

Mechanical Properties and Lifecycle Assessment of a Green Alkali-Activated Mortar Based on Biomass Wood Ash

Yiying DU^{1*}, Ina PUNDIENE², Jolanta PRANCKEVICIENE³,
Aleksandrs KORJAKINS⁴, Modestas KLIGYS⁵

^{1–3,5}*Laboratory of Concrete Technology, Institute of Building Materials, Vilnius Gediminas Technical University, Linkmenu St. 28, LT-08217 Vilnius, Lithuania*

⁴*Institute of Materials and Structures, Faculty of Civil and Mechanical Engineering, Riga Technical University, Kipsalas iela 6A, Riga, LV-1048, Latvia*

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Abstract – Portland cement (PC) is the most commonly used binder material for producing concrete. Nonetheless, increasing concerns have been attached to its manufacture which is highly energy-intensive and generates a large quantity of greenhouse gases. Developing alkali-activated materials as eco-binders is a sustainable replacement for PC and many investigations have been reported successfully utilizing industrial wastes as precursors. However, owing to the low reactivity, studies regarding biomass wood ashes (BWA) are still limited. To produce a green cementless alkali-activated mortar material, in this study, biomass fuel by-products – biomass wood bottom ash and biomass wood fly ash – were binarily used as precursors. Sodium hydroxide NaOH at 10 mol/L and calcium hydroxide Ca(OH)₂ at 20 % by binder mass were applied as alkali activators. Recycled sand, substituting natural sand, was adopted as fine aggregate with an aggregate/binder ratio of 2 to reduce the consumption of non-renewable natural resources. Compressive and flexural strength were tested to evaluate the mechanical performance. A cradle-to-gate lifecycle assessment was conducted to analyse the environmental impacts. The results reveal that the alkali-activated mortar has less environmental impact compared to the traditional PC mortar. NaOH solution is the primary source of environmental influence and BWA only contributes to very limited impacts. The usage of Ca(OH)₂ effectively improves the mechanical strength and compared to NaOH, it leads to decreased energy demand, requires fewer preparation steps and is less dangerous for operation.

Keywords – Calcium hydroxide; compressive strength; flexural strength; lifecycle assessment; recycled sand; sodium hydroxide.

1. INTRODUCTION

Nowadays, the building and construction sector is challenged by attempts to minimize the environmental influence and reduce the consumption of energy and non-renewable natural resources. As the traditional binder material, the production of Portland cement (PC) is not only highly energy intensive and consumes a large quantity of natural raw materials, but also it is responsible for the release of a great amount of greenhouse gas into the atmosphere, accounting for the environmental problem connected with global warming [1]. Every

* Corresponding author.
E-mail address: yiying.du@vilniustech.lt

production of 1000 kg of PC clinker results in 866 kg of carbon dioxide (CO₂) generation, corresponding to 5 to 8 % of the CO₂ emitted by mankind [2], [3]. Therefore, to improve sustainability in civil engineering, more eco-friendly low-carbon alternative binder materials are needed to replace PC. To aid in this effort, alkali-activated binder (AAB) materials have been developed, which are formulated by blending solid aluminosilicate powders with an alkaline solution [4]. According to Duxson *et al.* [5], the substitute of PC by AAB contributes to an estimated reduction of CO₂ footprint by 80 %. Additionally, the possibility of recycling and reuse of both industrial and agricultural by-products in producing AAM has also enhanced the sustainability of the material. This leads to growing research interests in AAB, especially in the current context of the concept of developing cementless eco-binder materials.

As effective aluminosilicate sources which are of high chemical reactivity, by-products, such as coal fly ash, ground granulated blast furnace slag, metakaolin and rice husk ash, have been widely investigated and applied in the field of AAB [6]–[9]. Unlike these traditional precursor materials, the utilization of biomass wood ash (BWA) is still rare and the study regarding it is at the beginning stage. Along with the growing popularity of biofuel energy as a clean renewable energy source [10], especially in northern European countries where forest resources are abundant, the waste management of BWA becomes an environmental problem. BWA is composed of organic and inorganic residues generated in the wood-burning process. Usually when wood is combusted, an estimated 6 to 10 % of the weight converts into ash [11]. There are approximately 4600 million tons of BWA generated globally every year and only 30 % of them are recycled for agricultural purposes [11], [12]. Most of the ashes are disposed of in landfills, which increases the cost of waste management and leads to concerns about the conservation of land resources [13]. Meanwhile, leaching of the hazardous elements from BWA can contaminate groundwater and fine particles can become airborne, causing potential health risks [14]. Comparatively, reusing BWA in building and construction materials is a more sustainable alternative.

