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ORIGINAL ARTICLE

The Impact of Waste Marble Powder as A Partial Alternative Material for Cement

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ABSTRACT – Waste marble powder generated from the sawing and the mining activity of marble stone caused environmental problems that harmed human health. This current research aimed to investigate the influence of waste marble powder to substitute cement partially in mortar production. The mortar preparation with the mixture compositions of 0–50% marble powder was investigated for their resistance to compression and porosity. The characterization was assessed with X-ray fluorescence (XRF), scanning electron microscopy-energy dispersive X-ray (SEM-EDX), Fourier-transform infrared (FTIR), and X-ray diffraction (XRD). The finding of this research revealed that a replacement of up to 10% marble powder obtained an increase in compressive strength after 28 days. A low level of marble powder to replace cement led to a less porous microstructure of the mortars. These confirmed that waste marble powder could be applied to manufacture mortars. In addition, the utilization of waste marble powder as an alternative building material would reduce the damage to the environment.

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INTRODUCTION

South Central Timor Regency has various potential natural resources, including minerals. Minerals are widely used in construction as building materials, one of which is marble. Marble is a coarse crystalline rock derived from limestone undergoing a metamorphic process. This process is caused by increased temperature and pressure in the earth's core. Marble stone is chemically comprised of calcium carbonate or calcite ($CaCO_3$), dolomite, and serpentine minerals [1]–[2].

Naitapan Stone Mountain, located in Tunua Village, Fatumnasi Subdistrict, South Central Timor District, is one of the sources of marble stone production in East Nusa Tenggara Province. Marble stone mining in Mount Batu Naitapan produces powder waste from the residue of sawing marble stones, which is still abandoned around the mining area and has not been utilized optimally and appropriately. This phenomenon has led to significant environmental problems due to the enormous waste. The waste reduces the soil's porosity and lowers its fertility by increasing the alkalinity. If the waste marble powder gets suspended in the air, it will cause hazards to human health [3]. Thus, it is necessary to have a solution to overcome that problem.

Previous studies had utilized waste marble powder, so it was more beneficial and was not harmful to the environment, particularly its benefit on mortar production. Marble powder is an inert material [4] and has been globally beneficial for use as cement replacement in the production of mortars [3], [5]–[6]. Alyamaç et al. studied the influence of marble powder replacement at 10%, 20%, 30%, 40%, 50%, and 90% by volume. They concluded that up to 20% marble powder was suitable to improve the compressive strength of the mortars [5]. Kabeer et al. proved that replacing cement with up to 20% marble powder increased the compressive strength of the mortars after 28 days of curing [6]. A study by Vardhan et al. reported that marble powder could be used as a partial substitution for cement at a percentage of up to 10% [3]. Perkasa et al. revealed an increase in the compressive strength value of paving blocks with the usage of waste marble powder at 10%. The maximum compressive strength value was obtained using a percentage of 10% marble powder, with a compressive value of 29.97 MPa [7].

This present study focused on the utilization of waste marble powder to overcome environmental problems in Tunua Village, Indonesia, by carrying out a partial replacement for cement with waste marble powder, with the composition of marble powder varied from 0%, 10%, 20%, 30%, 40%, and 50%. The properties of the mortar mixes were studied, i.e., compressive strength and porosity at the curing age of 7, 14, 21, and 28 days. X-ray fluorescence (XRF), scanning electron microscopy-energy dispersive X-ray (SEM-EDX), Fourier-transform infrared (FTIR), and X-ray diffraction (XRD) analyses were performed to examine the microstructures and their characteristics.

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EXPERIMENTAL METHOD

Materials and Instruments

Marble powder waste was obtained from Tunua Village, North Mollo District, South Timor Tengah Regency. Portland cement (Tiga Roda) was purchased from a local building material store in Kupang.

The samples' morphology and element content were determined by PhenomProX Desktop scanning electron microscopy-energy dispersive X-ray (SEM-EDX). X-ray diffraction (XRD) PANalytical AERIS was used to record the diffraction pattern of the samples. The composition of marble powder was analyzed using X-ray fluorescence (XRF) S2 PUMA Bruker.

Method and Procedure

Preparation of mortars

The procedure for preparing mortars was determined by following ASTM C-305-06 [8]. The mortars were prepared by mixing Portland cement, sand, and water in a proportion of 1:1:0.6, respectively, using a mechanical mixer at 285±5 rpm for 90 seconds. The sand was replaced by marble powder from 0% to 50% in the steps of 10%. The mortar mix was poured into a cube container (5×5×5 cm). The mortar without marble powder (0%) was utilized as a control.

Compressive strength test

Three cubical samples (5×5×5 cm) of each mixture were assessed for their compressive strength. The specimens were cured for 7, 14, 21, and 28 days, then tested using a hydraulic press, following the procedure ASTM C 109/C 109M [9].

