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Sustainability evaluation of alkali-activated mortars incorporating industrial wastes

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ABSTRACT

In this study, the alkali activated mortars (AAMs) were synthesized from waste ceramic tile powder (WCP), ground blast furnace slag (GBFS) and fly ash (FA). The effects of binder composition on the sustainability of AAMs were investigated. GBFS was replaced by FA at different percentages of 10%, 20%, 30%, and 40%. In contrast, the WCP content of the binders was kept at 50% in all AAMs. Engineering properties like compressive strength, drying shrinkage and sulfuric acid resistance were evaluated in this study. From the study, the compressive strength of proposed mortar was found to be decreased with a higher content of FA in the alkali-activated matrix. Besides, AAMs with a higher content of FA demonstrated a lower drying shrinkage and better performance under sulfuric acid environments. Other than enhanced durability properties, a lower energy consumption, smaller carbon dioxide emissions, and cheaper production cost of proposed mortars were shown when GBFS was replaced by FA.
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1. Introduction

Alkali-activated binder is an effective alternative for traditional Portland cement. It not only saves energy and reduces carbon dioxide (CO_2) emissions, but also enhances the durability performance of concrete and resolves landfill issues. It is well-recognized that the ceramic industry generated a large amount of calcined clay waste every year and a large amount of the clay wastes were mainly disposed of in the landfills. Instead of being sent to the landfills, such wastes should be reused appropriately and efficiently. Lately, the research areas of alkali-activated concrete and mortars have been progressed substantially and novel concrete and mortar products with far less CO_2 emission and more environmentally friendly characteristics have been developed [1–4]. It has also recently attracted the interests of many academic experts and industrial specialists. Conversely, concerns have been growing over limited landfill areas in many developing countries like Malaysia due to a thorough understanding of the environmental issues. As waste management costs rising, industrial manufacturers are seeking cost-effective alternatives for such waste. In the literature, few recent works have successfully demonstrated the production of concrete materials from ceramic wastes for the construction industry. However, many researchers have evaluated the robustness and durability properties of the ceramic materials as a binder agent or fine and coarse aggregates, which partially replaced the environmentally unfriendly OPC [5–9].

Furthermore, many research works have been attempted to replace the OPC with a more sustainable alkali-activated material as an effort to mitigate the CO_2 emissions. However, the high production cost and adverse environmental impacts of such materials have hindered its commercial production and potential market penetration. Thus, this study attempted to evaluate the effect of alkali-activated ternary blended composite binder on different sustainability factors like production cost, CO_2 emission, durability properties, and energy usage. Besides, the flexibility of AAMs was also studied extensively by replacing the FA with several loadings of GBFS, and the resulting products were activated with a low concentrated alkaline solution. During the production process of

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AAMs, ambient heat is typically only applied to cure and precast members, and the usage of such heat can theoretically be expanded to more areas, which subsequently reduces the energy consumption and production costs. As a result, this study proposed different compositions of AAMs that could be effective under extreme environments with the consideration of CO_2 emissions, energy usage, and manufacturing costs. Lastly, this study also analyzed the compressive strength and sulfuric acid (H_2SO_4) attack resistance of the resulting AAMs products.

2. Methodology

2.1. Materials

The waste ceramic tiles were sourced from a local industrial ceramic manufacturer, White Horse from Pasir Gudang, Johor Malaysia. All ceramic tiles collected possessed the same thickness without the coating of a glassy protective layer. A jaw crusher was used to crush the ceramic tiles before passed through a 600 μm sieve for the removal of coarse particles. A Los Angeles abrasion machine was used to ground the sieve particles for 6 h. It consists of 20 stainless steel balls and each ball has a diameter of 40 mm. The resulting product was known as WCP. Fig. 1 shows the production of WCP. For the pure GBFS, it was collected from a supplier who was located at Ipoh, Malaysia, and was used as received without any pre-treatment. Also, the FA materials with low calcium content were collected from the Tanjung bin power station and used an aluminosilicate materials for the synthesis of AAMs. It was observed that the colors of FA, WCP, and GBFS were grey, light grey, and white, respectively. Table 1 shows the physical properties of WCP, GBFS, and FA. From Table 1, FA had a lower specific gravity of 2.2 than WCP (2.6) and GBFS (2.9). Moreover, WCP, GBFS, and FA possessed medium particle sizes of 35, 12.8, 10 μm, respectively.

