

Highlights:

- Feasibility of industrial wastes in brick production is reviewed
- Design and development of alkali-activated bricks are studied
- Physico-mechanical and durability properties are reviewed extensively
- The factors responsible for the brick performance are examined
- Alkali activation provides sustainable solutions for brick production

Abstract

Brick is one of the majorly used building materials for masonry construction. Unlocking the potential to deliver significant impact against India's current housing and agro-industry waste challenges is crucial. This can be met by developing sustainable products using industrial wastes. Alkali-activated products are claimed to be sustainable and cost effective, giving rise to Portland cement free products. The paper presents a state-of-the-art review on the development of sustainable bricks by alkali-activation of industrial wastes. Physical and chemical characterisation of industrial wastes are discussed in order to check its feasibility for the development of alkali-activated bricks. The influence of parameters on physico-mechanical and durability related properties are evaluated. Previous studies show that bricks with the appropriate values of molarity, alkali modulus, liquid-binder ratio and water glass-NaOH ratio (5 M-15 M, 0.15-0.9, 0.2-0.48 and 0.5-2.5 respectively) achieves a compressive strength of 5-60 N/mm². Though considerable research has been carried out, application of industrial wastes in the manufacturing of alkali-activated bricks are still limited and some recommendations are suggested. Geopolymeric bricks seems to be the most advantageous as they can be incorporate high content of wastes. It signifies the potential use of waste materials to form alkali-activated brick as an alternate sustainable masonry option.

Keywords: geopolymerization; masonry unit; waste; industrial by-product; physico-mechanical characteristics; eco-efficiency

1. Introduction

Over the years, there has been a considerable growth in the brick industry due to increasing demand for housing and infrastructure in developing countries. Moreover, rapid industrialization in these countries has generated a substantial amount of waste that affects the environment. Hence, with the increasing housing demand and industrial waste, it is desired to use these wastes as raw material in production of bricks [1]. Just in India, for example, 20 million housing units need be constructed by 2022 in urban regions (MoHUPA), where the majority is for low-income urban populations.

Zhang [2] have classified eco-efficient bricks as per the manufacturing process, which are fired in kilns, with cementation and alkali activation. In the first process, traditional fired clay bricks are replaced by bricks made up of clay but with a partial substitution of waste. Though there is a decrease on the use of raw materials (replaced by the waste), the kiln creates heavy air pollution and increases coal or other combustible consumption [2]. Even compacted earth bricks are generally cemented. Nevertheless, the use of cement or other chemical binders increase the carbon footprints, as an average cement industry presents an embodied carbon of 0.95 kg CO₂ per kg of cement that is produced [3-5]. Hence, the trend of research is shifting towards the investigation of sustainable alkali-activated bricks. It offers the

possibility of using 100 % waste materials as the base material, giving rise to OPC-free bricks or hybrid cements. Activation of alumina and silicate solids by alkaline liquids leads to geopolymeric gel formation followed by setting and hardening at a particular curing condition [6]. Geopolymer are mineral polymers that results from the chain formation of mineral molecules by covalent bond, i.e. geosynthesis [6]. It consists of a polymeric framework of Silica- Oxygen- Aluminium (Si-O-Al) with alternate sharing of four oxygen atoms (SiO_4 and AlO_4). The four-coordinated oxygen of Aluminium (Al) imparts free negative charge that should be balanced with positive ions in order to have stable matrix [7-8]. These essential cations are calcium (Ca^{2+}), sodium (Na^+), potassium (K^+) and lithium (Li^+) ions. Thus, geopolymerization (alkali activation) process is nothing but the synthesis of aluminosilicates in strong alkaline media. The prime steps of this are: a) dissolution of solid alumino-silicate oxides in alkali hydroxide solution; b) diffusion of dissolved alumino-silicates from the surface to inter particle space; c) polymerisation with silicate solution that results the formation of gel and d) hardening of the matrix [4]. Van Jaarsveld & Van Deventer [9] stated that the reaction of pozzolanic materials with alkaline solutions produces polysilicates that are amorphous and stable compounds. It forms an artificial stone like structure under 100°C temperature [6]. Previous studies showed that, to have an alkali-activated product, three components are important: raw material (source of alumina and silicates), filler and alkali-activators. The various raw materials used for the alkali activation are industrial wastes or by-products such as fly ash (FA), ground granulated blast furnace slag (GGBS), boiler ash (BA), rice husk ash (RHA), sugarcane bagasse ash (SCBA), mining waste tailing (MT) and fillers like kaolinite, lateritic clay, stone dust and others. Hydroxides and silicates of alkali (namely Na^+ , K^+ , Ca^{2+} , Li^+) are the most used alkali-activators. The hardened inorganic polysilicates (geopolymer) have wide use in construction applications such as soil stabilization [10], masonry units [11], mortar [12], concrete [13], precast panels [14] and other structural components [15]. This is due to improved mechanical and chemical performance, durability and eco-efficiency. Therefore, the application of alkali-activated bricks or masonry blocks in building construction proved to be efficient, sustainable and cost competitive.

The present study briefs the properties of alkali-activated bricks that are produced from various raw materials and alkali-activators. The article evaluates the influence of parameters such as concentration of alkali-activator, liquid to binder ratio, alkali modulus, forming pressure and curing temperature on physico-mechanical and durability related properties of alkali-activated bricks. The study states the potential of different industrial wastes in high strength, durable alkali-activated bricks for better masonry option in sustainable construction.

2. Development of alkali-activated bricks

Alkali-activated brick, which is also known as geopolymer brick, are formed by the process of mixing, moulding and curing either at ambient or elevated temperature. The brick consists of source materials containing silica, alumina and fillers or fine aggregates, which are activated by alkali-activators such as sodium hydroxide (NaOH), potassium hydroxide (KOH), Lithium hydroxide (LiOH) and silicates of potassium and sodium. Industrial wastes such as ashes (FA, RHA, SCBA, BA), GGBS, surkhi (waste from fired clay brick), are the materials that contain silica and alumina in significant amount, acting as source materials for alkali activation.

2.1 Raw materials characterisation

Raw material characterisation is one of the important parameters that affects the physical and mechanical properties of alkali-activated bricks. The physical characterisation of raw materials includes particle size distribution, morphology, density and specific gravity. Whereas, chemical characterisation includes chemical composition and loss on ignition (LOI) value of constituent materials. The most common tests that are used for categorization are X-ray diffraction (XRD), X-ray fluorescence (XRF), scanning electron microscopy (SEM) and particle size distribution (PSD). Table 1 shows the chemical composition of various industrial wastes that are used as raw materials for polymerisation.

Table 1 Characterisation of raw materials used for alkali-activated bricks

Raw Materials	SiO ₂ (%)	Al ₂ O ₃ (%)	Fe ₂ O ₃ (%)	MgO (%)	CaO (%)	Na ₂ O (%)	K ₂ O (%)	SO ₃ (%)	LOI (%)	Density (g/cc)	Ref.
BA	82.7	2.8	1.9	1.35	2.24	0.45	4.68	0.97	13.1	2.49	[1]
BA (from SCBA+ RHA +Coal)	81.9	3.87	1.17	1.24	1.55	0.49	3.55	2.84	34.9	2.23	[1]
FA	63.9	20	6.64	1.25	3.84	-	1.08	1.32	2.01	2.08	[16]
GGBS	43.4	12.5	1.3	1.5	40.3	0.9	0.6	-	-	2.9	[17]
RHA	95.6	-	0.24	-	0.7	-	2.66	0.15	2.67	2.18	[18]
Kaolin	48.28	35.69	0.97	0.07	0.05	0.28	1.42	0.11	12.7	2.6	[19]
Copper MT	64.8	7.08	4.33	4.06	7.52	0.9	3.26	1.66	-	2.83	[20]
Cement kiln dust (CKD)	11	3.9	2	3.6	42	-	0.6	-	36	3.15	[21]

In addition to source materials, it is important to decide the type of alkali-activator to be used in the manufacturing of bricks. As previously mentioned, the alkali-activators used are hydroxides and silicates of alkalis. Table 2 presents raw materials and alkali-activator used in previous studies for the development of geopolymeric bricks.

