The Effect of Precursor-Activator Ratio and Activator Type on the Bulk Density, Compressive Strength, and Microstructure of Abuan Pumice-Based Geopolymer Binder

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ABSTRACT

This research developed a geopolymer binder based on pumice waste from Abuan Village, Kintamani, which is rich in silica and alumina, with a composition of SiO_2 55.90%, Al_2O_3 12.00%, CaO 6.25%, and other metals around 8.61%. The activators used are Na_2SiO_3 combined with either NaOH or KOH at a concentration of 12 M, the precursor-activator (P/A) ratios used were 75%:25%, 70%:30%, and 65%:35%, with 75%:25% representing the highest P/A ratio and an alkali ratio of NS/NH at 2:1. Cube samples 50 x 50 x 50 mm were analyzed to measure bulk density and compressive strength at 7, 14, and 28 days after being cured in an oven at 80°C for 72 hours. The bulk density test results for samples using NaOH showed an increase as the P/A ratio increased, but a decrease as the testing age increased. Then, the bulk density results for KOH samples showed the opposite trend both in terms of P/A ratio and age. The average compressive strength test for NaOH samples increased with the P/A ratio, and while it fluctuated by age, it continued to increase overall. The KOH samples followed a similar trend in P/A ratio but tended to decrease over time. This behavior is influenced by the chemical and physical properties of the activators used. Microstructural analysis was conducted using X-Ray Diffraction (X-RD) on the samples with the lowest and highest compressive strength. New minerals formed in NaOH samples, including Sodium Aluminosilicate, Albite, and Anorthite, while KOH samples showed formations of Dipotassium Sulfate (VI) – Alpha, Ht, Albite, and Cristobalite.

Keywords: Bulk Density, Compressive Strength, Geopolymer Binder, Microstructure, Pumice

1. INTRODUCTION

Indonesia is known using concrete as the primary construction material in various projects. Concrete typically consists of aggregate, cement, and water, with cement acting as the adhesive that binds other components to form a strong structure [8]. However, the production of cement,

which requires high-temperature firing, it causes CO_2 emissions, contributing to global warming. For every ton of cement produced, approximately 1 ton of CO_2

To mitigate environmental impact, alternatives to cement, such as geopolymer binders, are needed. The term "geopolymer" was introduced by Davidovits in 1978 [3] and involves the polymerization of materials such as fly ash and rice husk ash. Geopolymers reduce dependence on Portland cement, making them a more environmentally friendly option [4]. Pumice, which contains silica and alumina, can serve as a geopolymer precursor. In Bali, pumice is abundant in Abuan Village, Kintamani, though its usage remains limited.

This research aims to explore the use of local materials in producing geopolymer binders. The main focus will be on testing bulk density, compressive strength, and microstructure with variations in the types of activators used, with the hope of providing a new, more sustainable alternative in concrete technology.

2. THEORY AND METHODS

2. 1 Theory

A. Geopolymer

Geopolymer is an inorganic alumino-silicate material synthesized from natural resources or industrial waste rich in silica (Si) and alumina (Al). In the synthesis process, these atoms form a rock-like structure through a chemical reaction between alkali and Si-Al, resulting in consistent Si-O-Al-O bonds [3]

The term “geopolymer” was first introduced by Professor Davidovits in 1978 [3]. Geopolymer concrete does not use cement as a binder; instead, fly ash, which is rich in Si and Al, reacts with an alkaline solution to produce a binding material.

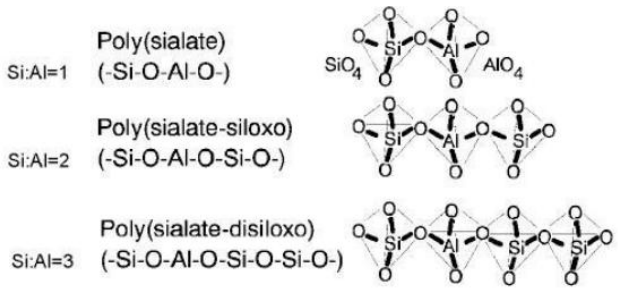

Figure 1 polymerization reaction

B. Precursor

A precursor is the primary material used as a binder in the formation of geopolymers, sourced from natural materials or industrial waste. The formation process (synthesis) involves the activation of alumina-silica materials by alkaline ions and a curing process to promote the polymerization of alumina-silica monomers into a three-dimensional network [10. The precursor can come from materials that contain high levels of alumina and silica [1].

