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Research Article

Exploring Metakaolin and Coconut Shells as Sustainable Alternatives in Concrete: Physical, Mechanical and Microstructural Properties

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ISSN: 2639-0574

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Submission: February 07, 2025

Published: April 07, 2025

Volume 6 - Issue 4

How to cite this article: John Newton Antoh, Humphrey Danso* and Russell Owusu Afrifa. Exploring Metakaolin and Coconut Shells as Sustainable Alternatives in Concrete: Physical, Mechanical and Microstructural Properties. Adv Civil Eng Tech. 6(4). ACET.000645.2025.
DOI: 10.31031/ACET.2025.06.000645

Abstract

One of the most promising solutions to mitigate the depletion rate of non-renewable components in concrete is to partially or completely replace them with renewable plant-based alternatives. The main purpose of this study is to evaluate the synergistic effect of substituting cement with metakaolin and natural aggregate with coconut shells on concrete properties. The novelty of this research lies in the determination of the physical, strength and microstructural properties of varying concentrations of Meta Kaolin (MK) and a constant Coconut Shell Aggregate (CSA). Six groups of concrete test samples were designed and tested, which contained the control, CSA set at 10% with natural coarse aggregate and four different MK contents and constant CSA contents. The density, split tensile strength, compressive strength and microstructural properties were studied. The study found the highest split tensile strength of 3.32MPa, compressive strength of 32.45MPa and density of 2464kg/m^3. There was a decrease in split tensile strength of 6.63-18.98% and compressive strength of 11.02-25.63% as the MK content increased as compared with with the control. The density of concrete decreased to 7.3% with an increase in the MK contents to 20%. Among the concrete with MK and CSA, the synergistic of 5% MK and 10% CSA showed the best physical and mechanical properties. An analysis of the microstructure of concrete with 5% MK and 10% CSA content exhibited a relatively compact microstructure and minimal internal porosity. The study concludes that the synergistic use of 5% MK and 10% CSA is feasible for concrete production.

Keywords: Compressive strength; Coconut shell aggregate; Concrete; Metakaolin; Split tensile strength

Introduction

Worldwide, concrete remains the most widely used building material because of its significant demand in the construction sector. However, the cement production required for concrete contributes to 5-7% of global carbon dioxide (CO_2) emissions, causing negative environmental impacts [1-3]. Davidovits [4] discovered that producing 1kg of cement emits around 0.95kg of CO_2. It is projected that by 2060, the annual consumption of port land cement will rise to 6 billion tons, driven by the increasing demand in the construction industry [5]. To achieve sustainability, reducing these emissions to lessen their detrimental impact on the ecosystem is crucial. One of the main strategies to lower energy consumption and mitigate the effects of cement production on global warming is to decrease the use of energy-intensive cement and natural coarse aggregate. The quest for sustainable materials to substitute cement and aggregate in concrete production will also contribute to Sustainable Development Goals 7 and 11 (SDGs 7 and 11) which advance access to affordable and clean energy and foster the development of sustainable cities and communities. Therefore, partially replacing Ordinary Portland Cement (OPC) with supplementary Cementitious Materials (SCMs) like, ground granulated blast-furnace slag [6], metakaolin (MK) [7], silica fume [8] and fly ash [9], is seen as an optimal solution. This approach can create concrete with an amorphous structure and high hydraulic reactivity in alkaline conditions. SCMs enhance the physical and mechanical properties and the durability of cement when exposed to fire and harsh environments. Moreover, SCMs can help decrease cement-related environmental pollution, including solid waste production, air pollution and water pollution [10]. Metakaolin is obtained by heating kaolin clay to elevated temperatures ranging from 600-800 °C [11-12]. Metakaolin is a highly responsive pozzolanic which generates a significant area of C-S-H and phases containing alumina along with calcium hydroxide ($Ca(OH)_2$) [13]. Metakaolin exhibits even higher pozzolanic activity than silica fume [14] as a result, it can participate in secondary reactions with $Ca(OH)_2$ during the hydration process, producing significant amounts of calcium silicate hydrate (C-S-H) and hydrated calcium aluminate and calcium sulphoaluminate hydrate. Jiang et al. [15] found that the hydration products can decrease the porosity of the concrete and improve the cementitious properties of the aggregates and slurry [16]. Among the various SCMs, incorporating metakaolin in cement can cut CO_2 emissions by up to 170kg per tonne. Furthermore, as an environmentally friendly SCM, metakaolin significantly influences the workability, mechanical strength and durability of concrete. Metakaolin can enhance the mechanical properties of concrete in its early stages [17], effectively addressing the negative impacts caused by traditional mineral admixtures such as ordinary coal fly ash and blast furnace slag. Several studies have been conducted regarding the impact of Meta Kaolin (MK) on concrete mechanical related-properties [18-22]. Shekarchi et al. [23] revealed that incorporating 15% MK replacement led to a 20% improvement in the concrete's compressive strength. Raveendran and Vasugi [24] examined the combined effect of Metakaolin and Nano Silica on concrete's mechanical and microstructure properties. Their study revealed that the synergistic effect between 12.5% MK replacement and 2% Nano Silica resulted in enhanced concrete compressive and split tensile strengths. Bheel et al. [25] investigated the synergistic effect of coconut shell ash, metakaolin and calcined clay on the fresh and mechanical properties of concrete. The study found a rise in compressive, split tensile and flexural strengths at 9% of metakaolin, coconut shell ash and calcined clay. In addition, the study revealed that increasing the MK, coconut shell ash and calcined clay content decreases the water absorption, density and porosity of the concrete. Brooks and Johari [26], Dinakar and Manu [27] and Li and Ding [28] studies concluded that a 10% MK content provided the optimal replacement for cement, resulting in the highest achievable compressive strength. Shehab El-Din et al. [29] revealed that MK can be incorporated into concrete up to 20% without compromising the concrete compressive strength. On the other hand, when combined with glass powder in concrete, a higher MK content of 20% results in a significant reduction in compressive strength [30].

Moreover, Keleştemur and Demirel [31] and Bhat and Naqash [32] indicated that replacing cement with MK up to 15% could enhance corrosion resistance, as well as provide protection against acid and sulfate attacks on the concrete. Substituting cement with MK has been proposed as a method to improve the transport properties of concrete, such as water penetration ionic diffusion gas permeability and water absorption. Courard et al. [33] analyzed the transport properties and chemical behavior of mortar modified with MK. Nawab et al. [34] assessed the physical and mechanical properties of mortar with coconut fibers, silica-fume and metakaolin as cement replacement and found that the combination of 10% silica fume 10% metakaolin and 6% coconut fibers showcased superior mechanical and physical properties. The study showed that the migration coefficient of concrete decreased by 170% when MK was substituted in amounts between 10 and 15%. Additionally, a careful examination after a year revealed no diffusion in concrete containing 20% MK. Research conducted by Poon et al. [35] and Gruber et al. [36] confirmed these findings by demonstrating that adding 5-20% MK to concrete reduced chloride diffusion and penetrability. However, Xupeng et al. [37] found that the presence of excessive MK (above 15%) can lead to agglomeration and cause the surface aggregates to peel off by creating a loose and porous micro-morphology.