The physicochemical characteristics of BWA significantly vary depending on the biomass source and combustion settings. Commonly, the ash particles have a porous structure and are more irregular and non-homogeneous than PC particles [15]. And the chemical reactivity of the ash is comparatively low [16], [17]. Thus, in most of the existing literature, due to the purpose of guaranteeing the mechanical properties, BWA is used as a supplementary binary precursor together with other materials. The partial addition of BWA can facilitate the degree of alkali activation and the secondary hydration of CaO source in BWA reacting with silicate and aluminate can increase the formation of calcium silicate hydrate (CSH) and calcium aluminium silicate hydrate (CASH), generating a dense binder structure [18]. Also, unreacted BWA particles can act as microfillers in the voids and pores, which enhances the densification of the microstructure of the binder [19]. This favours the strength development of AAB when BWA is used as a binary precursor [20]–[22]. BWA can be divided into two categories according to its biomass combustion procedure: biomass wood bottom ash (BWBA) which consists of fully and partially burnt materials discharged during the combustion, and biomass wood fly ash (BWFA) from the control system of air pollution [17]. BWFA have smaller particles and higher reactivity in contrast with BWBA [23]. Thus, there are more investigations regarding BWFA than BWBA in AAB materials [16], [20], [24], [25]. In some studies, BWBA is only used as aggregates to replace natural sand due to its limited cementitious characteristics [26]–[28]. To understand the possibility of single usage of BWA at a large ratio, especially when BWBA is utilized as a precursor in AAB, still more research is needed.

Another concern in manufacturing concrete is the consumption of natural aggregates that belong to non-renewable resources. Aggregates generally consist of 60 % to 75 % of the volume of concrete and approximately 48.3 billion tons of aggregates are consumed to produce concrete materials every year, which presses great pressure on the conservation of natural resources [29]. Furthermore, the process of mining, processing and transportation of such large quantities of natural aggregates involves abundant energy consumption and CO₂ emissions [30]. From this perspective, the reuse of recycled aggregates from biomass fuel plants as a substitute for natural sand is a sustainable attempt to improve the eco-efficiency of the material and can avoid the landfill dumping of waste.

Aligned with the demands of sustainable development and circular economy in civil engineering, a green BWA-based alkali-activated mortar (AAM) has been developed using industrial wastes from biomass fuel energy. Flexural and compressive strength were examined to analyse the mechanical properties of the produced mortar. A cradle-to-gate lifecycle assessment (LCA) was performed to evaluate the greenhouse gas emission and energy consumption of the mortar. The aim of the study is to investigate the influence of the content ratio of BWBA and BWFA, as well as the effects of Ca(OH)₂ introduction as a binary activator, on the mechanical behaviours and environmental impacts of AAM.

2. MATERIALS AND METHODOLOGY

2.1. Materials

In this study, industrial waste from biomass fuel was used to produce a sustainable AAM. Two types of BWA from pine tree combustion -biomass fly ash (BWFA) and biomass bottom ash (BWBA)- were collected in Vilnius, Lithuania and incorporated as precursor materials. The recycled sand (RS) generated during the combustion was reused to replace natural river sand as fine aggregates. Before the preparation of mortars, both BWFA and BWBA were subjected to ball mill equipment and ground into fine powder in order to enhance their chemical reactivity. Afterwards, BWBA powder was sieved through a 0.5 mm sieve to eliminate the unground impurities like stones. The RS passed through a 1.25 mm sieve to obtain the required particle size. The bulk density of BWBA and BWFA after processing is 1.03 and 0.68 g/cm³, respectively, and the main particle size diameter (D₅₀) is 56.62 and 43.26 µm. The specific surface area of BWBA is 978.68 m²/kg and that of BWFA is 493.27 m²/kg. RS has a bulk density of 1.23 g/cm³, a specific area of 38.40 m²/kg, and a D₅₀ value of 237.20 µm. Table 1 illustrates the main chemical composition of the raw waste. SiO₂ and CaO are the two primary oxides in all three wastes. Especially for BWBA and RS, SiO₂ takes up more than 50 % of the oxide proportions. Fig. 1 shows the micrographs of BWA. It can be seen that both ashes are of rough surfaces with unburnt wood residues, and the particles are irregular and non-homogeneous. NaOH and Ca(OH)₂ from Lerochem (Klaipeda, Lithuania) were utilized as alkali activators.

