



Contents lists available at ScienceDirect

Construction and Building Materials

journal homepage: www.elsevier.com/locate/conbuildmat

Effect of industrial waste-based precursors on the fresh, hardened and environmental performance of construction and demolition wastes-based geopolymers

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ARTICLE INFO

Keywords:

Construction and demolition waste
Industrial waste
Fresh properties
Hardened properties
Geopolymer
Life cycle assessment

ABSTRACT

The main goal of this study is to evaluate the effects of incorporation of the industrial wastes into Construction and Demolition Waste (CDW)-based geopolymer mixtures. To do this, a series of CDW-based geopolymer paste mixtures were produced and blast furnace slag (BFS), fly ash (FA), and silica fume (SF) were used to partially replace the CDW-based materials at the substitution rate of 10 and 20 % by weight. Flow table, buildability, vane shear, ram extruder and compressive tests were performed to assess some engineering performances of produced mixtures. Besides that, life cycle assessment analysis was conducted to reveal both the environmental impact of CDW-based mixtures and effects of industrial wastes. The results demonstrated that the geopolymer mixture with higher flowability and longer open time can be obtained by the incorporation of industrial waste precursors. Among them, FA incorporation resulted in the highest improvement in flowability while more decrement in shear yield stress was obtained from the SF-incorporated mixtures. Substitution of CDW-based precursors with industrial wastes provided higher compressive strength test results. Although 100% CDW-based geopolymer paste had lower environmental burden, the mixture using FA, BFS and SF followed very closely behind it. The transportation-related impacts were the most determinative factor for the differences between the environmental performance of geopolymer mixtures containing and non-containing industrial waste-based precursors. Overall, the main sources of environmental burdens of CDW-based geopolymer systems were the alkaline activators, electricity, and transportation. Results showed that industrial waste-based precursor can be successfully used in CDW-based geopolymer matrix to adjust engineering properties without endangering the environmental burden of the mixture.

1. Introduction

The generation of construction and demolition wastes (CDWs) is increasing globally as a consequence of the demolition of end-of-life structures, the construction of new buildings and infrastructure, and the expansion of existing structures in favor of providing more livable areas for the growing population. However, such a large quantity of generated CDWs was not properly managed in the circular value chain and were rather deposited in clean landfilling areas or used in low-tech applications such as filler materials in basement construction or as

aggregates in non-structural concrete [1]. Considering the 10 billion tons of CDWs generated annually worldwide, it is an urgent need to develop innovative, sustainable, and feasible solutions to solve CDW generation-based problems and to address the environmental, economic, and social impacts of these wastes [2]. In this way, the problems associated with the CDWs can be solved, creating a more livable world for people. Additionally, CDWs can be converted into high-value-added products resulting in economic benefits for countries. To this end, recent studies have focused on valorization of CDWs in the construction industry in order to reduce the current environmental burden of this sector

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<https://doi.org/10.1016/j.conbuildmat.2023.132265>

Received 1 June 2023; Received in revised form 20 June 2023; Accepted 22 June 2023

Available online 28 June 2023

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[3].

Geopolymers, which can be defined as inorganic polymers produced by a reaction between aluminosilicate materials (i.e., precursor) and alkaline activators [4], have been shown to be a promising alternative to cement-based systems in literature studies [5–10]. Most of the studies related to the geopolymers have focused on the use of mainstream precursors such as fly ash (FA), blast furnace slag (BFS), silica fume (SF), and metakaolin (MK), etc. However, the cement and concrete industry are increasingly relying on these wastes, as they are beneficial, and are therefore demanding more of them. Therefore, the prices of these precursors increase, and the availability of these materials in the market decreases, which can pose an obstacle to the production of geopolymers being a worldwide alternative to the cement-based systems. By taking these facts into consideration, studies began to be conducted on the use of different kinds of waste materials as precursors for geopolymerization. In view of the current situation and the quantities of CDWs available, researchers started to explore the use of CDWs as precursors for the production of geopolymers, since the chemical composition of CDWs (which mainly consists of aluminum, silicon, and calcium sources) was found to be suitable for geopolymerization [11–17]. The current studies related to the CDW-based geopolymers showed that CDW-based geopolymers can exhibit comparable performance to that of conventional cementitious systems and geopolymers made from mainstream precursors [9,13,18,19]. However, the development of CDW-based geopolymers needs to be further investigated in detail as they are less explored compared to the geopolymers based on mainstream precursors and the knowledge about CDW-based geopolymer systems are limited.

The studies conducted on the development of CDW-based geopolymer binder systems showed that the ultimate performance of the mixtures can be modified by adjusting the alkaline content, water-to-binder ratio and aggregate-to-binder ratio, chemical composition of the precursor, curing period, and temperature. In this context, the study conducted by Ulugöl et al. [9], showed that for a CDW-based geopolymer mixture composed of red clay brick (RCB), roof tile (RT), and hollow brick (HB), the main geopolymerization products were N-A-S-H gels with different zeolitic polytypes ranging from amorphous to polycrystalline structure. However, for the geopolymers made of glass waste (G) precursor, the main geopolymerization product was comparatively weaker and unstable sodium silicate gels. And it was concluded that increasing the NaOH concentration, curing period and temperature, as well as the content of SiO₂ and Al₂O₃ resulted in an increase in the compressive strength up to a certain point. It was also stated that fineness was effective on the geopolymerization and the compressive strength of CDW-based geopolymer mixtures could exceed 45 MPa depending on the given conditions. In the study of Yıldırım et al., [13] it was stated that the CDW-based masonry units (RCB, RT, HB) can be effectively used to produce geopolymers with a compressive strength of up to 80 MPa at elevated temperature curing, provided that the mixture design parameters are optimized. Microstructural investigation results showed that, regardless of the chemical content of the CDW-based precursors, the main reaction product was N-A-S-H gels. In another study, Mahmoodi et al. [20] found that the maximum compressive strength for CDW-based geopolymers (concrete waste (C)) after 28 days ambient curing and 100 °C heat curing was 20.5 and 37.1 MPa, respectively. The results obtained showed that the fresh and hardened properties of CDW-based geopolymers are highly dependent on the chemical ratios of Si/Al and Na/Si. Additionally, it was concluded that the inclusion of slag had a greater impact on the mechanical properties of CDW-based geopolymers compared to FA and MK, as slag led to formation of more C-A-S-H (tobermorite type) and N-A-S-H gels in the CDW-based geopolymer matrix. In the studies of Ilcan et al., [1,15] the effects of alkaline content from different sources (NaOH, Ca(OH)₂, and Na₂SiO₃) on the fresh and hardened properties of CDW-based geopolymer mortars composed of RCB, RT, HB, G, C and recycled concrete aggregate (RCA) were investigated. Results showed that alkaline content

has a significant influence on the properties of CDW-based geopolymers. It was revealed that, by arranging the alkaline types, combinations, and concentrations, it is possible to design different kinds of construction materials that meet specific application requirements. And the compressive strength values of CDW-based geopolymer mortars were found to be between 11 and 36 MPa at the end of 28-day ambient curing. The study conducted by Khan et al. [21] on the life cycle assessment (LCA) of CDW-based geopolymer system found that the environmental burden of CDW-based geopolymer binder (RCB-, RT-, HB-, G-based) was lower than that of equivalent cement-based materials. Results showed that the electrical energy requirement for CDW processing (grinding, crushing etc.) had the highest environmental implications, accounting for more than 64% of the global warming potential (GWP) while NaOH production was the second most energy-intensive process. In another study [22], LCA analysis was conducted on ceramic tile and brick waste-based geopolymers and results showed that heat curing had a negative environmental impact, although it was not significant. In this study, the highest environmental impacts were recorded from the Na₂SiO₃, NaOH and electricity consumption for CDW-processing.