Porosity

The examination of porosity was carried out following the standard as described in ASTM C642-06 [10]. The mass of dry specimens was determined and noted as A. Next, the specimen was soaked in water for <48 h at 21°C, then the surface of the specimen was dried and weighed (B). Water was poured into specimen B until it got soaked, then it was boiled for 5 h and left until 20–25°C. The weight of the specimen was noted as C. Furthermore, the specimen was hung in water, and the weight was determined (D).

Specimen design and preparation

Response surface methodology (RSM) and Design Expert 13 were used to optimize the proportional mixture of mortar materials to obtain an optimum compressive strength of mortars cured for 7, 14, 21, and 28 days.

RESULT AND DISCUSSION

The Composition of Marble Powder

The X-ray fluorescence (XRF) analysis was conducted to get information on the composition of marble powder. In general, marble powder was composed of Si, Al, Fe, and Ca as the main elements in the form of oxides. Table 1 summarizes the oxide compounds that formulated marble powder using the XRF analysis. The composition of marble powder showed that it contained a high amount of CaO (97%). The concentration of CaO in this study was higher than the result obtained by Susilowati, which was 54.22% [11].

Table 1. The composition of marble powder analyzed by XRF

No.	Compounds	Percentage (%)
1	Silicon dioxide (SiO_2)	0.9
2	Aluminum dioxide (Al_2O_3)	0.1
3	Ferric oxide (Fe_2O_3)	0.2
4	Calcium oxide (CaO)	97.0
5	Magnesium oxide (MgO)	0.5
6	Potassium oxide (K_2O)	0.1

The compressive strength results can be influenced by curing time and high levels of CaO. The increase in the CaO content of marble powder caused an increase in the compressive strength of mortar, consequently [12]. With a relatively high calcium content in marble, marble powder can be beneficial to manufacturing mortars as an alternative material. Hence, the waste marble powder could be processed optimally, increasing the economic value.

The Characteristics of Cement and Marble Powder

The microstructural analysis using scanning electron microscopy-energy dispersive X-ray (SEM-EDX) revealed that cement and marble powder had spherical shapes and similar surface morphology (Figure 1). EDX analysis reported the respective element contents of cement and marble powder (Figure 2).

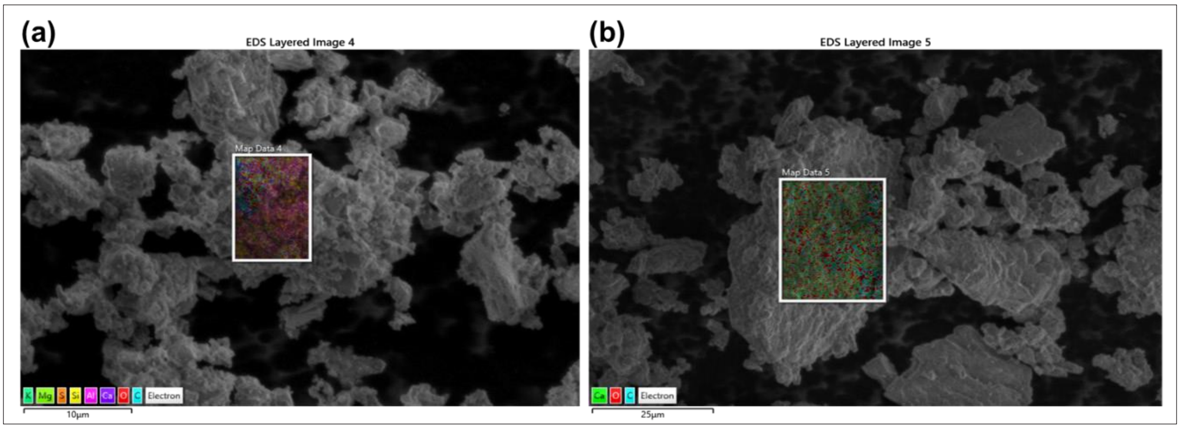


Figure 1. Morphology of (a) cement and (b) marble powder analyzed by SEM-EDX

Figure 2 (a) showed that the cement samples comprised O, Ca, and C with the wt% values of 42.4%, 35.5%, and 14.89%, respectively. Meanwhile, the waste marble powder contained 49.3%wt of O, 36.0%wt of Ca, and 14.7%wt of C (Figure 2 (b)). The similarity in their respective properties allowed waste marble powder to partially replace cement in the mortar preparation [6].

