DOI: https://doi.org/10.24425/amm.2024.150903

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A PREFATORY SUSTAINABILITY ASSESSMENT OF ONE-PART GEOPOLYMERS

This study investigated the sustainability aspect of the fly ash (FA)-based one-part geopolymers (OPGs) with various combinations and amount of alkali activators (AA). The three groups of OPG were: the M-OPG activated with solely sodium metasilicate-anhydrous (Na_2SiO_3), the MH-OPG with Na_2SiO_3 and sodium hydroxide (NaOH), and the MC-OPG with Na_2SiO_3 and sodium carbonate (Na_2CO_3). The compressive strength, embodied carbon, embodied carbon index, embodied energy and embodied energy index were identified. Test result shows that the M-OPG and MC-OPG with the AA/FA ratio of 0.20 attained 83 MPa and 75 MPa of compressive strengths, respectively. The MH-OPG with AA/FA ratio of 0.15 attained 72 MPa of compressive strength. The embodied carbon and embodied energy of the OPGs were mainly contributed by the Na_2SiO_3-anhydrous. The values were lowered when the Na_2SiO_3-anhydrous were partially substituted with NaOH or Na_2CO_3. Increasing the AA content increased the embodied carbon and energy. The embodied carbon index (3.80 kg CO_2/m^3/MPa) and embodied energy index (14.72 MJ/m^3/MPa) of the MC-OPG with AA/FA ratio of 0.20 were the lowest. The outcome of this study supports the utilisation of Na_2CO_3 to partially substitute Na_2SiO_3 for the development of OPGs.

Keywords: Geopolymer; embodied energy; embodied carbon; Fly ash, One-part

1. Introduction

Global urbanisation and economic development increase the demand for new buildings and infrastructure, which relies heavily on concrete. Today, concrete is the second most used resource on earth. The main ingredient of concrete is cement. Cement is produced through crushing, grinding and heating of limestone. Such approach accounts for 8% of global carbon dioxide emissions [1]. Recent advancement in building materials revealed the potential of geopolymer as a green alternative to the traditionally used cement. According to Neupane [2], the geopolymer emits around 5 times less CO_2 than Portland cement and consumes 3 times less energy. One-part geopolymer (OPG) is an inorganic polymer that possesses cementitious properties after treating the aluminosilicate waste materials in alkaline conditions. In addition to the significant reduction in environmental impact, the production of OPG is less complicated, which is by the "just-add-water" approach.

To response against climate change, sustainability assessment was greatly focused in recent research. Embodied carbon and embodied energy are the two most common indicators of the sustainability of building materials. Embodied carbon associates with the CO_2 emission while embodied energy indicates the amount of energy required for the production of the materials [3]. The problem arises when Ma et al. [4] discovered that the commonly-used alkali activator (AA), sodium metasilicate anhydrous (Na_2SiO_3-anhydrous) was the best-performing sodium silicates, but extremely harmful to the environmentally. The compressive strength of the slag-based OPG activated with Na_2SiO_3-anhydrous was the highest, which was 76.3 MPa. But among the various types of solid sodium metasilicates, the embodied carbon of Na_2SiO_3-anhydrous was 21.6% and 113.8% higher than Na_2SiO_3-pentahydrate and Na_2SiO_3-nanohydrate, respectively. To reduce the environmental impact of OPGs, the solid AAs could be altered. For example, sodium carbonate (Na_2CO_3) was applied in the development of slag-based OPG. It was found that increasing the share of Na_2CO_3 slightly reduced the compressive strength but significantly reduced the embodied carbon per unit volume per unit strength (from 4.79 to 3.57 kg/m^3/MPa), making the OPG a more sustainable material

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[5]. Other solid alkali activators, such as the sodium hydroxide, NaOH, yielded OPG with a compressive strength of 29.3 MPa [6], but the sustainability assessment was not carried out.

