

## Optimizing Mortar Mixtures with Basalt Rubble: Impacts on Compressive Strength and Chloride Penetration

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### Abstract

This research aims to establish a theoretical framework for developing binders from waste materials to reduce cement use in mortar production. It specifically examines the potential of ground basalt rubble (BS) as a supplementary binding material for partially replacing Portland cement Type 1 (OPC) in mortar mixtures. Various substitution ratios of BS, specifically 0%, 10%, 20%, 30%, and 40% by binder weight, were tested while maintaining a constant water-to-binder ratio (W/B) of 0.45. Superplasticizers (SP) were utilized to ensure consistent workability and flow of the mixtures. The SEM-EDS analysis was conducted to examine the microstructure of the cement paste, confirming the presence of calcium silicate hydrate (C-S-H) phases resulting from the pozzolanic reactions of BS. The findings showed that, at the 7-day test, replacing cement with 10% and 20% basalt rubble (BS) by weight of the binder yielded compressive strengths of 97% and 92% compared to the control (CT) mortar. In contrast, replacements of 30% and 40% BS resulted in compressive strengths of 72% and 60% of the CT mortar, respectively. Results from 28-day tests showed that replacing 10% of OPC with BS not only increased the compressive strength but also significantly decreased chloride penetration compared to the control mortar (CT). This enhancement suggests that BS can effectively replace 10%-20% of cement, with the compressive strength of the mortar ranging from 92% to 107% of that of the control. The findings accentuate the potential of using industrial by-products such as ground basalt rubble to reduce waste, alleviate environmental impacts, and promote the development of sustainable construction materials.

**Keywords:** Basalt Rubble; Construction Materials; Chloride Penetration; Strength.

## 1. Introduction

Researchers are currently exploring industrial and agricultural by-products as alternative binders to replace cement in mortar and concrete production while maintaining material quality. The use of these waste materials requires thorough investigation into the behavior of mortar and concrete to develop methods that ensure long-term performance, high efficiency, durability, and minimal maintenance. Chloride penetration in concrete significantly affects the durability and longevity of concrete structures by enabling the intrusion of chloride ions into concrete pores when water is present. Such penetration is influenced by several factors, including the water-to-cement ratio, compressive strength, type of cement or binding material, and curing conditions, all of which are critical in resisting the corrosive effects that compromise the structural integrity of reinforced concrete [1-3]. The corrosion process comprises two phases: the

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initiation period, where chloride ions reach a critical concentration and disrupt the protective oxide film on reinforcing steel, and the subsequent corrosion period, which directly diminishes the lifespan of the structure [4, 5]. To enhance concrete's resistance to chloride-induced corrosion, strategies such as optimizing water permeability, adjusting the water-to-cement ratio, increasing the concrete cover thickness over reinforcing steel, and incorporating filler materials to densify the concrete are essential [6].

Pozzolanic materials are characterized by their chemical composition, primarily containing silica or a combination of silica and alumina, which have few to no binding properties on their own. However, when finely ground and moistened, these materials react with calcium hydroxide at normal temperatures to form compounds with binding properties. In Thailand, several pozzolan materials such as fly ash, rice husk ash, palm oil fuel ash, bottom ash, sugarcane bagasse ash, and silica fume are utilized to enhance concrete's mechanical properties and durability. These materials contain vital compounds like silica and alumina, which contribute to the pozzolanic reaction, a chemical process where silica dioxide ( $\text{SiO}_2$ ) reacts to form calcium silicate hydrate (C-S-H). The formation of dispersed C-S-H in concrete not only leads to denser concrete but also results in structures that are stronger and more resistant to environmental degradation [7-9]. The use of these pozzolanic materials is particularly beneficial in resisting chloride penetration and reducing the rate of steel corrosion within concrete structures [4, 5].

Recent research has highlighted basalt, a by-product of the stone-crushing industry in northeastern Thailand, as a promising alternative pozzolan material. Both domestically and internationally, researchers have conducted studies on using stone powder to enhance the efficiency of cement-based materials. Predominantly composed of oxides such as  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$ , and  $\text{Fe}_2\text{O}_3$ , basalt offers potential advantages when used in concrete. Basalt rock fragments are naturally occurring materials and, as such, do not contribute to carbon dioxide emissions. A study by El-Didamony et al. [6] demonstrated that replacing 20% of Portland cement with basalt powder could enhance the compressive strength of concrete, despite an initial decrease compared to traditional cement formulations. The strength tends to increase with the progression of curing. Saudi et al. [10] explored the use of cement blended with basalt rocks and assessed the impact of electromagnetic radiation on the cement's performance. Their findings demonstrated that incorporating basalt rocks into cement effectively reduces radiation exposure and absorbs internal heat. Rashad et al. [11] examined the effects of mixing natural basalt powder (BP) with fly ash (FA) at varying ratios, ranging from 2.5% to 40%, to produce geopolymer cement. The results revealed that BP incorporation between 2.5% and 20% significantly improved the flowability, compressive strength, and hydration reaction of FA-based geopolymer cement, with 10% BP identified as the optimal ratio. Moreover, air curing proved to be more effective than water curing for this composite. Abo Hashem et al. [12] observed that partially replacing cement with basalt powder enhances the compressive strength of mortar and increases resistance to chloride corrosion, a result attributed to the presence of  $\text{Al}_2\text{O}_3$ , which reduces free aggressive chloride ions in the solution. Relatedly, Çelikten and Baran [13] investigated Waste Basalt Powder (BT) as a partial substitute for Portland Cement (PC) in high-strength mortar production. Their study revealed that a 15% substitution of PC with BT resulted in mortars that were stronger and more resistant to high temperatures, indicating the potential for effective recycling of BT in high-performance mortar applications.