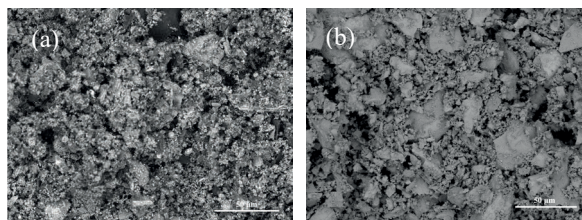


Fig. 1. Micrographs of BWA: (a) BWFA; (b) BWBA.

TABLE 1. CHEMICAL COMPOSITION OF RAW MATERIALS

Material	SiO ₂	CaO	Al ₂ O ₃	MgO	K ₂ O	Na ₂ O	Fe ₂ O ₃	MnO	P ₂ O ₅	SO ₃
BWBA, %	50.25	19.99	9.20	6.44	5.71	1.19	1.90	0.66	3.34	0.51
BWFA, %	22.91	31.50	2.63	3.57	4.00	0.26	2.32	2.07	2.71	0.88
RS, %	62.03	14.14	4.65	5.49	6.42	0.61	0.71	0.43	3.40	1.16

2.2. Preparation of Samples

The mix design is exhibited in Table 2. There are two groups including ten mixes of AAM samples experimented in this study. BWFA substituted BWBA at a ratio of 0, 25, 50, 75 and 100 % by binder weight as a binary precursor. The water/binder (w/b) ratio was 50 %. RS was utilized twice the mass of the binder as fine aggregate. NaOH solution at a concentration of 10 mol/L was used alone as an alkali activator in the first group. Ca(OH)₂ at a content of 20 % by binder weight was added as a binary alkali activator in the second group. NaOH solution was prepared in advance by dissolving NaOH powder into tap water. Ca(OH)₂ was used as dry powder. In Table 2, the mix ratio is presented by binder weight which is the mass sum of BWFA and BWBA. C represents the percentage of Ca(OH)₂ and F interprets the proportion of BWFA. For instance, C20F50 refers to the mix with 20 % Ca(OH)₂ and 50 % BWFA.

TABLE 2. MIX DESIGN

Group	Mix ID	BWFA, %	BWBA, %	RS, %	Ca(OH) ₂ , %	NaOH, mol/L	w/b ratio, %
1	C0F0	0	100	200	0	10	50
	C0F25	25	75	200	0	10	50
	C0F50	50	50	200	0	10	50
	C0F75	75	25	200	0	10	50
	C0F100	100	0	200	0	10	50
2	C20F0	0	100	200	20	10	50
	C20F25	25	75	200	20	10	50
	C20F50	50	50	200	20	10	50
	C20F75	75	25	200	20	10	50
	C20F100	100	0	200	20	10	50

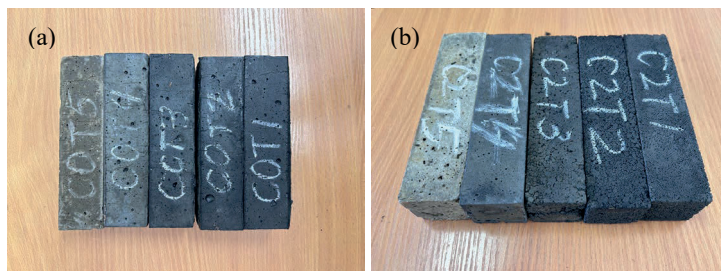


Fig. 2. AAM samples after 28-day curing: (a) group 1; (b) group 2.

