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Development of cementless mortar produced from calcined kaolin clay and granite dust powder with chemical activator

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Abstract: Geopolymers have garnered worldwide interest as an alternative binder to ordinary Portland cement (OPC) due to their environmental benefits, improved durability, and acceptable mechanical properties. Large amounts of aluminosilicate waste materials, often discarded and causing pollution, are generated globally. This study aims to develop a cementless binder (CLB) as a 100% OPC replacement by using pozzolanic and waste materials—calcined kaolin clay (CKC) and granite dust powder (GDP)—activated chemically with sodium hydroxide (NaOH). Six mortar mixes were prepared with CKC partially replaced by GDP at 0, 5, 10, 15, and 20% by weight. The mixtures were evaluated for various properties, including consistency, setting time, workability, bulk density, compressive strength, and flexural strength. Results showed increased consistency but decreased setting time and flow compared to conventional mortar. Setting time increased with higher GDP replacement. All alkali-activated CKC+GDP mixes demonstrated improved workability. The mixture containing 10% GDP exhibited the highest compressive strength (42.7 MPa) and flexural strength (6.69 MPa) after 28 days of air curing at 80°C, outperforming the control mixture. The findings suggest that the CLB developed with CKC and GDP activated chemically is a promising sustainable alternative to cement, especially for structures where early high strength is not critical. This approach can reduce cement demand, lower CO_2 emissions, and mitigate environmental pollution by utilizing waste materials.

Keywords: *Alkali-activators, Cementless binders, Hardened characteristics, Physical properties, Pozzolanic and waste materials, Workability.*

1. Introduction

In recent years, the global consumption of cement within the construction sector has risen sharply, particularly in developing nations. The global demand for cement is anticipated to continue rising due to factors such as population growth and the continuous development of infrastructure. In response to this escalating demand, the use of pozzolanic materials and industrial by-products as partial substitutes for cement in concrete formulations has become increasingly widespread. This practice has particularly gained significant momentum in recent decades, especially in regions like North America and Europe. This practice is also observed in Asian countries occasionally. Simultaneously, the imperative of environmental protection is emphasized by international summits that set clear eco-sustainability goals. These goals focus on lowering harmful CO_2 emissions, minimizing waste production, developing innovative recycling strategies to repurpose materials previously classified as waste, and establishing effective systems to alleviate the environmental impact of mining, industrial activities, and energy supply. Achieving the 45% reduction target for CO_2 emissions, limiting the global temperature rise to below 2 degrees, and reaching net-zero emissions is not feasible unless sectors with high energy consumption, like cement, implement strategies to curb their CO_2 emissions [1, 2]. Although cement production has made significant environmental improvements, it remains a major source of CO_2

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emissions. Therefore, efforts should focus on reducing cement consumption by incorporating alternative, less polluting materials. Additionally, substituting traditional cement with recycled eco-friendly products or industrial waste in concrete and high-performance mortar production could further mitigate its environmental impact [3, 4]

Furthermore, replacing a part of the cement with waste materials can reduce the amount of cement required for production, potentially improving the characteristics of both fresh and hardened concrete. This approach minimizes the need for extensive industrial processes, leading to cost and time savings as well as a reduction in environmental pollution [3]. Given the high global demand for cement, incorporating industrial and agricultural waste as partial replacements offers significant economic and ecological advantages worldwide. Pozzolanic and waste materials refer to mineral additives made from finely ground substances that are incorporated into concrete and mortar mixes to improve certain fresh and hardened characteristics of ordinary Portland cement (OPC) concrete. These materials are typically sourced either from natural origins or as by-products of industrial processes. When used as partial replacements for OPC, they are commonly referred to as OPC substitute materials. Pozzolanic materials are composed of siliceous or alumino-siliceous substances that, by themselves, exhibit little to no cement-like qualities. When these materials are finely milled and mixed with moisture, they react chemically with alkali and alkaline earth hydroxides at ambient temperatures, leading to the creation of compounds that exhibit cement-like properties. As a result, to reduce these issues, researchers have focused on exploring the use of waste materials like ground granulated blast furnace slag (GGBS), fly ash (FA), rice husk ash (RHA), marble powder (MP), and others as viable substitutes in the construction industry, particularly in concrete production.

Various pozzolanic materials occur naturally, including metakaolin (MK) and calcined shale or clay. Moreover, various industrial by-products, including marble powder (MP) and granite powder (GP), are produced daily as waste materials. Their incorporation into concrete and blended cement manufacturing is becoming more common, as it generally helps to decrease the amount of cement needed. This not only lowers production costs but also helps decrease CO_2 emissions [5]. Moreover, incorporating high volumes of these materials in cement and concrete contributes to the conservation of natural resources like sand and stone, supporting more sustainable construction practices [6]. The effective use of waste materials is a critical component of waste management approaches worldwide. Recycling offers several benefits, including minimizing environmental pollution, decreasing the volume of waste sent to landfills, and conserving natural resources [7]. In a similar vein, using waste by-products like marble and granite powders as partial replacements in mortar and concrete, once their compatibility is verified through testing, offers a potential strategy for reducing waste while possibly improving the characteristics of both fresh and hardened mortar and concrete. Motivated by this, the present study conducted an experimental investigation into the development of a cement-free binder (CLB) using these pozzolanic waste materials combined with chemical activators, aiming to provide a sustainable alternative to traditional cement.

Geopolymer materials represent a cutting-edge approach gaining significant attention within the construction sector, particularly due to the increasing emphasis on environmental sustainability. In contrast to conventional Portland cement, which is widely used in the industry and requires significant energy, contributing substantially to carbon dioxide emissions (responsible for approximately 85% of energy consumption and 90% of CO_2 emissions in typical ready-mixed concrete), geopolymers primarily rely on natural raw materials or industrial by-products as their main binding agents. This substitution offers substantial potential reductions in both energy use and carbon footprint Consequently, geopolymers are being increasingly explored for applications in transportation infrastructure [8-10].

