Kastamonu University
Journal of Engineering and Sciences
e-ISSN 2667-8209
http://dergipark.gov.tr/kastamonujes

Multifunctional Utilization of Marble Wastes in One-Part and Two-Part Geopolymer Production Methods: Applications as Filler and Fine Aggregate

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Received: **January 28, 2025** ◆ Accepted: **June 18, 2025** ◆ Published Online: **June 25, 2025**

Abstract: This study compares the one-part and two-part geopolymer production methods, both of which utilise marble powder as a filler material. Marble powder, when used as a filler, is a promising strategy that can significantly enhance the environmental sustainability of construction materials and reduce carbon emissions. The study produced different geopolymer mixtures by replacing ground granulated blast-furnace slag (GBFS) with marble powder at 0% and 30% levels and using sodium metasilicate as an activator. These mixtures' mechanical properties, workability, bulk density, water absorption, and compressive strength were evaluated, with a specific focus on the role of marble powder as a filler material. The results indicate that the addition of marble powder decreases the viscosity of the geopolymer mixtures, with the one-part method exhibiting increased fluidity. However, it was observed that adding marble powder reduced compressive strength. In contrast, the mixtures produced by the two-part method, while more costly, provided materials with a high degree of reliability in their resistance to elevated temperatures and mechanical loads, demonstrating higher durability and thermal stability. The one-part method, with its increased fluidity, offers a practical and easily applicable solution, while the two-part method, despite its higher cost, provides materials with superior resistance to elevated temperatures and mechanical loads. However, both methods offer the unique advantage of significantly reducing carbon emissions when marble powder is used. These findings suggest that marble powder can be used in the production of sustainable construction materials, and the two different production methods offer distinct advantages.

Keywords: Carbon emissions, Environmental sustainability, Geopolymer production methods, Marble powder

Öz: Bu çalışma, mermer tozunun dolgu malzemesi olarak kullanıldığı tek bileşenli ve iki bileşenli geopolimer üretim yöntemlerini karşılaştırmaktadır. Dolgu olarak kullanılan mermer tozu, inşaat malzemelerinin çevresel sürdürülebilirliğini önemli ölçüde artırabilecek ve karbon emisyonlarını azaltabilecek umut verici bir stratejidir. Çalışmada, granüle yüksek fırın cürufu (GBFS), %0 ve %30 oranlarında mermer tozu ile ikame edilerek farklı geopolimer karışımları üretilmiş ve sodyum metasilikat aktivatör olarak kullanılmıştır. Bu karışımların mekanik özellikleri, işlenebilirlikleri, hacim yoğunlukları, su emme kapasiteleri ve basınç dayanımları değerlendirilmiş; özellikle mermer tozunun dolgu malzemesi olarak oynadığı role odaklanılmıştır. Sonuçlar, mermer tozu ilavesinin geopolimer karışımlarının viskozitesini azalttığını ve tek bileşenli yöntemde artan akışkanlık sağladığını göstermektedir. Ancak, mermer tozu ilavesinin basınç dayanımını azalttığı gözlemlenmiştir. Buna karşın, daha maliyetli olan iki bileşenli yöntemle üretilen karışımlar, yüksek sıcaklıklar ve mekanik yükler karşısında daha yüksek dayanıklılık ve termal kararlılık sergileyerek malzemelere yüksek bir güven seviyesi sağlamaktadır. Tek bileşenli yöntem, artan akışkanlığı sayesinde pratik ve kolay uygulanabilir bir çözüm sunarken, iki bileşenli yöntem ise yüksek maliyetine rağmen üstün mekanik dayanım ve yüksek sıcaklıklara karşı direnç sunmaktadır. Bununla birlikte, her iki yöntem de mermer tozu kullanıldığında karbon emisyonlarının önemli ölçüde azaltılması gibi benzersiz bir avantaj sağlamaktadır. Bu bulgular, mermer tozunun sürdürülebilir inşaat malzemelerinin üretiminde kullanılabileceğini ve iki farklı üretim yönteminin kendine özgü avantajlar sunduğunu ortaya koymaktadır.