The dry materials, BWBA, BWFA, RS and $\text{Ca}(\text{OH})_2$, were mixed at first and homogeneously stirred for 2 min at a low speed. Then the prepared NaOH solution was poured into the dry mixture and stirred for 1 min, followed by the addition of the extra water and the mixing for 2 min via the electronic mixer to homogenize the mixture. Afterwards, the mixture was poured into stainless steel prism moulds and subjected to 1 min of vibration on a shaker table to eliminate bubbles and achieve good compaction. The size of the mould was 160 mm × 40 mm × 40 mm. Then the moulded mortar was sealed via plastic film and cured for 24 hrs under 60 °C due to the consideration to minimalise the energy consumption. After 24 hrs, the hardened mortar was de-moulded, labelled, covered with plastic film and put at room temperature for 28-day curing. Fig. 2 shows the examples of samples after 28 days.

2.3. Testing Methods

After 28 days, the specimens were subjected to a flexural strength test on the WDW-20 universal testing apparatus. After that, the compressive strength test was performed on the CONTROLS 50-C56G2 strength testing apparatus. The particle size analysis of raw wastes was conducted on the Cilas 1090 LD apparatus and the chemical composition was tested on the AXIOS-MAX spectrometer.

2.4. Lifecycle Assessment

In order to evaluate and compare the environmental impacts of produced AAM mixes, a cradle-to-gate LCA was performed. Two impact assessment indicators were analysed, including the energy consumption during the production process via cumulative energy demand (CED) and the global warming potential (GWP) by the IPCC 2013 GWP 100a method. Modelling was constructed via OpenLCA software. The functional unit (FU) of the assessment was determined as the manufacture of 1 m³ of BWA-based AAM, as is commonly adopted in the LCA investigations regarding solid waste management [31], [32]. Fig. 3 depicts the system boundary of the study. The fabrication flow is composed of four main stages: (1) the recycling of raw waste; (2) the transportation of the raw waste to the production site; (3) the pre-processing of the raw waste to raw materials; (4) the fabrication of AAM products.

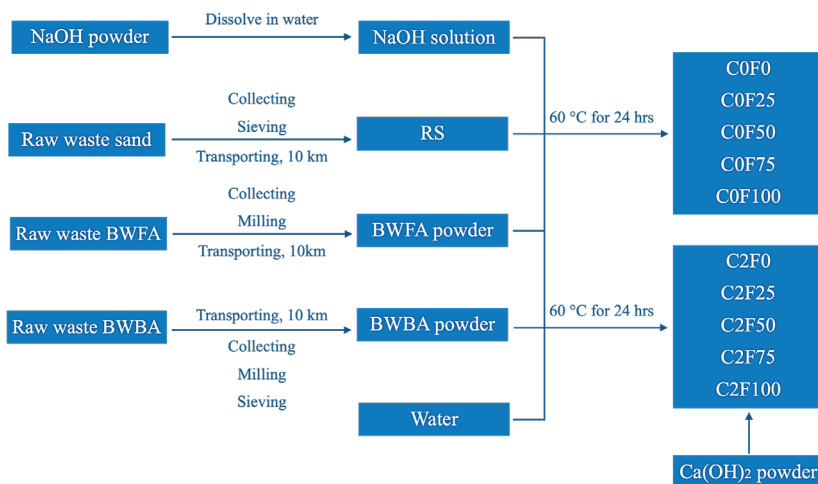


Fig. 3. Flow chart of the production system of AAM.

In this study, the lifecycle inventory (LCI) was mainly obtained from the *Ecoinvent* 3.01 database and those excluded in the database were acquired from relevant literature. The LCI is composed of transportation and pre-treatment of raw waste, preparation of NaOH solution, production of mortar mixture and temperature curing. BWA was considered pure waste and did not afford economic values, therefore, no flows were allocated to BWBA and BWFA from the biomass thermal plant [33], [34]. Only the environmental impacts of milling and sieving BWA were considered. Similarly, RS used in the study was also regarded as pure waste and merely its sieving procedure was taken into account in the assessment. The quantity of each type of raw material required to produce per FU of AAM was estimated based on the laboratory data. The average transportation distance from the biomass plant that produced raw BWA and sand waste to the mixing site of AAM was 10 km. The milling and sieving process, respectively, required an average electricity of 11.9 and 2.2 kWh/t [35], [36]. The temperature treatment of 60 °C for 24 hrs in the curing chamber consumed approximately the energy of 146 MJ/m³ [37].

