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Red-mud additive geopolymer composites with eco-friendly aggregates

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ABSTRACT

Geopolymers have an increasing importance and a crucial task in engineering, especially structural engineering. Geopolymers are sustainable composites with low carbon emissions rather than cement composites. It leads to be an alternative to cement composites and the structure of geopolymer composites can allow to utilize various industrial and non-industrial wastes and by-products. In this point, there is an unclarity of waste material contributions to the geopolymeric composites and this issue is worth to study and an area of ongoing research. In this paper, it is aimed to produce red mud additive sustainable geopolymer composites with various eco-friendly fillers (brick powder, waste marble powder, glass powder, ceramic powder, and rice husk ash). 28 mixtures were designed, many specimens produced, and tested. In the first step of the experimental stage, the physical, the mechanical and the durability properties were determined and evaluated. At second, the best geopolymer composites were determined with a multi-criteria decision-making method (MCDM). 22 geopolymer parameters were employed in MCDM. As a results, 25% marble dust, 25% brick powder, 75% ceramic powder, 50% rice husk ash and 50% glass powder were the optimum ratios to obtain maximum compressive strength in terms of waste types and the highest compressive strength is obtained for 25% brick powder among the results. The best geopolymer composite was determined as GWGP50 with 50% glass powder + 50% recycled aggregate in a holistic manner with TOPSIS. Results showed that geopolymer GWGP50 had aspects that were open to development.

1. Introduction

Geopolymer composites with up to 100% recyclable materials keeps a permanent place in researcher's mind [1,2]. Sustainable materials (geopolymer is one of them) have an increasing importance for structural engineering. The demand of the sustainable materials comes from the sustainability trend to achieve and use low carbon materials. Waste materials and by-products have emerged as the most popular materials tested in the geopolymer mixture. Developing a sustainable material with low carbon emission alternative to cement is a crucial task in structural engineering. These types of materials compatible with other blended ingredients of geopolymer composite is expected to provide enough carrying capacity of geopolymer composite. In the world, 150 Mt waste of aluminum production facilities is occurred and stocked [3] and is named as red mud (RM) being a by-product from the processing of bauxite ore. "Red" meaning of RM comes from the color of chemical content of RM (i.e., iron oxide). The chemical content of RM may vary and may include heavy metals. In addition, the physical properties of RM may vary, and thin material structure make easy to disperse in the atmosphere. Thus, disposal and storage of RM requires to take security measures/precautions, and this threatens groundwaters, organisms, and soils [3,4]. Disposal of RM in cement production is limited by high alkalinity of RM.

High alkalinity required process (i.e., in geopolymer production as a precursor) is suitable for RM with other reactive materials (metakaolin (MK), ground granulated blast furnace slag (GGBFS), and fly ash (FA), although RM is a material with low reactivity [3,5]. Geopolymer with RM is a way to reduce the amount of RM in the world [5] and MK+RM can be utilized as a good binary material. MK from calcining kaolin with heat process ranging 600–900 °C can reduce the worse impacts (i.e., carbon emissions, dust) of cement and its production and can be an alternative to cement to construct net-zero buildings/cities. Sustainable carbon neutrality construction in the future requires the design change not only to material mixtures, but also to building system and urban planning [6]. Thus, the needs to compose the environmentally friendly designs is related to the material design. The sustainable material design

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gain importance while the topic is worth to study.

Accordingly, geopolymer and its applications in the engineering area are broadened by the new investigations. The studies considered the geopolymers with RILEM sand (i.e., [7–9]) and other waste materials (i.e., [10–14]) and geopolymers included waste fillers came ahead in terms of sustainability although the waste materials may have worse properties compared to natural ones. Replacement of natural resources with wastes to complete the life cycle of materials bring the investigators to design sustainable materials for low CO_2 emission/net-zero buildings/cities nowadays [15,16] and geopolymer is one of the sustainable materials. In this point, composing the structure and components of geopolymer mostly with materials (i.e., construction and demolition waste) that has reached the end of its life change the human minds into recycling. In addition, according to the goal of the European Green Deal, EU's Horizon 2030 will welcome 100 smart and climate-neutral cities [17].

There are a little number of studies to investigate the consideration of waste concrete (WC) as an aggregate/component of fibrous and non-fibrous geopolymer. A systematical investigation on types and properties of aggregates is meaningful and the researchers formed a composite of MK based-geopolymer [18]. In the investigation, the findings showed that the impact of recycled aggregate (RA) sourced from WC decreased the mechanical performance of composite and it was tried to enhance the performance parameter by adding fibers. RA has porous structure due to attached old mortar, and it leads a relatively lower physical and mechanical properties compared to natural aggregate [19,20]. In addition, RA with low properties causes low physical and mechanical properties of concrete when RA is included [21,22]. Despite the observation of disadvantages of RA, the aim of contribution of RA in sustainability and for designing a sustainable material for low carbon footprint buildings/cities was undeniable. It has low travel distance to achieve the material with low release of dust into atmosphere contributed to this topic. In addition to commonly known wastes (i.e., FA, MK, GGBS, RM), some specific wastes produced in various industrial branches (i.e., ceramic waste dust (CWD), brick waste dust (BWD), marble dust (MD), wood dust (WD) and glass dust (GD)). The wastes have a possible role being a component of sustainable materials for the buildings/cities and widely available on the world.

The wastes given above have been attracted the attention of researchers and the use of wastes was found suitable for mortars as an aggregate/filler. The suitability of the wastes as an aggregate/filler was considered in relation to chemical components of the wastes (i.e., BWD has an alumina-silicate chemical structure [3]) and large amount of the wastes around the world. It may accelerate the use process into a material to recycle them. Combining/separately using the powder of WC with BWD was an example of the topic and tried by Tang et al. [23]. The findings showed that despite high utilization ratios of the waste materials with mechanical performance decrease disadvantages, the topic is worth to study and there are a few studies on geopolymer with the mentioned wastes. In this point, the release of high amount of RM in the world reveals disposability in areas where it can be used at a high rate, and especially the construction industry in terms of sustainability is a good example and has high potential.

