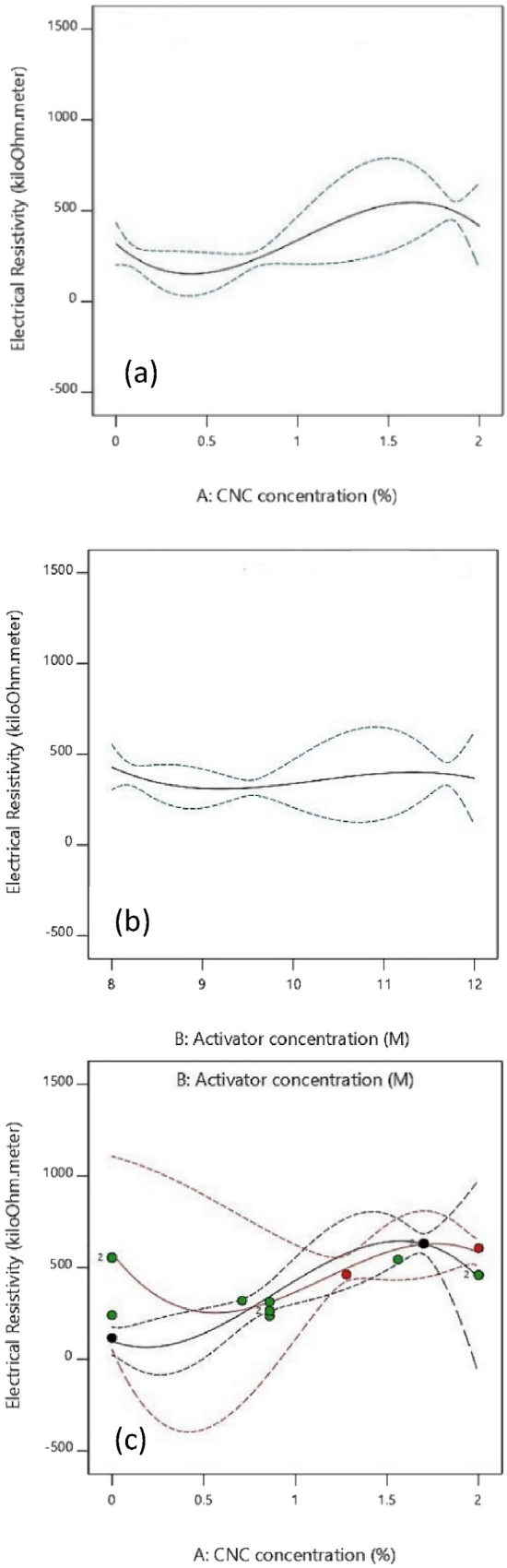


Fig. 15. Electrical resistivity interaction panel plots (48-hour curing condition).