Anahtar Kelimeler: Geopolimer Üretim Yöntemleri, Çevresel Sürdürülebilirlik, Karbon Emisyonları, Mermer Tozu

1. Introduction

Cement-based composites are among the fundamental construction materials in the modern construction sector, attracting attention due to their wide range of applications. However, cement production significantly contributes to environmental concerns and is associated with high energy consumption and greenhouse gas emissions. The production of Portland cement accounts for approximately 8% of global carbon dioxide (CO_2) emissions, which is considered a significant disadvantage in terms of sustainability [1]. Moreover, traditional concrete has been shown to lack long-term resistance to chemical attacks and to be susceptible to corrosion. These disadvantages have accelerated the development of more environmentally friendly and durable alternative materials [2].

In this context, geopolymers have attracted attention in recent years as environmentally friendly and high-performance binder materials. Geopolymers are aluminosilicate-based materials that are typically derived from industrial by-products. Geopolymers require less energy in their production processes and emit significantly lower CO_2 emissions their production processes [3]. Geopolymers offer several advantages such as higher chemical resistance, lower porosity, and long-term mechanical performance compared to traditional concretes. Consequently, geopolymers are regarded as a promising solution to address the need for sustainable construction materials [4].

In the process of geopolymer production, two main methods are utilized: the one-part and two-part methods. The one-part method signifies an approach in which all components are prepared as a single dry mix and activated by the addition of water. The one-part method is notable for its ease of application and simplicity in production processes, thus offering a reassuringly practical alternative. This method also enables more efficient use of industrial waste and provides significant advantages in terms of environmental sustainability.

Ground granulated blast-furnace slag (GBFS), marble powder, and sodium metasilicate are notable among the binder materials employed in geopolymer production. Despite being an industrial by-product, GBFS demonstrates considerable binding potential, thus establishing it as a sustainable alternative [5]. Marble powder accumulates as a large-scale by-product and contributes to environmental and economic sustainability when used in geopolymer systems [6]. Sodium metasilicate has been demonstrated to accelerate the geopolymerization process, enhancing the early strength of the material [7].

Furthermore, the selection of aggregate in geopolymers significantly impacts performance. Marble powder, utilized as an aggregate, is a cost-effective and readily available material that enhances the mechanical strength of geopolymers while contributing to the effective management of environmental waste. The incorporation of marble powder as an aggregate promotes the conservation of natural aggregate resources and facilitates the recycling of waste materials.

The present study examined the physical, mechanical and durability properties of one-part and two-part geopolymers produced using marble powder. Furthermore, the study investigated the effectiveness of marble powder as a filler in these two mixing methods. In addition, the study analysed the economic and environmental impacts of these composites.

2. Material and Method

2.1. Material

This study utilized ground granulated blast-furnace slag (GBFS) as the primary binder, with marble powder measuring less than 250 microns was employed as a filler material in the mixture. Marble chips ranging from 0.250 to 2.36 mm were utilized as aggregate. The specific gravities of these materials are as follows: GBFS 2.84 g/cm^3 , marble powder 2.66 g/cm^3, and marble crumbs 2.63 g/cm^3. The chemical and physical properties of the GBFS utilized as a binder and the marble powder used as a filler are presented in Table 1. Two types of geopolymer production methods were used for comparison in the study. In the one-part method, sodium metasilicate, with a specific gravity of 2.61 g/cm^3, was used as the activator for forming the geopolymer matrix. Moreover, a superplasticizer (polycarboxylate-based) was used in the one-part method to adjust workability. Conversely, in the two-part method, NaOH with a specific gravity of 1.25 g/cm^3 and Na_2SiO_3 with a specific gravity of 1.42 g/cm^3 were used for matrix formation.

Table 1. Chemical and Physical Properties of GBFS and Marble Powder [8]

Oxides	CaO	SiO_2	Al_2O_3	Fe_2O_3	MgO	Na_2O	K_2O	SO_3	LOI	Color
GBFS	40.6	35.5	11.2	0.7	10.1	0.2	0.4	0.5	0.6	Cream
WMP	53.2	0.4	0.1	0.1	0.4	0.2	0.1	0.1	45.1	White

2.2. Mixture Design

The mixtures that formed the subject of this study were prepared for comparison by means of volume-based measurements. Mixtures produced by the one-part method were coded with the letter 'O,' while those made by the two-part method were coded with the letter 'T.' The total binder content in all mixtures was set at 800 kg/m^3. While the O-0 and T-0 mixtures contained 100% GBFS, the O-30 and T-30 mixtures included 70% GBFS and 30% marble powder. A superplasticizer was used to adjust the workability of the mixtures produced using the one-part dry mix method. While, no superplasticizer or water was used in the two-part method, where liquid activators were employed. In two-part mixtures, the Na_2SiO_3/NaOH ratio was set to 2.5. In one-part mixtures, sodium metasilicate was added at 15% by weight of GBFS. In two-part mixtures, the amount of alkaline solution consisting of NaOH+Na_2SiO_3 was added as 50% of the amount of GBFS. The material quantities for the mixtures are presented in Table 2.