In recent research, different types of industrial waste have been substituted into CDW-based geopolymer mixtures in order to tailor the fresh and hardened properties of the mixtures. In one of this study, Mahmoodi et al. [12] investigated the effects of Class C FA, Class F FA, MK and granulated BFS on the fresh and hardened properties of the ceramic tile waste-based geopolymer. The results showed that the inclusion of different types of industrial wastes had a significant and varied impact on the properties of the CDW-based geopolymer by providing extra dissolved quantities of Ca²⁺, Si⁴⁺ and Al³⁺ in reaction medium, which then produce geopolymerization product, and by forming different physical interactions due to the varied surface texture. Higher compressive strength was achieved for the mixtures cured under ambient condition when calcium sources materials (Class C FA and BFS) were added to the mixtures. In another study, Hwang et al. [23] analyzed the performance of brick and ceramic waste-based geopolymer mixtures containing varying levels of BFS (0–50%). The results revealed that the higher substitution of BFS resulted the formation of densified C-S-H and C-A-S-H gels, binding and covering the unreacted particles. Moreover, the higher levels of CaO in the BFS led to denser hydrated gels, resulting in better pore refinement. In addition, substitution of BFS was found to be favorable in increasing strength. In the study of Tan et al. [24], mitigation of efflorescence for CDW (consisting of concrete, mortar, bricks and tiles, glass, aggregate)-based geopolymer mixtures through the inclusion of MK and BFS was investigated. Results showed that substitution of MK and BFS significantly decreased the efflorescence formation. Additionally, it was revealed that substitution of MK and BFS increased compressive strength and reduced flowability of the mixtures.

Although several studies have been conducted on the effects of industrial waste substitution on the properties of CDW-based geopolymer mixtures, these studies are much fewer in comparison to those conducted on mainstream precursor-based geopolymers and tend to focus on specific topics. The novelty of this study lies in its comprehensive and comparative experimental and environmental impact analysis on CDW-based geopolymer mixtures containing industrial wastes. Additionally, special attention was given to tests conducted in the scope of this study to explore the possibility of using these materials in advanced manufacturing technologies. Experimental studies evaluated in detail the effects of industrial waste-based precursors on the fresh properties of the CDW-based geopolymer mixtures, by conducting flow table, buildability, vane shear, ram extruder test methods. Additionally, compressive strength test was conducted to determine the effects of industrial wastes on the mechanical performance of mixtures. To assess the environmental burden of the developed geopolymer mixtures and substitution of industrial wastes, a life cycle assessment was performed in the scope of this study. CDW-based clay-originated masonry units, including hollow brick (HB), red clay brick (RCB), and roof tile (RT) along with waste glass (G) and industrial waste-based precursors including BFS, FA,

and SF were used as precursors in geopolymer mortar mixture designs. For the alkaline activation of geopolymers, sodium hydroxide (NaOH), and calcium hydroxide [Ca(OH)₂] were used in binary combinations. It is believed that the findings of this research will contribute to the current literature by demonstrating the effects of industrial waste-based precursors on the properties of the CDW-based geopolymer systems. Besides that, the outcomes of this study are believed to make a significant contribution to the current state-of-the-art, as the findings provide comprehensive insights and knowledge about tailoring the fresh and hardened properties of the CDW-based geopolymer mixtures considering environmental burdens. In addition to that, developed products will be further considered and optimized for novel sources to provide requirements for advanced manufacturing processes.

2. Experimental program

2.1. Materials

The CDW-based materials used in this study as precursor material (including hollow brick [HB], red clay brick [RCB], roof tile [RT] and glass [G]) were obtained from the demolition sites in Turkey. Once the process of selective demolition was completed, since the CDW elements were obtained in large sizes, they were further broken down into smaller pieces. Then, they were subsequently reduced in size with a jaw crusher and milled during one hour in a laboratory-type ball mill to obtain powdery materials. Industrial waste materials such as BFS, FA and SF were also employed in geopolymer precursor blends. Fig. 1 displays the visual representations of CDW-based materials in their raw, crushed, and ground forms and their scanning electron microscopy (SEM) images. Additionally, the figure also features the general views and SEM images of industrial waste materials being used in the geopolymerization process. X-ray fluorescence (XRF) analysis was conducted to determine the oxide compositions of the used precursor materials (Table 1). For the CDW-based precursors, the main oxides available in the chemical

Table 1
Chemical compositions of precursors.

Oxides, %	HB	RCB	RT	G	BFS	FA	SF
SiO ₂	39.7	41.7	42.6	66.5	38.2	60.1	81.2
Al ₂ O ₃	13.8	17.3	15.0	0.9	13.0	21.4	0.5
Fe ₂ O ₃	11.8	11.3	11.6	0.3	0.7	7.4	1.3
CaO	11.6	7.7	10.7	10.0	35.3	1.0	0.4
Na ₂ O	1.5	1.2	1.6	13.6	0.1	1.0	0.6
MgO	6.5	6.5	6.3	3.9	6.5	1.8	5.8
SO ₃	3.4	1.4	0.7	0.2	0.5	0.2	0.5
K ₂ O	1.6	2.7	1.6	0.2	0.9	2.9	2.6
TiO ₂	1.7	1.6	1.8	0.1	1.0	0.9	0.1
P ₂ O ₅	0.3	0.3	0.3	0.0	0.0	0.2	0.0
Cr ₂ O ₃	0.1	0.1	0.1	0.0	0.0	0.0	2.5
Mn ₂ O ₃	0.2	0.2	0.2	0.0	1.4	0.1	0.1

composition of the clay-originated precursors (RCB, RT, and HB) were SiO₂, Al₂O₃ and Fe₂O₃, while G contained SiO₂, Na₂O and CaO as the main oxides. For industrial waste materials, the main oxides available in the chemical composition of BFS were SiO₂, Al₂O₃ and CaO, while FA contained SiO₂, Al₂O₃ and Fe₂O₃ and SF contained high amount of SiO₂ as the main oxides. Particle size distributions (PSD) of the various precursor materials were also determined by conducting laser diffraction analysis having particle size range sensitivity of 0.02–2000 μm (Fig. 2). While clay-originated wastes including HB, RCB, and RT, and industrial waste-based BFS had similar PSD, G and FA were in coarser grain sizes and SF had the finest grain size.

For alkaline activation of waste-based precursors, upon previous studies conducted and proven achievement in literature [1,14,15], two different alkaline activators were utilized: (i) sodium hydroxide as flake form with a minimum of 98% sodium hydroxide, maximum 0.4% sodium carbonate, 0.1% of sodium chloride and 15 ppm of iron, and (ii) calcium hydroxide in powder form with 87% purity.