Several research studies have investigated the use of basalt powder, fly ash, and slag in the production of mortar. The experimental results revealed that the amounts of basalt powder and fly ash significantly influence both early and long-term strength. Nonetheless, slag primarily affects the early strength of the mortar. Basalt powder, fly ash, and slag at proportions of 10%, 5%, and 25.11%, respectively, achieved the optimal compressive strength and met the specified requirements [14]. Previous studies have conducted limited research on the use of basalt as a binder to replace cement, specifically examining the durability of mortar in terms of chloride resistance and corrosion. Therefore, this research aims to further develop ground basalt rubble as a sustainable replacement for Portland cement in Thailand, investigating its effects on compressive strength, the chloride ion diffusion coefficient of mortar, and the microstructure of cement pastes, thereby contributing to the innovation of new construction materials from waste products.

## 2. Materials and methods

### 2.1. Materials

In this study, binder materials consisting of Portland cement type 1 (OPC) and ground basalt rubble (BS) were used to investigate the potential of BS as a supplementary cementitious material. Portland cement, with a specific gravity of 3.14, was finely ground, and basalt rubble was processed to retain 10% by weight on sieve No. 325, a measure to assess the fineness of the material. Sand with a specific gravity of 2.58, typically used in general construction works, served as the fine aggregate. This study utilized a constant water-to-binder ratio of 0.45. To enhance the workability and flow properties of the mortar, a superplasticizer (SP) was employed, adjusting the mix to achieve the desired consistency.

The physical properties of the basalt rubble were further characterized by grinding the material using a pozzolan grinding machine, ensuring that 10% remained on standard sieve No. 325. The specific surface area of the basalt was measured using the Blaine fineness method [15], detailed in Table 1, which presents the physical properties of the BS. Moreover, Figure 1 displays a scanning electron microscopy (SEM) photograph of the very fine ground basalt rubble, illustrating that the BS primarily comprises fine, irregular-shaped particles. These characteristics are significant for understanding how BS integrates into the cement matrix and influences the overall properties of the mortar.

**Table 1. Physical properties of OPC and BS**

Physical properties	OPC	BS
Retained on a sieve No. 325 (%) or 45 $\mu\text{m}$	N/A	10
Specific Gravity (S.G.)	3.14	2.92
Blaine Fineness ( $\text{cm}^2/\text{g}$ )	3,400	7,500

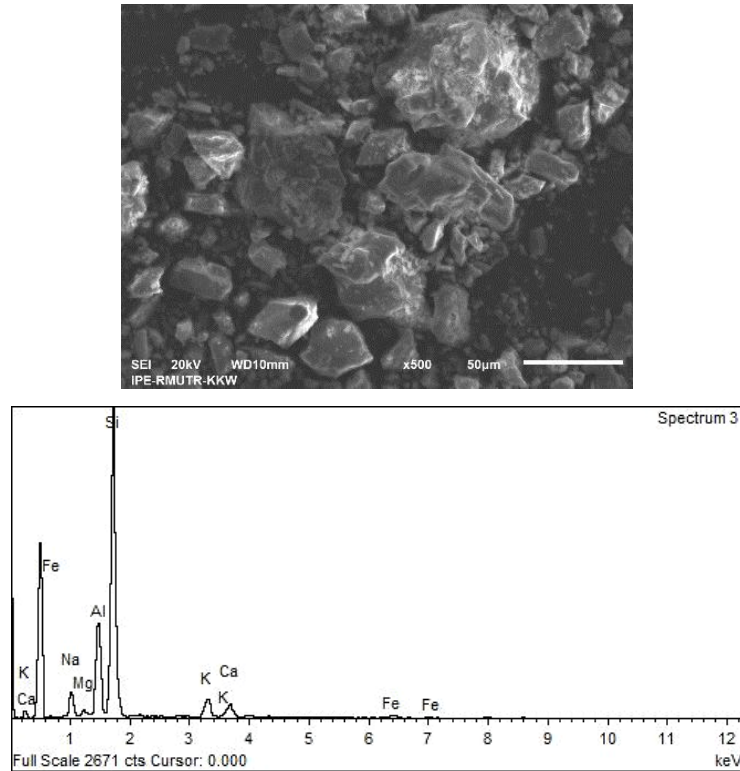
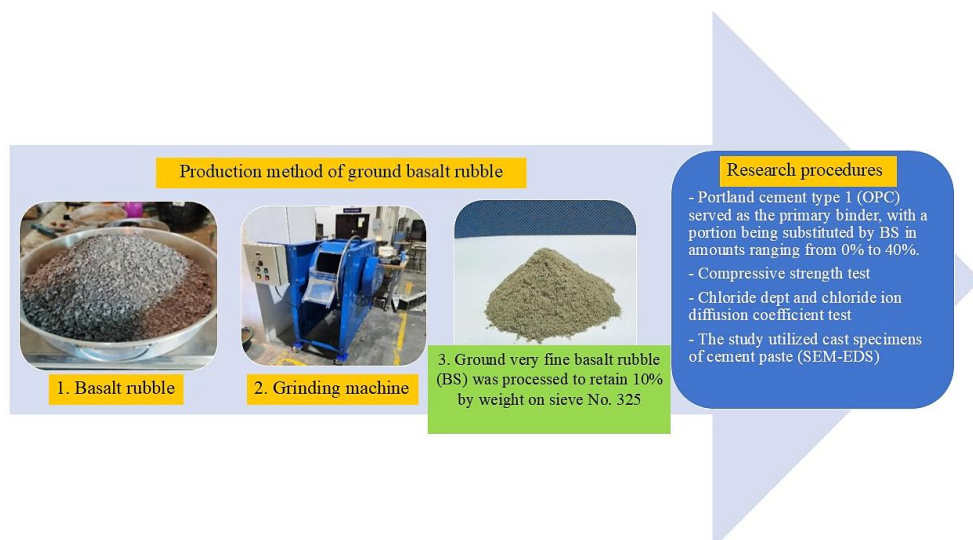
**Figure 1. SEM-EDS analysis of BS**