Despite there are some technical difficulties of RM for cement production, RM can be an alternative to cement. The chemical content of RM is a key factor for being a potential binder and RM has been interested in inorganic polymers, such as geopolymers recently [12,24]. Although alkalinity of RM is itself naturally and RM has Al, Si, Fe, and Ca as main products of geopolymerization, activation reactivity should be improved by high reactive components (i.e., FA, MK, and GGBS). Because it was found that compressive strength of geopolymer included RM as a binder was 2.5 MPa [25] and, for instance, adding MK to RM-based geopolymer caused a high strength improvement (from 10.6 MPa to 51 MPa, see ref. [26]).

In this point, it was concluded that low range strength development was owing by iron/resulting geopolymer structure and iron prevented MK from dissolving in the medium. Huge amount of MD and WC produced by various activities spirits the researchers to utilize as an aggregate instead of RILEM sand to compose a wholly recycled building material. As well-known, dust wastes can be flow into the atmosphere by winds and threatens the environment. To break/slow down the mentioned impacts, substitution of aggregates with MD including cement was investigate by Khyaliya et al. [27] and assessed the strength and durability performance of the mortars. It was reported that performance improvement due to the MD was at enough level. Besides, and fly ash-based alkali activated concrete with marble aggregate was investigated by Lim et al. and marble aggregate addition from 0% to 50% improved the mechanical performance and low penetration depth was occurred [28].

Rice husk ash (RHA) burning and then milling to obtain an alternative/supplementary binding material has been tried and application area is wide. The findings showed that replacement of cement with 10% RHA caused maximum strength and the burning/milling process has an impact on the quality of RHA [24,29–31].

Three thousand six hundred years ago, glass production has been started and glass is usually consumed as a material of window and bottles [32]. The consumption of glass as non-returnable increases the amount of waste glass around the world. Issues of waste glass occurs worldwide and the difficulties in fragmentation of waste glass into low size particles is one of them. Disposal of waste glass as an aggregate replacing natural gravels in mortar and concrete comes into mind and can be geopolymeric material after milling process [33]. Accordingly, the researchers have been inspired from the findings on the geopolymeric feature of powder of waste glass (WGP) and used WGP in the geopolymer production. The reduction in open porosity of geopolymer based on MK with WGP was observed and reported in the literature [33–37]. This advance showed that reduction in the open porosity may cause a durable geopolymer.

Despite the geopolymer composites included waste materials with durable behavior, ceramic waste powder (CWP)-based geopolymer investigations were rarely studied. CWP of the ceramic industry can be a by-product for construction industry and is the significant waste. CWP occurs while the polishing coating processes of the ceramic industry and refilling excavation pits is a common way to disposal CWP. However, disposal of ceramic waste in landfill may not be a solution due to taking a large space and disposal mechanisms should be implemented finding new ways to save the environment. In this view CWP is considered as a supplementary material for concrete due to pozzolanic properties and strength/durability gain compared to control was reported [38,39]. Here, many papers on binder replacement in the mixtures included CWP in the literature, but limited studies on investigation of CWP in alkali activated materials/geopolymer as a filler or aggregate [3,24]. The employment of CWP in ongoing studies to investigate the reuse impact on properties of geopolymer composites and to research the recycle mechanism is thought as a crucial topic.

The waste brick powder (WBP) is another essential topic in the literature and exploration of the utilization of WBP as a supplementary cementitious material inspires the researchers. In this point, the calcination process of the brick with an optimum calcination temperature range is the key factor to shape the crystalline structure of the medium into amorphous structure (silica + alumina) [24,40]. WBP is used after the life end of the brick and this will may help the environment protection while we go on the net zero/low carbon cities. However, the utilization of WBP is rarely investigated and has a great potential to mentioned sustainable net zero material topic. It is aimed in the goal of the European Green Deal that EU's Horizon 2030 will welcome 100 smart and climate-neutral cities.

In this comprehensive study, a sustainable composite design with geopolymeric binding medium and waste fillers/aggregates aim was preferred to contribute the mentioned sustainable low carbon material topic of EU's Horizon 2030. Thus, many wastes such as waste glass powder, waste brick powder, waste marble powder, ceramic waste

powder, and rice husk ash were used with recycled aggregate. Red metakaolin-mud-based geopolymeric composites were produced. Due to the utilization of many types of components in the composites and their impact on the resulting properties of the sustainable geopolymer, a holistic evaluation strategy was adopted in this comprehensive study, and a decision support method was used to find the best geopolymer in consideration of all findings.

2. Materials and methods

2.1. Materials

Geopolymer had two phases in this study and the phases had duties such as binding and filling. The binding phase had metakaolin and red mud binders. The producer details of metakaolin and red mud were Kaolin EAD (Turkey) and Eti Aluminum plant in Seydisehir (Konya/Turkey), respectively. The additional binder with a low use ratio was a ground granulated blast furnace slag (GGBS) obtained from Bolu Cement Factory. Hence, the total number of binders considered in this paper was three and the chemical details of the binders were presented in Table 1.

