Review article

A review: Alkali-activated cement and concrete production technologies available in the industry

Isabel Pol Segura [a],[*], Navid Ranjbar [b], Anne Juul Damø [a], Lars Skaarup Jensen [c], Mariana Canut [c], Peter Arendt Jensen [a]

[a] Department of Chemical and Biochemical Engineering, Technical University of Denmark, Kgs. 2800, Lyngby, Denmark
[b] Department of Mechanical Engineering, Technical University of Denmark, Kgs. 2800, Lyngby, Denmark
[c] Department of Research and Development, FLSmidth, Valby, 2500, Denmark

A B S T R A C T

A growing interest in alternative cements has emerged with the sole purpose of reducing the environmental footprint associated with cement production. One of the promising alternatives is to use non-carbonate materials such as alkali-activated materials. They have demonstrated to have a similar performance as traditional Portland cement and have the potential to significantly reduce CO_2 emissions. This paper reviews the main relevant technologies that are already available in the construction industry and explains how to consider them for alkali-activated cement and concrete production. This includes aluminosilicate pre-treatment methods (drying, grinding, and calcining) to increase the precursor's reactivity and degree of amorphization, alkali activation by two-part or one-part mix, as well as, mixing and casting fresh alkali-activated concrete ensuring low porosity and adequate strength development. This review also presents an overview of the alkali-activated cements market, providing examples of commercialized products, estimating related CO_2 and costs, as well as future considerations for standardization and commercialization. Most of the commercialized alkali-activated materials are two-part mixes despite their limitations for in-situ applications. CO_2 emissions can be reduced by more than 68% when compared to Portland cements. However, they have been estimated to be 2 to 3 times more expensive and the cost is primarily dependent on the aluminosilicate and alkali activators source.

1. Introduction

Concrete is the backbone of modern construction and is the second most used commodity in the world after water [1]. Cement is the binder of concrete, which brings together the inert aggregates. Among the different types of cement, Portland cement accounts for most of the world's cement production [2]. This worldwide reliance on Portland cement derives from the ease of handling it, the good performance (strength and durability) and the abundance of raw materials, which are mainly limestone, clays, sand and iron ore.

Around 4 billion tonnes of Portland cement are manufactured worldwide every year [2,3]. The tremendous environmental concern lies in the fact that for every tonne of Portland cement produced, around 0.5–0.6 tonnes of CO_2 are released into the atmosphere [4,5]. This leads to an annual contribution of approximately 8% of the total CO_2 anthropogenic emissions [6–8]. In addition, if there is no major change in the current cement production, the estimated contribution in 2050 has been estimated to rise up to 26% of the total

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* Corresponding author.
  E-mail address: isapol@kt.dtu.dk (I. Pol Segura).

CO_2 anthropogenic emissions [2]. Accordingly, after the Paris Agreement in 2015, which pursues maintaining the global temperature increase below $2 \, °C$, the cement industry focused its attention on different options including low- CO_2 alternatives to reduce the overall CO_2 emissions derived from cement production.

Most carbon emissions in cement production are inherent to the chemistry of cement and the high temperatures required for its processing (up to $1450 \, °C$). More than half of the emissions derive from burning the primary raw material constituent of cement, limestone (essentially calcium carbonate -$CaCO_3$), which decomposes into calcium oxide (CaO) and carbon dioxide (CO_2) [9–11]. This means that by changing the main raw material with a non-carbonate based precursor, the CO_2 emissions could be significantly reduced.

The cement industry has been working on different alternatives to lower the CO_2 emissions, such as energy efficiency measures, alternative binders or fuels, and carbon capture [12]. One of the promising binders are alkali-activated materials (AAMs), which also include the so-called geopolymers that are based on low-calcium precursors. AAMs and geopolymers are a Portland clinker-free alternative binder, which are synthesized from either aluminosilicate or calcium aluminosilicate precursors in highly alkaline conditions [13]. AAMs offer the advantage of being a versatile type of material where production can rely on local materials, as they can be synthesized by a great variety of aluminosilicate-based precursors (depending on availability, cost, reactivity, and application) [14]. Moreover, they can often achieve a comparable performance to Portland cement when using adequate mix proportions and outperform them with respect to acid and fire resistance [1,15,16]. On top of that, several investigations indicate that alkali-activated cements can reduce the CO_2 emissions by more than 50%, when compared to Portland cement [13,17,18].

However, it is important to note that to commercially embrace alkali-activated cements, there are several regulatory, supply chain, product durability, cost-effectiveness and sustainability issues that must be overcome. The entire supply chain and construction standards of the cement industry are highly associated with Portland cement production, and with a certain refusal to new types of cements especially without clinker.

Most of the AAMs scientific studies focus on enhancing their properties, long-term durability and developing optimal mix formulations [15,19]. However, there is a lack of documentation on how alkali-activated cement and concrete can be scaled-up from the laboratory to the existing construction industry. The main challenge for upscaling alkali-activated concrete is that there is no unique straightforward flowsheet as there is to produce Portland cement, and that mainly relies on their raw material flexibility. As mentioned before that has the advantage of using abundant raw materials available locally, which reduces transport costs and CO_2 emissions, however, it brings also higher degree of confusion to the concrete industry who does not know what kind of processes or equipment are needed to produce geopolymers or alkali-activated concrete.

This review intends to provide insightful information on the production technologies that are already available in the cement industry and could be used in the large scale manufacturing of alkali-activated cement and concrete, with limited investment [20]. Moreover, this review is completed by providing a summary of the current commercialized AAMs and geopolymers, as well as a CO_2 and cost assessment.

2. Background

AAMs are synthesized by the reaction of an aluminosilicate-based precursor ($xSiO_2 \bullet yAl_2O_3$), alkali activators (MOH and M_2SiO_3; where M is either alkali ion Na^+ or K^+) and water (H_2O). No Portland cement is required to produce alkali-activated binders, but AAMs could also be added as a supplementary cementitious material (SCM) and would then constitute a hybrid cement [13]. However, the focus of this review paper will solely be on pure alkali-activated cements and concrete.

Equations (1) to (4) show the sequence of reactions that occur during alkali-activation [21]. Eq. (1) presents that the reaction begins as soon as water is added to the mixture of aluminosilicates and alkali activators. Thereafter, Eq. (2) represents the mixing of
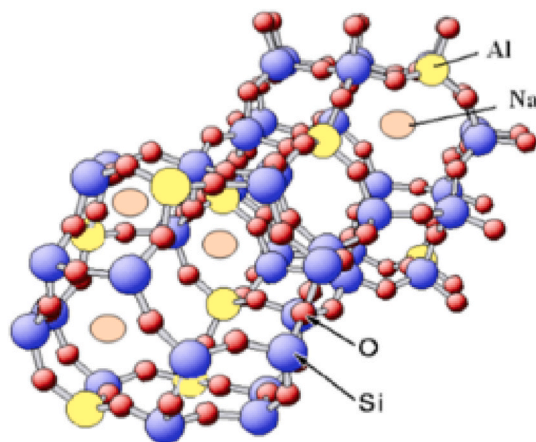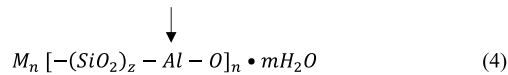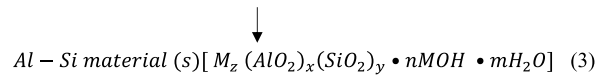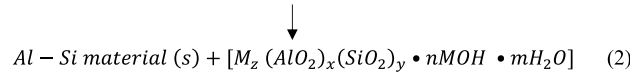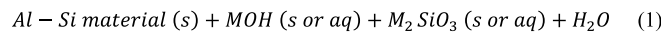

Fig. 1. Amorphous three-dimensional structure by the combination of alumina and silicate species with Na^+ ions in the interstices maintaining the electrical balance (N-A-S-H gel) (adapted from Ref. [23]).

aluminosilicates with the dissolved alkali activators, Eq. (3) the beginning of gel formation, and Eq. (4) shows the formation of a solid alkali-activated binder.

$$Al - Si\ material\ (s) + MOH\ (s\ or\ aq) + M_2\ SiO_3\ (s\ or\ aq) + H_2O \quad (1)$$

$$\downarrow$$

$$Al - Si\ material\ (s) + [M_z\ (AlO_2)_x(SiO_2)_y \bullet nMOH \bullet mH_2O] \quad (2)$$

$$\downarrow$$

$$Al - Si\ material\ (s)[\ M_z\ (AlO_2)_x(SiO_2)_y \bullet nMOH \bullet mH_2O] \quad (3)$$

$$\downarrow$$

$$M_n\ [-(SiO_2)_z - Al - O]_n \bullet mH_2O \quad (4)$$

Alkali-activation of low-calcium precursors leads to an amorphous three-dimensional structure by the combination of silica $[SiO_4]^{4-}$ and alumina $[AlO_4]^{5-}$ tetrahedra (Fig. 1), commonly known as N-A-S-H gel (sodium aluminosilicate hydrate). The alkalis, such as Na^+ or K^+, are embedded in the interstices of the structure, maintaining the electrical neutrality as a result of aluminium ions substituting silica ions in the tetrahedral layer [21,22].

For the alkali-activation of high-calcium precursors, calcium (Ca^+) has shown that it can act as a charge balancing cation like Na^+ and K^+. If some of the Na^+ is replaced by Ca^+, a C-A-S-H gel is formed (calcium aluminosilicate hydrate). Recent studies, show that the higher pH of the medium, the more C-A-S-H will prevail over N-A-S-H [24,25]. Likewise, the lower the concentration of alkalis and the more calcium-rich environment, the more C-A-S-H gel will be formed. The C-A-S-H gel chains present in alkali-activated cements have a similar tetrahedral structure as the C–S–H gel in Portland cement. The difference lies in the fact that C-A-S-H gels include aluminum in their structure, replacing some of the silicon in the bridging position [24–27].

2.1. Precursors and alkali activators

Common high- and low-calcium precursor materials include industrial wastes such as fly ashes, blast furnace slags, or natural sources such as clays [28]. Fly ashes and blast furnace slags have a limited availability, since they are by-products from other industries and they are already widely used as supplementary cementitious materials (SCMs) in Portland cement production. Meanwhile, clays are a natural occurring source that is widespread over the Earth's crust [29].

Regarding the most used alkaline reagents, they are generally a mixture of alkali hydroxides and silicates [9]. The alkali hydroxide increases the pH of the solution to favour the dissolution of aluminosilicates and help maintain the electrical neutrality of the structure as a result of aluminium ions substituting silica ions [23]. Meanwhile, the alkali silicate is generally added as a secondary source of silicates, which tunes the setting time of AAMs. An excess of silicate amount in the system, reduces the reaction rate and delays the setting time [25]. Most studies reveal that sodium-based alkaline reagents are the preferred activators, due to their greater capacity of liberating silica and alumina monomers, higher abundancy in natural resources, and their lower cost [14].

2.2. Alkali-activation process

The alkali activation process consists in mixing the aluminosilicate precursor with a mixture of alkali hydroxides and alkali silicates. In 1 m^3 of alkali-activated concrete, alkali hydroxide typically constitutes 1–5%wt, while the alkali silicate constitutes 3–10%wt [30–32].

A typical alkali hydroxide used is NaOH, which is commonly produced from sodium chloride (NaCl) by the chlor alkali process, in which chlorine gas is also a by-product. Another option for NaOH production is by reacting Na2CO3 with hydrated lime (Ca(OH)$_2$) to form NaOH and unreactive calcite (CaCO$_3$). In 2018, the global NaOH production was 83.6 million tonnes, and it is expected to increase to 98.76 million tonnes by 2024 [33]. The mainly used alkali silicate is sodium silicate (Na$_2$SiO$_3$). Commercial Na$_2$SiO$_3$ production involves the fusion of silica sand and sodium carbonate by the Solvay process, which requires temperatures of 1400 °C and has as a byproduct CO$_2$ [34]. The annual Na$_2$SiO$_3$ production in 2019 was 27.68 million tonnes and is expected to increase to 33.6 million tonnes by 2026 [35]. Due to the high energy intensity and CO2 emissions associated especially with sodium silicate production, there are also other options that are being investigated as a secondary silica source in AAMs, such as amorphous silica [34]. Amorphous silica can be obtained from thermal waste by-products such as silica fume, rice husk ash, or ground waste glass among others [36,37].