Table 2 presents the chemical compositions of Portland cement type 1 (OPC) and ground basalt rubble (BS), where OPC is primarily composed of calcium oxide ( $\text{CaO}$ ) at a concentration of 65%. In contrast, BS is characterized by a high silica ( $\text{SiO}_2$ ) content, also at 54%, with its total composition of  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$  and  $\text{Fe}_2\text{O}_3$  reaching 77%. This composition aligns BS with the characteristics of a typical pozzolan material as defined by the ASTM C618 standards [16]. The chemical analysis of BS conducted using SEM-EDS, shown in Figure 1, confirms that silica remains the predominant component. These findings suggest that BS, derived from industrial basalt products, meets the criteria for use as a pozzolanic material in cement formulations, offering potential enhancements in cement's structural and durability properties. Figure 2 illustrates the research procedures and the production method of basalt rubble, respectively.

**Figure 2. Research procedures and production method of ground basalt rubble**

**Table 2. Chemical compositions of OPC and BS**

Oxides (%)	SiO <sub>2</sub> (%)	Al <sub>2</sub> O <sub>3</sub> (%)	Fe <sub>2</sub> O <sub>3</sub> (%)	CaO (%)	SiO <sub>2</sub> + Al <sub>2</sub> O <sub>3</sub> + Fe <sub>2</sub> O <sub>3</sub> (%)
OPC	20	5	3	65	-
BS	54	6	17	1.9	77

## 2.2. Mix Proportions

The mortar mixture was designed with a fixed water-to-binder ratio of 0.45 and a composition of binder to fine aggregate at a ratio of 1:2.75. A superplasticizer (SP) was employed to optimize the flow and workability of the mortar in accordance with ASTM C230 standards [17]. Portland cement type 1 (OPC) served as the primary binder, with a portion being substituted by ground, very fine basalt rubble (BS) in amounts ranging from 0% to 40%. After the mortar was cast in molds, it was stored at a controlled temperature of  $25 \pm 3^\circ\text{C}$  for 24 hours in a laboratory setting. Subsequently, all samples were cured in clean water at a stable temperature of  $23 \pm 2^\circ\text{C}$  until they reached the designated ages for testing. The evaluation process included compressive strength tests and chloride ion diffusion coefficient assessments. The specific proportions of the mortar components are presented in Table 3.

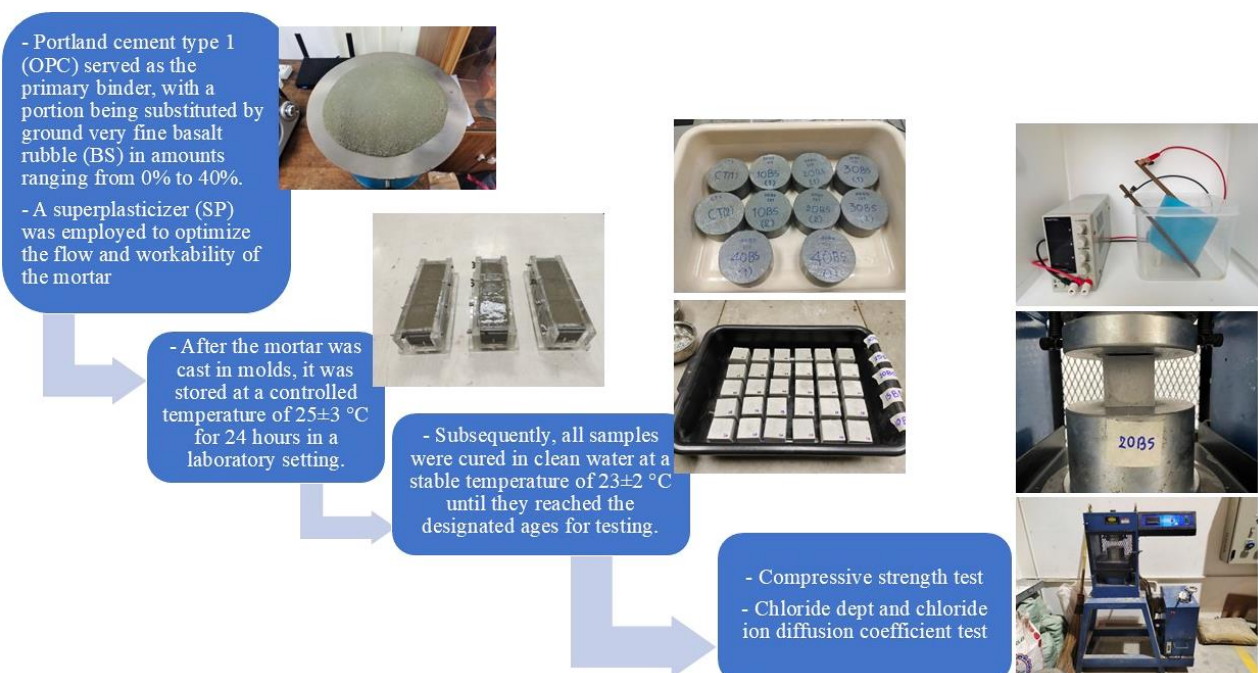
**Table 3. Mix proportions of mortars**

Mix	Mix Proportion (% by Weight)					
	W/B	Cement (OPC)	BS	Sand	SP	Flow (%)
CT	0.45	100	-	275	0.37	115
10BS	0.45	90	10	275	0.36	105
20BS	0.45	80	20	275	0.32	115
30BS	0.45	70	30	275	0.31	108
40BS	0.45	60	40	275	0.30	109

**Note:** W/B = Water cement to binder ratio (Cement = Portland cement type 1 (OPC)), CT = Control mortar, SP = Superplasticizer (by percentage of binder), BS = Ground basalt rubble.