The chemical compositions of FA, WCP, and GBFS were evaluated by using X-ray fluorescence spectroscopy (XRF). From the results, it was revealed that each material exhibited different characteristics in accord with the chemical composition. For example, the major chemical constituents of WCP and FA was SiO_2, with the respective content percentage of 72.6 and 57.8%. On the contrary, the main chemical compound of GBFS was CaO with a content percentage of 51.8%. As compared to GBFS, both WCP and FA possessed a lower CaO content of 0.02 and 5.2%, respectively. On the contrary, FA offered a higher Al_2O_3 content of 28.8% than WCP (12.6%) and GBFS (10.9%). It is well-known that the production of C-(A)-S-H gels during the hydration process requires crucial oxide components like SiO_2, Al_2O_3, and CaO. Thus, the blending of WCP and materials enrich in Al_2O_3 and CaO is essential because it allows the production of an alkali-activated concrete and mortars products with superior properties [10].

Table 1
WCP, GBFS and FA chemical and physical characterizations.

Material	GBFS	FA	WCP
Physical traits			
Medium particle size (μm)	12.8	10	35
Specific gravity	2.9	2.2	2.6
Chemical composition (% by mass)			
SiO_2	30.8	57.20	72.6
Al_2O_3	10.9	28.81	12.6
CaO	51.8	5.16	0.02
Others (Fe_2O_3, K_2O, Na_2O, MgO)	6.28	8.71	14.65
Loss on ignition (LOI)	0.22	0.12	0.13

More calcium (aluminum) silicate hydrate gels will be formed from the pozzolanic reaction due to a higher calcium content in GBFS and higher aluminum content in FA. Besides, both WCP and FA are pozzolan in the class F category because the total percentage of SiO_2, Al_2O_3, and Fe_2O_3 is more than 70% according to ASTM C618-15.

The sodium silicate solution (NS) used in this study composed of 29.5 wt% SiO_2, 14.70 wt% Na_2O, and 55.80 wt% H_2O. Whereas, the sodium hydroxide pellets (NH) were obtained in analytical grade with the purity of 98%. The NH solution was prepared at a concentration of 4 M by dissolving the sodium hydroxide pellets into the water. Besides, an alkaline solution with a SiO_2:Na_2O ratio of 1:02 was prepared when the dissolved solvent was mixed with the NS solution after cooled for 24 h [11]. In this study, a NS: NH ratio of 0.75 was kept constant in all alkaline solutions to reduce the adverse environmental impacts of sodium silicate (Na_2SiO_3) [12]. In addition, sieve analysis was performed according to the standard procedures of ASTM C33-16 by using sands obtained from the local rivers. The bulk density, specific gravity, and fineness

Fig. 1. Process of preparation of WCP in the lab.

modulus of the sand particles were determined as 1614 kg/m^3, 2.6, and 2.8, respectively. Furthermore, it was found out that all sand particles were successfully passed through a 2.36 mm sieve.