The chemistry behind geopolymer synthesis is relatively straightforward. Aluminosilicate-based geopolymers require source materials rich in amorphous silicon and aluminum compounds. These materials can be obtained from natural minerals and clays, such as kaolinite, or industrial by-products like low-calcium ASTM C618 Class F fly ash (FA) or ground granulated blast furnace slag (GGBS), or combinations thereof. The most frequently produced low-calcium alkali-activated binders, known as geopolymers, are generally created by utilizing fly ash or metakaolin (MK) as solid aluminosilicate sources [11-15].

Calcined Kaolin Clay (CKC) is a form of mineral admixture also referred to as MK, one of the more recently created supplemental cementing ingredients. It is made by calcining pure kaolin clay at a particular temperature range in order to release the chemically bonded water and dissolve the crystalline structure. A substance that is reactive with lime is created as a result of this method. Due to the quasiamorphous character of the collapsed structure, the calcined clay reacts differently with lime. There is, thus, a preferred calcination temperature for each clay; at temperatures higher than the preferred level, re-crystallization starts, whilst at lower temperatures, the clay lattice structure remains unaltered. The combination of sodium silicate and sodium hydroxide solutions for activating metakaolin (MK) results in materials exhibiting significantly higher mechanical strength than those activated with sodium hydroxide alone. Additionally, flexural strength improves as the volume of the activator is reduced or the sodium concentration is increased [16-20].

Granite dust (GD) is waste from crusher units and quarries from rock processing. The penalties are currently being eliminated of by Filling in the odd soil. This causes severe climate change issues. If this material is utilized as a partial substitute in concrete, it could provide significant economic and environmental advantages. sludge/rock dust, which is an easily available Such material is waste from granite rock quarries and crushers. The sludge disposal by landfill is a cause of Important environmental factors. If waste materials can be utilized as partial substitutes for cement, it would contribute to a sustainable and resource-efficient concrete technology. GD is consisting mainly of alumina and silica and therefore complies with the Chemical demands for Pozzolanic components [21-24].

The advancement of cement-free binders derived from pozzolanic and waste materials holds considerable importance for several reasons. Recently, the incorporation of pozzolanic and industrial by-products into cement and concrete production has surged, owing to numerous advantages such as reduced cement consumption, lower manufacturing costs, and enhanced concrete performance [4, 25]. As a result, the need for alternative, sustainable binders has become increasingly important to address the depletion of the world's limited fossil fuel resources and to reduce the CO_2 emissions linked to cement production. The efficient use or recycling of these waste materials as full (100%) substitutes for cement, activated with minimal alkaline additives and cured at ambient temperatures, presents a promising approach to minimize waste disposal and lower the carbon footprint. Moreover, employing such materials as substitutes for cement is both practical and timely, aligning with the global objective of achieving sustainable concrete and eco-friendly binding solutions. Against this backdrop, creating new binders from readily available local waste sources is both relevant and impactful. In this study, granite dust powder (GDP), a by-product produced during the cutting, shaping, and polishing of granite, was incorporated as a partial pozzolanic substitute in geopolymer formulations based on CKC. The inclusion of GDP showed a notable enhancement in compressive strength, indicating its promising potential as an effective alternative raw material for geopolymer binder production.

This chapter highlights how the inclusion of pozzolanic materials affects the strength of both mortar and concrete, focusing on the advantages of their supplementary use. Additionally, it emphasizes the substantial impact these materials have on the durability characteristics of mortar and concrete as they accumulate. Finally, a discussion on the geopolymer concrete, chemical activators and mechano-chemical activation technique has been introduced to find a guideline for development of non-cement binder (NCB) as a finding of this research.

2. Experimental Program

The evaluation of various mortar formulations was conducted by analyzing both their fresh and hardened characteristics based on experimental data. To assess the fresh properties, workability was examined through fresh flow table and slump tests. The mechanical properties of the hardened state were evaluated through tests measuring the dry unit weight, compressive strength, and flexural strength. A comprehensive experimental program was developed for this characterization process.

2.1. Materials

The cement utilized in all the mixtures was locally sourced Portland cement CEM1, complying fully with the Iraqi Standard No. 5/2019. Usually, this brand of cement is considered as the better quality and available at local market in Iraq. The cement was stored in airtight plastic containers and put in dry place to avoid exposure to atmospheric conditions.

The CKC was derived from Iraqi Kaolin clay collected from the Dewekhla region in the Al Ramadi desert, situated to the west of Baghdad, Iraq. To create the CKC, the kaolin clay was first ground into a fine powder and subsequently heated in a furnace at a temperature of 800 ºC ± 20 ºC for two hours, with a heating rate of 5 ºC per minute. After the heating process, the material was allowed to cool gradually to ambient temperature over a period of 24 hours [26]. The final reactive material, with a finer consistency, was then produced in Baghdad's Al-Zahra'a Shop using the air blast method.

In this study, granite dust powder (GDP), a waste material generated from the granite processing industry, was utilized. The use of granite dust described above as a filler for concrete, mortar and dry building mixtures of multiple reasons is a promising direction of this waste recycling. It has been characterized from a chemical and physical viewpoint.in order to evaluate its use in the development of mortar and concrete. Fig. 1 shows photographs of calcined kaolin clay (CKC) that was produced and granite dust powder (GDP) as received from the factory. The granite dust powder was analyzed in the laboratory to determine its physical and chemical properties.