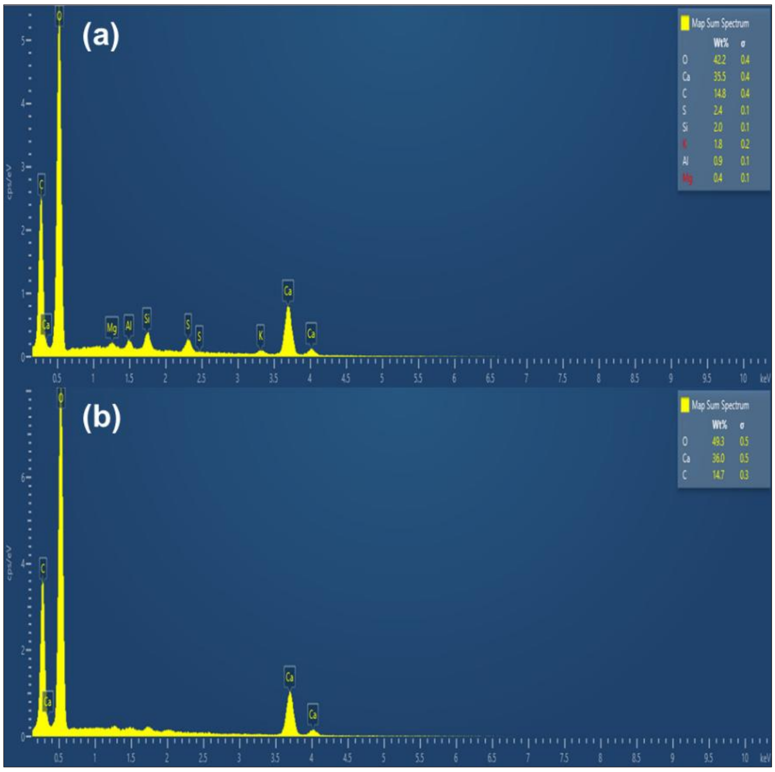


Figure 2. The elemental composition of (a) cement and (b) marble powder

The X-ray diffraction (XRD) analysis investigated the compounds contained in cement and marble powder. XRD showed the peaks attributed to the correlation of diffraction angle (2θ) and intensity (I) on the diffraction pattern. This analysis was performed for 2θ between 10° to 80°. The intensity of every peak reflected the amount of photons detected by the XRD detector. It was observed from Figure 3 (a) that the cement sample had a crystalline solid level of 85.067%

and an amorphous solid level of 1.313%. Similar results were obtained from the marble powder sample, where the crystalline solid content is 88.038%, and the amorphous solid content is 2.193% (Figure 3 (b)).

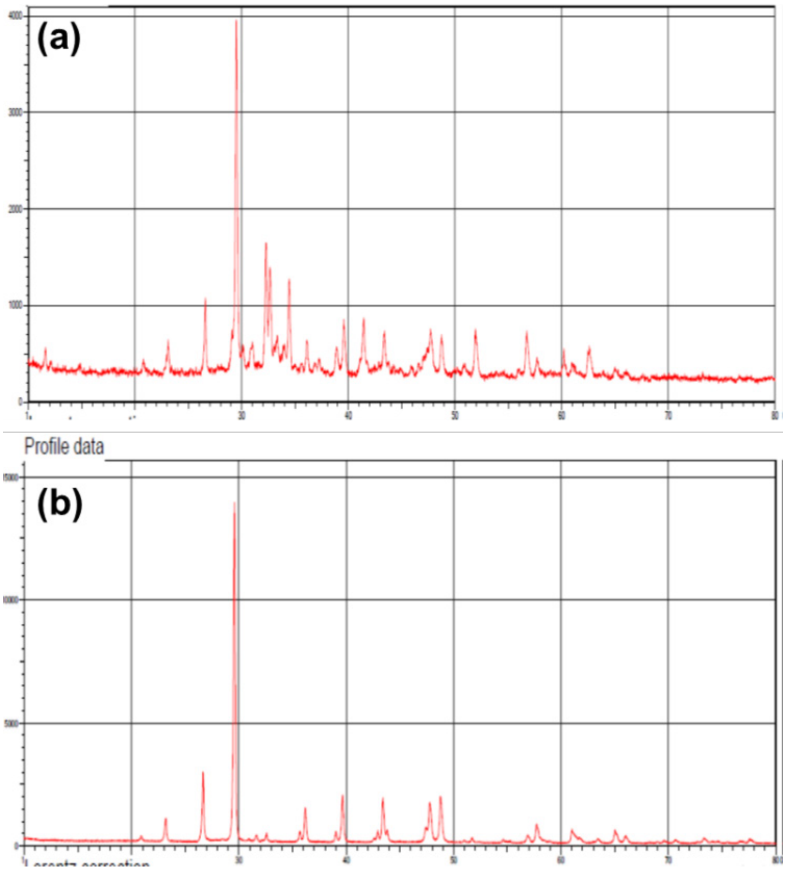
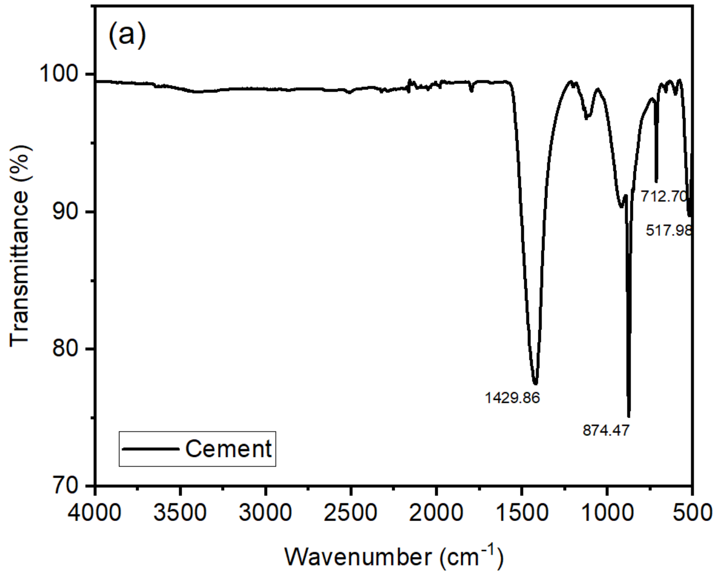