Conventionally, alkali activators have been mixed with the precursor in the liquid phase, as a solution (two-part mix). However, in the recent years, the potential of adding the alkali activators in the solid phase (one-part mix) has increasingly gained attention, as it implies selling a "dry bag" of alkali-activated cement similar to how Portland cement is sold [10]. In this way, two different production methods can be identified within alkali-activated cements.

2.3. Production methods

Fig. 2a and b shows a two-part and one-part mix, respectively. In Fig. 2a The precursors need to be pre-treated, and then they are

mixed with an alkali-activating solution. Meanwhile, in Fig. 2b, the pre-treated precursors are mixed with solid alkali activators in the mixer. They are generally pre-ground together with the precursor to decrease their particle size and promote their subsequent dissolution..

In both types of production methods (Fig. 2a and 2b), the pre-treatment of industrial by-products such as coal fly ash and ground granulated blast furnace slag (GGBFS) is often sufficient with drying and grinding steps to homogenize the material and increase their reactive surface. However, when pre-treating natural mineral clays, a thermal treatment known as calcination is commonly used to increase their amorphous content [38]. Another option to improve clay mineral reactivity would be a mechanical processing. Grinding reduces particle size and may contribute to nanostructural disorder in crystalline networks [39]. However, some believe that only grinding cannot induce full amorphization [40]. Instead, some propose that mechanochemical activation may be more effective. In this way, successively combining different pre-processing treatments (thermal, mechanical, and chemical) would be very beneficial to further explore [20,40].

After alkali-activating the pre-treated precursors, inert aggregates such as sand and gravel are added to the mixer to form alkali-activated concrete. They increase the stability of hardened concrete, especially as they reduce the shrinkage [41]. Admixtures are commonly included to improve the durability of concrete or control setting and hardening (examples of common additives are water reducers, superplasticizers, accelerators or retarders) [42].

After homogenously blending all the raw materials, the fresh alkali-activated concrete can be either pre-cast with controlled conditions or cast-in-situ at the construction site. Subsequently, the alkali-activated concrete will begin to set, harden and gain strength over time. During this time, known as the curing period, the surrounding environment and protective measures adopted are crucial for the long-term properties of the concrete [43].

The commonly applied industrial technologies available for pre-processing, alkali-activating, and mixing the three main aluminosilicate precursors are presented in this review (coal fly ash, GGBFS, and clays) as well as casting of alkali-activated products and some CO_2 and economic considerations regarding alkali-activated cement and concrete production.

3. Aluminosilicate pre-processing technologies

Coal fly ash and slags are aluminosilicate by-products from the coal combustion and iron manufacturing industry, respectively. Meanwhile, clays are natural resources that can be mined and quarried from underground or open pits. Those materials are already
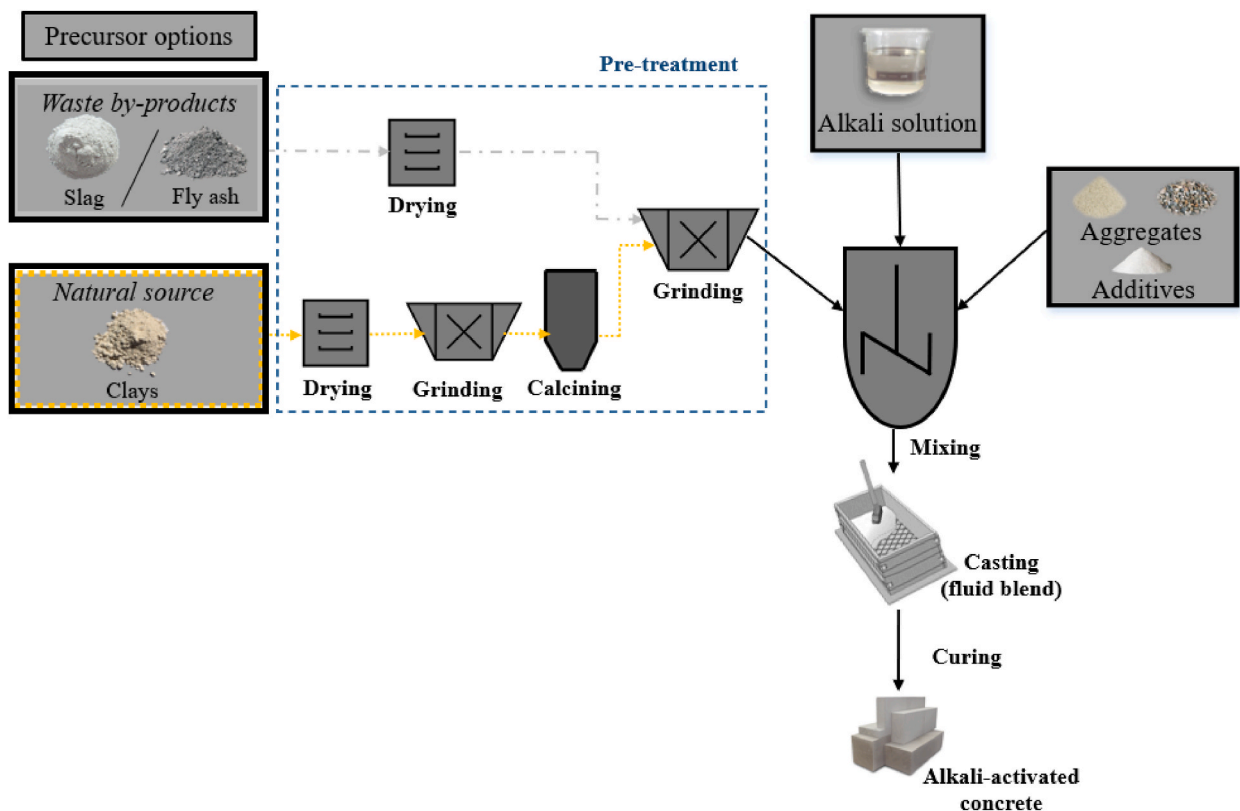

Fig. 2a. Production of alkali-activated concrete by a two-part mix. The aluminosilicate precursors undergo a pre-treatment to increase their reactivity and then they are mixed with an alkali solution, aggregates and additives. The graphical symbols of the pre-treatment and mixing units follow the ISO standards.
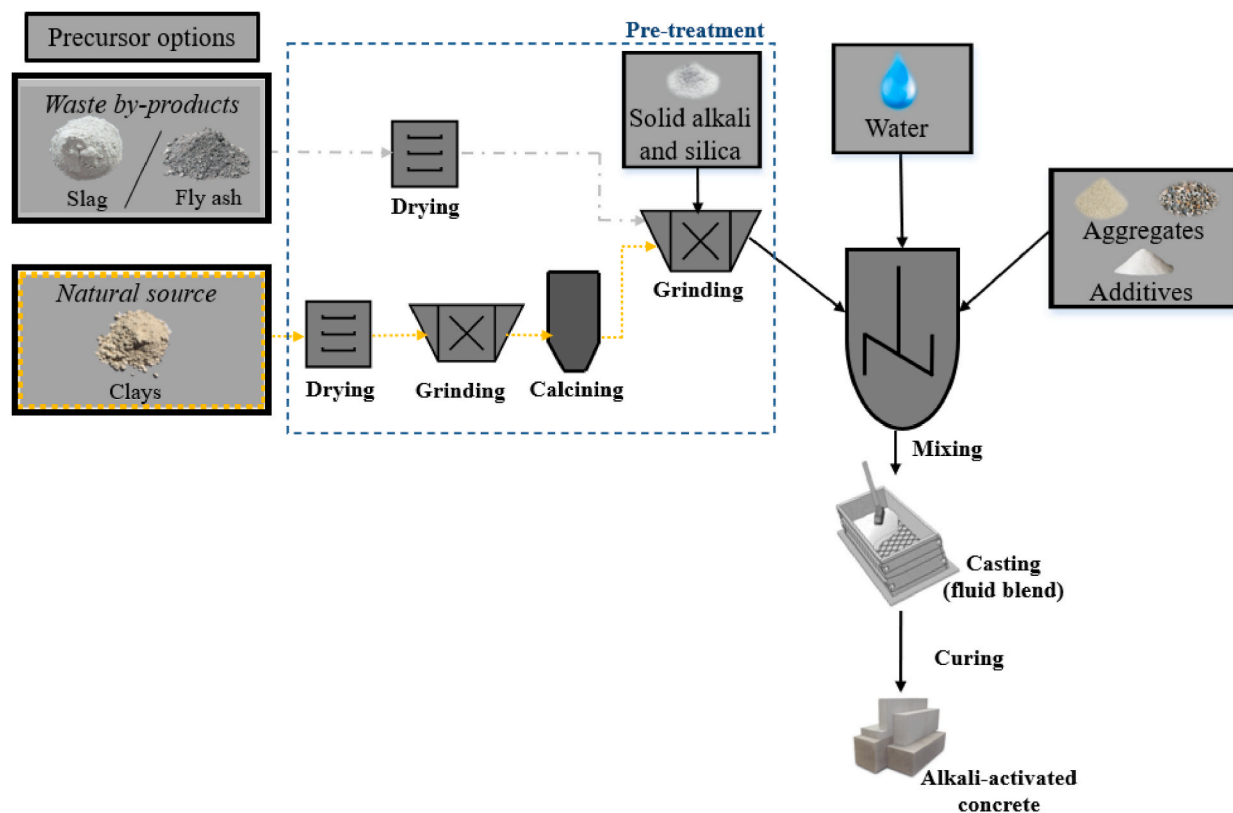
Fig. 2b. Production of alkali-activated concrete by a one-part mix. The solid alkalis are pre-ground to favour their dissolution and thereafter, they are mixed with the pre-treated aluminosilicate precursors, water, aggregates and additives. The graphical symbols of the pre-treatment and mixing units follow the ISO standards.

used as SCMs in blended OPC production, and several studies [44,45] focus on improving their pre-treatment technologies to enhance their reactivity, increase the degree of clinker substitution, and reduce the clinker-to-cement ratio to achieve lower carbon emissions. However, in this review, the focus is on technologies pre-treating those materials to produce clinker-free binders. Depending on the type of aluminosilicate source, different extraction methods are used and different pre-processing technologies (mechanical, thermal and/or chemical pre-treatments) can be employed. Since chemical activation is required to synthesize the binder in AAMs, this will be covered individually in Section 4. Meanwhile in this section, the potential mechanical and thermal pre-treatments for the main types of precursors are presented.

3.1. Coal fly ash

Coal fly ash is a byproduct of coal combustion plants. Annually, 700 to 750 million tonnes of coal fly ash are produced, and approximately 6 billion tonnes are in stockpiles [2]. The pulverized coal is burnt in a boiler, and fine particles (fly ash) are carried off in the flue gas, from where they typically are collected by means of electrostatic precipitators, baghouses, or mechanical collectors such as cyclones [46]. When fly ash is collected from coal combustion plants, it has between 50 and 90% mineral matter in the shape of glassy amorphous particles. The formation of those glassy amorphous particles is due to the fast cooling after the combustion, and they have been found to enhance alkali-activation [47]. Typical bituminous coal fly ash has a composition that mainly contains 30–60% SiO_2 and 20–30% Al_2O_3, however, it can also contain more than 10% of CaO [48]. Particle size of fly ash ranges from 0.3 to 250 μm [49, 50] with a major fraction in the range of 20–25 μm.

After fly ash has been collected, a series of steps of disagglomerating, drying and grinding are performed to remove moisture and decrease the particle size. It has been reported that the higher the fineness, the greater the reactive surface area [51–53]. However, fly ash particle size below 45 µm should be sufficient for AAMs production [50,53].