## 2.3. Compressive Strength Test

The compressive strength of the mortar was evaluated using molds measuring  $50 \times 50 \times 50 \text{ mm}^3$ , in accordance with the ASTM C109 standard [18]. Testing was conducted at two critical curing ages, specifically at 7 and 28 days, to assess the development of the material's strength over time. This method provides a standardized approach to measure the ability of the mortar to withstand loads, which is essential for determining its suitability for structural applications. Figure 3 indicates an example of mortar production and testing instruments.

**Figure 3. Mortar production and testing instruments**



## 2.4. Chloride Dept and Chloride Ion Diffusion Coefficient Test

The depth of chloride penetration was assessed using the rapid migration test chloride depth (RMTCD). Mortar samples, initially prepared with a diameter of 100 millimeters and a height of 200 millimeters, were cut to a reduced height of 50 millimeters after curing for 7 and 28 days. These samples underwent an accelerated chloride penetration test in a 3% sodium chloride solution. The test setup involved submerging both the cathode and the anode in limewater, as illustrated in Figure 4, and applying a 30-volt DC electrical voltage across them for 30 days. Post-testing, the samples were bisected and treated with 0.1 M  $\text{AgNO}_3$ , following the methodology established by Otsuki et al. [19]. The chloride depth, indicative of the chloride distribution coefficient, was subsequently calculated using the Nernst-Plank equation [20, 21], providing a quantitative measure of the mortar's resistance to chloride ingress. The chloride depth of RMTCD can be used to find the chloride distribution coefficient using the calculation principle of the Nernst-Plank equation [20, 21], shown in Equation 1.

$$D = \frac{RTL}{zFE} \times \frac{x_f}{t} \quad (1)$$

When  $D$  is Chloride ion diffusion coefficient ( $\text{cm}^2/\text{s}$ ),  $R$  is gas content ( $\text{J/K mol}$ , 8.314),  $T$  is temperature ( $\text{K}$ ),  $L$  is mortar thick ( $\text{cm}$ ),  $z$  is ion valence,  $F$  is Faraday content ( $\text{C/mol}$ ,  $9.65 \times 10^4$ ),  $E$  is values of potential ( $\text{V}$ ),  $x_f$  is inflection point of chloride ion profiles that needs to be related to the depth given by the colorimetric technique and  $t$  is time of test ( $\text{s}$ ).

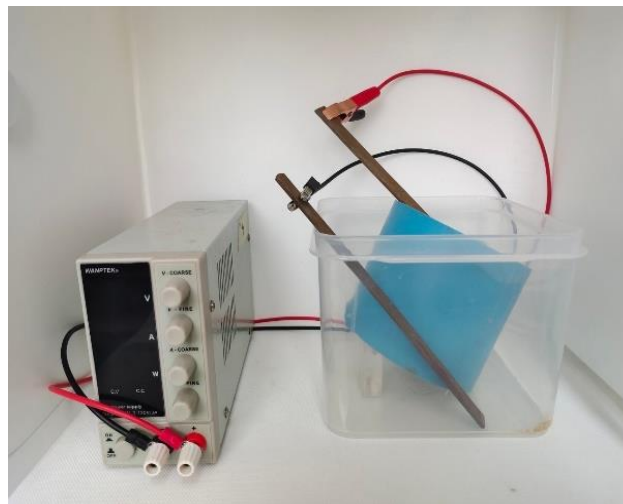


Figure 4. Rapid migration test chloride dept (RMTCD)

## 2.5. SEM-EDS Analysis

The study utilized cast specimens of cement paste, each measuring  $50 \times 50 \times 50$  millimeters, where ground basalt rubble replaced cement at rates of 10% by weight of the binder, with a water-to-binder ratio maintained at 0.35. After a 24-hour casting period, the specimens were demolded and cured in clean water for 28 days to ensure proper hardening. To arrest further chemical reactions, the specimens were subsequently immersed in acetone within sealed glass jars for a duration of seven days. After the acetone treatment, the specimens were crushed into small fragments for detailed analysis using Scanning Electron Microscopy with Energy Dispersive Spectroscopy (SEM-EDS). This comprehensive analysis aimed to assess the microstructural effects of basalt substitution in the cement matrix.

## 3. Results and Discussion

### 3.1. Results of Compressive Strength and Strength Activity Index of Mortars

Figures 5 and 6 present the results of the compressive strength tests and the strength activity index for mortars that incorporated ground basalt rubble (BS) in proportions ranging from 0% to 40% by weight of the binder. At the 7-day testing mark, mortars with cement replaced by ground basalt rubble at 0%, 10%, 20%, 30%, and 40% by weight of the binder exhibited compressive strengths of 43.5 MPa, 42 MPa, 40 MPa, 31.5 MPa, and 26 MPa, corresponding to 100%, 97%, 92%, 72%, and 60% of the control (CT) mortar, respectively. As testing progressed to 28 days, the compressive strengths of the same mortar samples increased, measuring 53 MPa, 56.5 MPa, 49.5 MPa, 38.5 MPa, and 33.5 MPa, which represented 100%, 107%, 93%, 73%, and 63% of the CT mortar. The results indicate that while higher basalt replacement ratios reduce early strength, a replacement of 10% basalt rubble can enhance long-term compressive strength beyond that of the control.