Figure 3. X-ray diffraction pattern of (a) cement and (b) marble powder (b)

The analysis using Fourier-transform infrared (FTIR) examined the molecule bonds in cement and marble powder. Figure 4 and Table 2 summarize the minerals consisting of cement and marble powder. According to a report by Hulungo et al., each peak appearing in the FTIR spectra was owing to a different mineral [13]. A hematite mineral peak appeared in the cement spectra at 517.98 cm^{-1} with an intensity of 77.98%. A peak at 712.70 cm^{-1} (cement) and 712.18 cm^{-1} (marble powder) was attributed to quartz. Aragonite was shown by a peak at 874.47 cm^{-1} and 873.03 cm^{-1} in the cement and marble powder spectra, respectively. The palygorskite peak in the cement and marble powder spectra was at 1429.86 cm^{-1} and 1405.98 cm^{-1}, respectively.

SEM-EDX analysis was performed to examine the particle size distribution, the surface morphology, and the components of the marble-cement composites. Figure 5 revealed the morphology of marble-cement composites, which had dissimilar shapes and size distributions of 18.26 ± 11.39 nm. Figure 6 and Figure 7 below show the element distribution maps of composite incorporating marble powder as a partial substitution of cement. The intensity level of each component contained in the marble-cement composites is shown in Figure 7. The contents of O and Ca elements in the composites were relatively high compared to other elements. Özkılıç et al. reported that the high concentration of Ca created a density in the porosity of the mortar's internal structure; it also affected the mechanical performance of the mortar concrete [14].



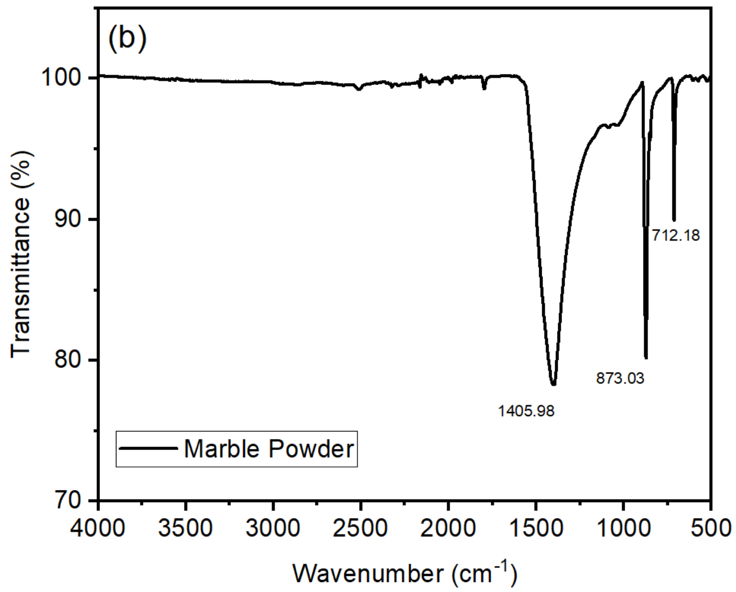


Figure 4. FTIR spectra of (a) cement and (b) marble powder

Table 2. The FTIR analysis shows the minerals consisting of cement and marble powder

Peak Number	Compound/Mineral	Cement		Marble Powder	
		X (cm^{-1})	Y (%T)	X (cm^{-1})	Y (%T)
1	Palygorskite	1429.86	76.47	1405.98	77.70
2	Aragonite	874.47	71.31	873.03	78.87
3	Quartz	712.70	86.66	712.18	88.21
4	Hematite	517.98	77.98		

