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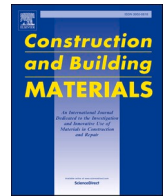
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Review

Production, characteristics, and utilization of rice husk ash in alkali activated materials: An overview of fresh and hardened state properties

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ABSTRACT

Vast amounts of rice husk ash (RHA) are generated annually, with most of them unutilized or disposed to landfills, resulting in serious environmental degradation. To avoid this, the use of RHA as precursors or co-binder in the development of alkali activated materials (AAMs) is viewed as a viable alternative to avert this significant problem. In this paper, the generation, properties, and utilization of RHA in AAMs are systematically and comprehensively reviewed. Furthermore, the physical, chemical, and mineralogical composition of several RHA were critically examined, and their impact on fresh and hardened state properties of AAMs and blended formulations are presented. Generally, the properties of RHA are influenced by the source material and the production process (pre-treatment, burning time, burning temperature, cooling rate, cooling duration and milling) which in turn affect their pozzolanic reactivity. Due to its high pozzolanic nature, low energy requirement and greenhouse gas emission during production, RHA is an environmentally friendly and cost-effective alternative cementitious material to produce AAMs. Various studies have reported the beneficial role of RHA on the mechanical, microstructural and durability properties of AAMs, especially when used at an optimal level. Overall, this review will provide valuable insight, direction, and recommendations for researchers and industrial sectors on the generation, recycling, characteristics, and utilization of RHA in the development of AAMs for potential construction applications.

1. Introduction

Portland cement (PC) is the most widely used cementitious material in the construction and building industry [1]. However, their continuous production and usage have resulted in adverse environmental impacts. For instance, the global production of PC was estimated to be 4.1 billion tons in 2017, which account for about 5-8% of global anthropogenic CO₂ emissions [2,3]. This amount is expected to rise due to a gradual increase in population and infrastructural development in developed and developing countries [4]. Besides the environmental impacts of PC, other limitations such as significant consumption of energy and excessive use of non-renewable, naturally occurring raw materials have raised serious concerns about its sustainability [5]. Therefore, there is

increasing demand for an alternative binder with lower carbon emissions that could at least partly or wholly replace PC.

In the search for alternative cementitious materials and binder, alkali-activated materials (AAMs) has received wide attention [6]. AAMs including those classified as geopolymers and inorganic polymers (IP) can be derived from a reaction between alkali activator (waste-based and non-waste based) and aluminosilicate precursors (such as fly ashes [7], metallurgical slags [6], mineral wools [8-10] and calcined clay minerals [11] or sometimes iron silicate precursors [12,13]). The development of AAMs using such materials can provide numerous benefits such as reduced CO₂ emissions and can also exhibit superior or similar properties in comparison to PC based binders [14-16]. For example, AAMs have been reported to exhibit higher resistance to

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thermal loads and aggressive conditions, making them attractive to the scientific and industrial community [17]. Over the years, ground granulated blast furnace slags (GGBFS) and fly ash (FA) are the most widely studied and utilized precursors for AAMs and are currently almost fully used as supplementary cementitious materials (SCMs) for cement production [1,18]. Therefore, other industrial residues such as rice husk ash (RHA) with different chemical and mineralogical compositions are currently being investigated for their suitability as alternative SCMs.

RHA is an agro-residue or byproduct generated from the milling of rice, which is often landfilled or underutilized, resulting in adverse environmental impacts and higher economic costs for the rice manufacturers [19]. The production of RHA has continued to increase due to the increased production of rice for consumption [20]. The global production of rice in 2020 was estimated at 499.31 million metric tons, and for every 1 kg of rice produced, approximately 0.28kg of rice husk is generated [20,21]. Over the years, rice husk produced has found limited application and are mostly used as fuel in the rice mills to generate steam for the parboiling process and approximately 25 % of the weight of this rice husk is converted into RHA [22]. With the ever-growing population and increasing demand for rice as important staple food worldwide, the amount of RHA emanating from cogeneration plants is expected to increase significantly. In the future, the number of available landfill spaces will be limited, making the disposal and effective management of the RHA difficult. Reuse and recycling of RHA as secondary raw materials in cement production or as precursors for AAMs have proven to be a sustainable alternative to avert this significant problem. Therefore, studies on the utilization of RHA as precursors and co-binders in AAMs have increased significantly over the last decade [19].

The chemical and mineralogical composition of RHA depends on the source material and the production process but usually contains around 85-90% amorphous silica; it is thus classified as class N pozzolan in accordance with ASTM C618 standard [23]. They exhibit excellent pozzolanic properties due to their high surface area and amorphous nature, making them suitable SCMs in cement and concrete production [19]. When incorporated as SCM in mortars and concrete, they could improve mechanical and durability properties, lower the cost of materials due to cement savings, and avoid landfilling costs and environmental degradation associated with the disposal of waste materials [20]. In this line, several studies have demonstrated that RHA can potentially replace PC partly or fully in a concrete mix, and the results were promising [24-27]. However, other factors such as physical and chemical properties of RHA, amount and concentration of the activating solution, curing conditions, and the water/cement ratio could also influence the mechanical and durability properties of the RHA-based mortars and concrete [19].

Despite the fact that a lot of studies and reviews have been carried out on the performance of RHA as a pozzolan, its impacts on fresh and hardened state properties of AAMs have not been sufficiently studied. The purpose of this review is to present a comprehensive, detailed, in-depth state-of-the-art review on the generation, characteristics, and utilisation of RHA in the AAMs. In addition, the review provides information about the origin, different generation techniques, processing conditions and properties of RHA used by various researchers and investigators and how it influences the properties of AAMs produced. This review will provide valuable information for researchers and industrial community on the utilization of RHA as precursors for AAMs for potential construction and building applications.

1.1. Objectives and methodology

The aim of the study is to review the generation, properties, and utilization of agro-industrial waste such as RHA in AAMs. The state-of-the-art review is presented on (i) origin and production of RHA (ii) physical and chemical characteristics of RHA (iii) impacts of RHA on fresh and hardened state properties of AAMs (iv) challenges and opportunities. In this review, all forms of RHA are considered including

low-carbon RHA, high-carbon RHA, ground and unground RHA as well as treated and untreated RHA. Scopus database being one of the most widely known and largest bibliometric information sources for peer reviewed studies was used to support the literature search. Science direct and Google Scholar were used as additional search engines. Several keywords were used to search for relevant and appropriate literature, and they include “alkali activated material” or “geopolymer” or “inorganic polymers” or “alkali activation” and this was combined with the word “rice husk” or “rice husk ash”. Also, we searched for specific words such as: generation, production, properties, utilization, earth construction, cement, concrete, mortar, building and construction. Furthermore, we searched based on the chemistry of the material to identify all studies related to alkali activation of silica-based materials to enable us cover large number of literatures on RHA utilization.

Original articles written in English and published in peer-reviewed journal were chosen to ensure high quality and proper overview. To ensure that the references found were related to RHA in AAMs, the abstract of all the references found using the search engines were read and the full text were accessed using the above-mentioned databases.

2. Rice Husk Ash (RHA)

2.1. Origin and production

Rice is an essential food item consumed by approximately half of the world's population [28]. They are grown in significant quantities in China, India, Indonesia, and other countries around the world. In many rice-producing countries of the world, rice husk is one of the most common waste or by-products generated in large quantities [20]. Rice husks are hard-protective layer that surrounds the paddy grain, which are separated from rice seed during the milling process [28]. They are lignocellulosic materials and it was estimated that during the milling process, about one ton of rice husk is produced from five tonnes of rice paddy and approximately 120 million tonnes of rice husk is produced annually [29]. The amount of rice husk generated is enormous, necessitating their reuse in bioethanol production and as fuel for heat production in industries [30]. The use of rice husk instead of coal as fuel in industries could lower the carbon emissions associated with coal combustion and support environmental preservation, thus complying with the European Union directive on waste management [31]. The combustion of rice husk when used as fuel in the boiler generates a by-product known as RHA, which accounts for about 20-25% of the weight of rice husk [19]. During the production of RHA, a combination of processing operations is involved which include pre-treatment, combustion method, separation process and milling. The combustion of rice husk is mostly carried out either by uncontrolled open combustion or controlled incineration and this influences the composition of RHA produced [32]. When produced through uncontrolled open combustion, the rate of crystallization is very high, resulting in ash with poor pozzolanic properties [33]. The significant amount of silica present in ashes originating from uncontrolled open combustion is mostly non-reactive [19]. Similarly, RHA produced from uncontrolled combustion in industrial furnaces also contains non-reactive silica which occurred as cristobalite and tridymite in the ashes [34]. They are mostly black in colour, attributed to the presence of excess carbon [35]. Furthermore, they have a considerably lower loss on ignition (LOI) compared to RHA from controlled combustion, indicating the presence of a higher amount of unburned carbon [34]. However, the emission of harmful gases associated with uncontrolled open combustion is a serious concern in many countries [36].

Controlled incineration of rice husk can improve the pozzolanic properties of RHA due to lower crystallization and higher formation of amorphous silica. Additionally, environmental issues associated with uncontrolled open combustion are minimized using this process. Controlled incineration of rice husk to produce RHA has been carried out in numerous controlled furnaces such as fluidized bed reactor, cyclonic

furnace, tubular reactor, muffle furnace, electric furnace, thermal gravimetric column, electric oven, fixed bed reactor, torbed reactor, and rotary tube furnace as reported by various authors [37,38].

The amount and nature of silica, carbon, and LOI of RHA is also influenced by the burning duration, burning temperature as well as the cooling rate and duration which in turn affects the physical and chemical properties of RHA [39]. It has been reported that when the combustion temperature is raised above 900 °C, the silica in RHA is transformed into crystalline nature [40]. Furthermore, prolonged heating above one hour at lower temperature of 700 °C can convert amorphous silica to crystalline silica [40]. A significant amount of crystalline silica was also formed when heated at an elevated temperature above 1150 °C [41]. The schematic diagram showing the generation process from rice plant to RHA are presented in Fig. 1. The origin and generation of RHA obtained from different studies are presented in Table 1.

During the combustion or burning process, substances such as alkali and metallic compounds present in rice husk can undergo melting reactions causing impurities in the RHA produced [20]. These impurities if not removed can cause a reduction in the specific surface area and affect the pozzolanicity of RHA.

Another important step in the production of RHA is milling. The milling of RHA is carried out for different purposes. The gradation and particle size to be achieved also determines the grinding method to be used. However, milling is important to decrease the particle size, increase the specific surface area and pozzolanic activity of RHA [53,60].

2.2. Characteristics of rice husk and RHA

2.2.1. Pretreatment of rice husk

Rice husk usually contains lignin, cellulose, hemicellulose, and moisture content [29]. However, the properties of rice husks obtained can vary widely with rice variety, geographical location, climate, soil chemistry, and type of fertilizer employed in paddy growth [19]. Compared to other silica-based waste materials such as sugarcane bagasse and coconut shells, rice husks have significantly higher ash content, limiting their utilization in certain applications [61]. Depending on the intended application, different properties are considered important in the selection of rice husks as suitable precursors. For

instance, in the production of biofuel, a lower ash content is required while in AAM production, a higher surface area is considered important [62]. In addition to other processing operations, pre-treatment of rice husk can also influence the physico-chemical properties of RHA. Some authors have reported that the ash content in rice husk can be lowered up to 92 wt.% by pre-treating them with NaOH, and this depends on the soaking time, pre-treatment ratio, alkaline concentration, and process temperature [63]. Also, the pre-treatment process using NaOH can cause a reduction or complete removal of silica from rice husks due to the reaction of silica with NaOH to form water soluble sodium silicate [63]. The sodium silicate produced is then extracted from the rice husks during the water washing stage [64]. Furthermore, leaching of rice husk using acid solutions prior to their combustion has also been explored and an increase in surface area, improved pozzolanicity and removal of metallic impurities was observed [65]. Meanwhile, the kind of pre-treatment carried out on rice husk can have a significant impact on the mineralogical and chemical composition of RHA produced.