Table 2 Raw materials used for alkali-activated bricks

Raw Material (RM)	Particle size	Alkali-activator (AA)	AA/RM (By wt.)	Ref.
FA (Class F)	Mean particle size: 20 µm	NaOH (12M) and sodium silicate	0.4	[22]
Kaolinitic clay	Diameter: 5–120 µm	NaOH and sodium silicate	0.15	[19]
Copper MT	Mean particle size: 120 µm	NaOH (10- 15 M) and de-ionized water	0.08-0.18	[20]
Circulating- fluidized bed combustion (CFBC) bottom ash	CFBC passing through 45 µm with 7 % retention on the sieve	NaOH (5 M and 10 M), KOH (5 M and 10 M) and 5 M	0.30	[23]

		LiOH and sodium silicate		
Red mud (alumina refinery waste) with RHA	Diameter: 5–300 μm	NaOH (4M) and de-ionized water	1.20	[24]
CKD with MT	Mean Particle size:120 μm	NaOH (10-15 M) and de-ionized water	0.12-0.20	[21]
FA with GGBS and foam	Diameter: 2–100 μm	NaOH (10 M) and sodium silicate	0.40	[11]
Unground RHA and FA and RHA	Mean Particle size: 15 to 21 μm	NaOH (10 M) and sodium silicate	0.51	[18]
FA with crumb rubber (1:1)	Diameter: 5–120 μm	NaOH (8- 14 M) and Sodium Silicate	0.60	[25]
BA with clay and lime				
a) 100 % RHA	Diameter: 5-600 μm	NaOH (2 M) and de-ionized water	0.45	[1]
b) 63 % SCBA, 27% RHA and 10% Coal				
Concrete residue (CR) with arc furnace slag (AFS)	CR passing through 75 μm AFS- passing through 150 μm	NaOH (10 M) and sodium silicate	0.90	[26]

2.2 Manufacturing process

The manufacturing process of brick involves activation of source materials by alkali hydroxide and silicate with or without additional water, followed by vibration, compaction and curing. Following section presents the procedure to develop alkali-activated bricks from various raw materials and alkali- activators, their proportion, type of compaction, curing temperature and duration. Figure 1 presents the process flow diagram of alkali-activated brick formation.

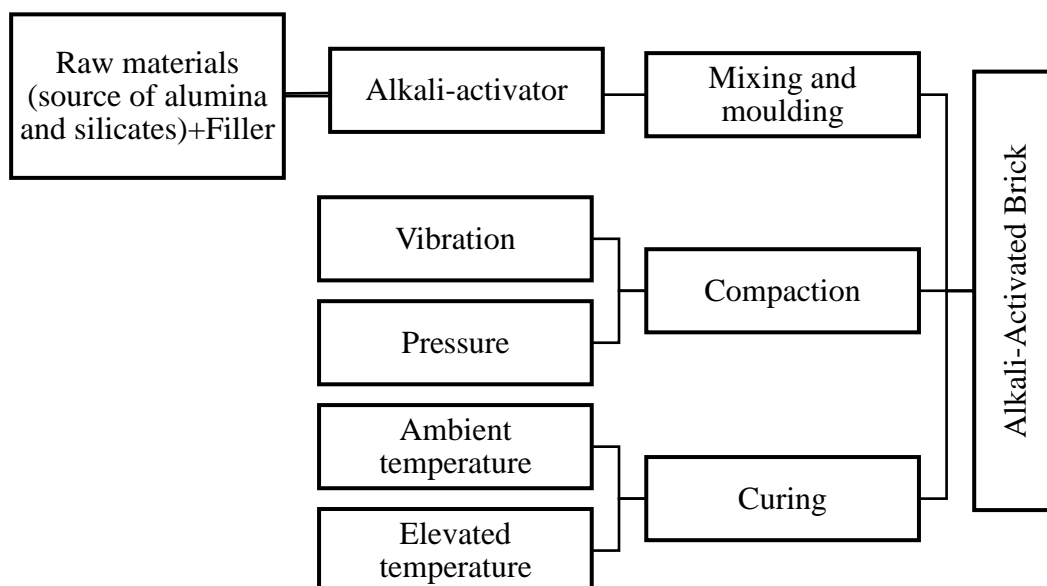


Figure 1. Process flow diagram of alkali-activated brick production

In order to study the physico-mechanical and microstructural properties of alkali-activated bricks, Arioiz et al. [22] developed a brick (size 19 cm x 9 cm x 5 cm) of FA class. The FA was activated by NaOH (12 M) and sodium silicate (water glass-WG) and mix was compacted with 30 N/mm² forming pressure. The bricks were cured in an oven as well as with steam treatment at 40°C, 60°C, 80°C and 100°C for various durations: 2, 4, 6, 24, 48 and 72 hours. Sukmak et al. [27] also used FA as a base material with silty clay as filler material and alkaline liquid, which was a mixture of NaOH (10 M) and sodium silicate. The ratio of Na₂SiO₃ to NaOH and liquid-FA were altered from 0.4 to 2.3 and 0.4 to 0.7, respectively, to identify the role of influential factors on the strength development. The developed brick samples were kept at normal ambient condition for one day, and then heated at 65, 75, and 85°C for 1, 2, and 3 days. It was observed that, for any specimen a liquid to FA ratio below 0.3 and more than 0.8 is not suitable for making compressed bricks. Abdullah et al. [28] evaluated the performance of FA based alkali-activated bricks. Researcher carried out trials for various mixes as FA-aggregate ratio in the range of 1:2 to 1:5 (by weight proportioning). Whereas, sodium silicate to hydroxide and liquid-binder ratios were fixed at 2.5 and 0.5 respectively. The binder, filler and activator were mixed and poured into the mould of size 21.5 cm x 10.25 cm x 6.5 cm with 10 N/mm² forming pressure. The samples were cured for 1, 3, 6, 12 and 24 hours at various temperatures ranging from 40-80 °C.

Pointot et al. [1] elaborated the alkali activation of BA with a) 100 % RHA and b) 63 % bagasse ash, 27% RHA and 10% coal that are wastes generated from paper mill industry, to develop alkali-activated bricks. The source material used possessed detrimental characteristics for polymerisation, such as large particle size (5-600 micron), varying morphology, loss on ignition (8-35 %) and less alumina content. The morphology of the ash was angular and rough surfaced that affected the workability of the mix. The BA has more LOI than the recommended value of 6 % [29]. Hence, to get the desired properties, supplementary materials, namely clay and lime, were added in the proportion of ash: clay: lime - 70:20:10 (by weight).

As red mud has the benefit of high pH value with amorphous alumina (Al₂O₃) that dissolves amorphous silica (SiO₂), which is present in RHA, He and Zhang [24] developed geopolymer using red mud. The red mud slurry has a pH of 12, dissolved NaAlO₂, NaOH in liquid phase, and hematite and alumina (Al₂O₃) in solid phase. To facilitate alkali activation and minimize the influence of compositional variation, the slurry was dried and ground to a precipitate until all the material passed through a 300-micron opening size sieve.

Chen et al. [23] determined the suitability of circulating fluidized bed combustion (CFBC) bottom ash in the manufacturing of alkali-activated bricks. The researchers studied the effect of various alkali-activators on the compressive strength of bricks. With variation in the concentration of KOH (5M and 10 M), NaOH (5M and 10 M), LiOH (5 M) and sodium silicate solution (SiO₂/Na₂O Modulus- 1.2, 1.5 and 2), the samples were cast for testing at the age of 3, 7, 14 and 28 days. The samples were cured at a temperature of 40°C and 100 % humidity. The bricks were produced by the application of 60 kN force for 10 seconds with liquid to solid ratio as 0.3. Singh et al. [30] produced bricks (size 23 cm x 11.5 cm x 7.5 cm) by mixing FA, BA and river sand with an alkali-activator. The samples were kept at 80°C temperature for two hours. Further, solid alkali-activated blocks size 30 cm x 15 cm x 10 cm were prepared using FA (23 %), fine aggregate (14 %), coarse aggregate (55 %), alkali-activator (5 %) and water (3 %).

Hwang & Huynh [31] focused on the application of FA and RHA in alkali-activated bricks. The mean particle size (D₅₀) of FA and RHA was 21.5 and 15.3 µm, respectively. However, from Chapelle test, FA was found to be more reactive in an alkaline environment than the RHA. It was observed that the particles of FA were rounded and smoother, whereas, RHA was porous and angular surfaced (Figure 2).