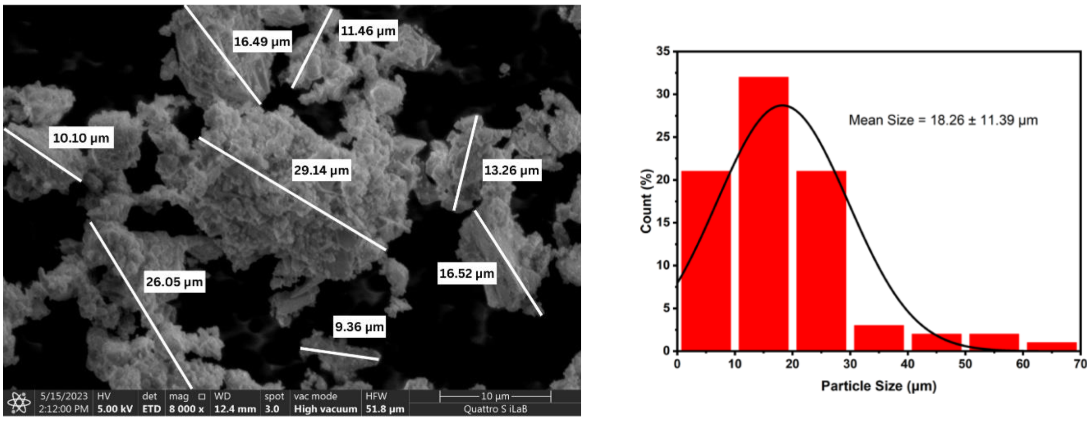
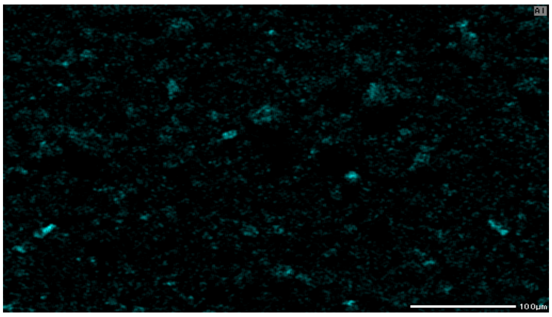
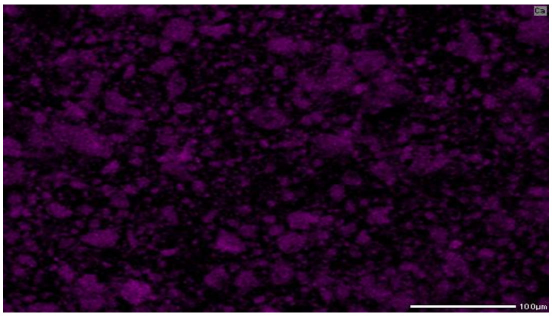


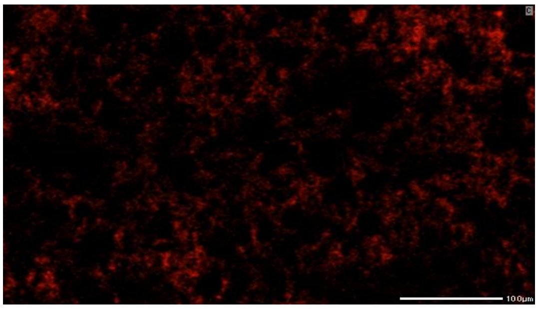
Figure 5. SEM image of marble-cement composites and the size distribution



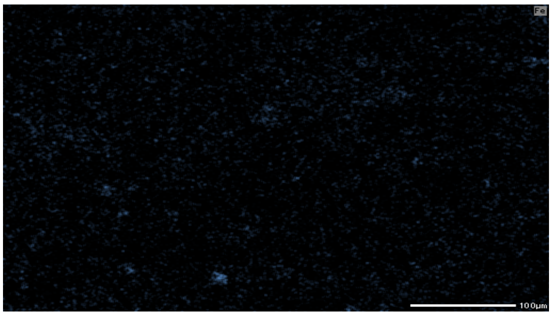
(Al)



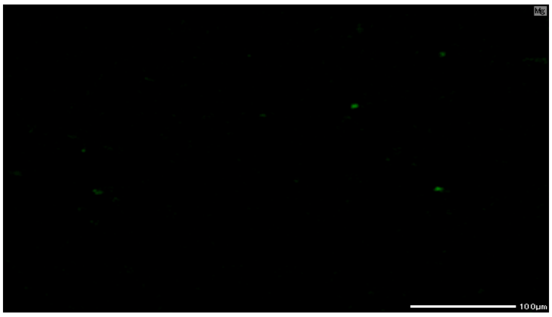
(Ca)



