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DOI

[10.1016/j.jobe.2024.110927](https://doi.org/10.1016/j.jobe.2024.110927)

Publication date

2024

Document Version

Final published version

Published in

Journal of Building Engineering

Citation (APA)

Liang, X., Li, Z., Dong, H., & Ye, G. (2024). A review on the characteristics of wood biomass fly ash and their influences on the valorization in cementitious materials. *Journal of Building Engineering*, 97, Article 110927. <https://doi.org/10.1016/j.jobe.2024.110927>

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A review on the characteristics of wood biomass fly ash and their influences on the valorization in cementitious materials

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ARTICLE INFO

Keywords:

Wood biomass fly ash
Waste valorization
Cementitious materials
Minerals
Ash classification

ABSTRACT

Wood biomass fly ash (WBFA) has emerged as one of the most dominant by-products in the biomass energy sector. Circulating WBFA for construction practice can mitigate the secondary pollution caused by improper ash management, and provide a new material source to compensate for the scarcity of raw materials in the construction industry. This paper reviewed the current research progress on recycling WBFA in cementitious materials. The physicochemical properties of WBFA were summarized based on the literature. Further, the implementations of WBFA for the development of cementitious materials were categorized into three binder systems: clinker, blended cement, and alkali-activated materials (AAMs). Owing to the large variation in chemical compositions of WBFA and strict requirements in clinkering parameters, employing WBFA in blended cement and AAMs seems to be more promising. A new classification approach for WBFA was proposed to divide WBFA into two categories. This helps to provide simple guidance for ash recycling in construction practice. Finally, the current research gaps in WBFA valorization in cementitious materials were summarized, outlining the research for further exploration.

List of abbreviation:

WBFA	Wood biomass fly ash
AAMs	Alkali-activated materials
SCMs	Supplementary cementitious materials
ESP	Electrostatic precipitators
CFA	Coal fly ash
XRD	X-ray diffraction
ASR	Alkali-silica reaction
LSF	lime saturating factor

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1. Introduction

Global warming is to date one of the main challenges worldwide. This is primarily due to the substantial emissions of greenhouse gas, especially CO₂, with the escalating industrial productions [1]. Statistical results evidence that approximately 30–40 % of CO₂ emissions originate from the power sector, as illustrated in Fig. 1. This should be predominantly attributed to the combustion of fossil fuels, particularly coal, for energy generation [2]. In recent decades, efforts aimed at reducing carbon emissions in the energy sector have led to a shift from fossil fuels to renewable energy sources. An illustrative instance is the European energy industry, which launched the coal phase-out initiative in 2015, proposing the closure of all coal-fired electricity plants by 2030. This initiative seeks alternative sources, especially renewable energy [3,4].

The emphasis on reducing fossil fuels while promoting renewable energy development has yielded significant results. Fig. 2(a) illustrates a steady increase in renewable energy development over the past 20 years in EU-27 countries. By 2022, the total renewable energy production had reached approximately 249 Mtoe, which is two times larger than the amount in 2002. Among the widely available sources of renewable energy, solid biofuel has emerged as a dominant component, constituting around 40 % of renewable energy sources in 2022, as depicted in Fig. 2(b).

Biofuels are composed of a wide variety of sources, including forestry biomass, agricultural biomass, and animal wastes etc [6,7]. Despite the complexity of the sources, it is acknowledged that wood biomass is the primary source of biofuels, and has been largely promoted in the last decades [8,9]. In the EU, wood biomass-derived energy accounts for more than 70 % of the total energy from biofuels, as depicted in Fig. 3. One of the main advantages of wood biomass energy is its carbon-neutral characteristic. It is recognized that with sustainable forest management, energy produced from wood biomass can achieve net-zero carbon emissions, as carbon circulates within the wood growth through the photosynthesis process [10]. In addition, biomass combustion techniques have been well developed, enabling its integration into existing coal-fired plants after proper retrofitting. This facilitates a seamless transition for the energy industry towards bioenergy orientation without extra costs and overhauls, which is a privilege from the industrial perspectives. Based on these advantages, a prospective development of the wood biomass energy industry can be anticipated. An estimation of wood biomass derived energy can reach about 18 % of total energy consumption by 2050 [11].

While the carbon neutrality of wood biomass energy greatly benefits the decarbonization of energy sectors, there are critical issues worth attention, particularly in the post-biomass energy production stage. Similar to coal combustion, it is inevitable for the generation of by-products, namely wood ash, after biomass incineration. In 2022, the annual fuel wood supply has reached approximately 0.72 billion cubic meters in European countries [13]. Based on the estimation of ash production ranging from 0.5 % to 7 % of the dry mass content of wood biomass [14], a massive amount of wood ash can be anticipated to be discharged in Europe.

As illustrated in Fig. 4, through the combustion process, two types of wood ashes, namely bottom ash and fly ash, are generated. The weight proportions between bottom ash and fly ash vary depending on the incineration techniques and feedstocks [15]. Unlike bottom ash that has great potential in road construction due to its geometric similarity to aggregate, the circulating of wood biomass fly ash (WBFA) remains challenging. Currently, most of the WBFA is directly landfilled or disposed of, which not only incurs economic costs due to land occupation but also presents environmental risks. The lightweight nature of WBFA makes it prone to becoming airborne, increasing the risk of air contamination through dust. Besides, there is the potential for heavy metals present in the ash to leach into groundwater, leading to ground water contamination [16]. Given the rapid development of the biomass energy industry, a surge in WBFA generation is anticipated. Thus, there is an urgent need to implement effective recycling methods for WBFA to integrate the sustainability of biomass energy.

The construction industry has become a preferred destination for a wide range of industrial by-products [17–21], mainly due to the imperative of reducing carbon footprints in the building sector and the high demand for raw materials. For now, the construction sector accounts for about 8–10 % of global CO₂ emissions, primarily due to the carbon-intensive production process of Portland cement, which involves limestone decomposition, clinkering at high temperatures, and grinding [22]. Viable solutions, such as the partial substitution of cement clinker with industrial by-products, often referred to as secondary or supplementary cementitious materials (SCMs), and the formulation of new alternative binders, have been proposed in recent decades, integrating industrial by-products to fulfil the raw material needs of the construction sector. Previous successes in recycling blast furnace slag and pulverized coal fly ash

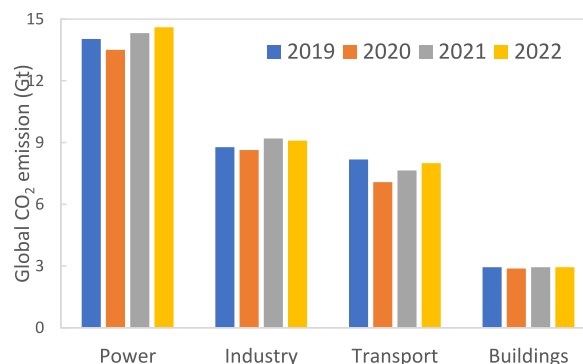


Fig. 1. Global CO₂ emissions by sector from 2019 to 2022 (adapted from Ref. [2]).

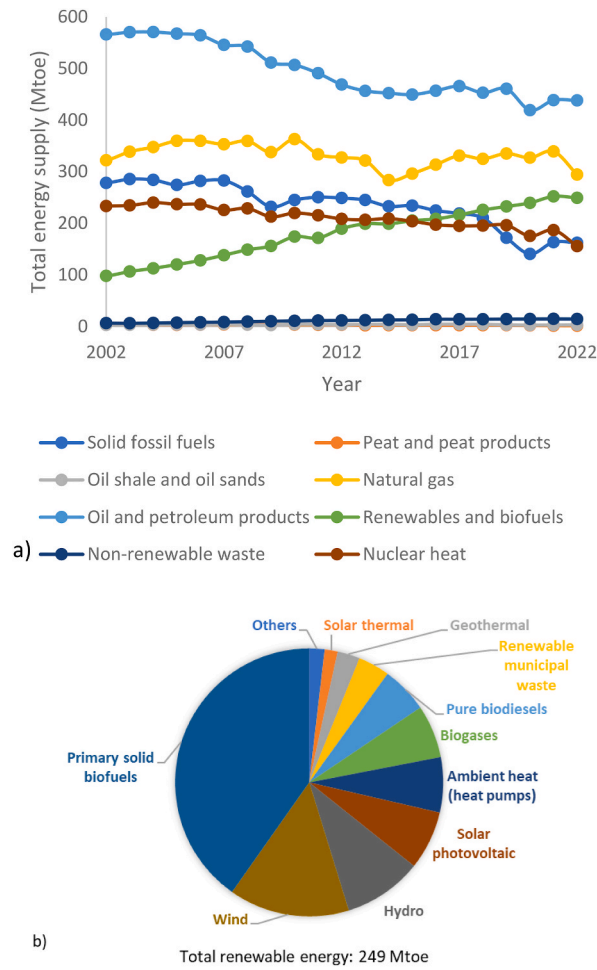


Fig. 2. EU total energy supply evolution (a) and distribution of renewable energies sources in 2022 (b) (data collected from Eurostat: Energy Balances [5]).

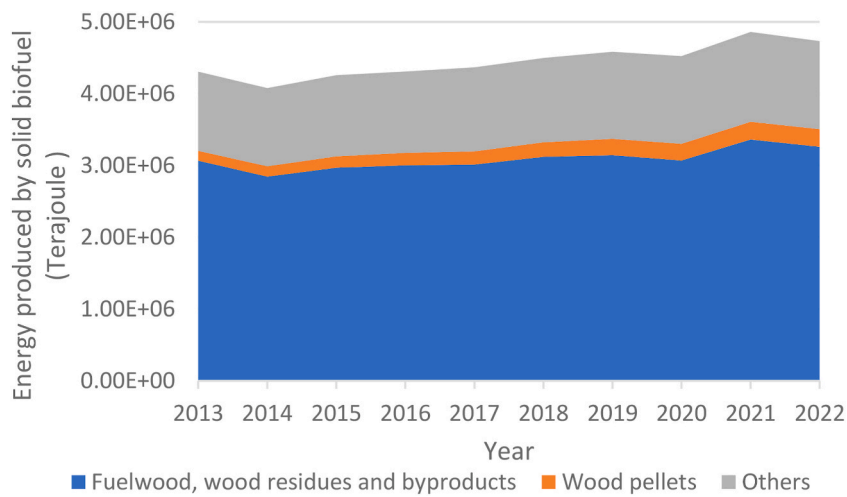


Fig. 3. Energy produced by different types of solid biofuels in EU (data collected from Ref. [12]).