Utilization of low-calcium fly ash seems to be more suitable for AAMs production than high-calcium fly ash. The major issue with high-calcium fly ashes is that they have an extreme fast setting also known as flash setting [54]. In addition, in the coming years, coal-power plants will shift to renewable sources such as wind, solar or biomass. Coal fly ash could still be obtained from the existing stockpiles for some years. By using the stockpiled fly ash as aluminosilicate source for AAMs, the environmental problem of landfill would also be reduced. However, the main drawback of using stockpiled fly ash is that it might contain impurities and moisture, meaning that they must undergo some extra pre-conditioning [18]. For this reason, most of the cement manufacturers prefer fly ash

coming directly from the power stations as lower handling costs are required.

Nevertheless, with the new renewable energy sources, biomass fly ash will be very probably an abundant by-product [2]. Hence, biomass ash is already being studied as aluminosilicate precursor for alkali-activated cements and it has been found to reach compressive strengths above 20 MPa [55,56]. Moreover, biomass ash has a certain advantage with respect to coal fly ash. Biomass ash has generally a higher content of potassium than coal fly ash, and this could lead to a potential reduction of the level of alkali activator used [57].

The company Agico Cement has designed a drying and grinding processing plant for fly ash using a horizontal dryer and a ball mill (Fig. 3) [58]. In this processing system, the stored fly ash is first sent to the dryer. Then, the dried fly ash is fed into a horizontal ball mill to be ground, and with an elevator, the material is led to a separator, where it is classified according to size. The material with particle size not greater than 45 μm is sent to the fine fly ash silo, while the coarser ash is sent back to the ball mill, and the process is repeated. The whole system is also equipped with a dust collector to avoid any dust emissions.

The Agico fly ash processing plant has a capacity of 100 tonnes/hour with a classification efficiency of up to 85% and provides a Blaine fineness in the range of 2000–5500 cm^2/g [58].

Other fly ash processing plants are also available, such as FEECO, which has a similar system as Agico Cement and can provide similar Blaine fineness [59]. However, they use a horizontal pug mill instead of a ball mill and their production capacity can be up to 250 tonnes/hour.

3.2. Ground granulated blast furnace slag (GGBFS)

There are different types of slags, but the most commonly used as an aluminosilicate precursor and in the cement industry is ground granulated blast furnace slag (GGBFS), which is a by-product from the iron industry. When the iron ore, coke, and limestone melt in a furnace at 1800 °C, molten iron and slag are formed [60]. The slag floats on the top surface of the furnace and is rapidly removed and quenched with water. This fast quenching forms granulated slag particles that can be collected as a by-product. Annually, the global production of GGBFS is around 300 million tonnes [2]. Its main constituents are mostly rich in CaO (40–50%) and SiO$_2$ (25–30%), with some Al$_2$O$_3$ (7–15%) [61,62], and is often referred to in the literature as calcium aluminosilicates, rather than just aluminosilicates. Since GGBFS is a semi-hydraulic material and has a high content of CaO, its hydration mechanism is similar to Portland cement, generally leading to a faster setting and earlier strength development than fly ash and clay alkali-activated cements [,63]. The morphology of GGBFS particles is also physically like limestone particles, which are characterized by an angular shape [64,65].

After collecting the quenched GGBFS particles, they are generally dried and ground to increase their reactive surface area and degree of amorphization.

Nevertheless, it is important to note that a high share of the total GGBFS is already being used as SCM in blended OPC cements. It is mainly used for high strength concrete applications and for fast setting purposes. Therefore, a main drawback of GGBFS is their lack of availability due to its extensive use as SCM. Moreover, the iron ore refineries are responsible for a 4–7% of anthropogenic CO$_2$ emissions globally [66], and many blast furnaces are already being substituted by greener alternatives (such as Electric Arc Furnace) which do not produce GGBFS [67,68].

The company Chaeng has designed a GGBFS processing plant (Fig. 4), in which they use a horizontal dryer and a vertical mill to grind GGBFS [69]. In this plant, the stacked slag is first dried using heat from a heat generator and then fed into a vertical mill. The ground particles are then sent to the separator. The particles that meet the requirements (<45 μm) pass through a breeze chute to the finished product storage silo, and otherwise, they are sent back to the vertical mill until the desired particle fineness is reached. Chaeng's plant can process 70 tonnes/hour and reach a Blaine fineness between 4200 and 4500 cm^2/g [69].

Among other GGBFS processing plant examples, there is ECOCEM's and FLSmidth's processing plant. ECOCEM's Moerdijk plant (Holland) is similar to Chaeng's, having a similar processing capacity (74 tonnes/hour) and Blaine fineness (around 4200 cm^2/g [44]). However, instead of using a vertical mill they use a horizontal rotating mill drum [70]. Comparably, FLSmidth uses typically a vertical flash dryer and a vertical roller mill. Their processing capacity can vary from 50 to 650 tonnes/h, depending on the product's fineness
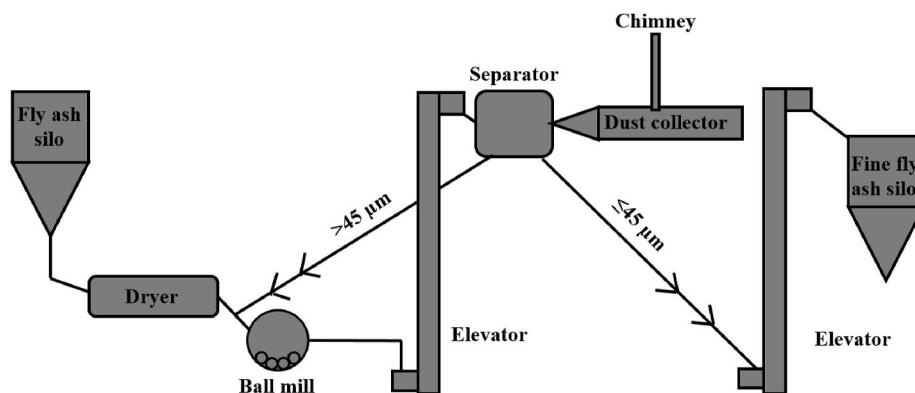

Fig. 3. AGICO Cement's fly ash processing plant flow chart using a horizontal dryer and a ball mill to grind (flow chart adapted from Ref. [58]).
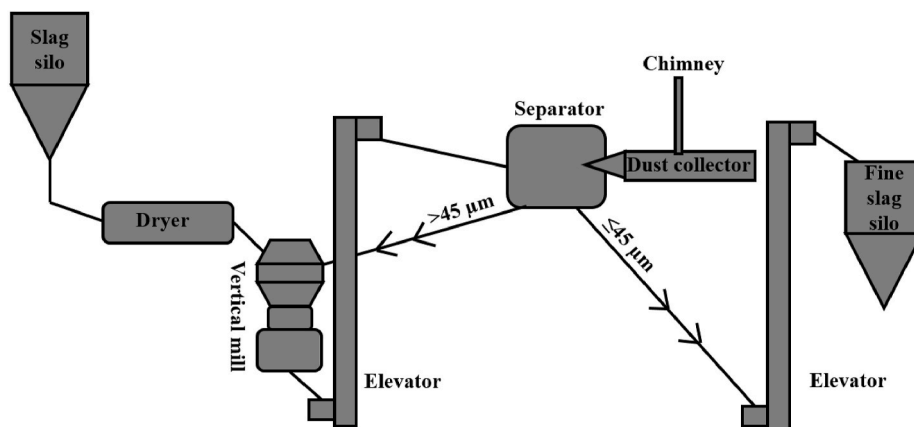
Fig. 4. Chaeng's GGBS processing plant using a horizontal dryer and a vertical mill to grind (adapted from Ref. [69]).

and grindability [71].

3.3. Mineral clays

The other most conventional aluminosilicate precursor is mineral clays, which are a naturally abundant resource covering 2/3's of the Earth's crust, and mainly composed of SiO_2 (50–70%) and Al_2O_3 (20–30%) [72–74]. They need to be either mined from underground or from open pits using surface mining equipment such as draglines, power shovels, front-end loaders, backhoes, scraper-loaders, or shale planers [75]. Recovery of mineable beds of clays in open-pits is generally 100% and for underground operations is around 75% [76]. The mineral clay most used in alkali-activated synthesis is known as kaolin clay, due to its suitable chemical and structural composition [21,77]. However, other mineral clays such as smectites and illites or a mixture of them are also being investigated [78,79]. There are then different types of clay pre-treatment technologies (chemical, mechanical, thermal, etc.) depending on their application, however, as construction material, the most investigated pre-treatments consist of mechanical grinding and/or thermal activation [21].

Mechanical treatment consists of drying and grinding the clays with balls, rods, pebble mills, or similar. The particle size after grinding can be in the range between 2 mm to a few microns, depending on the thermal activation treatment that will then be applied (rotary or flash calcination) [80]. Grinding the clays does not only reduce the grain size or deagglomerate but the clay is also delaminated and its structure becomes more accessible and with a higher reactive surface area [81].

The retention time and intensity of the milling are key parameters in the clay's reactivity. Long and intense milling of clays can result in a high amorphization of the clays, which positively influences the reactivity towards alkali-activation. However, very fine particles can lead to agglomeration and water absorption [82]. Thus, those two parameters have to be carefully balanced when grinding clays.

After the mechanical grinding of clay, around 40% of the initial raw clay is typically discarded as waste [76]. The discarded fraction is generally off-grade clays, quartz, mica, or feldspar. The clays are then usually thermally treated to extensively transform crystalline phases into reactive amorphous phases, which is the active constituent that determines the strength of alkali-activated cements. The thermal treatment is known as calcination, and it demands temperatures that can range between 600 and 950 °C [83], depending on the type of clay used. At these temperatures, the dehydroxilation of strongly bound hydroxyl ions on the aluminium layer occurs [22]. An optimal calcination temperature is essential, as higher temperatures than the optimal can induce recrystallization of phases and, thus, negatively affect the reactivity towards alkali-activation [16].

Several types of mineral calciners are used in the industry. However, for processing mineral clays as construction materials, the most typical are: rotary calciners and flash calciners.

- **A rotary calciner** installation conventionally includes 2–3 stages of preheating cyclones, a cylindrical rotary kiln, and a rotary cooler. The cylindrical rotary kiln generally has a length of 20–40 m, a diameter between 3 and 6 m and has a slight inclination [84, 85]. The material to calcine is fed at the elevated end of the cylinder at the top preheater stage by chutes, screw conveyors, or slurry pipes. After the kiln, the calcined product is discharged from the lower end of the cylinder into the cooler, where the material quenches rapidly with a counterflow of cold gas. Material movement through the kiln results from the combined effects of the kiln inclination and the rotation of the cylinder.

Fig. 5 illustrates a typical rotary kiln design that can operate as a rotary clay calciner [86].

Rotary clay calciners generally operate within temperatures of 700–900 °C, with flexible feed size and have residence times of 0.5–1h [80]. Most rotary clay calciners have countercurrent air and material flow to achieve the most energy-efficient moisture content reduction. Some of these calciners are refurbished idle rotary kilns from cement plants [83], which lowers the capital

expenditure (CAPEX) significantly.

- A **flash calciner** installation consists of a series of preheating cyclones, a cylindrical calciner vessel, and a series of cooling cyclones (Fig. 6 [87]). Raw dry feed material is introduced at the top stage preheater cyclone. Thereafter, the clay particles are carried throughout the preheating cyclones until the calciner stage, where temperatures can reach 850–1000 °C [82]. The fuel is injected directly into the calciner stage to complete the conversion in a few seconds. The flash calcination is mostly used for materials with particle sizes below 1 mm, as the finer the particles, the less time to reach their dehydroxilation temperature (600–950 °C, depending on the type of clay) [83,88]. Subsequently, the calcined clays leave the calcining stage and they are quenched at the cooling cyclones. Fresh air is introduced to the bottom cyclone to cool off the clays, simultaneously the air is heated. Heated air is then taken to the calciner as combustion air [87].

After cooling the calcined clays, they might need to be sent to another grinding unit to decrease the particle size (to reach 40-90 μm average particle size [89,90]) and resolve any type of agglomeration that might have occurred during calcination.