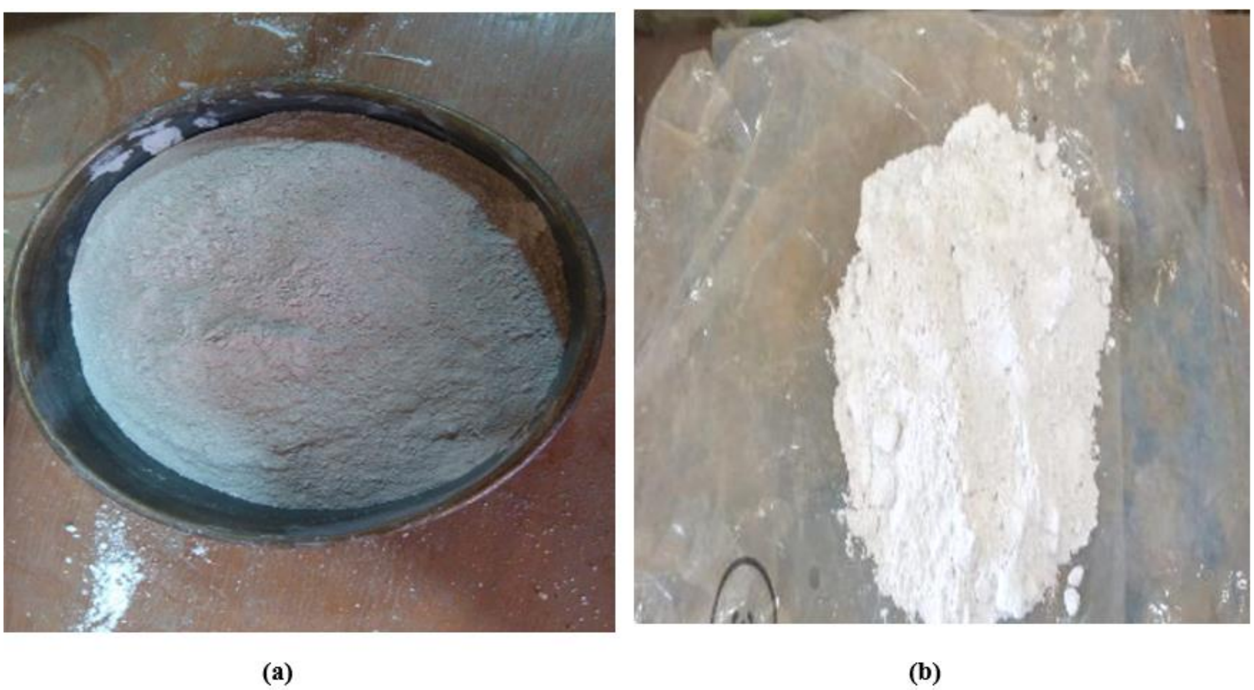

Figure 1.
Photograph of: (a) calcined kaolin clay powder and (b) granite dust powder.

Tables 1 and 2 display the chemical compositions and physical properties of Ordinary Portland Cement (OPC), Calcined Kaolin Clay (CKC), and Granite Dust Powder (GDP), respectively. For the fine aggregate, local river sand from the Al-Najaf Sea Region was employed, with a maximum particle size of 4.75 mm, in accordance with ASTM standards [27]. The sand exhibited a fineness modulus of 2.86, a

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water absorption rate of 1.72%, and a specific gravity of 2.65. To activate the CKC and GDP materials and enhance the binding properties of the cementitious material, a chemical activator was used, namely sodium hydroxide (NaOH). Sodium hydroxide, purchased locally in pellet flakes and granules form with 99% purity, was prepared for use. The solution prepared had a molarity of 12 M and was supplemented with additional water. After preparation, the solution was allowed to cool naturally over a period of 24 hours. To ensure the required workability for all mortar mixtures, a high-range water-reducing agent (HRWRA) based on polycarboxylate, conforming to ASTM C494/C494-05 [28] standards, was used. This superplasticizer, branded as Glenium 54, was supplied as a liquid with a color ranging from whitish to straw-yellow.

Table 1.
Chemical Composition of Cement, Calcined kaolin clay and Granite dust powder.

Chemical composition (%)	Cement	Calcined kaolin clay	Granite dust powder
Silicon dioxide (SiO_2)	19.36	53.6	67.7
Aluminum trioxide (Al_2O_3)	4.82	36.8	12.5
Iron oxide (Fe_2O_3)	3.28	1.97	4.64
Calcium oxide (CaO)	62.43	0.78	3.72
Magnesium oxide (MgO)	3.0	0.32	0.84
Sodium oxide (Na_2O)	0.07	0.84	4.22
Potassium oxide (K_2O)	0.44	0.17	3.89
Sulfur trioxide (SO_3)	2.26	0.43	0.21
Phosphorus pentoxide(P_2O_5)	-	0.26	0.14
Loss on ignition (LOI)	2.17	4.83	2.43
Lime Saturation Factor (LSF)	0.96		
PH	-		
Bogue Composition, (%)			
Tri-calcium silicate (C_3S)	58.49	-	-
Di-calcium silicate (C_2S)	11.38	-	-
Tri-calcium aluminate (C_3A)	7.22	-	-
Tetra-calcium aluminoferrite (C_4AF)	9.98	-	-

Table 2.
Physical Characteristics of Cement, Calcined Kaolin Clay and Granite dust powder.

Property	Cement	Calcined kaolin clay	Granite dust powder
Specific gravity	3.12	2.61	2.68
Fineness (SSA), m^2/kg	338	1650	569
Median particle size (μm) (d_{50})	16.8	13.3	24.8
Pozzolanic activity index 7 days (%) 28 days (%)	 100 100	 96 102	 68 83
Color	Grey	Off-White	Light grey

2.2. Mixture Proportions of New Binder

After assessing the physical and chemical characteristics of all raw materials, the preliminary tests focused on utilizing 100% CKC independently to assess its ability to act as a binder. Utilizing the reactive components identified in the CKC's chemical composition, geopolymer pastes and mortars were subsequently prepared, with detailed formulations presented in Tables 3 and 4, respectively. The research program was structured into two distinct phases. The initial phase focused on identifying the optimal percentage of granite dust powder (GDP) to be added to CKC that would maximize compressive strength. In the subsequent phase, the study examined both the fresh and mechanical properties of mortar mixtures where GDP replaced CKC at varying ratios of 0%, 5%, 10%, 15%, and 20%.

Paste and mortars were prepared by inclusion of chemical activators such as NaOH by weight of CLB and by molar concentration of activators. Firstly, the geopolymer paste samples were created by blending CKC and CKC+GDP with a specially prepared alkaline activator solution (NaOH) using a 5-liter mixer for approximately five minutes. Once thoroughly mixed, the resulting paste was transferred into cubic plastic molds measuring 50 mm on each side. To remove any trapped air bubbles, the molds were placed on a vibrating table for a duration of two minutes. Subsequently, a thin plastic film was applied over the specimens to reduce moisture evaporation during the curing process.