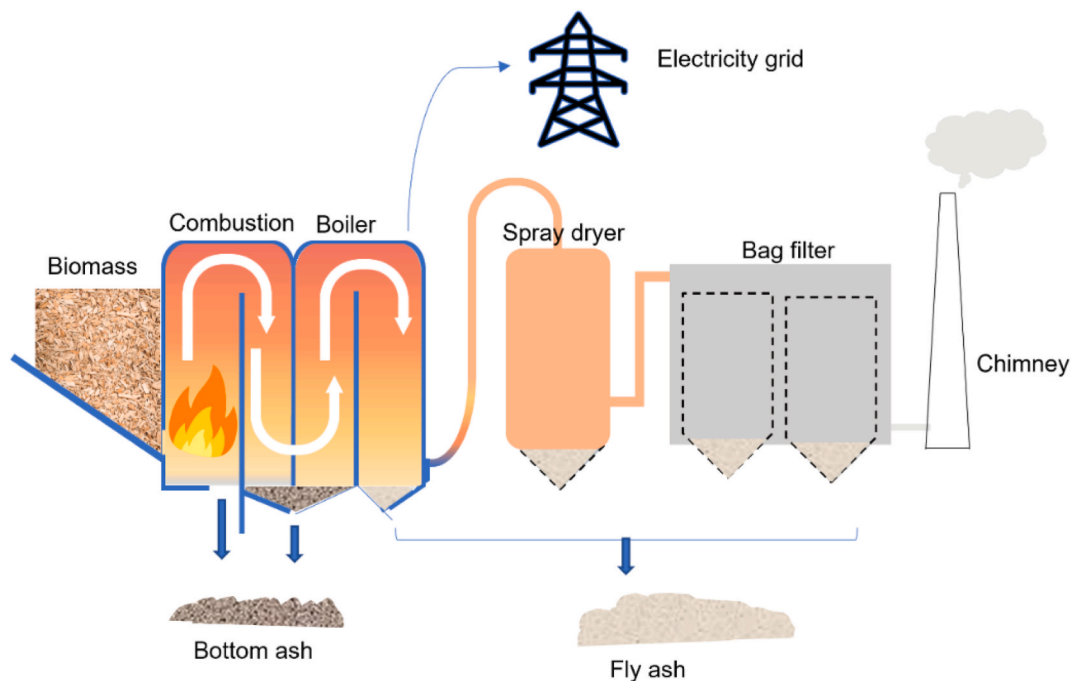


Fig. 4. Schematic figure of biomass combustion and ash collection.

have established these by-products as valuable mineral sources [23]. However, given the increasing demand for raw materials and the depletion of those ‘well-studied’ minerals, there is a pressing need to explore mineral sources from other industries. Wood biomass fly ash, on the other hand, shows significant potential as an alternative raw material for the construction industry. With the anticipated increase in WBFA availability, it could effectively supplement existing raw material sources, helping to ease the growing scarcity of conventional materials in construction industry. In addition, incorporating WBFA into cementitious materials allows it to be solidified into a monolithic structure, significantly minimizing the risks of air and water contamination compared to direct disposal or land-filling. This provides a win-win solution for both the biomass energy sector in managing WBFA and the construction industry in securing new material supplies.

Over the recent decade, numerous studies have explored the feasibility of using WBFA in construction materials. Despite notable progress and claims that WBFA holds promise for construction applications, inconsistencies in research findings persist, mainly due to the variation in ash properties. Currently, there is a lack of systematic analysis linking WBFA properties with its performance in cementitious materials, making it challenging to provide clear recommendations for the use of WBFA. To address this issue, it is necessary to classify WBFA and summarize its current utilization properties, thereby establishing a systematic ash classification for use in construction materials, a task that necessitates this review. This paper critically focuses on current recycling approaches of WBFA for the development of cementitious materials. The ash generation, physicochemical properties, and pretreatment methods are summarized based on the up-to-date research. The implementation of WBFA in three binder systems, namely cement clinker production, blended cement, and alkali-activated materials, were comprehensively discussed, highlighting the advantages and disadvantages of each. Further, a preliminary WBFA classification is proposed based on their chemical compositions and application preferences, aiming at providing a guideline for WBFA recycling. Finally, this review identifies the research gaps in developing cementitious materials with WBFA and outlines future perspectives on WBFA valorization.

2. Methods for literature selection

In this review, three databases, namely Scopus, Web-of-Science, and Google Scholar, were used to gather relevant literature. Considering that the terminology for wood biomass fly ash can vary across different studies, a systematic search was conducted using the following keywords: “wood ash,” “biomass ash,” “biomass fly ash,” and “wood fly ash,” “wood biomass fly ash.” These keywords were carefully selected to ensure comprehensive coverage of relevant literature. An exclusion process was carried out to eliminate duplicate literatures.

Subsequently, the literature underwent a rigorous evaluation process focusing on its “Abstract” and “Materials” sections to determine if the source of raw materials met the criteria of the present review study on WBFA. Articles lacking specification of the ash type or with a focus on ash derived from other biomass sources (such as agriculture ash, sludge ash etc.) were excluded from the review to ensure its exclusive focus on WBFA. The selected papers were then used for investigation on the physicochemical characteristics of WBFA. Following this, an investigation is carried out on the application of WBFA in construction materials, specifically focusing on the keywords of cement manufacture (clinker), blend cement (supplementary cementitious material), and alkali-activated materials

(geopolymer). This structured approach ensured that the review comprehensively covered both the properties and practical applications of WBFA in construction materials.

3. Fuels type, incineration techniques and ash collection

The variable properties of WBFA are dependent on multiple factors, including fuel feedstocks, incineration techniques, and ash collection methods [24]. This section provides information on these three aspects, associating these factors with the properties of WBFA. This information can facilitate the understanding of WBFA generation and how these factors can affect its valorization in cementitious materials.

The types of wood biomass can be one of the main factors affecting the properties of WBFA, particularly in terms of chemical composition. For instance, WBFA derived from poplar and eucalyptus was normally reported to contain a high amount of calcium, while WBFA from spruce wood and wood residuals was found to contain a higher proportion of silica [25,26]. Vassilev et al. [26] compared the chemical compositions of wood ash from 28 types of wood resources. Despite the large variations in wood biomass sources, a general trend in element enrichment follows the orders: $\text{CaO} > \text{SiO}_2 > \text{K}_2\text{O} > \text{MgO} > \text{Al}_2\text{O}_3 > \text{P}_2\text{O}_5$. Still, conflicting reports on element distribution can be found in different research studies, indicating the diversity of wood biomass sources contributing to the complexity of WBFA chemical compositions.

In addition to the sources of wood biomass, combustion techniques can also influence the properties of WBFA. To date, there are mainly three types of incinerators used in electricity production, as summarized below.

- **Grate furnace:** This is one of the most used combustors for biomass energy generation. It is feasible for small-scale biomass energy plants, showing high adaptability to a variety of biomass fuels in terms of size and moisture contents [27]. However, this combustion technique shows low burning efficiency, thus ashes generated from the grate furnace come with high carbon contents [28]. In the context of by-products produced, bottom ash comprises a substantial portion, ranging from 60 % to 90 % of the total weight fraction, while fly ash constitutes the minor by-products in this combustion technique [29].
- **Fluidized bed incinerator:** Compared with the grate furnace, the fluidized bed incinerator shows the advantage of high burning efficiency. As a result, ashes from the fluidized bed incinerator typically have lower carbon contents in comparison to those from grate furnaces. In this type of combustor, fly ash is the main residual [30,31]. Moreover, the fluidized bed incinerator is considered a more environmentally friendly option for biomass combustion compared to the grate furnace [32]. Therefore, the fluidized bed incinerator has become a more prevalent option for the newly developed biomass energy plants.
- **Pulverized firing system:** By pulverizing fuels into powders (typically with a size smaller than 0.1 mm [27]), this combustion method shows the advantages of having the highest burning efficiency among the three techniques. However, it is not applicable for the direct use of wood biomass as the main fuel, due to the challenges associated with fuel preparation and the potential corrosion of the combustor by inorganic salts from wood biomass [33,34]. Pulverized firing is typically used for the co-combustion process, where large proportions of coal are combusted alongside additional wood biomass (<20 % wt.). The co-fired fly ash from a pulverized firing system is within the scope of fly ash used for concrete, with great similarity to coal fly ash [35]. Consequently, it is not further discussed in the following review.

During biomass combustion, fly ash particles are carried by the flue gas through ash collection devices before being released into the air. Common devices for collecting WBFA include cyclones, bag filters, and electrostatic precipitators (ESP) [27]. Different ash collection methods initially determine the particle size of the WBFA. Among these systems, cyclones are normally used as the first step for ash filtering, and are reported to have the lowest ash collection capacity among the three [36]. WBFA collected from cyclones mostly consists of coarse fly ash, while WBFA collected from bag filters and ESP tends to have a relatively smaller particle size [37]. Additionally, the ash collection methods affect the carbon content. Since unburnt carbon normally presents in lightweights with large-size particles, WBFA with larger particle sizes is more likely to contain higher amounts of unburnt carbon. Cyclone WBFA is,

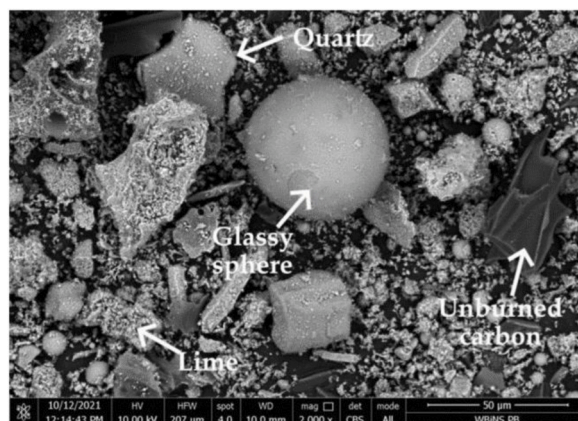


Fig. 5. Morphology of WBFA determined by SEM (picture from Ref. [45]).

therefore, observed to have higher carbon contents than those collected from ESP and bag filters [38].

4. Characteristics of WBFA

Despite the various influencing factors on the physicochemical properties of WBFA, as discussed in the previous section, it remains feasible to compile these properties from the available literature. Summarizing the physicochemical properties of WBFA is advantageous for better ash classification, which, in turn, aids in proposing a systematic valorization approach for WBFA in the development of cementitious materials.

4.1. Physical properties

The physical properties of WBFA include its particle size, morphology, texture, etc. The colour of WBFA can be an indicator of unburnt carbon content, as it turns darker with an increase in unburnt carbon content. Relating the appearance of WBFA to the combustion techniques, it is normally reported that WBFA from a fluidized bed tends to show a lighter colour compared with the ones from grate [38], due to the normally higher contents of unburnt carbon in grate-fired WBFA.