2.2. Mix design and test procedure

From Table 2, all five mixtures were prepared to study the effects of GBFS on the durability properties of the WCP-based alkali-activated cements. To ensure consistency and continuity throughout the experiments, the molarity of NH and WCP binder mass were maintained at 4 M and 50%, respectively. Furthermore, the ratio of binder agent to fine aggregates was kept at 1 whereas the ratio of alkaline solution to binder was kept at 0.40. Besides, the ratio of NS: NH and the modulus value of the solution were maintained at 0.75 and 1.02, respectively. The mixture with 50% GBFS was initially served as the control specimen and the mass ratio of GBFS and FA in other mixtures were varied consecutively in the following increasing order of 10%, 20%, 30%, and 40%. In a typical experiment run, a weighted solution was prepared and cooled to room temperature after both NH and NS were mixed [13]. Then, the fine aggregates were added into a mortar mixed machine to mix the AAMs. After that, all binder agents were subsequently loaded and mixed for 2 min under the dry condition to promote homogeneity before performing the activation process using the alkaline solvent. After further 4 min, the resulting mixture was poured into the molds with two layers according to ASTM C579-18 before it was placed on a vibration table for 15 s to remove any trapped air bubbles [14]. By considering the Malaysia climate, all AAM specimens were kept under the storing temperature of 27 °C ± 1.5 and relative humidity of 75% for 24 h to allow the curing process. Finally, the specimens were left in the same conditions until the testing day.

By adding the H_2SO_4 into mortar, it helped to disintegrate the binder paste solution and weaken the structure of AAMs. 10% H_2SO_4 acid solution was prepared from deionized water and the effects of 10% H_2SO_4 acid solution on the composition of AAMs were investigated. After 28 days, a total of 6 specimens were weighted for each AAMs before each specimen was immersed into the alkaline solvent for 365 days. In order to maintain a constant pH level throughout the experiment, the solution was changed periodically once every 2 months. After 180 days and 365 days, the AAMs specimens were collected for further analysis. Based on the standard specifications of ASTM C267, several factors like surface change weight loss, and residual strength were considered in evaluating the durability performance of AAMs [15]. The effect of FA content on drying shrinkage performance of proposed mortar were evaluated following the ASTM C157.

2.3. Environmental benefits

Besides, the production cost, energy usage, and CO_2 emission were calculated based on the life cycle of each material used. Table 3 presents the life cycle of FA, GBFS, and WCP. In this study, the collection cost of all three materials were assumed to be zero Ringgit Malaysia (RM) because the materials were considered as wastes. However, the transportation distance of each material was included for life cycle assessment. Other than the transportation distance, other factors like production cost, diesel consumption, type of truck engine, collection volume, traveling speed, charge of 1 tonne/m were kept the same for all materials. Besides, the production cost and electricity consumption were calculated based on the volume, function time, and power usage of each machine.

The total production cost, CO_2 emission, and energy consumption of all three materials are presented in Table 3. From Table 4, the electricity and diesel consumption during the preparation stages of GBFS were significantly higher than WCP and FA, which subsequently increased the production cost, CO_2 emission, and energy consumption. From the results, GBFS exhibited a higher energy consumption of 2.37 GJ/tonne as compared to WCP (1.11 GJ/tonne) and FA (0.17 GJ/tonne). A similar trend was also observed in the CO_2 emission and production cost, where the production process of GBFS generated the highest CO_2 emission of 0.152 tonnes/tonne than WCP (0.045 tonnes/tonne) and FA (0.012 tonnes/tonne). Likewise, GBFS exhibited a higher production cost of 449 RM/tonne than WCP (169 RM/tonne) and FA (34 RM/tonne) due to long-distance transportation. From the results, it is evident that the essential sustainability factors like CO_2 emission, production cost, and energy usage can be improved signifi-

Table 4
WCP, GBFS and FA greenhouse, energy and cost necessitate [16]

Materials	WCP	GBFS	FA
CO_2 emission (t/t)	0.045	0.152	0.012
Energy (GJ/t)	1.113	2.379	0.173
Cost (RM/t)	169.02	449.74	34.35

Table 2
AAMs mix design prepared with various level of GBFS as FA replacement.

Mix	Binder composition (kg/m^3)				Binder to Sand, %	River sand(kg/m^3)	Alkaline activator solution (kg/m^3)	
	WCP:GBFS:FA	WCP	GBFS	FA			Na_2SiO_3	NaOH
$AAMs_1$	50:50:0	550	550	0	1.0	1100	188.6	251.4
$AAMs_2$	50:40:10		440	110				
$AAMs_3$	50:30:20		330	220				
$AAMs_4$	50:20:30		220	330				
$AAMs_5$	50:10:40		110	440				

Table 3
WCP, GBFS and FA' preparation stages.

