



Performance of eco-friendly fly ash-based geopolymer mortars with stone-cutting waste[☆]

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HIGHLIGHTS

- The potential of using stone-cutting wastes (SW) in geopolymer mortars was investigated.
- SW increased mechanical strength by up to 40% to ≈ 30 MPa from 16.4 MPa.
- SEM/EDS and XRD analyses were performed on the microstructures of geopolymer mortars.
- Elevated temperatures caused the large acicular crystals in SW-based samples to undergo amorphous transformation.

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ABSTRACT

In this study, the physical and mechanical properties of the geopolymers formed by substituting red (RSW) and yellow (YSW) stone-cutting wastes into geopolymer mortars based on class F fly ash at 10–40 wt% rates were investigated. Sodium hydroxide (NaOH) solution was used as an activator in the production of the mortars. Produced samples were thermally cured at 90 °C for 24 h. Workability, unit weight, flexural strength, compressive strength, water absorption, porosity, and elevated temperature resistance tests (400, 600, and 800 °C) were applied to the produced geopolymer mortars. In addition, the chemical analysis (XRF), crystal phase analysis (XRD) of wastes/selected mortars, and microstructural analysis (SEM) of before and after elevated temperature resistance tests were carried out to investigate the effect of stone-cutting wastes on geopolymer mortars. SW up to 40% enhances mechanical strength up to 26–30.7 MPa from 16.4 MPa due to its high Si/Al ratios and Ca content and needle-like crystals formed by stone-cutting waste. The crystalline phases are determined to be mullite, quartz, anorthite, and zeolite derived from stone-cutting waste in the selected mortars. As a result, it was seen that the use of stone-cutting wastes up to 40% improved the physical, mechanical, and microstructural properties of fly ash-based geopolymer mortars.

1. Introduction

The requirement for raw materials increases day by day due to the uncontrolled consumption of natural raw materials. The scarcity of raw material resources leads to the search for new resources. As a result, the evaluation of solid wastes as alternative raw materials comes to the fore. Although the storage of these solid wastes poses environmental problems, significant gains are achieved by evaluating them in different sectors. Nowadays, waste reduction and disposal in the industrial and mining sectors is an environmental and climate change priority. The main demands for sustainable production have been the recycling of

industrial wastes, conserving natural resources, and minimizing the need for final costs [1,2]. When the construction sector is taken into account, greenhouse gas emissions occur during the production of Portland cement and concrete. Considering the damage caused by this gas emission to the environment, geopolymers can offer alternative solutions to such problems using secondary industrial products and the production of heat and abrasion-resistant materials by trapping industrial wastes in the structure [3,4]. The most crucial advantage of geopolymers compared to Portland cement is the meager amount of CO₂ released in the production of geopolymers. The absence of a high-temperature calcination step during geopolymer synthesis, it's a

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Fig. 1. Stone-cutting waste (SW) preparation process in factory and mortar production in the laboratory.

low-carbon binder, and clinker-free creates this advantage. Increase in the importance of the construction industry and technological development, the importance of lighter and better mechanical properties geopolymers has also increased [3,4]. Geopolymer, a new environmentally friendly inorganic binder produced with activators that activate aluminosilicate-sourced materials (such as metakaolin, fly ash, and slag), has received significant attention as an alternative to cement in recent years [5,6]. Geopolymerization is a chemical process that involves the formation of a three-dimensional polymeric network known as a geopolymer. It serves as an alternative to traditional cement-based materials for construction applications. The process utilizes aluminosilicate source materials, such as fly ash, metakaolin, or slag, and an alkaline activator solution [6]. Various applications have been made in non-combustible plastics, polymer production with high-temperature resistance, aircraft cabins, and runways. Some research uses decorative stone products, building materials, ceramic tiles, refractory materials, biotechnology, composite materials for infrastructure, repair and reinforcement, automobiles, high-tech resin products, radioactive and toxic waste prevention, old historical masonry buildings, etc. [7,8]. It's crucial that the specific geopolymerization process may vary depending on the chosen aluminosilicate source material, activator solution, and desired properties of the final product. Research and development in geopolymer technology continue to expand, aiming to optimize the process and explore new applications for this sustainable construction material. In addition to the concrete industry, geopolymers in different industrial and scientific fields are also being investigated [8].

Fly ash is an industrial waste formed by coal combustion, mixed with the atmosphere as dust, and obtained by holding micron-sized particles by electrostatic precipitating filters or filter bags in the chimneys of thermal power plants stacking them in fly ash silos [9]. The cost of fly ash, which is one of the primary materials used in the production of geopolymer concrete, has increased with the increase in the value of the geopolymer and the distance to the thermal power plants. Moreover, as fly ash can be used in concrete, cement, aggregate, foam concrete, brick production, road, and ground improvement applications, the production cost of the material has increased. The manufacturers have turned to different raw materials or solid waste materials [10]. Considering the studies in the literature, the effects of red clay ceramic powder [11], fine waste dust from brick-tile production [12,13], ceramic tile waste [14–17], and ceramic sanitaryware waste [18–20] on the performance of geopolymers have been evaluated in the last decades.

Another waste material on which research has been done in the literature is stone-cutting waste. Natural stones are natural pozzolan materials. According to ASTM C 618–03, pozzolans are siliceous or aluminous materials that non-binding, fine-grained state due to their natural structure or as a result of grinding. In humid environments, it reacts with lime (calcium hydroxide) and thus, gains binding properties [21–23]. When the studies made with stone-cutting wastes are investigated, it is seen that these wastes are used in application areas such as lightweight wall blocks, composite materials, brick components, and road stabilization materials [23–29]. It can be seen that the most commonly used stone-cutting waste in geopolymer studies is marble powder [25,30]. It is seen that geopolymer studies including granite [31], feldspathic [25], clay-waste [32], and volcanic cutting wastes,

have generally been carried out in the last few years. Depending on the cutting activities carried out in the quarries and factories, natural stone waste occurs at the rate of 40–70% of the production [33]. Nowadays, large amounts of waste are produced worldwide in the extraction and conversion of natural stones, representing an important sector of the economy. The possibility of recovery and the characteristics of these wastes show that the construction and ceramic industry can absorb them for new applications [34]. The stone-cutting wastes pose a serious environmental problem and new studies regarding wastes will prompt researchers to further research and development in this direction. The production and properties of wastes for stones-based geopolymer are still needed to be studied.

The history of stonework in the Cappadocia region of Central Anatolia (Turkey) dates back to ancient civilizations and has substantial potential for the region. Nevsehir stone, which is the common architectural material of the region, can be quickly processed because of its soft structure when it comes out of the quarry due to the volcanic structure of the region. However, it hardens after contact with air and turns into a very durable building material. Due to the large amount of material used and its easy processing, stonework, which is unique to the region, has developed and become an architectural tradition. There are many companies in the region where these stones are produced. In these companies, a high amount of cutting waste is generated when forming stones into products [35,36]. In this study, the use of volcanic tuff stone cutting wastes released after stone-cutting (SW) in Nevsehir province in geopolymers produced with fly ash and its effect on the mechanical and microstructural properties of geopolymers were investigated. Cutting wastes (red and yellow stone) obtained from the stone enterprise where a high amount of cutting waste is generated were used in geopolymer mortars. The wastes in specific proportions instead of fly ash and the physical, mechanical, and microstructural properties of the formed geopolymers were compared with the fly ash-based geopolymer.