The particle size of WBFA is largely dependent on the ash collection techniques, as previously illustrated in Section 2. Although the particle size of WBFA largely varied case by case, most of the particles maintain sizes within 100 μm [37,39,40], assuring the feasibility of WBFA being able for the homogeneity in mixing when blended with other materials. Still, there are small portions of WBFA remains large in size, which are mainly composed of unburnt carbon. Fig. 5 shows the representative scanning electron microscope image of WBFA. Unlike coal fly ash (CFA) that is mostly spherical in shape, most of the WBFA particles are angular, with only a small fraction being spherical. This distinction arises from the lower burning temperature of wood biomass (normally below 1000 $^{\circ}\text{C}$) compared to coal combustion (above 1200 $^{\circ}\text{C}$). The low combustion temperature of wood biomass is less effective in melting the solids and subsequent condensation into spheres [41]. The lignocellulosic nature of wood biomass contributes to the formation of fibrous particles with highly porous structures [42]. The heterogeneity and porous structure of WBFA particles lead to large variations in density and specific surface area ranges, with a range of 2.2–2.7 g/cm^3 [43,44] and 8–40 cm^2/g [40], respectively.

4.2. Chemical compositions

4.2.1. Major elements

To date, the supply of forest residues (e.g., barks and branches), waste woods (e.g., discarded furniture and demolished wood constructions), as well as wood-based by-products from the pulp and paper industry, has become the main stream for wood biomass energy [46]. The diversified feedstocks of wood biomass contribute significantly to the considerable variation in the chemical compositions of the WBFA. We have summarized the chemical compositions of 97 types of WBFA reported in the literature, as presented in Fig. 6. The box covers the ranges of first quartile and third quartile of the contents of each element, and the whiskers represent the maximum and minimum value of contents of elements. Although wide variations are reported for these elements, based on the mean contents of each element, a general trend for the major elements follows the order: $\text{CaO} > \text{SiO}_2 > \text{K}_2\text{O} > \text{MgO} \approx \text{Al}_2\text{O}_3 \approx \text{SO}_3 \approx \text{P}_2\text{O}_5 \approx \text{Fe}_2\text{O}_3$.

Despite coal being considered the fossilized form of biomass, this biological relationship between these two fuels does not translate into similarity in their ashes regarding chemical compositions. It is well known that CFA in cementitious materials is mainly rich in aluminosilicates. Even for high-calcium CFA, the sum of silica, alumina, and iron oxide is normally greater than 50 % of the total weight [90]. Comparatively, as shown in Fig. 6, WBFA contains a high amount of calcium and magnesium while having a relatively low content of aluminosilicates. The medium values of silica and alumina in WBFA are only 19.30 % and 3.74 %, respectively. This information suggests the possible latent hydraulic properties and low pozzolanic reactivity of WBFA. However, the high availability of

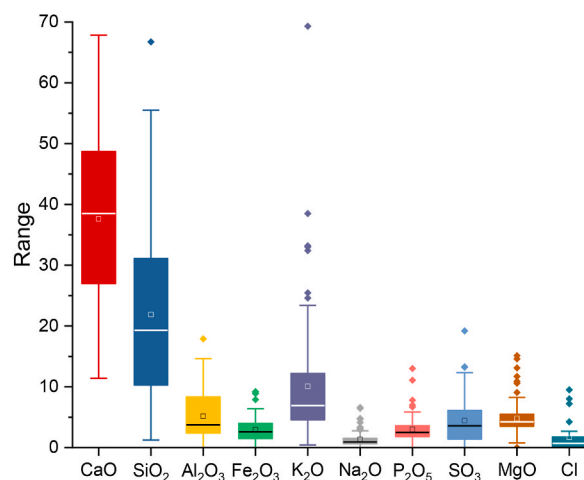


Fig. 6. Boxplots of chemical compositions of 97 types of WBFA with data collected from literature [25,38,39,45,43,44,47–89]

calcium and magnesium also raises concerns about expansion and cracking potential of cementitious materials if they are in the form of free lime or free MgO.

Due to its high alkali content, WBFA is normally reported to be alkaline in nature, with a pH value ranging from 9 to 13.5 [91]. However, when measuring the pH value of WBFA, different water-to-ash ratios are normally employed in various studies. This renders it inappropriate to directly compare the pH values of WBFA across various studies. Fig. 7 plots the measured pH values of WBFA from three research articles, along with their corresponding equivalent alkali contents. It is interesting to note that even in the same studies, the pH values and equivalent alkali content in WBFA are not linearly correlated; WBFA with higher alkali contents may potentially display lower pH values, as seen from the works by Carević et al. [47], and Sigvardsen et al. [66]. This phenomenon may be attributed to the presence of alkalis in the form of insoluble salts or in amorphous phases that are less susceptible to dissolve in water. Additionally, the elevated pH values in WBFA can also be influenced by the presence of free lime in the ashes, increasing the complexity of relating the alkalinity of WBFA to their chemical compositions.

Chloride is a potential threat to steel bar corrosion in reinforced concrete structures. Hence, the chloride content in cementitious materials must be strictly controlled. However, in WBFA, chloride content is typically reported to be high due to the wood feedstock. As indicated in Fig. 6, the median chloride content is 0.70 %, with the highest content reaching about 9.50 %, which is much larger than the required maximum chloride content (0.10 %) in cementitious materials. This elevated chloride content would present a limiting factor for the use of WBFA in structural concrete.

To assimilate the chemical compositions of WBFA into cementitious materials, the classical normalized $\text{CaO-SiO}_2\text{-Al}_2\text{O}_3$ ternary diagram is employed to compare the composition of 97 types of WBFA (illustrated as black dots) with those of conventional cementitious materials, including Portland cement, slag, class C (C) and class F (F) CFA, and natural pozzolans, as shown in Fig. 8. The distribution of WBFA in the diagram is similar to the results found by Carević et al. [47]. It is evident that the representative area for WBFA covers a significant expanse, implying its inherent heterogeneity in chemical compositions. In addition, there are observable overlaps between WBFA and both Portland cement and slag. This suggests that WBFA may possess latent hydraulic properties and possibly limited pozzolanic properties.

In summary, despite the variations in chemical compositions, WBFA contains higher amounts of calcium and alkalis but fewer aluminosilicates compared to CFA. It is, therefore, not applicable to directly adopt the valorization methodology of CFA in cementitious materials. Instead, new strategies are needed for the valorization of WBFA.

4.2.2. Heavy metals

Owing to the limited content, it is difficult to precisely measure the contents of heavy metals from XRF techniques. Instead, leaching test is normally applied to determine the concentration of heavy metals in WBFA. The heavy metals in WBFA are largely influenced by the types and sources of wood biomass. As wood biomass grows, it absorbs heavy metals from the soil, which subsequently remain in the WBFA following wood combustion. In addition, heavy metals can also originate during the service life of wood. Demolition wood, for instance, is reported to contain high amounts of Pb and Cd due to its coating with paints [84,94].

Table 1 provides a concise overview of the findings from several studies regarding the measurement of heavy metals in WBFA. While the leaching results may vary depending on the experimental conditions and deviations in wood biomass, Cr, Cd, Pb, and Zn were consistently identified as the most commonly reported heavy metals in WBFA that warrant further attention. It is essential to note that the evaluation criteria for heavy metals also differ in different countries, lacking unified requirements. Nevertheless, it is encouraging that several studies have shown positive outcomes of using WBFA in cementitious materials in relation to heavy metal leaching [38,75,95,96]. This is because, on the one hand, the heavy metal leaching potential can decrease with aging as a result of the formation of new mineral phases that capture the heavy metals through their precipitation [97]. On the other hand, the

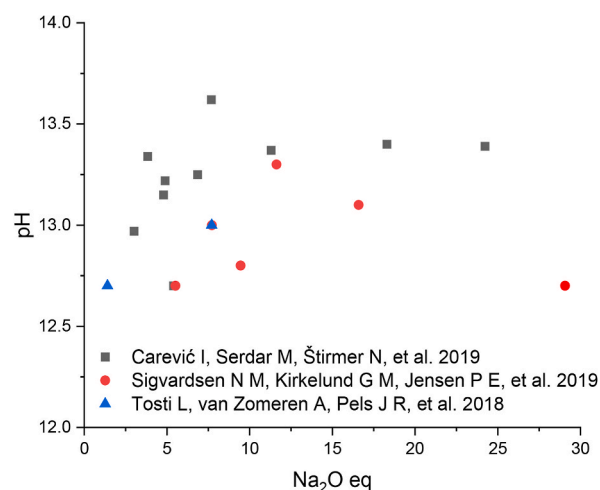


Fig. 7. pH values of WBFA vs. their equivalent alkalis content (data collected from [47,66,75], equivalent alkalis content ($\text{Na}_2\text{O}_{\text{eq}}$) = Na_2O wt.% + 0.658 K_2O wt.%).

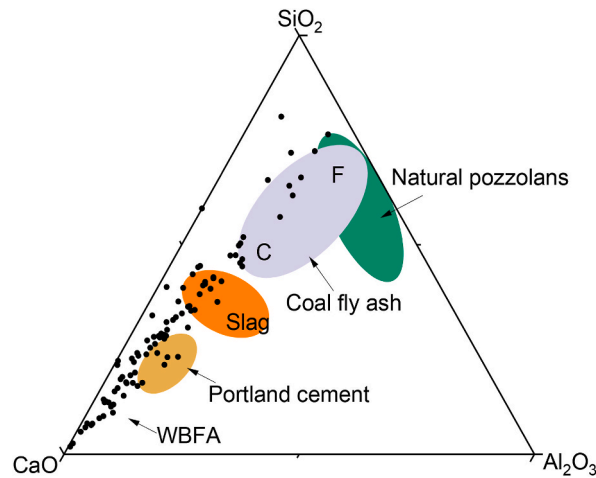


Fig. 8. CaO-SiO₂-Al₂O₃ ternary diagram of WBFA (data collected from Ref. [38,39,45,43,44,47-59,62-66,68,89,92]) with comparison of conventional cementitious materials [93].

Table 1

Main conclusions of heavy metals leaching from WBFA.