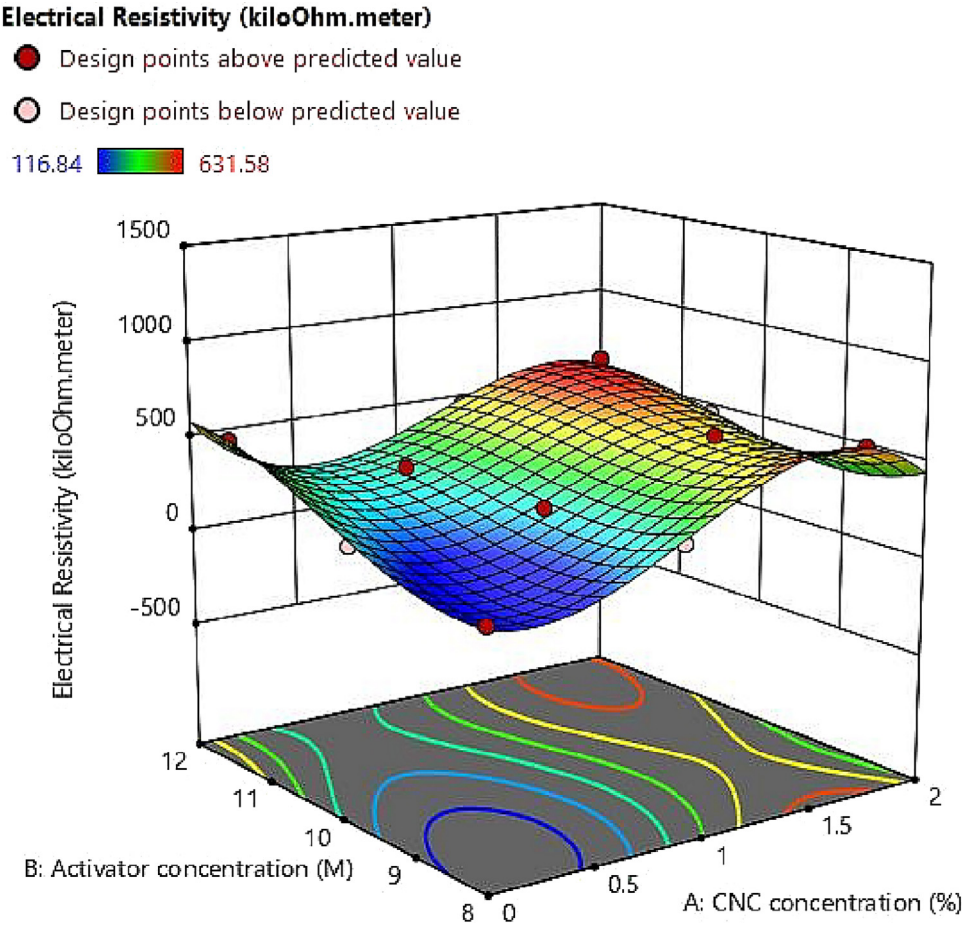


Fig. 16. Electrical resistivity three-dimensional surface response curve (48-hour curing condition).

conditions implied negligible corrosion resistance [29,31,33]. From the perspective of corrosion resistance, it is therefore recommended that the 48-hour curing conditions be implemented in the development of the geopolymers.

4.5. Overall correlation between the mechanical properties

Under certain conditions of cellulose nanocrystals and alkaline activator concentrations, the compressive strength of the geopolymer is inversely proportional to the density. In addition, the geopolymer electrical resistivity is inversely proportional to the concentration of cellulose nanocrystals and directly proportional to the alkaline activator concentration. However, the relationship between the electrical resistivity and cellulose nanocrystals concentration was more pronounced than the relationship between the electrical resistivity and the alkaline activator concentration. Hence, the overall correlation between the mechanical properties of the geopolymers was that the electrical resistivity is directly proportional to the compressive strength, which is inversely proportional to the density.

4.6. Geopolymer development framework

As per the literary recommendation for uniformity in the geopolymer production process [4,17], the empirical framework to aid the development of geopolymer construction materials is recommended below:

1 Identify and quantify the mechanical properties required in the final product.
2 Select the industrial waste aluminosilicate precursor material. Typical cement replacement materials include fly ash, blast furnace slag and silica fumes [1,2,16].
3 Select the alkaline activator. To improve the production economics, consider waste alkaline materials (such as green liquor dregs from kraft pulp mills).
4 Consider the use of organic or inorganic reinforcement agents, such as fibres [14,35,36,37,38,39].
5 Perform a statistical design of experiment to optimize the experimental runs.
6 Select the curing conditions (duration, temperature, and sample rotations). Strive to use ambient temperature curing. However, if this is not possible, try to not exceed 60 °C curing temperature. Observe the effect of the implemented curing temperature on the formed geopolymer samples. If the presence of micro cracks is observed, adjust the temperature and redo step 6.
7 Perform the mechanical testing.
8 Insert the mechanical testing results in the statistical experimental design model and generate the interaction plots and the three-dimensional response plots. Observe the regions of the three-dimensional response plots corresponding to the desired range of the mechanical properties. If the mechanical results do not meet the required specifications, then revert to step 2.
9 If the mechanical results meet the required specifications, the geopolymer development procedure can be accepted.