2.2. Mixture proportions

In total, seven different geopolymer paste mixture designs were set in this study. First, a dry base paste mixture design containing entirely (100%) CDW was devised, and the binder material blend used for this paste mixture was composed of the combination of 30% RCB, 30% HB, 30% RT, 10% G, by the total weight of precursor materials according to the preliminary study published by the authors [14]. Then, in addition to the 100% CDW-based system, different industrial waste-incorporated paste mixtures were designed. To do this, clay-originated waste powders of HB, RCB, RT (equal replacement amounts for each type of clay-originated wastes) were replaced by BFS, FA and SF separately at two

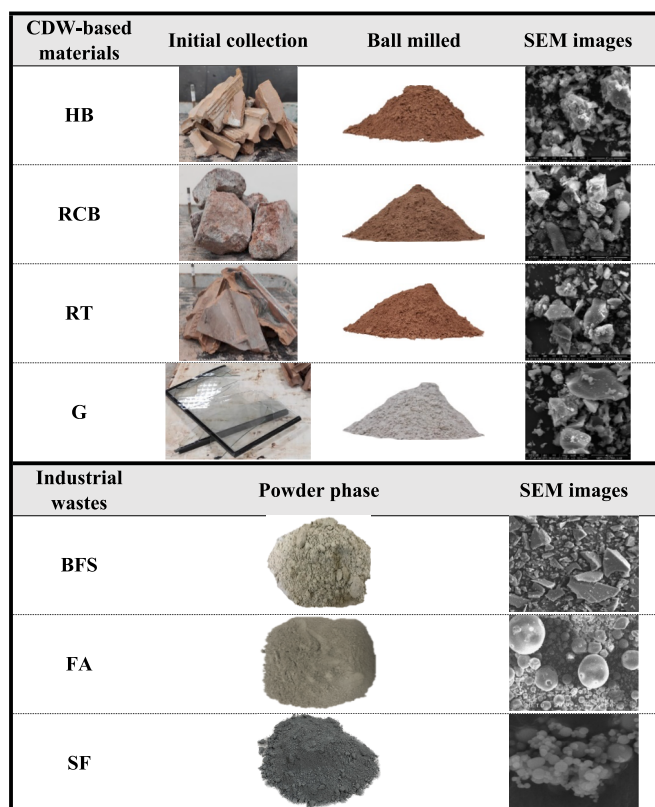


Fig. 1. Images of the CDW-based and industrial waste-based precursors.

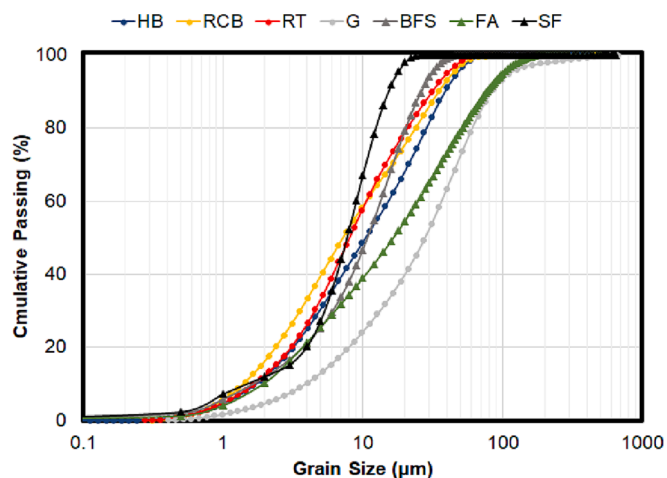


Fig. 2. Particle size distribution curves of CDW-based precursors and industrial wastes.

different replacement levels of 10% and 20%, by weight. All paste mixtures were produced with NaOH molarity of 6.25 M and the Ca(OH)₂ utilization rate of 10% and the water-to-binder ratio (by weight) (w/b) was fixed at 0.33 for all mixtures. Table 2 presents the mixture proportions of geopolymers prepared with the combination of CDW-based powders and industrial wastes.

2.3. Mixture preparation and curing

The production procedure of geopolymer paste mixtures began with preparing alkaline solutions. The required quantities of NaOH in the form of solid flakes were dissolved in tap water, then left to cool since the dissolution reaction of NaOH is exothermic. A mortar mixer was used to combine all ingredients into a homogeneous mixture. The mixing process involved following steps: (i) all of the solid components were softly mixed [precursors, and Ca(OH)₂] for a minute at a low speed (100 rpm); (ii) the NaOH solution was then added and the mixture was allowed to stir for another 60 s; (iii) mixing was continued for a further 90 s at low speed (100 rpm); (iv) finally, the mixture was blended at a medium speed (150 rpm) for the last 60 s, and that concluded the mixing process.

50-mm cubic specimens were produced by pouring fresh geopolymer paste mixtures into pre-oiled molds. These specimens stayed in their molds for 24 h at the laboratory environment with the temperature of 23 ± 2 °C and relative humidity of 50 ± 5%. Subsequently, they were cured in ambient condition in same laboratory environment until they reached the required testing ages of 7 and 28 days. The remaining fresh mixture was utilized for the assessment of rheological properties.

2.4. Testing methods

2.4.1. Flow table test

To evaluate the flowability of geopolymer paste mixtures, the flow table test was conducted according to the ASTM C1437-15 standard. Kazemian et al. [25] found a strong relationship between increased spreading diameter and yield stress. Therefore, it can be possible to state that this test assesses the yield stress of the mixtures which is crucial for fresh state property assessment. To measure the yield stress of the mixture, the flowability index (Γ) was determined by using Eq.1 [26] taking the spreading diameters of the geopolymer paste mixtures into account.

$$\Gamma = \frac{d_1 d_2 - d_0^2}{d_0^2} \tag{1}$$

where the inner diameter of the mold is d₀ (100 mm) while d₁ is the maximum spreading diameter and d₂ is the spreading diameter perpendicular to d₁.

2.4.2. Buildability test

A modified version of mini-slump test (complying with ASTM C1437 standard) proposed by Nematollahi et al. [27] was used to determine the

buildability performance of geopolymer mortar mixtures in this study within the purpose of investigating the response of fresh state mixtures under static load. This test involved placing the mixture in the mini-slump cone followed by a one-minute resting period. Then a glass plate was placed on the mixture, with a static load of 600 g including the weight of the plate, for one minute. The deformation of the fresh geopolymer mixture was then measured in two perpendicular directions and the average height was noted. The results were evaluated by considering that lower deformation or higher final height indicates higher yield stress of the mixture.

2.4.3. Vane shear test

Vane shear test was also conducted to assess the flow properties of freshly prepared geopolymer paste mixtures. The pocket-type vane shear tool (with the shear stress capacity of 0–10 N/cm² and vane diameter of 48 mm) was employed to measure the flow characteristics of fresh-state geopolymer mixtures. The test was performed to determine the resistance of the fresh state material to shear forces and to evaluate the rheological properties of the geopolymer mixtures. This single shear procedure was mainly used to compare the yield stresses of geopolymer mixtures, as yield stress values can vary according to the test methods employed [28]. The vane shear test was also performed on the mixtures after 30, 60, and 120 min from the end of mixing to give insight into how the shear yield stress changes over time and determine the workable time of the geopolymer mixtures.