Ref	Measured heavy metals	Employed standards	Heavy metals beyond limit	Results remarks
[37]	Ti, Pb, Cr, Ni, Cd, As, Hg	Fertilizers Regulation Act in Japan	None	The heavy metal levels in WBFA met the regulatory standard, satisfying the requirements for fertilizer production
[38]	Sb, As, Ba, Cd, Cr, Hg, Pb, Mo, Ni, Se, Zn	Measured according to EN 12457; Evaluation based on SQD	Cr, Pb	Cannot be used for construction material due to the exceeded upper limits for Cr and Pb
[47]	Zn, Cd, Cr, Ni, Pb, Mn, Co, Ba, Bi, Sr, Cu, Hg	Measured according to EN ISO 16968:2015 and ASTM D 6722-11; Evaluation based on European Commission: Heavy metals and organic compounds from wastes used as organic fertilisers	Zn, Ni, Cd, Pb	The leaching of Cd surpassed the maximum allowable value in all 10 categories of WBFA.
[96]	Cd, Cr, Cu, Ni, Pb, Zn	Measured according to NEN 7345; Evaluation based on SQD	None	WBFA blended with cement showed good environmental stability
[97]	Cd, Cr, Cu, Ni, Pb, Zn	US EPA 3015A	n/a	Carbonation and hydration processes can reduce the leaching of heavy metals in WBFA

implementation of WBFA in cementitious materials can significantly immobilize heavy metals through hydration products and a condensed microstructure [98]. Consequently, the monolithic leaching of cementitious materials containing WBFA has a limited leaching potential. For instance, Berra et al. [96] employed WBFA to replace 30 % of cement to prepare paste samples. While the leaching of Zn (2274 mg/kg), Pb (177 mg/kg), and Cr (101 mg/kg) from WBFA exceeded the upper limit of IBC building materials (Zn < 14 mg/kg, Pb < 8.3 mg/kg, Cr < 7 mg/kg) according to the Dutch requirement (SQD), the monolithic leaching of paste was found to comply with the SQD requirements. Cement pastes containing WBFA showed no adverse environmental effects in terms of heavy metal contamination. Tosti et al. [75] measured the leaching of heavy metal from cement mortar containing up to 40 % of WBFA. The crushed mortar was verified to comply with the leaching requirement. These results further highlight that the use of WBFA in cementitious materials is advantageous from an environmental perspective.

4.3. Mineral compositions

The mineral compositions in WBFA are closely relevant to its chemical compositions. Therefore, it can be anticipated that the major mineral phases are rich in calcium and alkalis, given the high content of these elements in WBFA, as indicated in Fig. 6. Vassilev et al. [99] have provided a comprehensive list of minerals reported in biomass ash. However, the data encompasses a broad range of biomass sources without further distinguishing WBFA.

In light of the complexity of mineral compositions in WBFA due to the variation of wood sources as well as the limited availability of quantitative phase analysis in WBFA, it is unjustified to compare the abundance of minerals in WBFA. To better specify the most likely present mineral in WBFA, we have conducted a frequency analysis on mineral phases reported in WBFA. In total 70 types of WBFA from 27 research papers were analyzed by regarding their mineral compositions determined by X-ray diffraction. For specific minerals, the reported frequency was calculated according to Equation (1):

$$M_i(\%) = \frac{\text{Numbers of WBFA samples containing mineral } i}{\text{Total numbers of WBFA samples}} \times 100\% \quad (1)$$

where M_i is the reported frequency of mineral phase i , and the total numbers of WBFA samples are 70, which is collected from 27 research papers. The reporting frequencies of the prevalent phases in WBFA are present in Fig. 9.

It is evident that calcium-bearing phases are frequently reported in WBFA, with calcite, portlandite, and lime being the most commonly observed. Additionally, quartz is the primary silicon crystalline phase in WBFA. Due to the presence of sulfate, potassium, and chloride, minerals such as sylvite, anhydrite, and arcanite are also commonly noted in the literature [66,72,86]. These phases contribute to the primary crystalline minerals in WBFA.

In addition to crystalline phases, the amorphous phase is another important component in WBFA. Amorphous phase is more prone to react in an alkaline environment compared to crystalline phases, thus indicating the reactivity of WBFA to a certain extent [101, 102]. Since the combustion temperature of wood biomass is mostly within 1000 °C, which is insufficient for melting the ash particles to form a high content of amorphous phases, a generally low content of amorphous phases can be expected. This can be supported by the X-ray diffraction (XRD) patterns of WBFA, as shown in Fig. 10. Materials with high amorphous content often exhibit a hump in their XRD patterns, whereas the XRD patterns of WBFA typically show highly crystalline features with flat patterns. Quantified phase analysis, although with limited data available from the literature, has shown a low amorphous content in WBFA. For instance, the amorphous content in WBFA was reported to be 27.6 % in Ref. [103] and a range of 24–61 % in Ref. [86] from QXRD analysis, and 11 % from acid attack dissolution test [104].

5. Pretreatment of WBFA

The purpose of pretreatment is to improve the feasibility of materials for developing cementitious materials, where sieving and washing were the most commonly adopted pretreatment techniques for WBFA.

The primary goal of sieving is to remove the larger particles associated with lower reactivity in cementitious materials. For WBFA, it is noted that the coarse particles mostly composed of unburnt carbon, which may be efficiently removed by sieving [45,105,106]. Given the large specific surface area of these unburnt carbon particles, the water requirement of cement would be greatly increased with the introduction of raw WBFA. Therefore, sieving can somewhat reduce the higher water requirement, so facilitating the attainment of satisfactory workability when utilizing WBFA in cementitious materials.

The purpose of washing is to eliminate soluble salts that could be detrimental to cementitious materials. Berra et al. [48] applied washing to remove alkalis, sulfate, and chloride in WBFA. Since these elements exist primarily as soluble salts, as previously demonstrated in Fig. 6, such as sylvite and arcanite, washing is efficient in improving the quality of ash to meet the requirements of chemical compositions in fly ash used for concrete, as specified in EN 450–1 [35]. Meanwhile, washing can also reduce the concentration of heavy metals [38]. However, it is important to note that washing only transfers the contaminating substances from WBFA into water. This means that further actions should be taken for waste water management; otherwise, secondary pollution may occur after the washing process, which is also unfavourable from an environmental perspective. However, there is limited information provided on wastewater management after washing WBFA, which deserves future research attention.

In addition to sieving and washing, there are other methods employed to enhance the quality of WBFA. One such method is grinding, which has been shown to effectively increase the reactivity of WBFA. Through grinding, the crystalline phases might be melted due to the localized high temperature, thus forming amorphous phases. This is also referred to as mechanical activation [107, 108]. Research by Kaminskas et al. [109] revealed that WBFA after grinding showed a higher pozzolanic reactivity compared to the raw WBFA. Meanwhile, grinding can improve the reactivity of calcium-bearing minerals in WBFA, facilitating the formation of portlandite and C-S-H gels. By reducing the particle size of WBFA, WBFA particles can serve as the filler effect to improve the compactness

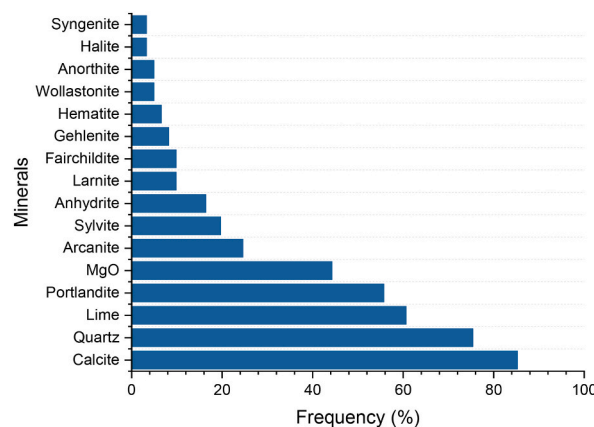


Fig. 9. Minerals in WBFA reported from literature and their reported frequency (data collected from Refs. [38,45,47–51,53,54,56,57,60–63,65,66,70,72,77,80,82, 85–87,89,100]).

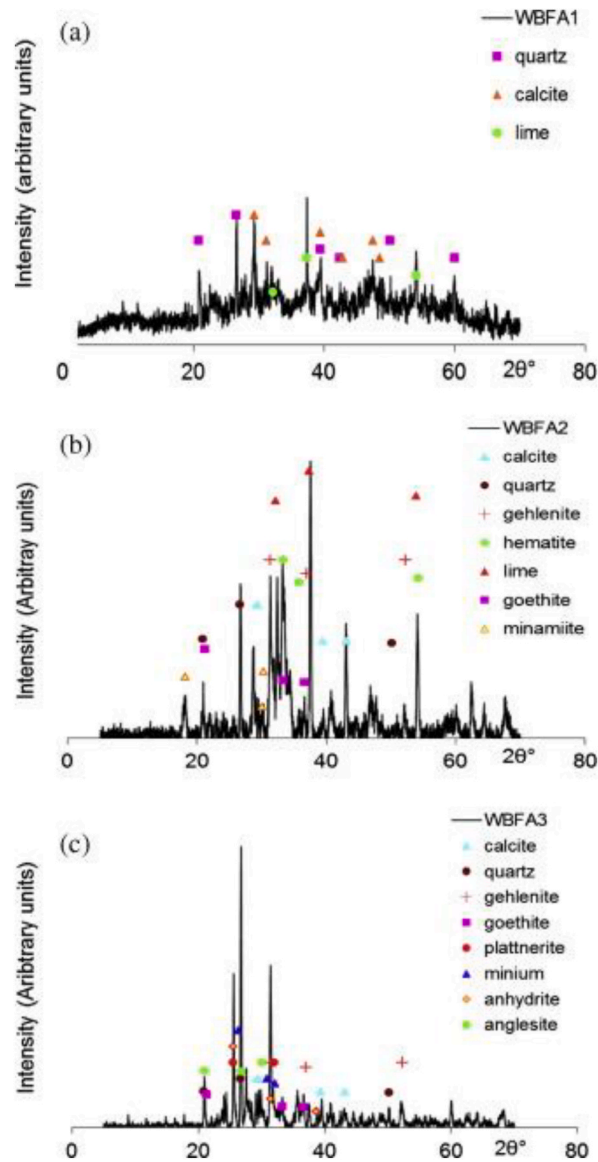


Fig. 10. XRD patterns of WBFA (picture adopted from Ref. [48]).

of cement matrix, promoting the strength development of cementitious materials [64,110].

Thermal treatment is another option to improve the reactivity of WBFA. The range of temperatures might vary considerably depending on the purposes. Regarding the calcination of WBFA at temperatures ranging from 700 to 800 °C, the unburnt carbon in WBFA can be further removed [38,111]. Meanwhile, the decomposition of mineral phases, especially calcite, can take place. This led to more reactive calcium oxide that can be used for prepare cementitious materials. For instance, Ates et al. [55] demonstrated that geopolymer mortar made with calcined WBFA at 800 °C exhibited greater strength compared to mortars made with WBFA without such treatment. Furthermore, in order to produce amorphous phases with greater reactivity, the crystalline phase can be melted at a high temperature, often above 1000 °C. Existing studies have focused on by-products like municipal solid waste incineration (MSWI) ash [112], while such elevated temperature treatment for WBFA has rarely been reported yet.