2.2.2. Physico-chemical properties of RHA

RHA is a residual ash formed from the combustion and burning of rice husk [66]. RHA is mostly X-ray amorphous, ultrafine in size, higher specific surface area, highly porous and chemically reactive [19]. Specific gravity, specific surface area, particle size, and pozzolanic activity index of RHA are important physical properties that needs to be investigated prior to its usage as SCMs/ precursors. In addition, particle size is crucial to the pozzolanic activity of RHA. The lower the particle size, the higher the surface area and reactivity. Meanwhile, a study reported that milling does not have any significant influence on the chemical composition of RHA. However, milling can decrease the particle size and increase the surface area of RHA [53]. For instance, Cordeiro et al. (2011) reported that the milling of RHA lowers the pores in the microstructure because of the disintegration of the silica walls in RHA [67]. The surface area and water demand were also reported to increase with the increase in milling time, thereby influencing the behaviour of RHA in AAMs [68].

Thus, the physical properties of RHA must be optimized to achieve suitable properties, especially when it is to be used in AAMs. The physical properties of RHA used by various researchers are shown in

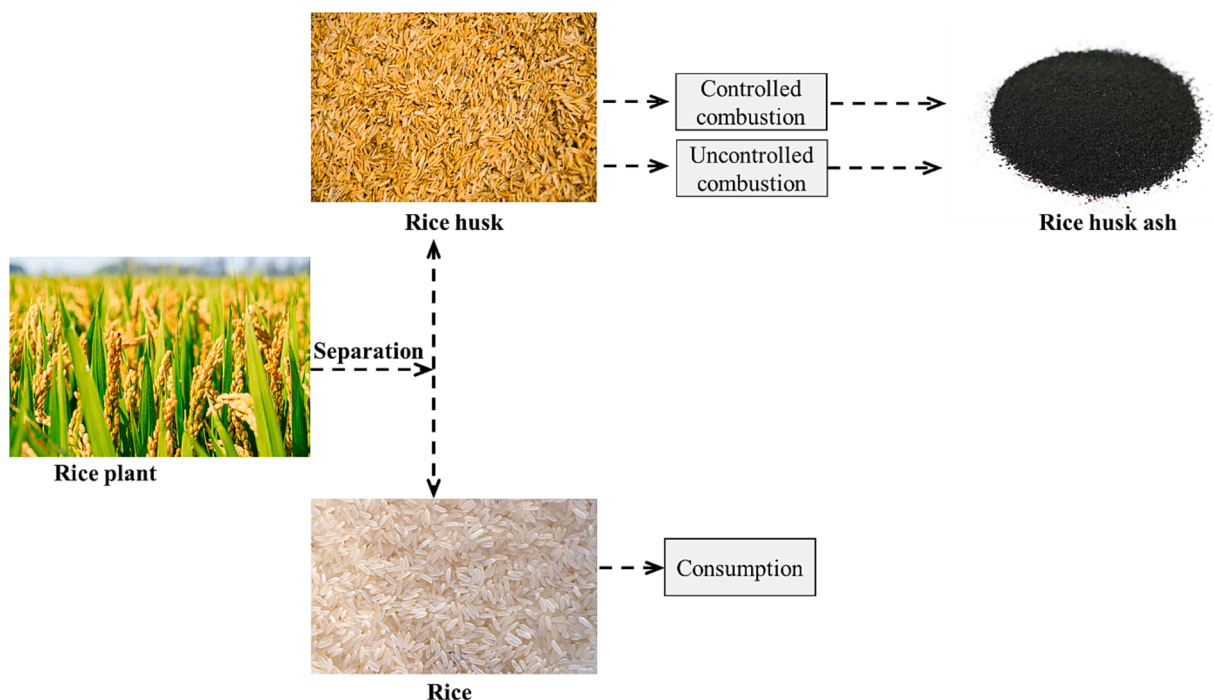


Figure 1. Schematic diagram of the generation of rice husk ash.

Table 1
Origin and production of RHA reported by different researchers.

Burning method	Burning temperature	Burning duration	Colour	LOI	References
Incineration	800-900 °C	7 h	White	4.04	[42]
Incineration	500 °C	2 h	Off-white	2.27	[43]
Controlled burning	600 °C	3 h	White	-	[44]
Open burning	-	1 h	Grey	3.83	[45]
Controlled combustion	650 °C	1 h	-	2.10	[35]
Burning in muffle furnace	700 °C	2 h	-	-	[46]
Controlled burning in gas furnace	800 °C	-	White	-	[47]
Controlled burning	450 °C	-	Brownish white	5.32	[48]
Controlled burning	500–700 °C	30 min	White	3.21	[39]
Open burning	650 °C	-	-	3.7	[49]
Open burning	422 °C	72 h	-	-	[50]
Controlled burning	650 °C	1 h	Whitish grey	5.91	[49]
Combustion in fluidized bed reactor	-	-	-	5.1	[37]
Combustion in Torbed reactor	850 °C	-	-	2.3	[37]
Open burning	600 °C	-	Whitish grey	-	[51]
Pyrolysis	800 °C	40 min	Off-white	0.54	[52]
Open burning	-	1 h	Grey	3.83	[45]
Controlled burning	700 °C	-	Grey	4.05	[53]
Controlled burning	600-850 °C	-	Dark grey	11.9	[54]
Controlled burning	500 °C	2 h	Brown	-	[55]
Burning in muffle furnace	600 °C	2 h	-	4.77	[56]
Uncontrolled combustion	600- 8500 °C	60 h	Gray	-	[57]
Burning in automatic microwave incinerator	500 °C	2 h	-	5.02	[58]
Burning in annealing furnace	660-750 °C	1 h	-	2.65	[59]

Table 2. As shown in this table, specific gravity of RHA vary between 1630 to 3100 kg/m³ depending on its fineness. The specific surface area and pozzolanic activity index also varies from one study to the other and this is expected to affect the performance of AAMs produced.

RHA produced from uncontrolled combustion are mostly crystalline in nature and exhibit poor pozzolanic properties [20]. On the other hand, RHA produced under controlled combustion contains 80-96% amorphous silica, making them suitable as cementitious binders [54]. The pozzolanic activity of RHA is intimately linked to the amorphous structure of the silica. Due to the high pozzolanic activity and fineness, RHA has potentially been used in high strength concrete and possibly as filler in polymers [83]. Typically, RHA is mainly composed of organic matter, unburned carbon, metallic oxides, and silicon dioxide [19]. However, the chemical composition of RHA may vary from one region to the other depending on the source material and the processing operations. The chemical composition of RHA extracted from publications of various investigators are listed in Table 3. As shown in Table 3, silica is the principal oxide present in RHA, and this varies between 80 to 95%. Furthermore, the LOI also varies from 1 to 12%.

3. Utilization of RHA in AAMs

The need to reduce the emissions caused by cement production and usage, as well as incessant exploitation of non-renewable natural resources provide justifiable reason to find an alternative binder that is sustainable and has less carbon footprint. The use of agro-industrial wastes such as RHA as precursors for AAMs could be an alternative pathway to valorise industrial wastes and reduce environmental pollution associated with cement production. The presence of the significant amount of silica and alumina in RHA makes it suitable for use as a sole precursor or as co-binders in AAMs. Like other AAMs, the properties of RHA-based AAMs are influenced by the activator types and concentration, curing conditions, water to binder ratio, and RHA replacement level, which is discussed in detail in Section 4. Meanwhile, RHA-based AAMs can exhibit superior properties compared to PC-based concrete in terms of mechanical and durability properties [28]. Table 4 shows the mix design and curing regimes of RHA based AAMs studied by different researchers.

4. Influence of RHA on fresh and hardened state properties of AAMs

4.1. Workability

Workability is an important parameter that can be used to assess the ease and homogeneity of fresh matrix immediately after mixing [15,20]. The workability of mortar or concrete must not too fluid or excessively stiff otherwise production efficiency will be adversely affected [99]. Meanwhile, it has been reported that the workability of AAMs or geopolymer is mostly influenced by precursor particle size, alkali amount, type and concentration, water to binder ratio, characteristics of the ingredient, and the mix design. Depending on the fineness and replacement level in the matrix, the use of RHA can significantly influence the workability of the mixture [20]. With increasing fineness and replacement level, the workability of the mixture is decreased significantly [100]. The higher the fineness, the higher the specific surface area and the higher the water demand. Furthermore, due to the high specific surface area of RHA, it tends to have higher reactivity which can consequently lower the flowability of the mix. Nuaklong et al. (2020) studied the effect of RHA on the workability of high calcium based geopolymer concrete containing recycled coarse aggregates; their results indicated that with an increasing percentage of RHA incorporation in the concrete matrix, the slump flow decreased [101]. Similar observations were reported in another study when assessing the effect of RHA on properties of FA-GGBFS based AAMs. The slump value of the resulted mixture gradually decreased with an increasing percentage of RHA incorporation. The detrimental effect of RHA on the workability of the concrete is due to its high water demand [102] and angular particle shape [103]. Furthermore, it is also stated that when FA is replaced by RHA in the range of 25-30%, a significant reduction in slump values are noticed, hence it is suggested that the use of superplasticizers or high range water reducing admixtures could improve the workability of the matrix [104]. In addition, another study conducted by Detphan and Chindaprasirt (2009) using FA and RHA, revealed that the flow of geopolymer mortar reduced significantly with higher content of RHA, ascribed to the high fineness and cellular structure of RHA [105]. Das et al. (2021) studied the effect of ultrafine RHA in FA-GGBFS based AAM, with RHA incorporated into the matrix in the proportion of 1%, 3%, 5%, 7% and 10% to replace of FA [106] (Fig. 2). The results obtained from this study show a reduction in the slump values with increase in RHA addition which is in line with other research findings discussed earlier. The primary reason for the reduction in slump is attributed to the high fineness of RHA. In addition., it is also suggested that the high specific surface area of RHA could also be responsible for the reduction in workability [101].

Table 2
Physical properties of RHA used by various investigators.