Materials	Type	Collection	Transport	Lab treatment		
				Crush	Sieve	Grind
WCP	Waste	√	√	√	√	√
GBFS	Waste	√	√	–	√	√
FA	Waste	√	√	–	–	–

cantly by incorporating a lesser GBFS content in the alkali-activated concretes.

3. Results and discussion

3.1. Compressive strength development (CS)

Fig. 2 illustrates the CS of different proposed mortar specimens at 1, 7, and 28 days. It was observed that all mortar specimens exhibited strength over the curing ages. The initial strength of the AMMs with 0% FA content was determined as 30.6%. In the first day, the initial strength of the specimen was found to be decreased when the GBFS was replaced by FA. When the FA content was increased further to 10%, 20%, 30%, and 40%, the strength of the AAMs specimens was found to be decreased to 28.2 MPa, 25.7 MPa, 22.5 MPa, and 17.8 MPa, respectively. All AAMs specimens with high FA contents exhibited similar trends at 7 and 28 days of aging. However, the strength of AAMs specimens with FA content of 10%, 20%, 30%, and 40%, were determined as 66.2 MPa, 60.2 MPa, 56.5 MPa, and 45.9 MPa, respectively. In contrast, the control sample had a greater strength of 72.1 MPa. The total CaO content was affected by the addition of FA content up to 40%. The development of C-(A)-S-H gels was consequently restricted and led to poor strength properties. Previous studies [17,18] have reported that a reduction in the formation of reaction products could decrease the strength of AAMs significantly. On the contrary, an increase in the Ca content in the AAMs could improve the strength properties of FA under the co-existence of C–S–H and C–A–S–H gels, and N–A–S–H gel [19–21]. On the other hand, AAMs with an acceptable strength of 45.9 MPa could be used for a few construction applications despite their high WCP (50%) and FA (40%) contents.

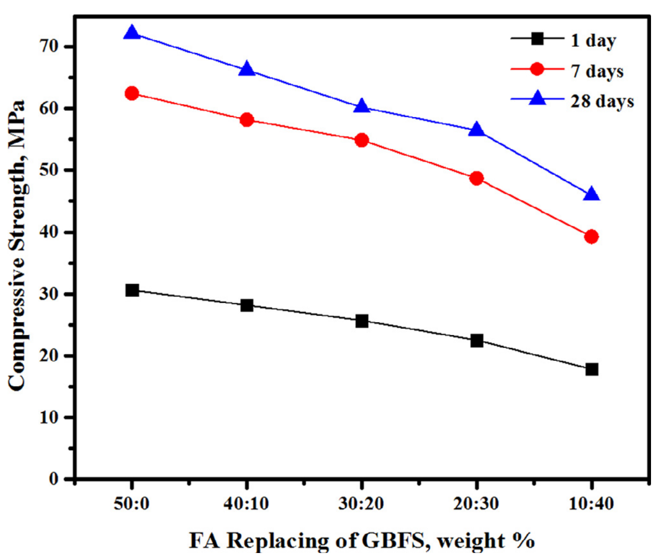

Fig. 2. Effect of FA and GBFS content on AAMs compressive strength development.