It is noteworthy that both grinding and thermal treatment are high-energy demanding. The acknowledgement of the need to enhance the quality of WBFA necessitates a comprehensive environmental analysis focusing on the pretreatment process and performance improvement in cementitious materials. Thus, it can justify the necessity for these treatment process. However, this research is limited at this moment and is of great interest for further research.

6. Valorization of WBFA in cementitious materials

6.1. Clinker production

As presented in Fig. 7, the normalized CaO-SiO₂-Al₂O₃ ternary chemical compositions of WBFA have a large overlap with Portland cement. This brings up the possibility of WBFA as one of the raw materials for cement clinker production. The benefits of applying WBFA in this way can be concluded as follows.

- 1) Reducing the need for limestone quarrying, considering the abundance of calcium in WBFA;
- 2) Decreasing energy costs associated with raw materials grinding, considering the particle size and grindability of WBFA compared to that of limestone;
- 3) Lowering the CO₂ emission during clinkering, given the presence of carbonate-free calcium minerals such as lime and portlandite.

To obtain high quality cement clinker, several factors should be considered when designing mixtures of raw materials. The lime saturating factor (LSF), as defined in equation (2), is one of the pivotal parameters. It is recommended to maintain a range of 0.92–0.98 in LSF for Portland cement to enlarge the amount of C₃S without yielding free lime [113]. Based on the chemical compositions of WBFA in the literature, the LSF of WBFA from 88 samples is calculated in this review. The distribution of LSF values and the frequency analysis are plotted in Fig. 11. It can be seen that the LSF in WBFA exhibits a large variation. By comparing the recommended LSF values for Portland cement clinker production (plotted in grey area) with the distribution of LSF in WBFA, it is noticed that most of WBFA falls outside the recommended LSF range. Therefore, it is impractical to apply WBFA as the main component for Portland cement clinker production. Instead, it can only be used in a smaller quantity as a partial replacement for calcium and silicon sources, depending on the chemical compositions.

$$LSF = \frac{CaO}{2.8 \times SiO_2 + 1.2 \times Al_2O_3 + 0.65 \times Fe_2O_3} \quad (2)$$

Besides the elements with interests in clinker preparation (Ca, Si, Al, and Fe), the impact of other elements in WBFA should also be addressed. For instance, the common presence of potassium, sulfur, and chloride in WBFA, as indicated in Fig. 6, may lead to the corrosion of kilns [114] further negatively influencing the properties of clinker. It is, therefore, necessary to assess these elements before applying WBFA to clinker production.

Experimental research on the implementation of WBFA for clinker production is so far limited. Tosti et al. [115] conducted a feasibility analysis regarding WBFA for clinker production. They emphasized the significance of controlling the properties of biomass ash with desirable CaO/SiO₂ ratios to satisfy the requirements of clinkering parameters for cement production. However, as shown in Fig. 11, it is difficult to maintain the parameters in a stable condition. Instead of producing Portland cement, Carr [116] directly fired WBFA in order to obtain mineral phases with hydraulic properties. By subjecting WBFA to high temperatures at about 1200 °C, wollastonite and β-C₂S were formed, imparting hydraulic properties to the ash. The hydraulic property can be further enhanced by fixing the mixture designs to produce more hydraulic phases. Carr extended this work by introducing pure chemicals to generate more alite and belite at the lab scale. Another study by Buruberri et al. [117] adopted other industrial wastes to fix the mixtures and produce belite clinker at about 1390 °C. Still, the compressive strength of the waste-derived cement was less satisfactory, reaching only about 10 MPa at 28 days.

To date, it is still challenging to apply WBFA to clinker production. One reason is attributed to the large variations in chemical compositions of WBFA. To prepare high-quality clinker, the designed clinker parameters such as LSF, alumina ratio, and silica ratio are extremely stringent. The imbalanced and inconsistent chemical composition of WBFA makes it difficult to meet these requirements in raw material design, thus complicating the mixture formulation for clinker production. Besides, there is a lack of research on the impact of unwanted chemicals in the WBFA on clinker formation and kiln corrosion, which should be further addressed. In addition,

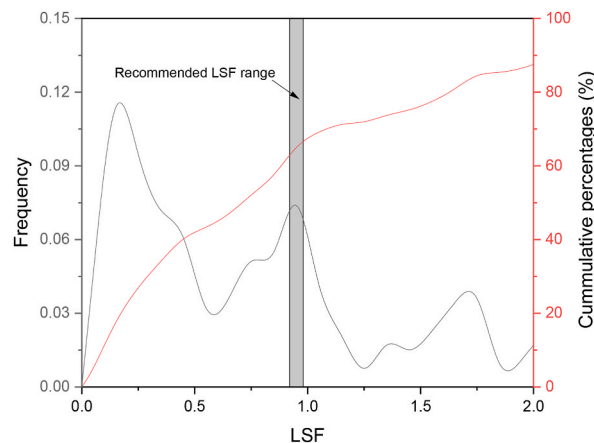


Fig. 11. Distribution of LSF and their occurrence frequency in 88 types of WBFA from literature (data collected from Refs. [45,39,43,44,47–58,62–66,68–89,92]).

there is no quantified environmental analysis of the valorization of WBFA in clinker production. It is crucial to evaluate whether the environmental burden relief from recycling WBFA in clinker production justifies the high-temperature process, especially considering the low-grade cement as reported in the literature.

6.2. Blended cement

Partially replacing cement with industrial by-products has become a well-established approach for waste recycling in the construction industry. A wide range of industrial by-products have been turned into valuable SCMs to partially replace cement, aiming at diminishing the carbon footprint of cement [118–121]. In this section, the implementation of WBFA in blend cement is discussed.

6.2.1. Roles of WBFA

For most of the SCMs in blended cement, we can generally categorize their roles in the following three aspects.

- Pozzolanic material

The pozzolanic reaction primarily involves the interactions between portlandite, one of the primary hydrates in cement hydration, and reactive aluminosilicate phases in SCMs [122]. This reaction ultimately facilitates the formation of C-(A)-S-H gels, leading to a densification of the microstructure of the cement matrix [93]. The pozzolanic reactivity of the WBFA is therefore of great interest to evaluate the potential of WBFA in this regard.

The governing elements for pozzolanic reactivity are primarily aluminosilicates and iron [123]. To ensure the pozzolanic reactivity, the amount of those aforementioned elements shall be above certain criteria. For instance, it is suggested in coal fly ash with a lower limit of the sum of aluminosilicate and iron at 50 % and 70 % for C and N types, respectively [90]. In addition to the chemical compositions, it is preferable that these elements exist in an amorphous state rather than in an inert crystalline form like quartz, mullite etc., as amorphous phases are more conducive to reaction [101]. According to the chemical compositions provided in Fig. 6, however, the mean value of the sum of silicon, alumina, and iron in WBFA is only about 29.77 %, implying the possibly low pozzolanic reactivity of WBFA in general.

Besides the evaluation of WBFA in terms of pozzolanic reactivity from chemical compositions, direct experimental assessments of the pozzolanic reactivity of WBFA have been reported in a few papers. Among the available test methods [124], the saturation lime method (Frattini method) and the Chappelle test are seen as the most commonly adopted methods for measuring WBFA in the literature. Even though WBFA normally contains a lower aluminosilicate than conventional SCMs, it has been reported with satisfactory pozzolanic reactivity in literature [48,70,74,125]. Moreover, it is important to note that these methods may lead to an underestimation of the pozzolanic reactivity of WBFA, as the calcium leached from WBFA is overlooked. The example can be referred to in Ref. [104], where the Chappelle test result for WBFA shows a negative value.

When comparing different types of WBFA, it is normally found that WBFA with higher proportions of aluminosilicates tends to present greater pozzolanic reactivity [66,74]. However, compared to commonly used SCMs such as metakaolin and coal fly ash, WBFA demonstrates significantly lower pozzolanic reactivity [70,89]. Thus, less compelling properties are present in cement blended with WBFA compared with those blended with coal fly ash and slag.

- Mineral with synergistic effect

Even for WBFA with low pozzolanic reactivity, it is still possible to be used in blended cement as a mineral additive, provided that it can positively contribute to the hydration of cement and/or other SCMs, instead of promoting pozzolanic reactions. This phenomenon is termed the “synergistic effect” in this review, a concept that has been extensively reported in the literature on other industrial by-products [126–128].

One representative case can be the limestone powder, which has been shown to react with aluminate and sulfate phases in blended cement, ultimately contributing to the increased monocarbonaluminate and stabilizing the presence of ettringite [129,130]. In this context, it is worth emphasizing that calcite is one of the main mineral phases in WBFA, as illustrated in Fig. 2.4. The results in Ref. [105] found that when WBFA was blended with cement, there was an observed increase in the formation of ettringite content, accompanied by a decrease in calcite contents. The depletion of calcite in WBFA also led to the formation of mono/hemi-carbonaluminate. Consequently, in their research, WBFA was found to exhibit a similar function to limestone powder in reacting with the aluminate phase, suggesting its synergistic effects on cement hydration.

Considering the latent hydraulic property of WBFA in the presence of lime and periclase, a minor addition of WBFA can also positively contribute to the strength development of cement. Meanwhile, since WBFA is highly alkaline, the introduction of WBFA can increase the pH value of the pore solution, thereby promoting the hydration of cement and other components such as slag and coal fly ash [79,131,132]. However, it is necessary to note that the high alkali content might lead to side effects on the properties of cementitious materials, for instance, the decreased long-term strength [133], and possibly increased alkali-silica reaction potential [133].

- Filler

Filler is considered an inert component in blend cement, as it barely contributes elements to the formation of hydrates. Still, it can accelerate cement hydration by serving as a nucleation site for the growth of hydrates. For example, the addition of rutile [134] and quartz powder [135] has been shown to promote cement hydration through the nucleation effect. However, it is important to note that in order to achieve the quasi-chemical filler effect, it requires an extremely fine particle size. Until now, there has been limited research reporting the filler effects of WBFA in blended cement systems, mainly due to the typically large particle size of WBFA. Bajto et al. [44]

reported the possible nucleation effect of one type of WBFA in blended cement, observing an enhanced heat flow peak. In their research, WBFA presents a smaller particle size than cement, with the d_{50} of about 6.10 μm . Maschowski et al. [110] also reported that up to 5 % of cement replacement by WBFA led to a significant strength increase due to the pore-filling effect. To improve the feasibility of non-reactive WBFA as a filler, grinding techniques can be recommended [53]. Still, an important consideration should be that when a large proportion of filler is added, the dilution effect of filler would eventually lead to performance degradation of binder [136,137]. Therefore, the amount of WBFA used for filler purposes should be carefully restrained.

