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Innovations in the Cement Industry: Advancing Towards Sustainable Concrete Production

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Abstract

Slag, an artificial pozzolan derived from the by-product of metal ore smelting, exhibits notable richness in active silica and alumina. This investigation scrutinizes the influence of slag as a clinker substitute on cement quality. The study explores the composition range of slag from 15.69% to 20.39% of the total cement mass. Other constituent materials, including gypsum (2.07%), limestone (17.24-22.54%), and clinker (55-65%), are compared with commercial cement available in the market, featuring 73.09% clinker, 2.07% gypsum, 11.89% limestone, and 12.95% trass. The study employs rigorous physical tests, encompassing Blaine, residue at 45 μm (325 mesh), flowability, setting time, and compressive strength assessments at 1, 3, 7, and 28 days. The Blaine testing, the residue at 45 μm, and other tests were conducted meticulously to ensure the accuracy and reliability of the results. Blaine testing measured specific surface area, residue at 45 μm assessed fineness, flowability evaluated paste workability, setting time determined paste stiffness, and compressive strength assessed material durability. All tests followed ASTM standards, providing reliable insights into cement performance. Compared to the commercial sample, there is a decrease in compressive strength in all results with an increase in the amount of slag. This condition is because components such as tricalcium silicate (C3S) and dicalcium silicate (C2S), which play a crucial role in producing hydration strength, decrease with the reduction of clinker. However, the compressive strength specified by ASTM C 109/109M-01 280 kg/m^2 standard is still exceeded. Optimal results are obtained with a slag substitution of 16.64%, resulting in compressive strength of 305 kg/m^2 at 28 days. This investigation highlights slag as a promising substitute for clinker in cement production due to its high silica content. Slag offers several advantages, including enhanced sustainability by reducing environmental impact and lowering production costs. Its utilization diversifies raw material sources, promoting industry resilience.

Keywords: composite cement; greenhouse gas; slag; XRD

1. Introduction

In the dynamic landscape of the global cement industry, a paradigm shift toward sustainable practices is unfolding, with a particular focus on the development of environmentally conscious concrete products. This progressive trend is evident in ongoing innovations, both on a global scale and specifically within the context of Indonesia (Ahmad et al., 2022; Rahman and Maulana, 2023). One crucial aspect of this innovation is exploring and implementing alternative materials aimed at replacing clinker as a shift from traditional cement production methods. (Rahman and Mulyani, 2023).

Clinker, renowned as the primary raw material for cement, has long been associated with substantial energy consumption in its production (Cicek et al., 2020). Recognizing the adverse environmental impacts of processes that consume substantial energy, the cement industry has strategically implemented alternative materials to reduce energy consumption significantly during cement production (Tole et al., 2022). In addition to providing direct benefits to energy efficiency, such substitution also has the potential to reduce greenhouse gas emissions, mainly CO_2, thus aligning with global goals to mitigate environmental impacts from industrial processes (Sivakrishna et al., 2020). Therefore, a transformative transition towards replacing clinker with alternative materials such as slag is important in efforts to produceproducing environmentally friendly cement.

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Among the alternative materials that stand out in this context are fly ash, silica fume, trass, and slag. However, they focused more on investigating the effect of a combination of fly ash and slag as a substitute for clinker (A.A. Shubbar et al., 2020; Saly et al., 2018). Other researchers mention that these materials exhibit pozzolanic properties comparable to traditional cement components, indicating their suitability for integration into the production process. (Tole et al., 2022). Other researchers have reported that slag is a by-product of blast furnaces, an integral part of iron production. The chemical composition of slag includes the main components of cement, such as CaO, SiO2, Al2O3, and MgO. (Czop and Lazniewska-Piekarczyk, 2020).

Distinguished by its distinctive physical characteristics' white appearance, fineness exceeding 350 m^2/kg, and a bulk density of 1200 kg/m^3, slag emerges not only as a potential clinker substitute but also as a reservoir of active silica and alumina compounds instrumental in the final cement grinding process (Mardani-Aghabaglou et al., 2017). The chemical interactions of the active silica within slag, particularly its reaction with calcium hydroxide, culminate in the formation of calcium silicate hydrate (CSH) and calcium hydroxide (Arabi et al., 2015). While hydrated calcium silicate contributes to enhanced concrete compressive strength and decreased susceptibility to cracking (Czop and Lazniewska-Piekarczyk, 2020), the resulting calcium hydroxide may introduce contrasting effects. This dichotomy underscores the imperative need for precise calculations when integrating alternative materials to ensure the produced cement aligns with stringent quality standards.