3. RESULTS AND DISCUSSIONS

3.1. Compressive Strength

Fig. 4 illustrates the test results of the compressive strength of all the mixes with different BWFA and BWBA ratios and alkali activators at 28-day curing. When BWFA or BWBA were used alone as precursors, for NaOH-activating mortar, with 100 % BWBA, the compressive strength was only 1.0 MPa, whereas with 100 % BWFA, the strength was around four times greater (4.2 MPa). When the proportion of BWFA was higher than BWBA, C0F75 had larger compressive strength, compared to C0F25 with more BWBA. This can be attributed to the higher chemical reactivity of BWFA, which can be more effectively activated by alkaline solutions [23]. When NaOH and Ca(OH)₂ were used together as activators, the compressive strength of BWFA-based mortar was 4.1 MPa, similar to that of BWBA-based mortar. Mix with more BWBA showed better compression performance, which is connected with the extra addition of Ca(OH)₂. Regardless of the activator type, the highest compressive strength was achieved for the mix with 50 % BWFA and 50 % BWBA, with a value of 4.8 and 6.3 MPa, respectively. This is similar to the results of AAB materials with 100 % of BWA [16]. The addition of BWFA facilitates the activation degree and the filling effects of BWA refine the microstructure due to the difference in particle sizes of BWBA and BWFA, favouring strength development [20], [21].

Compared to the single use of NaOH, the extra usage of the Ca(OH)₂ activator improved the strength of AAM, as is proven in the existing studies [38], [39]. This can be related to the increment in hydration and polymerization generating more gels in the matrix [40]. The greatest enhancement of 320 % in strength value to 4.2 MPa was observed for samples with 100 % BWBA, and along with the increase of BWFA ratio in the mortar, the growth of the compressive strength demonstrated a downtrend. When 75 % of BWFA was added, by using Ca(OH)₂, the strength was only enhanced by 9.8 % from 4.1 to 4.5 MPa. And with 100 % BWFA, Ca(OH)₂ led to a slight strength decrease by 0.1 MPa. This indicates that Ca(OH)₂ exhibited more outstanding activating effects at a larger amount of BWBA incorporated in the precursor. This is aligned with the finer particles of BWFA which require more water in the binder system and its higher Ca content contributes to fast-setting, especially plus the addition of an extra 20 % Ca(OH)₂, adversely affecting the densification of AAM [21], [41].

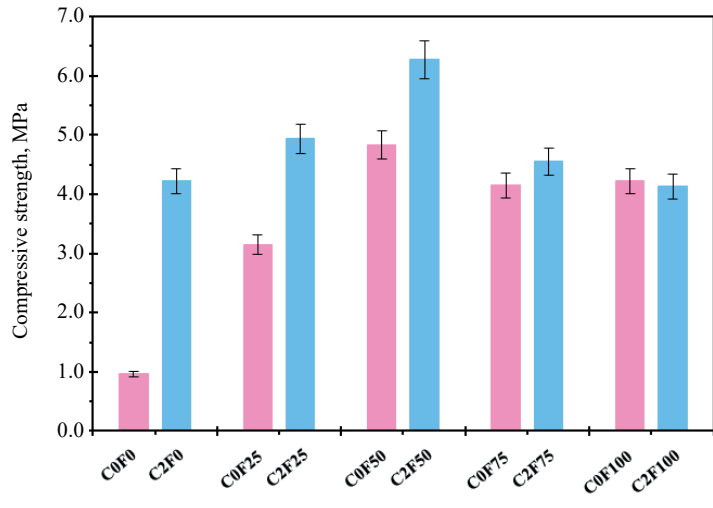


Fig. 4. Compressive strength of AAM.

TABLE 3. CHEMICAL COMPOUND RATIOS OF AAM SAMPLES

Ratio	C0F0	C0F25	C0F50	C0F75	C0F100	C2F0	C2F25	C2F50	C2F75	C2F100
CaO/SiO ₂	0.28	0.31	0.34	0.37	0.41	0.37	0.40	0.44	0.47	0.52
Al ₂ O ₃ /SiO ₂	0.11	0.10	0.09	0.09	0.08	0.11	0.10	0.09	0.09	0.08

In AAB materials, the content of CaO, Al₂O₃ and SiO₂ in the chemical composition significantly influences the hydraulic and pozzolanic activity of the material [16], [42]. Table 3 concluded the oxide ratios of AAM. Due to the higher CaO content in BWFA and greater Al₂O₃ amount in BWBA, along with the increment of BWFA proportion in AAM, the CaO/SiO₂ ratio increased, while the Al₂O₃/SiO₂ ratio was decreased. This indicates that the binary usage of BWFA at a proper dosage has positive effects on BWBA-based AAM and can enhance the degree of chemical reaction of the mortar during the activation process. The introduction of Ca(OH)₂ as a binary alkali activator improved the CaO/SiO₂ ratio of the mortar due to the extra source of CaO, enhancing the hydraulic activity of the material and further favouring the strength gain.