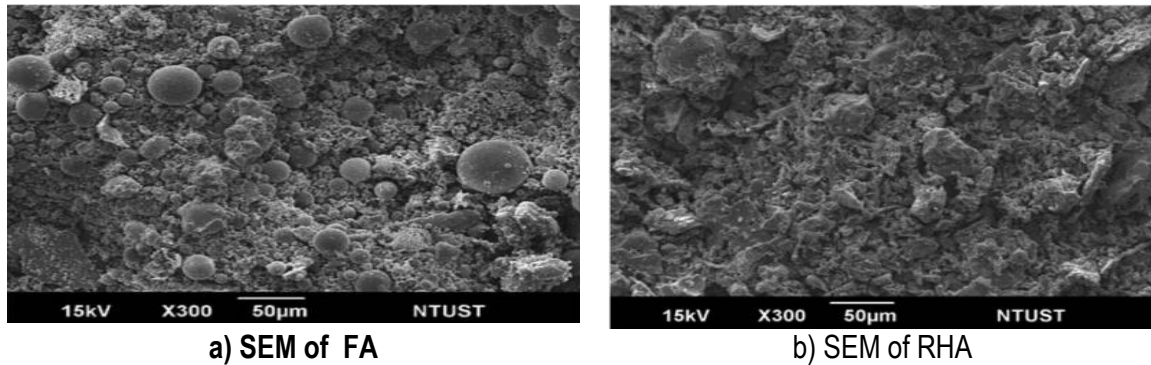


Figure 2. SEM images of a) FA and b) RHA [31]

Maulana et al. [32] developed hybrid composite rice husk-alkali activated bricks. The FA and RHA (as-received, washed and dried at 100°C for 4 hours) were used as a binder and synthesized at 60°C for 1 hour by alkali-activation with its molar oxide ratio of $\text{SiO}_2/\text{Al}_2\text{O}_3$ as 3, $\text{Na}_2\text{O}/\text{SiO}_2$ as 0.2, and $\text{H}_2\text{O}/\text{Na}_2\text{O}$ as 10. The RHA was mixed in varying proportion at 5 %, 10 % and 15 %. Ferone et al. [33] used weathered coal ash as a raw material for low temperature alkali-activated ceramic brick production. The alkali-activators used were NaOH (10 M and 14 M) and water glass (SiO_2 -27 %, Na_2O -8 % and Water-65 %, by weight). The samples were cured for different curing conditions. The water to total solids (w/s) ratio (by weight) fixed for 10 and 14 M were 0.31-0.49 and 0.28-0.45 respectively.

Banupriya et al. [17] experimented the use of FA, GGBS, fine aggregate (sand and quarry dust) to produce alkali-activated masonry and paver blocks. FA, GGBS, aggregates and NaOH (5 M) were mixed together to form alkaline condition. The specimens were tested for compressive strength, split tensile strength and flexural strength. Radhakrishna [34] designed and developed compressed masonry bricks (20 cm x 10 cm x 6 cm and 23 cm x 19 cm x 8 cm) with FA, GGBS, metakaolin and silica fume as binder and sand, quarry dust and pond ash as fine aggregates. The alkali-activators used for synthesis were KOH (8 M) and NaOH (8M – 14 M). Venugopal et al. [35] determined the properties of masonry bricks made up of class F FA (specific gravity 2.4), GGBS (specific gravity 2.9) as binder, artificial sand of zone II as filler and NaOH (8 M) and sodium silicate as an alkali-activator. The FA to GGBS ratio was taken as 4:1, aggregate to binder as 1:1, liquid to binder as 0.2 and NaOH to sodium silicate as 1:1.5. The bricks cured for 28 days at room temperature were tested in conformance to IS 2185 [36]. Venugopal et al. [37] also developed alkali-activated bricks (solid as well as hollow) by activating FA, GGBS and artificial sand with WG and NaOH (8 M).

In addition, Ezzat et al. [38] used water-cooled slag and surkhi as a binder in a ratio with sand 85:15 (weight %) and NaOH (8 %) as an alkali-activator. Zawrah et al. [43] also utilized surkhi and GGBS in the production of geopolymer using 8 M NaOH and water glass in the ratio of 2.5 (by volume). The researchers studied the effect of curing with incorporation of GGBS in surkhi by 20, 40, 60 and 80 % replacement. The weight ratio of alkaline liquid to powder mix used was fixed at 0.30 for all samples. Besides these, Mohsen and Mostafa [19] investigated the use of Kaolinitic clay in the production of alkali-activated bricks. Locally available clay was calcined at 700°C for two hours, becoming a metakaolin, and grounded in a ball mill. The clay particles passing through 120 µm were used in proportion with NaOH and water glass solution. The pressure of 15 N/mm² was applied to form the test samples that were allowed to dry at ambient temperature for a period of three days and then cured for one day at both temperatures of 75°C and 150°C.

Khater et al. [39] demonstrated the alkali-activation of slag and metakaolin, which resulted in the formation of calcium and sodium aluminosilicate gel. The activators used were 10% NaOH solution in addition to 5% liquid sodium silicate. The developed bricks were suitable for heavy works. In addition to electric arc furnace slag (EAFS), Apithanyasai et al. [26] included concrete residue to produce bricks. The sources of alumina and silica, which are CR (crushed and passed through a 75-µm sieve) and EAFS

(passed through a 150- μm sieve) were mixed as 60:40, 70:30, 80:20 and 90:10 (weight %) and activated by NaOH (10M) and sodium silicate in the ratio of 1:2.5 with 0.9 alkali-binder ratio.

In addition to previously mentioned wastes, Ahmari & Zhang [20] investigated the feasibility of copper MT to develop sustainable alkali-activated bricks by performing compressive strength tests, water absorption and micro structural analysis by SEM and XRD. The researchers analysed the effect of four major parameters that are molarity of NaOH, water content, compacting pressure and curing conditions on physico- mechanical properties of bricks. The samples were tested for NaOH (10 and 15 M), curing temperature (60 to 120° C), water content (8% to 18%), and compaction pressure (0-35 N/mm²).

2.3 Properties of alkali-activated bricks:

The physico-mechanical, durability and functional properties of alkali-activated bricks primarily depend on the characterisation of raw materials, concentration and proportion of alkali-activators and curing conditions. The physico-mechanical properties, such as dimensionality, density, water absorption, initial rate of absorption (IRA), mechanical strength (compressive, flexural, shear and bond) and micro structural properties were evaluated. Many factors, such as liquid-binder ratio, alkali silicate-hydroxide ratio, binder-aggregate ratio, forming pressure, temperature conditions and duration, influence the properties of bricks [40].

Arioz et al. [22] developed bricks that has compressive strength in the range of 5- 60 N/mm² at an age of one week, one month and three months in various temperature conditions. The density of the bricks were in the range of 1400-1500 kg/m³. Densities of alkali-activated bricks produced by steam-treatment were lower than that of oven treatment. However, there was no significant effect of heat treatment and duration on the density of the bricks. To get 25 N/mm² strength, the brick needed to be in the oven for 6 hours at 40 °C. Similar bricks at 60 °C for 24 hours treatment achieved only 2 % more compressive strength than the former one and were, therefore, less eco-efficient.

Sukmak et al. [27] used FA as source material, silty clay as filler material and an alkaline liquid (L) resulting from a mixture of NaOH (10 M) and sodium silicate. Figure 3 indicates the test results of compressive strength at 90 days with L/FA of 0.4-0.7 and WG/NaOH 0.4-1.5. Irrespective of alkali modular ratio, compressive strength increased with the increase in L/FA ratio until it reaches 0.6. The WG/ NaOH ratio of 0.7 gives the optimum results for the clay-FA brick. Whereas, for clay-free FA brick, the optimum value was 1.5 that shows the advantage of addition of clay to FA.

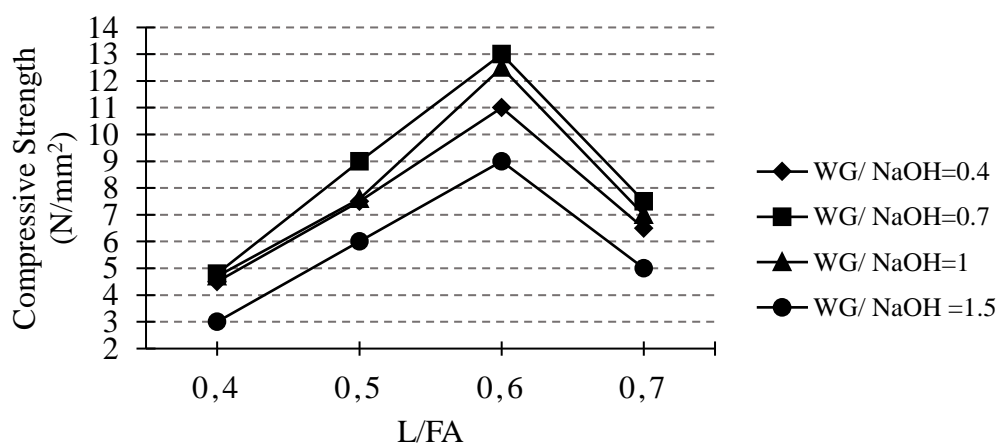


Figure 3. Effect of liquid/fly ash (L/FA) and water glass/sodium hydroxide (WG/NaOH) on compressive strength of clay-FA brick [27]

Figure 4 shows the SEM of specimen at 0.3, 0.6 and 0.7 L/FA ratio. The alkali-activated products, hydration product (A) and unidentified spherical accumulations (B), are detected on the particles. However, micro cracks were observed on at high L/FA specimen (C). From the results, $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio of 0.7 and the liquid / FA ratio of 0.6 was found to be the optimum dosage for alkali-activated bricks. It was found that 65 to 75 °C is the optimum range of temperature that could control the polymerisation reaction, which results in the development of the strength. Overheating causes deterioration and develop micro cracks over the brick sample.