Table 2. Material quantities of mixtures (kg/m^3)

Mixtures	GBFS	Marble Powder (under 250 microns)	Water	Plasticizer	Sodium Metasilicate	NaOH	Na_2SiO_3	Marble crumb
O-0	800	0	240	40	120	0	0	980
O-30	560	240	240	40	12	0	0	985
T-0	800	0	0	0	0	120	280	1062
T-30	560	240	0	0	0	120	280	1067

2.3. Mixing, Molding and Curing

In the geopolymers produced using the one-part method, GBFS, marble powder (if applicable) and sodium metasilicate were mixed for 1 minute to ensure homogeneity. Following this, water was added, and the mixture was stirred for 1 minute at a slow speed, followed by 1 minute at a high speed in order to prepare the paste. Then, the marble crumb was added to form the composite material. In the two-part method, GBFS and marble powder (if applicable) were mixed at a slow speed for 1 minute to achieve homogeneity, followed by the addition of NaOH + Na_2SiO_3 and further mixing for 1 minute at a slow speed and 1 minute at a high speed to obtain the paste. A marble crumb was added to the paste to produce the composite material. The geopolymer composites obtained through both methods were molded and kept at 70℃ for 24 hours. After 24 hours, the samples were demolded and subjected to an additional 48-hour curing period at 70°C before being tested.

2.4. Test Procedures

A series of tests were conducted in order to ascertain the samples specific properties. All of these tests were conducted after 48 hours. The details of these tests along with the standards according to which they were performed can be found in Table 3. The high-temperature test was conducted at the temperatures specified in Table 3. The samples were heated at a rate of 5℃ per minute and then held at the specified temperature for 2 hours. Subsequent exposure to elevated temperatures, the samples' weight loss and compressive strength were measured.

Table 3. Applied test, sample size and standards

	Physical Properties	Mechanical Properties	Sorptivity	High-temperature (Durability)
Sample size (mm)	50x50x50	50x50x50	50x50x50	50x50x50
Standard	ASTM C642	ASTM C109	ASTM C1585	300℃-450℃-600℃

3. Results and Discussion

3.1. Physical Properties

As demonstrated in Figure 1, the flow diameters of the mixtures. Based on this graph, the O-0 mixture contains 0% marble powder, while the O-30 mixture contains 30% marble powder. In terms of flow diameter, it is observed that O-30 (232 mm) has a larger flow diameter compared to O-0 (187 mm). The addition of marble powder to the mixture leads to a reduction in viscosity, resulting in a more fluid consistency. The incorporation of marble powder, a spherical and fine filler material, has been shown to reduce internal friction within the mixture, thereby increasing the flow diameter.

The flow diameter in the T-0 mixture is 155 mm, whereas the T-30 mixture increases to 165 mm. This indicates that marble powder also imparts a certain fluidity to the two-part geopolymer mixtures. This phenomenon can be explained by the particle size.

Overall, the flow diameters of the one-part geopolymers (O-0 and O-30) are greater than that of the two-part geopolymers (T-0 and T-30). The O-30 mixture demonstrates the most substantial flow diameter at 232 mm. The incorporation of a dry mix in the one-part method along with the addition of water, which reacts homogeneously with the binder components, enhances the fluidity of the mixture. The utilization of a superplasticizer in the one-part method significantly affected this outcome.