2.4.4. Ram extruder

This study used a ram extrusion test to evaluate the shear yield stress of geopolymer paste mixtures. The ram extruder used in the study was designed based on literature studies [29–31], and consisted of four components: piston, chamber, nozzle, and stands (Fig. 3). It was set up on a universal testing device. Although the diameter of the chamber was 4 cm, mixtures flowed through a 1.5 cm-diameter nozzle. The piston speed was adjusted to 2.25 mm/s by considering this difference. This piston speed resulted in a flow speed of 16 mm/s for the extruded material.

Extrusion pressure was determined using the equation proposed by Chen et al. [29] (Eq.2). The measured extrusion pressure or yield stress when the mixture is flowed through the chamber was represented by σ₀, and the average extrusion force used to obtain the predetermined piston speed was denoted by F. The inner diameter of the chamber was equal to 4 cm (D₀).

$$\sigma_0 = 4F/\pi \times D_0^2 \tag{2}$$

Extrusion pressure obtained from the ram extruder were transferred into the shear yield stress, in accordance with Von Mises criterion [32], by using the Eq.3.

$$\tau_0 = \sigma_0/\sqrt{3} \tag{3}$$

where, τ₀ is the shear yield stress and σ₀ is the elongational yield stress.

Table 2
Proportions of paste mixtures.

Mixture Code	Alkali activators				Precursor materials (1000 g)						
	NaOH		Ca(OH) ₂		HB	RCB	RT	G	BFS	FA	SF
	Molarity (M)	Amount (g)	Rate (%)	Amount (g)							
100CDW	6.25	82.5	10	100	300	300	300	100	–	–	–
90CDW-10BFS			10	100	266.7	266.7	266.7	100	100	–	–
80CDW-20BFS			10	100	233.3	233.3	233.3	100	200	–	–
90CDW-10FA			10	100	266.7	266.7	266.7	100	–	100	–
80CDW-20FA			10	100	233.3	233.3	233.3	100	–	200	–
90CDW-10SF			10	100	266.7	266.7	266.7	100	–	–	100
80CDW-20SF			10	100	233.3	233.3	233.3	100	–	–	200

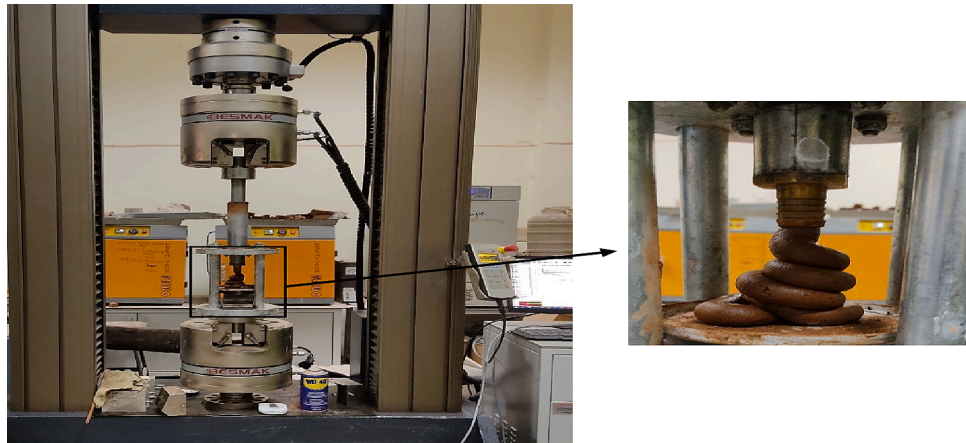


Fig. 3. View of the ram extruder setup.

The performance of geopolymer mixtures was also assessed over time in terms of their shear yield stress using a ram extruder. This was done after 30, 60, and 120 min from the end of initial mixing, similar to the vane shear test. This was done because geopolymerization reactions might cause these mixtures to lose their consistency.

2.4.5. Compressive strength test

The test was performed according to ASTM C109 standard at a loading rate of 0.9 kN/s, was used to evaluate mechanical properties of the $50 \times 50 \times 50$ mm (cubic) geopolymer paste specimens. Compressive strength tests were performed on three replicates of the cubic specimens for each mixture at curing ages of 7- and 28-day. The mean of the test results obtained from the three replicates were reported as compressive strength.

3. Life cycle assessment

3.1. System description

LCAs were performed following thoroughly International Organization for Standards (ISO) 14,040 and 14,044 criteria for the methodology of this study. LCA studies were prepared in four stages including goal and scope definition, inventory analysis, impact assessment, and interpretation in ISO guidelines as shown in Fig. 4.

3.2. Goal and scope

The aim of this study was to evaluate the environmental effects and sustainability performance of producing geopolymer pastes for using CDWs with various industrial waste-based materials such as BFS, FA and

SF at different rates (90% CDW-10% industrial waste-based material, 80% CDW-20% industrial waste-based material) with the suitable blend (6.25 M-NaOH and 10% $\text{Ca}(\text{OH})_2$) of alkali activators. The LCAs were performed with Simapro software in an iterative manner.

3.3. Life cycle inventory

The system's inventory data made of the material and energy inputs at the system boundary were gathered and presented in this part. Material procurement processes were carried out in different cities. CDW-based materials were gathered from various demolition sites and alkaline activators were taken from the chemical manufacturer in Ankara, Turkey. While FA was provided from İSKEN Sugözü Thermal Power Plant, BFS was obtained from İskenderun Iron and Steel Inc., and SF was brought from Antalya-Etibank Ferro-Krom Factory. The information regarding the distances and vehicles required for the transportation of each material is given in Table 3.

To prepare geopolymer mixture, CDWs were subjected to the crusher and milling processes to obtain powdery form. Then, geopolymer pastes were obtained through mixing process with the mixer. During analysis, crusher, ball mill and mixer devices were selected as large-scale devices used in the sector. The information regarding to the devices used and the electrical energy consumptions are also presented in Table 4. The energy consumption values were calculated by multiplying the number of kg of material entered into each mixture at these stages by the consumption values per kg.

3.4. Life cycle impact assessment (LCIA)

The impact assessment results were obtained by using CML IA method [33] on Simapro software [34], and the Ecoinvent 3.0 database. The main impact assessment categories used in this study are global warming (kg CO_2 eq.), ozone depletion (kg CFC-11 eq.), acidification (kg SO_2 eq.), eutrophication (kg PO_4 eq.) and photochemical oxidation (kg C_2H_4 eq.).

3.5. Assumptions & limitations

LCA of this study aims to systematically assess the environmental impacts and overall environmental sustainability of producing geopolymer pastes. The assumptions for the LCA study were as follows:

- The LCA study was a cradle-to-gate analysis.
- The functional unit was set to be 1 m^3 of geopolymer pastes.
- The process was determined to be located in Turkey, however, due to the lack of available data in the software, RoW geography was

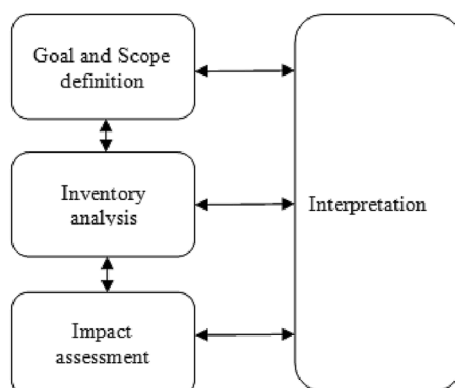


Fig. 4. LCA framework based on ISO standards.