Fly ash is an industrial by-product, the embodied carbon and energy were minimal, the subsequent building materials would also be more sustainable [3]. Thus, this study aims to synthesise fly ash-based one-part geopolymers using the combinations of various solid alkaline activators, and then identify the extent of sustainability advancement achieved. For this purpose, three groups of fly ash-based OPGs were developed using varying amounts of Na_2SiO_3, NaOH and Na_2CO_3. The compressive strengths of the OPGs were identified and a simplified sustainability assessment based on the embodied carbon and energy was carried out.

2. Experimental method

2.1. Materials

The fly ash (FA) used in this study was obtained from the coal-fired power plant at Manjung, Perak, Malaysia. The chemical composition as obtained by X-ray fluorescence (XRF) is displayed in TABLE 1. The FA conforms with the requirement of Class C fly ash in ASTM C618-19. The FA was activated by three combinations of solid alkali activators (AAs). The solid AAs include sodium metasilicate-anhydrous (Na_2SiO_3-anhydrous), sodium hydroxide (NaOH) and sodium carbonate (Na_2CO_3). The Na_2SiO_3 – anhydrous of brand Alfa Aesar, in the form of 18 mesh granular was supplied by Fisher Scientific (M) Sdn. Bhd. The NaOH pellet with 97.0% purity was supplied by Progressive Scientific Pvt. Ltd. The Na_2CO_3 in powder form with 99.5% purity was acquired from Chemiz (M) Sdn. Bhd.

TABLE 1

Chemical composition of fly ash

Oxides	Weight percent (wt%)
SiO_2	36.7
CaO	19.1
Al_2O_3	18.7
Fe_2O_3	17.2
SO_3	3.04
K_2O	1.78
TiO_2	1.68
others	1.85

2.2. Sample Preparation

TABLE 2 lists the formula of the one-part geopolymers (OPGs). The samples were named in accordance with the solid AA involved and the solid alkali activator-to-fly ash (AA/FA) ratio. Three combinations of the solid AAs were prepared: Na_2SiO_3 only; Na_2SiO_3 + NaOH (Na_2SiO_3-to-NaOH ratio of 4.0); and Na_2SiO_3 + Na_2CO_3 (Na_2SiO_3-to-Na_2CO_3 ratio of 1.0). The AA/FA ratio was fixed at 0.15, 0.20 and 0.25 for all the combinations. The sample preparation began with the mixing of dry materials (FA + solid AAs) until homogeneous. Then, water was then added to the dry mix and mechanically mixed for 3 minutes to obtain geopolymer paste. The water content added into the mix varied from one combination to another due to the different water requirements. The water-to-dry materials (W/M) ratios for Na_2SiO_3-activated OPG (M-OPG), Na_2SiO_3 and NaOH-activated OPG (MH-OPG), as well as Na_2SiO_3 and Na_2CO_3-activated OPG (MC-OPG), were 0.25, 0.30 and 0.20, respectively. The fresh pastes were then cast into 50 mm^3 plastic moulds and compacted. The samples were wrapped with plastic sheets to prevent moisture loss. Samples were demoulded after 24 hours and aged for 28 days in ambient condition (30°C).

TABLE 2

Mix proportion of one-part geopolymers

Samples	AA/FA ratio	Alkali activators (AA)	Precursor
M0.15	0.15	Sodium metasilicate (Na_2SiO_3)	Fly ash (FA)
M0.20	0.20		
M0.25	0.25		
MH0.15	0.15	Sodium metasilicate (Na_2SiO_3) + sodium hydroxide (NaOH)	
MH0.20	0.20		
MH0.25	0.25		
MC0.15	0.15	Sodium metasilicate (Na_2SiO_3) + sodium carbonate (Na_2CO_3)	
MC0.20	0.20		
MC0.25	0.25		

2.3. Testing and analysis

After 28 days of ageing, three cube samples from each mixture were collected for compressive strength test using Universal Testing Machine (UTM) modelled Shimadzu UH-1000 kNI. The loading placement rate was 0.083 mm/s. The prefatory sustainability assessment of the OPGs was identified based on the embodied carbon and embodied energy. The sustainability appeal of the OPGs was made significant by calculating the embodied carbon index and embodied energy index using Equations 1 and 2, respectively.