In recent years, researchers have focused on exploring the potential of utilizing various industrial wastes such as ceramic wastes, red mud, and slags, as well as natural pozzolans. Turkey, known for its abundant deposits of natural stones like marble, granite, and travertine, has also become a focal point for investigation. This study aims to investigate the effectiveness of using cutting tuff stone wastes from the Cappadocia region in the production of geopolymer mortars. By utilizing these waste materials, not only can the benefits of waste reuse be realized, but also the mechanical and microstructural aspects of geopolymerization in cutting waste-based mortars are thoroughly examined. Furthermore, unlike previous literature, this study delves into the relationship between microstructure and mechanical properties, specifically focusing on the discussion of crystal phase formation at high temperatures ranging from 400 to 800 °C.

2. Materials and methods

Yellow (YSW) and red (RSW) stone-cutting waste, fly ash (class F), river sand, sodium hydroxide, and water were used to create an alkali-activated mortar. The fly ash was obtained from Sugozu thermal power plant (Adana/Turkey). The river sand's saturated dry surface-specific gravity is 2.66 g/cm³ and the water absorption rate is 1.90%

Table 1
Chemical compositions of FA, YSW and RSW.

Chemical composition (%)	FA	YSW	RSW
SiO ₂	60.51	70.14	65.39
Al ₂ O ₃	21.69	14.48	15.64
CaO	1.52	2.12	4.31
K ₂ O	2.58	3.00	4.45
Na ₂ O	0.92	0.13	–
Fe ₂ O ₃	7.85	1.23	1.57
TiO ₂	–	0.26	0.33
MgO	1.55	–	0.42
P ₂ O ₅	–	–	0.18
BaO	–	–	0.15
MnO	–	–	0.06
SO ₃	0.53	0.69	0.08
SiO ₂ /Al ₂ O ₃	2.79	4.84	4.18
L.O.I	3.30	7.99	7.42

L.O.I: loss of ignition.

Table 2
Mixture proportions of geopolymers (g).

Sample codes	RSW	YSW	River Sand	Fly ash	NaOH solid	Water
100 FA(Conrol)	–	–	1350	450	87	180
10 RSW	45	–	1350	405	87	180
20 RSW	90	–	1350	360	87	180
30 RSW	135	–	1350	315	87	180
40 RSW	180	–	1350	270	87	180
10 YSW	–	45	1350	405	87	180
20 YSW	–	90	1350	360	87	180
30 YSW	–	135	1350	315	87	180
40 YSW	–	180	1350	270	87	180

[37]. It obtained from the Nevşehir region in Turkey was employed for the manufacture of mortars. The purity of sodium hydroxide pellets used as an activator is 98.27%. The mixtures used tap water from Nevşehir [38]. Stone-cutting wastes in red (RSW) and yellow (YSW) colors were obtained from Ozkapadokya Stone (Nevşehir/Turkey) company in the form of dust and broken stone pieces. Stone waste preparation process in

the stone production factory and SW-based mortars in the laboratory are shown (Fig. 1). Unit volume weights of stones were 1.50 g/cm³ (YSW) and 1.58 (RSW) g/cm³. The total porosity of stones rate was 30–32% [39] and for fly ash 6.97% [40]. The stone-cutting wastes were ground to pass through a 125-µm sieve for use in the experiments. Chemical analysis of the stone-cutting wastes and fly ash (FA) was done using an X-ray fluorescence (XRF, Rigaku Bruker Tiger S8) device, whose results are shown in Table 1. The crystal phases of stone-cutting wastes were determined by X-Ray Diffraction Device (XRD, Rigaku Miniflex).

The amounts of ingredients used to prepare the mixture are given in Table 2. The liquid/binder (180g/450g) ratio was 0.4 and the sand/binder (1350g/450g) ratio was 3 in the prepared mortars. The concentration of sodium hydroxide solution (NaOH) used as an activator was 12 M. The determined amount of water (180 g) and sodium hydroxide pellets (87 g) were added to the glass jars, and the chemical was entirely dissolved by shaking for about 1 min. Since the reaction of sodium hydroxide with water is exothermic, the solution was waited to cool down to room temperature. With these solutions at room temperature, mortar samples of 40 mm × 40 mm × 160 mm were produced [41]. The solution, ash and wastes were first placed in the mixing bowl and mixed for 30 s, and then the sand was added in the 30th second. Afterward, the mixing continued for another 30 s. Then, the mixing process was completed by maintaining the mixing process at high speed for another 60 s. A flow table test was carried out on the mortars in their fresh state by TS EN 1015-3 standard [42]. The mixtures poured into the 3-cell mortar molds were placed in a laboratory oven with their molds and subjected to thermal curing at 90 °C for 24 h (optimum curing conditions) [19,20]. After thermal curing (at 90 °C, for 24 h), the mortar samples removed from their molds were kept in laboratory conditions at approximately 23.0 ± 2.0 °C for 28 days. The geopolymer chemical reaction period is fast, and the required curing period may be within 24–48 h [43]. The unit weight (UW) test was applied to the geopolymer samples 24 h after the heat-curing. Flexural and compressive strength tests were performed on the same samples following TS EN 1015-11 standard [44]. Apparent porosity (AP) and water absorption (WA) tests were conducted on mortars according to ASTM C642 [45]. In order to determine the physical and mechanical structure changes on

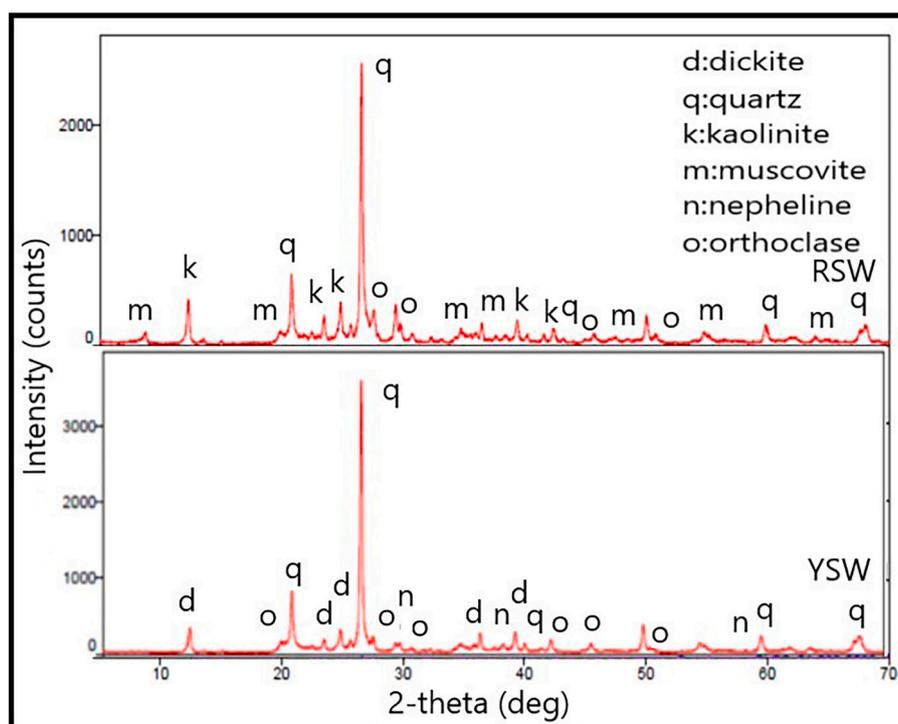
**Fig. 2.** XRD analysis of stone wastes.