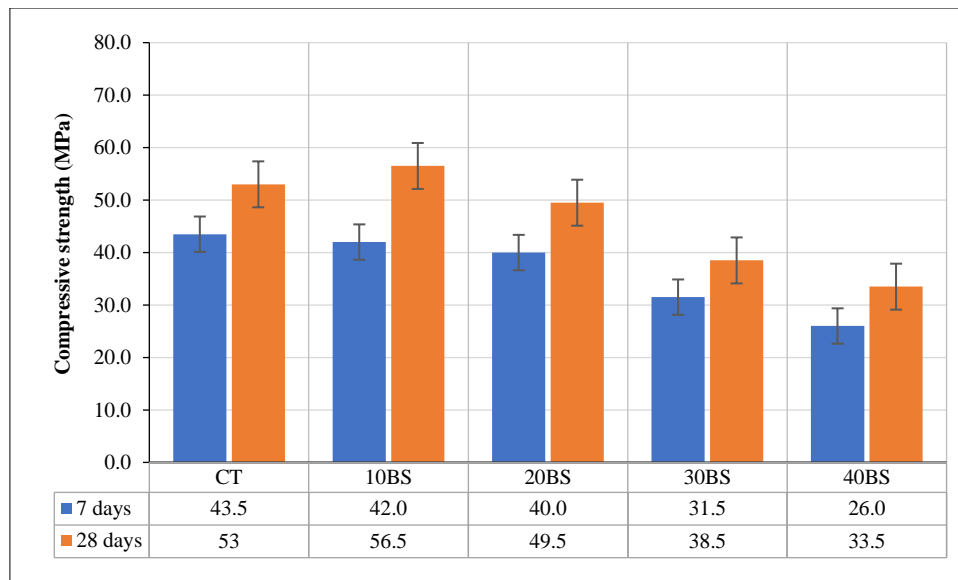


Figure 5. Compressive strength of mortar

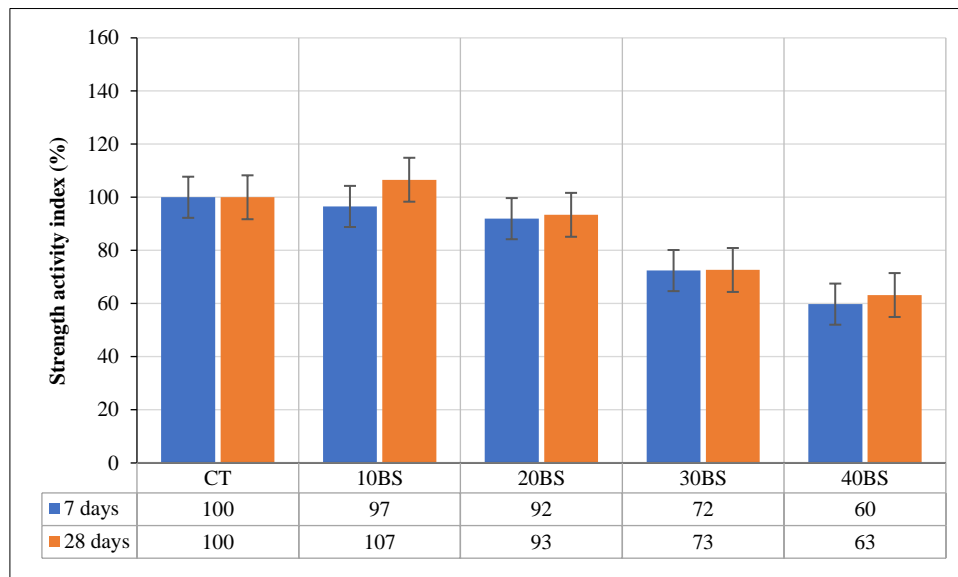


Figure 6. Strength activity index of mortar

Initial findings at a 7-day curing age revealed a reduction in compressive strength for mortar mixtures incorporating basalt rubble (BS) compared to those made with traditional cement, aligning with observations from previous studies [6]. Specifically, when BS replaced cement at 10% and 20%, the compressive strengths recorded were 97% and 92% respectively, relative to the control mortars made with traditional cement (CT), as specified by ASTM C618 [16]. Despite this initial reduction, the strength values suggest that BS can effectively function as a pozzolanic material, maintaining more than 75% of the compressive strength of the CT mortars. Yet, a further increase in BS content to 30% and 40% resulted in a decrease in strength to 72% and 60% of the CT values, respectively.

Over a longer curing period of 28 days, the mortars with 10% BS demonstrated superior compressive strengths compared to the CT mortars. The enhancement in strength over time can be attributed to the finer particle size of BS due to grinding, which promotes more effective pozzolanic reactions as the material ages [6]. The compressive strengths of mortars containing 10% and 20% BS were 107% and 93%, respectively, of those of the CT mortars. These findings emphasize that while BS can be beneficial up to a certain threshold, increasing the proportion of BS beyond 30% to 40% leads to a reduction in compressive strength, suggesting an optimal upper limit for BS incorporation in mortar mixtures. The study revealed that incorporating basalt stone (BS) as a partial cement replacement resulted in a slow pozzolanic reaction during the early stages, followed by a gradual increase in compressive strength as curing time progressed, consistent with earlier research [6]. Yet, higher proportions of ground basalt rubble led to a reduction in compressive strength due to the decreased release of  $\text{Ca}(\text{OH})_2$ , which is essential for the pozzolanic reaction. This indicates that increasing the amount of ground basalt rubble reduces the overall cement content in the mixture, thereby weakening the structure. Previous studies [13, 22, 23] observed a decline in strength when using pozzolan beyond optimal levels, which aligns with these findings.