6.2.2. Effects of WBFA on the fresh properties of blended cement

Regardless of the diverse properties of WBFA and its different functional roles in blended cement, it is interesting to observe that the results regarding the impact of WBFA on the fresh properties of cement concrete are largely consistent. It is generally acknowledged that WBFA can increase the water demand for cementitious materials [138]. Thus, the decrease in flowability is reported when WBFA is introduced as a partial cement replacement [48,79,103]. The reasons are attributed to water adsorption by unburnt carbon and particles with porous structures. The increased interparticle friction due to the angular WBFA particles is also one of the contributing factors [73].

The rheology properties of cement influenced by WBFA are less investigated yet. One study [73] concluded that due to the water adsorption by WBFA particles and the agglomeration of WBFA particles, the yield stress of cement mortar was found to be largely increased with the addition of WBFA. The increased yield stress with WBFA could also be attributed to the potassium salts that led to the early precipitation of portlandite and gypsum [72]. In terms of setting time, an extend in both initial and final setting time is widely reported [44,73,77]. The phenomenon was explained from the physical perspectives: competition for water, reduced water for cement hydration [73], and chemically retarding effects: the high sulfate and phosphorus content-caused slower hydration of cement.

6.2.3. Effects of WBFA on hydration and microstructure alteration of blended cement

Isothermal calorimetry test is normally used to investigate the hydration process of cementitious materials. When WBFA is introduced as a cement replacement, contrary results are reported. For instance, in Refs. [82,89], a decrease in the main hydration peak with the increment of WBFA was reported. This phenomenon was explained by the low reactivity of WBFA, which led to the dilution effect. However, in Ref. [44], when 15 % of cement was replaced by different types of WBFA, it was found that all these mixtures showed a higher main exothermic peak than the reference mixture, despite that the occurrence of the main peak can be delayed or accelerated depending on the different ashes. By relating the chemical compositions of WBFA with the heat flow, it is seen that for WBFA with high calcium and alkali content, the exothermic peak tends to increase in isothermal calorimetry, and vice versa. The abundance of alkalis and calcium can help with the initial cement hydration to a certain extent, which can explain these results. The adiabatic calorimetry shows similar results. Rajamma et al. [73] measured the temperature evolution of cement pastes with two types of WBFA. They observed that pastes containing less than 20 % of WBFA experienced an accelerated temperature rise during early hydration (before 10 h). Furthermore, pastes with a higher calcium content exhibited a more pronounced increase in temperature. Combining the information in short, it can be seen that the compositions of WBFA largely determines its effects on the hydration process when blended with cement. Higher calcium WBFA can slightly accelerate the early hydration process, while WBFA with high silicon tends to dilute the hydration intensity with its increased dosage.

As WBFA changes the hydration kinetics of cement, undoubtedly the phase assemblages shall alter when WBFA is introduced. From a qualitative analysis point of view, cement blended with WBFA typically contains similar types of hydrates. The main distinction lies in the inert phases introduced by WBFA, such as quartz and calcite [53,65], present in the matrix. However, the introduction of WBFA can influence the quantity of hydrates. For example, considering the availability of sulfate in WBFA, an increased amount of ettringite might form when a large amount of WBFA is incorporated [65,74]. This could result from increased $\text{SO}_3/\text{Al}_2\text{O}_3$ ratios with WBFA, and the presence of calcite in WBFA can also help with the stabilization of ettringite. In addition, it had been reported that the content of portlandite generated in cement pastes increased with WBFA at early hydration [48,65], as the calcium-bearing phases in WBFA could lead to a higher accumulation of portlandite. However, this increment effect was found to decrease with hydration. Considering the limited pozzolanic activity in WBFA, its contribution to the formation of C-S-H gels was minor. In Ref. [65], pastes blended with WBFA were observed to contain a similar amount of bound water and portlandite content, but a higher amount of ettringite at later stages of hydration (>28 days). Consequently, it was deduced that WBFA did not promote the formation of C-S-H gels. Fort et al. [89] determined the content of amorphous phases in cement blended with WBFA using QXRD. They observed a linear decrease in the amorphous content with the increment of WBFA, which can be attributed to the decrement of C-S-H gels.

The phase assemblages influenced by WBFA in blended cement would eventually lead to changes in pore structure. Yet, only limited investigations have been conducted regarding the effects of WBFA on the pore structure of blended cement. Maschio et al. [72] observed a significant increase in water adsorption when cement was replaced by WBFA by >5 %, indicating the increase in open porosity with the increment of WBFA. Fort et al. [89] applied the MIP technique to determine the cumulative pore volume and pore sizes of blended cement samples with WBFA. The results showed a gradual increase in total porosity with an increment in cement replacement level from 10 % to 70 % with WBFA. Particularly, capillary pores ranging from 10 nm to 1 μm showed the most significant increase in volume. Similar results were also reported in Ref. [139]. In all, due to the generally low reactivity of WBFA and coarse particle size, WBFA tends to cause a more porous microstructure in blended cement. This would be a significant factor affecting the mechanical properties and long-term performance of blended cement.

6.2.4. Effects of WBFA on the compressive strength of blended cement

The compressive strength of cementitious materials is a result of their phase assemblage and microstructure compactness. Therefore, it can be anticipated that WBFA may result in a negative impact on the compressive strength, given that in most cases WBFA

leads to a reduction in hydrate phases and increased porosity. Experimental results in general align with this expectation. When more than 10 % of cement is replaced by WBFA, in most research, a negative strength impact is observed [73,76,89,140]. Explanations for strength loss often point to the slow pozzolanic reaction of WBFA, which impedes strength development. Other factors such as the large WBFA particles with porous textures, and adverse effects on some alkali salts may also lead to a strength decrease, especially at a later stage.

Fig. 12 summarizes the effects of different WBFA percentages on the compressive strength of blended cement. There are optimal dosages recommended for WBFA from the strength point in different studies, claiming that within these criteria, WBF can positively contribute to strength development, or only lead to negligible strength compromise. Although the optimal dosages varied across different studies due to the variations of WBFA, a general 5–10 % was suggested [43,59,73,82]. By adding WBFA more than 10 %, a more noticeable strength decrease is commonly seen, as shown in Fig. 12. This dosage is much lower than the upper limit criterion of conventional SCMs in blended cement [141]. The efficiency of WBFA used in blended cement is relatively restrained.

6.2.5. Effects of WBFA on the durability of blended cement

Durability determines the service lifespan of cementitious materials. As durability encompasses a wide range of topics, the evaluation of durability should be based to the specific application of the cementitious materials with its associated requirements. While numerous studies on the feasibility of using WBFA in cementitious materials, relatively little attention has been given to durability, with only a few investigations addressing the durability of WBFA blended cement across various issues.

The transport property is one of the most important factors affecting the durability of cementitious materials, as it implies the pathway for aggressive agents to migrate into the materials [142]. Carević et al. [43] determined the capillary water absorption of cement mortar with WBFA. The results showed that introducing 5–10 % WBFA slightly reduced the water absorption of the mortar. However, increasing the WBFA content beyond this range led to higher water absorption. Similar trends were reported in other studies [44,64], where the use of WBFA at dosages of 15 % or more resulted in increased water absorption. From a transport property perspective, the recommended WBFA content should be within 10 %, which aligns with the strength perspective. An excessive dosage can create a more porous microstructure, leading to decreased strength and increased transport coefficients, compromising the durability of the cementitious material.

An elevated alkali concentration in WBFA may raise concerns about alkali-silica reaction (ASR), which can result in expansion and volume stability issues in cement concrete. Esteves et al. [70] substituted cement with WBFA containing 4.14 % potassium content. The results showed that mortar containing 30 % WBFA had a reduced ASR expansion compared to ordinary cement mortar. Furthermore, Acordi et al. [63] also concluded similar results when they substituted cement with WBFA and replaced aggregate with bottom ash in the production of concrete. A substantial decrease in ASR growth was noted in concrete containing WBFA as compared to the reference mixture. From the current research, WBFA appears to have no adverse effect but rather a comparable or even reduced potential of ASR cracking.

WBFA normally contains elevated concentrations of chloride, raising concerns about steel bar corrosion in structural concrete. While some studies have investigated external chloride transport in WBFA blended cement, little attention was given to the self-contained chloride. Therefore, the obtained results in chloride migration coefficient is often more closely associated with the pore structure of the cementitious materials [53]. Rumman et al. [103] found that concrete with 15 % of WBFA presents a same chloride penetrability class as reference, while 30 % of WBFA leads to a worse performance. Regarding the internal chloride content in WBFA, nevertheless, one notable argument is that the chloride from WBFA may be chemically captured to form Friedel's salt [143], which could reduce the amount of free chloride available to induce corrosion. Still, research in the self-contained chloride caused steel bar corrosion should be further investigated in future research if WBFA is intended to be used for structural concrete.

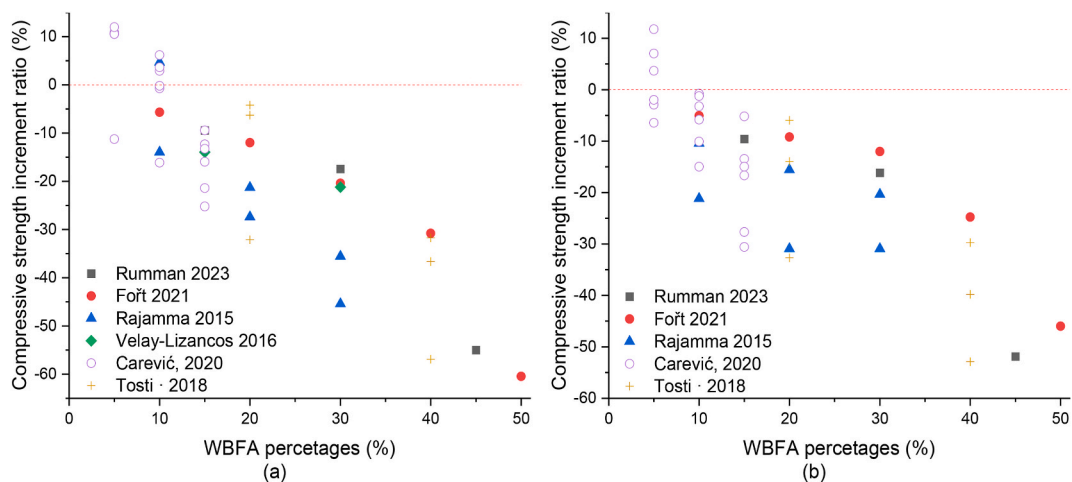


Fig. 12. WBFA contents vs. compressive strength increment ratios (a: 28-day specimens, b: 90-day specimens; data collected from Refs. [43,73,75,89,103,140]).