Considering this advancement's growing significance, this study explores the effects of slag as a replacement for clinker in cement production. By rigorously examining physical and chemical parameters and comparing them against established standards, the research aims to contribute substantially to the scientific comprehension of slag's role as a clinker substitute in modern cement manufacturing.

2. Methodology

2.1. Material

The raw materials used are Portland Composite Cement (PCC) from a cement company in the Cibinong area, Bogor, Indonesia, including clinker, limestone, and slag. All materials are ground to a target Blaine value of 4000 cm^2/g, except for gypsum, which has a Blaine value of 5000 cm^2/g to ensure uniform dispersion in the cement mixture. The method employed is separate grinding using a tube mill with grinding time based on the target Blaine.

2.2. Blaine Fineness Evaluation

The assessment of cement fineness, specifically Blaine, was conducted utilizing the Tony Blaine apparatus. This instrument incorporates air suction and a particle size function to measure the fineness of the cement particles quantitatively. The Blaine test methodology adhered to the specifications outlined in the ASTM C204-00 standard. This standard delineates the prescribed test procedure for evaluating the fineness of hydraulic cement using an air permeability apparatus. The Tony Blaine tool, by ASTM C204-00, played a pivotal role in elucidating the crucial parameter of Blaine fineness during this investigation.

2.3. Residue Analysis at 45 μm

The assessment of cement fineness encompasses not only the Blaine value but residue testing using an Alpine sieve with a mesh size of 45 μm (325 mesh). This method provides a complementary perspective by evaluating the portion of cement particles exceeding the specified size. Cement particles with a broader size distribution indicate varied retention characteristics on the sieve surface. Residue testing refers to ASTM standard C430-96, which outlines a systematic procedure for assessing the fineness of hydraulic cement. This methodological approach contributes to a comprehensive understanding of cement particle size distribution, ensuring a thorough evaluation of its fineness characteristics.

2.4. Flowability Examination

The assessment of flowability refers to ASTM C230 for hydraulic cement mortar flow, conducted using a precision flow table. This equipment provides a detailed evaluation of the flow characteristics of mortar mixtures. Flowability is critical in hydraulic cement as it directly impacts workability during construction. High flowability ensures easy placement and compaction, improving construction efficiency and concrete quality. Conversely, high flowability may lead to better consolidation and compromised structural integrity. Overall, assessing flowability is essential for optimizing construction practices and ensuring the success of hydraulic cement applications.

2.5. Setting Time Analysis

The evaluation of setting time refers to ASTM standard C191-01a for the setting time of hydraulic cement using a Vicat needle apparatus. Setting time is a vital parameter in cement characterization, indicating the duration for the mixture to transition from a plastic to a specific stiffness level. This test determines both the initial and final setting times. The initial setting time measures the time between the initiation of the cement-water mixture and the point where the material reaches a particular hardness. This determination utilizes a Vicat needle to measure the time required for penetration to a depth of 25 mm into the cement paste.

On the contrary, the final setting time indicates the point where the Vicat needle penetration is no longer visible. Careful preparation of the paste, which is an integral part of this evaluation, is required. This research necessitates precisely 500 g of cement with an accuracy of 0.1 g. A systematic approach to setting time analysis is based on ASTM C191-01a.

2.6. Compressive Strength

The determination of compressive strength for PCC mortar refers to ASTM standard C 109/109M-01. This method utilizes cube molds with a side size of 50 mm. The material mass ratio for the mortar is cement: sand: water = 1: 2.75: 0.485. To prepare the test samples for this study, 740 g of cement, 2035 g of sand, and 359 g of water are used. Immediately after the test specimens are molded, they are stored in a humid room for 24 hours.

2.7. Chemical Composition Analysis

The examination of the chemical composition of the samples involved a comprehensive set of analyses, including Lost on Ignition (LOI), Insoluble Residue (IR), X-ray diffraction (XRD), and X-ray fluorescence (XRF) tests. In the LOI test, the specimen underwent ignition within a controlled heating furnace set at 950 ± 50 °C. Pre- and post-annealing weight measurements were taken, and the disparity in weight delineated the quantifiable presence of water and carbon dioxide within the sample.