Fig. 3. Workability values of mortars.

geopolymer surfaces exposed to elevated temperature (400-600-800 °C), flexural, and compressive strength losses were determined at ambient temperature by exposing the samples to high-temperatures separately at 400, 600, and 800 °C for 30 min with a temperature increase of 5 °C/min in the high-temperature furnace (the elevated temperatures were applied to samples after 28 days).

Three temperatures (400, 600, and 800 °C) were selected to investigate the bonding properties of the mortar. The first two temperatures correspond to temperatures both below and above the decomposition temperature of the river sand, which is 570 °C. The last temperature represents typical fire rates that could be encountered in residential and industrial environments.

3. Results and discussion

While the amount of $\text{SiO}_2 + \text{Al}_2\text{O}_3$ is 82.2 in the chemical analysis of fly ash, this ratio is 81.03 for RSW and 84.62 for YSW (Table 1). RSW and YSW have pozzolanic activity due to the high SiO_2 content as well as the $\text{SiO}_2 + \text{Al}_2\text{O}_3 + \text{Fe}_2\text{O}_3 > 70\%$ according to ASTM C 618.23 Moreover, the total alumina-silica ratios are close to each other and also, the

iron oxide content of the fly ash is higher than the stone-cutting wastes, and on the other hand, the amount of CaO is low. Furthermore, while the $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratios of stone-cutting wastes are 4.84 and 4.18, this value is 1.52 for fly ash. This comparison indicates that mortars containing stone waste form a more stable silica-rich gel structure [46]. As a result of phase analysis, quartz (SiO_2 , Jcpds:01-083-0539), muscovite ($(\text{Si}_{3,38}\text{Al}_{0,62})\text{O}_{10}(\text{OH})_2$, Jcpds:01-079-6478), kaolinite ($\text{Si}_2\text{O}_5(\text{OH})_4$, Jcpds:01-080-0886), orthoclase (KAlSi_3O_8 , Jcpds:01-075-1592) minerals in RSW, quartz, orthoclase, dikite ($\text{Al}_2(\text{Si}_2\text{O}_5(\text{OH})_4$, Jcpds:01-072-8193) and nepheline ($\text{Na}_3(\text{K}_{0,57}\text{Na}_{0,24})(\text{Al}_{3,8}\text{Si}_{4,16}\text{O}_{16})$, Jcpds:01-078-8390) minerals (minor phase) were determined in YSW (Fig. 2).

The workability test was first applied to the fresh mortar samples produced in the study and the results are given in Fig. 3. The workability values of the mortars prepared with stone-cutting wastes substituted by fly ash at the rate of 10–40% are 125–175 mm. It is seen that both stone-cutting wastes exhibit similar behavior and decrease the workability values when compared to the control sample (215 mm) containing 100% fly ash. This situation has also been indicated in previous studies [47, 48]. In the literature, the specific surface area, grain shape, activator

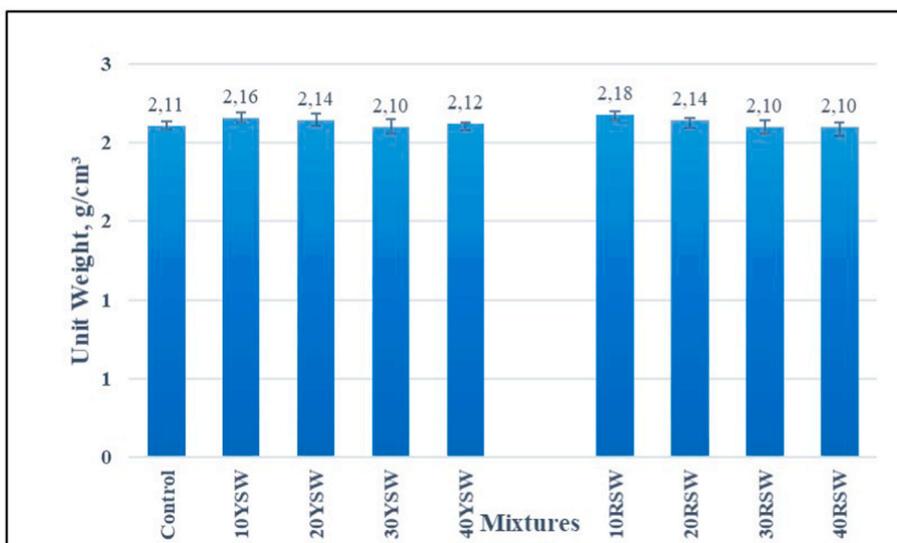


Fig. 4. Unit weight values of mortars.

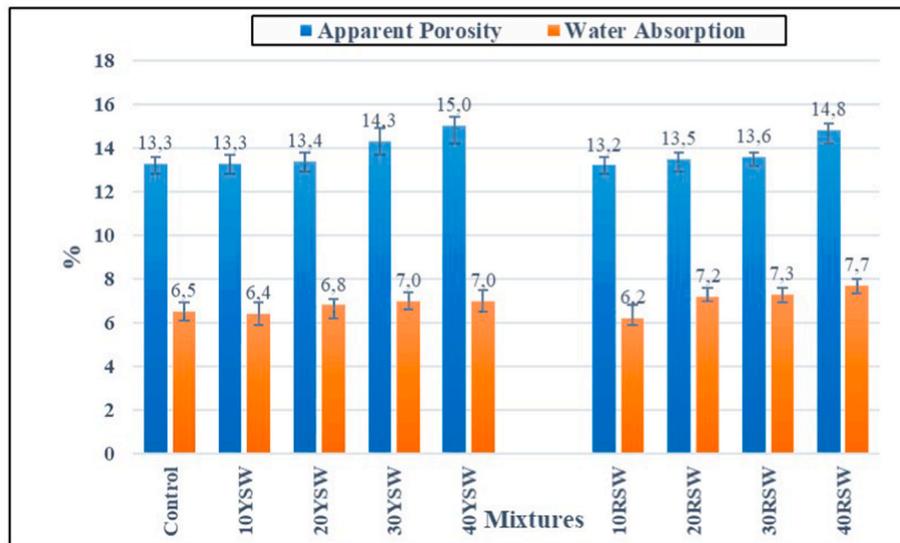


Fig. 5. Water absorption and apparent porosities of produced mortars.