12 Molar sodium hydroxide and sodium silicate were employed to activate the components of the geopolymer composite and were product of AS Chemistry Company (Turkey). The details of the chemical components of geopolymer is given in Table 2. To overcome the workability problem while the production of fresh geopolymer, polycarboxylate ether superplasticizer (SP) with high-water reducing feature was used and supplied from BASF. In this point some researches were enlighten the use of SP and the impact of SP was in relation with the considered activator solution structure [24]. In the mixture of geopolymer composite various wastes were employed and the ratio of the activator to binder changed (decreased or increased). This led a workability problem in the fresh state of the geopolymer composite and SP was added to the trial samples to observe the fresh behaviour. Then, 4.4% SP addition to the mixtures with 4.4% additional extra water solved the trouble and ideal consistency was easily achieved despite a decrease in the ratio of the activator to binder. Here, although sustainable approach was followed using waste materials in the composite, economy has also been considered using optimized and enough level expensive activator material. Filling ability of the waste materials was the second economy considered in the paper: High filler content, low cost, and high economy. Accordingly, the waste glass powder, waste brick powder, waste marble powder, ceramic waste powder, and rice husk ash contributed the topic mentioned above and the properties of them were presented in Table 3. Industries on glass, powder, waste brick powder, waste marble powder, ceramic waste powder, and rice husk ash in Turkey work globally and annually produce huge number of not only products but also by-products and wastes. In addition, the wastes are not in amorphous form but in crystal form and hence these cannot be used in resulting products in their industry. Thus, according to sustainable mind and environmental concerns, the wastes mentioned above were employed to investigate.

Table 1
Composition details of the binders (red mud, metakaolin and ground granulated blast furnace slag).

Composition, %	Red mud	Metakaolin	Ground granulated blast furnace slag
SiO_2	16.2	56.1	40.5
Al_2O_3	22.9	42.25	12.83
Fe_2O_3	34.5	0.85	1.10
CaO	1.8	0.19	35.58
MgO	-	0.16	5.87
SO_3	-	-	0.18
Na_2O	8.7	0.24	0.79
Cl^-	-	-	0.0143
Loss of ignition	-	1.11	0.03

Table 2
Sodium hydroxide's and sodium silicate's properties.

Material	Features
Sodium hydroxide	Na_2CO_3 (g/kg) $\leq$ 4; NaOH (g/kg) $\geq$ 990; $SO_4 \leq 0.01$; Fe $\leq$ 0.002; $Cl^- \leq 0.01$; Al $\leq$ 0.002
Sodium silicate	Na_2O (%): 9.68; SiO_2 (%): 26.12; Density (20 °C) (g/ml): 1.367; Fe (%) < 0.005; Heavy metals value (pb) (%) < 0.005

Table 3
Properties of brick powder, ceramic powder, marble powder, rice husk ash, glass powder, recycled aggregate.

Oxides (% by weight)	Brick powder	Ceramic powder	Marble powder	Rice husk ash	Glass powder	Recycled aggregate
SiO_2	55.5	63.43	1.12	92.33	69.42	60.56
Al_2O_3	17.1	20.22	7.30	18.0	1.09	52.0
Fe_2O_3	5.8	6.40	0.05	0.17	0.48	5.82
CaO	10.5	0.66	83.22	0.02	8.27	12.01
MgO	4.5	3.13	–	–	4.25	1.83
SO_3	1.0	–	0.56	–	–	–
Na_2O	1.9	0.98	–	0.29	12.31	2.69
K_2O	2.8	3.96	–	–	–	1.30
TiO_2	0.8	0.77	–	–	–	0.75

2.2. Mixing procedure and curing specimens

Binding medium of the geopolymer composite was consisted of two main components such as RM+MK and their use ratio in the mixture was constant and equal to each other. Only a little rate GGBS was employed aiming to obtain a little setting time. The preparation methodology for geopolymeric materials was deeply investigated in the previous papers [41,42] and the findings were followed by the authors properly. Accordingly, 24 hours waited 12 Molar NaOH was cooled in the laboratory under standard laboratory conditions. On the one hand, the sodium solution consists of sodium silicate + sodium hydroxide was prepared and waited 24 hours before the mixing day. On the second hand, the binders (RM and MK) were dry mixed together until a uniform composition was obtained. Then, at the third stage, the sodium solution and dry form uniformly mixed binders were mixed. In this point, 13% GGBS was added to all geopolymer composites. For constant consistency, 4.4% additional water and 4.4% plasticizer were added to the mixture, and it was by weight of the binder. Despite varying raw materials in the mix, the constant workability was ensured with varying sodium silicate and sodium hydroxide solutions while fresh geopolymer production process. Thus, 4.4% plasticizer + 4.4% additional water was added and required workability can be achieved. Here, the Then, the last stage (4th stage) the mortar was composed with the filler components (it was aggregates) and binder. The main filler was chosen as RA. The replacement of RA with other aggregates such as marble powder, brick powder, glass powder, rice husk ash and ceramic powder is applied by 0–25–50–75%. Thus, it is aimed to investigate the impacts of the wastes on the geopolymer composite.

16 geopolymer composites were produced and ingredients of the geopolymers were given in Table 4. After a careful casting process for fresh geopolymer composites, a proper settling by vibration table was applied. First 24 hours after casting process, the specimens were cured under laboratory conditions at 50% relative humidity @ 23∓2 °C. Beginning from 24 hours to 48 hours, the samples were heat cured 60 °C for MK, RM and GGBS based geopolymer [43,44]. However, some researchers tried different temperatures and molarity to obtain required strength grade for different materials (i.e., [45,46]). Then, the samples were cured under laboratory conditions at 50% relative humidity @ 23∓2 °C till the testing days (28 days).

Table 4
Ingredients of the geopolymer composites (g).