Another factor to consider in industrial clay calcination for construction purposes is the ability to keep a grey colour of the product. The iron (Fe) content often found in clay can result in the product being reddish depending on how it combines to oxygen (O). Iron compounds in the form of hematite (Fe_2O_3) have a reddish colour and when in the form of magnetite (Fe_3O_4) have a dark-greyish colour. At higher temperatures, the iron is mostly in the form of magnetite. The most important aspect to maintain the grey colour is to keep the iron in the magnetite form during the cooling of the clay after calcination. The colour control technologies applied in clay calcination can be either quenching, applying a controlled atmosphere or using organic modifiers [91]. A controlled atmosphere can be achieved by either operating at reducing conditions, injecting fuel close to the kiln discharge or in the cooler and/or injecting fuel mixed directly with the raw clay. Note, that it has been demonstrated that the binder's colour does not correlate with the concrete's performance, but such feature is commonly requested for end-user's market acceptance, although it increases the product cost.

3.4. Selection of aluminosilicate pre-processing technologies

The quality and reactivity of aluminosilicate precursors depends greatly on the pre-treatment applied (mechanical, thermal, chemical or a combination of those). Thus, a careful selection of the pre-treatment technologies should be performed, as it directly influences the properties of resultant alkali-activated concrete.

The selection of aluminosilicate pre-processing technologies involves making decisions on the type of dryer, type of mill and type of calciner. Those decisions shall be based on the feed size, feed composition, product fineness, moisture content, grindability, abrasiveness, degree of amorphization, installation space, and budget.

3.4.1. Dryers

The objective of the dryer unit is to remove the moisture from the raw materials. There are two main dryers used in large scale mineral processing industry: rotary and flash dryers. The rotary dryer consists of an inclined rotating cylinder with internal lifter or blades, where the material is dried through direct contact with hot gases. Rotary dryers are used for drying wet and/or abrasive materials with different granulometric properties [92,93]. In the flash dryer, the material is fed to a vertical vessel with a mixing helix and hot gasses flow from bottom to top to dry the material [94,95]. Flash dryers can rapidly treat sufficiently fine-grained materials. Table 1 compares the main features of both technologies.

From Tables 1 and it can be seen that flash dryers can be combined with ball/hammer mills, so that drying, and grinding are merged in a single unit. Integration of drying and grinding in one combined system is common practice in the cement industry and is encouraged for AAM greenfield applications. Another typical example of this combined systems are dryer crushers. They are suitable for soft, coarse, and moist materials that need to be dried, crushed and deagglomerated [96,97].
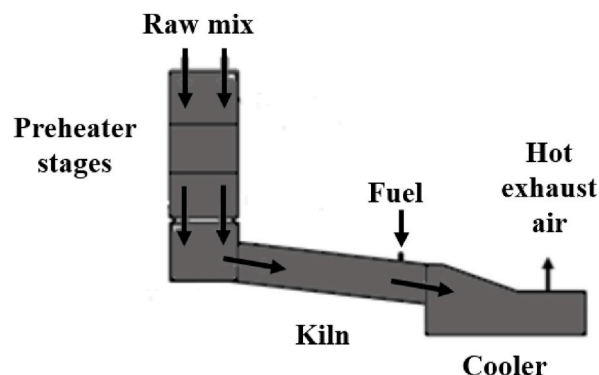

Fig. 5. Schematic of a rotary kiln that can operate as a rotary clay calciner (adapted from Ref. [86]).
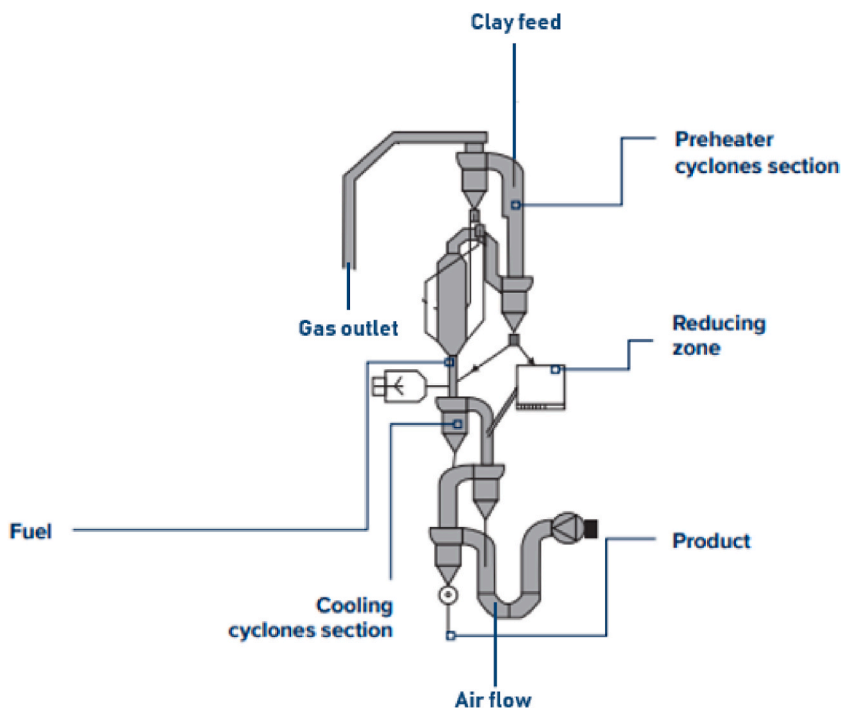
Fig. 6. Schematic of a flash calciner with preheater stages and cooling cyclones (adapted from Ref. [87]).

Table 1
Comparison of the main features between a rotary dryer and a flash dryer [93,95].

Rotary dryer	
• High flexibility for drying materials of varying size and moisture (fine and lumpy) • Controlled drying process while mantaining low fuel consumption • High throughput • Rugged design with high availability and low mantainace cost • Energy-efficient by reutilizing hot offgas from other processes	
Flash dryer • Low space requirement • Effective drying with lower OPEX[a] than rotary drying for same moisture level • Low maintenance and repair equipment • Feed size up to 2 mm and moisture up to 35% • Simple operation • Can be combined with a ball/hammer mill • Higher CAPEX[a]	

[a] OPEX: Operating expenditures, CAPEX: Capital expenditures.

3.4.2. Mills

The purpose of grinding is to produce homogenous raw material feeds with a proper fineness ($d_p < 45\mu m$) that will enhance the reactivity towards alkali-activation. The most common dry grinding systems in the industry are ball mills, vertical roller mills and high-pressure roller press.

i. **Ball mills** consist of a rotating cylinder in which the material is crushed by steel balls moved with the centrifugal force of rotation [98]. They were the most predominant type of mill until 1970s. Several types of ball mills are shown in Fig. 7.

ii. In **vertical roller mills** (VRM), the material is fed into a hydraulic system that pressures act on it between the roller and a grinding table [99]. They appear to have a better grinding efficiency and higher production capacity than ball mills, thus VRMs are the mill selected as the first choice for most of the new AAM installations. Several types of VRMs are presented in Fig. 8.

iii. **High-pressure roller presses** consist of two cylindrical rollers, one is fixed, and the other one is free to move. The material is fed above the rollers, so-called choke feed, and pulled in between the rollers. A very heavy grinding pressure (between 100 and 200
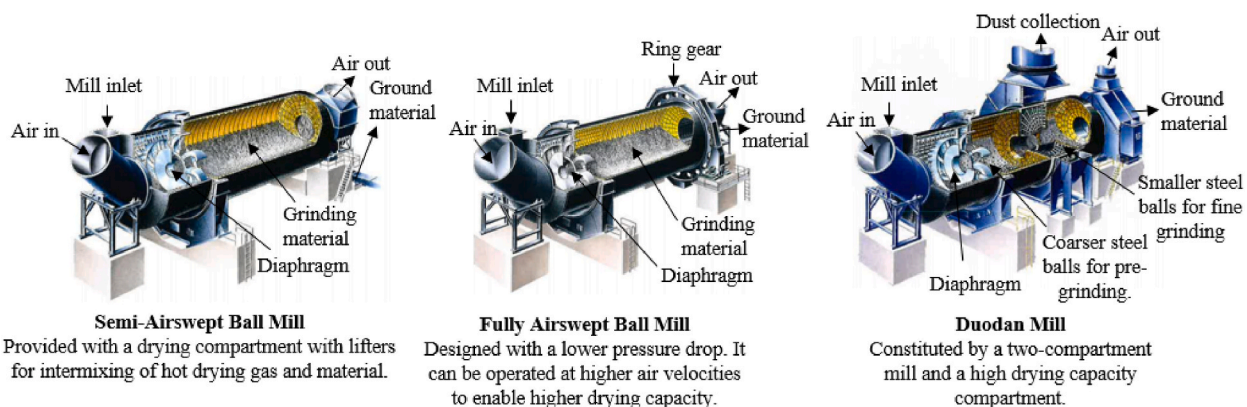
Semi-Airswept Ball Mill
Provided with a drying compartment with lifters for intermixing of hot drying gas and material.

Fully Airswept Ball Mill
Designed with a lower pressure drop. It can be operated at higher air velocities to enable higher drying capacity.

Duodan Mill
Constituted by a two-compartment mill and a high drying capacity compartment.

Fig. 7. Three types of ball mill: Semi-Airswept, Fully Airswept and Duodan Mill that can be used to grind aluminosilicate precursors [102–104].

MPa) is supplied from hydraulic cylinders working on the moveable roller [100,101]. The roller presses are used to finish grinding raw materials or pre-grinding in connection with a ball mill. Fig. 9 shows a sketch of a high-pressure roller press.

Comparing all the various grinding systems presented, different aspects should be considered when making a decision on which type of mill should be used to pre-treat precursors:
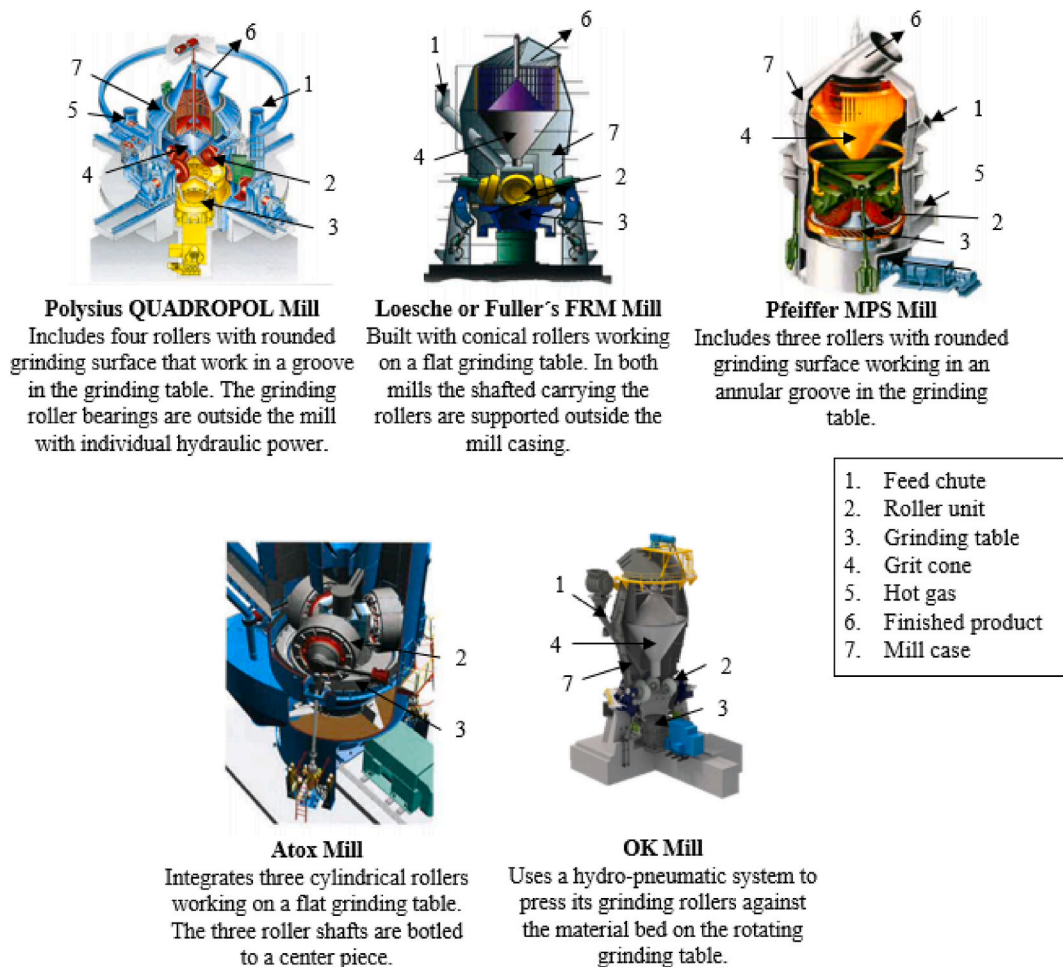

Polysius QUADROPOL Mill
Includes four rollers with rounded grinding surface that work in a groove in the grinding table. The grinding roller bearings are outside the mill with individual hydraulic power.