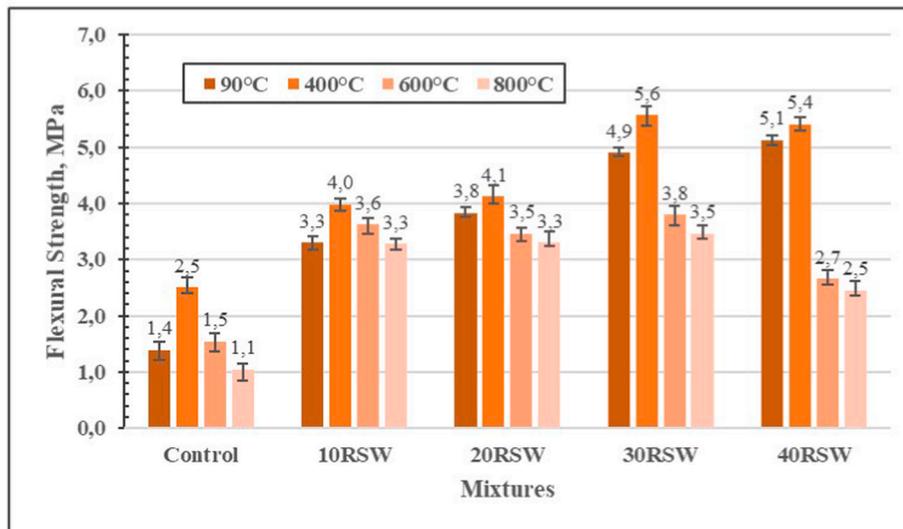
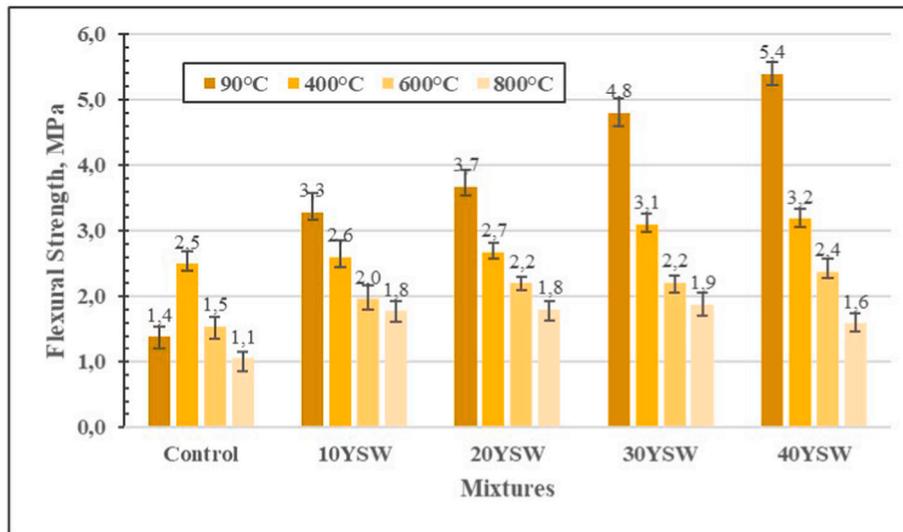


Fig. 6. Flexural strength values of mortars, a) YSW-based mortars, b) RSW-based mortars.

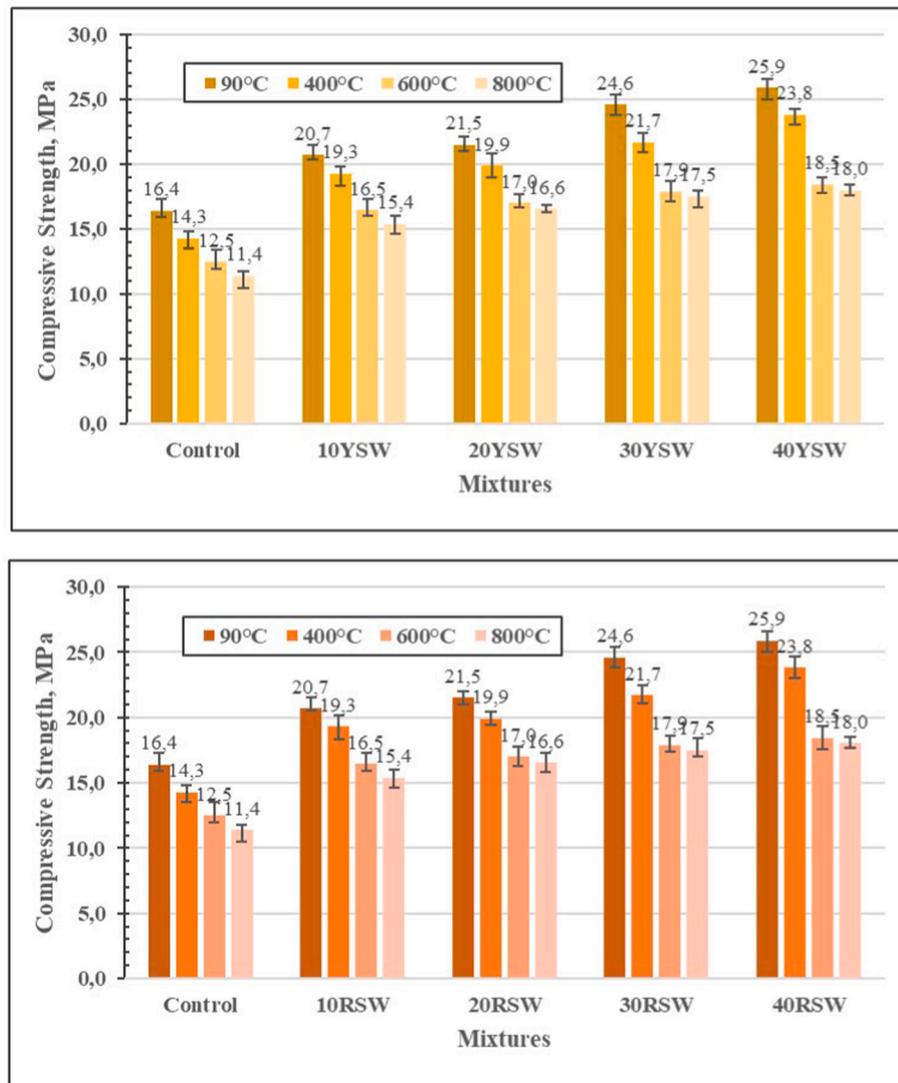


Fig. 7. Compressive strength values of mortars.

ratio, chemical content, and the development of the reaction rate have been shown as factors affecting the workability [48,49]. Unlike fly ash which properly flows due to the spherical particles, using stone wastes (pozzolan) reduced the workability due to the physical shapes. Considering the constant NaOH solution concentration, when the RSW and YSW ratio of the mortars increases, the water requirement for the flow diameter increases due to the high specific surface and shape structure of the RSW and YSW. Nevertheless, when the RSW and YSW ratio of mortars increases, strength values increase [22,25].

The unit weights slightly increased with the increase of both stone-cutting wastes up to with 30% in mortars. While the unit weights are between 2.16 and 2.12 g/cm³ in the samples containing YSW, it is between 2.18 and 2.10 g/cm³ in the samples containing RSW. However, the values in both groups are quite close to each other and control (Fig. 4). A similar variation in the unit weight of the geopolymers with the andesite, recycled sand content was reported approximately 2.00–2.20 g/cm³ [50,51]. Unlike fly ash which properly flows due to the spherical particles, using stone wastes (pozzolans) reduced the workability due to the physical shapes. In particular, the aggregates from stone wastes such as granite, marble etc. are angular and rough [52].