Coconut shells represent a significant agricultural waste generated in numerous countries. On average, a coconut tree yields around 70 fruits each year, with the shells comprising roughly 15% of the fruit's total mass [38]. The coconut shell, situated between the husk and the central endosperm of the coconut fruit, serves as its tough outer covering. Due to its slow degradation, disposing of coconut shells can pose environmental challenges. However, due to its durability and resistance to biodegradation, coconut shells are viewed as a viable coarse aggregate for concrete production [39]. According to Nadir and Sujatha [40], the bulk density and specific gravity of coconut shells were found to be 650kg/m^3 and 1.15 respectively. In addition, Gunasekaran et al. [41] found that the average water absorption and moisture content of the coconut shell was 24.0% and 4.20, respectively. An experimental study revealed that the impact value and aggregate crushing value of Coconut Shells (CS) are 3.94% and 1.6, respectively [41]. it can be inferred that CS demonstrates superior resistance to impact and crushing. Coconut shells contain high lignin content making the composite more weather-resistant and less moisture-absorbent due to the low cellulose content compared to other agricultural waste [42]. Sugar significantly retards the setting and hardening of concrete. However, the sugar present in coconut shells does not impact the setting and strength of concrete, provided they are present in free form [43]. The smooth surface on the concave side enhances workability and binds with the concrete matrix without leaching [43]. Maheshwaran et al. [44] found that coconut shells contain amorphous silica which enhanced their bonding with the cement paste. Ting et al. [45] examined the physical and chemical properties of coconut shells. Their study revealed that CS are partly soluble in water and hygroscopic, because of the open structures containing many hydroxyl and acetyl groups. Kumar et al. [46] found that incorporating coconut shell aggregate in concrete reduces the workability of concrete due to the absorption of more water present in the mix for concrete preparation. This can be inferred that excessive content of water will weaken the structure of CS [45]. In addition, Tomar et al. [47] found that the flakiness of CS aggregates reduces the movement of aggregates hence reducing the workability. Numerous studies have explored the impact of incorporating Coconut Shell Aggregate (CSA) into concrete, with a specific focus on its influence on mechanical properties. There have been extensive discussions on the subject by [48-52]. In the studies [53-54], the authors found that the optimal percentage for replacing part of the coarse aggregate with coconut shell is determined to be up to 10%. In the study by Bhoj et al. [50], the authors successfully produced concrete without any performance deterioration by replacing 15% of the natural coarse aggregate with CSA.

Additionally, Tangadagi et al. [55] found through experimental studies that up to 20% of CSA can replace natural aggregate without significantly compromising strength characteristics. Reddy et al. [56] asserted that CSA can be integrated into concrete applications at levels of 25% or lower without compromising the mechanical properties of concrete. Hasan et al. [57] researched the rheological and strength characteristics using 10-50% coconut shell aggregate as a replacement for natural coarse aggregate in concrete. The study revealed that the strength values showed a slight reduction with CSA replacements of up to 20% with increasing percentages of CSA. Bhoj et al. [50] study concluded that a 15% CSA content demonstrated a 24% reduction in strength compared to conventional concrete. Additionally, Maheshwaran et al. [44] investigated the micro-structural characteristics of coconut shell-based concrete. They revealed that coconut shell concrete contains no major interfacial transition zone which is responsible for the lower strength properties compared to conventional concrete. Liu et al. [58] examined the micro-structural properties of biomass recycled concrete made using coconut shells. The study revealed the presence of loose C-S-H gels and pores adjacent to the coconut shells which contribute to reduction in strength. Scanning Electron Microscopy (SEM) analysis of the microstructure of CS revealed that the specimens consist of closely spaced discrete cells ranging from 16.36 to 29.33μm in size, with micropores varying from 760.6nm to 1.64μm within these cells. Additionally, some long, continuous chain-linked cells with widths between 7.35 and 8.88μm were observed. These discrete and chain-linked cells are likely responsible for the enhanced impact, crushing and abrasion resistance of the CS specimens [43].

Despite the increasing interest in using CSA as partial replacement material in concrete due to its high lignin and low cellulose content, minimizing agro-waste disposal issues and durability properties, CSA is not capable of filling the pores in concrete, which in turn leads to instability that affects the strength and micro-structural properties of the concrete. In this context, the incorporation of MK with micro-pore filling and high pozzolanic properties into concrete has emerged as a suitable option that garnered the attention of researchers to produce concrete with improved strength, density and microstructure properties.

Substantial information exists in the published literature regarding concrete's physical, mechanical and durability properties with either MK or CSA individually added. However, inadequate studies are addressing the combined effect of MK as a substitute for cement and CSA as a substitute for coarse aggregate on the mechanical, physical, chemical composition and microstructural properties of concrete. For instance, Panda [59] investigated the workability with the use of a slump test and some strength properties of concrete prepared with coconut shells and metakaolin in India. The aforementioned study focuses on addressing the combined effect of MK and CS on the mechanical and workability properties of concrete. There is, therefore, a need to clarify whether the synergistic of metakaolin and coconut shells in concrete sourced from a developing country such as Ghana, can maintain the same performance under compressive and tensile stresses and establish the chemical composition and microstructural analysis of the concrete. The novelty of the present research is to study the effect of varying the concentration of MK and a constant coconut shell aggregate on the density, strength and microstructural properties of the concrete with a cement-binder ratio of 0.5.

The purpose of this study is to evaluate the synergistic effect of substituting cement with metakaolin and natural aggregate with coconut shells on the physical, mechanical and microstructural properties of concrete. The study has practical implications for the construction and engineering industries. This can support practitioners in making informed decisions regarding using CSA and MK in concrete mixtures. Replacing cement with MK and coarse aggregate with CSA in concrete production will reduce cement and coarse aggregate production and therefore intensify reductions in the generation of CO_2 emissions in the environment and promote sustainable construction practices. The study used experimental research design to ascertain the density, split tensile and compressive strengths and the relationship among them as well as the microstructural properties and chemical composition. Six series of concrete test samples, which contained the control, CSA set at 10% with natural coarse aggregate and four different MK contents and constant CSA contents, were designed and tested.

Materials and Methods

Materials

The study utilized cement manufactured in Ghana conforming to BS EN 197-1 [60] as a binder, pit sand possessing the following properties was used as a fine aggregate: Absorption Capacity (AC)=3.65% and Specific Gravity (SG)=2.67.20mm. Crushed granite having the following properties was considered as conventional aggregates: SG=2.60 and AC=3.83%. The kaolin utilized in this experimental study was procured from Mfensi, a locality in Kumasi within the Ashanti Region of Ghana. Endene et al. [61] and Yaya et al. [62] assessed the Mfensi clay deposit's suitability as a construction material. The study indicated that silica, aluminum and iron were the predominant oxides in the chemical compositions of Mfensi clays making it a suitable pozzolana material to replace cement in concrete. Additionally, the study identified, kaolinite as a mineral in the clay deposits. After air-drying, the kaolin underwent calcination at 700°C to produce metakaolin, as outlined by Rashad [63]. The resulting metakaolin was cooled, ground using a pestle and mortar and sieved through a 75μm British Standard sieve size. The properties of the metakaolin used as partial cement replacement are SG=2.52 and Fineness Test (FT)=3.5. Figure 1a visually represents the metakaolin used as cement substitution in the present study. Waste Coconut Shells (CS) sourced from Roman

Hill in Kumasi, Ghana, underwent a preparation process. The outer part (the husk) of the coconut shells was removed and the shell was subjected to sun-drying for 8hrs before manual crushing. The shells were then broken into smaller sizes using a hammer. The crushed shells underwent a washing process and were then left to dry naturally at ambient temperature conditions for an additional 8 hours. The particle size range of the coconut shells as shown in Figure 2b for utilization in concrete. Before being used as aggregate, the coconut shells were submerged in water for 24hrs. They were conditioned to achieve a Saturated Surface Dry (SSD) state before mixing. An SSD condition prevents the aggregate particles from absorbing water during mixing. Figure 1b shows a sample of these processed coconut shells aggregate.