C. Activator

The production of geopolymers requires activators to facilitate the condensation polymerization reaction. The commonly used activator compounds are NaOH and KOH. Typically, NaOH activator yields higher compressive strength compared to KOH, even at the same concentration. Solid activators are dissolved in water according to the desired molarity [11].

D. Pumice

Pumice is a silica-rich rock formed from volcanic eruptions, characterized by its porous structure due to trapped steam and gas during cooling. Pumice is used as an alternative material in various industries, such as lightweight concrete aggregate and abrasive materials, and its high porosity allows it to float in water [9].

This research utilizes pumice from Abuan Village, Kintamani, Bangli, which is commonly used by artisans for making shrines (pelinggih). The production process of these shrines generates unused pumice waste, which the author intends to utilize as a precursor for geopolymer binders.

Figure 2 Abuan Pumice

E. Bulk Density

Bulk density is the value obtained from measuring the weight and volume of a sample. The calculation of bulk density is performed using the following formula:

$$\gamma = \frac{w}{v} \qquad (1)$$

Where : γ = bulk density (gr/cm^3)
w = weight (gr)
v = volume (cm^3)

F. Compressive Strength

Compressive strength is the ability of concrete to withstand compressive forces per unit area and serves as an indicator of structural quality. The higher the desired strength, the better the quality of the concrete required. Compressive strength is determined based on the maximum compressive stress (P) achieved after 28 days. The calculation of concrete compressive strength can be performed using specific formulas.

$$f'c = \frac{P}{A} \qquad (2)$$

Where: $f'c$ = Compressive strength (MPa)
P = maks. load (N)
A = area (mm^2)

G. X-RF (X-Ray Fluoresence)

The X-RF analysis method is used to identify elements within a material, particularly minerals or rocks. This testing can be performed qualitatively to recognize the types of elements present and quantitatively to measure the concentration of those elements in the material [5].

H. Microstructure

Microstructure testing involves analyzing the smallest structure of a material, providing insights into its chemical composition, crystal structure, and surface morphology [7]. This analysis is crucial for evaluating how the interactions between chemical components and physical properties affect the final characteristics of the geopolymer. Information obtained from this testing helps to understand how the internal structure and particle distribution influence the quality and performance of the geopolymer.

I. X-RD (X-Ray Diffraction)

X-RD is a characterization method that uses X-rays to analyze the structure and crystal size of a material through X-ray diffraction spectra [6]. This process generates an intensity peak graph that forms a diffraction pattern, which is then compared to patterns from known samples for identification. X-ray Diffraction is used to determine the structure of crystalline solids, with powdered samples placed on a glass plate with crystal sizes ranging from approximately 10^{-7} to 10^{-4} m. The X-rays are generated from electrons emitted from a heated filament under high voltage in a vacuum environment [2].

2.2 Material and Methods

A. Material

The precursor in this study is Abuan pumice powder, sieved to 200 μm (Figure 3). XRF analysis (Table 1) reveals its mineral content SiO_2 (55.90%), Fe_2O_3 (17.40%), Al_2O_3 (12.00%), CaO (6.25%), and other minerals (8.61%). With over 50% silica and alumina, the geopolymer precursor is expected to be high quality. The alkali activators used are sodium silicate (Na_2SiO_3), sodium hydroxide (NaOH) at 12 M concentration, and potassium hydroxide (KOH). The geopolymer binder ratios are based on prior research and include P/A ratios of 75%:25%, 70%:30%, and 65%:35%, with a sodium silicate to sodium hydroxide ratio of 2:1.

Table 1 XRF test result of Abuan pumice powder

Compound	Percentage (%)
SiO_2	55,9
Fe_2O_3	17,4
Al_2O_3	12
CaO	6,25
K_2O	4,70
TiO_2	1,74
P_2O_5	0,91
MnO	0,48

SrO	0,26
Eu_2O_3	0,26
Re_2O_7	0,1
CuO	0,064
ZnO	0,04
Cr_2O_3	0,039
V_2O_5	0,02

Figure 3 Abuan pumice powder

B. Methods

The research began with a literature review, analyzing studies on geopolymer adhesives and activator variations, followed by fieldwork to gather data on pumice waste. Primary data included Abuan pumice powder as precursor and activators (Na_2SiO_3, NaOH, and KOH), characterized using XRF, XRD, and tests for bulk density and compressive strength. Bulk density and compressive strength tests are conducted at 7, 14, and 28 days, with 5 samples for each variation as shown on Table 2

A trial-and-error process was used to determine the optimal geopolymer mix formulation. Pumice was used as the primary precursor, with Na_2SiO_3 combined with either NaOH or KOH as activators. The choice of activators was based on availability and ease of procurement to ensure smooth research progress. Precursor-activator ratios of 75%:25%, 70%:30%, and 65%:35% were selected based on literature and trial results. At a ratio of 80%:20%, the mixture's workability was poor, becoming too thick and difficult to mix. Conversely, at 60%:40%, the mixture was too runny, leading to activator leakage during drying and dimensional shrinkage. Therefore, the ratios of 75%:25%, 70%:30%, and 65%:35% were chosen as the most optimal in terms of workability, dimensional stability, and mechanical strength.