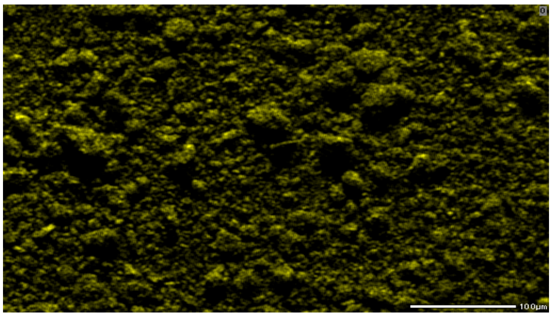
(C)



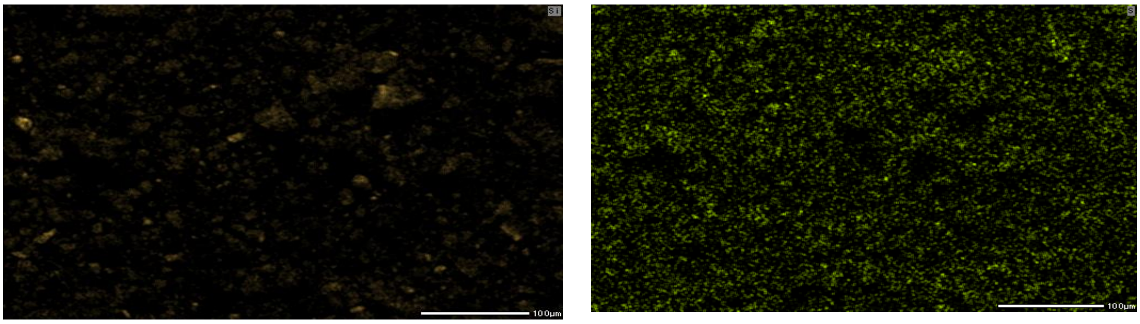
(Fe)



(Mg)



(O)



(Si) (S)

Figure 6. SEM-EDX analysis of marble-cement composites

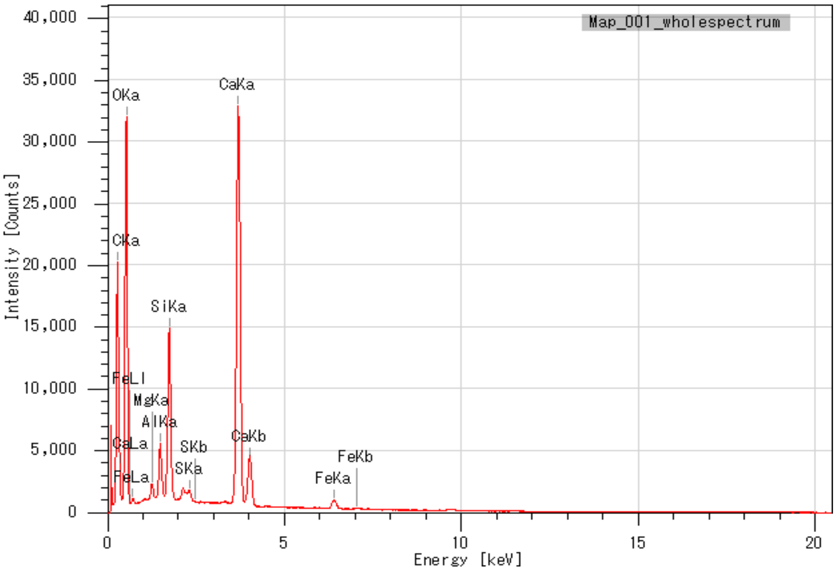


Figure 7. The elements composing marble-cement composites

Compressive strength

The compressive strength properties of the mortar mixes with various percentages of marble powder as a cement replacement at the curing ages of 7, 14, 21, and 28 days were plotted graphically in Figure 8. At 7 days, the maximum compressive strength was achieved by the mix containing 40% marble powder, while the control mix (0%) exhibited the minimum compressive strength. At 14 days, the compressive strength value with a mix composition of 20% marble powder experienced an increase in compressive strength, with a maximum compressive strength value of 26.67 kN. The composition of the mix with 50% marble powder waste produced a minimum value of 16.67 kN. However, after 21 and 28 days of curing soak, the mortar mix containing 20%, 30%, 40%, and 50% marble powder gained lower compressive strength value than previous curing time. Meanwhile, the control mix and the mix with 10% marble powder obtained an increase in the resistance of compression.