Table 3

Transportation details of the precursors and alkaline activators.

Transportation of CDW		Transportation of BFS		Transportation of FA		Transportation of SF		Transportation of Alkali Activators	
ton	km	ton	km	ton	km	ton	km	Ton	km
10	20	10	620	10	560	10	480	0.5	15

Table 4

Consumed electrical energies of machines.

#	Jaw Crusher		Ball Mill		Mixer	
	Amount	Unit	Amount	Unit	Amount	Unit
Capacity	20,000	kg	20,000	kg	600	kg
Duration	1	h	1	h	1	h
Consumption	15	kwh	280	kwh	7.5	kwh
	0.00075	kwh/kg	0.014	kwh/kg	0.01	kwh/kg

chosen for the LCA. Nevertheless, electricity data were modeled based on the Turkey electricity country mix.

- The energy input of large-scale devices used in the sector, such as a jaw crusher, ball mill, and mixer, was considered for the production of geopolymers and calculated based on their corresponding hourly power consumption to accurately reflect the energy consumption caused by the material’s life cycle. The energy consumption calculations were based on the production capacity and power consumption of the industrial-grade crusher, ball mill, and mixer, which had respective power consumption values of 15 kW, 280 kW, and 7.5 kW.
- A production process for CDWs was not included. The Ecoinvent database was only used for treatment process for these wastes. This treatment process involved energy for dismantling, particulate matter emissions caused by dismantling and handling.
- BFS, FA and SF were waste of the iron industry, coal thermal power plants and silicon alloy plants, respectively and do not require a separate process for production [35]. FA and SF, for example, were waste materials and had no bearing on the environmental impact of the end product, as determined by [36]. Their environmental impact was limited to the specific processing required for their use in concrete (grinding, drying and stock). The LCA data for these materials

were taken directly from the Ecoinvent database for waste and emissions to treatment.

4. Results and discussion

4.1. Fresh properties

Rheological test results of entirely (100%) CDW-based and BFS-, FA- and SF-substituted geopolymer paste mixtures produced with a blend of NaOH and Ca(OH)₂, are demonstrated in Fig. 5.

4.1.1. Effects of BFS

According to the results given in Fig. 5-a, in terms of the effect of the BFS substitution, it was observed that BFS substitution increased the flowability of the mixtures while decreasing buildability. This behavior was general and valid for all substitution levels (10% and 20%). This can be caused by the differences in the physical and chemical properties of the BFS and masonry-originated waste powders (HB, RCB and RT) and can be attributed to less water requirement because of the decrease in the amount of plate-shaped (layer-like structure) masonry-originated waste powders by BFS substitution [37–40]. These reasons also resulted in decrement in the vane shear stress and shear yield stress values of the mixtures by descending of the initial viscosity of the mixtures, and slower setting and hardening response (longer open time which means a longer duration during which a substance remains workable or usable) as can be followed from and Fig. 5-b. The vane shear stress and shear yield stress of the mixtures, regardless of the different mixture design parameters, continuously increased with passing time. This can be attributed to the ongoing geopolymer reactions resulting strengthening of the matrix with time [14,15]. As mentioned before, the results showed that, at the end of 120 min, BFS substitution resulted in lower vane shear stress values (compared to 100% CDW-based mixture), in agreement with previous studies [41–44]. On the other hand, contrary to expectations, the results showed that the substitution of BFS did not

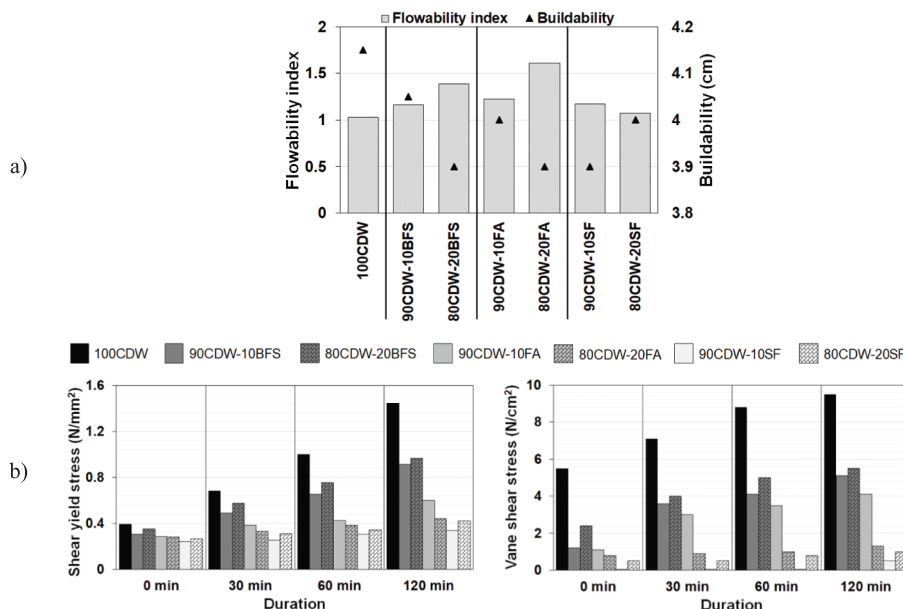


Fig. 5. Rheological test results of (a) flowability and buildability, (b) yield stresses.

yield rapid hardening response (Fig. 5-b), even though an excess of calcium ions in BFS leads to rapid gel formation in an alkaline environment, as reported in the current literature [40,45–47]. This opposite result can be attributed to the rapid formation of calcium-based gels in the alkaline medium, which covers the precursor materials and slows down the dissolution rate for the initial hours, thus delaying the hardening of the matrix [48]. Additionally, the higher flowability of the BFS-substituted mixtures may also be contributed to the extended open time. When the substitution level of BFS within the mixtures was increased from 10% to 20%, the vane shear stress and shear yield stress values of the mixtures increased. Possible reasons that can cause this behavior can be related to the increased surface areas with the further increase in BFS content and thereby increased water requirement for wetting the surface of the particles [49] and excessive increase in calcium content.

4.1.2. Effects of FA

When the test results of geopolymer pastes are evaluated from Fig. 5-a, it can be clearly observed that FA-substituted mixtures exhibited relatively higher flowability index values compared to entirely (100%) CDW-based pastes. Also, the buildability results decreased with the substitution of FA. As the substitution rate increased, the increase in the flowability (increase in flowability index and decrease in buildability) continued to increase. These results were also observable from the shear yield stress and vane shear stress results that can be seen from the Fig. 5-b. As mentioned before, masonry-originated waste powders (HB, RCB and RT) were replaced by FA. The increase in flowability of the geopolymer paste mixtures with the substitution of FA can be related to the differences in the physical and chemical features of the FA and masonry-originated waste materials (HB, RCB and RT), such as, lower porosity and relatively more smooth surface and spherical (round) structure of the FA particles compared to those of plate-shaped (layer-like structure) masonry-originated waste material particles. These may result in less fluid requirement to lubricate the particle surface, lower water absorption and lower particle-to-particle friction [37,50–54]. When evaluating the test results in terms of the time-dependent variability in the fresh properties of the geopolymer paste mixtures, it can be seen that shear yield stress and vane shear stress values of the FA-substituted mixtures increased over time. This was an expected result considering the ongoing geopolymerization reactions with time. At the end of 120 min, it was observed that FA-substituted paste mixtures had considerably lower vane shear stress and shear yield stress values compared to those of entirely (100%) CDW-based mixtures. This result can be attributed to the fact that relatively higher flowability of the FA-substituted mixtures arising from the shape of the FA particles preventing trapping of free water in agglomeration [55]. In literature, there are parallel results which indicate that incorporation of the FA increases the setting time of the geopolymer mixtures [56]. As a result, this finding means that the substitution of FA increased the setting time of the mixtures and positively affected the long-term workability performance of the mixtures.