Burningmethod	Millingtime (minute)	Specific gravity (kg/m ³)	Blaine fineness (m ² /kg)	BET specific surface area (m ² /kg)	Median particle size (µm)	Pozzolanic activity index (%)	References
Burning in boilers	120	2293	1564	33672	4.3	109	[69]
Burning in muffle furnace	-	2200	-	430	-	-	[70]
Incineration	-	-	-	67330	16.76	-	[71]
-	-	1630	-	-	150	-	[72]
-	-	2300	-	-	25	-	[73]
Controlled incineration	-	-	-	-	36.9	-	[74]
-	60	-	-	11000	34.9	-	[75]
Burning in automatic microwave incinerator	-	2230	-	7810	16.22	92.3	[58]
Incineration	-	3100	-	23320	-	-	[68]
-	-	2190	-	52280	-	-	[76]
-	-	2090	697.5	12700	6.72	-	[77]
Controlled burning	-	2270	-	25210	5.7	-	[78]
Burning in combustor	60	2040	-	11350	5	-	[79]
Controlled calcination	30	2140	-	78280	22.84	-	[80]
Controlled burning	15	2190	-	64700	-	123.7	[55]
Burning in furnace	-	-	-	18940	-	-	[81]
Burning in ferro-cement furnace	180, 270 and 360	2110	-	27400, 29100, 30400	-	-	[82]

4.2. Setting time

The setting time of concrete or mortar is an important parameter to be assessed, especially when it is to be considered for building and construction applications. It should be reasonable to allow suitable time for mixing and casting operation [99]. However, when the setting time is excessively fast or longer, i.e., more than a couple of hours and days, the production efficiency is adversely affected [99]. The use of RHA as precursors or co-binder can greatly influence the setting time of the AAMs due to its high silica content, which increases the Si/Al ratio of the mixture substantially. It is already established that when the Si/Al ratio increases in the alkali environment, the high silicate species available to develop oligomers of silicates by condensation has increased, and thereafter it takes more time for condensation of silicate oligomers to form geopolymer network which ultimately delays the setting process [107]. Zhu et al. (2019) investigated the setting times of MK geopolymer pastes as a function of RHA content and established a relationship between the RHA additions and the setting time of AAMs. In the mentioned study, it was clearly observed that the setting time was delayed with the incorporation of RHA (Fig. 3) [22]. The delay in setting time was attributed to the slower dissolution of SiO₂ from the layered structure of RHA which consequently retards the geopolymerization process [28]. Billong et al. (2018) investigated the consistency and setting behaviour of MK/RHA blend. The results showed that the consistency of MK/RHA blend was higher than the mixture having only MK, attributed to the dilution effect of RHA irrespective of whether a constant or lower solid to liquid ratio (S/L) was used [108]. Furthermore, an increase in RHA content was observed to delay the setting time of the mixture. For instance, blends containing 0, 12.5, 25, and 50% replacement of MK by RHA had 128, 144, 140 and 290 min as initial setting times, respectively (Fig. 3). The final setting time of the blends having 0 to 25% RHA was 230 mins, while that of the paste containing 50% RHA was 450 mins. This evidently shows that RHA acted as a retarding agent in MK based AAMs, with the effect more visible in mixtures containing large proportions of RHA. The retarding effect of RHA could be due to high SiO₂/Al₂O₃ ratio in the blend as discussed in several studies which contributes to the inhibition of alkali activation process or geopolymerization. During the alkali activation process, the precipitation of Si-Al species lowers the contact between the reactive precursor material and the alkali activating solution [108]. In contrast, a study reported that the incorporation of RHA into MK and red ceramic waste blend improves the setting time of the AAMs. A fast setting observed with high RHA percentage was ascribed to the high reactive silica content of RHA [109]. In addition to the silica content provided by RHA, it is also possible that the

setting performance of the blend may have been influenced by the two precursors used for the alkali activation process which can make the role of RHA in setting behaviour quite challenging to comprehend. On the other hand, the use of RHA as alternative activator can significantly influence the setting time of the matrix, with the initial and final setting time shorter than the samples activated with commercial activators. For instance, it was reported that using RHA-NaOH produced locally as an alternative activator for the synthesis of geopolymer cured at room temperature yielded promising results [28]. The setting time of the matrix was good, and the demolding of samples was possible after 24h, indicating the activation potential of RHA based activator.

4.3. Compressive strength

The compressive strength of mortars and concretes is perhaps one of the most important engineering properties of building and construction materials [110,111]. For instance, the minimum compressive strength requirement for building materials can vary depending on standards and the working conditions that will be applied. To this end, several residues such as RHA are being investigated as precursors and additives to improve the mechanical properties of AAMs. It is worth stating that the high specific surface area and reactivity of RHA (amount of amorphous silica present) play a significant role on the mechanical properties of AAM. Chao-Lung et al. (2011) evaluated the effect of RHA addition on the mechanical, microstructural, and durability properties of concretes [112]. They observed that increasing the RHA content from 10 to 20 wt. % caused an increase in the capillary pores due to the formation of the CH phase on the interface, resulting in a less compact structure at an early age. However, at later ages, the pozzolanic reaction increases, thus decreasing the CH phase formed and enhances the performance. In contrast to the previous reported studies, some literatures discovered that reactive silica from RHA generally improves the structural build up and mechanical performances of AAM during early and later ages when used at a certain dosage. For instance, Nbillong et al. (2001) replaced MK with RHA from 25 to 75 wt.% and activated with Ca(OH)₂ to produce mortars [113]. In the mentioned study, it was revealed that the mechanical performances increased for all the formulations from 7 to 56 days. This strength development was attributed to the reaction between reactive silica and hydrated lime, resulting in the formation of CSH and C₂ASH₈; consequently, improving the mechanical performance. Furthermore, the incorporation of RHA in alccofine based AAMs has shown promising results in terms of compressive strength enhancement. It was observed that when the replacement ratio of alccofine with RHA was increased, the compressive strength of the AAMs increased

Table 3
Processing parameters and chemical composition of RHA used by various investigators.

Burningmethod	Burningtemperature	Burningduration	Cooling	Elemental oxides (wt.%)										References
				SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	SO ₃	Na ₂ O	K ₂ O	Others	LOI	
Burning in boilers	600-850 °C	-	-	82.6	0.4	0.5	0.9	-	0.1	0.1	1.8	-	11.9	[69]
Burning in muffle furnace	-	4h	-	94.8	0.52	0.13	-	-	-	-	-	-	-	[70]
Incineration	800 °C	-	-	89.34	0.45	0.40	0.76	0.49	0.90	-	4.98	-	-	[71]
-	-	-	-	80.2	2.39	3.2	1.51	0.59	-	0.34	1.8	-	9.30	[72]
-	-	-	-	91.79	2.12	0.80	1.27	0.67	-	0.14	0.76	0.89	1.56	[73]
Controlled incineration	600 °C	3h	-	94.4	0.2	0.2	-	-	-	-	-	-	2.8	[74]
-	-	-	-	89.6	-	0.75	1.49	-	0.67	-	7.05	-	12	[75]
Burning in muffle furnace	600 °C	2h	Room temperature	86	1.16	1.85	0.64	0.77	0.32	1.14	2.54	-	4.77	[56]
Uncontrolled combustion	600-8500 °C	60h	-	90.16	0.11	0.41	1.01	0.27	0.12	0.01	0.65	-	-	[57]
Controlled combustion	-	-	-	92.46	0.26	0.30	1.63	0.38	0.11	1.24	-	-	-	[32]
Burning in automatic microwave incinerator	500 °C	2h	-	90.75	0.75	0.28	0.87	0.63	0.33	0.02	3.77	-	5.02	[58]
Burning in outdoor furnace	-	2h	Ambient temperature	86.02	0.36	0.16	1.12	0.39	-	1.15	-	-	-	[84]
Incineration	-	-	-	86.81	0.50	0.87	1.04	0.85	-	0.69	3.16	-	4.6	[68]
-	-	-	-	87.4	0.4	0.3	0.9	0.6	0.4	0.04	3.39	-	4.6	[76]
-	-	-	-	94	1.2	0.37	2.93	0.60	0.30	-	0.50	-	-	[85]
Burning in a furnace	-	-	-	87.55	0.39	0.20	1.03	0.67	0.57	0.05	2.85	-	3.14	[77]
Controlled calcination	600 °C	2h	-	83.05	1.82	0.58	0.69	3.59	0.34	0.13	5.65	-	-	[86]
Controlled burning	500-800 °C	-	-	87	0.8	0.4	1.2	0.6	0.4	0.4	2.63	-	3.7	[78]
Burning in combustor	-	-	-	90.93	0.11	0.19	0.36	0.33	0.15	0.02	1.97	-	5.1	[79]
-	-	-	-	94.1	0.037	0.046	0.49	0.27	0.057	0.052	1.79	0	-	[87]
Controlled calcination	600 °C	2h	-	92.33	0.18	0.17	0.63	0.49	-	0.07	0.15	2.57	-	[80]
Controlled burning	500 °C	2h	-	91.56	0.19	0.17	1.07	0.65	0.47	0.16	3.76	-	-	[55]
-	-	-	-	89.47	0.83	0.53	0.68	0.37	0.12	0.22	0.17	-	7.61	[88]
-	700 °C	3h	-	91.79	2.12	0.80	1.27	0.67	-	0.14	0.76	0.89	1.56	[89]
Burning in furnace	700 °C	1h	Room temperature	88.07	1.35	0.22	1.04	0.74	0.49	1.15	2.02	-	2.61	[81]
Burning in annealing furnace	660-750 °C	1h	Rapidly cooled with water	91.94	0.29	0.25	1.05	0.44	0.37	0.11	1.69	-	2.65	[59]
Controlled burning	650 °C	2h	Slow cooling	92	0.31	0.38	0.97	0.47	-	0.20	3.87	-	0.76	[90]
Controlled burning	650 °C	2h	Slow cooling	88.3	0.25	0.37	0.91	0.40	-	0.16	3.63	-	5.02	[90]
Uncontrolled combustion	400 °C	-	-	23.81	0.09	0.12	0.26	0.13	-	0.07	1.02	-	74.33	[90]
Uncontrolled + Controlled combustion	650 °C	4h 15 min	-	87.32	0.22	0.28	0.48	0.28	-	1.02	3.14	-	2.10	[35]
Controlled burning	650 °C	1h	-	89.61	0.04	0.22	0.91	0.42	-	0.07	1.58	-	5.91	[49]
Controlled burning	600-850 °C	-	-	82.6	0.4	0.5	0.9	-	0.1	0.1	1.8	-	11.9	[54]
-	-	-	-	92.95	0.31	0.26	0.53	0.55	-	0.08	2.06	0.12	1.97	[91]
-	-	-	-	91.3	1.4	0.6	2.4	2.1	-	0.3	1.9	-	-	[92]
-	-	-	-	93.00	0.35	0.23	1.31	0.41	0.09	0.15	1.61	-	1.90	[93]
Burning in muffle furnace	650 °C	1h	-	87.89	0.19	0.28	0.73	0.47	-	0.66	3.43	-	4.36	[94]
Controlled calcination	600 °C	2h	-	81.4	0.26	0.93	1.02	1.47	-	0.18	6.79	-	3.13	[95]
-	-	-	-	87.86	0.68	0.93	1.30	0.35	-	0.12	2.37	-	-	[96]
-	-	-	-	77.19	6.19	3.65	2.88	1.45	-	-	1.82	-	5.43	[97]
Controlled burning in muffle furnace	500 °C	5h	-	87.22	0.70	1.68	2.12	1.18	0.04	0.20	1.12	0.46	1.06	[98]
-	-	-	-	88.46	0.17	0.31	0.75	0.47	0.05	0.22	1.69	-	9.22	[27]

Table 4
Mix design and curing regimes of RHA based AAMs from literatures.