Loesche or Fuller´s FRM Mill
Built with conical rollers working on a flat grinding table. In both mills the shafted carrying the rollers are supported outside the mill casing.

Pfeiffer MPS Mill
Includes three rollers with rounded grinding surface working in an annular groove in the grinding table.

1. Feed chute
2. Roller unit
3. Grinding table
4. Grit cone
5. Hot gas
6. Finished product
7. Mill case

Atox Mill
Integrates three cylindrical rollers working on a flat grinding table. The three roller shafts are botled to a center piece.

OK Mill
Uses a hydro-pneumatic system to press its grinding rollers against the material bed on the rotating grinding table.

Fig. 8. Five types of vertical roller mills: Quadropol, FRM, MPS, Atox or OK mills that can be used to grind aluminosilicate precursors [71,105–108].

- **Grinding capacity.** Ball mills can operate at a maximum of 400 t/h, while vertical roller mills more than 500 t/h [110,111]. The roller press has generally a maximum capacity of 300 t/h [112,113].
- **Drying capacity.** For small and medium size ball mills, the internal drying capacity is adequate for most raw materials, however, as the mill size increases, then supplementary drying (for example, flash dryer) is needed. Vertical roller mills can be used for raw materials with more than 20% moisture contents independent of mill size, without any need for external drying [113]. In a roller press, the drying takes place outside the unit. The standard drying capacity is 5% [112].
- **Grindability.** For standard grindability and moderate moisture contents the total energy consumption of a ball mill is about 20 KWh/t, while the vertical and roller press have a total energy consumption of around 75% of the ball mill [101,114]. For increasing moisture contents, the energy consumption of vertical roller mills becomes even more advantageous.
- **Feed size.** The acceptable feed size of a ball mill is 25–30 mm, independent of mill size. The vertical roller mills can accept feed sizes up to 100–150 mm, depending on the mill size [111–113]. For the roller press, the feed size depends on the roller diameter. The maximum feed size is 30–60 mm [112].
- **Maintenance and reliability.** Ball mill, vertical roller mill and high-pressure rollers can be considered to be mechanically reliable. The wear rate in g/t is higher in the ball mill than in the vertical mill or high-pressure roller, but as the unit cost for wear parts is lower for the ball mill than for the vertical or high-pressure mill, the total cost of wear parts is similar for the three mill systems [113,115–117].
- **Investment cost.** Vertical roller mills have been reported to be approximately 20% cheaper than ball mill systems and high-pressure roller press [109,110,118].

3.4.3. Calciners

After drying and grinding, most mineral clays also need to be thermally activated to increase their reactive surface area. The working methods of a rotary calciner and a flash calciner have been explained in Section 3.3. Nonetheless, an overview of the characteristics and performance indicators of each calciner need to be considered before its selection (Table 2).

Flash calciners have a lower energy demand due to a good heat recovery, offer $\sim 10\%$ enhanced pozzolanic reactivity, and a higher degree of control (lower variability) over the clay conversion, and colour control minimizing the commercial risks from quality guarantees. In addition, it has limited maintenance as there are no moving parts and reduced footprint. Nevertheless, flash calciners require a smaller feed particle size demanding more intense grinding and have a higher capital cost than a rotary clay calciner.

In short, several factors should be considered when selecting a dryer, a mill, or a calciner for pre-treating the aluminosilicate precursors. Those factors substantially vary depending on the installation location, initial feed characteristics, product requirements, and specific project demands. The different possible process combinations needed to pre-treat aluminosilicates for subsequent alkali-activation shall be further investigated. Likewise, re-use of idle equipment from a well-established Portland cement plant shall also be considered.

4. Alkali-activation production methods

Depending on whether the alkaline activators are added in the liquid or solid phase there are two main alkali-activation methods as introduced in Section 2. Those are known as two-part and one-part, respectively. Two-part alkali-activation is formed by the reaction between a concentrated aqueous solution of alkali hydroxides and silicates, and a solid aluminosilicate precursor, which is two-part in addition to water. In one-part alkali-activation, a dry mixture constituted by a solid alkali activator and a solid aluminosilicate precursor is mixed with water [18,63,123].
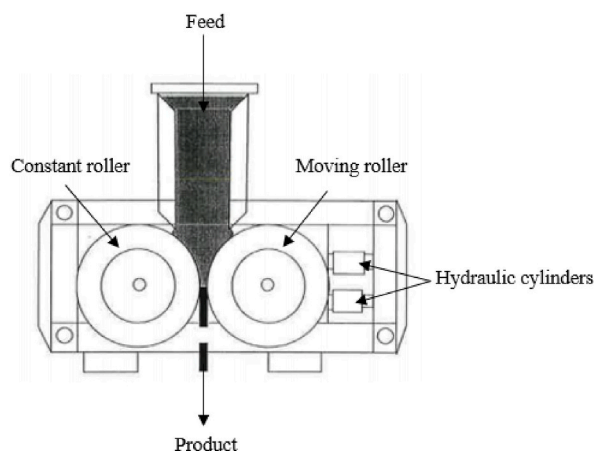

Fig. 9. High-pressure roller press sketch, mainly used for finish grinding or pre-grinding combined with ball mill [101,109].

Table 2
Characteristics and performance indicators for flash and rotary kiln clay calciners [91,119–122].

Characteristics	Flash calciner	Rotary kiln calciner
Heat source	Air atomized natural gas burner, and additional fuel is added by injectors (natural gas, propane) along the chamber	Dual fuel burner located outside discharge end of the kiln. It can fire natural gas, oil, solid fuels independtly or simultaneously
Optimal temperatures and residence times	Range 850–950 °C 0.5-3 s	Range 800–900 °C, 0.5–1h
Pozzolanic reactivity (%)	$\sim 80\%$ at 850 °C depending on clay	60–70% at 850 °C depending on clay
Colour control	Under reducing conditions very good results can be achieved	Under reducing conditions and 2% wet O_2 at inlet it can be achieved but with limited degree of control
Pre-treatment of clays before calcination	Dehydration and deagglomeration of clay to have <1 wt% moisture and <1 mm particle size	Flexible in moisture, size and quality of the feed material to kiln
Emissions	CO and SO_2 emissions at the flash calciners baghouse exit higher than at calciner exit, since feed releases organics and SO_2 in the upper stages	Very low emissions under oxidizing conditions. However, under reducing conditions, higher emissions of CO and SO_2 are observed, and need approppiate control provisions to be limited
Energy consumption	Low heat loss and energy consumption from calcination (450–600 kcal/kg)	Higher thermal energy consumption from calcination (700–800 kcal/kg)
Capital costs	High Capex	Low Capex (specially from kiln retrofits)
Other	Low mantainace as it has few rotating parts	Well-known established technology, simple and robust process

4.1. Two-part alkali-activation

The alkali-activation consists first in the dissolution of the solid alkalis and silicates with water and then mixed to constitute the alkali solution silo. The dissolution of alkalis in water is an exothermic reaction, and so it should be generally prepared 24h before mixing with the pre-treated aluminosilicate powder to allow heat dissipation [124]. Once the pre-treated aluminosilicate precursor is mixed with the alkali solution, the alkali-activated paste is formed. Shortly after, the inert aggregates (sand and gravel) and additives are added to the fresh paste to form alkali-activated concrete. Fig. 10 shows a schematic of two-part alkali-activated production.

There are several studies that show that two-part mixes have comparable mechanical properties as Portland cement and that they can even outperform in thermal and acid-resistance applications [66,125]. Furthermore, for similar compressive strengths they have also demonstrated higher flexural strength than for Portland cement [126].

Two-part mixes are the choice of most active AAMs companies, and constructions such as the Melton Public Library in Melbourne, Global Change Institute or the Brisbane Wellcamp Airport, which were built using two-part slag and fly-ash alkali-activated concrete. Nonetheless, they have also several disadvantages since handling large volumes of a viscous and corrosive solution is hazardous, and it is more expensive than transporting dry activators [127].

4.2. One-part alkali-activation

The powdered solid alkali activator is mixed with the powdered aluminosilicate precursor, provided that their particle size is small enough ($d_{max\approx} 45\mu m$). Thereafter, the water, aggregates, and additives are added to the solid mixture. Solid alkali activators enable the integration of the alkalis in the pre-treatment of the aluminosilicates as Fig. 11 presents. This means that grinding of alkalis and aluminosilicates can be done simultaneously (mechanochemical activation), or even include the alkalis in the clay calcination (thermal activation). Co-grinding aluminosilicates and alkali activators is commonly implemented by ball milling [128].

The addition of the alkalis in the clay calcination is known in the literature as alkali fusion [18] and it has been demonstrated to
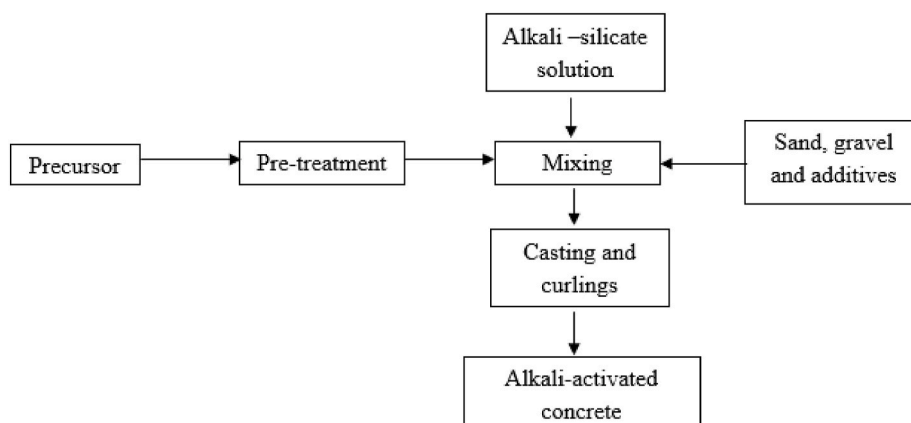

Fig. 10. Schematic of a two-part alkali-activated concrete production method.

favour alkali-activation [129–131]. This is probably due to sodium which increases non-bridging oxygen's in the clays, enhancing their dissolution and reactivity. Moreover, a study [132] has observed that the addition of NaOH in the clay calcination might be synthesizing sodium silicate as consequence of the reaction between NaOH and quartz (SiO_2).

$$SiO_2 + NaOH \rightarrow Na_2O \bullet nSiO_2 + H_2O$$
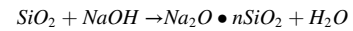
Few studies have focused on hydration kinetics and long-term properties of one-part mixes. Nevertheless, one-part alkali-activated pastes have been found to frequently set too fast due to the heat generated from the dissolution of the anhydrous NaOH and Na_2SiO_3 [133]. This can consequently increase the porosity, affect the compressive strength and durability. However, the setting time seems to be improved when preparing mortar or concrete, as the sand or admixtures absorb some heat and the settings are delayed [123].