3.2. Flexural Strength

The flexural strength of AAM samples is depicted in Fig. 5. The strength evolution is observed similarly to the trends of compressive strength. The greatest flexural strength was measured at the mix ratio of 50 % BWFA and 50 % BWBA, with the values of 2.4 (activated by NaOH) and 2.7 MPa (activated by NaOH and Ca(OH)₂). For mortar containing only NaOH as the activator, the lowest strength was recorded for C0F0 with 100 % BWBA, which only reached 0.4 MPa. While C0F100 encompassing 100 % BWFA exhibited a 450 % higher strength at the value of 2.2 MPa. Also, the strength of samples with greater BWBA content overpassed those with more BWBA. This results from the increased alkali activation degree after incorporating BWFA [21]. For AAM binarily activated by NaOH and Ca(OH)₂, C2F0 with 100 % BWFA presented the same strength value (1.8 MPa) as C2F100 with 100 % BWBA. And the 75 % BWFA content accounted for a lower strength of 1.9 MPa, decreasing by 24 %, in contrast with 75 % BWBA usage.

The introduction of $\text{Ca}(\text{OH})_2$ as a binary activator importantly enhanced the flexural strength of AAM samples which contained 100 %, 75 % and 50 % of BWBA, by a percentage of 140 %, 90 % and 30 %, respectively. The extra source of Ca introduced by $\text{Ca}(\text{OH})_2$ promotes the generation of hydrates, which greatly contributes to the strength gain [43]. However, for AAM including 75 and 100 % BWFA, the flexural strength, on the contrary, declined by 20.8 and 18.2 %, accordingly. This indicates that $\text{Ca}(\text{OH})_2$ exert more positive effects on AAM when BWBA takes up the majority content. This can be due to the decreased alkalinity which fails to meet the reaction of BWFA in the binder system, hindering the strength development [43].

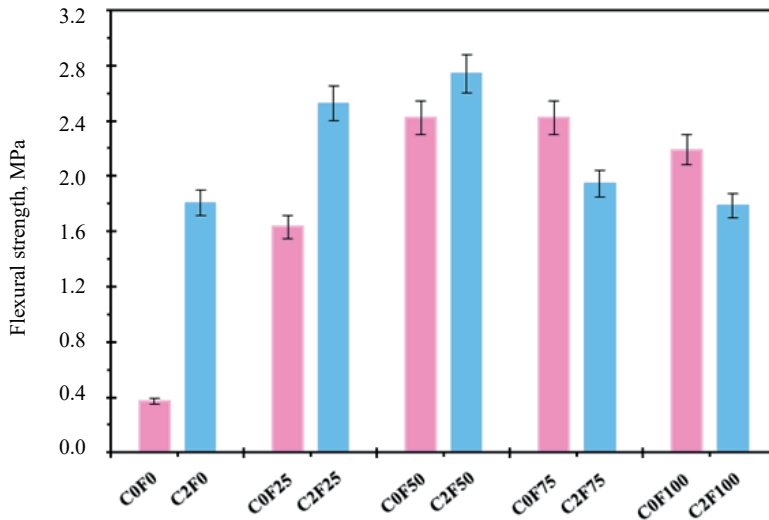


Fig. 5. Flexural strength of AAM.