Precursor	Activators	NaOH/ Na ₂ SiO ₃	Conc. NH/NA	Water/ BinderSolution/ Binder	Curing regime	Reference
RHA and basalt fibres	NaOH and Na ₂ SiO ₃	1.0	12M	0.5	Controlled room temperature at 25°C	[129]
FA, GGBFS and Ultrafine RHA	NaOH and Na ₂ SiO ₃	2.0	14M		Ambient curing	[106]
RHA and ultra-fine slag	NaOH and Na ₂ SiO ₃	2.5	16M	0.38, 0.42 and 0.46	Ambient curing	[123]
FA, GGBFS, RHA	NaOH and Na ₂ SiO ₃	.0	12M		Ambient curing	[103]
FA (class C), NS and RHA	NaOH and Na ₂ SiO ₃	0.6 and 1.0	10M	0.6	Ambient curing	[101]
Metakaolin (MK) and RHA	NaOH and Na ₂ SiO ₃	—	—	0.7	50°C steam curing	[22]
MK and RHA	NaOH and Na ₂ SiO ₃	—	—	0.5	Room temperature until demoulding and steam curing at 50°C until the date of testing.	[121]
MK and RHA	NaOH and Na ₂ SiO ₃	—	—	0.7	Room curing temperature for 24 h before demoulding and cured at controlled environment of 20 ± 0.5°C and 95 ± 2% relative humidity	[120]
Sludge and RHA	NaOH and Na ₂ SiO ₃	—	8-14M	1.0, 1.1 and 1.2	Ambient curing	[122]
Cement kiln dust, Silica fume and RHA	NaOH	—	10M	1.4	Cured at 60°C followed by ambient curing	[135]
GGBFS and RHA	NaOH and Na ₂ SiO ₃	2.50	10M	0.4	80°C for 24 h followed by ambient curing	[88]
FA and RHA	NaOH and Na ₂ SiO ₃	1.0	12M	0.6	80°C for 24 h followed by 40°C for 72 h then under open air till the day of testing.	[118]
GGBFS and RHA	NaOH and Na ₂ SiO ₃	2.50	12M	0.25	Ambient curing	[119]
RHA	NaOH	—	12-16M	0.6-0.7	80°C for 24h followed by ambient curing	[126]
RHA, GGBFS and FA	NaOH and Na ₂ SiO ₃ , Na ₂ CO ₃ and quick lime	—	—	—	Moist cured inside molds at room temperature for 24 h followed by room temperature curing in sealed condition (> 85% relative humidity)	[136]
Alcofine and RHA	NaOH and Na ₂ SiO ₃	—	8-16M	—	90°C for 24 hours followed by ambient curing and ambient curing of 27°C	[102]
FA and RHA	NaOH and Na ₂ SiO ₃	—	8-14M	0.37	35°C and 50% relative humidity	[117]
RHA (one-part AAM)	sodium aluminate	—	12M	0.44-0.54	40°C at oven until age of testing	[137]
GGBFS and Black RHA (BRHA)	NaOH and Na ₂ SiO ₃	2.5	8M	0.40	60 °C for 16 h (8 h with mould and 8h without mould) followed by ambient curing	[124]
Red mud (RM) and RHA	NaOH	—	2, 4 and 6M	1.2	Ambient curing	[115]
FA and microwave incinerated RHA	NaOH and Na ₂ SiO ₃	2.51	8M	0.51	Ambient curing (35+1°C); external exposure (55+1 °C), and oven curing (65°C for 24 h)	[71]
MK, RHA and RM	NaOH and Na ₂ SiO ₃	4	5M	0.97	(70 °C) for 24 h followed by ambient conditions curing for 60 days	[138]
Laterite and RHA	NaOH and Na ₂ SiO ₃	1	8M	0.6-1	First group cured at room temperature (28 °C)Second group cured at room temperature for 24 h followed oven curing at 80° C for 24 h	[130]
Laterite,MK,Basalt and RHA	NaOH and Na ₂ SiO ₃	0.56	6 M	0.6-0.75	24 – 72 °C for 8h for sample using unheated lateriteAmbient temperature for samples using calcined laterite	[139]
Waste fired clay brick andRHA	NaOH and Na ₂ SiO ₃	1	8 M	0.6-1	Room temperature (28 °C)	[140]

significantly [102].

Moreover, it was noticed that the curing temperature also plays a vital role in silica dissolution from RHA into the matrix, and thus when cured at elevated temperature, the compressive strength of AAMs amplifies notably. A compressive strength of 71 MPa was obtained from a mixture of 400kg RHA with 40kg of alcofine, cured at 90°C, and activated with 14M NaOH solution. However, when the mixture was cured at ambient temperature, the rate of strength development was quite slow and gradual, but when the molarity of the activating solution was increased, the strength development was hastened, which is in line with other findings [114]. The enhancement in mechanical properties due to the incorporation of RHA was partly ascribed to the fineness of RHA which plays an important role in strength development mechanism of the AAMs. Owing to the fine particle size of RHA, the specific surface area is higher, and the reactivity increases, thus enhancing the dissolution and the polycondensation reaction of the matrix, resulting in higher strength. From the literature examined so far, It is quite obvious that by increasing the RHA content and dosage in the mix, the total silica in the matrix is increased, forming more Si-O-Si bonds in the matrix; it is theoretically believed that Si-O-Si bonds are quite stronger than that of Si-O-Al or Al-O-Al bonds [115,116]. Therefore, the compressive strength

was enhanced due to the increase in a structural build-up of tetrahedral aluminosilicate gel phase, specifically N-A-S-H or C-A-S-H. For instance, the change in compressive strength with the incorporation of RHA as a replacement to FA in AAMs formulation are shown in Fig. 4. The compressive strengths of the samples containing 30%, 35%, 40%, and 50% RHA exhibited 72.1%, 97.8%, 87.7%, and 78.2% increase in compressive strength when compared to samples without RHA [117].

Yomthong et al. (2019) investigated the replacement of 3 wt.% FA with RHA, activated with NaOH and Na₂SiO₃ and cured at elevated temperature, and compared with reference samples (with no RHA). The AAMs containing RHA exhibited a higher compressive strength of 42.52 ± 4.86 MPa, whereas the reference samples showed a compressive strength of 35.91 ± 3.30 MPa [118]. A similar observation was reported when RHA was incorporated into GGBFS based AAMs. The compressive strength of the AAMs increased with an increase in RHA addition up to 15 wt.%, achieving a compressive strength of 69.4 MPa after 90 days of curing, which was approximately 10% higher than the reference mix containing no RHA [88]. The lower strength achieved in AAMs produced using GGBFS only was due to a lower amount of silica species. Thus, when RHA is incorporated into the GGBFS based matrix, the possible formation of siloxo bridges (Si-O-Si-O) chain is highly increased than the

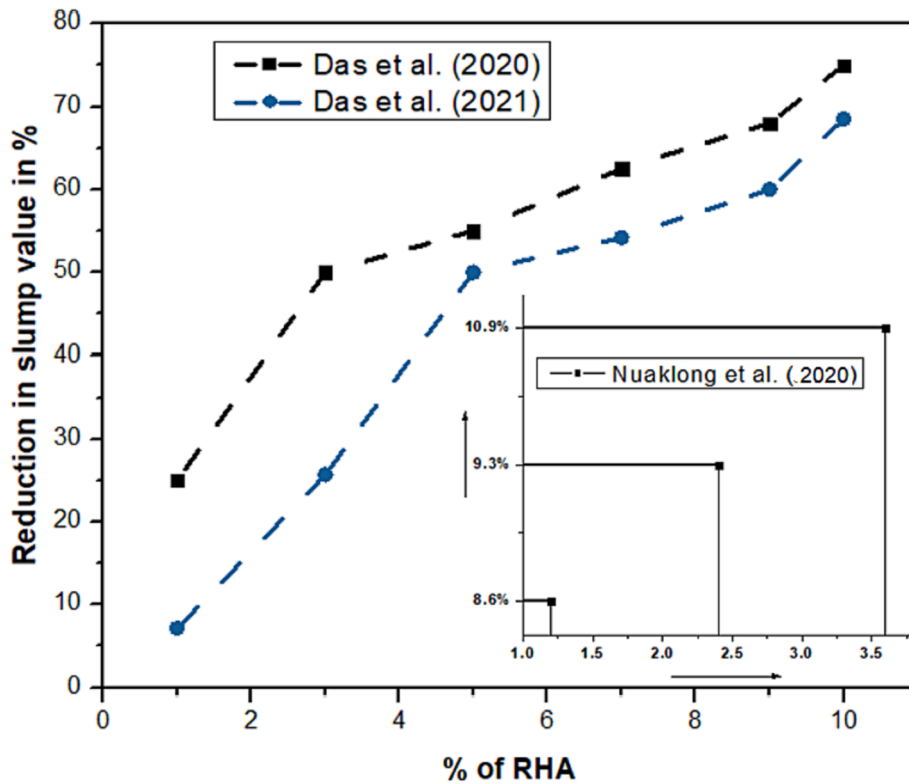


Figure 2. Effect of RHA on the slump values of AAC [101,103,106].

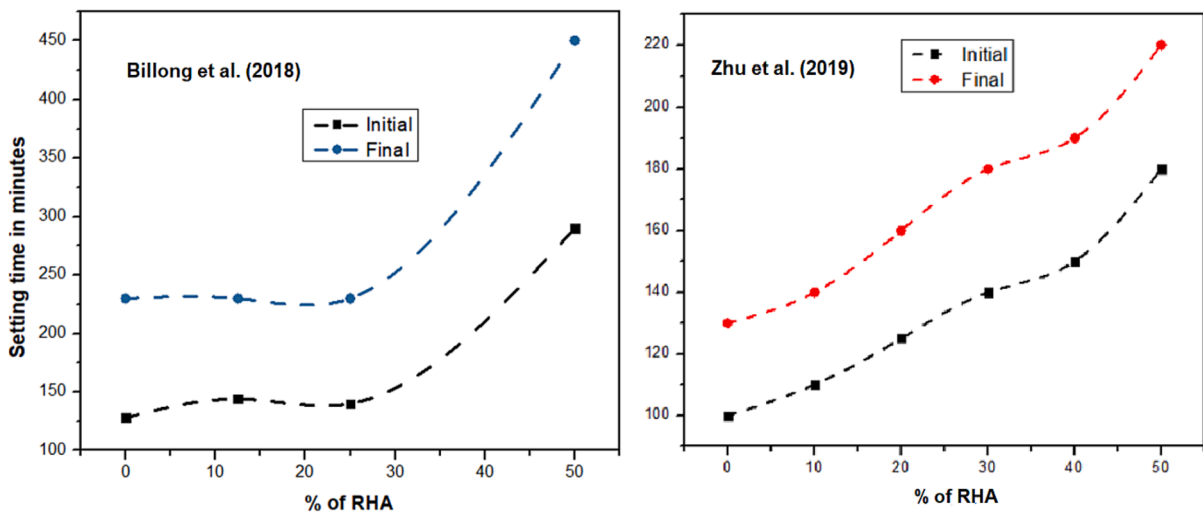


Figure 3. Effect of RHA on the setting time of AAMs [108].