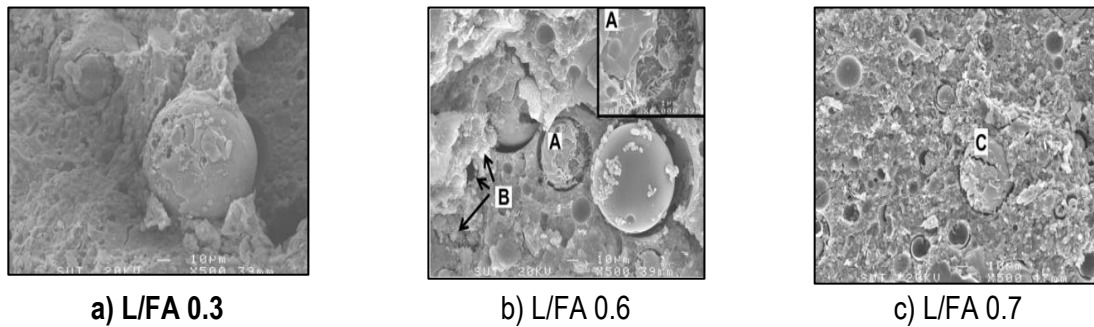


Figure 4. SEM of clay- fly ash brick with different liquid to fly ash ratio [27]

It was obtained that the mix with a ratio of one portion of FA and three parts of sand has optimum workability and strength (15.4 N/mm²) with appropriate water absorption (6.4 %). As sand ratio increased from 1:2 to 1:5, the strength reduced from 17 N/mm² to 9.2 N/mm², whereas water absorption increased from 3.7 to 9.6 % [28]. Up to 70°C, the compressive strength increased with the rise in temperature and further reduced by almost 6 %. Figure 5 represents the XRD pattern of original FA and the bricks developed at different conditions. It shows that the diffractograms changed due to alkali-activation of FA, where sharp peaks resulted to broader diffused halo that represents an amorphous material. Moreover, the peak shifted from 30 to 40° 2θ values with respect to temperature and number of days. This shift depicts the formation of aluminosilicate gel because of geopolymerization reaction. From SEM analysis, it was observed that samples aged 60 days were more compacted that imparts high density as well as mechanical strength than the samples aged 7 days (Figure 6). The densities of developed bricks were in the range of 1800-1900 kg/m³. Researcher also produced bricks of 1900-2100 kg/m³ density with 10–15 % of water absorption. Moreover, the compressive strength of bricks ranged from 12-25 N/mm². In addition, the obtained density, water absorption, compressive strength and drying shrinkage of solid geopolymer bricks were 2100 kg/m³, 8-10 %, 10 N/mm² and less than 1%, respectively [30].

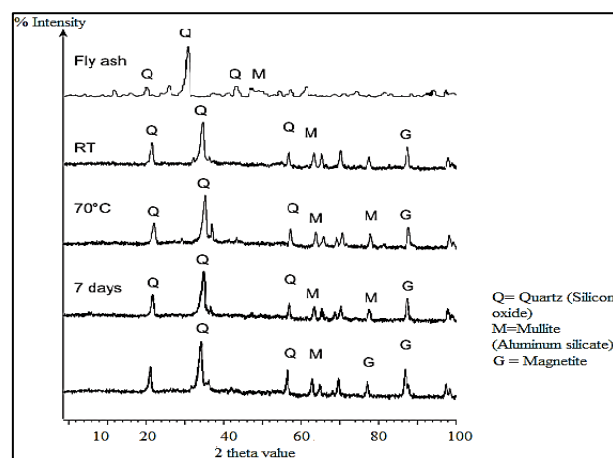
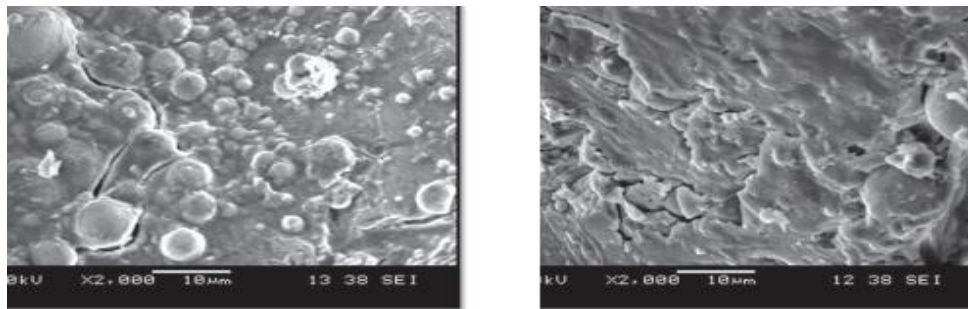


Figure 5. XRD pattern of brick at different conditions [28]

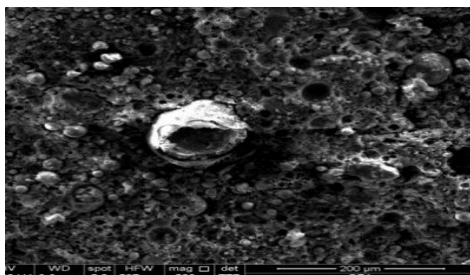


a) Sample cured for 7 days

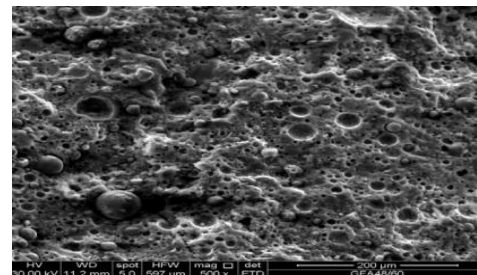
b) Sample cured for 60 days

Figure 6. SEM of developed alkali-activated brick [28]

It was found that LiOH with lesser concentration gives comparable results as samples with higher concentration of KOH and NaOH. The ascending order of compressive strength value for various alkali-activators obtained were 5 M NaOH, 5 M KOH, 5 M LiOH, 10 M NaOH and 10 M KOH [23]. Hwang and Huynh [18] observed that, as percentage of RHA increased, density and heat conductance decreased with increase in water absorption and void volume. The increased voids reduce compressive and flexural strength of alkali-activated bricks. The water absorption (7-10 %), density (1930-2090 kg/m³), volume of voids (1.4-1.7 %) and thermal conductivity (1.2 to 1.7 W/ (m.K). However, the 10 % RHA bricks show better results than no-RHA bricks due to lesser water absorption and density. A replacement of 10 % RHA by FA imparts maximum compressive strength of 30 N/mm² at the age of 28 days that decreased with the increasing percentage of RHA. Maulana et al. [32] attains a maximum compressive strength (7 days) of 26 N/mm² with a 10 % addition. The addition of RHA also increased its fire resistance. The brick with 10 % RHA gives better mechanical strength, thermal property, and acid resistance as compared to RHA free brick. Freidin [41] investigated the effect of sodium silicate content, compaction pressure and hydrophobic additive on alkali-activated bricks (size-30 cm x 14 cm x 9 cm) made up of FA and BA by using sodium silicate solution as an alkali-activator. The specimens were cured and tested at room temperature of 20–23 °C and relative humidity of 35–60 % for 28 days. With the increase in water glass content from 10- 30 %, strength also increased from 1.5 to 16 N/mm². The results indicated that due to high absorbing capacity of ash, water uptake was up to 40 % that can be reduced by addition of hydrophobic admixture. Ferone et al. [33] stated that utilization of as-received wet FA with optimized liquid mix design gives almost similar properties as the bricks with dry FA. In addition, it was observed that, an increase in the w/s ratio resulted in reduction of unconfined compressive strength (UCS). Curing at room temperature showed the unreacted spherical FA particles whereas the brick cured at 48 hours and 60 °C gives a compact structure that imparts high strength (Figure 7).



a) Cured at room temperature



b) Cured at 60 °C

Figure 7. Sample cured at a) room temperature and b) for 48 hours at 60 °C [33]

In order to produce higher-class brick that has high strength and low water absorption, researchers have used several waste raw materials like water-cooled slag, metakaolin, silica-fume, copper MT in addition or partial substitution with conventional industrial wastes like FA and RHA. The researchers observed that compressive strength of alkali-activated bricks have increased from 14 to 19 N/mm² with the GGBS increasing from 20 % to 35 % [17].