3.3. Lifecycle Assessment

The results of LCA are presented in Fig. 6 and Fig. 7. In Fig. 6, the GWP of two groups of mixes were calculated to compare the greenhouse gas emissions of the mortar production influenced by the introduction of $\text{Ca}(\text{OH})_2$ and the ratios of BWFA to BWBA. It is noted that the GWP is less than 300 kg CO_2 eq, significantly lower than that of PC-based mortar which varies from 700 to 1000 kg CO_2 eq [44]. Along with the increment of BWFA amount from 0 to 100 %, the GWP of the produced mortar declined moderately by 1.2 % for NaOH-activated mortar and 11.0 % for NaOH/ $\text{Ca}(\text{OH})_2$ -activated mortar. For both groups, the most significant contributor to CO_2 emission came from the alkali activator. When NaOH was used alone, it took up approximately an average of 44.3 %. The introduction of $\text{Ca}(\text{OH})_2$ enhanced the GWP by a maximal 42.3 %, observed for mix with 100 % BWBA in which $\text{Ca}(\text{OH})_2$ resulted in 31.4 % of emissions. Recycled materials from biofuel wastes accounted for very few environmental impacts, especially BWFA which consisted of a maximal 6.6 % (C0F100). This aligns to the results acquired in the former investigations [45]. Due to the sieving process, the GWP of BWBA was slightly higher than that of BWFA per the same amount. RS showed an average GWP of 13.6 % and 9.14 %, for mortar with and without $\text{Ca}(\text{OH})_2$, respectively. This was greater than the GWP of BWA owing to the large quantity of fine aggregates.

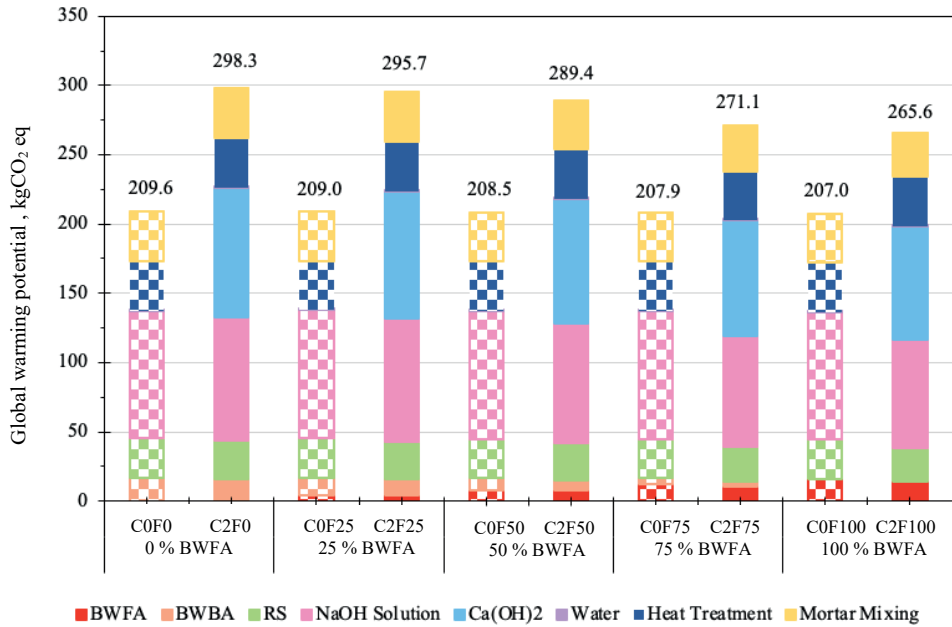


Fig. 6. GWP of AAM mixes.