Si-O-Al or Al-O-Al formation (since GGBFS has high alumina content, Al-O-Si or Al-O-Al would be generated in the absence of adequate silica species). Similarly, another study conducted using a mixture of GGBFS and RHA stated that the inclusion of RHA enhances the mechanical strength of GGBFS based AAMs. The CaO present in GGBFS reacts with the silica liberated from RHA to form C-S-H gel along with some amount of C-A-S-H. Moreover, the hydration process releases some heat which facilitates the formation of both C-A-S-H and N-A-S-H, which eventually creates a densified 3-D network of silico-aluminate structure improving the compressive strength [119]. It is obvious that such significant strength improvements in AAMs by addition of RHA observed in various studies may be attributed to: (a) the formation of stronger gel phases due to Si-O-Si bond synthesis mechanism owing to high silica species leached

from RHA (b) high RHA content associated with high specific surface area resulted in high ductile final products, thus improving the strength properties. Meanwhile, the change in compressive strengths with the incorporation of RHA as a replacement to MK in AAMs formulation at the curing ages of 7d and 28d (cured in steam curing conditions at 50 ± 2 °C for the entire period until testing) are displayed in Fig. 5. For samples made with 100% MK, the compressive strength was 30.08 MPa after 7 days of curing and this increased to 46.45 MPa as the curing age increased to 28 days. A sharp increase in compressive strength of the AAMs was noticed with the addition of RHA. The replacement of MK with 10, 20, 30, 40 and 50 wt.% of RHA resulted in 46.3%, 62.2%, 65.0%, 63.4%, and 65.0% increase in compressive strength respectively at 7 days of curing while after 28 days, the compressive strength further

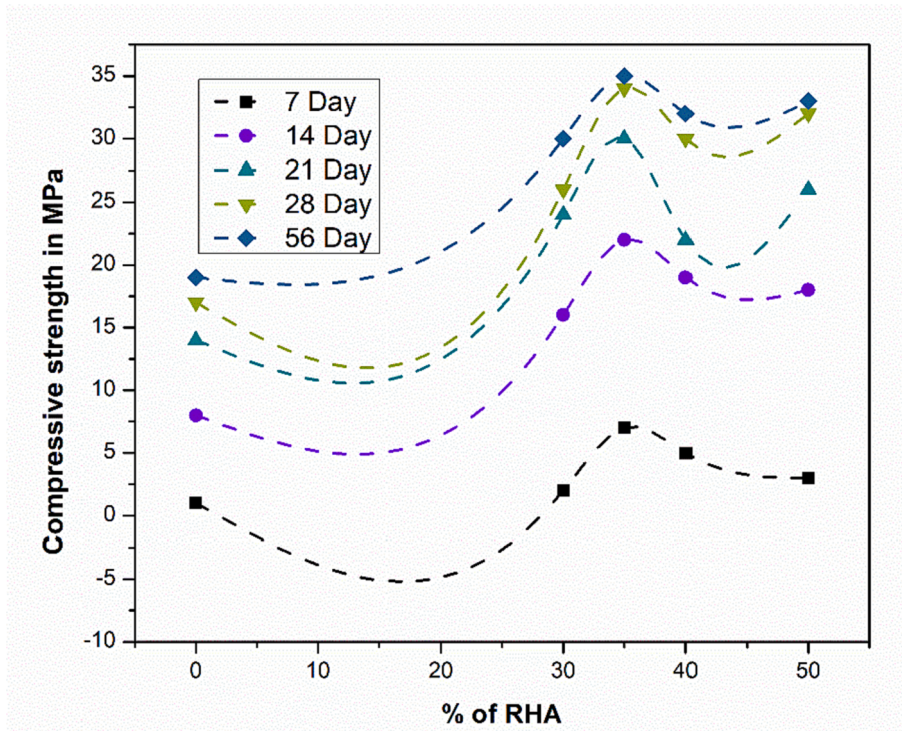


Figure 4. Effect of RHA on the compressive strength of AAM at different age [117].

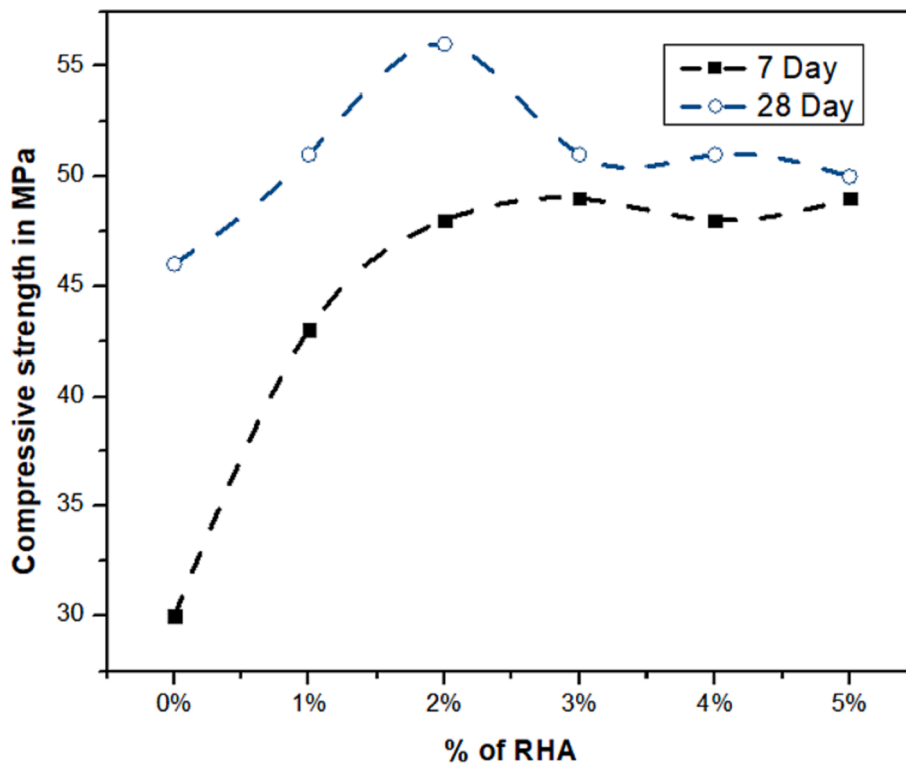


Figure 5. Effect of RHA on MK-AAM at different curing age [22].

increased by 10%, 21.7%, 8.9%, 9.5%, and 8.4% respectively. All mixtures containing RHA gained a considerable amount of strength as compared to the mix that contained no RHA, both at 7 and 28 days [22]. Nonetheless, the early strength gain mechanism of the AAMs is ascribed to the high reactivity of RHA.

Liang et al. (2019) investigated the effect of RHA incorporation on

the mechanical performance of MK based AAMs. It was reported that the compressive strength of AAMs improves with increasing dosages of RHA up to 30%, beyond which it starts to have a detrimental effect. As the dosage of RHA was increased to 40%, a sharp drop in compressive strength was observed as shown in Fig. 6[120]. It is worth stating that the mixture composition containing 30% RHA achieved the highest

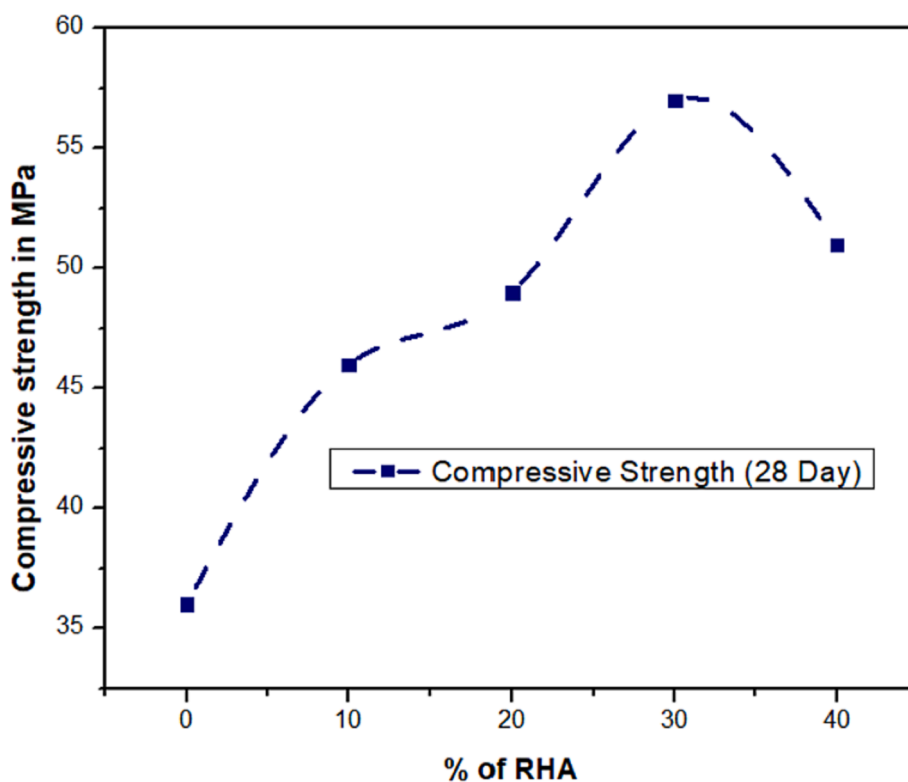


Figure 6. Compressive strength of MK-AAM with RHA [120].

compressive strength, which was 57.18% more than that of the strength achieved by the mix without any RHA. This specifies that the RHA made a significant contribution to the compressive strength development of AAMs, likely associated with the additional reactants provided by RHA.

Similar observations were reported by Liang et al. (2019) while investigating the influence of RHA addition on the waterproof property of MK based geopolymer. Since the replacement ratio was similar, the rate of compressive strength development was identical with previous observations. [121]. Furthermore, an enhancement in compressive strength development of non-calcined sludge (NCS) based AAMs was observed with the addition of RHA, and the compressive strength keeps increasing up to RHA/NCS ratio of 0.35. This improvement in strength is associated with the provision of reactive silica by RHA, which forms strong Si-O-Si structures or by the micro-filling effect of RHA, thus enhancing the degree of densification of the resulted matrix [122]. Also, Jindal et al. (2020) investigated the effect of ultrafine slag addition on the mechanical properties of RHA based AAMs. The results of the study indicated that the compressive strength of the mixtures increased with the increase in RHA and ultrafine slag addition. For instance, the early age compressive strength of the mixture, i.e., 3- and 7-days strength, increased by 50–100% and 120–180%, respectively, with an increase in RHA incorporation from 350 kg/m³ to 370 kg/m³ and 400 kg/m³. However, the late age strength development was not that affected by the RHA. Only 20–30% increase in compressive strength was noticed with increased RHA content for samples containing no ultrafine slag [123]. The incorporation of BRHA in GGBFS based AAMs, activated with a mix of NaOH and sodium silicate activators has also been investigated. The effect of BRHA was investigated with three different replacement levels, with the highest strength occurring in samples with 10% BRHA content. However, with an increase in replacement level to 20 and 30% BRHA content, strength reduction in AAM was noticed [124]. It is possible that 10% BRHA content is the optimum replacement level needed for adequate silica supply mechanism in the mixture. Above this, there is excess silica inclusion which might have caused strength reduction. Moreover, the BRHA contains an excessive amount of unburnt carbon,

and these carbon particles also might be responsible for such negative impact on compressive strength development.

On the other hand, some studies also investigated the use of RHA as the sole binder precursor in the development of AAMs. It was reported that a compressive strength of 35–45 MPa can be achieved using 100% RHA activated with alkaline activators, having a molarity in the range of 10 to 16M and cured at an elevated temperature between 60–80°C for 24 hours [125,126]. The elevated temperature and highly concentrated alkaline solution help in a higher degree of condensation and dissolution, respectively, thus attaining a higher compressive strength. It is believed that with higher molarity, the Si-leaching process gets hastened, leading to intensive polymerization [126,127]. In contrast, some studies also state that the higher concentration of the alkaline solution increases the viscosity, thus hindering the leaching of Si and Al from the source materials and hampering the degree of geopolymerization, which ultimately leads to a reduction in compressive strength development [105,117].

A study was conducted by Kishore and Gayathri (2017) on RHA-FA based geopolymers casted into 100 mm cubic moulds and varying the molarity of activating solutions (5M and 10M). The results in Fig. 7 showed that when the RHA content increases, the compressive strength decreases but with an increment in the molarity of NaOH solution, the strength increases. Meanwhile, up to a replacement level of 10 to 15%, the strength of the blended mix was comparable to the controlled mix [114]. The reduction in compressive strength with RHA inclusion was also indicated in a research conducted by Das et al. (2020), but their experimental investigation was conducted using FA-GGBFS based AAM where RHA was induced as a silica additive [103]. With an increasing percentage of RHA, the compressive strength of the resulted mix gradually reduces, likely attributed to the imbalance of Si/Al ratio or the improper dissolution of the silica present in RHA.