Radhakrishna [34] studied the effect of liquid to binder ratio (0.15-0.25), binder to aggregate ratio (1:1, 1:2 and 1:3), molarity of alkali-activator (8, 10, 12 and 14 M), age of the sample (1, 3, 7, 14, 28, 56, 90, 120 and 180 days) on compressive strength. The strength increased with the increase in molarity, whereas above 14 M, workability of the mix gets affected as the concentration of salts increases. It was witnessed that the addition of silica fumes increased the strength by almost 4 times with highest compressive strength of 23 N/mm². However, the use of silica fume increases the bricks cost. The addition of GGBS and metakaolin were found to be effective with FA. Furthermore, GGBS gives more strength as compared to metakaolin. Irrespective of mix proportions, the mechanical strength of bricks increased with age and decreased with an increase in the liquid-binder ratio [34]. The strength of masonry bricks in 7 days alternate drying and wetting was reduced by 23 %, which was still better than fired clay bricks. It was concluded that, with the increase in molarity of NaOH (8 M-12 M), compressive strength and density increases, whereas water absorption decreases [42]. Venugopal et al. [35] obtained the values of water absorption, IRA and density of the bricks as 8.5 %, 3 kg/m²/min and 1800 kg/m³, respectively. The strength of bricks increases with the age from 5-22 N/mm². However, it gains required minimum strength (5 N/mm²) within 24 hours that makes it handy. The Elastic modulus was found to be 9394 N/mm², which is higher than for reference bricks.

Researchers also proved the potential of fired clay brick's waste in the production of alkali-activated bricks. Replacement of slag with surkhi up to 40 % gives more than 40 N/mm² mechanical strength [38]. On the other hand, additional replacement leads to the reduced mechanical properties, because of increased crystalline content and insufficiency of activator to dissolve all crystalline fractures. Zawrah et al. [43] tested the bricks for bulk density, porosity, water absorption and compressive strength at 3, 7, 28 and 90 days. As the content of GGBS increased, the density of brick also increased due to reduced porosity. This inclusion of GGBS up to 60 % gives better results in terms of its mechanical properties, which has least porosity (Figure 8). However, further increase in GGBS percentage leads to cracks and deterioration of the material. In addition, it has increased the water absorption and porosity.

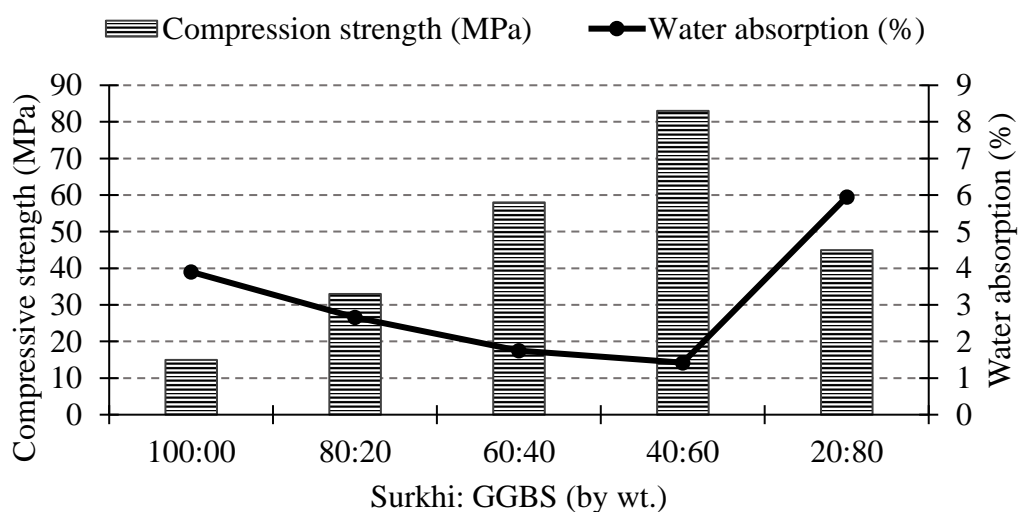


Figure 8. Compressive strength and water absorption of the alkali-activated brick at the age of 90 days [43]

Khater et al. [39] investigated that addition of metakaolin in slag gives more than 30 N/mm² compressive strength at the age of 28 days. Apithanyasai et al. [26] obtained the optimum ratio of 80 (CR):20 (EAF-slag) that gives maximum strength of 17 N/mm² with the lowest water absorption of around 1%. The mineralogical phases were analysed by XRD that showed the crystalline phase of silica as quartz (SiO₂), which is formed by the alkali activation of CR and EAF-slag. The CSH gel was formed at peaks 29.2°, 32°, 42.7°, 49.8° and 55.3° to develop an amorphous aluminosilicate gel that enhanced the compressive strength.

Ahmari & Zhang [20] observed that brick samples prepared by 15 M NaOH concentration have higher unconfined compressive strength than the 10 M concentration brick samples. From the study, optimum temperature for the brick development was found to be of 90° C. Durability and leaching behaviour of MT brick was studied. The leaching behaviour was determined by static leaching test that was performed by immersing the powder of brick sample and mine tailings. Further, durability of the specimen was evaluated through compressive strength, water absorption, loss in weight and concentration of heavy metal in two different solutions, at pH of 4 and 7. The results specify that though there is a considerable loss of strength after immersion in solution of pH as 4 and 7, the absorption of water and loss in weight were reported as small [44].

The cost of materials accounts for 92 % of the total cost of alkali-activated bricks out of which NaOH contributes to the highest percentage of cost [1]. Hence, it is necessary to reduce the consumption of alkali-activator without affecting the performance. Researchers also studied the use of CKD to reduce NaOH consumption and for improving physico- mechanical properties and durability of alkali-activated brick [21, 39]. Addition of maximum 10 % CKD to MT based alkali-activated bricks provide higher strength for 10 M NaOH brick than 15 M NaOH without CKD (Figure 9) [21]. It saves the additional requirement of NaOH and cost incurred due to it. Researcher also produced low cost alkali-activated brick using CKD as a source of free alkalis that reduces the concentration of other alkali hydroxides [39]. Alkali-activated bricks prepared by fractional binder replacement of water-cooled slag by CKD ratio ranged from 25 to 75 % with 25 % sand. The samples were cured and tested for 7, 28 and 60 days at 40°C and 100 % native humidity. The compressive strength increased with the increasing age of the brick. For 0, 25, 50 and 75 % substitution, 28 days strength was 7%, 4 %, 9 % and 21 % more than 7 days strength respectively. Furthermore, it achieves maximum strength at the age of 60 days around 50, 39, 20 and 15 N/mm² for 0, 25, 50 and 75 % replacement of slag by CKD respectively. The water absorption of the bricks was in the range of 10-20% at the age of 7 days that decreased with the time. The bricks up to 25 % CKD provides high mechanical strength that it goes under the category of heavy-duty brick [45].

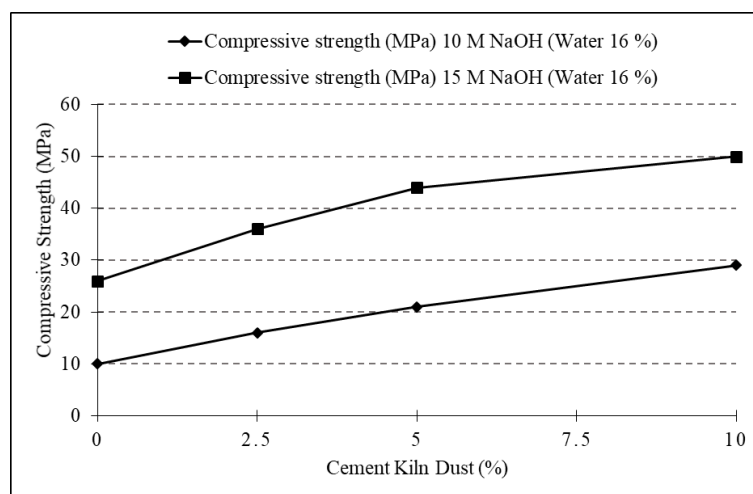


Figure 9. Effect of molarity and addition of cement kiln dust on compressive strength of mine tailings based bricks cured at 90°C for 7 days [21]

This section briefed about the constituent materials, method of production, design parameters and properties of alkali-activated bricks. Table 3 shows the list of constituents, parameters and properties that are of prime importance while discussing about the alkali-activated bricks.