The Insoluble Residue test necessitated dissolving the sample in hydrochloric acid (HCl) solvent, resulting in the formation of a precipitate. Subsequently, the precipitate was subjected to incandescence at a temperature of 1000 ± 50 °C, and its weight was measured. The weight of the remaining sample indicated the presence of free SiO_2, which exhibited insolubility in acid.

XRF analysis is conducted to determine the significant and minor oxide components according to SNI 15-2049 standard for Portland cement quality and ASTM SPT 985 for rapid analysis of hydraulic cement. The chemical reagents used in this analysis are Lithium Fluoride, with a purity level of up to 99.9%, and Lithium Bromide, with a concentration of 25%. This analysis provides an understanding of the chemical composition of the investigated samples.

3. Results and Discussion

3.1. Preparation of PCC Samples

In this investigation, a total mass of PCC weighing 5000 g was prepared consisting of clinker, gypsum, limestone, and slag. All components are milled simultaneously with a Herzog tube mill. The grinding process is carefully calibrated to achieve more than 2500 g/cm^2 fineness by SNI 7064:2014 standards. The PCC formulation was prepared with 6 samples labeled PCC1 to PCC6. In this study, the proportion of slag varied, ranging from 15.69% to 20.39%, clinker varied from 65% to 53%, and limestone varied from 17.24% to 22.54%, while the composition of gypsum was determined to 2.07%. This systematic approach shows the correlation of slag substitution in the overall composition of PCC samples, as presented in Table 1.

Table 1. Proportion of PCC

Sample Code	Total mass of mixture 5000 g			
	Clinker	Gypsum	Limestone	Slag
*Com.	3654.5	103.5	594.5	647.5 (trass)
PCC1	3250.0	103.5	862.0	784.5
PCC2	3150.0	103.5	914.5	832.0
PCC3	3050.0	103.5	967.5	879.0
PCC4	2950.0	103.5	1022.0	924.5
PCC5	2850.0	103.5	1074.5	927.0
PCC6	2750.0	103.5	1127.0	1019.5

*Com. = Commercial utilizes trass 12.95% to replace clinker

3.2. Physical Analysis of PCC Samples

In this study, the evaluation of PCC samples encompassed a comprehensive array of physical tests. These assessments comprised a determination of the Blaine fineness, an examination of the residue at 45 µm (equivalent to 325 mesh), an analysis of setting time characteristics, an assessment of flowability, and a quantification of compressive strength.

3.2.1 Blaine and Residue 45 µm Analysis

The fineness of cement, measured using the Blaine method, refers to ASTM standard C204, while the quantification of residue retained on the 45µm sieve follows the methodology outlined in ASTM C340. The results indicate that fineness significantly influences particle-to-particle bonding within the cement matrix (Faltin, 2022). Increased fineness is associated with greater surface area, thereby impacting the reactivity of cement with water. As a result, finer cement particle structures are linked to cement quality.

Blaine fineness and residue analysis at 45 µm are crucial in assessing cement quality, providing insights into particle size distribution and fineness. Blaine fineness measurement determines the surface area of cement particles, impacting hydration rate, strength development, and overall performance. A higher Blaine fineness signifies better reactivity and faster setting times. Meanwhile, residue analysis evaluates the proportion of coarse particles, influencing workability, setting time, and durability. Lower residue values indicate improved particle distribution and cement performance. These tests ensure uniformity and consistency in cement production, resulting in high-performance concrete suitable for construction applications (Faltin, 2022).

Table 2 delineated that the introduction of slag reduced clinker utilization while concurrently necessitating increased incorporation of limestone. The composite material, comprising clinker, limestone, and slag, underwent individual grinding to achieve a Blaine target of 4000 cm^2/g. By ASTM C 150, Blaine targets ranged between 3000 and 5000 cm^2/g. Exceeding the upper limit of this range, particularly with Blaine values surpassing 5000 cm2/g, significantly accelerated the hydration reaction of cement, resulting in a shortened setting time (Rahman and Rahayu, 2021). Notwithstanding, the specified Blaine target exhibited a correlated effect on energy consumption at the finish mill (Ghalandari and Iranmanesh, 2020). Elevated Blaine targets necessitated more significant energy input for material refinement owing to prolonged processing durations in the finish mill and heightened utilization of grinding media in the form of steel balls.

This investigation employs different grinding approaches to achieve uniform Blaine values for each constituent material. Differences in grinding capabilities arise from varying hardness levels caused by chemical composition variations. Slag is the most challenging material due to its high silica content. (Markandeya et al., 2018; Rahman and Rahayu, 2021).