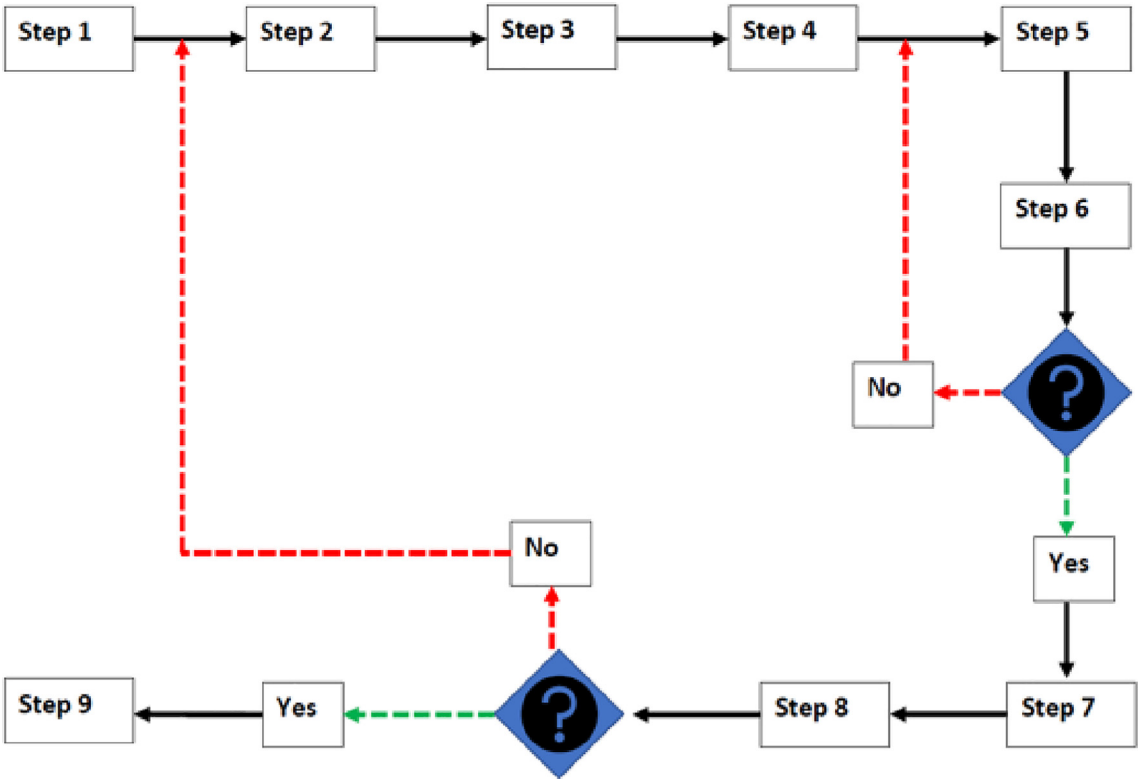


Fig. 17. Empirical geopolymer construction material development framework.