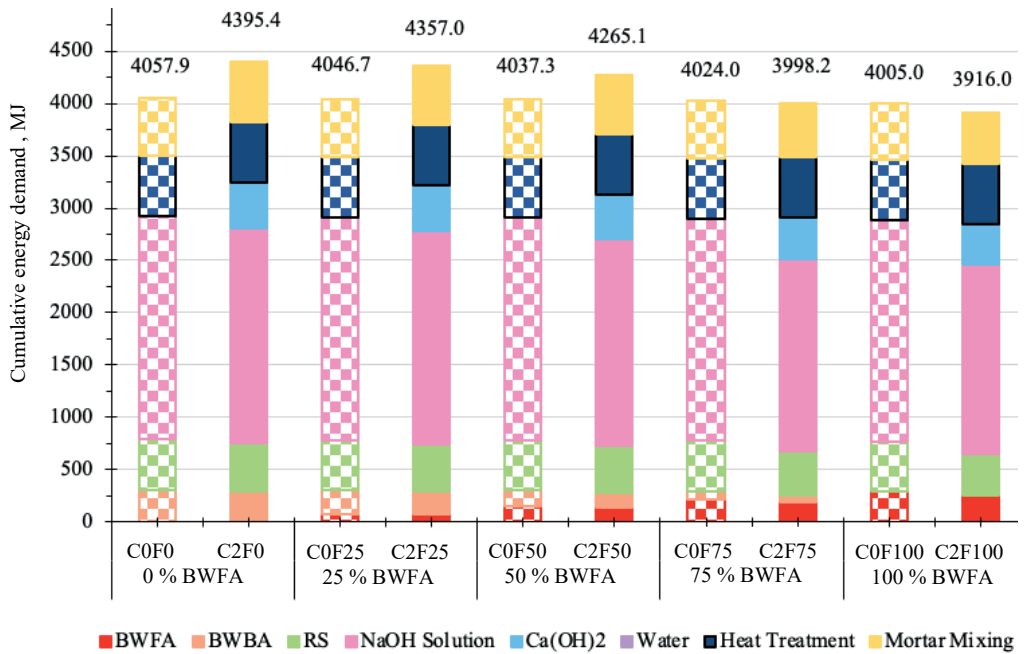


Fig. 7. CED of AAM mixes.

Fig. 7 illustrates the energy demand to manufacture the BWA-based AAM. From the graph, the energy consumption of one m³ mortar ranges between 4000 and 4400 MJ, less than the energy required for PC-based mortar which is around 4800 to 6600 MJ [44]. For all the mixes, the primary energy consumption belonged to the NaOH solution, consisting of 46.1–52.9 % of the total. The recycled materials, including BWBA, BWFA and RS, occupied a CEM within the range of 16.7 to 17.4 % which mainly came from the pre-processing of the wastes. The increment of BWFA content slightly decreased the CEM and the introduction of Ca(OH)₂ mildly increased the energy demand. Ca(OH)₂ accounted for an average of 9.9 % CED, which, however, is 35.0 % less than that of NaOH.

4. CONCLUSIONS

This study developed a green AAM with 100 % BWA as precursors. The effects of different BWFA-to-BWBA ratios, as well as the usage of binary alkali activator Ca(OH)₂, on the mechanical properties and environmental impacts of AAM were analysed. The circular economy is gaining popularity all over the world, and more sustainable biofuel energy, aiming at avoiding the use of non-renewable fossil energy for heating and electricity, is becoming increasingly significant. This study shows that previously unvalued wastes can be valorised for the construction industry. Especially under circumstances where natural resources are depleting and expensive to extract, the present study proves that the energy industries can benefit by recycling wastes for the construction sectors and converting waste to valuable products. Based on the obtained results, the primary conclusions are drawn as follows:

1. The highest compressive and flexural strength were both obtained for C2F50 which contained 50 % BWFA and 50 % BWBA as precursors. When the mortar was activated by 10 mol/L NaOH alone, the strength was 4.8 and 2.4 MPa, accordingly. With the introduction of 20 % Ca(OH)₂ as a binary activator, the values were measured to be 6.3 and 2.7 MPa, respectively.
2. The binary usage of BWFA led to the growth of the CaO/SiO₂ ratio and the decline of the Al₂O₃/SiO₂ ratio, improving the chemical activity of the mortar and thus benefitting the strength development. However, when its content is high, the great water demand due to its finer particles results in a fast setting, deteriorating the microstructure of AAM.
3. The introduction of Ca(OH)₂ enhanced the CaO/SiO₂ ratio of AAM and greatly improved the compressive strength when BWBA was included as a precursor and the flexural strength when the content of BWFA was less than 75 %. The most remarkable enhancement of compressive and flexural strength was 320 and 140 %, respectively, both observed for samples with 100 % BWBA.
4. The GWP of AAM varied from 200 to 300 kg CO₂ eq. Alkali activators contributed to the highest greenhouse gas emissions, with an average of 42.3 % and 31.4 % for NaOH solution and Ca(OH)₂, respectively. Biomass fuel wastes (BWA and RS) accounted for very few emissions, with the greatest GWP measured for RS up to 13.6%. The binary use of Ca(OH)₂ improved the GWP and by increasing BWFA content, GWP was decreased.
5. The production of AAM led to 4000 to 4400 MJ of energy consumption. Approximately half of the energy demand was attributed to the use of NaOH solutions. The binary introduction of Ca(OH)₂ only took up 9.9 % of the consumption. The CED of recycled wastes was less than 17.4 %.

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