Preparation of concrete specimens

The proportions 1:1.94:2.91 used in this study were adapted from existing literature [64]. A concrete grade strength of 20MPa with a target strength of 26.6MPa was used for the control mixes, binary mixes with 10% CS with coarse aggregate by weight and ternary mixes comprising 10% CS and 5-20% MK at 5% intervals replacement with natural aggregate and cement by weight, respectively. These mixes were prepared and cured at intervals of 7, 14, 28 and 56 days. MK replacement levels ranged from 0 to 20% with 5% intervals, while CSA remained constant at 10% of coarse aggregate replacement. Coconut shell aggregate was set at 10% of natural coarse aggregate is justified, as it does not significantly compromise strength characteristics, as reported in previous studies [59, 64]. The design mix proportion for control, MK and CSA specimens can be found in Table 1. Concrete was mixed in a pan mixer per ASTM C192/C192M-19 [65]. The pan mixer was first filled with premeasured sand, metakaolin and cement. After the mixer was rotated for about a minute, coconut shell and coarse aggregate were added and the mixture was then mixed for 2min before adding the requisite amount of water. The pan mixer was left to rotate for two more minutes to obtain a homogenous mixture. The freshly prepared concrete was then poured into 100x200mm steel cylinder steel molds and 100x100x100mm steel cube molds (see Figure3) conforming to ASTM C192 [66]. To eliminate trapped air, the molds were vibrated using a table vibrator. Ninety (90) concrete specimens were prepared for this study, 45 cylinders and 45 cubes. The samples were allowed to set for 24hrs and cured at intervals of 7, 14, 28 and 56 days by full water immersion. The concrete cubes and cylinders were cleaned with an absorbent towel to attain an SSD state after curing. Before the planned tests were carried out, the concrete samples were weighed to ascertain their densities.

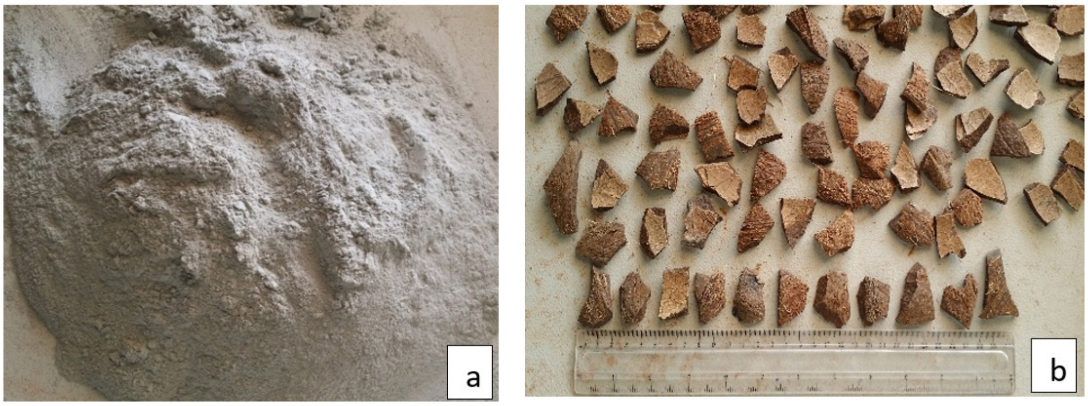


Figure 1: Materials for concrete production: (a) metakaolin used as cement, (b) coconut shells used as coarse aggregate.

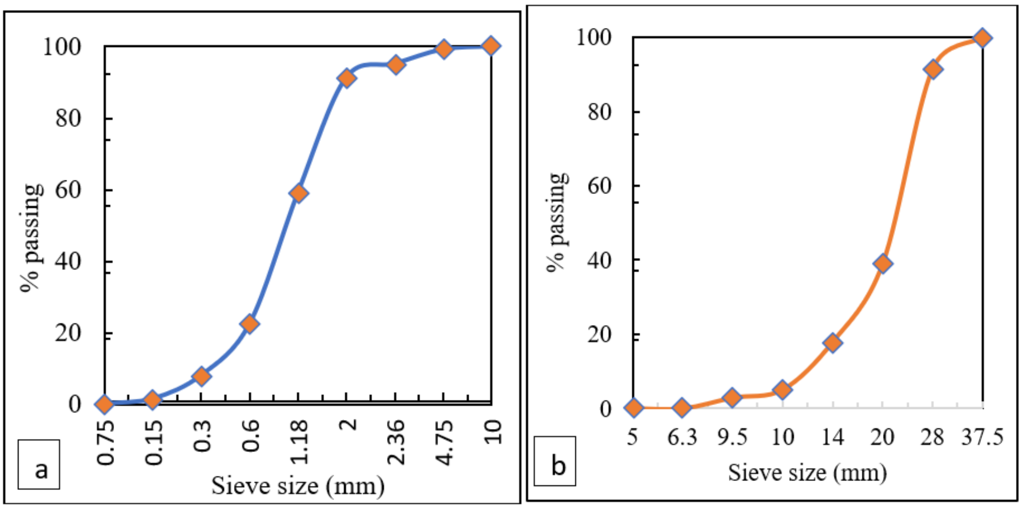


Figure 2: Particle size distribution: (a) sand, (b) CSA.

Figure 3: Moulded concrete speciemens: (a) cube speciemens, (b) cylindrical specimens.

Table 1: Proportions of concrete constituent kg/m^3.

Mix design	MK Rep. (%)	CSA Rep. (%)	Cement (kg)	MK (kg)	Sand (kg)	Gravel (kg)	CSA (kg)	Water (kg)
M0C0	0	0	367.27	0	712	1068.76	0	202
M0C10	0	10	367.27	0	712	961.88	106.88	202
M5C10	5	10	348.91	18.36	712	961.88	106.88	202
M10C10	10	10	330.54	36.72	712	961.88	106.88	202
M15C10	15	10	312.18	55.09	712	961.88	106.88	202
M20C10	50	10	293.82	73.45	712	961.88	106.88	202

Test procedures

Density test

The concrete specimens were tested at intervals of 7, 14, 28 and 56 days of curing. The cube specimens were used to test dry density. The dry density test was conducted on the cube specimens and determined in accordance with BS EN 12390-7 [67]. Three specimens from each mix were selected and weighed using the electronic weighing balance after drying in an oven at a temperature of 105 °C for 24 hours and their sizes were measured before. The specimens' weight and volume were determined and used to calculate the dry density. The density of the concrete was evaluated by the ratio of the weight of the cube to the volume of the cubes.

$$Dry\,Density\,(kg/m^3) - \frac{Weight\ of\ cube\,(kg)}{Volumeofcube(m\hat{}3)} \qquad (1)$$

Compressive strength test

The compressive strength test was carried out on 7, 14, 28 and 56 days, following the guidelines provided by the British Standard Institute BS EN 12390-3 [68]. Three concrete cubes were selected from each mix design for the test. The concrete cubes were dusted off after removing from the curing tank before being put into the crushing machine and each cube specimen was weighed separately. The test was conducted using a non-automated compression testing machine with a 2000KN capacity. The load was applied until the concrete cubes failed (Figure 4a) and the load at failure was noted. Figure 4b shows the cube failure. Equation 1 was utilized to ascertain the compressive strength.