Some one-part alkali-activated synthesis routes have been proposed and reviewed [18,134]. *Xun Peng* [132], calcined low-grade kaolin that contained a lot of quartz and integrated the solid NaOH in the calcination. The geopolymeric cement paste had a compressive strength of 63 MPa after 28 days [132]. Other studies, have added alkali-rich solid wastes such as silica fume, red mud (byproduct of the alumina production), biomass ashes or waste glass to the aluminosilicate precursors. The experimental studies show that they can reach 20–40 MPa after 28 days [132].

Recent investigations have used granular anhydrous sodium metasilicate powder to synthesize one-part mixes from fly ash blended slag and high calcium fly ash [63]. This led to 50 MPa compressive strengths after 28 days [63].

5. Alkali-activated concrete mixing technologies

Regardless of which alkali-activation procedure is used, one- or two-part mix, the construction industry has a great interest in resembling the mixing and casting of alkali-activated concrete to one of those commonly used in the Portland cement based concrete industry. Hence, Portland's mixing and casting technology can be reused to produce alkali-activated concrete, with the addition of storage tanks or silos for the alkali activators. The mixing unit can be a common cement and concrete mixer, such as those typically used in the construction industry. There are two main types of cement and concrete mixers: *batch and continuous*. The *batch mixers* are the ones that produce one concrete batch at a time, while in *continuous mixers,* the materials are continuously fed into the mixer at the same rate as concrete is discharged. *Batch mixers* can be horizontal (drum mixers, such as the truck mixers) or vertical (pan mixers). Truck mixers offer the advantage of homogenously mixing wet concrete while being transported to the construction site. *Continuous mixers* are used for applications, which require a short working time, they are placed at remote locations or have a long unloading time [115]. As in traditional cement and concrete mixing, steel-based mixers are preferred for alkali-activated materials since they can be highly corrosive to other metals (such as aluminium) when they are in a fresh state.

The selection of a mixer, mixing time, or stirring rate should be appropriate to have a homogenous alkali-activated concrete [116]. Nonetheless, it should already be noted that one-part mixing needs longer residence mixing times for dissolution and polycondensation. After mixing, the wet alkali-activated concrete should have a sufficient consistency and be able to be shaped in different formworks without segregation. Generally, alkali-activated cements' workability is lower than for Portland cement because the presence of silicate (SiO_2) in alkali-activated materials seems to bring a sticky characteristic, especially at low water/solid ratios [117]. Nonetheless, alkali-activated concrete has shown to compact well by using vibrators during casting or by adding appropriate admixtures (superplasticizers, etc.).

6. Alkali-activated concrete casting technologies

The casting of wet alkali-activated concrete can be done in two ways, *in-situ* at the construction site, or it can be *pre-casted* in specific shaped formworks such as bricks, beams or columns that can then be transported on-site [135].
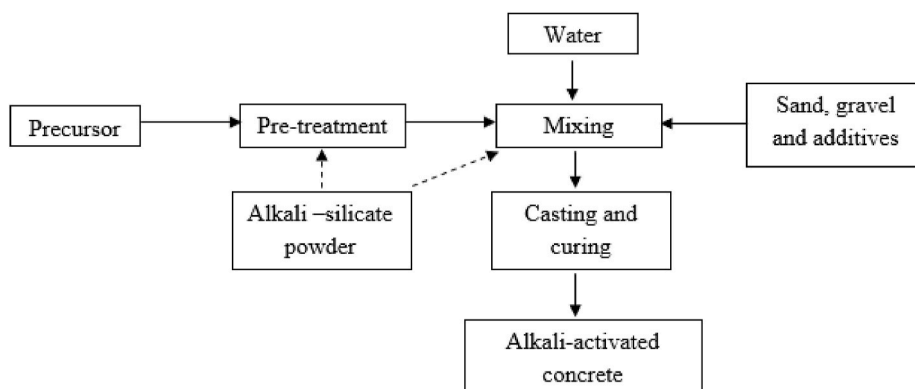

Fig. 11. Schematic of a one-part alkali-activated concrete production method.

6.1. In-situ casting

In-situ casting consists of placing the fresh alkali-activated concrete on-site in its permanent shape and location. Here, the material is transported by using pipes directly from the mixer and poured into formworks. For structural application, the alkali-activated concrete is reinforced with a mesh of steel bars or rebar to enhance its flexural properties. It has to be noticed that fresh alkali-activated concrete is not a liquid but a slurry so it should be carefully placed and handled to avoid segregation. It should not be placed more rapidly that it can spread, and it should be as near as possible from its final position [136]. Once the fresh concrete has been placed in the formworks, vibrators are used to compact the material and remove any trapped air bubbles [136]. This is mainly to prevent pores or voids that will overtime lead to loss of strength and induce corrosion, efflorescence, alkali-aggregate reactions, etc.

Thereafter, the wet alkali-activated concrete should be protected during the hardening and curing period to avoid moisture loss. This procedure is the same for Portland concrete's in-situ casting. Fig. 12 shows an in-situ casting, where blast furnace slag alkali-activated concrete is being poured in place and in between reinforced steel bars [137]. This cast-in-place concrete hardens at ambient conditions and should reach the determined compressive strengths after 28 days, both in Portland and AAMs systems.

Moreover, with the development of new technologies, there is also the possibility of casting AAMs on-site by a 3D extrusion printing technique, which consists in extruding the cementitious material from a nozzle to print the structure layer by layer (Fig. 13) [138–141]. The use of a formwork to provide lateral confinement is eliminated in 3D extrusion printing, and all the forces imposed on fresh structure need to be carried out by the material itself.

Ideally, in-situ casting would require a one-part mix, where the activator stays in the dry powdered shape. This would avoid transporting viscous solutions and corrosive hazards for large scale alkali-activated concrete manufacturing. However, in-situ casting has the disadvantage that there is not a controlled curing environment, and this can greatly affect the quality and durability of the binders especially when exposed to extreme conditions [142].

6.2. Pre-casting

Pre-casting consists of shaping the concrete into the formwork, and once it has hardened is transported on-site. The advantage of pre-cast manufacturing is that it has better quality control, improved environmental performance, improved site safety, and reduction of labour demand. Consequently, the quality and durability of alkali-activated concrete can be higher than casting in-situ and either one-part or two-part mixes can be used. In addition, several research findings calculate that pre-cast of alkali-activated concrete reduces the carbon emissions when compared to in-situ casting [143].

However, the specimens might have shorter time limitations to be fabricated when compared to in-situ. Thus, different accelerated curing methods have been applied in order to speed the alkali-activation and achieve high early strengths. These accelerated methods use elevated temperatures between 40 and 120 °C over the first hours after concrete casting [144–146]. Among those methods, the most common are steam curing and oven curing. Steam curing consists of placing the concrete formworks in a vaporous environment at temperatures around 60-110 °C [146]. This method has been seen to develop 70% of the ultimate compressive strength in 1 day, and the optimum operating temperature was found to be 80 °C [147]. Another fast strength development method is heat/oven curing, which consists of placing the samples in an oven at temperatures between 40 and 85 °C [146]. However, these techniques have the drawback that they take a few days and use convection-conduction methods to heat up a chamber, leading to a considerable energy consumption [124]. In this way, other efficient approaches have been studied, such as hot-pressing. Hot-press consists of subjecting the casted materials to a heat treatment at a fixed temperature ($\sim 350°C$) together with pressure for a short time. This causes a dense matrix, in which the extra liquid (mostly water) is forced out and the reaction is accelerated due to the increased temperature [148]. As a result of the hot pressing, the conversion of solid particles to a gel is accelerated by the formation of dissolved species that cross-link to form oligomers, which in turn produce AAMs. Hot pressing is an efficient method for curing AAMs, facilitating the production of high-strength binders in an extremely short time [145,148].

Fig. 12. Casting of an alkali-activated blast furnace slag concrete floor in Lohja, Finland, in 2020 [137].
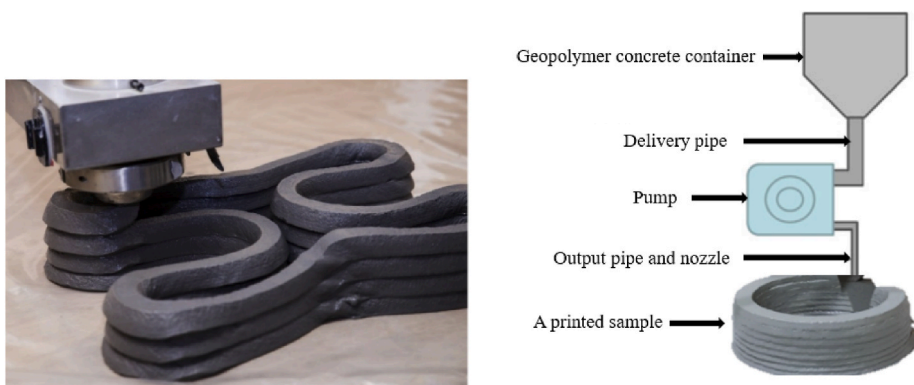
Fig. 13. Casting AAMs in-situ by 3D extrusion printing technique and a schematic of the concrete delivery system [138–141].

Fig. 14 shows an example of precast alkali-activated beams that can then be transported to the site [149].

The pre-cast can also be done by 3D printing technologies with a powder printing technique (Fig. 15a, and b [150]). The alkali-activated powder is prepared by mixing anhydrous Na_2SiO_3, slag and sand. Thereafter, the binder is an aqueous solvent (Zb®63, Z-Corp, US). This method is mainly used for small-scale building components such as panels, permanent formworks, and light-weight structural elements that can then be assembled on-site [150–152].

7. Alkali-activated cement and concrete market

The Pantheon and the Colosseum are monuments built by the Romans almost 2000 years ago. For their construction, they used a material with similar chemistry to alkali-activated cements. As aluminosilicate source, they used volcanic ashes from Pozzuoli (Italy), named pozzolans, and as alkali source they were using lime. They were then reacting it with water at ambient temperature [2].

The first modern combination of an alkaline reagent and an aluminosilicate to form cement, was in the 1900s when Kühl reported a patent using GGBFS and alkaline activators [153]. Later in 1940, Purdon published an article on slag cements and in 1950s they were commercialized in Belgium, including the construction of several buildings [6].

During the mid-1950s, with the shortage of OPC derived from the II-World War, Glukhovsky developed blast furnace slag cements and they were used in several buildings of the Soviet Union [154]. He termed those materials as "soil cements".

Research continued the following decades and in 1970′s Davidovits proposed that in addition to slag, natural minerals such as clays could also be used as aluminosilicate precursor [155]. At that time, he was investigating fire-resistant construction materials, as many buildings had to be torn down for having poor fire resistance. Therefore, to describe those fire-resistant inorganic resins, Davidovits coined the term "geopolymer" [155].

It wasn't until early 1990s that Wastiels first described fly-ash based geopolymers and an alkali activated GGBFS-fly ash binder [156]. Ongoing investigations and testing of alkali-activated cements have dramatically increased since then, both in the industry and in the academia. Most advances on these last decades have been leaded by Jannie Van Deventer, John Provis and Angel Palomo. The knowledge acquired has enabled an understanding on the manufacturing and performance of the alkali-activated cements. Australian universities and companies such as Zeobond, Wagners, CEMEX or Hoffmann have been active drivers on their developments and commercialization [2,18,28].