Notation	MK	RM	GGBS	NaOH	Na_2SiO_3	Water	SP	RA	MD	WGP	CWP	WBP	RHA
GC	225	225	60	165	330	20	20	1125	0	0	0	0	0
GMD25	225	225	60	180	360	20	20	853.75	281.25	0	0	0	0
GMD50	225	225	60	180	360	20	20	562.5	562.5	0	0	0	0
GMD75	225	225	60	225	450	20	20	281.25	853.75	0	0	0	0
GWGP25	225	225	60	180	360	20	20	853.75	0	281.25	0	0	0
GWGP50	225	225	60	180	360	20	20	562.5	0	562.5	0	0	0
GWGP75	225	225	60	195	390	20	20	281.25	0	853.75	0	0	0
GCWP25	225	225	60	225	450	20	20	853.75	0	0	281.25	0	0
GCWP50	225	225	60	225	450	20	20	562.5	0	0	562.5	0	0
GCWP75	225	225	60	240	480	20	20	281.25	0	0	853.75	0	0
GWBP25	225	225	60	195	390	20	20	853.75	0	0	0	281.25	0
GWBP50	225	225	60	210	420	20	20	562.5	0	0	0	562.5	0
GWBP75	225	225	60	225	450	20	20	281.25	0	0	0	853.75	0
GRHA25	225	225	60	225	450	20	20	853.75	0	0	0	0	281.25
GRHA50	225	225	60	240	480	20	20	562.5	0	0	0	0	562.5
GRHA75	225	225	60	315	630	20	20	281.25	0	0	0	0	853.75

2.3. Testing

50 mm cubic, 40×40×160 mm prismatic and 100×200 mm cylindrical geometries were employed to measure the physical and mechanical performance of the geopolymer composites. Unit weight and water absorption properties of the specimens were determined. ASTM C109, ASTM C348 and ASTM C469 were used to perform compression tests, bending tests, and splitting tensile strength tests. In addition, ultrasound pulse velocity (UPV) values of the geopolymer composites were properly determined on the specimens. Besides, unit weight, water absorption, and void rate were the other investigated parameters in the tests. Each test results was the mean of three specimen test results. Also, the durability performance of the high compressive strength geopolymer composites regarding the used waste powders was obtained. The tests of the high temperature (300–600–800 °C), the freeze-thaw (130 cycles), the sulphate attack (sodium sulphate and magnesium sulphate) were the employed. The specimens' exposure to harmful environments were subjected to compression tests to observe the remaining strength.

The high temperature tests with various temperatures up to 800 °C were applied to the oven dried geopolymer composites. An ash furnace was used in the tests with a temperature increment of 5 °C/min. The geopolymer composites were subjected to cold/hot cycles. A cycle has freeze impact (-18°C) with 12 hours and thaw impact (+4 °C) with 12 hours to the specimens. In total, 130 cycles were applied. The impacts of sodium and magnesium sulfates were tested for 90 days and % 10 concentration for the sulphates was considered.

2.4. Evaluation of test results

In this section, the best geopolymer composite was aimed to find among the bests by using a decision support method (TOPSIS) (Eqs.1–11). TOPSIS is widely used in many areas where a decision should be made to find the best choice [47–51]. Accordingly, in this paper, many specimens were produced and tested to determine the behavior under various impacts and a need is occurred for the selection of the best geopolymer composite but in a holistic manner. Thus, TOPSIS was used to evaluate the results. Here, weights of the criteria in Eq.4 was used the same for all.

Decision support systems such as TOPSIS can select the most appropriate/best option among multiple options with multiple criteria with holistic approaches. At this point, the user determines the input data based on the data the options have. In this study, physical, mechanical and permanence parameters along with the chemical content of the geopolymer were taken into consideration. The criterions of the alternatives (GC, GMD25, GWGP50, GCWP75, GWBP25, and GRHA50) were set as SiO_2, Al_2O3, Fe_2O3, CaO, MgO, SO3, Na_2O, K2O, TiO2, H2O, density, water absorption, UPV, compressive strength, splitting tensile strength, bending strength, compressive strength after high temperature apply, weight loss after freeze-thaw, sodium, and magnesium sulphate attacks. Having more parameters of the criteria may indicate that a holistic approach will yield better results. Because it is generally known both in statistics and in real life, decisions taken with a limited data set can often result in wrong or inadequate results.

On the other hand, in decision support systems such as TOPSIS, the weights of the criteria can be taken into account. In determining the weights, opinions of experts in the field, statistical surveys, compilation from similar studies in the literature and graphing from literature data are used [47,51]. There are reasons such as geopolymer studies are very new, different materials are being tested, and there may not be enough experts in the field for geopolymers containing different materials. For this reason, the weight coefficients of the parameters of the criteria could not be studied in detail and the weights of all parameters were considered equal. The authors plan to determine the weights by considering this detail in their future studies.

Decision matrix (Eq.1):

$$A_{ij} = \begin{bmatrix} a_{11} & \cdots & a_{1j} \\ \vdots & \ddots & \vdots \\ a_{i1} & \cdots & a_{ij} \end{bmatrix} \tag{1}$$

Normalization of decision matrix elements (a_{ij}) and normalized decision matrix (Eqs.2–3).

$$N_{ij} = \frac{a_{ij}}{\sqrt{\sum_{i=1}^{n} a_{ij}^2}} (i = 1, 2, 3, \ldots, m \quad ve \quad j = 1, 2, 3, \ldots, n) \tag{2}$$

$$N_{ij} = \begin{bmatrix} n_{11} & \cdots & n_{1j} \\ \vdots & \ddots & \vdots \\ n_{i1} & \cdots & n_{ij} \end{bmatrix} \tag{3}$$

Weighted normalized matrix with weighting coefficients (Eq. 4).

$$V_{ij} = \begin{bmatrix} w_1 \times n_{11} & \cdots & w_n \times n_{1j} \\ \vdots & \ddots & \vdots \\ w_1 \times n_{i1} & \cdots & w_n \times n_{ij} \end{bmatrix} \rightarrow V_{ij} = \begin{bmatrix} v_{11} & \cdots & v_{1j} \\ \vdots & \ddots & \vdots \\ v_{i1} & \cdots & v_{ij} \end{bmatrix} \tag{4}$$

Positive and negative ideal solutions given in (Eqs.5–8).
Positive ideal solution values:

$$A^+ = \left\{ (\max_i v_{ij}) \right\} \tag{5}$$

$$A^+ = \{ v_1^+, v_2^+, \ldots, v_n^+ \} \tag{6}$$

Negative ideal solution values:

$$A^- = \left\{ (\min_i v_{ij}) \right\} \tag{7}$$

$$A^- = \{ v_1^-, v_2^-, \ldots, v_n^- \} \tag{8}$$

Distance between alternatives and negative/positive ideal solutions are given in (Eqs.9–10).