Secondly, geopolymer mortar mixtures incorporating marble dust powder and metakaolin were prepared by combining calcined kaolin clay (CKC), a mixture of GDP and CKC, and a liquid alkaline activator (5.0% NaOH by binder weight). The ratio of sand to binder was fixed at 2.75, while the water to binder ratio was kept at 0.485. All components, including the cementless binder (CLB), sand, chemical activator, water, and superplasticizer (SP), were fully mixed using a Hobart mixer in accordance with ASTM C305-06 [29] guidelines after the alkaline activator had been diluted.

The mortar samples were prepared by casting into 50 mm cubic steel molds in three equally thick layers, each layer being thoroughly compacted. To eliminate trapped air and bubbles, the specimens were subjected to vibration immediately after casting. After vibration, the molds were immediately covered and transferred to an oven for curing at a temperature of 80 °C for 24 hours. Once the curing process was complete, the specimens were taken out of the oven and kept under normal environmental conditions, with an average temperature of 28 °C and 70% relative humidity, until the testing day.

Table 3.
Proportional Composition of Materials by Weight (%) for Paste Mix Preparation Utilized in this Study.

Mix no.	Type of binder (%)			NaOH (%)	SP (%)	W/B	S/B
	OPC	CKC	GDP				
OPC-GP	100	0	0	0	0	0.33	-
CKCGDP0	0	100	0	5	3	0.4	-
CKCGDP5	0	95	5	5	3	0.4	-
CKCGDP10	0	90	10	5	3	0.4	-
CKCGDP15	0	85	15	5	3	0.4	-
CKCGDP20	0	80	20	5	3	0.4	-

Table 4.
Proportional Composition by Weight of Materials Utilized in the Preparation of Mortar Mixes for the Present Study.

Mix No.	Type of binder (%)			NaOH (%)	SP (%)	W/B	S/B
	OPC	CKC	GDP				
OPC-GM	100	0	0	0	0	0.5	2.75
CKCGDP0	0	100	0	5	3	0.4	2.75
CKCGDP5	0	95	5	5	3	0.4	2.75
CKCGDP10	0	90	10	5	3	0.4	2.75
CKCGDP15	0	85	15	5	3	0.4	2.75
CKCGDP20	0	80	20	5	3	0.4	2.75

The mortar mixtures in this study were cast as specimens using the molds used (50×50×50) mm cubes to obtain specimen for compressive strength and density, determination (50×50×300) mm prism to obtain specimens for flexural strength. Before casting, the molds were thoroughly cleaned and their interior surfaces were coated with oil to prevent the mixture from sticking after curing. The concrete was then poured in successive layers, each approximately 50 mm thick. Each layer was compacted using a vibrating table to eliminate trapped air as effectively as possible. After compaction, the surface of the concrete was smoothed using a trowel.

In the final step, the specimens were wrapped in plastic sheets and maintained in a moist environment for approximately 24 hours to avoid plastic shrinkage cracks. Subsequently, the specimens were removed from the molds and placed in an oven set at 80°C for 24 hours, as depicted in Fig. 2. After this, the specimens were taken out of the oven and allowed to reach room temperature before being tested.

(a) (b)

Figure 2.
Curing Techniques: (a) Using an Oven, (b) At Ambient Room Temperature.

2.3. Fresh Properties of Pastes and Mortar

The standard consistency of cement paste serves as an important measure of its plasticity. For any specific type of cement, consistency is defined as the precise water content needed to achieve a paste with a predetermined standard flowability [30]. According to ASTM C187-09 [31] consistency is quantified by the volume of water necessary for the paste to allow a Vicat needle, with a diameter of 10 mm, to penetrate to a depth of 10 mm. To observe the water demand, normal consistency of the CKC, CKC+GDP and OPC was tested. In this study, cement pastes were prepared and tested following the ASTM C187-09 [31] standard. Setting time is a crucial parameter that indicates how long it takes for cement paste to transition from a fluid to a solid state. It represents the stiffening process of the paste, marking the change from a liquid to a rigid form [30]. According to ASTM C191-09 [32] the setting time of cementitious pastes is defined as the duration required for the paste to harden from its initial fluid condition to a solid state. The initial setting time is recorded when a standard paste-prepared with the water amount for normal consistency-allows a Vicat needle (1 mm diameter) to penetrate up to 25 mm. The final setting time is observed when the needle ceases to penetrate the paste under identical testing conditions. The setting times of the new binders were measured alongside ordinary Portland cement (OPC). The Vicat apparatus was employed to assess the setting times of CKC, CKC+GDP, and OPC pastes.

Flowability refers to the ability of a paste to deform and spread, reflecting its workability and suitability for shaping. To evaluate this property, mortar mixes were subjected to flow tests following the ASTM C1437-09 [33] standard.

2.4. Hardened Properties of Pastes and Mortar

Self-weight of any structure is completely dependent on unit weigh (bulk density) of the ingredient materials. Thus, it is a considerable parameter for mortar or concrete. The dry unit weight of the mortar was measured by calculating the mass of mortar per unit volume, following the guidelines specified in ASTM C642-03 [34]. This value was obtained by averaging the measurements from three separate specimens.

The specimens were placed vertically on one of their faces in the testing apparatus. A load was applied at a constant rate of 0.24 MPa per second. The mixtures were prepared with a set water-to-binder ratio of 0.485, while the mortar flow was kept within 110 ± 5 mm by modifying the superplasticizer content. A consistent fine aggregate-to-binder ratio of 2.75 was maintained across all mixtures. Both geopolymer and OPC mortars were mixed thoroughly before being poured into molds measuring 50 × 50 × 50 mm for compressive strength testing. The specimens were compacted using a vibrating table to ensure uniform consolidation. After casting, the specimens remained in their molds for 24 hours, followed by demolding. The samples were cured in ambient air at 60°C and covered with plastic sheets to reduce moisture evaporation. Compression strength tests were conducted using a standard control compression testing machine. The compressive strength was determined after measuring the density at 3, 7, and 28 days, in accordance with the ASTM standard [35]. The compressive strength values presented are the average results from testing three specimens for each mix. For each testing age, the mean strength of the three samples was recorded. Flexural strength tests were performed on 40 × 40 × 160 mm mortar prisms at the same curing ages, following the ASTM standard [36]. A testing machine with a 100 kN capacity was used to measure flexural strength, applying a two-point loading system. The test was conducted at 28 days, and the average result from the two prisms was recorded.