$$Compressive\,Strength\,(N/mm^2) = \frac{Crushing\ Load\,(n)}{Effective\ Area\,(mm^2)} \qquad (2)$$

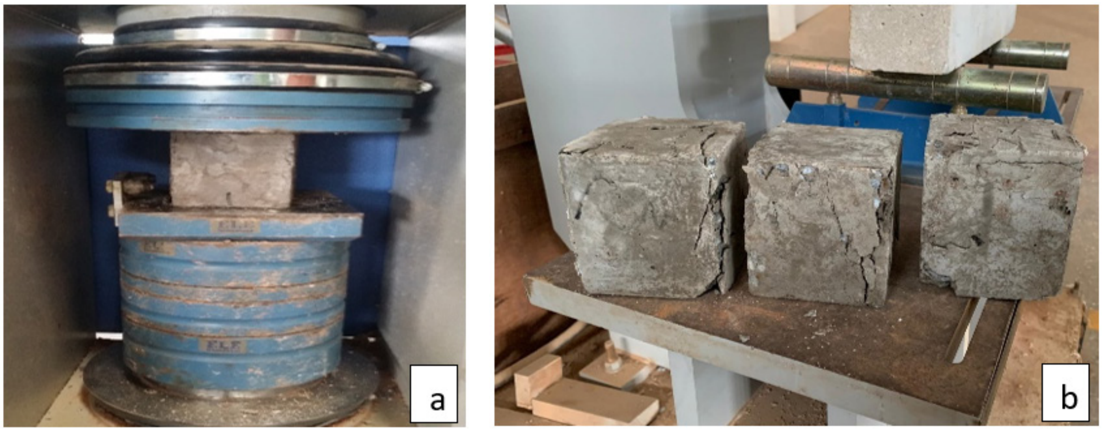


Figure 4: Compressive test process: (a) compressive test setup, (b) failed specimen.

Split tensile strength test

The split tensile strength test complied with the requirements in BS EN 12390-6 [69]. Three cylindrical concrete specimens were selected from each mix design for the test. A splitting jig was positioned centrally beneath and above each cylindrical concrete specimen. The test was conducted using a non-automated 2000KN capacity compression testing machine. The load was applied to the specimen until the concrete cylinder split into two (Figure 5a). In Figure 5b, the failed specimens are displayed. Equation 2 was utilized to ascertain the split tensile strength.

$$Split\,Tensile\,Strength\,(N/mm^2) = \frac{2P\,(N)}{\pi LD\,(mm^2)} \quad (3)$$

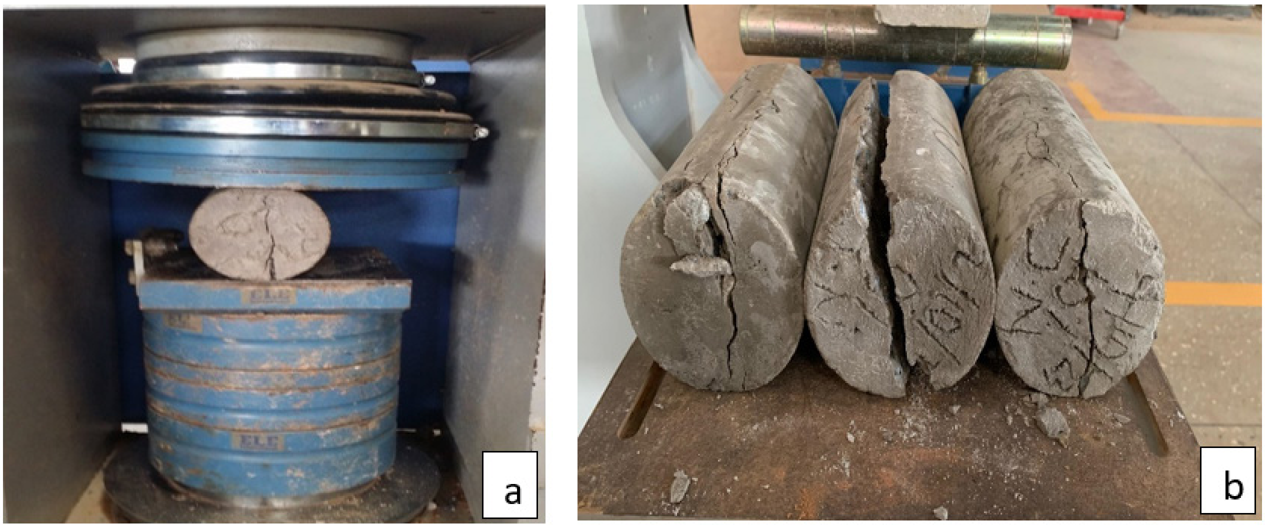


Figure 5: Split tensile test process: (a) split tensile test setup, (b) failed specimens.

EDS/SEM Analysis

Image analysis with Scanning Electron Microscopy (SEM) was performed following ASTM E1508-98 [70]. The study utilizes the Phenom-World ProX desktop scanning electron microscope situated at the Department of Earth Science, University of Ghana, Legon. To withstand vacuum conditions and high-energy electron beams, the samples were prepared in a solid, compact, dry state, devoid of any dust particles. Precision in sample size was ensured by careful cutting and trimming to fit the specimen stage, followed by mounting onto an aluminum stub using pelco double-sided carbon adhesive. An ultra-thin coating of gold was sputtered onto the samples with poor or no conductivity. Optical images were captured at the lowest magnification of 20x. Backscattered images were captured at different magnifications (minimum to maximum) using an image intensity and high-resolution voltage mode of 10kV using the backscatter detector until the best image focusing ends. Further, using the Phenom ProSuite software (element identification), the Energy Dispersive Spectroscopy (EDS) point analysis at 15Kv, duration of 30 seconds and map analysis at 15Kv, duration of 4 minutes 26 seconds were used for the elemental identification, distribution and concentration respectively.

Statistical analysis

After 56 days of curing, a One-Way ANOVA analysis was performed to evaluate if there were any statistically significant differences in split tensile and compressive strengths. Tables 2 and 3 display the obtained outcome.

Table 2: One-way repeated measures analysis of variance (ANOVA).

Compressive Strength (MPa)					
	Sum of Squares	df	Mean Square	F	Sig.
Between Groups	102.529	5	20.506	2.259	0.115
Within Groups	108.95	12	9.079		
Total	211.479	17	-	-	-

Table 3: One-way repeated measures analysis of variance (ANOVA).

Split Tensile strength (MPa)					
	Sum of Squares	df	Mean Square	F	Sig.
Between Groups	0.7	4	0.175	2.391	0.12
Within Groups	0.732	10	0.073		
Total	1.432	14			

Results and Discussions

Density of concrete

From Figure 6, it can be seen that with an increase in the percentage of MK as a replacement for cement, the density of the concrete decreases. The densities obtained were 2348, 2379, 2356, 2323 and 2285kg/m^3 for M0C10, M5C10, M10C10, M15C10 and M20C10, respectively. The control specimen recorded the highest density of 2464kg/m^3, followed by 2379kg/m^3 for the synergistic of cement with 5% MK replacement and CSA set at 10% with the natural aggregate of. The lowest density of 2285kg/m^3 was recorded by the synergistic of cement with 20% MK replacement and CSA set at 10% with natural aggregate. In our study, the decrease in density obtained was 4.71% for 10% of CSA instead of part of the natural coarse aggregate compared with the control sample. In Stelmakh et al. [54] the authors managed to obtain a 9.1% decrease in density with 10% of CSA introduced instead of part of the coarse aggregate. It is established in Bhoj et al. [50] and Tangadagi et al. [55] that the density of concrete decreases with replacement of CSA content.