Distance to positive ideal solution:

$$S_i^+ = \sqrt{\sum_{j=1}^{n} \left(v_{ij} - v_j^+\right)^2} \quad (9)$$

Distance to negative ideal solution:

$$S_i^- = \sqrt{\sum_{j=1}^{n} \left(v_{ij} - v_j^-\right)^2} \quad (10)$$

Calculation for relative proximity to the ideal solution (Eq.11):

$$G_i^* = \frac{S_i^-}{S_i^- + S_i^+} \quad (11)$$

3. Results and discussions

In the paper, a sustainable geopolymer composite design was performed to contribute the low carbon material topic of EU's Horizon 2030. Accordingly, the possible use of many wastes (waste glass powder, waste brick powder, waste marble powder, ceramic waste powder, and rice husk ash) with recycled aggregate in red metakaolin-mud-based geopolymer composites were investigated. Also, a holistic manner was followed in the evaluation of the test results part by using a decision support method (TOPSIS) and the best geopolymer composite was chosen, thus the optimum proportions was found/observed from the best geopolymeric mixture. The evaluations were in the sections given below.

3.1. Physical test results

Physical properties of geopolymer composites such as UPV, water absorption and unit weight were determined on 50 mm cubic specimens at 28-days as given in Table 5. When the results were examined, it is obviously seemed that the unit weights were ranged between 2.24 and 1.90 g/cm^3. The use of MD in geopolymer composites at 25–50–75%, the unit weight changed as 2.20–2.21–2.10 g/cm^3 while those of water absorption were determined as 16.46–17.88–18.48%. It was though that increasing MD use in the mixtures may cause settlement problem forming pores in the medium with an increase of open porosity. The values of UPV supported the results. In addition, the use of RHA in geopolymer composite had the same effect on the physical properties of those of MD. On the other hand, the use of WGP in the mixture at 25–50–75% changed the unit weight as 2.14–2.17–2.13 g/cm^3 and those of water absorption was decreased from 16.39% to 14.73% when the use of proportion was increased from 25% to 75% in the composite.

Table 5
Physical properties of geopolymer composites.

Notation	Unit weight, g/cm^3	Water absorption, %	UPV, km/s
GC	2.17	16.73	3472
GMD25	2.20	16.46	3987
GMD50	2.21	17.88	3900
GMD75	2.10	18.48	3432
GWGP25	2.14	16.39	3265
GWGP50	2.17	16.30	3519
GWGP75	2.13	14.73	2471
GCWP25	2.24	18.82	3599
GCWP50	2.24	18.99	3686
GCWP75	2.20	18.78	3880
GWBP25	2.18	16.79	3786
GWBP50	2.15	18.13	3526
GWBP75	2.24	18.19	3713
GRHA25	2.08	18.65	3372
GRHA50	1.98	19.46	3386
GRHA75	1.90	20.66	3025

However, the properties of CWP were changed due to CWP at any use proportion. UPV results were supported the results.

3.2. Mechanical test results

The mechanical test results were given in Table 6. The values of compression test results were the mean of the results of three specimens.

3.2.1. Compressive strength

Brittle and semi-brittle like materials (i.e., concrete, cement paste and geopolymer) has higher compressive strength with relatively low tensile strength. In engineering, it is aimed to use brittle and semi-brittle like materials in the structure and related locations considering the mentioned parameters [52]. Hence, the high strength material like geopolymer is desired and designed properly to obtain the required compressive strength level [46,53]. In this research paper, it is desired to achieve high compressive strength with high waste ratio in terms of sustainable approach.

The compressive strengths of geopolymer composites were increased with the substitution of RA with the wastes used (except for GMD75). The compressive strength of GMD25, GMD50 and GMD75 were found as 28.01, 25.04 and 21.93 MPa, respectively, while the control (GC) had 22.53 MPa. Increasing MD content in the mixture up to 50% increased the compressive strength and beyond decreased/marginally changed the strength. The strengths of GWBP25, GWBP50 and GWBP75 were increased and found as 39.20, 38.27 and 36.96 MPa respectively compared to GC and the increase was reduced beyond 50% use of WGP in geopolymer matrix. In this point, the use of MD and WBP increased the compressive strengths of geopolymer composites and however, the strength increase ratio of the WBP was higher than those of MD. In terms of chemical properties, WBP had 55.5% SiO_2 and 17.1% Al_2O_3 while MD had 83.22% CaO (Table 3). It can be said that the N-A-S-H formation may a role in the geopolymer strength gain and C-A-S-H may have a relatively low impact on the topic [45,54–56].

On the other hand, the use of CWP, RHA and WGP caused higher compressive strength than that of the control (GC). However, despite geopolymer composite included CWP with higher compressive strength than GC, CWP showed dissimilar impact than other wastes and increasing CWP content in the matrix increased the compressive strength of geopolymer. In this point the use of RHA and WGP up to 50% were increased the compressive strengths and beyond the strength values decreased.

In consideration of the wastes used in the geopolymer matrix and their impacts on the compressive strength, the optimum use values of them were able to be listed as 25% MD, 25% WBP, 50% CWP, 50% RHA, and 50 WGP. The maximum compressive strength value among the results was obtained for GWBP25 as 39.2 MPa with 25% WBP+75% RA content.