3. Analysis and Discussion of Results

3.1. Chemical and Physical Characterization of Materials

The chemical compositions of the calcined kaolin clay (CKC) and granite dust powder (GDP) are provided in Table 1. It was found that the main components of CKC are silica and alumina (53.6 % and 36.8 %), respectively, which are responsible for the pozzolanic reaction or secondary hydration in mortar or concrete.

The total proportion of key oxides (SiO_2 + Al_2O_3 + Fe_2O_3) in CKC exceeds 70%, surpassing the minimum threshold of 70% set by ASTM standards [37] for class (N) pozzolans, which are categorized as high-quality pozzolans. The chemical composition of the GDP was analyzed and is presented in Table 1. From the table, it is clear that GDP predominantly consists of SiO_2 and Al_2O_3, with small amounts of MgO, CaO, and Fe_2O_3, highlighting its predominantly siliceous composition. The chemical analysis indicates that GDP contains approximately 68.7% silica (SiO_2) and 13.6% alumina (Al_2O_3), making it highly suitable for concrete production. The loss on ignition (LOI), which represents the weight reduction observed when a sample is heated to 1000 °C, is a key factor in determining the material's composition. During this heating process, moisture and carbon dioxide are typically expelled. The LOI values for CKC and GDP are listed in Table 1. While the LOI for CKC is higher, it remains below the 10% threshold specified by ASTM [37]. In contrast, the LOI for GDP is 2.43%, well below the ASTM-recommended value of 10% [37].

Table 2 presents the physical characteristics of CKC and GDP. The specific gravities of OPC, CKC, and GDP are 3.12, 2.61, and 2.63, respectively, indicating that both CKC and GDP have lower densities compared to ordinary Portland cement. Notably, CKC exhibits a higher specific surface area (SSA) than GDP and OPC, which is attributed to certain material transformations. The SSA values were measured as 1650 m^2/kg for CKC, 1040 m^2/kg for GDP, and 386 m^2/kg for OPC. This clearly shows that all pozzolanic materials analyzed have a significantly greater surface area than OPC. The elevated SSA of these pozzolanic materials suggests they have favorable characteristics to act effectively as pozzolans. Furthermore, the particle size distributions for CKC and GDP span from 0.1 µm to 90 µm, with median particle sizes (d_{50}) of 12.8 µm and 14.3 µm, respectively.

3.2. Fresh Mortar Properties

Laboratory tests were conducted to evaluate the fresh properties of binders made from calcined kaolin clay (CKC) and granite dust powder (GDP), including normal consistency, setting time, and flow characteristics. Achieving proper normal consistency is crucial for the effective mixing of pastes and mortars. Table 5 summarizes the measured consistency values for Ordinary Portland Cement (OPC) and cement-less binders (CLBs) containing CKC and CKC combined with GDP. While the OPC paste exhibited a normal consistency of 33%, the CLB pastes containing CKC and CKC+GDP showed higher values, ranging from 34% to 42%, depending on the fineness and particle characteristics of the

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pozzolanic materials used. This improvement in consistency can be explained by the smaller particle size and larger specific surface area of CKC and GDP in comparison to OPC. Consequently, pozzolanic-based pastes generally require more water to achieve the desired workability, a trend consistent with earlier studies El-Diadamony, et al. [38] and Wianglor, et al. [39] which reported a gradual increase in consistency with higher CKC content. Thus, it is evident that pastes incorporating pozzolanic materials exhibit greater consistency than pure OPC pastes.

Furthermore, the initial and final setting times of OPC, CKC, and CKC+GDP pastes were measured under controlled laboratory conditions at a temperature of 25 ± 2 °C. The results are also shown in Table 5. Fig. 3 illustrates the comparative setting times for OPC and alkali-activated CKC pastes, both with and without the addition of granite dust powder, highlighting the influence of these supplementary materials on the setting behavior of the binders. In the case of alkali-activated by NaOH, setting time of the CKC pastes (CKCGDP0) was lesser but it increased with addition of 5%, 10%, 15% and 20% GDP as CKC replacement [23, 40, 41]. The addition of GDP influenced the setting times, both initial and final, of alkali-activated CKC pastes. An increase in the amount of marble dust powder led to a delay in the setting time of the CKC+GDP mixtures.

Table 5.
Normal Consistency and Setting Time of OPC, CKC and CKC+GDP Pastes.

Mix no.	Consistency (%)	Setting time (hour: min)	
		Initial	Final
OPC-GP	33.0	1:25	5:19
CKCGDP0	42.0	0:89	3:27
CKCGDP5	39.0	3:61	7:08
CKCGDP10	37.0	3:18	6:57
CKCGDP15	36.0	2:47	6:11
CKCGDP20	34.0	2:13	5:84

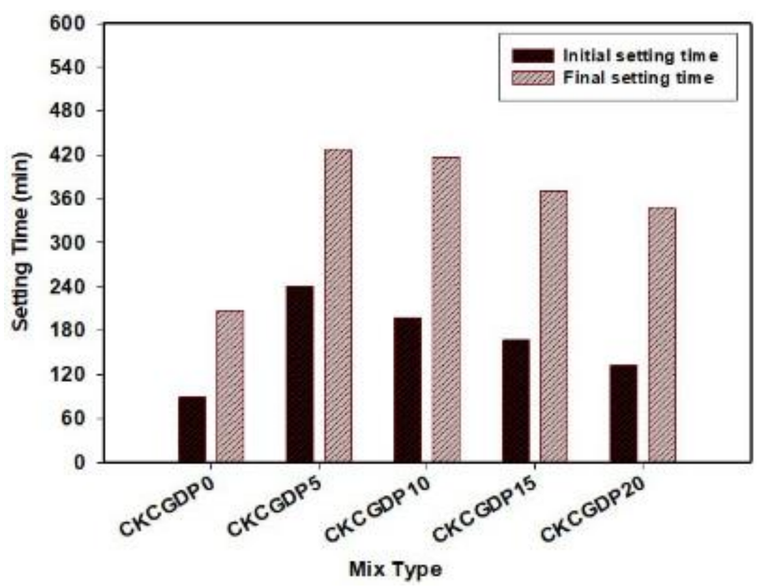

Figure 3.
Effect of Replacing CKC with GDP on the Setting Time of CKC-GDP Paste.