Fig. 14. Precast fly ash and slag alkali-activated floor beams used in the GCI building in Australia [149].
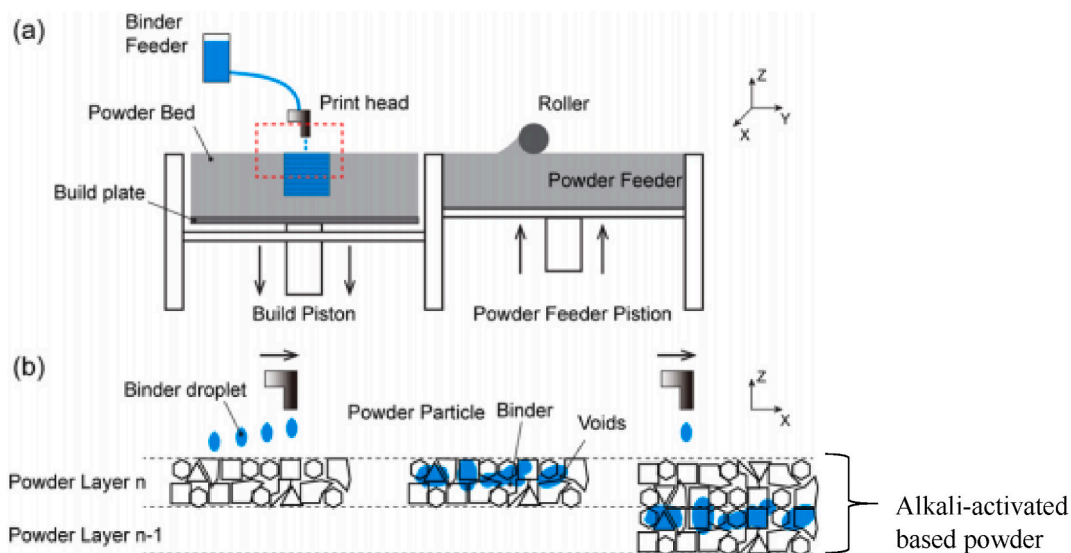
Fig. 15. Schematic illustrations of powder 3D printing process: (a) 3D powder inkjet printing system, (b) Enlargement of the area in the red rectangle: powder/binder interaction between adjacent layers [150]. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

7.1. Status of commercialized products

Most alkali-activated cement produced nowadays is primarily used in a niche market, for acid and fire protection. However, there is a growing number of companies (some examples shown in Table 3) that commercialize alkali-activated materials in high-performance applications, including the construction of buildings.

Table 3 shows that although some companies offer one-part mixes (Geopolymer Solutions, Hoffmann or Renca) for in-situ applications, most of them are commercializing two-part mixes. This is because even though one-part mix avoids handling alkali solutions, their properties and alkali dissolution mechanisms are still not well understood. In contrast, two-part mixes have been more broadly investigated, and they have been found to be suitable for both pre-cast and in-situ applications when adequate training and safety precautions are in place. Moreover, they have led to better compressive strength, workability, setting time as well as lower shrinkage when compared to one-part mixes [2,167]. In addition, the companies tend to have a preference to use industrial wastes such as GGBFS or coal fly ash, rather than metakaolin or other calcined clays due to workability-related issues and high-water demand [168]. Further investigations on natural resources and other waste sources are needed as there is a need to look beyond coal fly ash and GGBFS, whose availability is in decline [67,168].

7.2. CO_2 and cost assessment

Environmental and economic assessments are required when designing a new production process. They provide indicators for assessing the viability and the implementation of the new technologies. For the estimation of environmental impacts, life-cycle assessment (LCA) analysis is generally carried out, whereas profitability and cost-effective analysis are the common economic indicators.

LCA considers all the possible environmental impacts of the product, from the raw materials to final disposal. In the literature, most AAM environmental assessments [169,170] are focusing on the greenhouse gases (GHG) emissions and the global warming potential

Table 3
Some companies manufacturing alkali-activated concrete in 2023 [157–166].

Company	Location	Aluminosilicates	Alkali-activation type	Applications
Zeobond	Australia	GGBFS, Coal Fly ash	Two-part mix	Pre-mixed and pre-cast (E-crete)
Wagners	Australia	GGBFS, Coal Fly ash	Two-part mix	Pre-mixed and precast (Earth Friendly Concrete)
Geopolymer solutions	USA	GGBS, Fly ash and Metakaolin	Two-part and one-part mix	Powder and pre-cast (CFC)
CEMEX	Mexico	GGBS, Fly ash and Metakaolin	Two-part mix	Ready-mix concrete (Vertua)
ClockSpring| NRI	USA	GGBS, Metakaolin and Fly ash	Two-part mix	Mortars and grouts (GeoSpray)
Nu-Rock	Australia	Coal Fly ash	Two-part mix	Pre-cast
Hoffmann	France	GGBFS	Two-part and one-part mix	Powder, pre-cast (H–P2A)
Ceratech	USA	Coal Fly ash	Two-part mix	Pre-mixed and precast
Murray & Roberts	South Africa	Coal fly ash, Slag	Two-part mix	Pre-mixed
Renca	Russia	GGBFS, Coal Fly ash	Two-part and one-part mix	Powder and pre-cast

(GWP). However, there are some publications that reveal the importance of other impact categories when considering a complete LCA [171]. Those are abiotic depletion (ADP), ozone layer depletion (ODP), human toxicity (HTP), freshwater ecotoxicity (FAETP), marine ecotoxicity (MAETP), terrestrial ecotoxicity (TAETP), photochemical oxidation (POCP), acidification (AP), and eutrophication potential (EP). From the perspective of the greenhouse gas (GHG) emissions, and specifically CO_2 emissions, alkali-activated cement production has demonstrated to have the potential to reduce the CO_2 emissions to approximately an average of 50% [18,172]. However, it can greatly vary from almost no reduction to up to 90% CO_2 emission reduction [17,18]. The reduction depends on the raw materials, their composition and source location, as well as the processing applied for alkali-activation.

The differences in CO_2 emissions depending on the type of processing used, is directly linked with the aluminosilicate and alkali source and the energy efficiency of the technology. When re-using industrial wastes such as fly ash and slags in alkali-activated cements, their LCA generally leads to a lower environmental impact [75]. This is mainly because most LCA studies tend to allocate fly ash and GGBFS as wastes, and thus their environmental impact is limited to the pre-treatment and transportation [75,173]. However, coal-fired power generation is the largest emitter industry contributing to 30% of all energy-related carbon dioxide emissions (850 kg CO_2/MWh) [174], and steel has a contribution of 1.85 tonnes of CO_2 for every tonne of steel produced [175]. This means that the industries from which fly ash and GGBFS originate are high- CO_2 emitters, and by reusing their by-products in alkali-activated cements there is a positive re-valorisation of those wastes, as instead they would be landfilled.

When using natural raw clays as aluminosilicate precursors, the different calciner technologies must be considered. Table 4 presents the estimated CO_2 emissions to produce 1 tonne of calcined clay using two different technologies: rotary and flash calciner [83]. The flash calciner presents lower CO_2 emissions than a rotary calciner. Thus, flash calciner is generally the technology preferred for greenfield projects within clay calcination. Nevertheless, it must be noted that retrofitting an old cement kiln is faster than building a new flash calciner [83]. This could be an alternative path for low-income level countries, which cannot afford the capital costs of expanding with new capital investments but have an expanding cement demand to meet [176]. For considering the refurbishment, the cement plants need to have an old, outdated clinker kiln, which is set into operation in times of a peak cement demand, or when there are gaps in production.

Another factor that needs to be considered is the CO_2 emissions that derive from the alkaline activators. Several studies [177,178], claim that the largest environmental footprint of alkali-activated cement derives from sodium silicate, one of the most used alkali reagents in AAMs. If sodium silicate is produced using the Solvay process, the environmental impact is even more pronounced, since CO_2 is a by-product. Figs. 16 and 17 present an estimation of the energy demand (a) and the CO_2 emissions (b) derived from the production of 1 m³ of alkali-activated concrete using flash metakaolin and industrial wastes (fly ash or GGBS), respectively.

From Figs. 16 and 17, it can be seen that sodium silicate is not only the largest contributor to CO_2 (Fig. 16b and 17b) but also the one with highest energy demand (Fig. 16a and 17a), when compared to the rest of raw materials in an alkali-activated concrete [179]. In addition, the use of sodium silicate has been found not only to have high carbon emissions, but also to increase other environmental impacts such as ozone layer depletion and several toxicities (human toxicity, freshwater toxicity, or marine water toxicity) to levels higher than Portland cement does [177,182].

In contrast, Fig. 18 illustrates the estimated energy demand (Fig. 18a) and the carbon emissions (Fig. 18b) that derive from the production of 1 m³ of Portland concrete. Comparing Figs. 16 and 17 with Fig. 18, it is seen that alkali-activated metakaolin concrete can lower the energy demand by 43% and reduce the CO_2 emissions by 68% when compared to Portland concrete. On the other hand, when comparing alkali-activated industrial wastes concrete with Portland concrete, they can further reduce the energy demand by 54% and the CO_2 emissions by 75%.

The difference of carbon emissions and energy demand between the clay calcination process and the cement production, lies in the fact that the calcination of clays occurs at lower temperatures (700-900 °C) than the thermal process for forming clinker minerals in Portland cement ($\sim$ 1450°C). Moreover, in the calcination of clays there is no reaction that releases CO_2, while in Portland cement, 50% of the emissions is derived from the calcination of limestone (CaCO3→ CaO + CO_2).

From an economic perspective, it has also been observed that the cost of the alkali activators can greatly affect the cost of the alkali-activated concrete [183]. Table 5 shows the cost in euros (€) to produce 1 tonne of metakaolin, coal fly ash and GGBFS alkali-activated concrete with the mix proportions taken from Figs. 16 and 17. The raw materials prices represent approximate bulk chemical price levels.

It must be considered that the raw materials cost can greatly vary depending on the quality grade, the composition, the country produced, the availability or the specific supplier. Nevertheless, from Tables 5 and it can be observed that the sodium silicate solution is the most expensive material, followed by sodium hydroxide solution. Furthermore, pure metakaolin clay also seems to highly increase the alkali-activated concrete's cost. Hence, there are studies [177] investigating the performance of using of low-grade clays that could potentially reduce the total cost. Thereafter, Table 6 shows the cost in euros (€) to produce 1 tonne of Portland concrete (mix proportions taken from Fig. 18).

Comparing Tables 5 and 6, it can be seen that coal fly ash and GGBFS alkali-activated concrete are 1.9 and 2.15 times more expensive than Portland concrete, respectively, whereas metakaolin alkali-activated concrete almost triplicates the price.

Recent studies, aim to decrease the alkali-activated materials cost by using available silica-rich biomass ashes or industrial wastes without compromising the quality [188]. A water glass solution can be synthesized from silica-rich sources and its used instead of the commercial sodium silicate solution. Table 7 shows the hypothetical cost of producing a low-grade clay alkali-activated concrete, in which, sodium silicate is totally replaced by rice husk ash (RHA) water glass.

Replacing metakaolin with a low-grade calcined clay and sodium silicate with purified rice husk ash, results that the price for a clay alkali-activated concrete could drop from 134.68 €/t to 36.69 €/t, converting it into a more attractive alternative to Portland concrete. Nevertheless, this approach still needs to be further investigated to reach optimal mix designs that will lead to strong and durable

Table 4
CO_2 emissions derived from the kaolin calcination in a rotary and flash calciner [83].