The successful use of ground basalt rubble as a partial replacement for cement is critically dependent on the fineness of the material, which directly influences the pozzolanic reactions essential for strength development. Fine grinding of BS not only enhances these reactions but also allows the smaller particles to occupy voids within the mortar matrix [7, 9], contributing to a denser and structurally more cohesive material [6, 24, 25]. The silica in the basalt reacts with calcium hydroxide to form calcium silicate hydrate, thereby increasing the density of the mortar [26]. Nonetheless, excessive use of finely ground basalt rubble, particularly beyond 20% substitution by weight, tends to diminish compressive strength due to increased particle interlock and void formation, leading to less dense mortar with higher porosity. Therefore, the optimal substitution rate of finely ground basalt powder is recommended to be between 10% to 20% by weight of the binder [6, 13, 26, 27, 28].

### 3.2. Results of Chloride Depth and Coefficient of Chloride Diffusion

The chloride penetration depth in this study was evaluated using the rapid migration test for chloride depth (RMTCD) with a 30-volt DC setup, as depicted in Figure 7. Results demonstrated that a 10% substitution of Portland cement (OPC) with basalt rubble (BS) led to a lower chloride depth compared to pure cement and other mixed formulations. This reduction is attributed to the pozzolanic reactions facilitated by the basalt rubble, which enhance compaction and decrease chloride absorption [28, 29]. The silica released during these reactions forms salt crystals that effectively fill voids in the mortar, thereby reducing pathways for chloride penetration [21, 29].

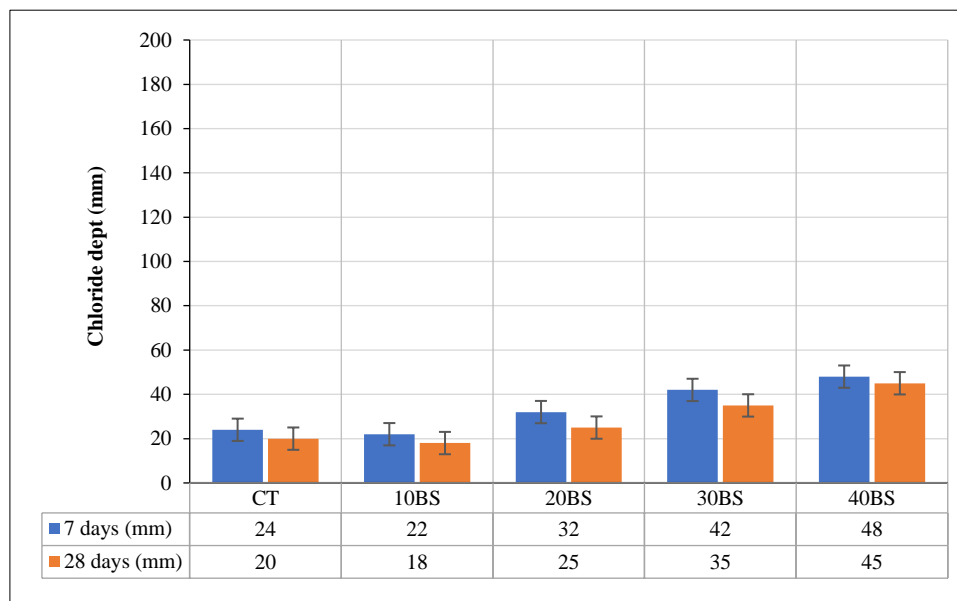


Figure 7. Chloride depth of mortar

Nevertheless, increasing the basalt substitution from 10% to 20%, 30%, and 40% correspondingly increased the depth of chloride penetration. This is likely due to the shape and high quantity of the ground basalt particles causing self-interference, which creates additional voids allowing for easier chloride ingress [6, 7, 21]. Despite this, the mortar with a 10% basalt content not only exhibited superior chloride resistance but also maintained a high compressive strength, achieving a strength activity index of 107% compared to the control mortar at 28 days. The chemical composition of BS, rich in  $\text{SiO}_2$ , reacts with  $\text{Ca}(\text{OH})_2$  to form calcium silicate hydrate (C-S-H), which improves the density of the mortar and its capacity to absorb chloride ions, as shown in the chloride depth results depicted in Figure 7 [24, 25].

Further testing revealed the coefficients of chloride diffusion, detailed in Figure 8. The control mortar (CT) had a diffusion coefficient of  $3.80 \times 10^{-6} \text{ cm}^2/\text{sec}$ . In contrast, the mixtures with 20%, 30%, and 40% BS substitution exhibited higher diffusion coefficients, specifically  $4.50 \times 10^{-6}$ ,  $5.20 \times 10^{-6}$  and  $6.10 \times 10^{-6} \text{ cm}^2/\text{sec}$ , respectively. Interestingly, the mortar with 10% BS substitution showed a reduced diffusion coefficient of  $3.20 \times 10^{-6} \text{ cm}^2/\text{sec}$ . This reduction aligns with the observed chloride penetration results, underscoring the effectiveness of a 10% BS content in enhancing chloride resistance. This enhancement is further supported by the pozzolanic reaction between  $\text{SiO}_2$  from BS and  $\text{Ca}(\text{OH})_2$ , forming C-S-H, which not only increases the density but also the fineness of the BS contributes to greater chloride resistance due to more extensive pozzolanic activity [6, 24, 25, 30, 31]. These findings confirm that finely ground basalt rubble is optimal at a 10% substitution rate for improving resistance to chloride penetration in mortar mixtures [24, 29]. This study focused on testing at 28 days of curing, and plans will be made to test chloride resistance over a longer period to gather data for the use of crushed basalt rock as a binder material in mortar or concrete in the future.