4.1.3. Effects of SF

To clarify the effects of SF on the fresh properties of CDW-based geopolymer mixtures, obtained results were examined by comparing the fresh properties of the geopolymer mixture composed of 100% CDW-based precursor. Flowability test results illustrated in the Fig. 5-a showed that 10% substitution of SF yielded an increment in the flowability of CDW-based geopolymer paste. However, 20% substitution of SF caused a decrement in flowability index and showed slightly higher flowability performance than the 100CDW coded mixture. According to the obtained results, the buildability performance of CDW-based geopolymer mixtures decreased with the substitution of SF, irrespective of usage rate. Although 10% SF substitution negatively affected the buildability, increment of SF usage rate showed an increment in buildability performance, yet 100% CDW-based geopolymer mixture showed higher buildability performance compared to the mixtures containing SF. Similar trends were obtained from both ram extruder and vane shear

test results. According to results, 10% substitution of SF considerable decreased the shear yield and vane shear stress values, but 20% SF containing mixture showed increment in both shear yield and vane shear stresses. However, 100% CDW-based geopolymer mixtures showed higher shear stress values than the mixtures containing SF. These trends were similar for each testing duration of 0-, 30-, 60-, and 120-min. Open time test results showed that the incorporation of SF into the CDW-based geopolymer system retarded the geopolymerization as can be seen from Fig. 5-b. There has not been observed considerable changes in shear stresses of mixtures containing SF until 120 min, although 100% CDW-based geopolymer mixtures showed an increment in shear stresses with elapsed time until 120 min.

Although it was expected SF to decrease flowability and increase shear stresses because of very fine particle size, high surface area and high-water absorption capacity, substitution of 10% SF caused increment in flowability, and therefore decrement in buildability performance and shear stresses. This could be attributed that the fine SF may densify the inner structure and reduce the amount of water filled in the voids and may show lubricating effect by preventing particles to sticking each other due fine spherical shape (as CDW-based precursors have rough surface) [14,57,58]. In addition, lower amount of OH⁻ ion existence on the surface of CDW-based precursor with the presence of SF particles in medium may cause retardation of dissolution and reaction [59]. Therefore, due to lower dissolution and reaction, weak bonding in medium may yield higher flowability. Besides that, the formation of silica-based complexes, because of the dissolution of SF at the beginning of mixing, may act as a superplasticizer by sterical repulsion and adsorption on the surface of CDW-based precursor, which can be another reason for the increment in the flowability [14,60]. However, substitution of 20% SF caused decrement in flowability and thereby enhancement of buildability and shear stresses compared to 10% SF substitution. So, the above-mentioned positive effects of SF on fluidity were replaced by the negative effects with the increase in the amount of use. This phenomenon can be attributed to increase in the number of fine particles, resulting in a larger surface area, an increased water absorption, and a denser pore structure. [61,62]. In addition, as observed in the study of Memon et al. [57], containing higher amount of SF yielded more cohesiveness and stickiness, which can also be reason of decrement in flowability for the 20% substitution.

In addition to the initial fresh properties, open time performance of mixtures was enhanced with the substitution of SF irrespective of usage rate. Such finding could be explained in a way of that the inclusion of SF may lower the OH⁻ ion presence on the surface of CDW-based precursor due to fineness, which may lead retardation of dissolution and geopolymerization [59]. Besides that, undissolved SF particles may be adsorbed on the surface of CDW-based precursors as a result of electrostatic interaction, which may also retard the rate of dissolution and geopolymerization [63].

4.2. Compressive strength

Compressive strength test results of entirely (100%) CDW-based and BFS-, FA- and SF-substituted geopolymer paste mixtures produced with a blend of NaOH and Ca(OH)₂, are demonstrated in Fig. 6.

4.2.1. Effect of BFS

As can be seen from Fig. 6, incorporation of BFS into the mixtures made monitorable enhancements in compressive strength results. Compressive strength results exhibited an incremental trend when the amount of BFS increased for all mixtures tested after 7- and 28-day curing ages. BFS substitution by 10% provided 23.8% increment in the 28-day average compressive strength of geopolymer mixtures, while the increment level was 69% for 28-day-old 20% BFS-substituted mixtures. This can be attributed to the enhancing the microstructure of the geopolymer matrix due to increased reactivity of precursor content with the inclusion of BFS [64–67]. Another possible reason may be due to the

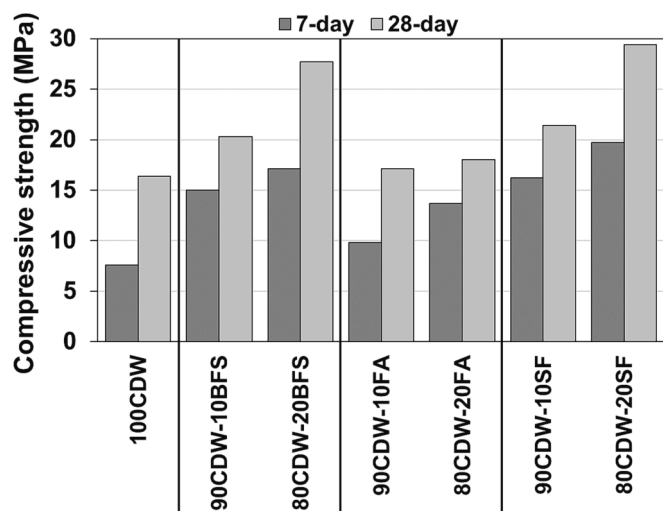


Fig. 6. Compressive strength test results.

increase in the calcium content of the mixtures providing higher availability of the Ca^{2+} ions in the reaction medium to take part into the geopolymerization and partially hydration (self-cementitious property), and thereby further formation of strength giving products [68,69]. By considering this, it is possible to state that BFS incorporation provided the formation of calcium-based gel structures (calcium silicate hydrate (C-S-H) and calcium aluminosilicate hydrate (C-A-S-H)) and also an intertwined gel ([C-(N)-A-S-H]) alongside with the sodium aluminosilicate hydrate (N-A-S-H) gels (main reaction product of CDW-based geopolymers) in favor of obtaining geopolymer paste mixtures with a denser and more compact microstructure [10,70,71].