6.2.6. Brief comments

To date, the integration of WBFA in blended cement remains one of the main approaches to its valorization. From the cement industry point of view, due to the scarcity of commonly used SCMs, there is an increasing demand to seek alternative raw materials, for the purpose of reducing the carbon footprint of cement. The substantial demand for alternative SCMs in cementitious materials can promote recycling efficiency in WBFA. These circumstances collectively build the foundation for the advantageous utilization of WBFA in blended cement.

The main challenge in this regard is associated with the low pozzolanic reactivity of WBFA. To avoid significant performance compromises in cement concrete, the amount of WBFA to be suggested in blended cement is restricted to 5–10 %. Thus, the recycling efficiency of WBFA is relatively low.

Apart from the pozzolanic reactivity, it is noteworthy that there are valuable elements (calcium, sulfate, and alkalis) and phases (lime, portlandite, calcite etc.) in WBFA that can facilitate the reaction of cement and other SCMs. While it may be too ambitious to directly replace cement with WBFA, a more practical approach could be using WBFA as a partial replacement for common SCMs. This approach has the potential to yield synergistic effects on the hydration of WBFA, SCMs, and cement, ultimately achieving satisfactory performance of cementitious materials while reducing the needs of conventional SCMs. Currently, there is only preliminary investigations on this perspective [104]. Further research is recommended to explore the synergistic effects of WBFA in SCMs blended with cement, thus expanding the recycling methodology of WBFA.

6.3. Alkali-activated materials preparation

Alkali activation is another prevalent technique for waste recycling. In this technique, the reactions between precursors, contributing aluminosilicates and calcium species, and activators, providing a strong alkaline environment, lead to the formation of different hydrate gels like C-A-S-H, N-A-S-H, and C-(N)-A-S-H [144] and other secondary products. These hydrates ultimately confer mechanical properties on monolithic materials [145]. Any material capable of serving as either precursors or activators can be used for AAMs production. This versatility has led to the exploration of a wide range of industrial by-products for their potential use in AAMs production [146,147], including WBFA [57,148,149].

This section provides an overview of current research on recycling WBFA in AAMs production. Given the distinct characteristics of WBFA in chemical compositions and high alkalinity, studies have focused on both the precursor and activator perspectives.

6.3.1. WBFA as a precursor

In the production of AAMs, the most commonly used precursors align with those frequently employed as SCMs in blended cement, such as slag, coal fly ash, metakaolin, etc [145]. This is because, for both alkaline activation and pozzolanic reactions, the reactive aluminosilicates are of great importance. Materials that present high pozzolanic reactivity can always be ideally used for AAMs preparation. Differently, materials with low or even poor pozzolanic reactivity, which may be considered less reactive in blended cement, could potentially be used for AAMs preparation. This is due to the fact that the highly alkaline environment can enhance the dissolution of the reactive phases [150]. Therefore, WBFA retains the feasibility of being used as one source of precursors in AAMs preparation, even though WBFA shows lower reactivity than those conventional precursors.

When WBFA is used as the singular precursor in AAMs, it is anticipated that a high activation energy, for instance, elevated alkalinity of activators, may be needed, considering the typically low reactivity of WBFA. Nevertheless, this assertion may not be consistently supported by the experimental findings. In Ref. [71], it was found that NaOH solution with a molarity of 10 M was optimal for alkali-activated WBFA mortar regarding compressive strength. Increasing the molarity of activator to 18 M, however, led to a strength decrease. Similar results were also presented in Ref. [62], the compressive strength of NaOH-activated WBFA pastes achieved its highest strength with a NaOH concentration at 2 M. Further increases in alkalinity barely promoted strength development but caused a slight reduction. These results can be explained from the dissolution point of view: within a certain limit, a higher alkaline environment facilitates the dissolution of WBFA. However, as the alkalinity increases, the dissolution of calcium species is impeded. Thus, it is not feasible for the precipitation of C-A-S-H gels, ultimately resulting in a strength loss [151]. Therefore, an optimal mixture design is highly demanded to attain satisfactory performance without wasting extra activator. Still, it is essential to note that in general, the compressive strength of pure alkali-activated WBFA is relatively low (≤ 10 MPa) [62,71,152]. This may restrain the application of alkali-activated WBFA from an engineering perspective.

Considering the limited strength of alkali-activated WBFA, it is viable to incorporate few other highly reactive precursors, such as metakaolin [71,149,153–155], glass powder [62], and blast furnace slag [156] to create hybrid systems. Partially replacing WBFA with reactive aluminosilicates has been reported to yield more reaction products, resulting in a denser microstructure and enhanced mechanical properties of alkali-activated WBFA [71]. However, it is worth noting that in this context, the improved performance appears to be primarily due to the external reactive aluminosilicates. The addition of more reactive aluminosilicates materials results in superior performance. Thus, it is challenging to justify the contribution of WBFA to the AAMs. The practical significance of recycling WBFA would be largely diminished, even if a higher content of WBFA can be used in this scenario compared to its use in blended cement, as most of WBFA was only treated as inert aggregate instead of a value-added mineral in the binder system.

Instead of employing WBFA as the main precursor along with additional reactive aluminosilicates to prepare AAMs, a more practical strategy involves partially replacing the conventional precursors with WBFA in AAMs. The distinction between this strategy and the former one is that in this scenario WBFA is deemed to function as a value mineral to tailor the reaction kinetics, influence the compositions of hydration products in AAMs, and subsequently improve their performance. This would enhance the practical significance of WBFA as the contribution of WBFA can be clearly recognized. There would be an optimal dosage for WBFA to justify its role in this system. For instance, in Ref. [157], when metakaolin was partially replaced by WBFA, the main reaction products evolved

from N-A-S-H gels in the pure metakaolin system to the (N,C)-A-S-H gels in the blended mixture. A more compacted microstructure can be observed in SEM with the introduction of WBFA, and the compressive strength increased with the WBFA replacement ratio up to 50 %. High calcium WBFA was also used to replace coal fly ash in AAMs, as reported in Ref. [158]. Due to the introduction of calcium from WBFA, the setting time of mixtures can be shortened. Besides, a strength improvement, especially at the early stage, was reported in the mixture with the optimal coal fly ash replacement dosage at 10 % by WBFA. Similarly, the presence of C-A-S-H gels are evident as one of the main hydrates together with N-A-S-H gels. WBFA effectively changed the hydrates compositions further enhanced the performance of AAMs.

6.3.2. WBFA as activator

Since WBFA is reported to have high alkalis salt contents, the anticipated high alkalinity of WBFA enables it to stimulate the alkaline activation process when blending WBFA with other precursors. Jurado-Contreras et al. [157] investigated the soluble alkali salts from WBFA on the properties of alkali-activated WBFA-metakaolin mixtures. By comparing WBFA before and after washing, it was reported that AAMs with raw WBFA obtained a higher mechanical property than those prepared with washed WBFA. The contribution of alkalis was evident to the reactions of AAMs.

Moreover, some research has attempted to directly employ WBFA as a solid activator for the preparation of one-part AAMs. In Ref. [159], the high calcium wood ash was used as a solid activator to activate coal fly ash. It was observed that by blending wood ash with water, the pastes had a pH value above 13, implying the feasibility of using wood ash as an activator. Meanwhile, due to the presence of high calcium, coal fly ash can be activated at ambient temperature, forming C-S-H gels as one of the main reaction products. In Ref. [160], WBFA was used as a solid activator to activate silica fume. A mixture of 50 % WBFA and 50 % silica fume can obtain a compressive strength close to 30 MPa at 28 days. Additional studies exploring WBFA as an activator to combine with other aluminosilicate precursors have also been reported [50,161].

6.3.3. Brief comments

Alkali-activation enhances the dissolution of precursors. When used as precursors, WBFA can release more amounts of aluminosilicates compared to its dissolution in cement pore solutions [150]. However, if WBFA is used as the sole precursor, it may be difficult to achieve satisfactory mechanical performance due to its inherently low reactivity. Hence, it is more practical to either introduce other highly reactive precursors, or partially replace traditional precursors by WBFA. Still, in the hybrid precursors, the recommended upper limit for WBFA is mostly larger than that reported in blended cement. This allows for a higher recovery efficiency when recycling WBFA through alkali activation compared with blended cement.

Furthermore, considering their predominantly high alkalinity, it is of great interest in valorizing WBFA as an activator. On one hand, the implementation of WBFA as a solid activator enables the production of one-part AAMs, which solves the drawbacks of conventional AAMs with great difficulties in handling corrosive liquid activators [162]. Since the primary carbon emissions of AAMs originate from activators [163], employing WBFA as an activator allows for the production of AAMs entirely derived from solid waste, resulting in significant environmental benefits. Owing to the complexity of WBFA in the chemical composition, when conducting mixture designs of the 'one-part' like AAMs, it can be feasible to consider WBFA into two parts: treat the aluminosilicates in WBFA as part of precursors in the hybrid systems and treat the alkalis in WBFA as the source of activator. This can be a strategy for the mixture design of one-part AAMs. However, recent research on WBFA in AAMs still largely focuses on preparing conventional AAMs by adjusting the mixture designs (WBFA proportions in precursors, alkali contents, and modulus), further concluding an optimal mixture containing a certain amount of WBFA. There is a need for comprehensive research on studying WBFA as a solid activator, addressing aspects such as reaction kinetics, reaction mechanisms, microstructure evolution, and the eventual upscaling practice. Such research is essential to effectively take the use of the chemical compositions of WBFA in developing AAMs and enhance its valorization efficiency.

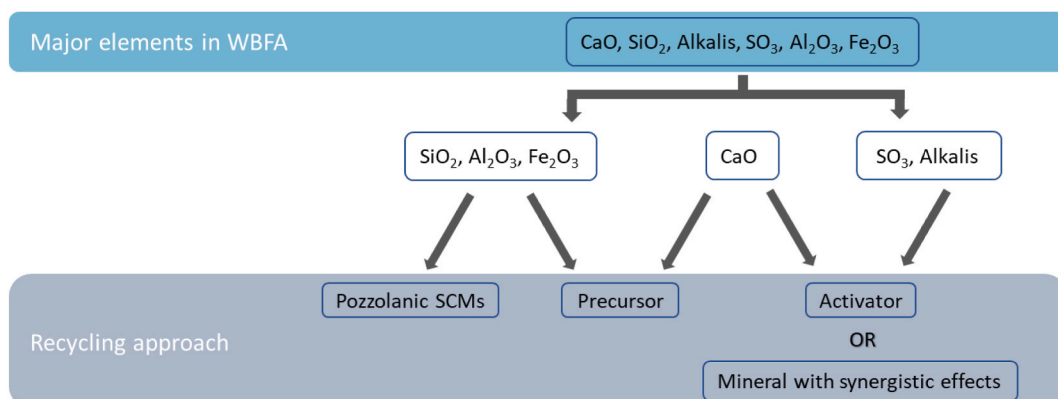


Fig. 13. Elements of interest from WBFA used in blended cement and AAMs.