3.2.2. Splitting tensile strength

Tensile strength is a parameter related to cracking of brittle and semi-brittle materials and the materials collapse due to tensile stress occurred under compression or shear loading. Accordingly, the undesired situation may cause durability concerns after cracking [51,57] and it is commonly known that the harmful environment impacts enter open pores of the material through the center destructing the material integrity. Usually, the paste is usually hosting and facing the mentioned effects due to its porous structure [58–66] and also sometimes porous aggregates (i.e., RA [67,68] and light-weight aggregate [69,70]) Hence, the lower strength is undesired for brittle/semi-brittle materials and the higher splitting tensile strength (STS) of geopolymer composite was aimed while the design of composition of geopolymer composition. Despite unavoidability of the low tensile strength of brittle/semi-brittle materials, the mentioned parameter focused on and intended to be developed was to increase the "splitting tensile strength" of the geopolymer composites. After a series of STS tests, the behavior of the

geopolymer composites were determined (Tables 6–7) and the investigated parameter was tried to be developed with various waste material content used.

In addition, in this comprehensive paper the waste increased the STS was investigated. Because the considered components/wastes can increase/decrease the brittleness of the designed geopolymers and, as well-known geopolymer is a brittle/semi-brittle material, the brittleness problem of the geopolymer should be solved or brittleness index should not be increased by the waste to prevent the durability problems. As mentioned above, the harmful environment impacts enter open pores of the material. Also, the more brittleness the more cracks.

Three approaches were followed in the literature to evaluate the tensile strength [67,71–73]. The first technique uses the ratio of STS to compressive strength of the material. The second uses the value of STS only and depends on the comparison of the results with control. The third one is the investigation of the relationship of material parameters such as STS-compressive strength by using a regression analysis determining a correlation coefficient (R or R^2). All of them have meaningful and fruitful steps and show the experimental gains in terms of components used and their impacts. The second one is the commonly known approach in the literature and the third one follows the second one in terms of popularity. In this paper the first and second approached were employed to evaluate and investigate the STS behavior of geopolymer composites included various wastes (MD, WBP, CWP, RHA and WGP) at different proportions.

Accordingly, at first only STS of geopolymer composites were evaluated and compared the results to achieve the best scenario for the use of wastes. The STS of control (GC) determined as 1.22 MPa and those of geopolymer composites modified with MD, WBP, CWP, RHA and WGP increased and found higher than GC. For instance, STS of GC, GCWP25, GCWP50 and GCWP75 were obtained as 1.22, 1.61, 2.45, and 1.86 MPa, respectively. Although the values of STS of geopolymers were higher than GC, the increase impact of wastes (MD, WBP, CWP, RHA and WGP) replaced by RA were not the same and the optimum use ratio may change with both waste replacement ratio and waste type. Accordingly, the maximum values of STS of geopolymers in terms of replaced wastes (MD, WBP, CWP, RHA and WGP) were determined as GMD25 with 2.37 MPa, GWBP25 with 2.05 MPa, GCWP50 with 2.45 MPa, GRHA75 with 2.17 MPa, GWGP25 with 2.89 MPa, respectively.

In addition, it can be noted that when CWP increased from 50% to 75%, the compressive strength was increased, however, the STS decreased. Whereas when RHA increased from 50% to 75%, the compressive strength decreased, however, split tensile strength increased. Accordingly, the findings showed that CWP and RHA increased and decreased the brittleness of the geopolymer, respectively. It can be said that different NaOH and Na_2SiO_3 contents of geopolymer mixtures may also affected the brittleness.

Table 6
Mechanical properties of geopolymer composites.

Notation	Compressive strength, MPa	Splitting tensile strength, MPa	Bending strength, MPa
GC	22.53	1.22	7.3
GMD25	28.01	2.37	6.66
GMD50	25.04	1.25	5.56
GMD75	21.93	1.82	4.6
GWBP25	39.2	2.05	7.74
GWBP50	38.27	1.9	6.98
GWBP75	36.96	1.76	4.6
GCWP25	32.37	1.61	5.32
GCWP50	34.67	2.45	4.45
GCWP75	37.27	1.86	3.9
GRHA25	27.23	1.83	6.25
GRHA50	33.84	1.49	4.97
GRHA75	26.29	2.17	7.68
GWGP25	32.92	2.89	6.92
GWGP50	33.29	1.88	6.92
GWGP75	24.93	2.18	5.38

Table 7
The ratio of compression to splitting tensile strength ratios of geopolymer composites.

Notation	Substitution ratio, %	Comp. Str./ Ten. Spl. Str. Ratio	Upper ratio	Lower ratio	Range between the upper and lower ratios
GC	0	0.054	-	-	-
GMD25	25	0.085	0.085	0.050	0.035
GMD50	50	0.050			
GMD75	75	0.083			
GWBP25	25	0.052	0.052	0.048	0.004
GWBP50	50	0.050			
GWBP75	75	0.048			
GCWP25	25	0.050	0.071	0.050	0.021
GCWP50	50	0.071			
GCWP75	75	0.050			
GRHA25	25	0.067	0.083	0.044	0.039
GRHA50	50	0.044			
GRHA75	75	0.083			
GWGP25	25	0.088	0.088	0.056	0.032
GWGP50	50	0.056			
GWGP75	75	0.087			

Table 7 presented the effect of waste contents on the ratio of STS to compressive strength. The findings from Table 7 demonstrated that the ratio of the geopolymer composites GMD25, and GMD75 (i.e., 0.085 and 0.083) were higher than GC (0.054) except for GMD50 (0.050) and the use of WGP in geopolymer composite caused the similar results (those of GWGP25, GWGP50 and GWGP75 were 0.088, 0.056 and 0.087). Although MD and WGP had an increase effect on the ratio, the fifty-fifty use of RA and MD, RHA or WGP had a marginal impact on the results and also 50% RHA use in geopolymer composite reduced the ratio down to 0.044. Opposite of this, the use of CWP had a low impact on the ratio, the use of 50% CWP increased the ratio up to 0.70. Besides, the use of WBP had a marginal impact on the ratio and the ratios were found lower (i.e., 0.052, 0.050 and 0.048 for GWBP25, GWBP50 and GWBP75, respectively) than the control (GC with 0.054). In addition, the lower and the upper bonds of the ratios for each waste (MD, WBP, CWP, RHA and WGP) were obtained as given in Table 7. According to the range between the upper and the lower ratios of geopolymer composites in Table 7, WBP and RHA change in the mixtures had a minimum and maximum effect on the ratio. Accordingly, different waste use in the geopolymer composite fluctuate the ratios and the use ratio of the waste also influenced the results as much as the use of waste. It was reported by Dilbas that the lower ratio of compressive strength to splitting tensile strength the higher brittleness [74].