$$\textit{Embodied carbon index}\ (\mathrm{kg\,CO_2 / m^3 / MPa}) = \frac{\textit{embodied carbon}\ \left(\mathrm{kg\,CO_2 / m^3}\right)}{\textit{compressive strength}\ (\mathrm{MPa})} \tag{1}$$

$$\textit{Embodied energy index}\left(\mathrm{MJ / m^3 / MPa}\right) = \frac{\textit{embodied energy}\ \left(\mathrm{MJ / m^3}\right)}{\textit{compressive strength}\ (\mathrm{MPa})} \tag{2}$$

3. Result and discussion

3.1. Compressive Strength

Referring to Fig. 1, the MH-OPG attained the highest compressive strength at the AA/FA ratio of 0.15, while the M-OPG and MC-OPG achieved the highest compressive strength at the AA/FA ratio of 0.20. The highest compressive strength was achieved by M0.15, which was 83.6 MPa, followed by MC0.20 with compressive strength of 75.1MPa. The lower compressive strength of the MC-OPG compared to the M-OPG was due to the lower alkalinity of the Na_2CO_3 [7], which discouraged the extensive geopolymerisation reaction. Geopolymerisation reaction was enhanced with the presence of NaOH, as a lower AA/FA ratio drove excellent compressive strength. For this reason, much geopolymer research utilised NaOH as one of the alkali activators [8]. Increment of the AA/FA ratio beyond 0.20 for M-OPG and MC-OPG, and 0.15 for MH-OPG caused compressive strength reduction. The increment of solid AAs leaves undissolved solid particles within the geopolymer matrix. Dissolution of the solid AAs during ageing through moisture ingression leaves voids in the structure, subsequently reduced the strength of the OPGs [9].

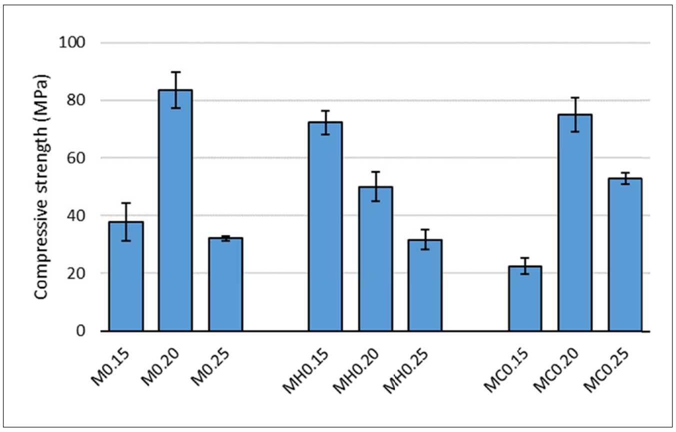

Fig. 1. Compressive strength of OPGs

3.2. Sustainability Assessment

The embodied carbon and energy of the raw materials used in this study are listed in TABLE 3. The contribution of embodied carbon and energy of fly ash was neglectable because fly ash was not purposely manufactured. Among the solid AAs, Na_2SiO_3 has the highest value of embodied carbon and embodied energy among raw materials. This is due to the manufacturing process of Na_2SiO_3-anhydrous involves the treatment of soda ash and silica sand at 1400°C. The process consumes a large amount of energy and releases a significant amount of greenhouse gases. The NaOH has the second highest embodied carbon and energy values, attributed to the production of NaOH which involved an electrolysis process. The Na_2CO_3 can be found naturally, thus the embodied carbon and energy are relatively lower [7].