The water absorption and apparent porosity test results are shown in Fig. 5. In the mortar groups containing YSW, the WA was calculated between 6.4 and 7.0% and the AP between 13.3 and 15%. In the mortar

groups containing RSW, WA was determined between 6.2 and 7.7% and AP between 13.2% and 14.8%. The WA and AP of the samples containing stone-cutting wastes, increased up to 13–20%, respectively, are higher than the control sample (WA:6.5; AP:13.3). An increase in WA values was determined due to porosity in samples containing 30–40% stone-cutting waste. The unreacted particles could serve as rigid fillers in geopolymer paste, however, more microcracks could be observed in the matrix. Different morphologies were obtained for the geopolymer mortars with different stone waste replacement ratios [25–28]. Some natural pozzolans may create problems because of their physical properties (angular and porous form, requires a high water content) [22,52]. When utilizing stone wastes in geopolymer mortars, their irregular and angular shapes, as opposed to rounded or smooth particles like fly ash, offer improved interlocking and mechanical stability. Additionally, their porous nature includes small voids or pores within their structure, enabling them to absorb and retain water.

The results in the flexural strength (FS) of the produced mortars are presented in Fig. 6. The measured FS of mortars prepared with the increasing addition of RSW are 3.3 MPa; 3.8 MPa; 4.9 MPa; 5.1 MPa, respectively. Due to the increase in RSW (30–40% RSW), an increase in FS of 250–264% compared to the control sample containing 100% fly ash was detected (1.4 MPa). This rate was determined as 242; 285% in samples containing YSW (30–40% YSW). It is seen that both stone-

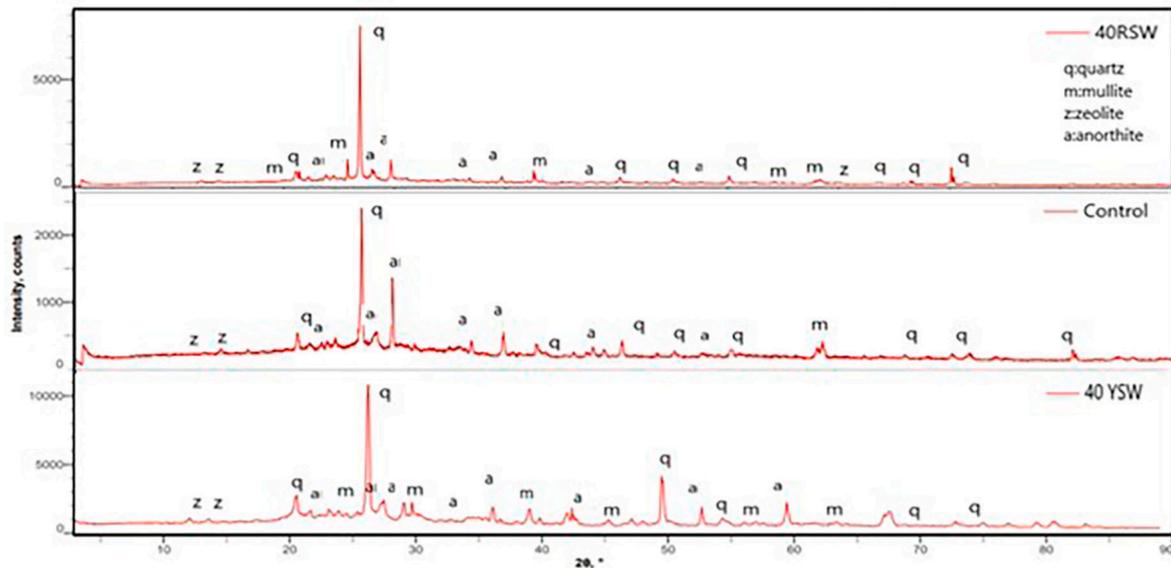


Fig. 8. XRD analysis of selected mortars at 90 °C.

cutting wastes increase the flexural strength (1.4 MPa) compared to the control sample. Fly ash, which can form reactive Si and Al in the reaction process with alkali solution, has an important role in mechanical strength in fly ash-based mixtures. The main reaction product is A-S-H type gel, which provides good mechanical strength. Fine filling materials increase the density of packaging [53]. There are two types of alkali aluminosilicate gel, Al-rich and Si-rich gel. The interaction between the Al-rich gel and the unreacted fly ash particles causes more microcracks to form in the gel matrix, reducing the strength [54]. As the Si/Al ratio in the mixture increases (Table 2), the FS of the mortars will increase thanks to the good bonding between the geopolymer mortar and the aggregate, and the strong Si–O–Si interaction [48]. It is seen that the increasing Si/Al ratio due to the increase in the amount of both yellow and red stone-cutting waste affects the flexural strength. As much as the Si/Al ratio, Ca in the gel structure is also important in forming C-A-S-H and N-A-S-H alkaline activated gels, and Ca has a positive contribution to the development of strength [55]. When the mortars containing the two wastes are compared with each other, it is thought that the high Si/Al content in the samples containing the RSW, and the high Ca content provide higher FS values.

In the literature, it is stated that the microstructure is affected by high-temperature and the formation of micro cracks is the reason for the decrease in flexural strength under the influence of temperature [56]. While the FS of the control sample at 400–600–800 °C was 2.5; 1.5; 1.1 MPa, respectively, the range of FS observed was from 2.6 to 3.2 MPa depending on the amount of waste increase in the samples containing YSW at 400 °C temperature application.

For the control sample, FS was observed to be 1.5 MPa and higher FS was observed from 2.0 to 2.4 MPa for the samples containing YSW at 600 °C. At 800 °C, 1.9 MPa(30YSW)-1.6 MPa(40YSW) values were observed compared to the control sample (1.1 MPa) (Fig. 6a). It is also observed that the increase in the RSW content in the mortar increases up to 60%(10RSW); 64%(20RSW); 124%(30RSW); 126%(40RSW) at 400 °C. The variation of values is determined as 140%; 133%; 158%; 80% according to the increase of the RSW amount at 600 °C. FS is observed that the increase of the RSW content in the mortar hikes up to 200% (10RSW); 200% (20RSW); 218% (30RSW); 127% (40RSW) at 800 °C in Fig. 6 b. This indicates that samples containing stone-cutting waste are less adversely affected by high-temperatures compared to the control sample.

The variation in the compressive strength (CS) of the prepared mortars is presented in Fig. 7. Depending on the amount of waste

increase, the CS of the samples containing RSW was determined as 20.7 MPa (10RSW); 21.5 MPa; 24.6 MPa; 25.9 MPa (40RSW). A higher increment rate of CS is observed from 26 to 58% for the samples containing RSW than control sample (16.40 MPa). For 30YSW and 40YSW, this rate is 50–58% in the samples containing YSW at 90 °C. According to the results, the compressive strengths of stone-cutting wastes-based mortars are increased over the control sample for all temperature conditions. In the literature, it is seen that the stone-cutting wastes such as marble-travertine, volcanic stone, etc. of stone cutting waste-based mortars instead of materials such as fly ash and slag have been investigated for the geopolymer mortars with different molarity, activator ratios. For SW-based mortars, maximum CS was observed to range from 28 to 38 MPa, with up to 50%, and the CS decreased to 14 MPa when used up to 75% in these studies [25,34,48,53] However, studies on the effect of high-temperature exposure on the microstructural variation of geopolymers formed by stone-cutting wastes are limited. In this study, the effect of composition and different temperature applications was examined in detail in microstructure analysis and how it reflected on strengths is also shown. When Fig. 7 is examined, it can be seen that the CS values of all samples decrease with increasing temperature, but the CS of the samples containing stone-cutting wastes is higher than the control sample.