Liu et al. (2020) in their study replaced MK with RHA up to 15% and found that the compressive strength increased by 24% caused by high content of Si- and Al- oligomers that ensured high degree of polymerization. After exposure to high temperatures (from 300 to 900°C), it was

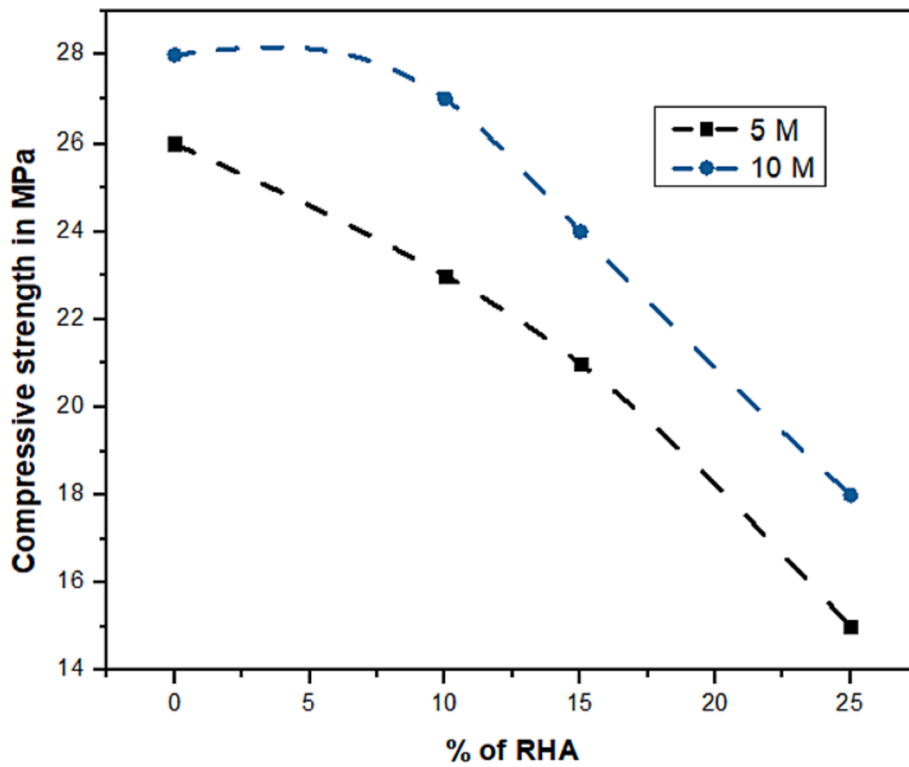


Figure 7. Compressive strength of AAM with RHA [114].

observed that using RHA as an additive in MK based geopolymer healed the microcracks and fissures within the matrix. They concluded that using RHA reduces the melting temperature of geopolymer, thus enhancing the formation of densified structure layer and preventing the high temperature spalling of geopolymer. From the findings of Borges

et al. (2016), the authors produced geopolymer from MK modified by RHA replacement up to 60 wt.%. They noticed that increase in RHA content from 0 to 30 wt.% does not cause a significant increase in the compressive strength, above which the compressive strength begins to decrease. Thus, in MK based geopolymer, incorporating 40 wt.% of RHA

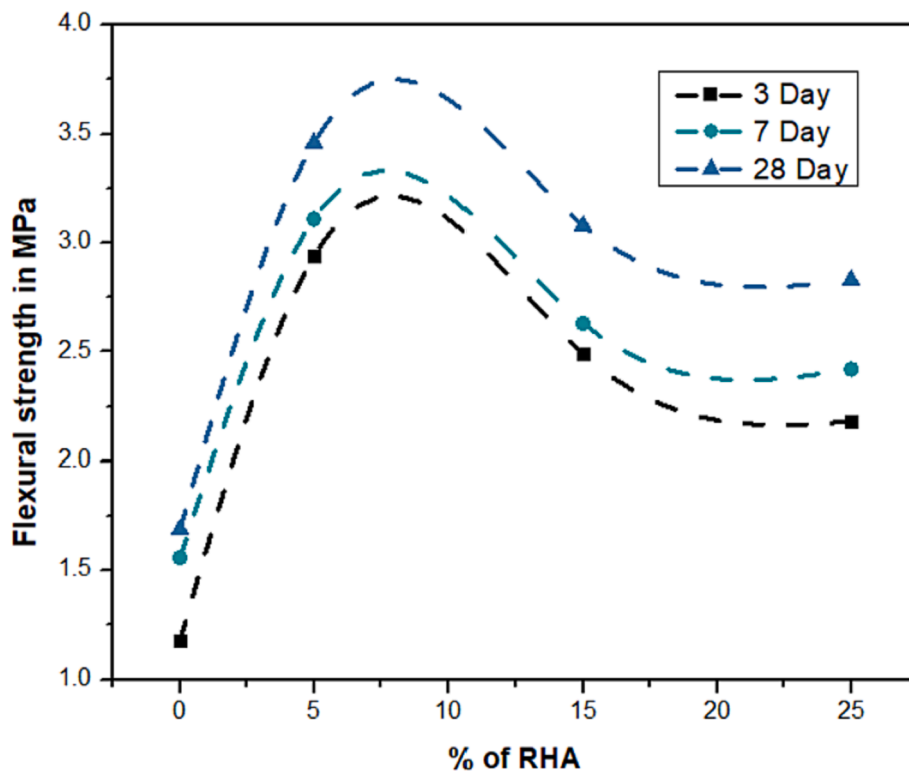


Figure 8. Development trend of flexural strength with RHA [119].

appears as the maximum to ensure stronger matrix resulting in high performance [128].

4.4. Flexural strength

Like compressive strength, the flexural strength is also an important parameter for building and construction applications, notably plain tiles, roofing tiles, and interlocking tiles. The flexural strength of GGBFS based AAM with RHA addition, casted in $150 \times 150 \times 700$ mm beam in accordance with IS: 516 standard were examined by Patel and Shah [119]. The flexural strength increases up to 5% RHA. However, when the RHA content was increased to 15% and 25%, the flexural strength decreased and was lower than those obtained for 100% GGBFS. On the other hand, the flexural strength of FA based AAMs containing RHA are higher than the control mix made with 100% FA. As shown in figure 8, the flexural strength of all the mixes increases with the curing age and the highest flexural strength of 3.56 MPa was obtained for the mix containing 5% RHA at 28 days [119].

The flexural strength of RHA based AAM reinforced with basalt fiber, activated with a mix of NaOH, and sodium silicate and cured at ambient temperature conditions was also investigated. The results showed that with increasing content of RHA, a reduction in strength was observed. Since basalt fiber contains a good amount of both Si and Al, the incorporation of RHA misbalanced the Si/Al ratio, which could be the possible reason for such a reduction in flexural strength [129]. Moreover, it is possible that the ambient curing temperature was not sufficient to enable the dissolution of the silica present in the RHA. Therefore, higher content of RHA acted more like a filler than a binder precursor for gel synthesis. Additionally, Ramani and Chinnaraj (2015) studied the effect of BRHA on flexural properties of GGBFS based AAM subjected to elevated temperature curing [124]. The flexural strength of the AAM containing 10% of BRHA increased by 13% when compared to the control specimen with no BRHA addition. However, a reduction in flexural strength was observed when BRHA content was further raised to 20 and 30% in AAM (Fig. 9). It is suggested that the strength reduction

beyond the optimum BRHA inclusion (10%) could be due to hindrance in the condensation process by the presence of surplus unburnt carbon particles in BRHA.

Generally, the flexural strength reduction in AAM with a higher amount of RHA can be attributed to increased SiO_2 content in the matrix, which ultimately misbalanced the Si/Al ratio, thus resulting in a less densified gel matrix. Therefore, the interfacial transition zone (ITZ) of these compositions are comparatively weak and produces cracks earlier than that of the other mixtures having an optimum amount of RHA during load applications. Recently Kaze et al. (2018) partially replaced raw corroded or iron-rich laterite with 10 to 40 wt.% RHA in order to produce sustainable geopolymer composites [130]. The weight ratio (laterite + rice husk ash)/ river sand was kept at 1/2, and the obtained products were cured at room temperature and at 80°C . The end products exhibited high stability in water, and the flexural strength increased from 3 to 12 MPa and from 5 to 40 MPa (Fig. 10), respectively at room temperature and oven cured at 80°C . From the XRD and microstructural analyses, the authors concluded that the strength development was linked to the formation of ferrosilicate phases between iron minerals and reactive silica that ensured good cohesion between different components within the matrix. In the same vein, Kamseu et al. (2020) studied the effect of curing cycles on the microstructural development of laterites based geopolymer composites containing RHA as an additive. They claimed that the application of low viscosity followed by oven curing resulted in a dense and compact matrix with few large open voids and pores. This was due to the formation of ferrisilicates phases like hinsingerite resulting from the development of Fe-O-Si bonds that favoured the rigidity of the matrix [131]. Recently, Nouping Fekoua et al. (2021) applied three curing modes comprising room temperature, oven curing and steam curing, and assessed their effect on strength development and microstructural evolution of laterite based geopolymer composites with RHA (Fig.11) [132]. The investigation revealed that applying a steam curing under controlled relative humidity of 65% yields best flexural strength of 56 MPa. Such improvement was linked to high degree of geopolymerization and the abrupt

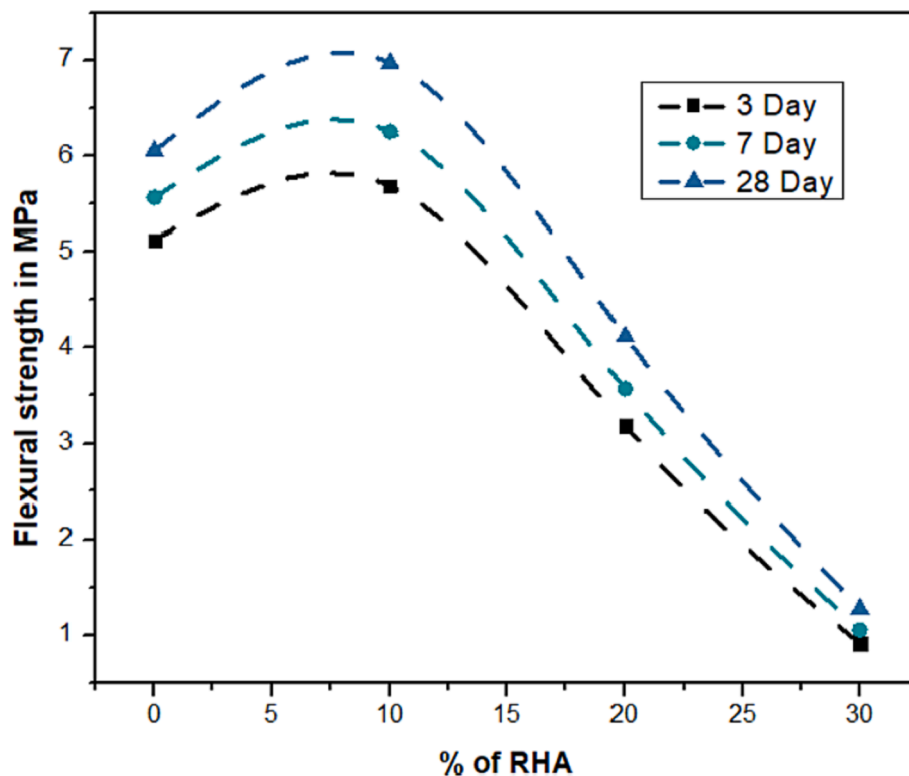


Figure 9. Effect of BRHA on flexural strength of GGBFS based AAM [124].