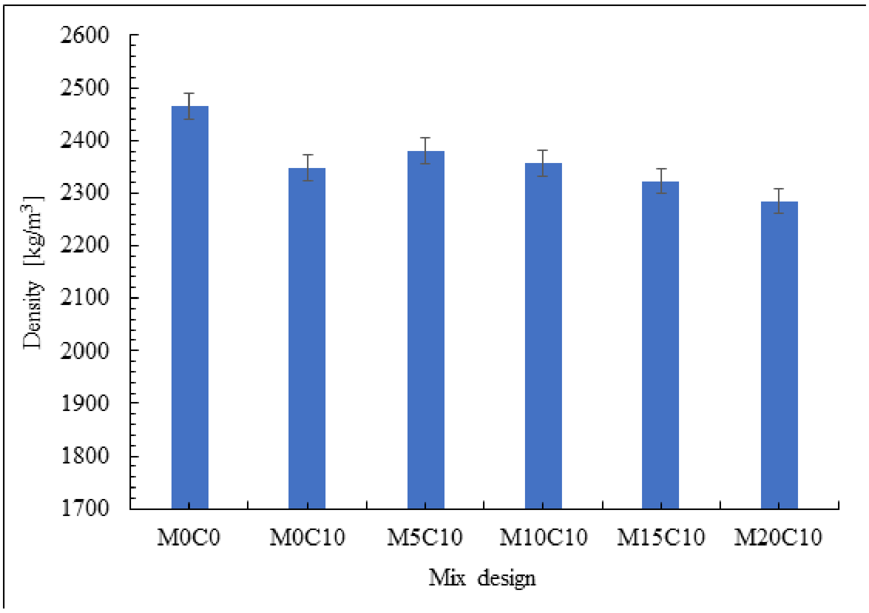


Figure 6: density of metakaolin concrete.

However, the introduction of cement with 5-10% MK to the concrete specimen with 10% CSA replacement with natural aggregate showed improved density compared with the M0C10 concrete samples. Moreover, incorporating cement with 15-20%MK into the concrete specimen with CSA set at 10% with natural aggregate decreased the density compared with the density of M0C10 concrete samples. On the other hand, the synergistic of cement with 5-20% MK and CSA set at 10% with natural decreased the density as cement's replacement with MK increased compared with the control samples. In the current study, the reduction in density of the synergistic of MK and CSA concrete compared with the control sample was 3.4, 4.4, 5.7 and 7.3% for M5C10, M10C10, M15C10 and M20C10, respectively. This observation suggests that the synergistic of MK and CSA in concrete decreased its density. This observation may be due to the significance of the characteristics and properties of MK compared with cement. MK has a lower specific gravity compared with cement and these may have contributed to the decrease in density. Prakash et al. [71] found a reduction in density as the replacement of MK increases. Their study attributed the reduction in density to MK having lower specific gravity than OPC. These findings were verified in the study [12]. In addition, the decrease in density could be attributed to the presence of lightweight porous coconut shell aggregate, because CSA has a significantly lower specific gravity than conventional coarse aggregate. Similar findings were reported in Bheel et al. [55]. In this current study, the densities obtained are more than 1900kg/m^3 and satisfied the density for normal-weight concrete as per ACI 318-14 [72].

Compressive strength of concrete

Figure 7 illustrates the compressive strength of all the concretes for 7, 14, 28 and 56 days of curing. The compressive strength of all the concrete increased with curing age. The test results revealed that CSA set at 10% of natural coarse aggregate is justified, as it does not significantly compromise strength characteristics. For example, in [54,71], an amount of CSA up to 10% was considered to be optimal. The 56-day compressive strength of the concretes was between 25.28-32.45 MPa. The highest strength was for the control concrete and the lowest strength was for the M0C10 concrete sample. In our study, the decrease in compressive strength when CSA is set at 10% with natural aggregate without MK replacement was 22.09% compared with the control sample. In [51,58,73], the compressive strength decreased to 25% when 10% CSA was used to replace natural coarse aggregate. Moreover, the drop in compressive strength in the current study 22.09% was greater than in Stelmakh et al. [54], which may be due to differences in characteristics of the coconut shell, proportions of the mixture and the density as well as the control composition of concrete. The decrease in the strength characteristics of concrete when 10% CSA is set to replace natural coarse aggregate could be attributed to the properties of CSA. CSA has significantly lower strength compared to natural coarse aggregate, this results in a notable reduction in the concrete strength of all specimens containing CSA [55,71]. In addition, the inter-particle bond plays a less crucial role in CSA concrete in strength development due to the relatively smooth surface of the CSA aggregate resulting in a decreased strength. From the results, it can be established that the introduction of cement with 5-20% MK content to concrete with a CSA content set at 10% which was used instead of a part of natural aggregate showed improved strength at 56 days as compared with the M0C10 samples. The slight increase in strength may be due to the small amounts of water absorbed by CSA during the production of the concrete mixture, then, during the hardening of the cement composite, this water acts as a reserve, intensifying the hydration process at the phase boundaries. Moreover, the strength gains at 56 days of curing for M5C10, M10C10, M15C10 and M20C10 compared to the M0C10 specimen could be attributable to the micropores filling and pozzolanic reactivity of MK, as it gains strength over time. In addition, the strength gain at 56 days could be ascribed to the coarse texture of the coconut shell, fostering strong cohesion with the matrix. In addition, this may be due to the high coefficient of adhesion of CSA to the cement-sand matrix of concrete, which significantly compacts the material at the interface of the mortar matrix. However, the study revealed that the compressive strength of M5C10, M10C10, M15C10 and M20C10 decreased as compared to the control across all curing periods. This observation suggests that the synergistic effect of cement with 5-20% MK at 5% intervals and CSA set at 10% of natural coarse aggregate systematically reduces the compressive strength as the MK percentage increases. The reduction in compressive strength at 56 days of curing for M5C10, M10C10, M15C10 and M20C10 was 11.02, 12.71, 17.87 and 25.63% for M5C10, M10C10, M15C10 and M20C10 as compared with the compressive strength of the control. Panda [59] recorded a 13.20% decrease in strength at 56 days when 10% of MK and CSA were replaced with cement and natural aggregate, respectively in concrete. The reduction in strength on the synergistic effect of MK and CSA concrete could be attributed to a decrease in cement hydration, as MK and CSA possess lower cementing properties compared to OPC resulting in a decrease in strength. Similar findings were reported by [25], their study revealed that when more than 9% of MK, CC and CSA are used as SCMs, the cement hydration decreases resulting in a decrease in its strength properties. Additionally, the drop in the strength properties of concrete with the introduction of CSA is associated with the properties of this material. CSA has a significantly lower strength than natural aggregate and is also lighter. Moreover, Samson [30] found that the combined effect of cement with 5-20% MK at 5% intervals and Powdered Glass (PG) set at 10% as SCMs decreases the compressive strength as MK content increases. The study further revealed that the decrease in compressive strength is a result of MK slowing down the early hydration of OPC, thereby affecting the rate at which concrete gains strength. In additionally, in addition, Subaşı and Emiroğlu [74] revealed that replacing cement with 5-30% MK by weight at 5% intervals decreases the compressive strength as MK replacement increases. According to Subaşı and Emiroğlu [74] two factors contribute to the decrease in compressive strength: first, substituting cement with MK replacement in concrete reduces tri-calcium silicates, a key compound contributing to the strength and second, MK fineness is lower than that of OPC and as a result, it cannot hydrate as rapidly as cement within the initial 28 days, leading to slower development of the desired strength compared to plain OPC concrete. Moreover, M5C10 concrete achieved better results than the targeted compressive strength of 26.6 MPa at 28 days. Further, the compressive strength of M5C10, M10C10, M15C10 and M20C10 specimens at 28 days was approximately 6.4-22.7% higher than the minimum requisite strength of 21 MPa for structural normal-weight concrete as per ASTM C330 [75]. In this study, the optimal percentage of replacing a part of the cement with MK in concrete with CSA set at 10% with natural aggregate is considered to be an amount of up to 10%. This allows for the production of high-quality concrete without a significant loss of strength. For example, in Panda [59] an amount of MK and CSA up to 10% was also optimal.