TABLE 3

Embodied carbon and embodied energy of raw materials

Raw materials data	Embodied Carbon (kg CO_2/kg)	Embodied energy (MJ/kg)
Fly ash	0.01 [10]	0.10 [10]
Na_2SiO_3-anhydrous	1.86 [4]	5.37 [11]
NaOH	0.63 [12]	3.50 [12]
Na_2CO_3	0.11 [7]	1.35 [7]
H_2O	0.00 [10]	0.10 [10]

As can be seen from Fig. 2, the total embodied carbon of the MC-OPG mixes were the least, while M-OPG contained the most embodied carbon. The embodied carbon of MC0.25 was around 44% lower than that of the M0.25. The observation corresponds to the one-part alkali-activated material developed by Yang et al. [13] where the use of Na_2CO_3 yielded lower CO_2 emission compared to those activated using NaOH and Na_2SiO_3. Since the use of Na_2SiO_3-anhydrous was inevitable in the production of fly ash-based OPGs, the current accomplishment was considered fulfilling. Besides, the embodied carbon increased by approximately 50% when the AA/FA ratio increased from 0.15 to 0.25, regardless of the combinations of the alkali activators. The Na_2SiO_3-anhydrous was the main contributor of embodied carbon in OPGs, while the embodied carbon contribution from fly ash, NaOH Na_2CO_3 and water were relatively low. The Na_2SiO_3 contributes to about 97% of the total embodied carbon for all the M-OPG mixes. The embodied carbon contribution of Na_2SiO_3 in the MH-OPG and MC-OPG mixes was about 90%. Although fly ash was the main material used to synthesised the OPGs, the embodied carbon of fly ash in all OPGs were less than 5.5%

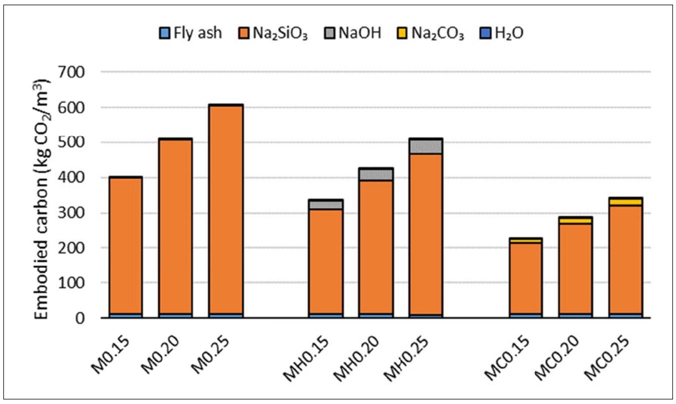

Fig. 2. Embodied carbon of OPGs

As shown in Fig. 3, the total embodied energy of these fly ash-based OPGs ranges from 908.7 to 1886.4 MJ/m^3. The embodied energy of these OPGs are significantly lower than the slag-based geopolymer (4690.42 MJ/m^3) due to the higher embodied energy of slag (1.60 MJ/kg) than fly ash [14]. The total embodied energy of the MC-OPG mixes were the least, while M-OPG contained the most embodied energy. Increasing the

AA/FA ratio from 0.15 to 0.25 increased the embodied energy of M-OPG, MH-OPG and MC-OPG by 45%, 44% and 42% respectively. So far, the Na_2SiO_3 was still the main contributor of embodied energy in OPGs. The embodied energy contribution of Na_2SiO_3 in the M-OPG, MH-OPG and MC-OPG mixes were about 89%, 75% and 67%, respectively. Generally, the reduction of AA content, as well as the substitution of Na_2SiO_3 with NaOH or Na_2CO_3, reduced the embodied carbon and energy, demonstrating significant benefits environmentally. However, the reduction of AA content might jeopardise the mechanical performances of the OPGs, thus it is advised to emphasise the embodied carbon and embodied energy of the samples with respect to the compressive strength.