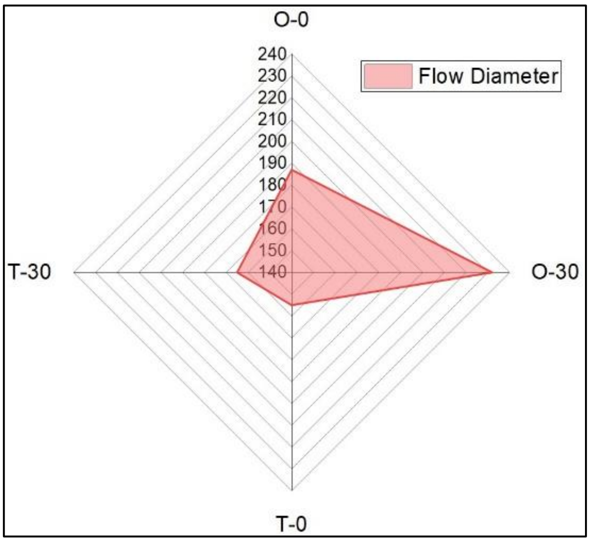

Figure 1. Flow diameters of the mixtures

As demonstrated in Figure 2, the fresh and dry bulk densities of the mixtures are exhibited. Considering the effect of marble powder in the produced mixtures, the increase in marble powder content in the O-0 and O-30 mixtures produced using the one-part method decreases bulk density. This can be attributed to the fact that the density of marble powder is lower than that of GBFS. Similarly, in the T-0 and T-30 mixtures produced using the two-part method, the increase in marble powder content resulted in a decrease in bulk density. However, it is observed that the fresh bulk densities in the two-part method are generally higher than those in the one-part method. This is thought to result from the different effects that the methods have on the reactive matrix structure. The increased use of aggregate in the two-part method during volume calculation is also a contributing factor to this observed difference.

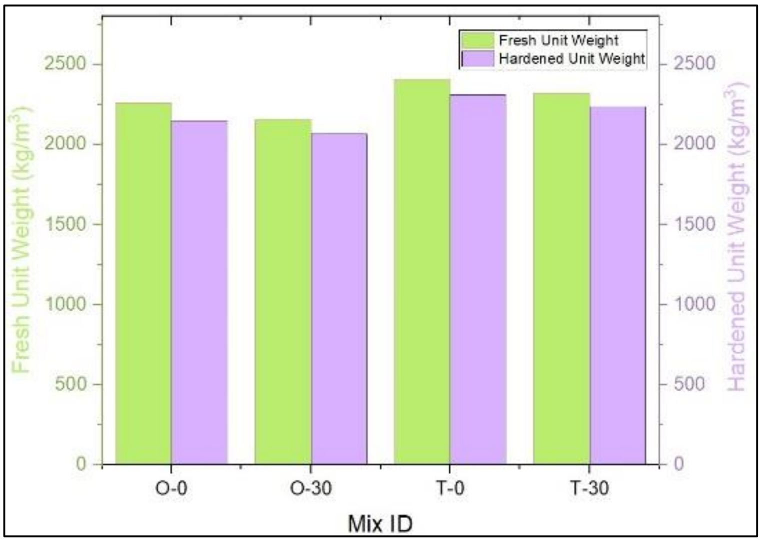

Figure 2. Fresh and hardened unit volume weight of the mixtures

3.2. Sorptivity Properties

Figure 3 below shows the capillary water absorption values of the mixtures. Upon examination of the graph, it is clear that an increase in marble powder content results in an increase in capillary water absorption values. This suggests that marble powder affects the pore structure of the mixture, leading to an enhanced capacity for water absorption. However, this increase is less pronounced in the two-part method compared to the one-part method. This observation indicates that the two-part method results in a more dense and less porous structure.