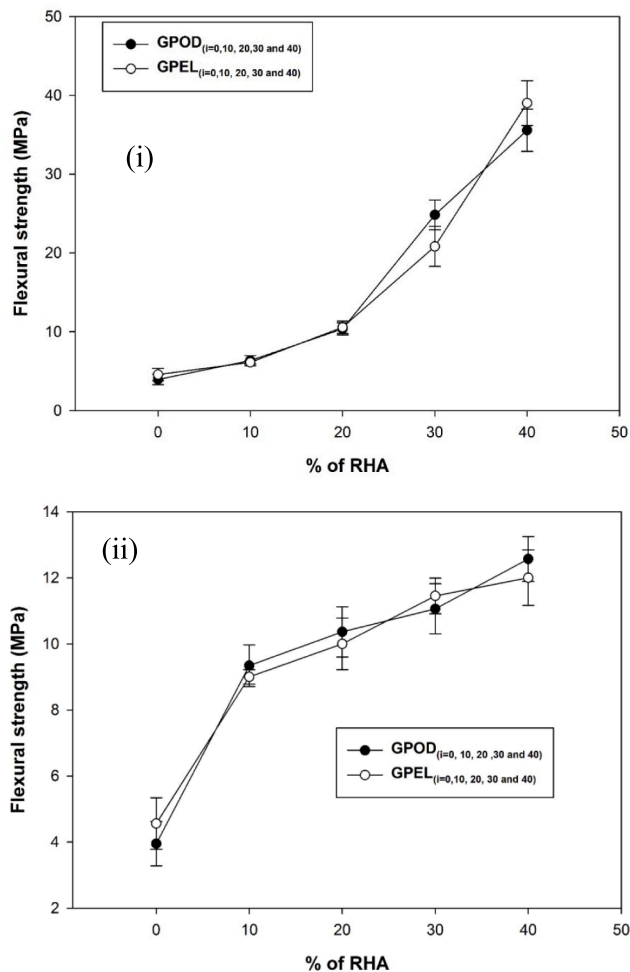


Figure 10. Flexural strength of geopolymer composite GPEL (laterite from Eloumden) and GPOD (laterite from odza) as function of RHA cured at (i) 80 °C (ii) room temperature [134].

departure of water molecules out geopolymer matrix resulting in densified and compact structure. In addition, Kaze et al. (2021) observed an improvement in the flexural strength and reduction in the water absorption percentage of laterite based geopolymer composite by combining RHA and quartz sand followed by curing at 80°C [133]. From all the studies examined so far, the improvement in the AAM samples containing RHA was associated with a higher release of Si-species from reactive RHA within the geopolymer matrix that contributed to more cohesion between different particles.

4.5. Fracture toughness

Billong et al. (2018) investigated the replacement of MK by 12.5, 25, and 50 wt.% of RHA and activated with 10 M NaOH (Fig.12) [108]. It was observed that samples containing 12.5 and 25 wt.% of RHA showed an increase in fracture toughness at an early age, but the values are still lower than that of samples made with 100% MK. However, after 28 days of curing, AAM samples containing RHA (12.5 and 50 wt.%) elongated more than pure MK based AAM used as a reference, ascribed to unreacted silica gel formed between reactive silica and NaOH.

4.6. Tensile strength

Tensile strength is one of the essential properties used for several design aspects of structural components. For instance, when designing

the shear and anchorage of reinforcing steel, the tensile strength of the concrete used plays a vital role. Therefore, it is essential to see the effect of RHA on the tensile strength of AAM taking into consideration the dosage and the content of RHA used. For example, it was reported in a study that the highest value of split tensile strength was achieved in samples containing 15% RHA in comparison with reference samples with no RHA after 90 days of curing. The development of higher tensile strength with the incorporation of RHA is attributed to the co-occurrence of C-S-H and N-A-S-H, produced by the additional silica content and the polymerization reaction. However, when the amount of RHA added to the matrix exceeds 15%, the tensile strength decreases gradually for all curing ages as shown in Fig.13 [88]. This was due to the difference in solubility rate of RHA and GGBFS and the presence of unreactive silica in the mixture. In addition, an increase in tensile strength of AAM was reported in another study when the RHA content was increased. In the mentioned study, about 4% and 7% increment in strength was observed when the RHA content was increased from 350kg to 375kg and 400kg, respectively, per each cubic metre of AAM [102].

Furthermore, the use of BRHA as an additive also had a positive impact on the tensile strength of AAM. When 10% of GGBFS was replaced by BRHA, 3% increment in strength was observed at 28 days (Fig. 14) [124]. Such incremental trend in tensile strength with the curing age observed was due to additional silica supplement for both C-S-H gel and Na/Ca- silicate gel synthesis, which densified the binder phase resulting in improved ITZ.

In contrast, another study stated that the inclusion of RHA had a negative effect on the tensile strength of the specimens. When the 5%, 15% and 25% of GGBFS was replaced by RHA, a reduction of 2%, 6% and 34% in tensile strength was observed after 28 days (Fig15). However, this is not the case with FA based AAMs. All the mixtures containing RHA had better tensile strength than that of the control mix (100% FA) [119]. The reason behind this negative effect of RHA on tensile strength in GGBFS based AAM could be due to the poor dissolution of RHA, and the difference in solubility of RHA and GGBFS in the alkali environment that led to a poor gel-matrix formation.

4.7. Microstructural development and durability properties

Due to high silica content, filler effect, pore refinement and pozzolanic nature of RHA, the microstructure of the concrete or mortar containing RHA is more homogenous and compact when compared to those without RHA. Furthermore, an increase in the formation of C-S-H products and decrease in the Ca (OH)₂ content of the matrix was observed when RHA was incorporated, and this led to the improvement of the pore structure of the concrete. The improvement in the microstructure was attributed to the high pozzolanicity and compatibility of RHA particles with cement [85,141]. Additionally, it was observed that the thickness of the ITZ close to coarse aggregate in concrete containing RHA is small, compared to the concrete without RHA. Moreover, it has also been reported that increasing the fineness and content of RHA results in a finer pore structure and reduction in total porosity of the ultra high-performance concrete [142]. Similarly, a significant reduction in porosity and densely packed microstructure was observed in RHA containing concrete, attributed to enhanced formation of C-S-H gels [85]. The microstructural transformation due to the incorporation of RHA is expected to influence the durability properties of the materials.

The durability properties of mortars and concrete is defined as the ability of material to withstand different environmental conditions such as physical, chemical, combined physical and chemical attacks, abrasions, and weathering conditions, while maintaining their desired engineering properties [143,144]. The durability properties commonly evaluated included resistance to acid attacks [145], sulfate attacks [51], freeze and thaw [146], chloride penetration [147], wet-dry cycles [148], biocorrosion [149] and carbonation [150]. In geopolymers or AAM, their durability properties largely depend on the material types and chemical composition since different material has different

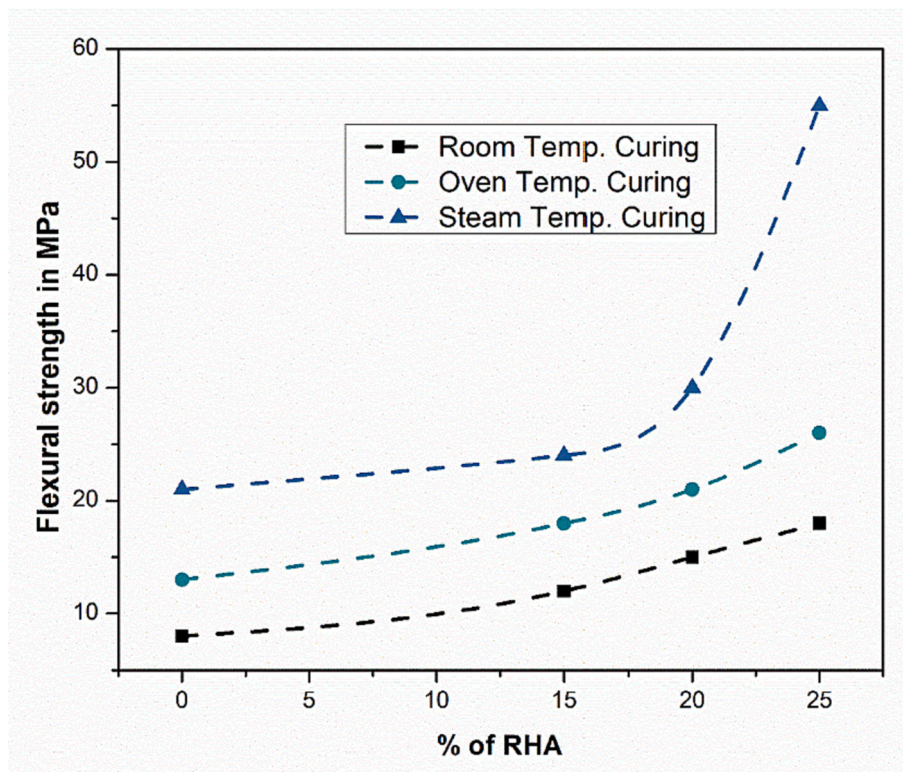


Figure 11. Flexural strengths of goethite-rich laterite based geopolymer composites cured under different curing cycles with permission from [132].

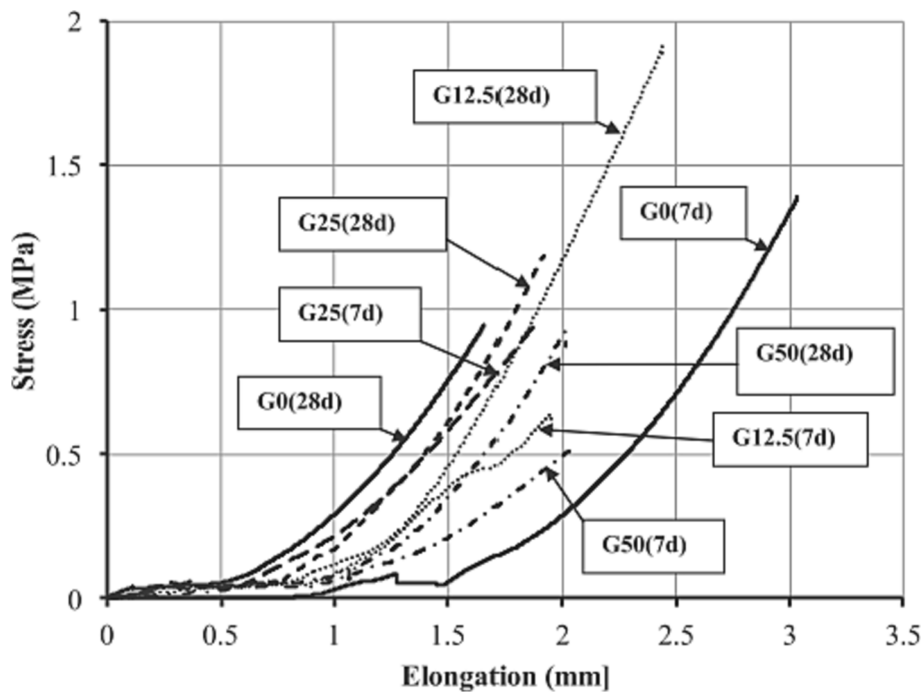


Figure 12. Stress-elongation curves of hardened geopolymer pastes at 7 and 28 days.