4.2.2. Effect of FA

When the compressive strength results of the FA-substituted and entirely (100%) CDW-based paste mixtures are examined from Fig. 6, it can be seen that, compared to other industrial wastes, there was not a significant influence of FA substitution on the 28-day compressive strength of the paste mixtures, irrespective of substitution rate. Although there were significant increases in the compressive strength of 7-day-old FA-substituted geopolymer mixtures, lower levels of strength increases were observed in those of 28-day-old geopolymer mixtures. As in the BFS-substituted mixtures, increases in the FA substitution rate was more beneficial for increasing compressive strength results. FA substitution by 10% provided 4.26% increment in the 28-day average compressive strength of geopolymer mixtures, while the increment level was 9.76% for 28-day-old 20% BFS-substituted mixtures. Although, increases in compressive strength with the incorporation of FA can be attributed to higher proportion of amorphous phase present in the FA [72] and the increased presence of Si and Al in the reaction medium [70,73], the main reason for the limited improvement in 28-day compressive strength of the geopolymer pastes with the inclusion of FA can be related to the decreased calcium content in the reaction medium because of the replacement of FA with masonry-originated waste materials (HB, RCB and RT) [56].

4.2.3. Effect of SF

Compressive strength test results showed that incorporation of SF into the CDW-based geopolymer system increased the 7- and 28-day compressive strength test results, regardless of SF usage rates. At the end of 28-day ambient curing, the compressive strength increased by ~30.5% and 79.3% and reached 21.4 and 29.4 MPa for 10% and 20% SF substituted CDW-based geopolymer mixtures, respectively. Increment in compressive strength with the inclusion of SF into alkali activated medium can be attributed to (i) increment in the quantity of reactive Si in

reaction medium to form strength giving geopolymer products [14,15,57,74], (ii) high pozzolanic reactivity of SF with the available $\text{Ca}(\text{OH})_2$, resulting in more C-(A)-S-H production [59,62,75–77], (iii) densified micro pores and more compact matrix because of both more geopolymer product formation and the behaving of SF as a micro-aggregate [57,62,78].

4.3. Environmental impacts - LCIA results

Seven different geopolymer paste mixtures produced in this study were evaluated by considering five impact assessments and the main components affecting each category including NaOH, $\text{Ca}(\text{OH})_2$, transportation, electrical energy, water consumption, CDW treatment and the industrial wastes such as BFS, FA, SF were also investigated. The results obtained were demonstrated in the Fig. 7.

In accordance with the global warming impact category, which is related to CO_2 equivalent emission results, it can be stated that the industrial waste incorporation resulted in increment of global warming potential (GWP) values of CDW-based geopolymer mixtures. The geopolymer paste mixture containing 100% CDW had the smallest value of 310 kg CO_2 eq. It was followed by the 90CDW-10SF coded mixture with the value of 317 kg CO_2 eq. The highest impact was observed in the mixtures containing BFS. When the most important influencing components including NaOH, $\text{Ca}(\text{OH})_2$, and electricity were evaluated, it can be seen that their shares were between of 42–47%, 36–40%, and 8–10%, respectively. The main reason for differences between the GWP of the industrial waste-based precursor included and non-included mixtures was the transportation-related environmental burden. By taking the GWP impact of transportation into account, it can be seen that while 7.3 kg CO_2 eq. recorded for the mixture made of 100% CDW, this value ranged between 19 and 40 kg CO_2 eq. levels for mixtures containing industrial wastes. In all geopolymer paste mixtures, the main contributor to the ozone depletion impact category was NaOH with values ranged from 85% to 89%. The primary factor contributing to ozone depletion potential (ODP) of the mixtures can be related to the use of carbon tetrachloride to recover chlorine from gas streams in the chlor-alkali process to produce NaOH [79]. It was followed by $\text{Ca}(\text{OH})_2$ with values of approximately 7% for all mixtures. Electricity values ranged from 0.9 to 1.0%. As in the GWP, the effect of transportation on ODP values were seen as the determining factor, which resulted in the differences between the mixtures (e.g., 1% for 100CDW, 7% for 80CDW20BFS). Similar trend with GWP were recorded for the industrial waste incorporation on the ODP results. In the acidification category, NaOH has the highest impact with values between 60 and 64%. $\text{Ca}(\text{OH})_2$ followed with values between 14 and 15%, while the electrical energy consumed by the devices was also an important factor with values between 12 and 14%. The eutrophication impact category is related to the potential for excessive nutrient loads, such as nitrogen and phosphorus, to cause environmental problems like harmful algal blooms in bodies of water. In this category, NaOH has the highest impact with values in the range of 51–68%, followed by electricity with values in the range of 14–20%. Although in the other impact categories, the influences of industrial wastes on the paste mixtures were negligible and very close to each other, this difference was greater in the eutrophication category as FA showed high influences on eutrophication potential. The higher eutrophication impact of FA compared to SF and BFS could be due to its higher content of phosphorus and/or nitrogen as it is produced from the combustion of coal. In the photochemical oxidation impact category, NaOH and $\text{Ca}(\text{OH})_2$ were found to have almost similar levels of impact, with values in the range of 38–45%. Electricity is the third most impacting category with values ranging between 7 and 9%, followed by transportation with values between 2 and 9%.

In general, NaOH, $\text{Ca}(\text{OH})_2$, transportation and electricity are the components that have the greatest effect on the impact categories, while water, CDW treatment and industrial wastes (BFS, FA and SF) are almost negligible. It was observed that alkaline activators, especially NaOH,