The test findings revealed that, despite a marginal reduction in the Blaine value with the inclusion of slag, it remained significantly above the stipulated threshold outlined in SNI 7064:2014, set at 2800 cm^2/g. Conversely, a decrease in Blaine value corresponded to a slight elevation in the residual value. The intricate nature of slag, surpassing that of clinker and limestone, imparted this dual effect, a reduction in Blaine value, and an increase in residual value (Czop and Lazniewska-Piekarczyk, 2020). The hygroscopic characteristics of limestone further contributed to this phenomenon by attenuating Blaine and elevating residual values, as the water content hindered the air permeability of the cement.

Table 2. Physical Properties of PCC Samples

Sample Code	Blaine	Residue 45 µm	Setting Time (min.)	
	cm^2/g	%	initial	final
Com.	4154	13.04	130	330
PCC1	4174	17.33	140	350
PCC2	4186	13.55	140	350
PCC3	4160	13.92	140	350
PCC4	4171	17.03	140	350
PCC5	4163	15.63	140	350
PCC6	4093	15.47	140	350

3.3. Flowability Analysis

The assessment of workability in this study follows ASTM C230. Workability, evaluated through slump tests, indicates the cement's capacity for easy placement (Rahman and Mulyani, 2023). As explained by (Singh et al., 2015), the concept of flowability is closely related to water usage, so increasing the water content in cement paste production results in commendable workability (Dai et al., 2020). According to ASTM C230, the recommended slump flow value is set at (110 ± 5) cm.

Figure 1 shows that substituting clinker with slag up to 16.64% results in nearly the same water quantity as the commercial sample for mortar production. This observation indicates the potential feasibility of slag as an alternative material for eco-friendly concrete formulations. (Czop and Lazniewska-Piekarczyk, 2020). Importantly, all test outcomes align with the established standards, demonstrating consistent achievement of the prescribed workability threshold of 110 ± 5 cm.

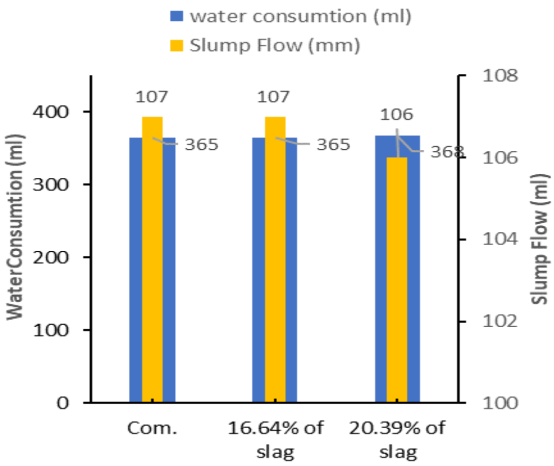

Figure 1. Effect of Slag on Water Consumption and Slump Flow

3.4. Setting Time Analysis

The determination of setting time in this investigation adhered to the guidelines outlined in ASTM C191-01a, the Standard Test Method for Setting Hydraulic Cement by Vicat Needle. A paste is formed upon the amalgamation of cement with water, and the ensuing rigidity in the paste, denoted as setting time, is expressed in minutes. The onset of stiffness is termed the initial setting time, while the final setting time signifies the point at which the cement attains complete stiffening. Notably, in contrast to gypsum, the inclusion of slag material does not impart any discernible impact on the setting time of cement (Ofuyatan et al., 2020).

To maintain consistency, a constant gypsum composition was employed in this study to ensure its neutrality toward setting time. The binding time assessments across all sample types consistently yielded results within the range of 130-140 minutes for the initial setting time and 330-350 minutes for the final setting time, as elucidated in Table 2 and Figure 2. This compellingly suggests that substituting slag for clinker does not introduce any significant alterations to the setting time of the cementitious system under examination.