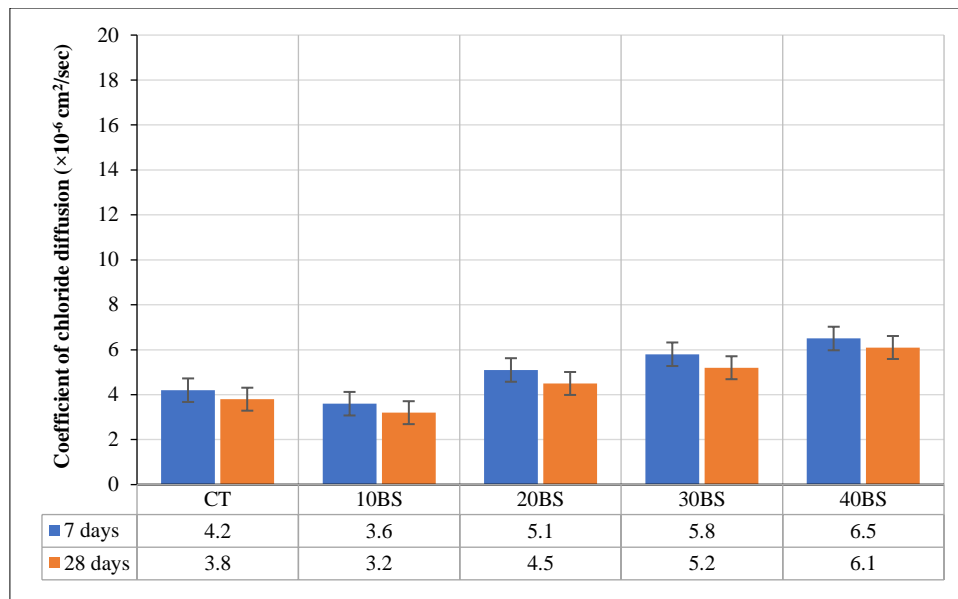
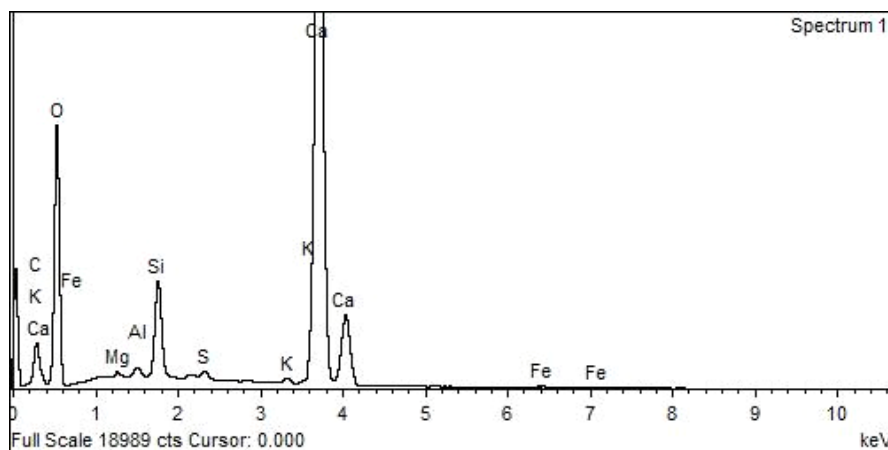
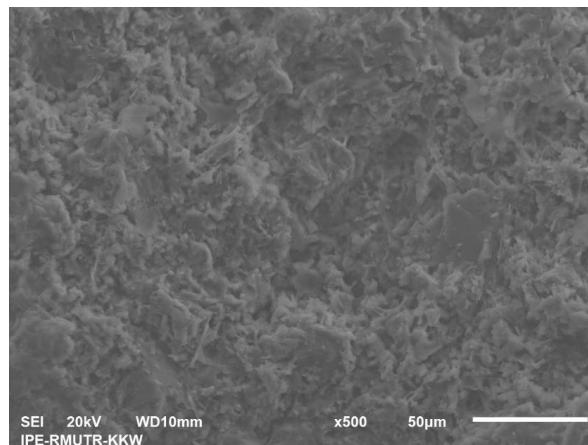


Figure 8. Coefficient of chloride diffusion of mortar

### 3.3. Results of SEM-EDS Analysis of Paste

Figures 9 a) and b) present the SEM-EDS analysis results of two types of cement paste: one composed solely of cement (CT) and the other with 10% ground basalt rubble (10BS) substituting cement by binder weight. This substitution choice was prompted by the enhanced compressive strength observed in the 10BS mix compared to the traditional cement mix. The analysis was conducted to explore how this substitution affects the microstructure, which is closely tied to the observed increases in compressive strength and resistance to chloride penetration. For the CT paste, the analysis highlighted an even roughness distribution, indicating a homogeneous hydration reaction across the sample.