<p>Constituent materials</p>	<ul style="list-style-type: none"> • Source material • Filler • Alkali-activator
<p>Design parameters</p>	<ul style="list-style-type: none"> • Raw material charecteristics • Type of alkali-activator • Concentration of alkali-activator • Binder to aggregate ratio • Liquid to solid ratio • Alkali modulus ratio • Forming pressure • Curing condition • Curing duration
<p>Properties</p>	<ul style="list-style-type: none"> • Dimension • Water absorption • Density • Compressive strength • Flexural strength • Shear bond strength • Flexural bond strength • Efflorescence • Durability • Thermal conductivity

Table 3 Constituents, design parameters and properties of alkali-activated bricks

3. Discussion on design, development and properties of alkali-activated bricks

Various researchers have identified the potential use of industrial wastes and stated the optimum mix design for sustainable, strong and durable alkali-activated bricks. The discussion is presented in three categories as constituent materials i.e. industrial wastes and alkali-activators, physico-mechanical properties of brick and durability related properties.

3.1 Materials for alkali activation

3.1.1 Industrial Wastes

It is evident from the Table 1 that various industrial wastes, which are rich in alumina and silicates, have the potential to be used as raw material for alkali activation. Materials such as artificial pozzolans and other wastes have different physical and chemical characteristics, which varies according to their source or origin [1, 46]. Physical properties such as particle size distribution, surface area, bulk density, morphology and chemical composition, as the amount of oxides (mainly $\text{SiO}_2 + \text{Al}_2\text{O}_3 + \text{Fe}_2\text{O}_3$), loss on ignition, decides the type, concentration and proportion of alkali-activator required to form the calcium alumino silicate gel that gets hardened. From the chemical characterisation of raw materials used for development and optimisation of bricks, it was observed that the summation value of SiO_2 , Al_2O_3 and Fe_2O_3 ranges from 75-95 % that confirms the chemical requirements of natural pozzolans according to ASTM C618 [29]. Larger the particle size, lesser will be the total surface area for reaction, which reduces reactivity. In addition, smaller the particle, faster will be the reaction as particle size less than 20 micron mostly comes under amorphous category that ultimately improves the mechanical properties of bricks [47]. The rounded and smooth surfaced source materials require less alkali-activator to give the desired workability due to their lesser surface area. The unburnt constituents of source materials affect the alkali activation suitability, as they are non-reactive and liquid absorber.

As shown in section 2, FA seems to be the most commonly used pozzolana for activation. However, in recent years the trend has been shifting to use other waste materials as well and optimize the mix design to achieve minimum requirement of properties as per the standards. Though RHA is the richer source of silica in comparison to other pozzolans, its porous and angular surface affects the polymerisation process (Table 1). Researchers also proved that FA is more reactive than RHA in an alkaline environment from the Chappelle test as the major crystalline phase of RHA is cristobalite, whereas FA has quartz and mullite [16]. Addition of RHA (up to 10 %) in FA alkali-activated bricks improved the mechanical strength and durability in comparison to RHA free bricks [16, 32]. In addition, incorporation of RHA also reduces the density, thermal conductivity and increases acid resistance of developed bricks. The use of metakaolin with GGBS found effective as far as compressive strength and durability is considered [32, 34]. Metakaolin alone does not provide sufficient strength due to its high porosity that makes the bricks soft. Hence, synthesis of metakaolin with FA and GGBS improves the mechanical properties [8]. GGBS is mostly used for producing high strength bricks as it has high specific gravity and achieve strength ranging from 35 -50 N/mm² [38, 43]. Bottom ash, BA, locally available calcite clay, concrete residue, surkhi, MT and other source materials need to be treated mechanically, to reduce particle size and, therefore, increase surface area, or thermally, to achieve the desirable properties for synthesis [48] and increase reactivity. Otherwise, addition of supplementary materials such as clay, quick lime, CKD, water-cooled slag, improves the mechanical properties with optimum proportioning [1, 21, 27]. CKD acts as an additional source of silicates and alumina as well as it raises alkalinity, which reduces the amount of NaOH that is needed. As the particle size of CKD is finer, it contributes to a denser structure by filling the pores. During the hydration process, pH of the mix increases due to the formation of calcium hydroxide and carbonate resulted from the reaction of calcium and pozzolanic materials. Furthermore, CaCO_3 imparts strength and reduces water absorption of brick [39, 44]. Disposal of MT is a major problem due to its impacts on the environment such as land disposal, contamination of surface and ground water, dust that causes air pollution. Frequently the MT have hazardous components, such as arsenic. Hence, the utilization of such MT (in alkali-activated bricks could stabilize those hazardous components and reduce the impact on the environment [20-21, 44]. Use of sand [28], silty clay [27], bottom ash [49], crumb rubber [25] and ungrounded RHA [18] as filler materials give minimum required strength with the reduced proportion of alkali-activators [41].

3.1.2 Alkali-activators

Generally, utilization of hydroxides of any alkalis such as sodium, potassium, calcium, lithium and silicates of sodium and potassium are suitable for activation of waste materials. However, most of the researchers have used sodium and potassium based alkali-activators to form alkali-activated bricks. Chen et al. [23] compared various activators with different molar concentrations and sodium silicate solution for the development of bricks. The lithium hydroxide followed by potassium-based activator (KOH) gives higher strength as compared to sodium hydroxide. This is because the size of potassium ion (K^+) is larger than the cation of sodium (Na^+) that forms the larger silicate oligomers, whereas sodium ions favour the formation of monomers. The larger oligomer easily bind with aluminium hydroxide that result in better setting and hardening [4]. NaOH has the capability to dissolve more aluminium and silica species, whereas, potassium hydroxide imparts more compressive strength. NaOH is a preferable activator as the potassium hydroxide is expensive that makes the brick costly [50]. The activators shall be used as per the properties of raw materials and the class of alkali-activated bricks. Another factor that influences the properties of bricks is the modulus ratio of alkali silicate that is nothing but the ratio of SiO_2 and M_2O (where M is the alkali metal). As the silicon dioxide increases the rate of reaction decreases. Excess of silica content makes the brick elastic that it deforms rather than fracture. Researchers have investigated the appropriate range of modulus as 2-5 to gain the desired strength [23, 51]. In addition, the ratio of SiO_2 to Al_2O_3 affects the strength of a brick. High content of Al_2O_3 results in lower strength products and therefore, SiO_2/Al_2O_3 should be in the range of 3.3-4.5 [4]. Molarity of the alkaline activator plays a significant role to achieve the required strength.

3.2 Physico-mechanical properties of alkali-activated bricks

Dimensionality, initial rate of water absorption, dry density, compressive strength, flexural strength, bond-strength, modulus of elasticity are the various physical and mechanical properties that decide the feasibility of bricks for construction. The major factors that have influence on the performance are physical and chemical properties of the raw materials, binder to aggregate ratio, type and concentration of alkali-activator, liquid to solids ratio, modular ratio, curing temperature and its duration.

The brick density increases with the increase in amount of alkali-activator, as it dissolves more silica and aluminium that fills the voids of alkali-activated brick structure. The molarity of alkali-activator is directly proportional to the unit weight. The brick becomes denser with age until the activation is completed. It has been found that the effect of temperature and duration on the density of alkali-activated bricks is not that significant. Ahmari et al. [20] studied the effect of forming pressure on brick density with varying initial water content. The unit weight of the block increases as the forming pressure increases, but the rate of increase decreases after a specific limit, as excess of pressure squeezes out the liquid (Figure 10).

The denser the brick, harder and stronger it is.