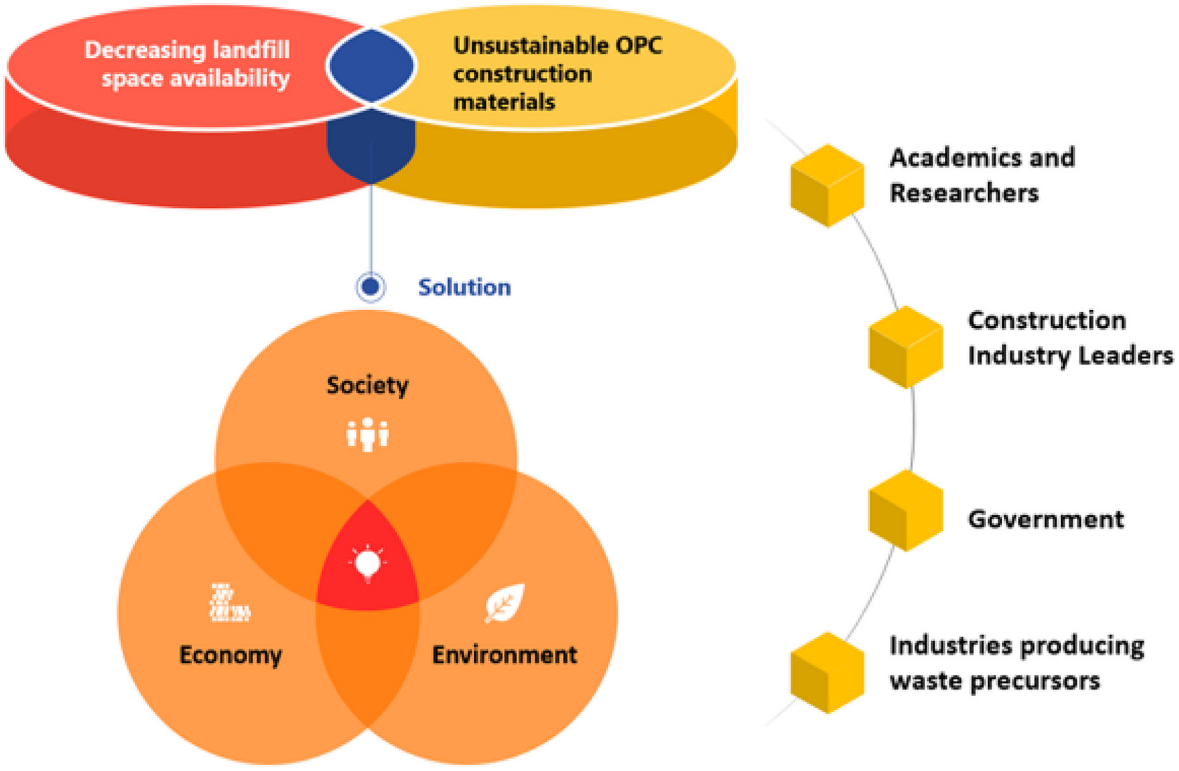


Fig. 18. Conceptual framework for green construction Materials.

A graphical flow diagram is shown in Fig. 17.

5. Conclusions

There is a strong need for novel green construction materials. The methodology reported in this study can be used to efficiently develop geopolymers with a significant degree of statistical accuracy. In addition, the effects of varying cellulose nanocrystals concentrations on the mechanical properties of the formed geopolymers were realized. It is significant that lower concentrations of cellulose nanocrystals (<0.5%) yielded higher strength geopolymers and, in some cases, corrosion resistance. Considering that higher cellulose nanocrystals concentrations prevented the cracking of geopolymers in unstable curing environments, this presented another application for the cellulose nanocrystals. Consequently, the first research aim was met.

The statistical methods enabled the custom development of specific mechanical properties as required by the application. The results of the statistical experimental design yielded the detailed experimental database of the mechanical properties of the cellulose nanocrystals-reinforced fly ash-based geopolymer construction materials in the form of interaction plots and three-dimensional surface response plots. Hence, the second research aim was met.

Due to the experimental optimizations offered by the statistical experimental design and the iterative nature of the proposed universal empirical model, it is recommended for the development of novel green geopolymer construction materials to meet specific mechanical requirements. The results of each iteration can be recorded to guide the inputs of the subsequent iteration until the desired mechanical properties are attained.

In a broader context, the proposed empirical framework can potentially solve the two-fold problems of depleting landfill space and the unsustainability of traditional construction materials (see Fig. 18). The solution is born at the overlap of these problems and entails using aluminosilicate industrial wastes to produce alternative green construction materials per the proposed framework. This approach would yield three sustainability benefits to the environment, society, and economy. The environmental benefit is that industrial wastes (fly ash and sawdust in this case) can be used to produce geopolymer construction materials with relatively minimal greenhouse gas emissions. Secondly, society could benefit from the use of high-quality geopolymer construction materials for housing. Thirdly, the economic benefit is that geopolymers are cheaper to produce than their traditional counterparts and can be easily customized to target specific construction applications.

A strategic partnership is recommended between the key stakeholders: industries producing the precursor waste materials for the green construction materials, academics and researchers, industry leaders of the construction sector and the government. Per this mutualistic partnership, the precursor waste materials can be supplied by the industries producing them to the researchers undertaking the development work. The findings can then be shared with the construction industry leaders for potential implementation and commercialization. Post commercialization, a steady input of precursor waste materials would be required. Government funding can be offered for start-up projects and incentives can be offered to bodies demanding the green construction materials.

The overall conclusion is that the threefold research aims were met, thus supporting the argument that geopolymers are strong contenders in green construction applications.

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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