(a)



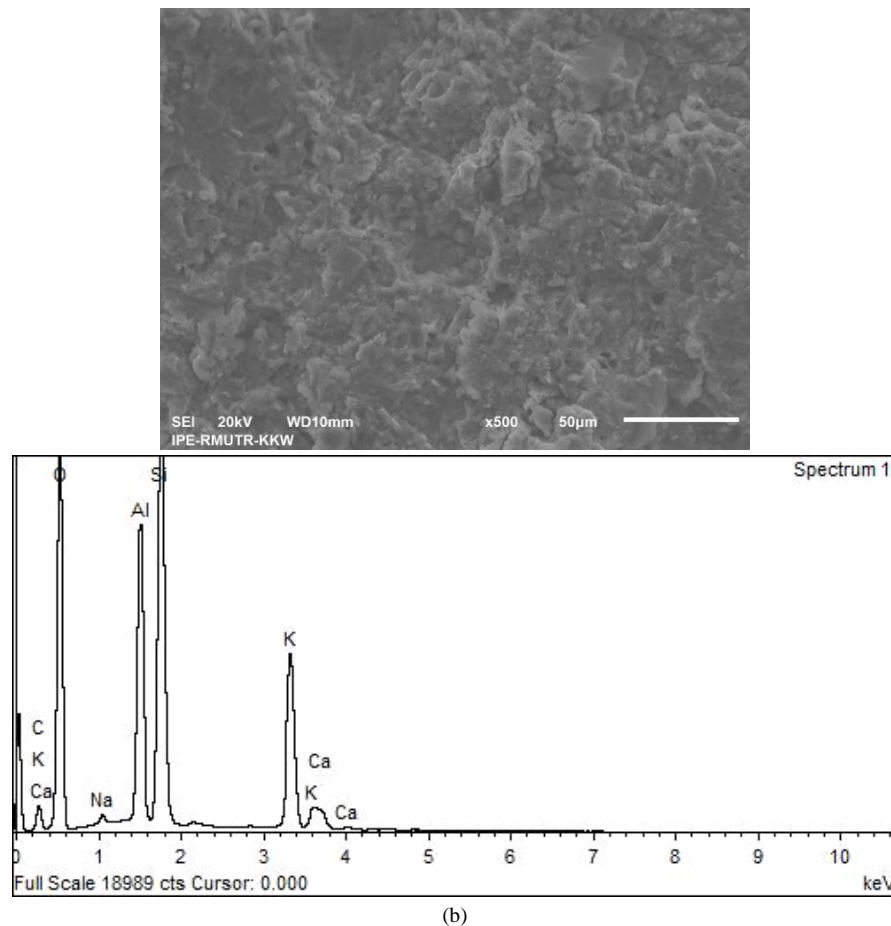


Figure 9. SEM-EDS Analysis of a) CT and b) 10BS pastes

The SEM-EDS results for the 10BS paste revealed the presence of calcium silicate hydrate (C-S-H) as a product of the pozzolanic reaction, which was not as evident in the CT paste. The substitution with very finely ground basalt rubble increased the silica ( $\text{SiO}_2$ ) content in the paste, corroborating previous findings that such substitution not only boosts compressive strength but also diminishes chloride penetration in comparison to control samples [6, 24, 25]. This effect is attributed to the smaller particle size of the BS, which fills voids within the mortar, thus increasing its density and enhancing water resistance. Furthermore, the significant surface area provided by the fine BS particles intensifies the pozzolanic reaction [28]. Similar to the findings of Serhat Çelikten & Bilal Baran [13], the primary factors influencing the effect of basalt powder on the strength and density of the mix are the fineness of the material, which enables it to fill voids effectively, and its pozzolanic reaction with  $\text{Ca}(\text{OH})_2$  [13]. Such a reaction, which occurs as a byproduct of cement hydration, contributes to a denser, more homogeneous mixture, enhancing both the structural integrity and durability of the mortar. The silica and alumina in the basalt react with calcium hydroxide to form C-S-H [7, 9, 30], thereby increasing the density of the paste and reducing its porosity, which in turn improves compressive strength and reduces chloride penetration [16, 26-28].

#### 4. Conclusion

The findings provide valuable insights that can enhance confidence in the construction industry regarding the use of basalt rubble in mortar or concrete. Basalt rubble, containing silica or a combination of silica and alumina, can serve as a pozzolanic material due to its ability to react alongside the hydration process, thereby increasing the strength of mortar or concrete. Silica and alumina are key components in the pozzolanic reaction, contributing significantly to the improved mechanical properties of the mix. Pozzolanic materials are also cost-effective, offering advantages such as enhanced compressive strength and durability in concrete production. The present study utilized replacement ratios of 0% to 40% to observe and compare the effects of substituting cement with ground basalt rubble at 10% increments by weight of the binder. Future research could investigate higher replacement levels beyond 40% to explore the impact of high-volume basalt rubble as a cement replacement.

This study conclusively demonstrates that ground basalt rubble can effectively replace 10% of cement in mortar mixtures, resulting in higher compressive strength and reduced chloride penetration compared to mortar made exclusively with cement. The findings also indicate that while substituting 20% to 40% of cement with basalt rubble leads to a reduction in compressive strength, the resulting strength values, ranging from 33.5 to 56.5 MPa, remain

sufficient for a wide range of construction applications. By introducing ground basalt rubble as a viable binder material, this study not only provides an innovative use for waste materials but also promotes efficient resource utilization. It exemplifies a balanced approach to addressing social and environmental challenges by reducing energy waste, mitigating the effects of global warming, and fostering sustainability from the present into the future.

## 5. Declarations

### 5.1. Author Contributions

Conceptualization, S.R.Z. and S.R.R.; methodology, R.T. and U.C.; software, S.R.Z.; validation, S.R.Z., S.R.R., and P.C.; formal analysis, S.R.Z.; investigation, R.T. and U.D.; resources, S.R.Z.; data curation, S.R.Z.; writing—original draft preparation, S.R.Z.; writing—review and editing, S.R.Z. and P.C.; visualization, S.R.Z.; supervision, S.R.Z.; project administration, S.R.Z.; funding acquisition, S.R.Z. All authors have read and agreed to the published version of the manuscript.

### 5.2. Data Availability Statement

The data presented in this study are available in the article.

### 5.3. Funding

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### 5.5. Conflicts of Interest

The authors declare no conflict of interest.

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