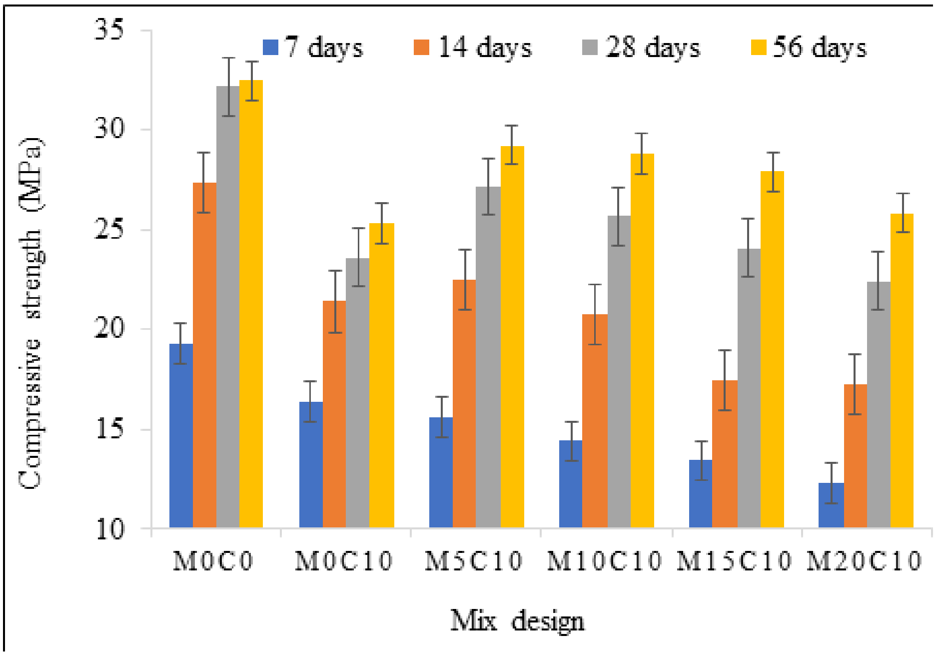


Figure 7: Compressive strength of MK and CSA concrete.

Additionally, after 56 days of curing, a One-Way ANOVA analysis was performed following Danso and Appiah-Agyei [76] to evaluate if there were any statistically significant differences in compressive strengths. Table 2 displays the obtained outcome. Although the compressive strength of MK and CSA specimens were marginally lower than the control specimens, at the 5% significance level, there was no statistically significant difference (F=2.259; p=0.115) between the compressive strengths of the control and all MK and CSA concrete (ρ>0.05).

Correlation between compressive strength and density

Figure 8 depicts the 56-day correlation test conducted between compressive strength and density. It reveals a notably linear positive relationship. The coefficient of determination (R^2) calculated is 0.7776 revealing a strong association. This suggests that the rise in compressive strength of the control specimen can be credited to the heightened density while the decrease in compressive strength of MK and CSA specimens was a result of the decrease in density. As seen in Figure 8. When one of these properties is identified, the formulas in Figure 8 can be used to evaluate the density or compressive strength. The finding in this study is comparable to what was revealed by Bheel et al. [25] and Prakash et al. [71]. In addition, Walker [77] revealed that compressive strength and density were positively correlated.

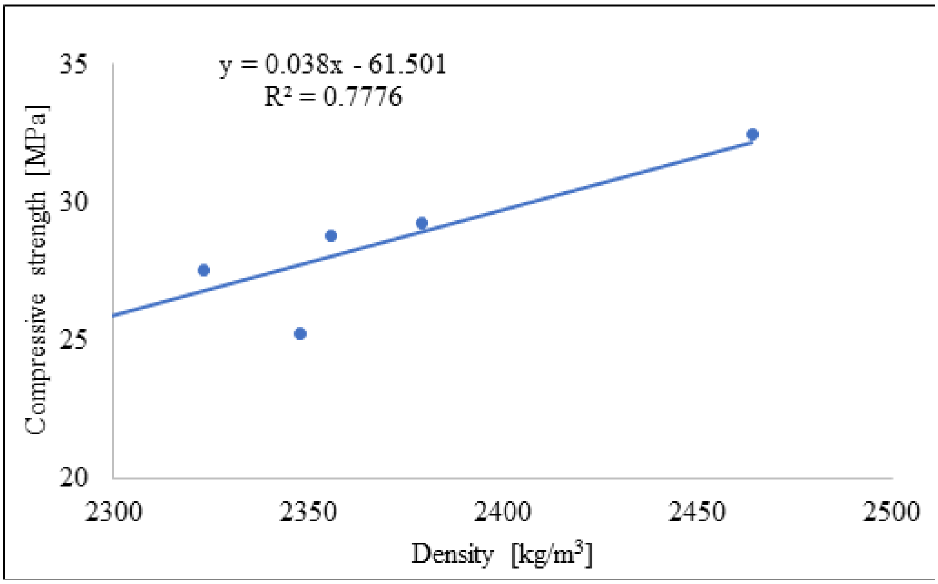


Figure 8: Relationship between compressive strength and density.