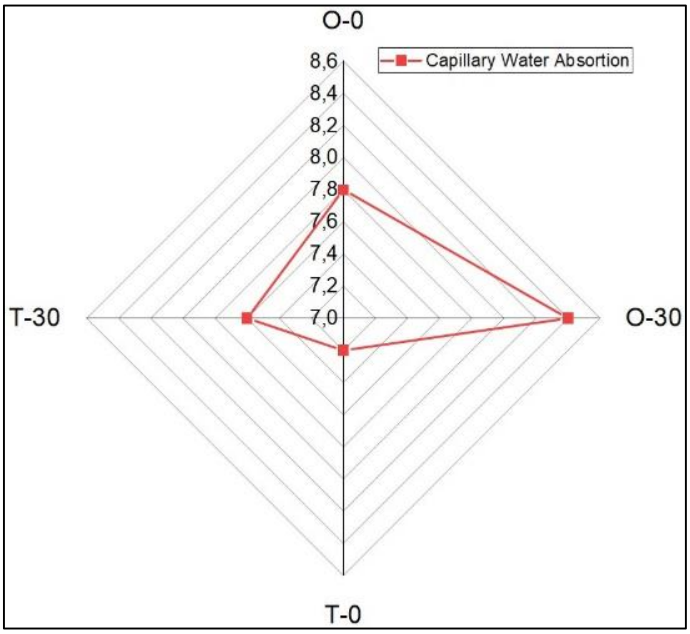

Figure 3. Capillary water absorption values of the mixtures

3.3. Mechanical Properties

Figure 4, shows the compressive strengths of the mixtures, are expressed in MPa. A close examination of the graph, it can be observed that an increase in marble powder content results in a decrease in compressive strength. This finding suggests that marble powder weakens the bonds within the matrix, thereby reducing structural integrity. This observation indicates that marble powder lacks binding properties. A comparison of the two production methods reveals that the two-part method, incorporating marble powder yields composites that exhibit higher strength than those produced by the one-part method.

While the compressive strength values obtained in this study provide valuable insights into the performance of the developed geopolymer composites, it is essential to interpret these results in the context of practical field applications. According to EN 206 and relevant standards for structural concrete, a compressive strength of at least 25–30 MPa is typically required for non-prestressed structural elements. In this context, the two-part geopolymer mixtures, particularly T-0 and T-30, exceed this threshold, indicating their potential suitability for structural or load-bearing applications under specific design conditions. Conversely, the one-part mixtures, particularly those with 30% marble powder substitution, may be more appropriate for non-structural applications such as paving blocks, partition walls, or thermal insulation layers, where mechanical strength requirements are comparatively lower.

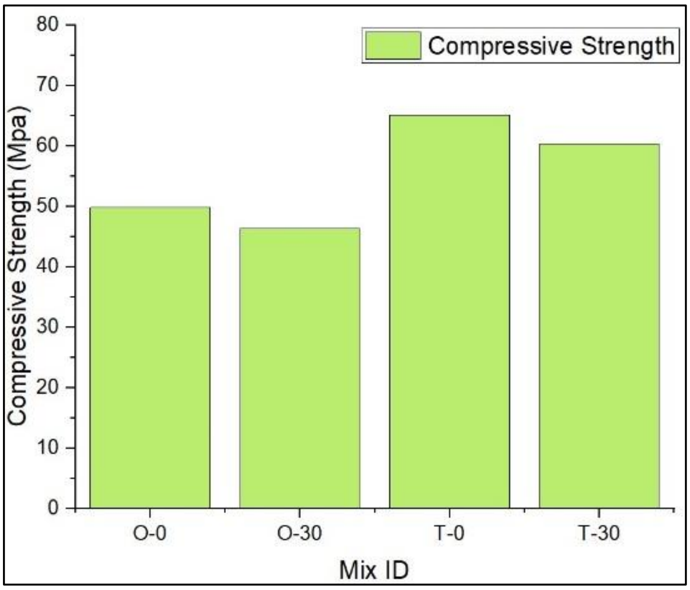

Figure 4. Compressive strength of mixtures

3.4. Elevated-temperature Durability Properties

As demonstrated in Figure 5, the percentage weight changes of the mixtures following exposure to temperatures of 300℃, 450℃, and 600℃ are presented. The graph indicates that mixtures incorporating marble powder (O-30 and T-30) undergo more substantial weight loss at elevated temperatures in comparison to those without marble powder (O-0 and T-0). This observation signifies that marble powder exerts a considerable influence on the thermal behavior of the material under such conditions.

Furthermore, it is also noted that the mixtures prepared using the two-part method (T-0 and T-30) generally exhibit less weight loss than those prepared using the one-part method (O-0 and O-30). This finding suggests that the two-part method forms a more stable structure and performs better under high-temperature conditions.