3.3. Durability test results

In this comprehensive study, the durability tests were applied to the geopolymer composites with highest compressive strength values. Accordingly, for each group in terms of waste types, 6 geopolymer composites GC (as a control), GMD25, GWGP50, GCWP75, GWBP25, GRHA50 were selected. The results of the durability tests (high temperature resistance, freeze-thaw resistance, and sulphate attack) were evaluated in the following.

3.3.1. High temperature

The specimens of selected best geopolymer composite series were subjected to 300–600–800 °C temperature and the mechanical results of the heated specimens at the room temperature were given in Table 8. It was observed that the compressive strength of the control had a gradual decrease from 27.76 MPa to 4.07 MPa, and GMD25 and GWBP25 showed a similar decrease behavior. However, it was interesting that GWGP50 and GCWP75 had higher fire resistance. Although a decrease in compressive strength was observed for both of GWGP50 and GCWP75 due to high temperature effect, the remaining strengths were still

Table 8
Mechanical results of specimens after 300–600-800 °C heating.

	Compressive strength, MPa			Bending strength, MPa		
Notation	300 °C	600°C	800°C	300°C	600°C	800°C
GC	27.76	21.31	4.07	1.47	1.03	0.32
GMD25	27.45	23.08	6.41	1.58	1.22	0.54
GWGP50	46.91	45.45	32.40	1.74	2.01	1.79
GCWP75	53.48	32.48	25.90	0.82	0.63	0.26
GWBP25	29.76	24.32	5.70	1.28	1.20	0.60
GRHA50	30.69	11.35	19.55	1.57	0.95	1.34

>25 MPa. In addition, GRHA50 was another fire resistant geopolymer and the decrease of the compressive strength was the lowest among the results and was obtained as −11.14 MPa. Besides, the bending strength decrease of the geopolymer composites showed similar decrease and the bending strength over 1 MPa after 800 °C was found for GWGP50 and GRHA50 as 1.79 MPa and 1.34 MPa, respectively. It can be stated that the remaining of compressive and bending strengths of GWGP50 and GHRA50 after 800 °C were sparkling and GWGP50 can be offered as the optimum geopolymer composite in terms of high temperature.

3.3.2. Freeze-Thaw

The specimens of selected best geopolymer composite series were subjected to 130 freeze-thaw cycles and the weight loss results of the specimens were given in Table 9. It was obviously seemed that the weight loss of geopolymers were higher than the control (GC) and lowest weight loss was found for GMD25 as 2.07% among the results, and GRHA50 with 2.26% weight loss followed the first one. The highest weight loss was determined for GWGP50 as 3.85% among the results. It was reported that the compactness was in relation with the porosity of the material [52,75]. When the compactness increased, porosity decreased [51,76]. Also, the high strength of the material can be obtained with low porosity and high compactness ratios in the medium [77]. In contrary to this, the porous environment allows frozen water and ice to expand during freezing and thawing, and the porous environment has a positive effect on freeze-thaw resistance [78]. Thus, high-strength materials with low void content may have lower freeze-thaw resistance and the materials can experience large mass losses during freezing and thawing [78–80]. It can be argued that this situation is valid for the high mass losses obtained in Table 9.

3.3.3. Sulphate attack

The specimens of selected best geopolymer composite series were subjected to sodium and magnesium sulphate attacks and the mechanical results of the specimens at the room temperature were given in Table 10. Harmful effects of sulphates were clearly observed, and some samples were damaged at such a high level. Thus, GMD25 and GWGP50 subjected to sodium sulphate, and GWGP50 and GRHA50 subjected to magnesium sulphate were damaged at high level and couldn't be tested. However, the geopolymers GCWP75 and GWBP25 could be tested after sulphate attacks and the weight loss of them were found as 17.62% and 10.79% after sodium sulphate attack and 14.54% and 13.14% after magnesium sulphate attack. In addition, the control (GC) had the lowest weight loss among the results and found as 10.14%.

Table 9
Weight loss of specimens after freeze-thaw cycles subjection.

Notation	Weight loss, %
GC	1.42
GMD25	2.07
GWGP50	3.85
GCWP75	2.85
GWBP25	3.58
GRHA50	2.26

Table 10
Mechanical results of specimens after sulphate attacks.

	Weight loss, %	
	Sodium sulphate	Magnesium sulphate
GC	11.62	10.14
GMD25	N/A*	15.32
GWGP50	N/A*	N/A*
GCWP75	17.62	14.54
GWBP25	10.79	13.14
GRHA50	11.81	N/A*

* No data obtained due to damage.

3.4. Results of decision support method (TOPSIS)

In this experimental research, a holistic examination approach depended on chemical compositions, physical properties, mechanical properties, and durability properties of geopolymer composites was adopted to select the best among them. The chemical composition of geopolymer composites were depended on the components of geopolymer (red mud, metakaolin, recycled aggregate, etc.) and calculated with the data given in Tables 1–4. TOPSIS is a decision-making support system and is used in many areas to select the suitable and the best choice in the alternatives according to their criterions. TOPSIS was employed to find the best choice in this paper and the criterions were thought to have the same importance selecting the same weights for each one. The criterions of the alternatives (GC, GMD25, GWGP50, GCWP75, GWBP25, and GRHA50) were set as SiO_2, Al_2O_3, Fe_2O_3, CaO, MgO, SO_3, Na_2O, K_2O, TiO_2, H_2O, density, water absorption, UPV, compressive strength, splitting tensile strength, bending strength, compressive strength after high temperature apply, weight loss after freeze-thaw, sodium, and magnesium sulphate attacks. After a series of calculation steps of TOPSIS (Eqs. 1–11), the results of TOPSIS for geopolymer composites GC, GMD25, GWGP50, GCWP75, GWBP25, and GRHA50 were presented in Table 11.