3.2. Sulfuric acid resistance

The resistance of AAMs toward sulfuric acid attack was evaluated in terms of strength loss, external cracks, and weight loss when FA was replaced by GBFS. Fig. 3 shows the residual strength of AAMs specimen after being immersed in a 10% H_2SO_4 solution at the age periods of 12 months. From the results, it was revealed that the strength loss of AAMs was directly associated with the amount of FA added. Furthermore, all AAMs exhibited a lower residual strength after the immersion time as compared to the control sample. When the FA content of AAMs specimen was increased from 0 to 10%, 20%, 30%, and 40%, the loss on strengths of the specimens after 12 months of exposure duration were found to be decreased from 32.1% to 23.8%, 14.2%, 13.8%, and 10.7%, respectively. As shown in Fig. 3, the weight loss percentage of the AMMs specimen was decreased from 2.1%, 0.86, 0.52, 0.39, and 0.32% when the FA content was increased from 0% to 10%, 20%, 30%, and 40%, respectively. From Fig. 4, an increase in FA content was observed to reduce the severity of surface deterioration and cracks. This can be explained by the formation of gypsum ($CaSO_4 \cdot 2H_2O$) via the reaction between the $Ca(OH)_2$ compound in the mortar and SO_4^{-2} ion when the AAMs were exposed to sulfuric acid, which leads to the expansion of the geopolymer matrix and formation of cracking within the specimen interiors as shown in Fig. 4. Few previous studies have also reported that the durability of AAMs was promoted by the presence of Na, Si, and Al contents, which subsequently reduced the formation of gypsum [22–26].

3.3. Drying shrinkage

The influence of FA content as GBFS replacement on drying shrinkage performance of AAM s were evaluated and presented in

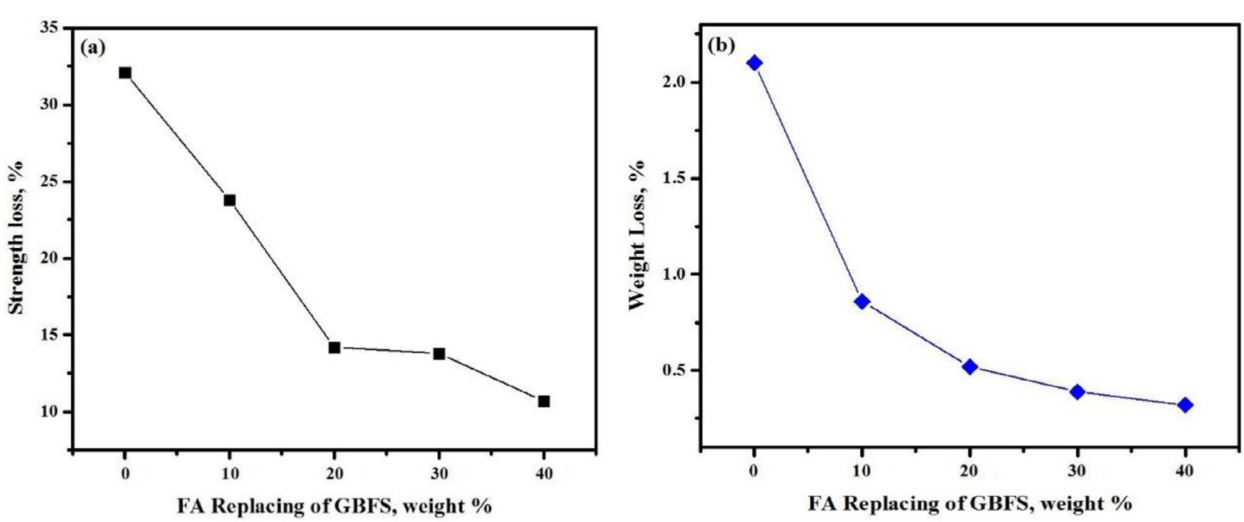

Fig. 3. Effect of sulfuric acid on AAMs' (a) strength loss and (b) weight loss.