The most significant extension in setting time occurs when 20% of GDP is used as a replacement in the binary system with CKC, compared to the other mixtures. This phenomenon is primarily attributed to the silica content in GDP. Additionally, the relatively larger particle size of GDP may contribute to the longer setting duration of the paste. On the other hand, when CKC completely replaces the material at a 100% ratio, the setting time decreases, which is probably attributed to the smaller particle size of CKC [39] similarly observed a decrease in the setting time of calcined kaolin clay paste with complete substitution by CKC.

3.3. Flow of Mortars

The flow results of alkali-activated CKC, alkali-activated CKC+GDP and cement mortars mixtures are presented in Fig. 4. Mortars were prepared using 5% NaOH with a water to binder ratio of 0.5 and sand to binder ratio of 2.75, and using local river sand. This section explores how the inclusion of GDP affects the flow characteristics of the pure CKC mixture while maintaining a fixed water-to-binder (w/b) ratio. As shown in Fig. 4, the neat CKCGDP0 mixture-containing no GDP-demonstrated a flow value of 48±5%, indicating moderate workability. In contrast, all alkali-activated CKC+GDP mixtures exhibited improved workability compared to the neat CKCGDP0 mix. This improvement can be credited to the smaller particle size of CKC when compared to GDP. Furthermore, the higher inclusion of CKC leads to an increased water requirement for the concrete, which can be attributed to factors such as the replacement ratio, the elevated specific surface area (SSA) of CKC, and the increased silica content within the mixture.

The fineness of the particles, which is closely related to CKC, is a key factor in influencing the cementitious properties of blended systems that incorporate pozzolanic materials. Previous studies Wong and Razak [41] and Arikan, et al. [42] have reported that pozzolans generally require additional water to achieve the desired consistency, often resulting in reduced flowability. However, in this study, an increase in GDP content corresponded to an improvement in flowability. The mixture with the highest GDP proportion showed the best workability among all tested samples. Workability increased with GDP content up to 20% by weight. For comparison, the flow value of the ordinary Portland cement (OPC) mortar without superplasticizer (SP) exceeded 80%.

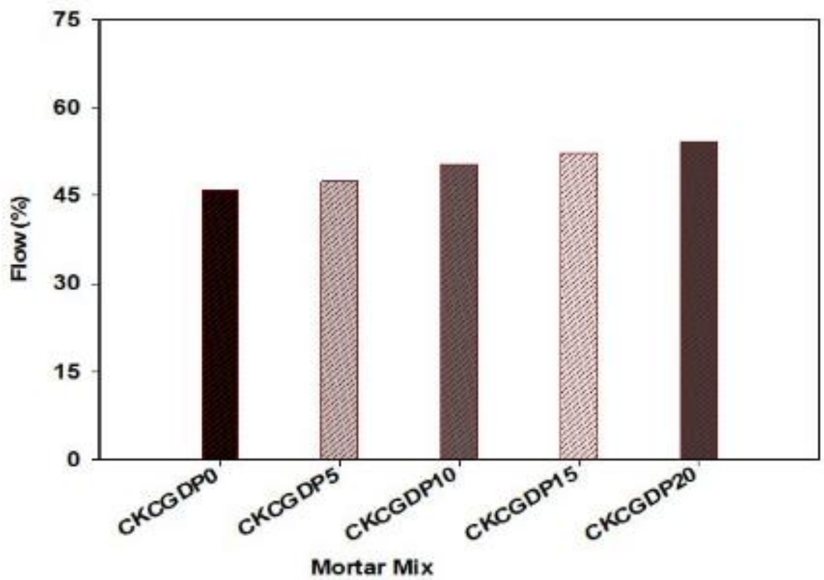

Figure 4.
Effect of GDP Substitution on the Performance of Alkali-Activated CKC Mixtures.

3.4. Hardened Properties of Mortar

The mechanical strength or hardened characteristics of cement, or any binding material, are essential for structural applications. Mortar strength is influenced by the cohesion of the cement paste, its bonding with the aggregate particles, and, to some degree, the strength of the aggregate itself [43].

The hardened properties, such as unit weight (density), compressive strength, and flexural strength of OPC and CLB-mortars (alkali-activated CKC, alkali-activated CKC+GDP), are discussed in detail below.

3.5. Unit Weight of Mortar

Self-weight of any structure is completely dependent on the unit weight (bulk density) of the ingredient materials. Thus, it is a considerable parameter for mortar or concrete. The bulk density of alkali-activated CKC, alkali-activated CKC+GDP and OPC cured at 80 °C as shown in Figure 5. The figure demonstrates that the density of all mortar types-whether containing pozzolanic and waste additives or not-increased over time. Additionally, Fig. 5 illustrates how different binders influenced the bulk density of the blended mortars at 3, 7, and 28 days of curing. The results showed an increase in bulk density with increasing the rates of the CKC substitution by GDP used in the preparation of the geopolymer mortars in comparison with CKCGDP0 mixture.

The specific weight of the marble powder material is higher than that of the metakaolin, therefore, the bulk density values of the samples increased with an increase in the substitution levels. The highest bulk density values were obtained from the mixtures with 20% GDP substitution. This is due to that the particle size and specific gravity of GDP was considerably higher than that of CKC, which increased the mass per unit volume. It was found that OPC mortar showed a unit weight of 2241 kg/m^3, while values of CLB were in the range of 2047 kg/m^3 to 2130 kg/m^3 for the binary mortar blends (alkali-activated CKC, alkali-activated CKC+GDP).