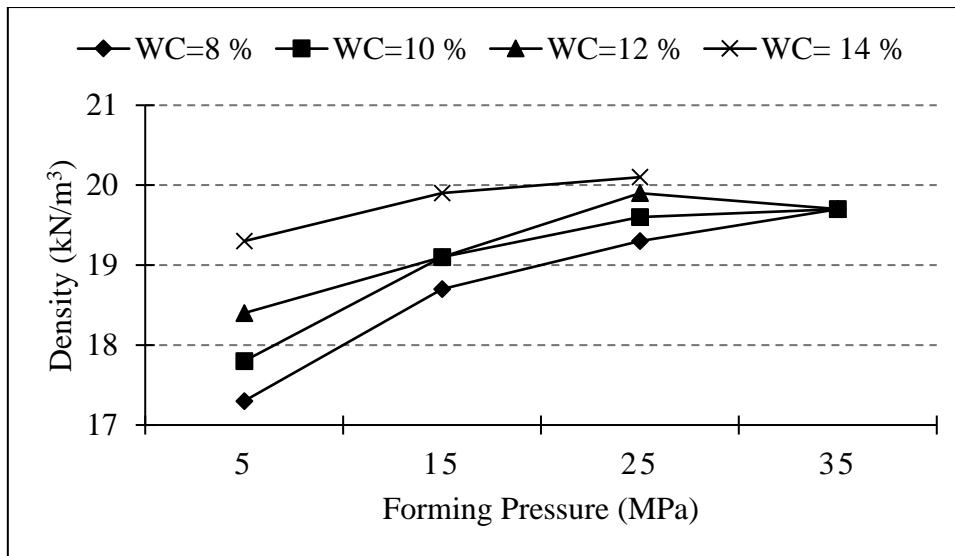


Figure 10. Effect of initial water content (WC) and forming pressure on density [20]

With optimum molarity and temperature, the higher degree of aluminosilicate gel results in dense and strong structure [20, 37, 44]. The higher the molarity, higher is the dissolution of aluminates and silica that makes the brick less porous and permeable. It is the case of high temperature (Figure 11) as well. Water absorption of the bricks depends on the geopolymer structural formation. Venugopal et al. [37] concluded that the water absorption of GGBS brick reduced from 10 % (6 M) to 6 % (12 M) with the increase in molarity of an alkali-activator. Ahmari et al. [20] found that the rate of absorption decreased with the time as the reaction gets completed. The water absorption varies from 2 % to 5 %, corresponding to forming pressure from 0.5 to 15 N/mm². At higher compaction, leaching of liquid from the paste leads to reduction of geopolymeric gel that makes brick porous and permeable. Freidin [41] concluded that the addition of a hydrophobic agent reduced the water absorption of bottom-ash alkali-activated brick by almost 10%.

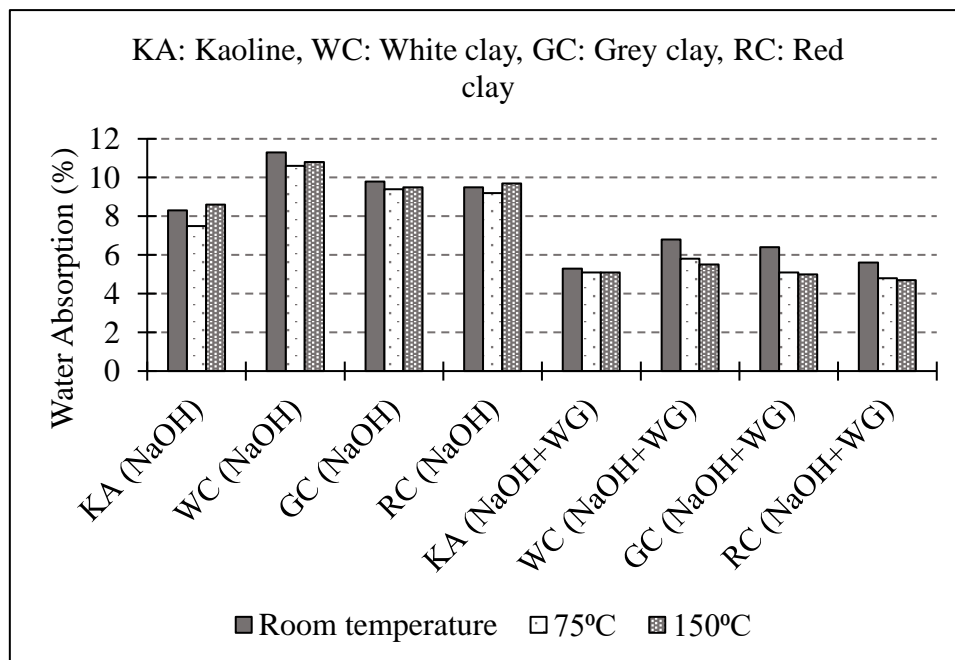


Figure 11. Effect on geopolymeric brick water absorption due to varying curing temperature [19]

Figure 12 indicates an increase in strength of bricks with the increase in proportion of aggregates. The increased filler content makes the brick brittle causing failure with lower compressive force. It also affects the workability of the mix to produce masonry bricks [28]. Irrespective of the materials used, compressive strength of the masonry bricks increases with the age. Figure 13 shows the effect of curing duration on mechanical strength of bricks cast and cured at ambient temperature.

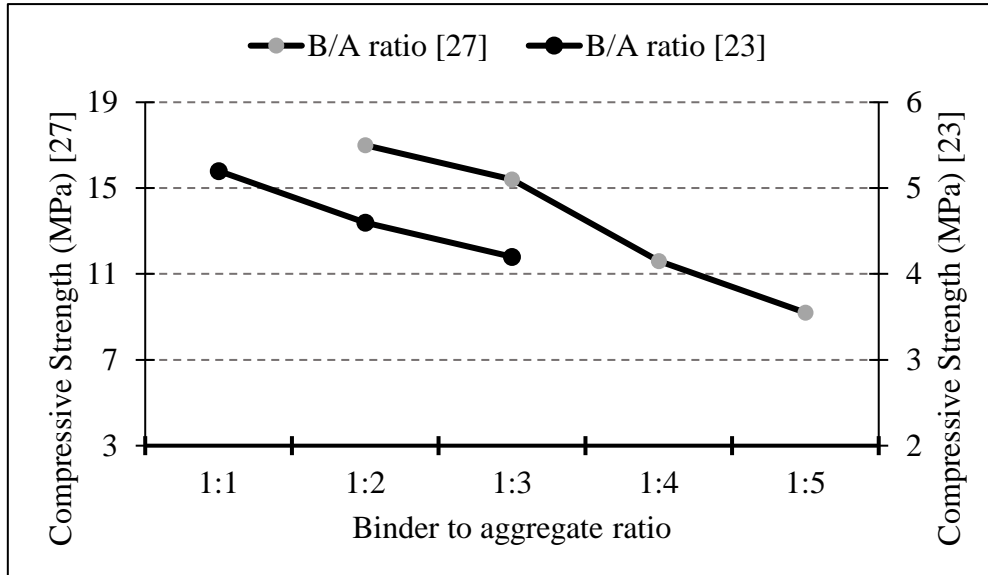


Figure 12. Binder to aggregate ratio vs. compressive strength

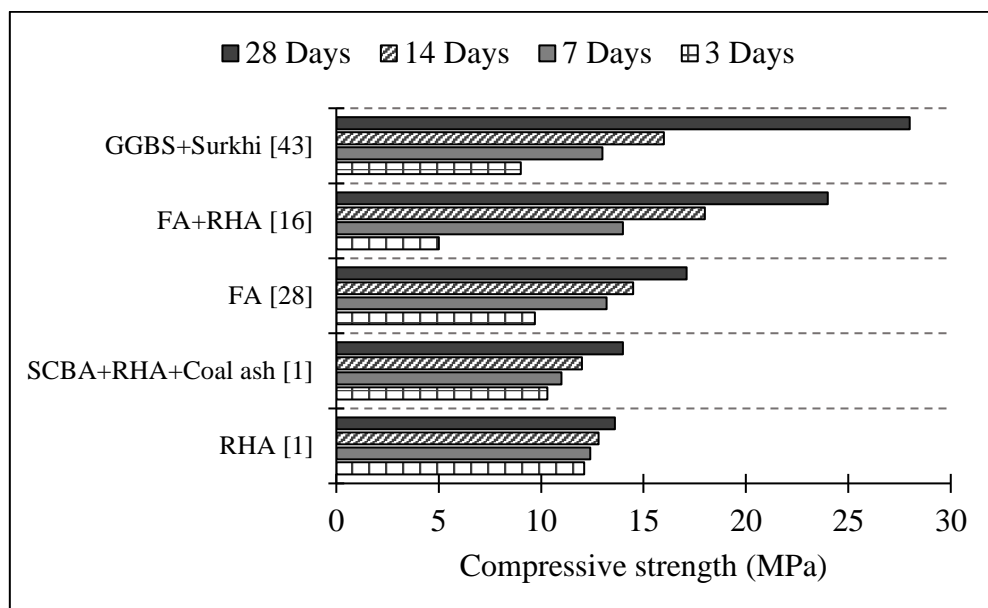


Figure 13. Compressive strength vs. curing duration

Arioz et al. [22] studied the effect of curing temperature by treating FA brick samples at 40°C, 60°C, 80°C and 100°C for 2, 4, 6, 24, 48 and 72 hours. The elevated temperature causes evaporation of water from the bricks reducing the bulk density. In addition, the heating temperature fastens the rate of reaction and gives early strength. However, above certain temperature, polycondensation and fast gel formation may obstruct the dissolution of alumina and silicates that reduces the strength. Researchers reported that

60°C is the optimum temperature that provided higher compressive strength [22]. Mohsen & Mostafa [19] studied the curing temperature effect on calcined clay-based alkali-activated bricks and reported an optimum temperature of 75°C. Ahmari & Zhang [21] tested copper MT bricks and found 90°C as optimum temperature for 10 and 15 M NaOH alkali-activator. For tuff-based alkali-activated bricks, the optimum temperature was found to be 40°C for 8–12 M and 80°C for 4 M NaOH concentration [52]. Figure 14 presents the variation in compressive strength of the FA, MT and calcined clay (CC) brick with respect to temperature.