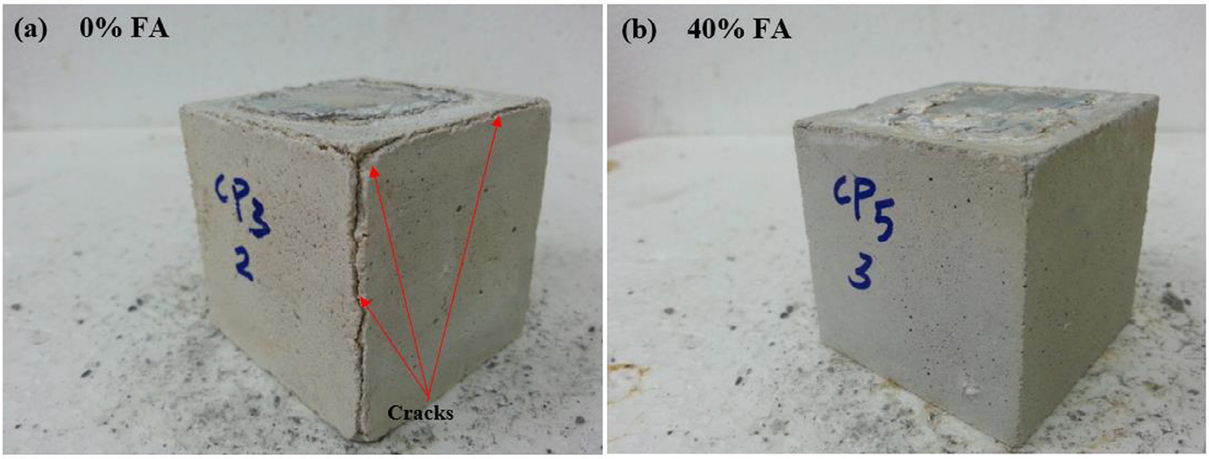

Fig. 4. Surface texture of mortar specimens exposed to 10% H_2SO_4 (a) 0% FA (b) 40% FA.

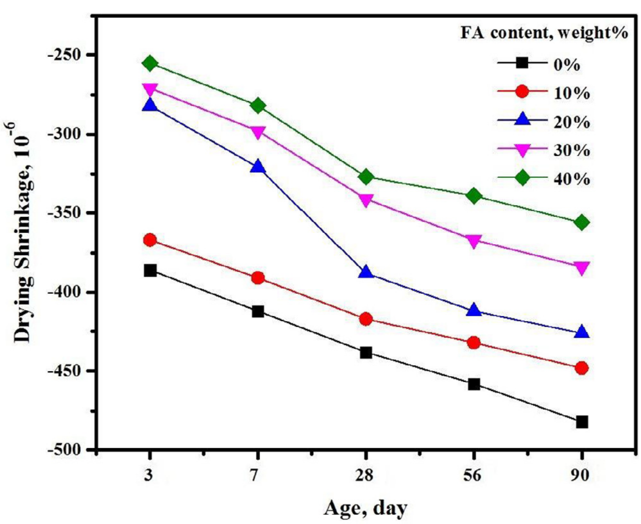

Fig. 5. Effect of FA content on drying shrinkage of AAMs.

Fig. 5. It was found the inclusion FA as part from binder composition of proposed mortars impact positively on specimens' durability and reduce the drying shrinkage at early and late ages. The change in length trend to decrease with increasing content of FA as GBFS replacement. At early age (3 days), 386 microstrain to 367, 282, 271 and 255 microstrain with increasing content of FA as GBFS replacement from 0% to 10, 20, 30 and 40%, respectively. Similar trend of results was observed after 28 days of age and the value of drying shrinkage trend to decrease with increasing content of FA in the AAMs matrix. The increasing level of replacement GBFS by FA from 0% to 10, 20, 30 and 40% led to drop the drying shrinkage value from 438 microstrain to 417, 388, 341 and 327 microstrain, respectively. The enhancement on proposed mortar durability was attributed to the high content of FA affected to reduce the surface tension and reduce the total drying shrinkage for all ages compared to high content of GBFS. Results indicated that pore size distribution and characteristics of geopolymerization products are the critical factors affecting the shrinkage in AAMs [27].