7. WBFA classification towards cementitious materials development

To date, research on the valorization of WBFA is primarily regional, often presented as case studies. Previous attempts to classify biomass ash have been conducted in a few studies. Vassilev et al. [99,164] proposed an initial classification of biomass and ashes based on chemical compositions. From a phylogenetic perspective, Zhai et al. [165] reviewed the chemical compositions of biomass ash from different sources. However, it is difficult to directly adopt these ash classifications to help the design of WBFA recycling methodologies in cementitious materials, as these classifications only aim to predict ash compositions based on the sources. To facilitate the valorization of WBFA in developing cementitious materials, therefore, it is essential to develop ash classification methods from the cementitious materials perspective.

In the current review, the primary roles of WBFA in cementitious materials may be categorized into four key aspects: as pozzolanic materials and minerals contributing to synergistic effects in blended cement, as precursors and activators in AAMs. The chemical compositions of WBFA are highly correlated with the functions of WBFA in the development of cementitious materials. Hence, it is possible to propose the WBFA classification method from an application-focused standpoint, linking the chemical compositions to the functions of WBFA. Fig. 13 illustrates that the anticipated recycling methods for WBFA can be determined based on its primary chemical compositions. For example, in order to utilize WBFA as pozzolanic materials in cement, a greater quantity of aluminosilicates is required. When adopting WBFA for precursors in AAMs, in addition to aluminosilicates, calcium species can also involve in the reaction. Furthermore, if WBFA is to be utilized as activators, it is crucial to emphasize the presence of alkalis.

Fig. 14 provides a simple application-oriented WBFA classification method, derived from the modified ternary diagram, referring to the work from Vassilev et al. [99,164] and Zhai et al. [165]. The coloured areas in the figure represent the estimated distribution of WBFA, with the data plotted as a modified version of Fig. 6. To further correlate the chemical compositions and their associated applications, the distribution of 20 types of WBFA from the literature identifying different roles in developing cementitious materials is specifically plotted. Thus, a simplified two types of WBFA are summarized. Type I WBFA primarily includes a high content of aluminosilicates and iron. The normalized weight proportions of these elements are above 50%. This makes it suitable for use as either pozzolanic material or precursors in AAMs production. Conversely, Type II WBFA primarily comprises a high content of calcium, alkalis, and sulfate, which make it a suitable activator or calcium source in AAMs production, or SCM with synergistic effects, as previously stated in Fig. 13.

Similar to the classification of CFA into type C and type F, the classification of WBFA into two different categories can be advantageous for efficiently screening the quality of WBFA and providing guidance for its recycling in the production of cementitious materials. Still, it is worth noting that this classification remains limited. For instance, if quartz presents as the only silica source, the recommended recycling methodology of using WBFA as precursor in AAMs or SCMs in blended cement is then invalid, even if the quantity of silica is dominated in the bulk chemical compositions of WBFA. Therefore, further refinement of ash classification is needed when more data, particularly in terms of the quantified mineral compositions and chemical composition of the amorphous phases, becomes available. Despite these limitations, this methodology can still be applicable for an initial evaluation of the properties and potential recycling approach of WBFA.

8. Suggestions for future investigations

Based on the current review, the following aspects were summarized as the research gaps that should be addressed for further consideration in future research.

- To control the quality and refine the classification of WBFA

Based on the current review, the typical physicochemical properties of WBFA are summarized. Still, the variation of WBFA in terms of chemical compositions is commonly seen, which prompted the initial ash classifications discussed in Section 7 of this review. These classifications are preliminary and require further refinement. Additional data on the quantitative analysis of chemical and mineral compositions, and their impact on the performance of cementitious materials, is needed. This requires intensive experimental characterization and quality analysis of WBFA collected from diverse sources. Through this approach, it will be possible to establish specific classifications and requirements for WBFA, similar to those for coal fly ash, thereby enhancing its industrial application in cementitious materials.

Moreover, it is imperative to investigate novel pretreatment techniques in order to enhance the quality of WBFA, thereby improving the utilization efficiency. Thorough consideration of the environmental and economic impact of pretreatment is necessary to achieve a harmonious balance between the enhanced performance and treatment expenses of WBFA cementitious materials.

- To improve recycling efficiency of WBFA in cementitious materials

The low recycling efficiency of WBFA for the development of cementitious materials is mainly due to the improper recycling methodology adopted. Normally, the conventional approaches for recycling of aluminosilicate-rich materials were directly adopted in the WBFA valorization, such as directly substituting WBFA for cement or using it as a precursor. New strategies are needed to exploit the major components (alkalis, sulfate, and calcium-bearing phases) in WBFA for designing cementitious materials. For example, it might be interesting to design the cementitious materials on the leveraging of sulfate, alkalis, and calcium-bearing phases to promote the reaction of other aluminosilicates, thereby forming cement-free binders.

- To explore the reaction mechanisms of WBFA in cementitious materials

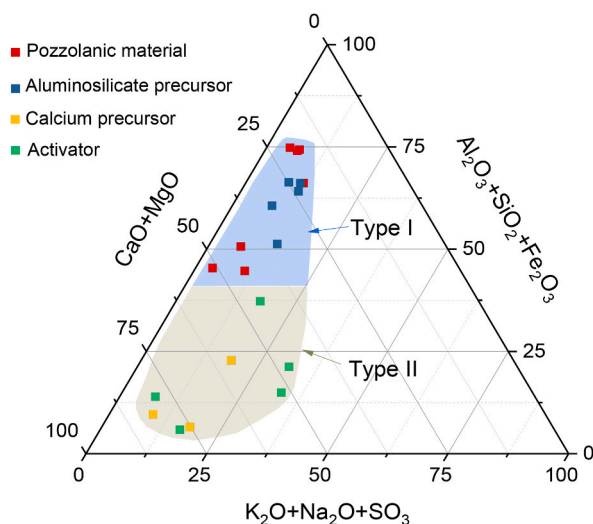


Fig. 14. Application-oriented classifications of WBFA based on chemical compositions (WBFA with pozzolanic activity in Refs. [61,63,70,76], WBFA used as aluminosilicate precursors in Refs. [71,149,153–155], used as calcium-based precursor in Refs. [62,158], used as activator in Refs. [50,157,161]).

Previous studies have widely investigated the performance of cementitious materials containing WBFA, while the reaction kinetics and microstructure evolution is not well understood. When WBFA is used for developing new types of cementitious materials, a more comprehensive investigation should be conducted to understand the role of WBFA and other components on the reaction mechanism perspective, enables providing a fundamental basis for designing the reaction products and further tailoring the performance of cementitious materials.

- To conduct pilot scale research for products development

Upon the development of cementitious binders with WBFA, research on a pilot scale is essential to convey the feasibility of WBFA in producing construction products from an engineering perspective. By producing construction products, for instance, concrete, bricks, and grout, the technical performance, such as mechanical properties and application-demanded long-term performance, can be exclusively evaluated. These properties, together with the environmental analysis, including heavy metal leaching and life cycle assessment, can prompt industrial interests and facilitate the valorization of WBFA for practice.

9. Conclusions

The prospective development of biomass energy leads to the accumulation of WBFA as one of the major by-products. Still, it also presents an opportunity for the construction industry to access a new source of raw materials. By integrating WBFA in cementitious material, it avoids the contamination of WBFA with the environment and further reduces the carbon footprint of construction materials by reducing the use of Portland cement. This can be advantageous for attaining waste-to-resource integration, therefore promoting the circular economy in the construction industry. In this paper, we comprehensively reviewed the up-to-date research on the physico-chemical properties of WBFA and its utilization for the development of cementitious materials. Based on the review, the following conclusions can be drawn.

- Despite the considerable variability in the physicochemical properties of WBFA, the predominant chemical composition tends to follow the sequence: $\text{CaO} > \text{SiO}_2 > \text{K}_2\text{O} > \text{MgO} \approx \text{Al}_2\text{O}_3 \approx \text{SO}_3 \approx \text{P}_2\text{O}_5 \approx \text{Fe}_2\text{O}_3$. The crystalline phases predominantly consist of calcium-bearing phases, and the amorphous content in WBFA is generally lower than that in conventional materials employed in the cement concrete industry, resulting in limited pozzolanic reactivity.
- Three main approaches for valorizing WBFA in cementitious materials, namely clinker production, cement substitution, and development of AAMs, were categorized. While clinker production demands high material stability and high energy cost in clinkering process, which is not particularly advantageous for WBFA valorization, incorporating WBFA in blended cement and preparing AAMs remain as two mostly used approaches.
- In blended cement, owing to the generally low pozzolanic reactivity of WBFA, it is recommended to use only 5–10 % WBFA as a cement replacement to avoid strength loss. In the production of AAMs, WBFA can be used as aluminosilicates and calcium sources depending on its chemical compositions. Besides, owing to the high alkalinity of WBFA, it is possible to recycle WBFA as a solid activator. Nonetheless, limited research has been conducted in this aspect.
- To enhance the understanding of WBFA and facilitate its application, an application-oriented WBFA classification method was proposed, based on the chemical compositions and their correlation with their reported applications in the literature, aiming to provide guidance for WBFA valorization in construction materials. A further refinement for the ash classifications is suggested in future research when more information of WBFA, especially the quantification of mineral compositions, become available.

In light of the information from this review, it was found that the utilization efficiency of WBFA in cementitious materials remains limited, necessitating the exploration of new recycling methodologies. To promote the WBFA recycling in construction materials, it is recommended to implement refined quality control and classification approaches for WBFA, establishing a general recycling framework. Additionally, more research should focus on developing new types of cementitious materials using WBFA by advancing the use of the main components and understanding the fundamental reaction mechanisms. Future pilot-scale research should include both technical and environmental performance assessments on the construction products developed with WBFA to promote its practical applications.

CRediT authorship contribution statement

Xuhui Liang: Writing – review & editing, Writing – original draft, Investigation, Formal analysis, Conceptualization. **Zhenming Li:** Writing – review & editing. **Hua Dong:** Writing – review & editing, Supervision. **Guang Ye:** Writing – review & editing, Supervision, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgement

Xuhui Liang would like to acknowledge the financial supports from the Chinese Scholarship Council (201806050051) and Mineralz (Part of Renewi). Zhenming Li would like to acknowledge Guangdong Provincial Key Laboratory of Intelligent and Resilient Structures for Civil Engineering (2023B1212010004).

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