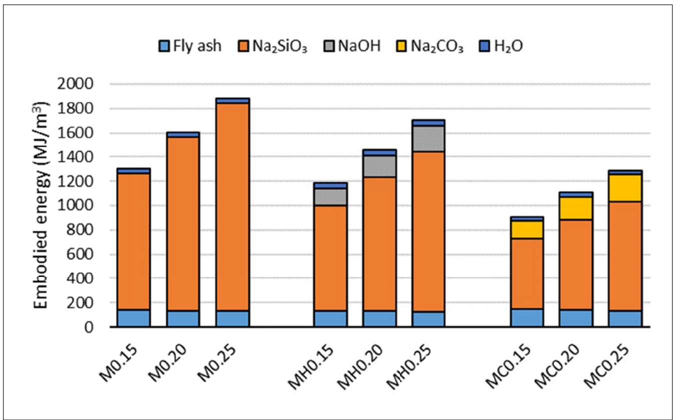

Fig. 3. Embodied energy of OPGs

TABLE 4 lists the calculated embodied carbon index and embodied energy index of the samples. The embodied carbon of M-OPG corresponds to lithium slag and blast furnace slag-blended one-part geopolymer activated by Na_2SiO_3-anhydrous (10-30 kg CO_2/m^3/MPa) [15]. The smaller the values, the more sustainable the materials are. Comparatively, the embodied carbon and embodied energy indexes of the OPGs were significantly lower than the commonly-used Portland cement products, which were 23.93 kg CO_2/m^3/MPa and 128.3 MJ/m^3/MPa, respectively [3]. Showing that the OPGs are a more sustainable construction material than the Portland cement. As indicated in the table, there were no clear correlation between the embodied carbon and embodied energy indices with the AA/FA ratio. In fact, these indices strongly correlate with the compressive strengths. Within the groups, samples M0.20, MH0.15 and MC0.20 that have the lowest embodied carbon and embodied energy indices were associated with high compressive strengths. Amongst the mixes, MC0.20 exhibited the lowest embodied carbon index (3.80 kg CO_2/m^3/MPa) and embodied energy index (14.72 MJ/m^3/MPa). The embodied carbon index and embodied energy index of the MC0.20 were 60% and 30% lower than M0.20, respectively. Similar to the study of Patrick et al. [16], the ECI of NaOH alkali-activated mortar was approximate 4 times higher than the Na_2CO_3 alkali-activated mortar. In addition to the reduction in embodied carbon and energy, the mechanical performance of the materials was not jeopardised. The sustainable assessment suggests that Na_2CO_3 is a potential alkali activator that can be used to partially substitute Na_2SiO_3 to produce a sustainable OPG.

TABLE 4

Embodied carbon and embodied energy indices

Samples	Embodied carbon index (kg CO_2/m^3/MPa)	Embodied energy index (MJ/m^3/MPa)
M0.15	10.60	34.47
M0.20	6.07	19.20
M0.25	18.87	58.75
MH0.15	4.63	16.35
MH0.20	8.49	29.08
MH0.25	16.03	53.93
MC0.15	10.03	40.32
MC0.20	3.80	14.72
MC0.25	6.44	24.41

4. Conclusion

This study focused on the sustainability aspect of the one-part geopolymers (OPGs) with various combinations and amount of solid alkali activators (AAs). The OPGs developed were the M-OPG (Na_2SiO_3 only), the MH-OPG (Na_2SiO_3 + NaOH), and MC-OPG (Na_2SiO_3 + Na_2CO_3). The alkali activator-to-fly ash (AA/FA) ratio were 0.15, 0.20 and 0.25 for all combinations. The following conclusion can be made based on the study.

- The 28-day compressive strengths for M-OPG with an AA/FA ratio of 0.20 reached 83 MPa. The MH-OPG wit AA/FA ratio of 0.15 and MC-OPG with AA/FA ratio of 0.20 caused minor strength reduction, with the highest compressive strength achieved were 72 MPa and 75 MPa, respectively.
- The embodied carbon and energy of the OPGs were mainly contributed by Na_2SiO_3-anhydrous.
- The MH-OPG and MC-OPG have generally less embodied carbon and energy compared with those of M-OPG, due to the lower embodied carbon and energy of NaOH and Na_2CO_3.
- Considering the compressive strength, the MC-OPG with an AA/FA ratio of 0.20 exhibited the lowest values of embodied carbon index (3.80 kg CO_2/m^3/MPa) and embodied energy index (14.72 MJ/m^3/MPa), signifying a high-performing and sustainable OPG.

Acknowledgements

The author would like to acknowledge the Ministry of Higher Education, Malaysia, for providing the support (FRGS/1/2020/TK01/UNIMAP/02/2).

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