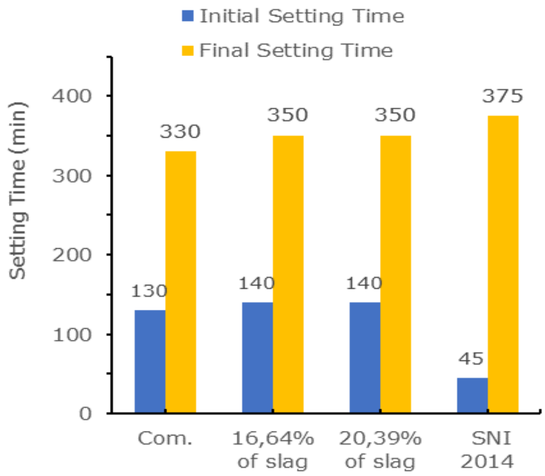

Figure 2. Effect of Slag on the Initial and Final Setting Time

3.5. Compressive Strength Analysis

The determination of mortar compressive strength in this investigation adheres to the ASTM C109/109M-01 standard, which is the standard for hydraulic cement compressive strength also conducted by (Yang et al., 2023). Compressive strength, indicating the ability to withstand loads, is crucial in assessing cement quality. Testing is conducted at mortar ages of 1, 3, 7, and 28 days (Penpichcha et al., 2019), with this study focusing on the analysis of composite mortar compressive strength consisting of cement, sand, and water.

The test results indicate a decrease in compressive strength with increased slag proportion and decreased clinker, as detailed in Table 3. In other words, substituting slag for clinker in the cement mixture can affect compressive strength. This case is because clinker is a primary material in cement production and directly contributes to its mechanical properties. Clinker contains components such as tricalcium silicate (C3S) and dicalcium silicate (C2S), which play a crucial role in forming strong hydration products, such as calcium silicate hydrate (CSH) and calcium hydroxide (CH) (Jifeng et al., 2022).

However, the CaO content in limestone interacts with SiO_2 to produce the formation of Calcium Silicate Hydrate (CSH), so the decrease in compressive strength does not fall below the minimum compressive strength standard for PCC of 250 kg/cm^2 (Zunino and Scrivener, 2022).

Slag, recognized for its pozzolanic attributes, boasts a composition rich in SiO_2, CaO, MgO, and Al_2O_3 (He et al., 2022). Notably, the Al_2O_3 content in slag contributed to a reduction in compressive strength values (Ofuyatan et al., 2020). This nuanced interplay of constituents underscores the complex influence of material composition on mortar compressive strength, elucidating the multifaceted considerations integral to optimizing concrete formulations for desired mechanical properties.

Table 3. Compressive Strength of PCC samples

Sample Code	Compressive Strength (kg/cm^2)			
	Day 1	Day 3	Day 7	Day 28
Com.	104	197	248	341
PCC1	88	176	231	319
PCC2	81	161	213	316
PCC3	74	151	202	305
PCC4	65	140	188	299
PCC5	56	128	178	287
PCC6	50	119	164	264

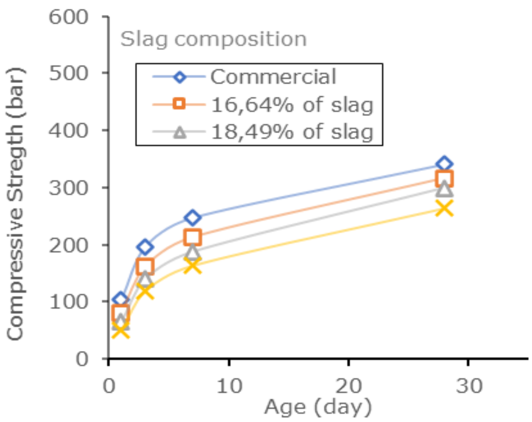

Figure 3. Effect of Slag Substitution on Compressive

The interaction among elements that influence compressive strength in cement-based materials provides a profound understanding of their performance. Figure 3 illustrates a systematic decline in compressive strength as the clinker percentage decreases, highlighting the importance of studying the underlying chemical mechanisms. This phenomenon can be linked to decreased concentration of crucial compounds in cement, particularly tricalcium silicate (C3S) and dicalcium silicate (C2S), as depicted in Figure 4. These compounds play a crucial role in the hydration process of cement, forming calcium silicate hydrate (C-S-H) gel and calcium hydroxide (CH), which significantly contribute to the development of concrete strength. However, with the introduction of slag as a clinker substitute, there is a simultaneous decrease in the concentrations of C3S and C2S, thereby affecting compressive strength (Zunino and Scrivener, 2022).

Nevertheless, adding slag up to 20% remains within the specified compressive strength criteria, according to the SNI 7064:2014 standard, at various test ages. This resilience underscores the feasibility of utilizing slag as a partial clinker substitute while meeting industry standards' compressive strength requirements. Understanding these intricate chemical interactions provides valuable insights into the performance of cement-based materials and the potential of alternative materials like slag in cement production.