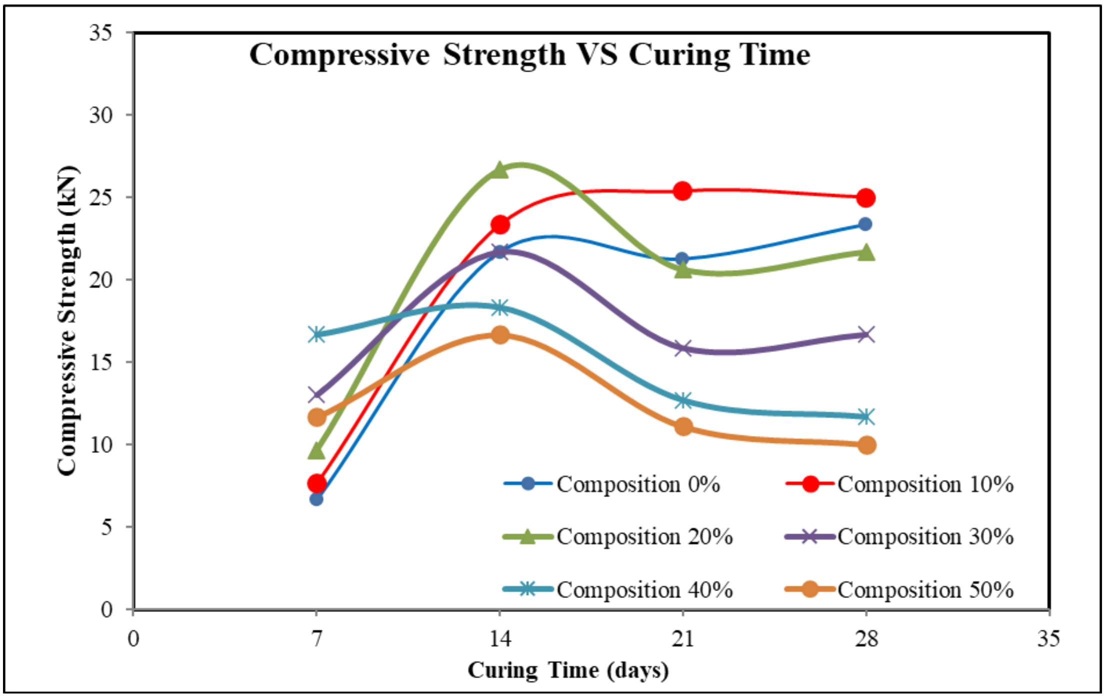


Figure 8. The graph represents the compressive strength of each marble powder percentage in the mortar mix over curing time

The phenomenon shown in Figure 8 explained that the compressive strength value of mortar mix at all ages had increased slightly until the 10% replacement level by marble powder. In contrast, a further increase in the percentage of marble powder caused a decrease in the compressive strength value. It can be concluded that marble powder can be utilized up to 10% as the partial replacement of cement to get the maximum benefits in compressive strength. Previous studies reported that marble powder was used to substitute the use of cement [15]–[20] and sand [21]–[24] in the production of mortar because marble powder could fill pores that could improve compressive strength. However, this property was only possible with a low replacement of cement in the range of 5% to 15% [16]. The decrease in strength also explained that the addition of marble powder only acted as filler and had no significant role in the hydration process [3]. However, the use of waste marble powder improved the physical characteristics of mortar, and they provided an eco-friendly way to produce more sustainable mortar concrete [25]. This statement was also reported by Kherraf et al. that the mortar of waste marble powder at 20% gave the best performance in terms of consistency, air content, flexural strength, and water absorption [26].

Porosity

The porosity of mortars indicated the percentage of void space between material phases in the mortar. In general, the compressive strength is inversely proportional to the porosity value. Based on the test results of mortars with various marble powder percentages, Figure 9 showed that the porosity of the mortar increased as the curing time increased.