3.4. Carbon dioxide emission

The production of cement and concrete is being explored by scientists in order to find more environmental-friendly method of producing materials that will do the same job. This has come out as people are becoming more aware of the harms to society that is caused by CO_2 emissions; a by-product of cement and concrete [28,29]. Fig. 6 depicts the effects of substituting GBFS with FA on the CO_2 emission of the ternary blended AAMs. From Fig. 6, the initial CO_2 emission of the pristine OPC was calculated to be 108.3 kg/m^3. The CO_2 emission was found to be declined to 92.9, 77.5, 62.1, and 46.8 kg/m^3 when the FA content in the AAMs matrix increased to 10%, 20%, 30%, and 40%, respectively. The new ternary binders like WCP, GBFS, and FA exhibited a much lesser CO_2 emission than the conventional binder (OPC) for all alkali-activated matrixes with more than 75% reduction in the CO_2 emission achieved. This finding was rather significant as the production of 1 m^3 OPC usually produces 421 kg/m^3 CO_2 emission. With a lower CO_2 emission from the alkali-activated binders, a simple sustainability comparison can be made between the OPC and alkali-activated binders.

3.5. Cost effective and energy saving

Fig. 7 illustrates the effect of substituting the GBFS with FA on the production cost of ternary blended composite AAMs and conventional OPC binder. From Fig. 7, it was revealed that the production costs can be lowered down by replacing 40% of the GBFS with FA. Based on the life cycle as shown in Table 3, the ultimate production cost of the ternary blended composite AAMs was affected

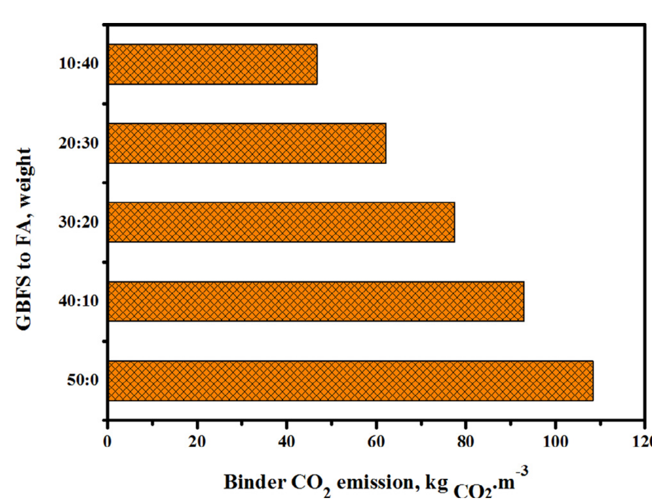

Fig. 6. Carbon dioxide emission of proposed mortars.

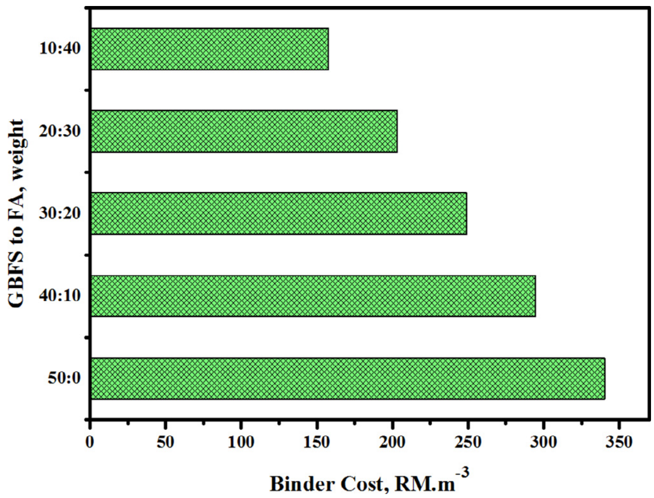

Fig. 7. Influence of AAMs' cost by content of FA as GBFS replacement.