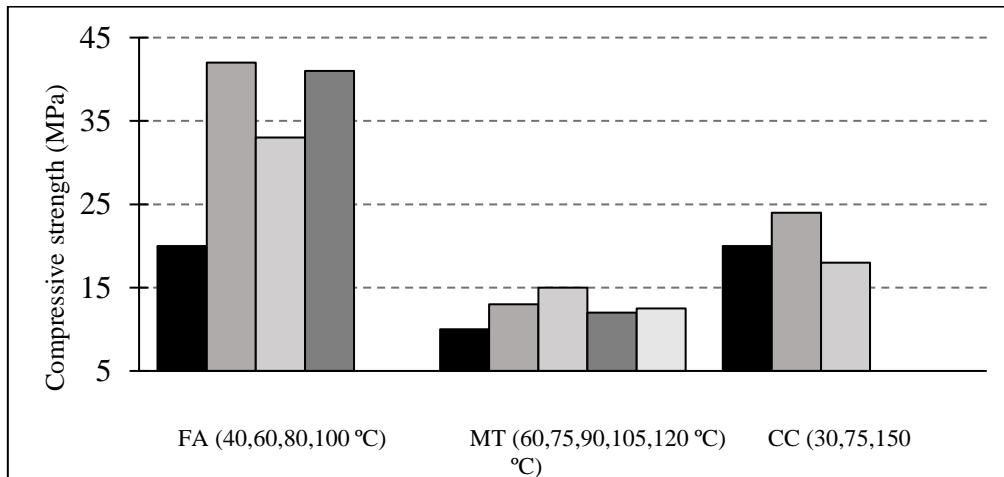


Figure 14. Effect of curing temperature on compressive strength [2, 19, 20, 22]

Furthermore, other physico-mechanical properties such as IRA, flexural and bond strength of bricks were studied. The optimum range of IRA is 0.2-2 kg/m²/min. Beyond these limits it affects the bond strength of masonry. Excess of IRA causes suction of water from mortar joint and makes the joint brittle. However, brick with lower IRA floats on mortar that makes the bond weaker. In addition, it affects water tightness and durability of masonry [25, 53]. Mohammed et al. [25] found that the IRA of interlocking alkali-activated brick was 0.18 kg/m²/min that gave the lower modulus of rupture as 0.26 N/mm² only. The IRA of alkali-activated GGBS solid and hollow bricks were found to be 2.7 and 2.5, respectively, which was found to be within permissible limit of Indian standard [36]. It was found that flexural bond strength of the alkali-activated hollow bricks were higher than that of solid bricks [37].

3.3 Durability related properties

The durability and leaching behaviour of MT-based alkali-activated bricks were studied [44]. It was observed that there was a substantial loss in compressive strength and water absorption and weight loss were negligible after the immersion of brick in solutions (pH of 4 and 7) for different periods. The loss in strength was due to the dissolution of aluminosilicate gel. However, metals are effectively mobilized that induces heavy metals in the gel. The leaching of metals was higher in a solution of pH-4 in comparison to a pH-7 due to higher solubility of MT in acidic conditions. The FA based alkali-activated bricks were tested for acid resistance and result of weight loss was in the range of 0.3-0.5 %, which was reported within permissible limit [54]. The loss in weight (%) of alkali-activated bricks was comparatively lower than conventional fired clay and FA bricks, when immersed in different concentration of sulfuric and hydrogen chloric acid [55]. Venugopal et al. [35] studied the structural feasibility of GGBS hollow and solid bricks for which the alternate drying and wetting test was carried out for durability assessment. The percentage gain in weight after 7 cycles was less than 6 % and strength loss were 28 % and 26 % for hollow and solid bricks, respectively, that was better than for the conventional clay brick. Moreover, the brick was not effloresced. The main properties of alkali-activated bricks that makes it superior than conventional bricks

was better compressive strength, resistance to acid attack and weather action, soundness and high thermal resistance [56-57].

Based on the reviewed literature, following recommendations are suggested:

- As there is a significant variation in the properties of industrial wastes, it is necessary to carry out the benchmarking for better utilization of these raw materials. Though materials like RHA, BA, and bottom ash are rich in aluminosilicates, the physical properties like shape, size and texture shall be considered while designing the mix proportions. Accordingly, preliminary treatment shall be given.
- The use of sodium-based alkali-activators rather than potassium and lithium gives cost effective product as well as comply the desired standards for physico-mechanical and durability properties. Hence, the use of NaOH and sodium silicate as an alkali-activator is recommended for development of alkali-activated bricks. Based on the properties required, the dosage of alkali-activator shall be decided. From the review, optimum range of liquid to solid ratio was found to be 0.4-0.8 (by weight). To reduce excess requirement of alkali-activators, supplementary materials, like CKD, clay, water-cooled slag or, lime, can be added into the mix. Incorporation of such materials, namely of wastes, (up to 20 %) reduces the cost of the bricks.
- From the study, it was observed that compaction is an important step on manufacturing bricks. Nevertheless, it is necessary to be within limits as excessive compaction removes liquid from the plastic or moistened mix and affects brick properties. The recommended value of forming pressure to be applied is maximum 25 N/mm².
- Based on the time constraint, the alkali-activated bricks can be made either at ambient or high temperature curing. The recommended higher value of temperature is 100° C, above which the rate of increase in strength reduces. Further, excessive heating induces cracks. In addition, it is an energy intensive process.
- It is necessary to carry out the product evaluation over a period of varying seasons for different geographic location. Life cycle analysis should be carried out along with the estimation of energy and carbon footprint study of alkali-activated brick manufacturing.

4. Conclusion

After an extensive literature review on the use of wastes to produce eco-efficient masonry bricks, the following can be stated:

Geopolymeric bricks seems to be the most advantageous because they can be produced with a high content of wastes. In comparison to cemented bricks, geopolymeric do not need a binder addition and, therefore, should be ecological. In comparison to fired clay bricks, do not have energy consumption for being fired.

Variation in the source materials significantly changes the physical, chemical and mechanical properties of alkali-activated bricks. After preliminary treatment, wastes generated from industries such as: agro, metallurgy, mining, ceramic, cement, construction and demolition have the potential to be used for the production of alkali-activated bricks. These industrial wastes are source materials of alumina and silica that helps geosynthesis. The physico-mechanical properties of bricks are greatly influenced by the concentration of alkali-activator, liquid to binder ratio, alkali modulus and sodium silicate to NaOH ratio. The ranges of molarity, alkali modulus, liquid to binder ratio and water glass to NaOH ratio are frequently in the range of 5 M-15 M, 0.15-0.9, 0.2-0.48 and 0.5-2.5, respectively. Forming pressure shall be decided according to the initial water content of a plastic or moistened production brick mix. Excess pressure causes squeezing of alkali-activator from the mix that adversely affects the brick properties. With the increase in curing temperature, density and mechanical strength of bricks increase, whereas the water absorption and porosity decreases. From the review, it is concluded that 75-90°C seems to be the optimum temperature range. In order to design and develop sustainable alkali-activated bricks, suitable recommendations were drawn. Compared to conventional bricks, alkali-activated bricks found to be

stronger, durable and sustainable. Though alkali-activated bricks found better over conventional masonry units, there are still some limitations, i.e. poor legislation to use wastes, mix design complexity and lack of available standards.

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