Split tensile strength of concrete

Figure 9 revealed the results of split tensile strength for various concrete mixes incorporating cement with 5-20% MK and CSA set at 10% with natural coarse aggregate. As expected, extension curing days increased the split tensile strength of synergistic of MK and CSA concrete specimens. At 28 days of curing the split tensile strength of the control specimens was 3.24 MPa, compared to 2.53, 2.81, 2.78, 2.64 and 2.38 MPa for M0C10, M5C10, M10C10, M15C10, M20C10, respectively. At 56 days of curing the split tensile strength of the control specimens was 3.32 MPa, compared to 2.62, 3.10, 2.91, 2.86 and 2.69 MPa for M0C10, M5C10, M10C10, M15C10, M20C10, respectively. From Figure 9 it was established that the highest split tensile was recorded by the control specimen and the lowest split tensile strength was recorded by concrete with a CSA content of 10% which was used instead of a part of natural aggregate without MK replacement. The reduction in the split tensile strength at 56 days of curing, was 21.08, 6.63, 12.35, 13.86 and 18.98% for M0C10, M5C10, M10C10, M15C10 and M20C10 as compared to the control specimens. Panda [59] managed to obtain a minimum decrease in split tensile strength of 3.32 and 2.25% after 28 and 90 days respectively for 10% MK and 10% CSA concrete samples. From the results, it can be established that the introduction of cement with 5-20% MK content to concrete with a CSA content set at 10% which was used instead of a part of natural aggregate showed improved strength compared with the M0C10 concrete sample. This observation may be due to MK's interaction with the calcium hydroxide released during pozzolanic reactions, leading to the formation of C-S-H and thereby enhancing strength [78-80]. Additionally, this can suggest that the MK introduced as a replacement for cement and CSA as a replacement for part of the natural aggregate has a fairly significant degree of adhesion to the mortar part and acts as an additional sealing component in the cement-sand mortar coarse aggregate. This is also in good agreement with [54,81]. However, the synergistic effect of M5C10, M10C10, M15C10 and M20C10 showed a slight decrease in split tensile strength as compared to the control specimen. The reduction in strength properties with the introduction of CSA set at 10% of natural aggregate and cement with 5-20% MK is linked to the properties of these materials. CSA has significantly lower strength compared to natural coarse aggregate and is also lighter. This inferred that incorporating CSA into heavy concrete will inevitably result in a reduction in strength characteristics. This is in good agreement with other studies [50,82]. ASTM C330 [75] specifies that the minimum requisite split tensile strength for structural normal-weight concrete is 2.1MPa. The synergistic effect of cement with 5-20% MK and CSA content set at 10% of natural coarse aggregate concrete achieved better results than the minimum split tensile strength specified by ASTM C330 [75]. The split tensile strengths of the synergistic effect of cement with 5-20% MK and CSA content set at 10% of natural course at 28 days were 11.8-25.3% higher than the minimum requisite strength of 2.1MPa for structural normal-weight concrete. Additionally, a one-way ANOVA test was performed to determine whether there is a statistically significant or no significant difference between the split tensile strength of the control specimen and 5-20% MK with 10% CSA specimen at 56 days. The results and calculations are presented in Table 3. The study revealed that the split tensile strength of the concrete with MK and CSA replacement was lower than that of the control specimen. However, at the 5% level of significance, no statistically significant differences were found (F=2.391; p=0.120) between the tensile strengths of the control specimen and the concrete containing MK and CSA (ρ>0.05).

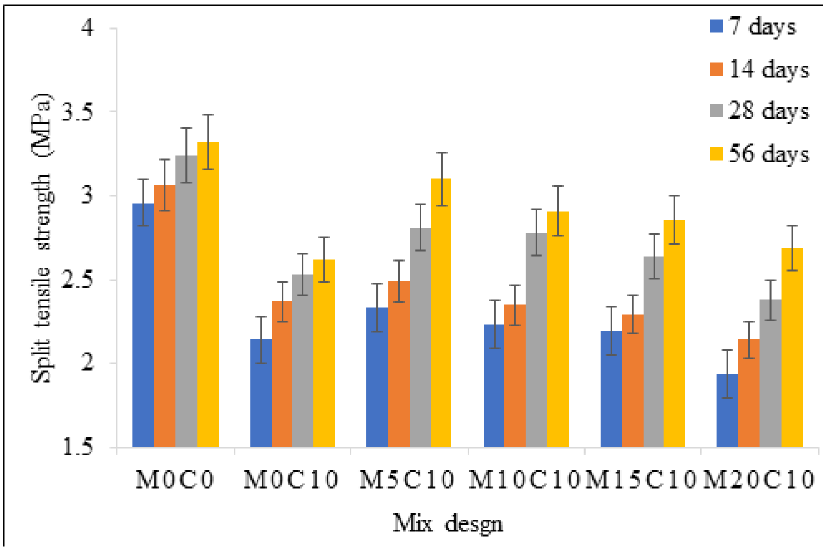


Figure 9: Tensile strength of MK and CSA concrete.

Correlation between compressive and split tensile strengths

Figure 10 summarizes the association between the tensile and compressive strengths of MK and CSA concrete cured at intervals of 7, 14, 28 and 56 days. The study revealed that the tensile and compressive strengths of each specimen group had a strong and positive linear relationship. The coefficient of determinants (R^2) obtained are 0.87, 0.88, 0.92, 0.96 and 0.97 respectively, for 0, 5, 10, 15 and 20%MK addition. This suggests that as the compressive strength rises, so does the tensile strength. The observations in our study are in good agreement with the studies reported in Bheel et al. [25] and Stelmakh et al. [54]. In addition, the correlation between increased compressive and tensile strength aligns with findings from previous studies [21,83-84]. Consistent trends were also observed in earlier studies [85-86].

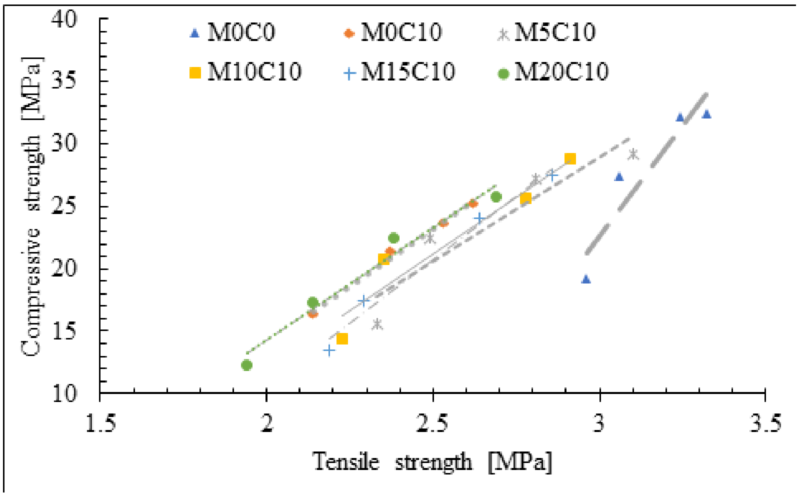


Figure 10: Relationship between tensile strength and compressive strength.

Microstructural and chemical composition analysis of concrete

Figures 11 and 12 present the results of synergistic of 5%MK and 10% CSA (M5C10) concrete specimens that underwent Scanning Electron Microscopy (SEM) and Energy Dispersive Spectroscopy (EDS), respectively. Figure 11 clearly illustrates the porous structure of the CS, into which the cement paste penetrates, thereby creating good adhesion of this aggregate into the cement-sand matrix. this observation inferred that the rough and porous surface of coconut shells indicates a favorable characteristic of a composite material. Similar results were reported in a previous study [76]. The combined MK and CSA concrete comprises the following chemical elements by weight percentage: oxygen (O) 50.67%, Sulphur (S) 0.28%, calcium (Ca) 10.43%, silicon (Si) 7.93%, carbon (C) 29.10%, aluminum (Al) 0.83%, iron (Fe) 0.31%, sodium (Na) 0.17% potassium (K) 0.05% and magnesium (Mg) 0.24%. The identified oxide content in combined MK and CSA concrete consists of Ca (38.67%), Si (31.49%), Al (2.81%), Fe (1.61%), Na (0.34%), S (1.18%), K (0.16%) and Mg (0.63%). The EDS analysis indicates that the material was mainly composed of O, Ca, Si and S. Therefore, it can be inferred that the hydration product formed is C-S-H. From Figure 11, the microstructure of M5C10 is more compact, this is attributable to the comparatively reduced number of pores. Additionally, most of the anhydrate particles and hydration products were closely connected resulting in a more compact structure. Similar findings were reported in previous studies [32,80]. Additionally, the absorption of a small amount of water by CSA particles during the hardening process intensifies hydration at the phase boundaries. This factor enables the concrete to maintain strength characteristics comparable to ordinary concrete when small percentages of this organic aggregate are used, effectively compensating for the lower strength of coconut shell relative to natural coarse aggregate. Figure 12 reveal a large quantity of calcium present in the matrix compared to aluminum and silica content, this can contribute to reduced durability. Additionally, the high percentage of oxides, specifically the Al/Si ratio of 0.11 and Ca/Si ratio of 1.23 are both within the limit of the cement-based matrix specified in the studies of Danso [87] and Kunther et al. [88], is responsible for the formation of calcium-silicate-hydrate in the cement-based mortar [89]. Incorporating 10% CSA and 5% metakaolin can enhance the internal microstructure of the test sample, thereby reducing pores.