The molarity of NaOH and KOH solutions was also varied to determine the optimal concentration. At 10M, the solution was too dilute, resulting in low compressive strength. At 14M, the workability decreased significantly, making the mixture difficult to mix and too dry. Tests showed that 12M molarity provided the best balance between ease of mixing and mechanical performance. Additionally, curing temperature variations were tested. At 70°C, drying was slow, leading to incomplete geopolymerization and inadequate drying. At 100°C, rapid water evaporation caused thermal shrinkage and potential microcracking. Based on the trials, a curing temperature of 80°C was selected

as it was high enough to accelerate geopolymerization without causing structural damage. The curing duration was set at 3×24 hours based on trials ranging from 1×24 to 4×24 hours. At 1-2×24 hours, the material remained moist, while at 4×24 hours, the material became overly dry and brittle, making it prone to cracking when removed from the mold.

Table 2 The mixing proportion of Geopolymer Binder

Group	Code	Ratio P/A	Activator
I	S11	75% : 25%	NaOH
	S12	70% : 30%	NaOH
	S13	65% : 35%	NaOH
II	S21	75% : 25%	KOH
	S22	70% : 30%	KOH
	S23	65% : 35%	KOH

Figure 4 Mixing geopolymer binder

Figure 5 Casting the sample

3. RESULTS AND DISCUSSION

3.1 Bulk Density

The bulk density test results are shown in Figures 6 and Table 3, presenting the bulk density measurements at 7, 14, and 28 days for each sample.

Figure 6 Bulk density sample

The graph illustrates a decrease in bulk density as the P/A ratio decreases (from 65%:35% until 75%:25%) for samples S11, S12, and S13, with reductions of 0.3%, 0.8%, and 3% at 7, 14, and 28 days from a P/A ratio of 75%:25% to 70%:30%. A further decrease from 70%:30% to 65%:35% results in reductions of 0.3%, 0.5%, and 5.8% at the same ages. Conversely, samples S21, S22, and S23 experience an increase in bulk density as the P/A ratio decreases, with increases of 0.9%, 1.1%, and 2.2% from 75%:25% to 70%:30%, and increases of 1.1%, 1.9%, and 0.6% at ratio 70%:30% to 65%:35%. Age also plays a role in bulk density changes; S11, S12, and S13 show a consistent decrease over time, while S21, S22, and S23 exhibit an increasing trend as the testing age progresses.

Figure 7 Bulk density test result

Table 3 Bulk density test result

Sample's Code	Average bulk density (gr/cm3)		
	7 Days	14 Days	28 Days
S11	1,651	1,637	1,592
S12	1,646	1,624	1,544
S13	1,642	1,616	1,454
S21	1,549	1,574	1,589
S22	1,562	1,592	1,624
S23	1,579	1,622	1,634

3.2 Compressive Strength

Figures 7 and Table 4 shown compressive strength at 7, 14, and 28 days. The highest strength was at a P/A ratio of 75%:25%, with strength decreasing as the P/A ratio lowered. From 75%:25% to 70%:30%, strength dropped by 56%, 63.7%, and 58.5% at each age, and from 70%:30% to 65%:35%, it fell by 68%, 60.5%, and 63.2%. Samples S21, S22, and S23 showed similar trends, with compressive strength reductions across all ratios and ages. Additionally, strength in S11-S13 raise till 28 days, while S21-S23 consistently decrease from 7 to 28 days. The compressive strength test results for the geopolymer binder, regardless of the type of activator used, indicated relatively low values compared to conventional cement. As a result, this geopolymer binder is unsuitable for structural applications but is adequate for use in non-structural adhesive purposes