While the CS of the control sample at 400–600–800 °C was 16.4 MPa; 14.3 MPa; 12.5 MPa; 11.4 MPa, respectively, the CS range of YSW-based samples reached 23.8 MPa from 19.3 depending on the amount of waste increase at 400 °C. For 600 °C, maximum CS was observed to surge from 16.5 to 18.5 MPa for YSW-based samples compared with the control sample with 12.5 MPa. At 800 °C, 17.5 MPa (30YSW)-18 MPa (40YSW) values were observed compared to the 11.4 MPa value of the control sample (Fig. 7a).

The measured increment of compressive strengths, mortars prepared with the increasing addition of RSW, are 86% (10RSW); 109% (20RSW); 121% (30RSW); 118% (40RSW), respectively at 400 °C. The variation of the values according to the amount of RSW at 600 °C was determined in the range of 82–108%. At 800 °C, an increase by more than 122% was observed in compressive strengths compared to the control sample (Fig. 6b). According to Davidovits [57], if Si/Al ratios are of 3.5–4.5, the molecular stability of geopolymer mortars is higher. An increase in SiO₂/Al₂O₃ ratio may be responsible for the augments in CS due to Si–O–Si bonds, which are more potent than Si–O–Al bonds [58–61] In the literature, the usage of Ca-rich materials in geopolymer mortars has been reported to improve strengths [47,61–64] Accordingly, it is

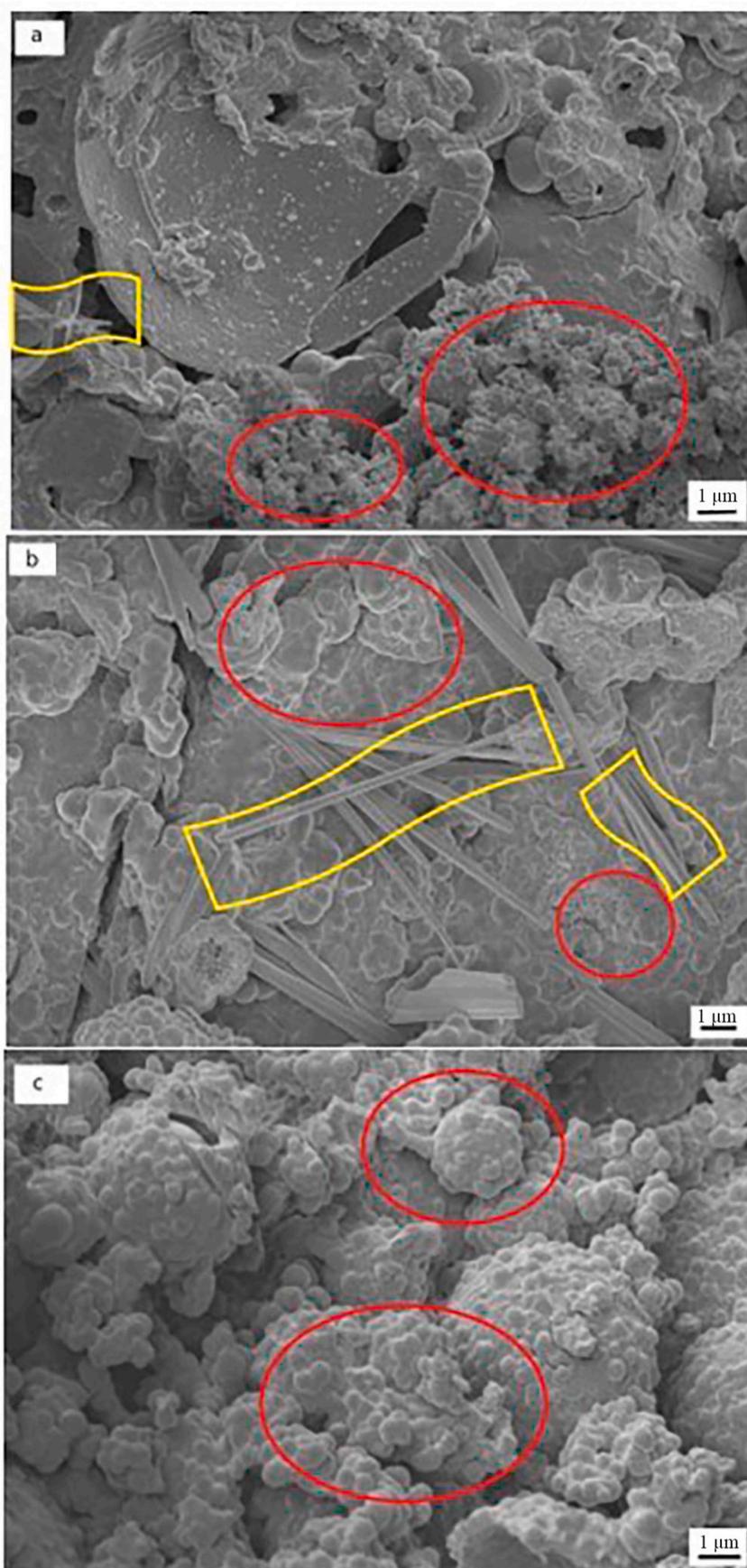


Fig. 9. SEM images of control sample a)at 90 °C, b)at 400 °C, c)at 600 °C.

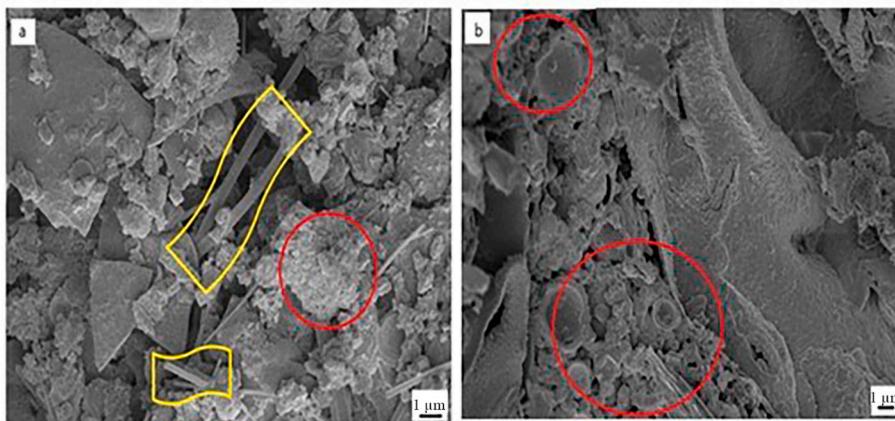


Fig. 10. SEM images of a)10RSW b)10YSW at 90 °C.

thought that both the high Si/Al ratio and CaO content of the samples containing stone-cutting wastes increased the compressive strength values compared to the control [57].

XRD patterns of selected mortars are presented in Fig. 8. Crystalline phases are identified as quartz (Jcpds:00-001-0649), zeolite (Jcpds:00-019-1183), anorthite (Jcpds:00-002-0523), and mullite (Jcpds:00-002-0415). Phase analysis of the samples stone-cutting wastes were included in the highest ratios instead of fly ash were compared; It is thought that especially in 40RSW and 40YSW, the prominent and numerous mullite and anorthite crystals develop due to the high Al_2O_3 , CaO, and SiO_2 content in the stone-cutting wastes compared to the fly ash in its composition. Quartz is recognizable, and the intensities of its diffraction peaks increased compared to raw stone wastes (before the preparation of mortars). Zeolite is a minor constituent in the selected mortars.