According to the TOPSIS results, the best geopolymer composite was determined as GWGP50 with 0.51 and the second was GWBP25 with 0.49 among the results while control had 0.28. Accordingly, GWGP50 had 50% waste glass powder + 50% recycled aggregate and GWBP25 had 25% brick powder + 75% recycled aggregate. On the other hand, the calculation of oxides for the best geopolymer composite GWGP50 showed that the composite included 49.3% SiO_2+CaO and it was the highest ratio in Table 11. It was well-known that C-A-S-H and N-A-S-H were the main structures constructed with SiO_2 and CaO [55,56,60,81] forming a durable medium of geopolymer and the results were supported by the literature.

The previously mentioned comments in Section 3.1., 3.2., and 3.3. included results that were made according to the related test results and

Table 11
TOPSIS results of geopolymer composites and other details.

	Main waste name and waste use ratio, %	Gi*	SiO_2 (%)	Al_2O_3 (%)	Fe_2O_3 (%)	CaO (%)	SiO_2 + CaO (%)
GC	-	0.28	37.8	29.3	5.8	6.4	44.2
GMD25	25% marble dust	0.48	30.3	30.4	4.9	13.7	44.0
GWGP50	50% waste glass powder	0.51	43.3	19.4	4.9	6.0	49.3
GCWP75	75% waste ceramic powder	0.46	41.9	19.4	6.2	2.7	44.6
GWBP25	25% waste brick powder	0.49	38.1	25.6	5.8	6.3	44.4
GRHA50	50% rice husk ash	0.35	45.0	21.1	4.4	3.6	48.6

*Higher is good.

related literature. Hence, the results were locally evaluated in consideration of each test results. Also, some fruitful comments were made, and the optimum proportions were determined and suggested. However, the suggestions may be different from the results of TOPSIS. Because TOPSIS-based results were depended on all physical, mechanical, durability and chemical properties, and holistic evaluation was followed for TOPSIS. According to the authors thoughts, the followed holistic evaluation may be more valuable than the singular evaluation less risk of making wrong decisions.

4. Conclusion

In this paper, a sustainable geopolymeric composite design with high waste content was aimed. Accordingly, design parameters were focused to lower the carbon emission and waste content around the world contributing the sustainable low carbon material topic of EU's Horizon 2030. Thus, waste glass powder, waste brick powder, waste marble powder, ceramic waste powder, and rice husk ash were preferred and used with recycled aggregate. Red metakaolin-mud-based geopolymeric composites were produced including the mentioned wastes. The properties of the sustainable geopolymer were obtained and evaluated solely. In addition, a holistic evaluation strategy was adopted in this comprehensive study and a decision support method was used to find the best geopolymer in consideration of all findings.

Despite inclusion of waste (i.e., RA) in geopolymer composite with low properties, marble dust, brick powder, ceramic powder rice husk ash and glass powder were the potential wastes to improve the properties of geopolymer composites. However, the optimum proportion of the waste in geopolymer mixture varied according to their chemical and physical properties. For instance, 25% marble dust, 25% brick powder, 75% ceramic powder, 50% rice husk ash and 50% glass powder were the optimum ratios to obtain maximum compressive strength. Splitting tensile strength evaluation can be made in many ways. When the ratio of splitting tensile strength to compressive strength were examined, 25% glass powder showed better results than others and it was calculated as 0.088 for GWGP25. However, the lower and upper values of the ratios were not the same in terms of waste type and the range between the upper and lower values of the ratios were highest for consideration of rice husk ash in geopolymer and it was the lowest for brick powder consideration in geopolymer.

The composed geopolymer mixtures with various types of wastes had different durability properties. In terms of high temperature resistance, the use of glass powder in the mixtures showed a satisfactory results and the higher compressive strength values were obtained for GWGP50. Besides, resistance to freeze-thaw of geopolymers had approximately the same values and the lowest weight loss after freeze-thaw cycles applied was found as 2.07% for GMD25 and GRHA50 followed the first with 2.26%. Although the test results gave different data to evaluate solely, the holistic approach depended on multi-criteria decision-making system evaluated the whole results and TOPSIS was used to find the best geopolymer composite. Accordingly, 22 parameters were regarded in TOPSIS. The criterions of the GC, GMD25, GWGP50, GCWP75, GWBP25, and GRHA50 were chosen as SiO_2, Al_2O_3, Fe_2O_3, CaO, MgO, SO_3, Na_2O, K_2O, TiO_2, H_2O, density, water absorption, UPV, compressive strength, splitting tensile strength, bending strength, compressive strength after high temperature apply, weight loss after freeze-thaw, sodium, and magnesium sulphate attacks. Thus, the best geopolymer composite was determined as GWGP50 with 0.51 and the second was GWBP25 with 0.49 among the results while control had 0.28.

Although the best geopolymer composite had the highest value in the analysis, it should be noted that the result of 0 in TOPSIS meant the absolute bad and the result of 1.00 in TOPSIS meant the absolute good, and accordingly the best geopolymer composite had 0.51. It showed that geopolymer GWGP50 had aspects that were open to development and thus the value can be come close to 1.00.

CRediT authorship contribution statement

Fatiha Bendjilali: Writing – review & editing, Writing – original draft, Investigation, Data curation. **Turgay Çoşgun:** Writing – review & editing, Methodology, Investigation, Data curation. **Hasan Dilbas:** Writing – review & editing, Writing – original draft, Validation, Methodology, Investigation, Data curation, Conceptualization. **Mucteba Uysal:** Writing – review & editing, Validation, Supervision, Methodology, Investigation.

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

Data will be made available on request.

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