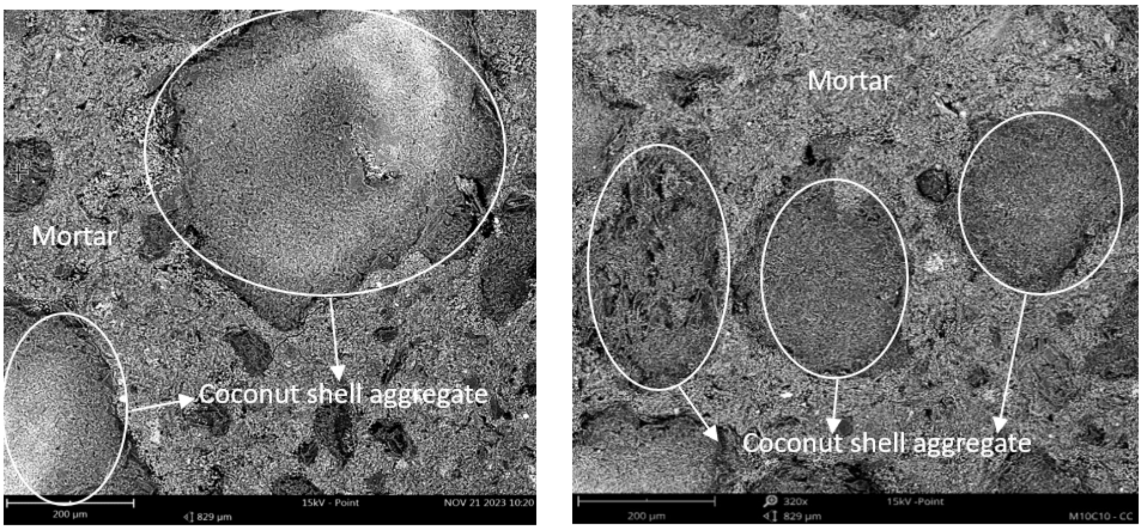
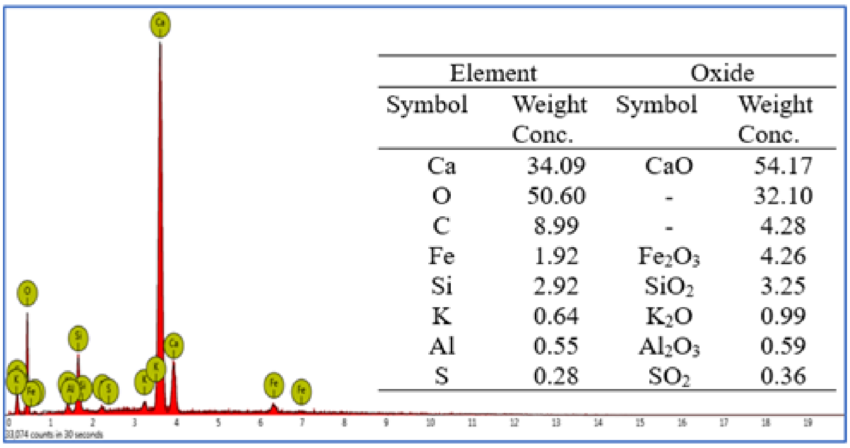


Figure 11: SEM images of MK and CSA concrete sample.



Element Symbol	Element Weight Conc.	Oxide Symbol	Oxide Weight Conc.
Ca	34.09	CaO	54.17
O	50.60	-	32.10
C	8.99	-	4.28
Fe	1.92	Fe_2O_3	4.26
Si	2.92	SiO_2	3.25
K	0.64	K_2O	0.99
Al	0.55	Al_2O_3	0.59
S	0.28	SO_2	0.36

Figure 12: EDS analysis of MK and CSA concrete sample.

Summary and Conclusion

This study was conducted to contribute to existing literature efforts of introducing MK as a cement substitute and CSA as a coarse aggregate substitute in concrete. The key characteristics such as split tensile strength, compressive strength, microstructural analysis and density of the concrete were investigated. The study used scanning electron microscopy and standard tests. it was established that:

a) Concrete specimens with 5% MK and 10% CSA replacement achieved better results than the targeted compressive strength of 26.6MPa at 28 days of curing.

b) The compressive strength of cement with 5-20% MK and CSA set at 10% with natural aggregate concretes observed at the age of 28 days was approximately 6.4-22.7% higher than the minimum requisite strength of 21 MPa for structural normal weight concrete as per ASTM C330.

c) The split tensile strengths of cement with 5-20% MK replacement and CSA set at 10% natural coarse aggregate obtained at the age of 28 days were approximately 11.8-25.3% higher than the minimum requisite strength of 2.1MPa for structural normal-weight concrete.

d) The density of concrete decreases with an increase in the metakaolin content.

e) A strong correlation was found, with an R^2 value of 0.78, between the density and compressive strengths of MK and CSA replacement concrete.

f) The study found that the split tensile and compressive strengths of MK and CSA concrete had a strong correlation, with R^2 values ranging from 0.9749 to 0.9896.

g) Statistical analysis revealed no significant difference between the compressive and split tensile strengths of the control and the synergistic of cement with 5-20% MK and CSA set at 10% with natural coarse aggregate.

h) An analysis of the microstructure of concrete with CSA replacing part of the natural coarse aggregate revealed that the cement paste penetrated the pores of the CSA, resulting in strong adhesion between the aggregate and the cement-sand matrix. When CSA was set at 10% natural coarse aggregate, this factor compensates for the lower strength of CSA compared with natural coarse filler, thereby maintaining the composite's strength without significant decline.

i) EDS analysis revealed high concentrations of Ca/Si oxides responsible for the C-S-H formation, resulting in increased strength and density.

j) Concrete with 5% MK and 10% CSA content exhibited a relatively compact microstructure and minimal internal porosity.

The study concludes that the synergistic use of 5% MK and 10% CSA with natural coarse aggregate are the most rational for normal concrete application. The current study mainly focused on the physical, mechanically and microstructural properties of concrete prepared with MK and CSA. It is, therefore, recommended that additional research be done on the durability and thermal conductivity properties of concrete with MK and CSA. Future research on MK and CSA's effects on the environment is also recommended.

Data Availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Declaration of Interest Statement

The authors declare that they have no competing interests.

Authors' Contributions

John Newton Antoh (JNA) carried out the investigation, experimental work and prepared the first draft. Humphrey Danso (HD) validated, confirmed, supervised, evaluated and edited the work. Russell Owusu Afrifa (ROA) analyzed the concrete properties, investigated other sections and validated the data. The final manuscript was read and approved by all authors.

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