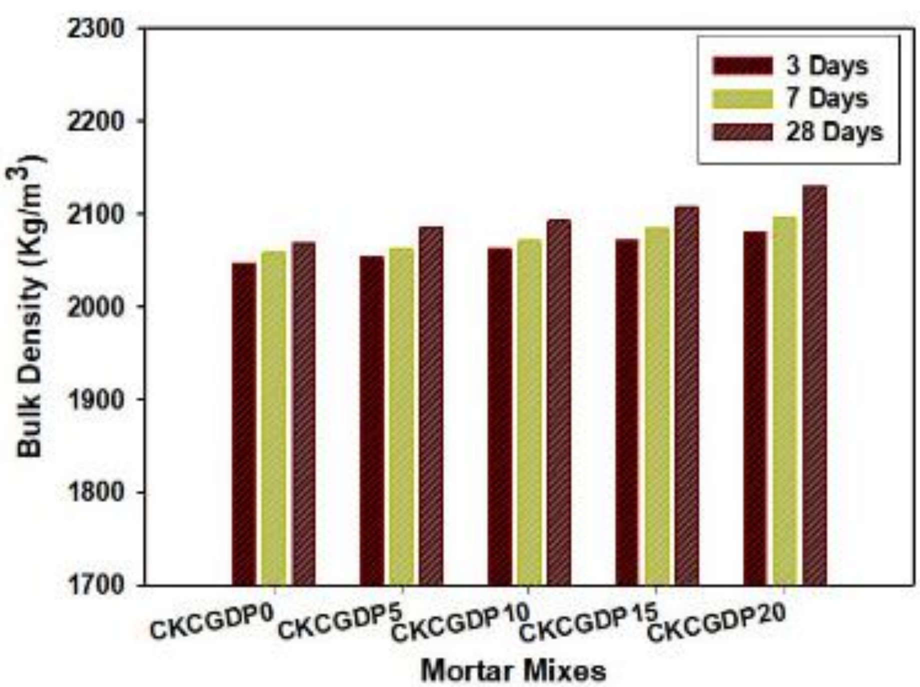

Figure 5.
Bulk densities of alkali-activated CKC, alkali-activated CKC+GDP mortars mixes.

3.6. Compressive Strength of Mortar

Compressive strength is a key design criterion for all types of concrete structures, as it directly influences both the structural integrity and overall project cost. In this study, the strength development of calcined kaolin clay-based binders (CLBs), specifically alkali-activated CKC and alkali-activated CKC blended with ground desulfurization product (GDP), alongside ordinary Portland cement (OPC) mortar, was examined at various curing ages. Fig. 6 illustrates the compressive strength results of binary mortar mixes consisting of CKC partially replaced by different proportions of GDP, tested at 3, 7, and 28 days. The data indicate that the level of GDP substitution plays a major role in compressive strength evolution. Initially, all mixes containing GDP showed a slight decrease in strength at early curing stages (3 and 7 days), with greater reductions observed as the GDP content increased compared to the control mix without GDP (CKCGDP0). However, by 28 days, mortars incorporating 5% and 10% GDP as partial replacements of calcined kaolin clay demonstrated a notable improvement in compressive strength relative to the control mortar.

In contrast, mortar samples containing 15% and 20% GDP as a replacement for CKC exhibited a decline in compressive strength starting from day 3 when compared to the CKCGDP0 sample. However, an overall increase in compressive strength was observed with higher GDP content. Specifically, the compressive strength of CKCGDP0 reached only 38.2 MPa, while CKCGDP10 achieved the highest strength of 42.6 MPa at 28 days. This indicates that GDP acts as a highly reactive material, significantly enhancing the reaction kinetics. The increase in reactive silica content at CKCGDP10 was identified as a crucial factor influencing the geopolymer's reactivity potential. These findings confirm that GDP is an important and compatible additive when used alongside other industrial by-products like RHA, FA, and POFA. The improved strength development in the mortar is attributed to the synergistic interaction between these materials in the binary blend [23, 44, 45].

As the percentage of GDP substitution increased, the compressive strength of the geopolymer mortar started to decrease. After a 28-day curing period at 80±2 °C, the compressive strengths of the mortar mixes CKCGDP0, CKCGDP5, CKCGDP10, CKCGDP15, and CKCGDP20 were recorded as 39.2, 40.7, 42.8, 35.6, and 34.7 MPa, respectively. Significantly, the inclusion of 5% and 10% GDP led to enhancements of about 3.8% and 8.4% in compressive strength at 28 days, respectively, when compared to the control mix. The contribution of granite powder to strength gain at later stages is primarily attributed to its rapid pozzolanic activity. This enhancement is closely linked to extended curing time, which allows the geopolymerization process to complete. In addition, increasing the concentrations of Al_2O_3 and SiO_2 facilitated the formation of geopolymers, resulting in the production of sodium aluminosilicate hydrate (NASH) and calcium aluminosilicate hydrate (CASH) gels, along with calcium silicate hydrate (CSH), which together enhanced the mechanical properties of the CKCGDP mortars.

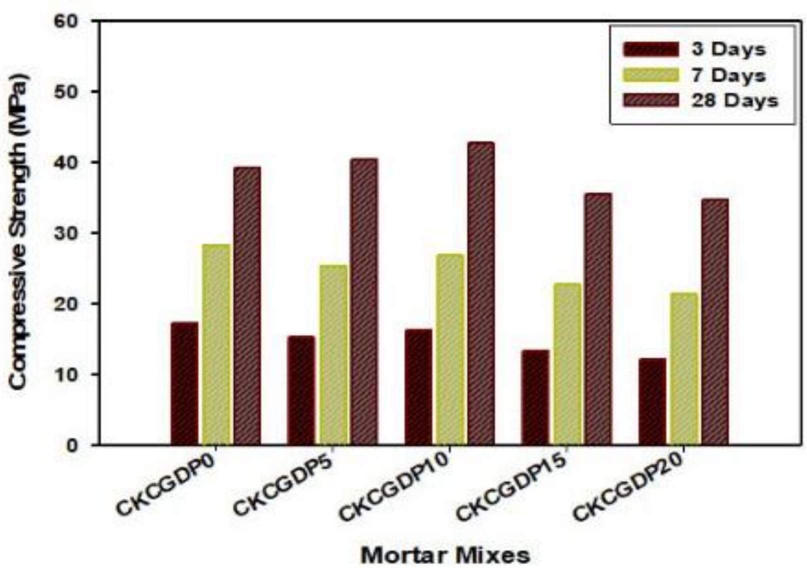

Figure 6.
Compressive Strength of Alkali-Activated CKC, Alkali-Activated CKC+GDP Mortars Mixes at Different Ages.