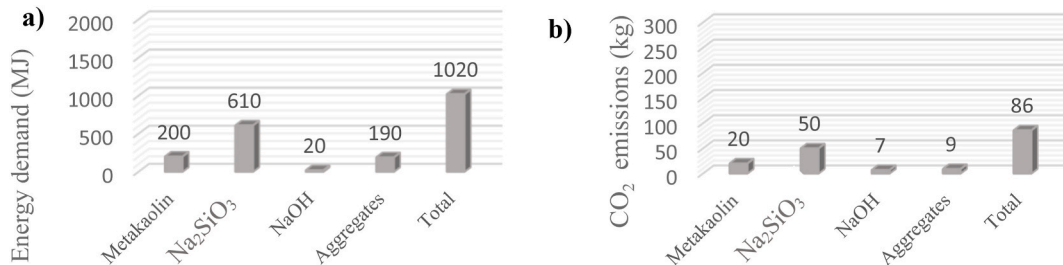| Technology	Emissions (kg CO_2/tonne of metakaolin)
Rotary calciner	393
Flash calciner	196

Fig. 16. Energy demand (a) and carbon dioxide emissions (b) in the production of 1 m^3 of a flash metakaolin alkali-activated concrete with a compression resistance of at least 40 MPa (2300 kg/m^3). Mix design used for calculations: 353 kg/m^3 metakaolin, 156 kg/m^3 alkaline reagents (NaOH + Na_2SiO_3), 304 kg/m^3 water, 611 kg/m^3 sand and 876 kg/m^3 gravel (adapted from Refs. [159,161,162]).
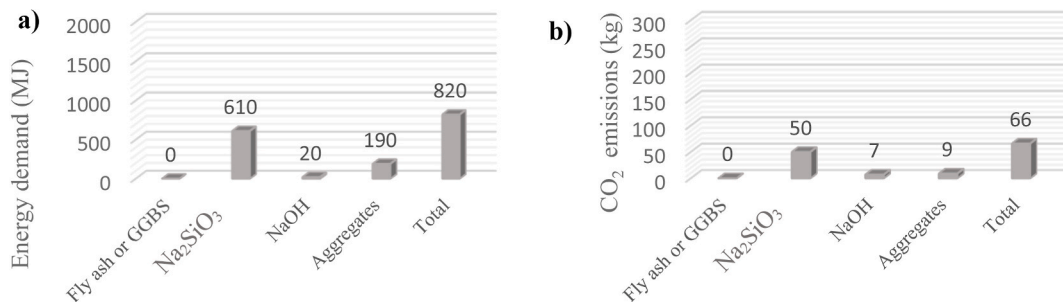

Fig. 17. Energy demand (a) and carbon dioxide emissions (b) in the production of 1 m^3 of an industrial waste alkali-activated concrete (fly ash/GGBS) with a compression resistance of at least 40 MPa (2300 kg/m^3). Mix design used for calculations: 353 kg/m^3 fly ash or GGBS, 156 kg/m^3 alkaline reagents (NaOH + Na_2SiO_3), 304 kg/m^3 water, 611 kg/m^3 sand and 876 kg/m^3 gravel (adapted from Refs. [179–181]).
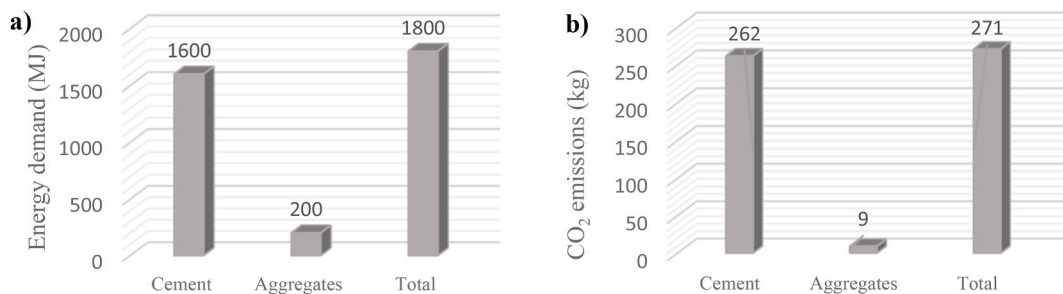

Fig. 18. Energy demand (a) and carbon dioxide emissions (b) in the production of 1 m^3 of a Portland cement concrete of 40 MPa strength class (2400 kg/m^3). Mix design used for calculations: 424 kg/m^3 cement, 174 kg/m^3 water, 4 kg/m^3 superplasticizer, 730 kg/m^3 sand and 1048 kg/m^3 gravel (adapted from Refs. [180,181]).

materials.

Other studies show that AAM can be profitable if a high aluminosilicate/activator ratio is used, implying a lower amount of alkali activators [191]. Moreover, the use of solid sodium silicate (one-part mixes) instead of a solution could also decrease the cost of the raw materials, as less volume of activator would be needed, and it would result cheaper to transport.

Table 5

Materials cost estimation for producing 1 tonne of metakaolin, coal fly ash and GGBFS alkali-activated concrete [32,184,185].

Material	Price (€/t material)	Cost (€/t concrete)
Metakaolin	300	45.9
Coal fly ash	39	5.9
GGBFS	120	18.36
Sodium hydroxide solution (10 M)	400	20
Sodium silicate solution ($SiO_2/Na_2O = 3.5$)	450	67.5
Aggregates	2	1.28

Estimated cost for 1 tonne of metakaolin alkali-activated concrete (€/t): **134.68**.
Estimated cost for 1 tonne of coal fly ash alkali-activated concrete (€/t): **94.68**.
Estimated cost for 1 tonne of GGBS alkali-activated concrete (€/t): **107.14**.

Table 6

Materials cost estimation for producing 1 tonne of Portland cement concrete [186,187].

Material	Price (€/t material)	Cost (€/t concrete)
Cement	104.9	48.79
Water	0.04	0.01
Aggregates	2	1.28

Estimated cost for 1 tonne of Portland concrete (€/t): **50.08**.

Table 7

Materials cost for producing a clay alkali-activated cement when replacing metakaolin clay by a low-grade calcined clay and sodium silicate by rice husk ash water glass [32,187,189,190].

Material	Price (€/t material)	Cost (€/t concrete)
Calcined clay	35	5.35
Sodium hydroxide solution (10 M)	400	20
Purified rice husk ash (RHA)	67	10.05
Water	0.04	0.01
Aggregates	2	1.28

Estimated cost for 1 m^3 of calcined clay alkali-activated concrete (€/t): **36.69**.

7.3. Future developments for standardization and commercialization

More alkali-activated cement and geopolymer companies are starting to enter the construction market, and an overall acceptance in the construction industry is starting to grow. Nevertheless, more efforts are to be made to overcome the limitations, standardize and commercialize alkali-activated materials worldwide.

A main obstacle to widespread alkali-activated cement is the issue of compliance with existing regulatory standards which have been written for the use of Portland cements. The world's cement production is mainly regulated by two big standards EN 197 (European Union standards) [192] and ASTM C150, C595 and C1157 (USA standards) [193]. Asia cement generally follows the European regulations and Oceania follows the USA standards. Those regulations contain prescriptive standards in which allowable compositions of cement are specified. In some countries, the prescriptive standards have constraints such as "minimum clinker content" which reduces the possibility of using novel-binders [194]. In this way, it is important to develop new standards that can allow clinker-free binders use. A satisfactory example of regulatory progress and commercial adoption of alkali-activated materials is Australia, a strong collaboration between academia, industry and authorities has led to many construction projects (private and public) using alkali-activated concrete (Wellcamp Airport, GCI building, VicRoads), and an Appendix to their Cement Standard has been included (AS3972). Australia has become a great reference for the RILEM committee and other public and private institutions around the world.

RILEM Technical Committee is an international union of construction and materials experts that is developing a framework to adopt alkali-activated materials. This framework is a performance-based approach instead of prescriptive [195]. For designing this approach, three aspects are considered. The first one is to fully understand the mechanical strength behavior of the alkali-activated concrete, so how the conditioning and raw materials affect the desired final strength. The second one is regarding sample preparation and conditioning. Different raw material mix designs will require different regulations. This means that the manufacturer should specify the mix designs and performance results for each product and report the outcome to show the compliance with the performance-based standards. The last key aspect is to prove that desired properties will be maintained over time, so what is known as the durability of concrete. However, this is done by accelerated tests and they only give an estimation of the expected performance. To verify the durability in the real world it can take approximately 20–30 years [196]. Therefore, at the moment durability is another limiting factor to widespread their use, as they don't have the support of decades of in-service testing and durability data to prove its long-term stability. This limitation has been mitigated by some cement companies that have opted to apply alkali-activated materials in low-risk applications first and after they have shown real world durability, high-risk applications have been considered [194].

The use of alkali reagent is another barrier towards commercialization. If the alkali reagent content was reduced, the interest of alkali-activated materials in the industry would probably increase. The amount can be greatly reduced or even eliminated if the correct glass can be selectively synthesized [197,198]. This can be done by blending different aluminosilicates that are rich in alkalis or by adding alkali admixtures. Moreover, a workable one-part mix ("just add water") must be developed to achieve a widespread market penetration, as they would largely simplify the logistics and no special training would be required for their handling [194].

While RILEM is working on a global standard long-term solution, there are short-term actions that can boost the use of alkali-activated cements worldwide. Those are driven by customers, construction consulting engineers and architects. Many customers are looking for innovative solutions that can reduce CO_2 emissions, being able to increase the demand on green products. The next most influential driver are the construction consulting engineers, who advise companies about new low CO_2 -technologies. As final short-term drivers, architects are determinant on the broader use of alkali-activated materials. They can promote the use of alkali-activated concrete to constructors as an innovative environmentally friendly solution. Market research shows that the key performance indicators for major projects now frequently include environmental innovations, meaning that a low CO_2 concrete is in demand even before a carbon trading system is in place. As soon as a price is set on CO_2 emissions and carbon taxes are implemented, low carbon products will have a price advantage and alkali-activated cement could be highly demanded [28]. Therefore, despite the market barriers that can be present today, the alkali-activated materials production technology for large-scale manufacturing shall be ready for when the standards and the customers are ready for clinker-free and low-CO_2 alternative construction materials.

8. Conclusions

There is a growing demand of low-CO_2 concrete and thus the production of alkali-activated concrete is gaining attention in the construction industry. However, the main challenge for upscaling alkali-activated concrete is that there is no unique straightforward flowsheet as there is for the production of Portland cement, bringing higher degree of confusion to the concrete industry who does not know what kind of processes or technologies are needed to produce geopolymers or alkali-activated concrete. This informative review has related common concrete technologies with the production of alkali-activated concrete and hereby presents the main conclusions:

- Selecting adequate aluminosilicate pre-treatment technology is essential for developing strong and durable alkali-activated materials. Accordingly, if a greenfield alkali-activated cement or concrete plant is to be built, a vertical roller mill that integrates drying and grinding in one unit (e.g.dryer crusher) would probably be the recommended choice to mechanically activate precursors such as coal fly ash or GGBFS. Thereafter, for clays being thermally treated, a flash calciner would generally be the preferred technology. Nonetheless, if existing cement plants would like to adapt their technology, retrofitting previous equipment could also be done.
- Regarding the alkaline activators, they can be added to the concrete mixers in either liquid (two-parts) or solid form (one-part). Two-part mixes are the most common practice since its chemistry and properties have been widely studied. However, large volumes of alkaline solutions are dangerous to handle at a construction site. Hence, one-part mixes with solid alkalis would be a better option and further investigations should be conducted to fully understand the alkali dissolution mechanisms and their long-term properties.
- Mixing and casting of alkali-activated concrete has the same considerations as for Portland concrete. A homogenous blend after mixing is necessary to ensure an adequate strength development and high extent of reaction. During casting, it is important to compact the wet concrete, to prevent pores or air voids that could lead to a loss of strength and affect the durability over time. As for Portland cement, with the development of new technologies, AAMs are also gaining attention in different 3D printing techniques such as extrusion and powder printing.
- Australian universities and some companies such as Zeobond, Wagners, CEMEX or Hoffman have been recent active drivers on the developments and commercialization of alkali-activated cements.
- From the environmental perspective, alkali-activated materials can lower the CO_2 emissions associated with Portland cement by 68% when using calcined clays and 75% when using industrial by-products (coal fly ash and GGBFS). However, it has also been observed that sodium silicate is responsible for most of the CO_2 emissions and energy demand in alkali-activated materials production.
- The cost estimation shows that alkali-activated concrete's cost depends on the type of raw materials, source and quality degree. Alkali-activated coal fly ash and GGBFS concrete are 1.9 and 2.15 more expensive than Portland concrete, respectively whereas alkali-activated metakaolin concrete almost triplicates the price. In any case, the most expensive raw material appears to be the alkali activator sodium silicate. Thus, a possible alternative would be to replace sodium silicate by alkali and silicate-rich biomass ashes, as they would decrease not only the cost but also the CO_2 emissions. Further use of low-grade clays instead of metakaolin could also prominently lower the price of alkali-activated calcined clay concrete.

Author contribution statement

All authors listed have significantly contributed to the development and the writing of this article.

Data availability statement

Data included in article/supp. material/referenced in article.

Declaration of interests

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.

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