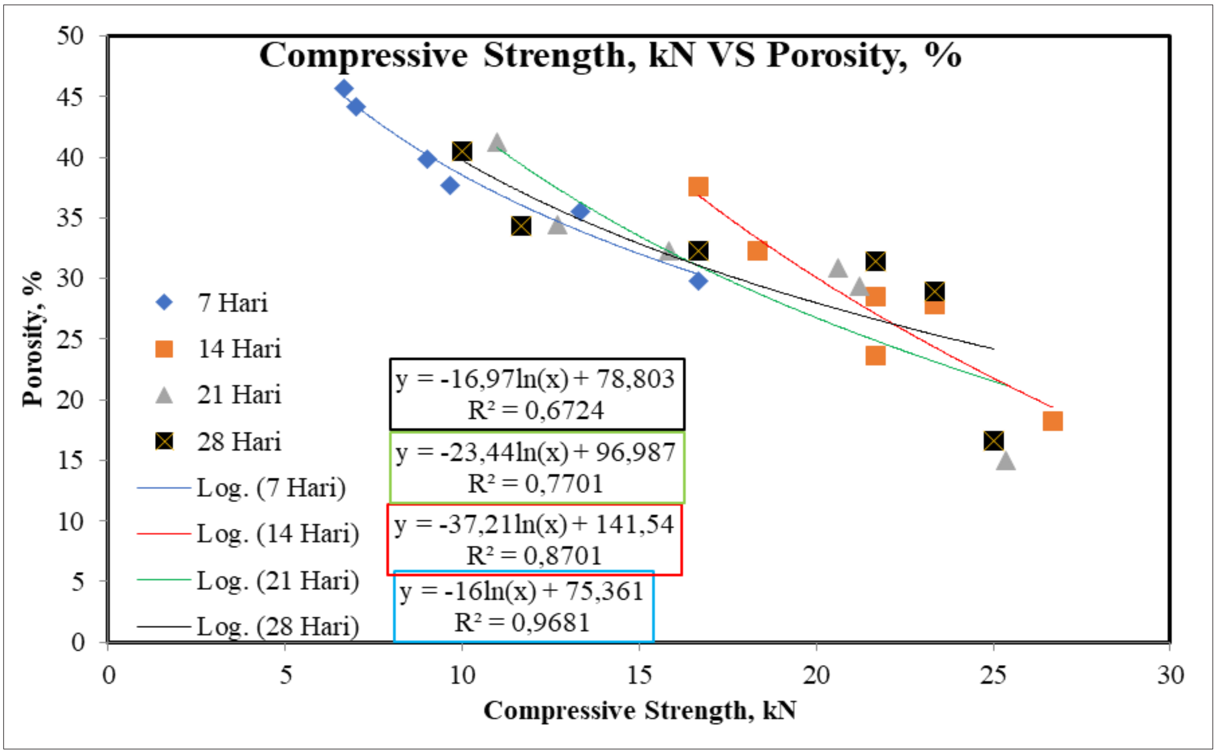


Figure 9. The correlation between compressive strength and porosity of mortars at the age of 7, 14, 21, and 28 days

The increase in mortar porosity was generally due to water's evaporation, which left empty spaces in the matrix and weakened the hydration product bond. Thus, the voids in the mortar continued to grow. The higher the temperature was, the greater the damage was. The percentage of porosity in the mortar would also increase sharply. A study reported by Keleştemur et al. obtained similar results that due to an increase in temperature, the pores containing air and water would gather and form capillaries, which resulted in mortar slip, reduced the binding capacity, and caused water penetration [21].

Contour and surface plot

Response surface methodology (RSM) is a method that combines mathematical techniques with statistical techniques in modeling and analyzing a response that is influenced by several independent variables or factors to optimize the response [27]. Figure 10 illustrates the resulting contour plot, consisting of various color variations that reflect the resulting response's magnitude. This color range would reveal the point of an optimum variable. The optimum condition determination was evidenced by the three-dimensional curve forming the optimum peak, as shown in Figure 10. The maximum condition of the plot was in yellow, with the WCR (water-cement ratio) value of 80% and the cement amount of 80 grams, resulting in a compressive strength value of 20–30 kN.

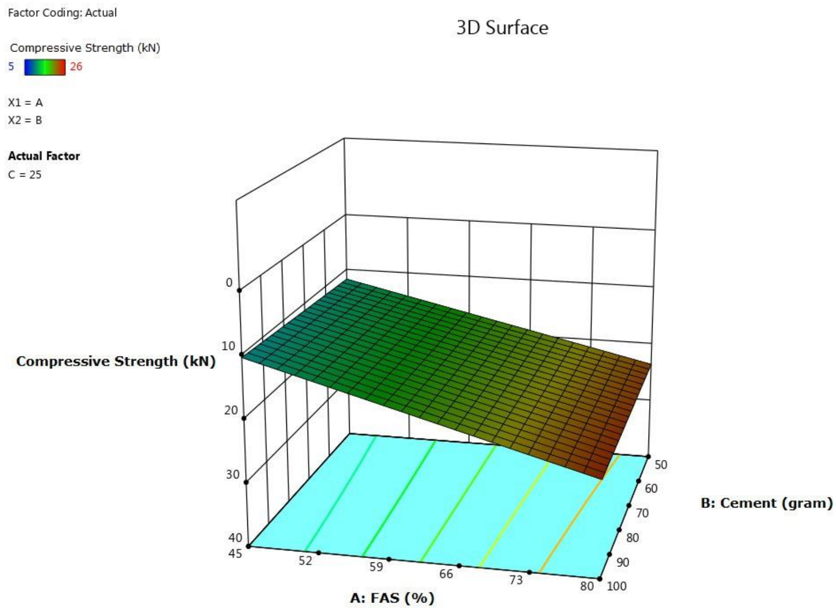


Figure 10. 3D contour plot representing the compressive strength of the mortars

CONCLUSION

The findings of this study concluded that up to 10% of waste marble powder could replace cement in mortar preparation. The substitution with up to 10% marble powder improved the compressive strength of the mortar mix until 28 days of curing time. A large replacement level of cement by marble powder could lead to high porosity. This current study proved that the produced waste from marble mining activity in South Central Timor Regency had a beneficial prospect in the construction field, i.e., utilization of waste marble powder as an alternative material to prepare concrete. A 10% usage of waste marble powder could reduce environmental degradation due to an accumulation of waste.

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