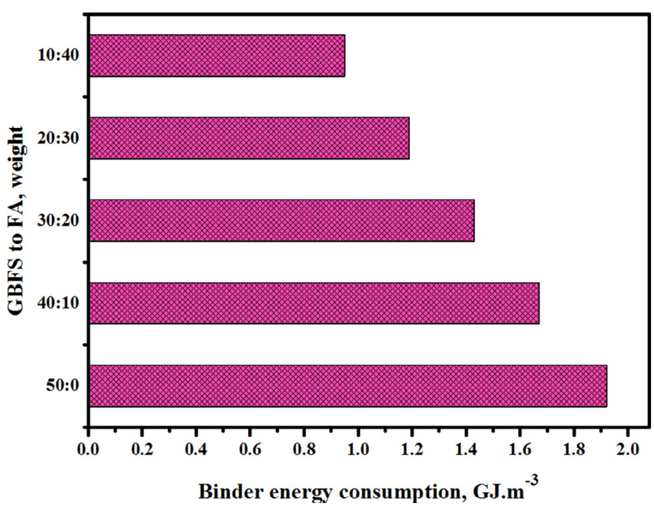

Fig. 8. Effect of FA content on energy saving of proposed mortars.

directly by the material costs by weight. From the results, the binder cost was found to be decreased from 340 RM/m^3 to 295 RM/m^3, 249 RM/m^3, 203 RM/m^3, and 157 RM/m^3 when the FA content was increased 0% to 10, 20, 30 and 40%, respectively. Furthermore, only mixtures with FA content higher than 20% demonstrated a lower production cost than the typical OPC cost of 275 RM/m3. Thus, it is showed that FA could be a sustainable binder alternative for GBFS.

Fig. 8 shows the total energy usage of each specimen. The energy usage was calculated from the life cycle and energy usage of each material used. From Fig. 8, the energy consumption of all alkali-activated AAMs was found to be decreased from 1.92 J/m3 to 1.67, 1.43, 1.19, and 0.95 GJ/m^3 when the FA content increased from 0% to 10, 20, 30, and 40%, respectively. Furthermore, a lower energy usage of 2.36 GJ/m^3 was obtained from all alkali-activated mixtures as compared to the pristine OPC. The low energy usage of AAMs can be attributed to the cheap diesel cost and low electricity usage. Overall, the production cost, CO_2 emission, and energy consumption are some of the main factors that could affect the sustainability of an alkali-activated binder.

4. Conclusions

With the objectives outlined in this study, the following conclusions are:

(1) Addition of FA can affect the development of compressive strength in the binders and a higher FA content generally tends to reduce the compressive strength of the WCP based AAMs. It was found out that AAMs with 40% FA content achieved a relatively higher compressive strength of 45.9 MPa, which allowed it to be used in several construction applications.
(2) A high-performance WCP based AAMs can be produced by replacing the GBFS with FA and it can be used under an aggressive environment. From the residual strength and mass loss, the resistance of AAMs against acid attack was found to be improved with a higher FA content. As a whole, AAMs with 40% FA content demonstrated the highest acid attack resistance among all mixtures. Furthermore, the formation of gypsum can be restricted by the presence of a lower CaO content in FA during the immersion period.
(3) All AAMs with FA as the substituent exhibited lower drying shrinkage compared to control specimens.
(4) From this study, the AAMs were cost-effective and eco-friendly as compared to the conventional OPC mortars. Furthermore, the products were more efficient with lesser CO_2 emission, lower production costs, and lesser fuel consumption as compared to OPC binders.
(5) It is recommended to replace the GBFS with 40% FA content as it allows to decrease the AAMs cost (RM 157) by 42.9% as compared to the typical OPC binder cost of RM275. Thus, the new mortar product can be served as a cost-effective alternative for the traditional OPC. Furthermore, the new composition of the mortar product required less than half of the fuel consumption required by the OPC, which subsequently reduces the CO_2 emissions.
(6) Apart from the environmental impacts, the AAMs prepared from this study also demonstrated superior mechanical and durability properties, which can be served as an attractive theme to many industrial mortar and concrete manufacturers.

CRediT authorship contribution statement

Zahraa Hussein Joudah: Investigation, Writing of the original draft preparation. **Ghasan Fahim Huseien:** Methodology. **Mostafa Samadi:** Supervision, Writing - review & editing. **Nor Hasanah Abdul Shukor Lim:** Supervision, Writing - review & editing.

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.

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