Figure 8 Compressive test

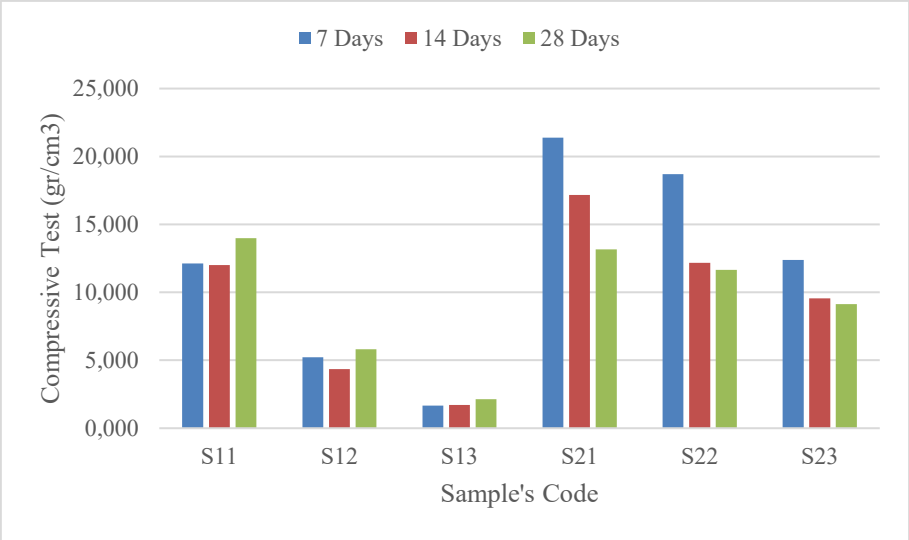

Figure 9 Compressive test result

Table 4 Compressive test result

Sample's Code	Average compressive test (MPa)		
	7 Days	14 Days	28 Days
S11	12,13	12,00	13,99
S12	5,23	4,35	5,81
S13	1,67	1,72	2,14
S21	21,38	17,16	13,15
S22	18,70	12,18	11,65
S23	12,38	9,57	9,14

3.3 Microstructure

The microstructure test used is X-Ray Diffraction (XRD). The XRD testing process, involving diffraction analysis, was conducted after the compressive strength tests. Initial XRD results for Abuan pumice powder revealed the presence of *Potassium Hexamanganoheptaarsenate* (V) ($As_7K_1Mn_6O_{24}$), *Anorthite* ($CaAl_2Si_2O_8$), *Magnetite low* (Fe_3O_4), and *Sodium Lanthanum Molybdenum Oxide* ($Na_{0.5}La_{0.5}(MoO_4)$). After being processed into geopolymer binder, new minerals formed in each tested sample, indicating significant changes in mineral composition due to the chemical reactions occurring during the setting and hardening of the geopolymer.

The samples tested included S11 with a precursor-to-activator (P/A) ratio of 75%:25% and a curing age of 28 days, S13 with a P/A ratio of 65%:35% at 7 days, S21 with a P/A ratio of 75%:25% at 7 days, and S23 with a P/A ratio of 65%:35% at 28 days. The results of the XRD analysis can be observed in the subsequent tables.

Table 5 X-RD result of KOH sample's

No.	Compounds	Chemical Formula	Intensity (%)	
			S21	S23
1	*Dipotassium Sulfate (VI) – Alpha, Ht*	$K_2O_4S_1$	16	40,4
2	*Albite*	$NaAlSi_3O_8$	82	49,5
3	*Cristobalite*	SiO_2	2	10,1

Table 6 X-RD result of NaOH sample's

No.	Compounds	Chemical Formula	Intensity (%)	
			S11	S13
1	*Sodium Aluminosilicate*	$Al_6Na_6O_{32}Si_{10}$	3	4,1
2	*Albite*	$NaAlSi_3O_8$	33	31,3
3	*Anorthite*	$CaAl_2Si_2O_8$	64	64,6

4. CONCLUSIONS

Based on the analysis and discussion results in this study, the conclusions that can be drawn are as follows:

1. In geopolymer binders with Na_2SiO_3 + NaOH activator, results show that higher P/A ratios lead to higher bulk density and compressive strength. Over time, bulk density decreases while compressive strength increases. After polymerization, compounds such as Sodium Aluminosilicate ($Al_6Na_6O_{32}Si_{10}$), Anorthite ($CaAl_2Si_2O_8$), and Albite ($Na(AlSi_3O_8)$) are formed.
2. In geopolymer binders with Na_2SiO_3 + KOH activator, results show that higher P/A ratios result in lower bulk density, while compressive strength increases. Over time, bulk density increases while compressive strength decreases. After polymerization, compounds such as Dipotassium Sulfate (VI) – Alpha, Ht (K_2O_4S), Albite ($Na(AlSi_3O_8)$), and Cristobalite (SiO_2) are formed.
3. The compressive strength test results for the geopolymer binder, regardless of the type of activator used, indicated relatively low values compared to conventional cement. As a result, this geopolymer binder is unsuitable for structural applications but is adequate for use in non-structural adhesive purposes.

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