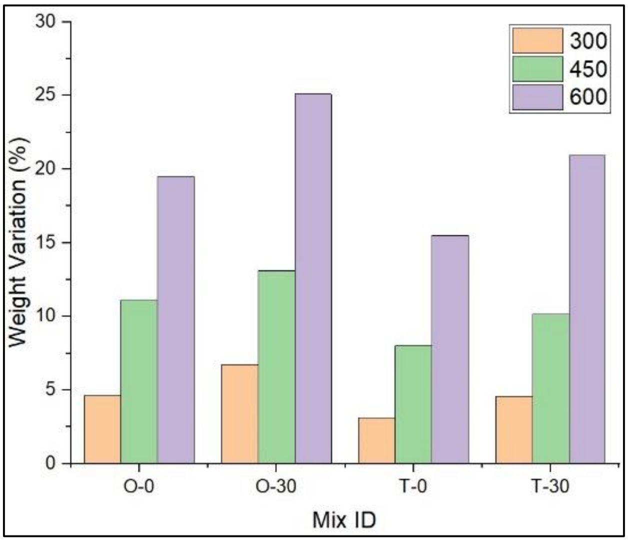

Figure 5. Weight changes after elevated temperature

As demonstrated in Figure 6, the compressive strengths of the mixtures, expressed in MPa, are observed to decrease following exposure to elevated temperatures. It is clear that the mixtures containing marble powder (O-30 and T-30) exhibit a reduction in compressive strength when compared to those devoid of marble powder (O-0 and T-0). Marble powder may weaken structural integrity and degrade rapidly at temperatures above 750℃ [9]. Therefore, the primary degradation hypothesized to occur within the matrix in this study. The findings indicate that all mixtures experience a reduction in compressive strength at elevated temperatures, suggesting that the materials undergo chemical degradation under elevated temperatures.

It has been observed that the mixtures prepared using the two-part method (T-0 and T-30) demonstrate superior performance at elevated temperatures. This method, even with the incorporation of marble powder, yields enhanced compressive strength and demonstrates reduced reduction in strength with increasing temperature. This suggests that the two-part method forms a more robust and thermally stable gel structure [10].

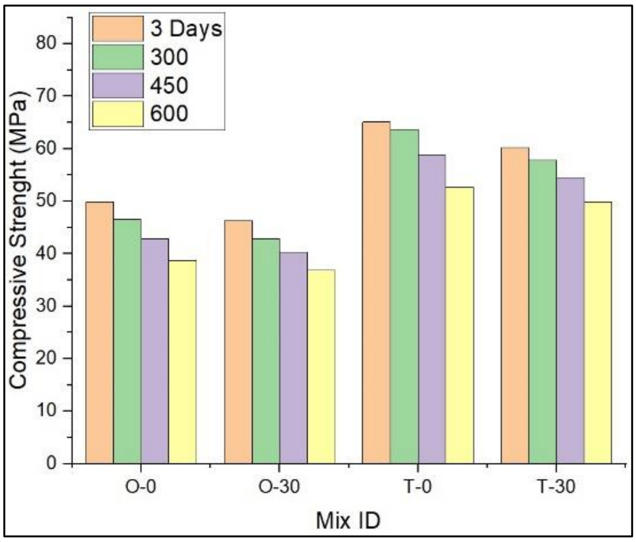

Figure 6. Compressive strength after elevated temperature

Figure 7 shows the appearance of the mixtures after exposure to 600℃. The samples are arranged from left to right as O-0, O-30, T-0, and T-30. In this context, the O-0 sample exhibits noticeable discoloration and surface darkening. This observation indicates that the material has darkened due to carbonization at elevated temperatures.

Upon examination, the O-30 sample appears to be less darkened than the O-0 sample, although small cracks and rough surface can be observed. Marble powder is thought to be the cause of surface deterioration due to thermal expansion at elevated temperatures [11].

The T-0 sample demonstrates a more homogeneous color and less surface degradation compared to the other samples. The two-part method provides enhanced integrity and stability for the material at elevated temperatures.

Despite the presence of marble powder, the T-30 sample appears to have as good an appearance as the T-0 sample.

Figure 7. Post-high-temperature views of the blends (from left to right: O-0, O-30, T-0, and T-30)

3.5. Sustainability and Cost Analyses

Table 4 presents the carbon emissions and unit costs of the materials utilized in the mixtures. The data presented in the table highlights the significance of environmental and economic factors in the selection of construction materials. While the carbon emission values were obtained from the literature, the material costs were sourced from manufacturers. Notably, marble powder and water are distinguished by their low costs and CO_2 emissions, while materials such as metasilicate and superplasticizers are less preferred due to their higher costs and CO_2 emissions.

Table 4. Carbon emissions and unit costs of materials used in mixtures [12–15]

Materials	GBFS	Marble Powder	Metasilicate	Plasticizer	NaOH	Na_2SiO_3	Water	Marble Crumb (Fine Aggregate)
($KgCO_2$/kg)	0.019	-0.08	1.9	1.88	0.46	0.65	0.0003	-0.08
Cost ($/kg)	0.05	0.02	0.044	0.148	0.18	0.125	0.001	0.02

Figure 8 shows the carbon emissions and unit costs of the mixtures. The graph shows that mixtures containing 30% marble powder in both production methods have lower CO_2 emissions than those containing 0%. This demonstrates that marble powder is an effective material in reducing CO_2 emissions.