The microstructure images of the control sample were examined before and after the high-temperatures at 400 °C and 600 °C. A-S-H (Al-Si-H₂O) gel matrix was formed before high-temperature and a small number of acicular crystals (anorthite and mullite), 1 µm in size, which is indicated by the marking (≈), were determined. At 400 °C, acicular crystals reached 5–10 µm dimensions for the control sample. It is monitored that the spherical fly ash particles start to be included in the amorphous matrix structure in the round-marked areas [19] It is observed that acicular crystals disappear, and spherical particle agglomerations of 0.5 µm form at 600 °C. A-S-H gel formation is observed in all three images, but it can be seen that A-S-H gels cannot significantly contribute to the densification, and the porosity increases with the

increase in temperature. It explains the strength decrease due to the temperature increase in the control sample (Fig. 9). Si/Al ratios of binders such as slag and fly ash used in the geopolymer mixture and high-temperature affect the bond structure and thus on the strength [65, 66]. When the effect of temperature change on the microstructures of the control sample is examined, it is viewed that there are few acicular crystals before high-temperature and at 400 °C. After being subjected to 400 °C, it can be seen that the acicular crystals dissolve into the amorphous phase and the gel structure containing round particles embedded in the matrix dominates the general microstructure.

Microstructures of the samples stone-cutting wastes were included in the highest and lowest ratios instead of fly ash were compared; It is thought that especially with the increase of RSW waste from 10% to 40%, the large and numerous acicular crystals (anorthite and mullite) that develop due to the high CaO and SiO_2 content in the RSW compared to the fly ash in its composition, interlock with each other and improved the strength of mortars. In Fig. 10, the regions where densification started with the fly ash and stone waste reaction and a small number of acicular crystals in the 10RSW sample are marked. When the microstructure of the sample containing 40% RSW waste is examined, the interlocking of many acicular crystals more significant than 1 µm with the increase in the number of additives are given with the marking (≈), and the regions where densification occurs with alkaline reaction with fly ash and stone wastes are given with round markings. In the sample containing 40% YSW, a small number of acicular crystals more prominent than 1 µm and a dense gel structure are observed (Fig. 11). The

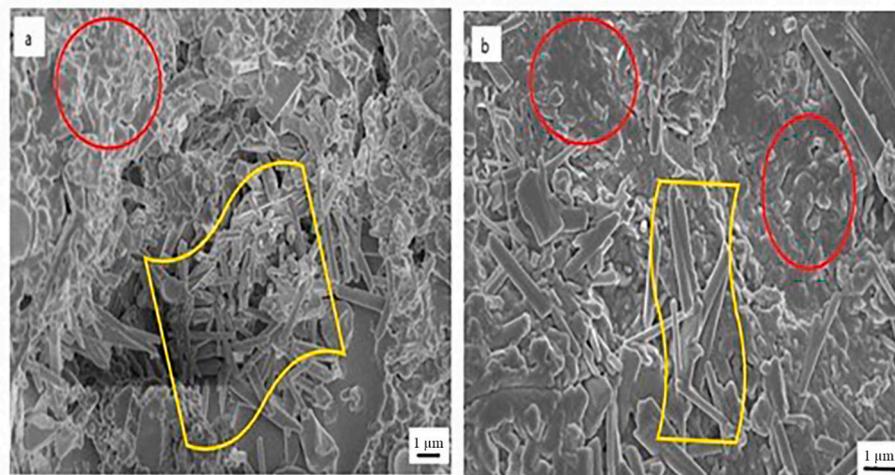


Fig. 11. SEM images of a)40RSW b)40YSW at 90 °C.

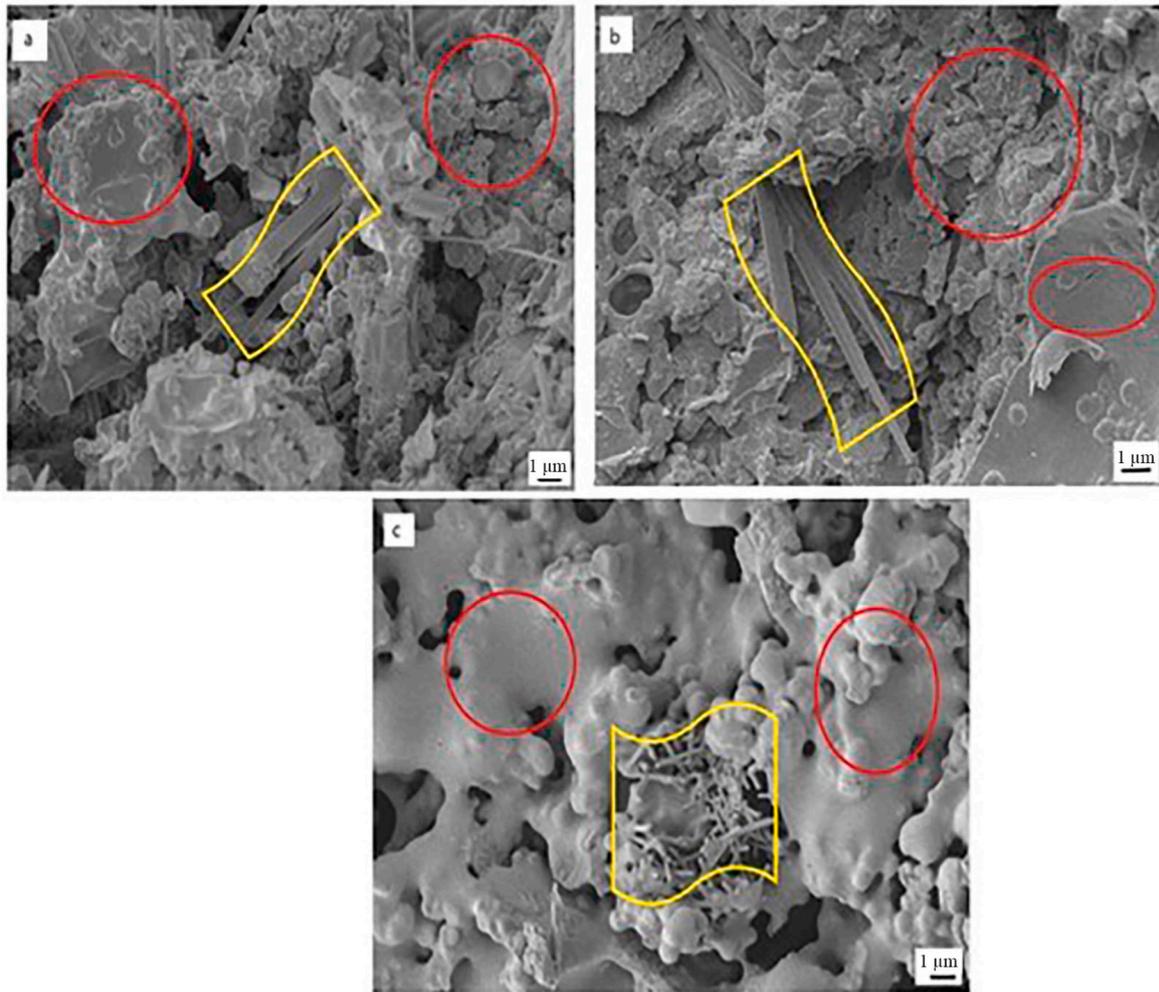


Fig. 12. SEM images of a)30 RSW-400 °C b)30 RSW-600 °C c) 30 RSW-800 °C.

literature states that the Si/Al atomic ratio is effective on the strength [67,68] In addition, with the increase in the Ca/Si ratio, the morphology of the calcium silicate hydrate gel, which is the hydration product, transforms from a thin sheet to a long fibrous structure and the compressive strength values increase in previous studies [69]. Moreover, the anorthite dissolution rate increases in geopolymerization process [70]. It explains why more acicular crystals are prominent in the samples containing RSW than in the samples containing YSW and the increase in strength due to the interlocking crystal structure.