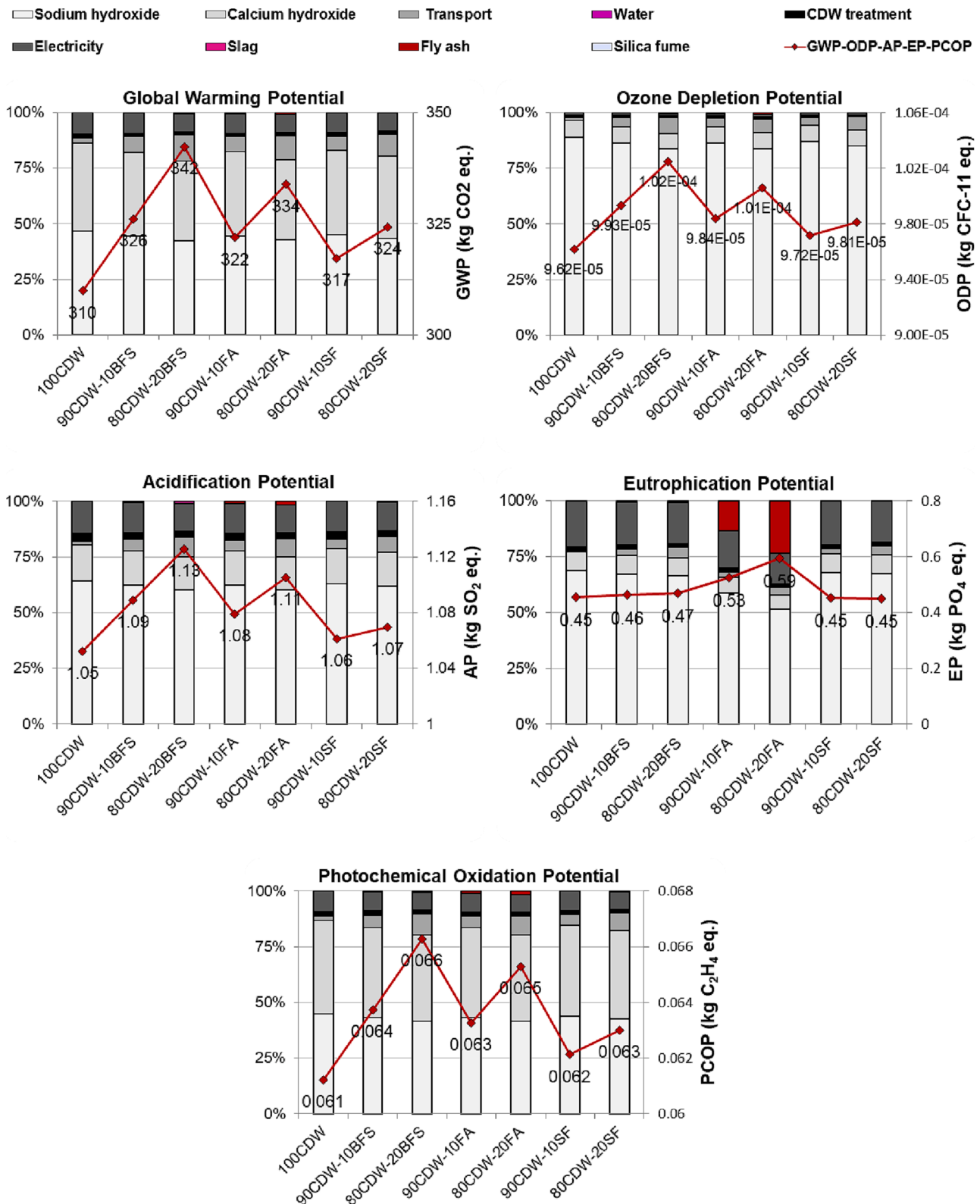


Fig. 7. Distribution of the input variables for the various impact categories.

had the greatest effect, while transportation was the most decisive factor on the results of BFS, FA and SF included mixtures compared to 100CDW coded mixture. Depending on the locations where these industrial wastes were obtained, as the transportation values increased, the impact values increased accordingly. Although the best mixture in terms of environmental performance was 100% CDW-based paste (except

eutrophication), industrial waste-incorporated mixtures were very close with it, and if the transportation values can be reduced, even better results may be obtained compared to the mixture containing entirely CDW-based materials. For all impact categories, the mixture made of 100% CDW containing higher levels of CDW-based materials, had higher emissions in terms of electricity values compared to the mixtures

containing BFS, SF and FA. This result can be attributed to that more electricity was consumed in the crusher and grinder devices used for precursor preparation with CDW-based materials including HB, RCB and RT. According to the findings, it can be said that the 100% CDW-based geopolymer paste exhibited lower environmental impacts, except for eutrophication. These outputs revealed that the geopolymer system containing entirely CDW was more advantageous in terms of environmental impact, but the use of FA, BFS and SF followed very closely behind.

The results obtained showed that the inclusion of industrial waste-based precursors resulted in the enhancement of engineering properties of CDW-based geopolymer mixtures. However, the environmental burden of developed CDW-based geopolymer mixture was increased at the same time with the inclusion of those wastes because of mainly procurement-related (transportation) energy consumption. But in this point, it should be considered that if those mixtures have similar engineering properties compared to 100% CDW-based geopolymer mixture, it is believed to observe similar environmental burden. The utilization of industrial waste-based precursors allows for a reduction in water usage within the matrix. This is because the inclusion of these waste materials has been found to enhance the workability performance. As a result, an improvement in the mechanical performance of the mixture becomes apparent since the presence of excess free water, which contributes to weakness, can be effectively minimized. Besides that, the compressive strengths of industrial waste included mixtures (especially for BFS and SF included) were already higher compared to the non-included one. Therefore, the use alkaline activators can be mitigated to adjust the mechanical performance of the mixture similar to the 100 %CDW-based geopolymer. In this way, the environmental burden of industrial waste-based geopolymer can be reduced and be similar to the 100% CDW-based geopolymer mixture. When considering environmental impact per MPa (in terms of compressive strength), the addition of industrial wastes was very beneficial as the strength of the mixtures enhanced remarkably while the environmental impacts depended on mainly transportation and were not significant. However, the wider availability of CDWs compared to industrial wastes bring environmental advantages considering the transportation-related impacts. Overall, results obtained suggest that industrial waste-based precursors can be successfully used in CDW-based geopolymer matrix to adjust engineering properties without endangering the environmental burden of the mixture.

5. Conclusion

This study aimed to investigate the compatibility of incorporation of industrial waste-based precursors including BFS, FA, and SF into the CDW-based geopolymer mixtures by analyzing some engineering properties and environmental impacts. To do this, a series of geopolymer mixtures were produced and their fresh and hardened property assessment was performed. In addition to this, a comprehensive LCA analysis was also performed to examine environmental burdens of the mixtures containing CDW and industrial waste in terms of different impact categories including global warming potential (GWP), ozone depletion potential (ODP), acidification potential (AP), eutrophication potential (EP) and photochemical oxidation Potential (PCOP). The following conclusions have been drawn from the experimental and assessment studies performed within the scope of the current research:

- Inclusion of industrial waste-based precursors into the CDW-based geopolymer mixtures yielded an increment in the flowability of the mixtures and provided longer open time performance. While the FA was the most effective precursor in terms of enhancement of workability, SF incorporation resulted in relatively lower enhancement among industrial wastes. However, SF incorporation caused more decrement in shear yield stress compared to the others.
- Industrial waste containing mixtures exhibited higher mechanical performance compared to the 100% CDW-based geopolymer

mixtures, regardless of type and substitution rate of industrial wastes. Among industrial waste substituted mixtures, the compressive strength results of SF-incorporated ones were the highest, while those of FA-incorporated ones were the lowest.

- Although 100% CDW-based geopolymer system had less environmental burden, the mixture using FA, BFS and SF followed very closely behind. The main distinct parameters on the LCA results were raised from the transportation related impacts. While influence of water, CDW treatment and BFS, FA and SF on the environmental impact categories were almost negligible, NaOH, Ca(OH)₂, transportation and electricity were the components that have the greatest effects.
- The promising results were obtained from the industrial waste-based precursors by using them in CDW-based geopolymer providing enhancement of engineering properties (e.g., better workability and enhanced compressive strength) without endangering the environmental burden of the mixture which can be used further in the studies related with advanced manufacturing technologies.

CRedit authorship contribution statement

Huseyin Ilcan: Conceptualization, Writing – original draft. **Oguzhan Sahin:** Conceptualization, Writing – original draft, Methodology. **Zeynep Unsal:** Writing – original draft, Investigation. **Emircan Ozcelikci:** Writing – original draft, Visualization. **Anil Kul:** Writing – original draft, Investigation. **Nazım Çağatay Demiral:** . **Mehmet Ozkan Ekinci:** . **Mustafa Sahmaran:** Conceptualization, Writing – original draft.

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

No data was used for the research described in the article.

Acknowledgement

The authors gratefully acknowledge the financial assistance of the Scientific and Technical Research Council (TUBITAK) of Turkey provided under Project: 119 M630 and 119 N030. This publication is a part of doctoral dissertation work by the first author in the Academic Program of Civil Engineering, Institute of Science, Hacettepe University.

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