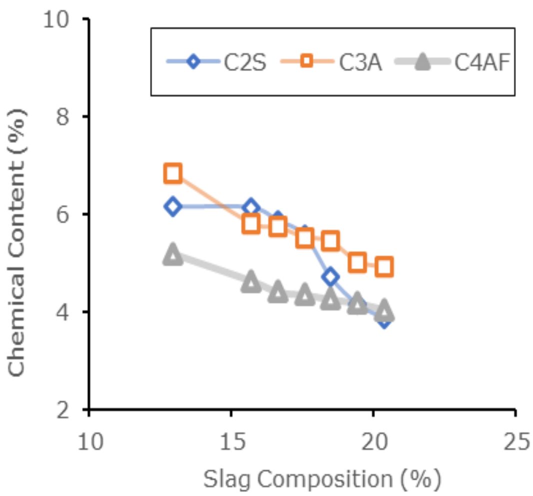

Figure 4. Effect of Slag Substitution on Main Mineral Cement

3.6. Chemical Content Analysis of PCC Samples

Chemical Content Analysis of PCC Samples: The chemical composition of the cement samples was assessed using X-ray fluorescence (XRF) to quantify major and minor oxides, following the methodology established by (Selma et al., 2017; Vicenzi & Lam, 2017). Principal oxides, encompassing SiO_2, Al_2O_3, and CaO, were examined, while minor oxides, specifically MgO, SO_3, F-CaO, K_2O, Na_2O, and total alkali (TA), were also considered by the framework outlined by (Galloway et al., 2015). However, this study focused solely on analyzing SO_3 to ascertain its compliance with the stipulated requirements (Nadezhda et al., 2018). The chemical test adhered to the regulatory provisions outlined in SNI 7064-2004, wherein the maximum allowable SO_3 content is stipulated at 4.0% (Rahman et al., 2021; Simoni et al., 2021).

Table 4. Chemical Contents Analysis

Sample Code	Chemical Contents			
	SiO_2	Al_2O_3	CaO	SO_3
Com.	21.28	6.09	55.73	1.78
PCC1	21.20	6.32	54.20	1.86
PCC2	21.22	6.38	54.19	1.89
PCC3	21.26	6.40	54.15	1.86
PCC4	21.14	6.45	54.01	1.89
PCC5	21.09	6.44	53.89	1.85
PCC6	21.08	6.49	53.61	1.86

The results of the chemical content analysis for PCC1 – PCC 6 are presented in Table 4, elucidating the adherence of the samples to the prescribed limits for SO_3 content as per the regulatory standards.

The composition of SO_3 (sulfate) in composite cement significantly influences its quality due to its impact on several fundamental properties. Excessive SO_3 levels can accelerate the setting time of the cement, leading to difficulties in handling and application. High SO_3 content can also contribute to ettringite formation, a compound that may cause expansion and cracking in concrete, particularly in sulfate-rich environments. On the other hand, controlled amounts of SO_3 can enhance the strength and durability of the composite cement by improving its resistance to sulfate attack and mitigating the risk of alkali-silica reaction (Rahman et al., 2021). Therefore, maintaining the SO_3 composition within specified limits is essential for ensuring the composite cement's desired performance, workability, and durability in construction applications.

4. Conclusion

A comprehensive analysis of the testing and observations indicates that replacing clinker with slag up to 20% of content consistently meets the established standards. Despite a decrease in compressive strength, one of the primary indicators of cement quality, across all testing stages, it still fulfills the criteria set by ASTM C 109/109M-01. The highest compressive strength result, reaching 305 kg/m^2, was achieved with a slag substitution of 16.64%. Furthermore, the fineness results are highly satisfactory, with values exceeding 4000 cm^2/g, significantly surpassing the established standard of 2500. This research holds significant value in the context of sustainable development through the more efficient utilization of natural resources.

Author contribution statement

Conceptualisation, H.R.,; Formal analysis, M.R,; Investigation, M.R.,; Project administration, M.R.,; Supervision, M.R.,; Writing – original draft, M.R.,; Writing – review and editing, H.R..

Data availability statement

Available on request to the corresponding Author.

Declaration of Competing Interest

The author declares that there is no conflict of interest regarding the publication of this manuscript.

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