In order to evaluate the effect of high-temperature, the microstructures of samples containing 30% YSW and RSW were examined. The size of the needled mullite affected by the temperature increased [19]. In particular, for 30 RSW, needle-like crystals (mullite) more prominent than 5 µm, indicated by the markings (≈), were detected in microstructures at 400 and 600 °C with increasing temperature (Fig. 12 a-b). It can be seen in Fig. 12 c that after 600 °C needle-like crystals begin to dissolve, shrink and become amorphous. When the microstructure of the 30YSW sample is examined, long acicular crystals can be seen at 400 °C. Notably, at 600 °C and 800 °C, the needle-like crystals disappear, and an amorphous structure containing round crystals in the markings with rounds develop (Fig. 13).

Previous studies showed that under alkaline, ambient conditions, Ca, Al, and Si ions may be reactive to the formation gel. Different from the literature, crystals' size and formation were evaluated at different temperatures and mixture ratios. Thus, the relationship between microstructure and mechanical properties was investigated in this study.

4. Conclusion

This study of stone-cutting wastes' influence on the physical, mechanical, and microstructural properties of geopolymers concludes that:

- Workability values of the produced geopolymer mortars decreased as the stone-cutting waste ratio increased. Unit weight and density values of the SW incorporated samples and the control sample were relatively close to each other.
- The samples containing RSW had higher flexural strength values than the samples containing YSW. The FS of the samples decreased with the increase in temperature, but the SW-based samples had higher flexural and compressive strength than the control sample.
- Compared to the control sample, the apparent porosity and water absorption values of the SW-based mortars were slightly higher.
- Quartz, anorthite, mullite, and zeolite crystals were detected. Anorthite and mullite peak intensities were increased with the increase in stone-cutting waste (40RSW and 40YSW).
- With an increased amount of SW (especially RSW) in the mortars, significant and many acicular crystals interlocked and increased the strength of the structure due to the high CaO and SiO₂ in its composition. After high-temperature application; It was observed that the large acicular crystals in the microstructure of the SW-based samples became amorphous with the increase in temperature.

In short, SW of the Cappadocia region was revealed to be a promising waste for geopolymer mortar, reducing the amount of fly ash in

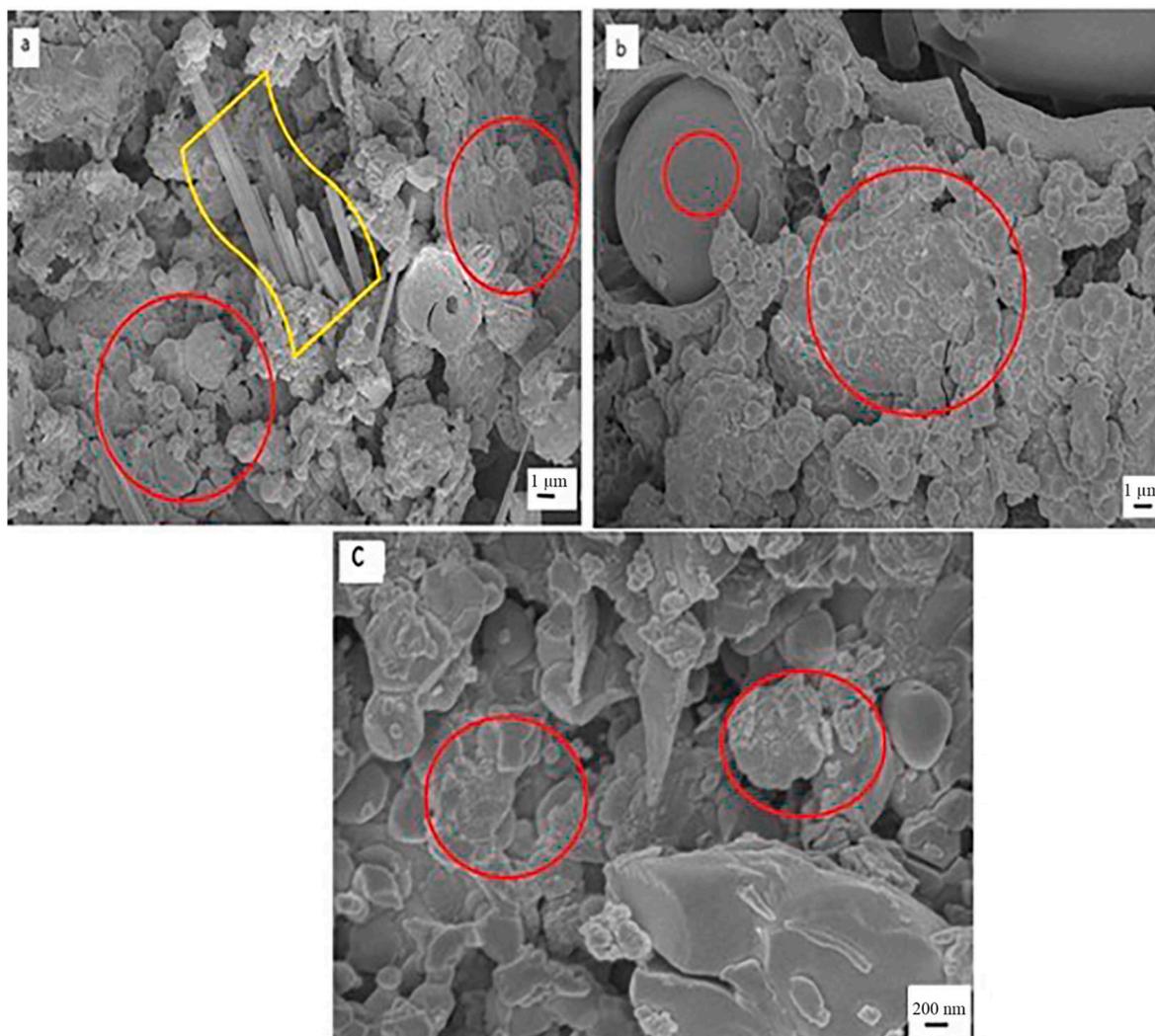


Fig. 13. SEM images of a) 30 YSW-400 °C, b) 30 YSW-600 °C, c) 30 YSW-800 °C.

compositions for high-temperature applications. Its Si-rich and Ca-rich-based chemical composition is the first step for more advanced studies and improved mechanical-microstructural properties of geopolymer mortars with other industrial wastes.

CRediT authorship contribution statement

Zahide Bayer Öztürk: Conceptualization, Methodology, Writing – original draft, Writing – review & editing, Writing – review & editing. **Tugrul Çam:** Sample preparation, Results and Investigation, Investigation, Writing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The data that has been used is confidential.

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