aluminosilicates resulting in different Si/Al ratios and geopolymer network [151]. Thus, the use of silica based RHA as precursors or co-binders in geopolymer mortars or concrete is expected to present different durability properties when compared to other calcium rich materials such as BFS. Meanwhile the use of RHA as precursor in concrete has been reported to reduce the absorption and penetration of liquids and ions in concrete after exposure to different environmental

conditions. Saraswathy and Song (2007) reported that the addition of 30% RHA lowered the total porosity of the concrete which in turn reduced the absorption and penetration of water considerably [91]. Venkatanarayanan and Rangaraju (2015) observed a 13 % and 12 % reduction in the water absorption values when 7 % and 15 % of milled RHA was incorporated into the concrete in comparison to the reference concrete without RHA [52]. Other authors have also observed a

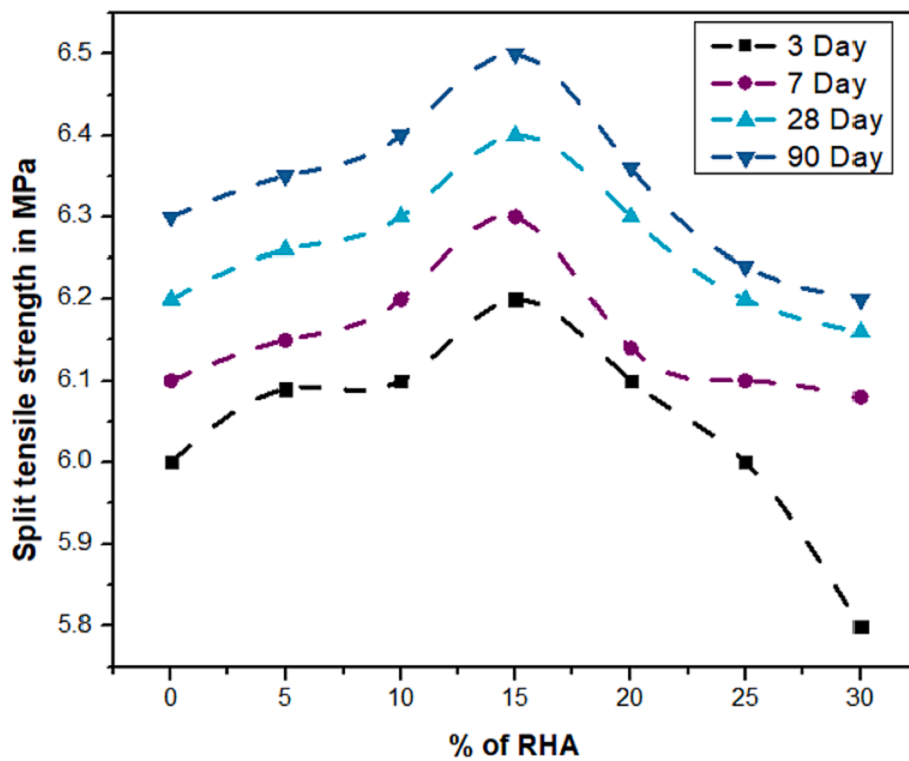


Figure 13. Effect of RHA content on tensile strength of GGBFS based AAM [88].

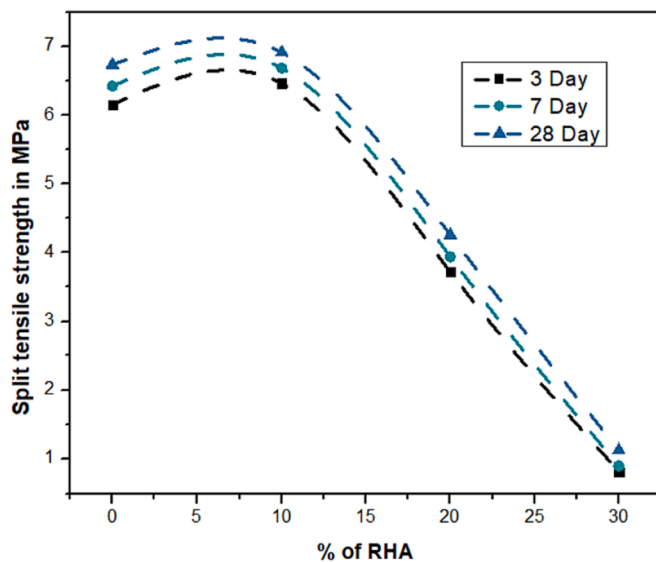


Figure 14. Effect of BRHA on tensile strength of GGBFS based AAM [124].

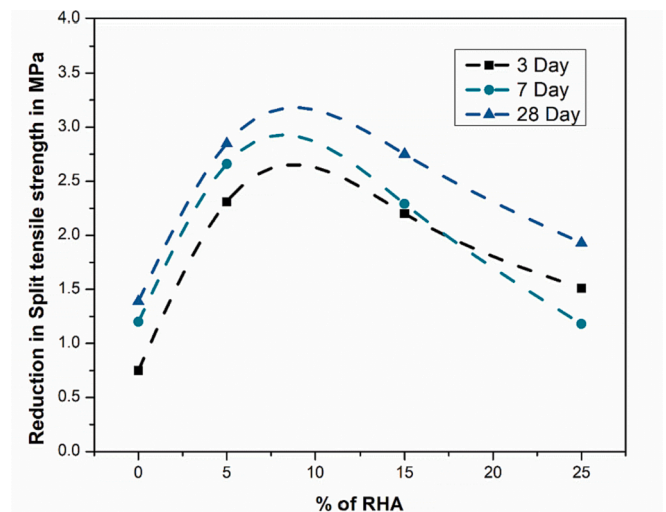


Figure 15. Reduction in tensile strength of slag based AAM with RHA.

reduction in the water absorption values of concrete containing RHA [44,152]. However, the porosity of the concrete is significantly reduced with increase in fineness and pozzolanic reactivity of RHA used. Furthermore, the rate of acid and chloride ions diffusivity in concrete is dependent on the porosity of the concrete [153]. Compared to PC-based concrete, the chloride diffusivity in blended cement containing RHA was significantly lower due to the dense microstructure [44]. The reason for lower diffusivity is due to the ability of the incorporated RHA to modify the gel formed thereby lowering the ingress of acid and chloride into the concrete. Similarly, the blending of RHA with fly ash and PC reduced the chloride penetration rate and better diffusion resistance, ascribed to the denser microstructure and higher number of C-S-H products formed

[154,155]. Thus, concrete or mortar containing RHA has lower chloride penetration rate when compared with PC-based concrete. The resistance of concrete to sulfate attacks has also been improved with the incorporation of RHA as reported by several researchers [34,92]. The expansion caused by sulfate attacks was reduced when 15% RHA was blended with PC even though the expansion rate can be influenced by other factors such as the type of salt and their concentration. For example, Chindapasirt et al (2007) observed that replacing cement with RHA up to 40% was very effective in providing resistance to sulfate attacks. In the mentioned study, the incorporation of RHA lowered the pH level, which inhibits the reactivity of sulfate ions [51]. The effect of RHA incorporation on the carbonation resistance of concrete has also been reported in literatures. Excessive carbonation can lead to higher shrinkage and corrosion of the reinforcement [156]. Compared to PC-based concrete,

the blending of RHA with cement can reduce the amount of unreacted Ca (OH)₂, ascribed to high pozzolanic nature of RHA and suitable curing condition [157,158]. Praveenkumar and Vijayalakshmi (2019) concluded that concrete containing more than 20% replacement of cement with RHA exhibited better carbonation resistance than normal PC-based concrete [159]. In the mentioned study, the incorporation of RHA is presumed to have refined the pore structure, resulting in lower pore connectivity and lower penetration of CO₂. The use of RHA in concrete can also reduce the expansion caused by alkali-silica reaction. Givi et al. (2010) observed that by replacing cement with 15% RHA can reduce damaging expansion caused by alkali-silica reaction, ascribed to the reduction in pH level and the entrapment of the alkalis by the supplemental hydrate formed [160]. Le and Ludwig (2020) concluded that curing condition and high fineness of RHA can also improve the resistance of the concrete to expansion caused by alkali-silica reaction [161]. It was noted that the rate of formation of alkali-silica reaction is higher and quicker in coarser RHA compared to finer RHA. However, the formation of alkali-silica reaction in concrete is inversely proportional to the reaction product formation.

5. Challenges and opportunities

It is envisaged that by 2050, the concrete industry will have to considerably decrease its CO₂ emissions, aiming for more environmentally friendly solutions and complying with the United Nations sustainable development goals (SDGs). Therefore, AAM is viewed as a viable and sustainable alternative to reducing the direct use of PC and its derivatives. The ability of AAM to utilize many industrial wastes such as slags, fly ashes, mineral wools, and natural materials such as laterite soil, marble/granite powder, basalt rock, will fulfill the requirement of a clean construction materials. Owing to the extensive use of AAM for different applications, the additives for strength and other property enhancement will be of great use. RHA can fulfill such requirements if appropriately used in consideration of its physicochemical properties and dosages. Using RHA for the preparation of sodium silicate would be beneficial with lower carbon footprints and costs than the commercial activators used at present, obtained from melting of a mixture of sodium carbonate with quartz sand heated at 1600 °C. Sufficient properties have been obtained in AAMs using alternative sodium silicate prepared by mixing NaOH with RHA in defined proportions. However, the durability of such materials such acid resistance, sulphate resistance, permeability, freeze-thaw cycle, carbonation, chlorine test penetration is not fully understood and should be studied and compared to those activated with commercial sodium silicate existing in the literature. In addition, life cycle analysis (LCA), exploitation effects of RHA deposits, and cost analysis at the industrial level, including the logistic cost of exploitation of rice husk ash vs. commercial silicate production, should be investigated.

6. Conclusions

To reduce the impact of CO₂ emissions, and support environmental preservation, sustainable approaches are necessary. Several approaches have been proposed to reduce CO₂ emissions associated with cement production and usage and address waste issues; it includes efficient waste management, waste recycling, and reuse. The utilization of RHA as a precursor or co-binders in AAMs is seen as a sustainable and eco-friendly alternative to PC. Thus, a critical review of current literatures on the production, characteristics, and utilization of RHA in AAMs are presented and the conclusions drawn from the review are explained below

- Significant amount of RHA is generated annually with 90% of them unutilized or stored in landfills resulting in higher economic cost for the producers as well as causing significant damage to the environment.

- The chemical and mineralogical composition of RHA depends on the source material and the production process, but usually contain around 85-90% amorphous silica, making them suitable secondary raw materials for AAM production.
- To be used as binder precursors for AAMs, proper processing is required to improve the pozzolanic performance of RHA. Controlled burning of RHA preferably around 700-800 °C reduces its crystallinity and enhances its reactivity. Milling of RHA has been suggested to reduce the particle size, increase the specific surface area, and attain highly reactive RHA.
- Being a good source of reactive silicate, the specific surface area, particle size and solubility of RHA are determinant for the end properties of the synthesized products.
- The pozzolanic reactivity of RHA is comparable with other widely used pozzolans and SCMs.
- RHA can either be used as a sole binder precursor or as a co-binder in AAMs. When used as a co-binder, the optimum amount is preferably in the range of 10-20% and this significantly improves the fresh and hardened state properties of AAM through the filling effects and increasing the number of strong Si-O-Si bonds.
- This review provides valuable insights on the feasibility of upcycling RHA as precursors in AAMs and can help achieve efficient waste management and social sustainability objectives.

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.

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