3.6. Flexural Strength

The correlation between tensile and compressive strengths of concrete or mortar is influenced by the overall compressive strength of the material. This research focused on measuring the flexural strength of OPC mortar, as well as geopolymer mortars formulated from CKC and a combination of CKC with GDP. Fig. 7 illustrates the outcomes of flexural strength tests performed on alkali-activated GDP and the alkali-activated CKC+GDP blended binder mortars following 28 days of curing.

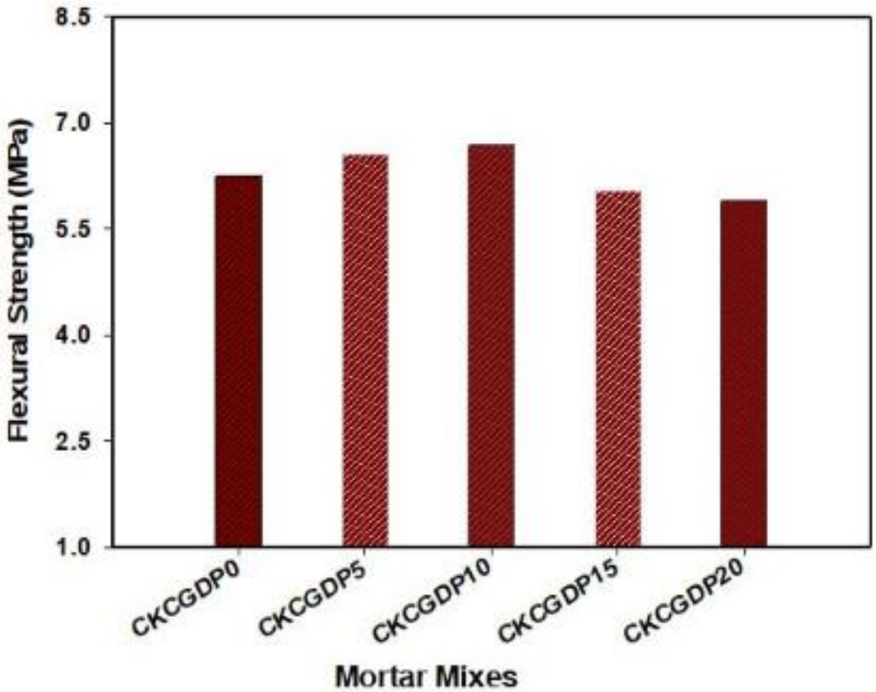

Figure 7.
Flexural Strength of Alkali-Activated CKC, Alkali-Activated CKC+GDP Mortars Mixes at 28 Days.

At 28 days, the mortar with a 10% replacement of CKC by GDP demonstrated a notable improvement in flexural strength compared to the control mortar (CKCGDP0), as illustrated in Fig. 7. The geopolymerization process enhanced the dissolution of SiO_2 and Al_2O_3, resulting in the formation of both CASH and CSH gels. This dual gel formation accounts for the superior strength observed in the CKCGDP10 sample, whereas the CKCGDP0 sample relies solely on calcium silicate hydrate (CSH) for its strength. The flexural strength trend aligns closely with the compressive strength results, consistent with findings reported in previous studies [46]. Generally, the ratio of flexural strength to compressive strength in concrete typically falls within the range of 10% to 15% [47] although this ratio varies for mortar. In the case of CKCGDP10 mortar activated with NaOH, the ratio was measured at 15.4%.

4. Conclusions

Based on the data obtained and the results of this study, the following conclusions can be drawn:

1. The results of specific gravity showed that considered pozzolanic and waste materials) calcined kaolin clay (CKC) and granite dust powder (GDP)) were lighter than cement.
2. The results for flow and slump demonstrated that by adjusting the water-to-binder ratio, both the flow and slump of the mortar mixes could be effectively maintained within the target ranges of 40±5% and 65±20 mm, respectively. Binary and ternary cementitious blends of calcined kaolin clay and granite dust powder one thing was noticeable that while increasing the replacement level

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of CKC and GDP, the slump and flow values are gradually decreasing in comparison with high performance control mortar (HPCM). High performance mortars with binary blends of 20CKC showed lowest flow and slump values followed by mixtures incorporating binary blends of 10CKC, 10GDP, 20GDP and ternary blends of 10CKC10GDP, 10CKC20GDP in comparison with control mixture.

3. The bulk density of the samples was found to rise as the proportion of CKC substituted by GDP increased, when preparing the geopolymer mortars, in comparison to the CKCGDP0 mixture.
4. Mixes containing GDP showed a slight decrease in strength at early curing stages (3 and 7 days), with greater reductions observed as the GDP content increased compared to the control mix without GDP (CKCGDP0). However, by 28 days, mortars incorporating 5% and 10% GDP as partial replacements of calcined kaolin clay demonstrated a notable improvement in compressive strength relative to the control mortar. Conversely, mortar samples containing 15% and 20% GDP as a replacement for CKC exhibited a decline in compressive strength starting from day 3 when compared to the CKCGDP0 sample.
5. The flexural strengths value of mortar with a 10% replacement of CKC by GDP demonstrated a notable improvement compared to the control mortar (CKCGDP0) at 28 days.
6. The findings suggest that CLB developed with CKC and GDP activated chemically is a promising sustainable alternative to cement, especially for structures where early high strength is not critical.
7. Generally, CLB developed with CKC and GDP activated chemically is a promising sustainable alternative to cement, especially for structures where early high strength is not critical. The development of could help reduce the demand for cement and consequently lower CO_2 emissions and environmental pollution by utilizing these waste materials effectively.

Symbols and Abbreviations

OPC	Ordinary Portland Cement
CKC	Calcined Kaolin Clay
GDP	Granite dust powder
CLB	Cementless Binder
SP	Superplasticizer
W/B	Water-to-Binder ratio
S/B	Sand-to-Binder ratio
LOI	Loss on ignition
RHA	Rice Hush Ash
FA	Fly Ash
POFA	Palm Oil Fuel Ash

Transparency:

The authors confirm that the manuscript is an honest, accurate, and transparent account of the study; that no vital features of the study have been omitted; and that any discrepancies from the study as planned have been explained. This study followed all ethical practices during writing.

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