The CO_2 emissions of mixtures produced using the two-part method are significantly lower than those made using the one-part method. However, the costs of these mixtures are also considerably higher.

The correlation between reduced CO_2 emissions and elevated costs is clear in the T-0 and T-30 mixtures. However, in cases like O-30 mixtures, where both cost reduction and a decrease in CO_2 emissions occur, it suggests that marble powder can be an economically and environmentally advantageous additive material.

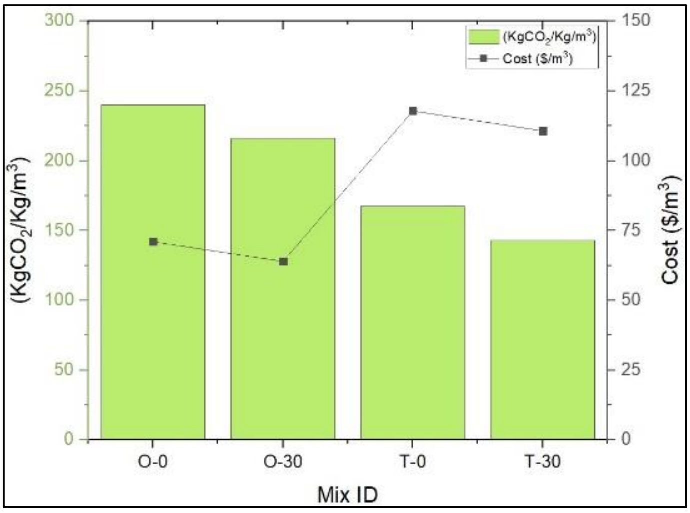

Figure 8. CO_2 emissions and unit cost of mixtures

In addition to the unit costs presented in Table 4, it is important to emphasize that the prices of raw materials were obtained from suppliers operating in the domestic market and reflect the average market values as of the first quarter of 2022. For instance, sodium metasilicate and polycarboxylate-based plasticizer prices were derived from technical-grade industrial quotations, whereas marble powder and marble crumb prices were based on local quarry outputs. These cost values do not include transportation, storage, or operational labor costs, which may vary significantly depending on regional supply chains. Therefore, while the presented cost analysis provides comparative insight into material efficiency, further comprehensive life-cycle cost assessments would enhance the practical applicability of these findings.

4. Conclusions

The addition of marble powder has been demonstrated to enhance the flow diameter in both one-part and two-part geopolymers, thereby improving the mixture's fluidity. One-part geopolymers have been observed to larger flow diameters and provide a more homogeneous mixture. The O-30 mixture has been identified as the most fluid, with the highest flow diameter. The bulk densities indicate that the physical properties of the raw materials and the production methods influence the microstructure and mechanical properties. It is evident that the values of capillary water absorption have a significant impact on material porosity and water permeability. These parameters are affected by the presence of additives and the methodologies employed during the manufacturing process. The incorporation of marble powder into the mixture has been shown to result in a decrease in compressive strength, however the two-part method has been demonstrated to be more effective in mitigating this adverse effect. It has been demonstrated that resistance to elevated temperatures decreases in proportion to the marble powder content; the two-part method produces materials with enhanced heat resistance. It is evident that surface degradation is less pronounced in the two-part method. Conversely, the addition of marble powder to the one-part method has been shown to increase surface degradation. A cost- benefit analysis reveals that the two-part method is shown to be the more expensive method, while the one-part method is the more economical choice.

Conflict of Interest

All authors certify that they have no affiliations with or involvement in any organization or entity with any financial interest or non-financial interest in the subject matter or materials discussed in this manuscript.

Ethics Committee Approval

Ethics committee approval is not required.

Author Contribution

Conceptization: BB, MAMİ, GK, OYB; methodology and laboratory analyzes: BB, MAMİ; writing draft: BB, GK, OYB; proof reading and editing. All authors have read and agreed to the published version of manuscript.

Acknowledgements

It was published in the abstract book presented at the 2nd International Symposium on Innovations in Civil Engineering and Technologies.

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