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Review Article

Alkali-Activated Slag Cement and Concrete: A Review

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ABSTRACT

Recently, Alkali-Activated Concrete (AAC) as an appropriate alternative to Portland Cement Concrete (PCC) has drawn considerable attention owing to its acceptable properties and less environmental impacts. The current study presents a review of previous researches on Alkali-Activated Slag Cement Concrete (AASCC). The following issues are also covered in this study: an introduction to AASCC and its constituents, environmental impacts, hydration reaction, hydration products, mechanical properties, high temperature resistance, attack resistance of acids and sulfates, water absorption, shrinkage, and its challenges in application. The results from different studies revealed a research gap to be filled. To be specific, several different parameters affect AASCC properties; however, given that its properties are still partially unexplored, more researches are needed to find out its best engineering properties.



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1. INTRODUCTION

Nowadays, Portland Cement (PC) is one of the most extensively used materials in construction. The usual consumption rate of PC is approximately one ton per person a year [1], and the demands for it are still increasing every year. Currently, the amount of PC production is more than three billion tons per year which, as predicted, may reach six billion tons in the next 40 years [2]. Figure 1 shows the increasing trend of PC production until 2014 [3].

PC production has some environmental impacts such as greenhouse gas emissions and air pollution. In addition, this industry is highly energy-consuming. In

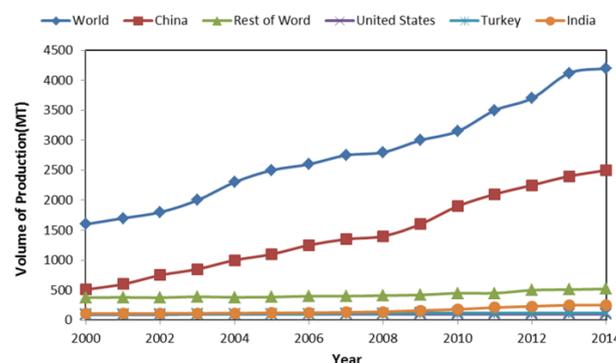


Figure 1. PC production per year for countries [3]

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this regard, given the increasing demand for PC, it is recommended that PC be substituted with another type of cement, especially Alkali-Activated Slag Cement (AASC) owing to its superior properties [4].

Although AASCC has been frequently explored in many studies in recent years, it is an old technology. In 1908, Kuhl confirmed the stabilization of the blast furnace slag powder with sodium hydroxide. In 1940, Pordon published more extensive researches on alkali-activated clinker-free cement. Two decades later, Glukhovskiy investigated the alkaline activation of different organic and inorganic aluminosilicates [5-7]. Industrial production of AASC first began in Ukraine in 1960-1964. It was obtained through activating the slag, a by-product in the production of iron, using alkaline materials. In this process, aluminosilicate monomers dissolve in the slag structure and produce a cementitious and adhesive composition. Use of this type of cement is economically affordable. In addition, compared to PC, it is characterized by more acceptable properties [8].

2. THE ENVIRONMENTAL IMPRESSION AASC

CO₂ emission from the PC industry is an important environmental issue to be concerned about. As shown in Figure 2, about 3 % of the world's greenhouse gases are generated by cement industries.

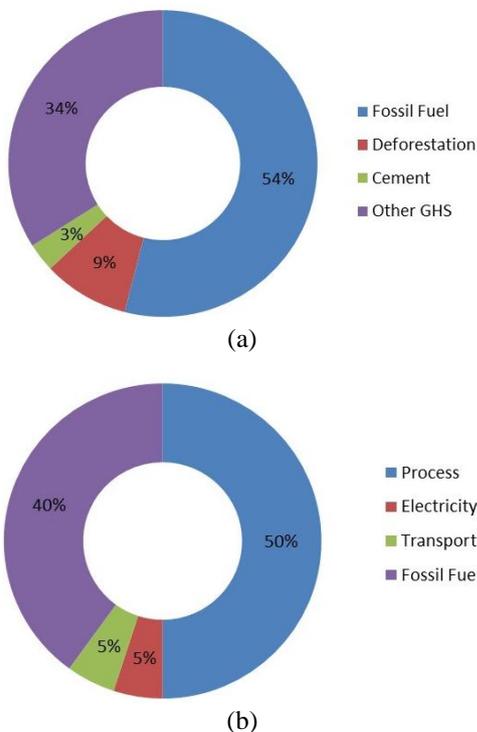


Figure 2. (a) Different sources of CO₂ gas emissions and (b) Contribution of pollutions of different parts of the cement production process

According to the reports, the cement industry alone can generate 7 % of CO₂ in the world [9]. The fabrication of one ton of PC releases approximately 0.7 to 1 ton of CO₂ gas into the atmosphere [10,11]. Between 1930 and 2013, cumulatively, cement industries contributed to about 43 % of CO₂ emissions [12].

The main advantage of AASC, compared to PC, is its notable effect on reducing the environmental pollutants. To be specific, the CO₂ emission in the AASC production is 50-80 % lower than that in the PC production [13,14], hence it can be regarded as an operative technology that can considerably reduce the CO₂ emissions. The key material in the AASC production is slag, which is actually a by-product of pig iron; therefore, the production of this type of cement also reduces the waste materials. According to the reports, the global warming potential in AASC production is 70 % lower than that in PC production [15].

3. INGREDIENTS OF AASC

3.1. Slag

In the production process of pig iron in the blast furnace, limestone reacts with Al₂O₃ and SiO₂ rich components in iron ore and coke and produces molten slag. If the molten slag cools rapidly below 800 °C, it becomes a suitable glass material to be used in the cement industry. Powdered slag is a material characterized by hydraulic properties only in the presence of an appropriate alkaline activator. AAS cement is a delayed hydraulic material. However, other hydraulic materials have this property when mixed with water [16]. Blast furnace slag with a C/S ratio between 0.2 to 2 and A/S ratio between 0.1 to 0.6 are commonly used in AASC production. A, S, and C are abbreviations of Al₂O₃, SiO₂, and CaO, respectively.

3.2. Alkaline Activators

Once the slag powder is mixed with water, the hydrated thin layers containing Ca⁺² and SiO₂ are formed on the external surface of the particles. At this point, the amount of pH reaches 10. This hydrated film on the surface of slag particles interrupts the hydration reaction; therefore, a suitable activator should be added to the mixture to have a continuous reaction. Alkaline activators increase the pH value of the liquid phase and prevent the formation of a protecting layer on the slag surface, thus preserving the solubility of the slag glassy phase. Alkaline activators break the bonds in the slag glass phase structure and increase the dissolution rate of the ionic components [17].

Sodium silicate in either powder or liquid forms, sodium hydroxide, and sodium carbonate are usually used as the activator. Sodium sulfate, potassium hydroxide, gypsum, sodium aluminate, and hydrated lime are also used in different studies. Activators are

added in small amounts of about 2-10 wt. % of the slag. Since the slag compositions differ depending on the raw materials and production process, the most appropriate activator should be selected. The type and amount of activators are both essential factors that affect the properties of AASC. For example, the results revealed that the pastes prepared by Na_2SiO_3 activator had shorter setting time than Na_2CO_3 and NaOH activators [18-22].

3.3. Chemical Admixtures

There are several types of superplasticizers including lignosulfonates, naphthalene, vinyl copolymer, melamine, and polycarboxylate derivatives. The chemical admixtures are absorbed into the cement particles. As a result, they create an electrostatic repulsion among the particles, thus preventing their agglomeration [23].

Shrinkage-reducing chemical admixtures such as polypropylene glycol derivatives reduce the surface tension of water in concrete pores. Consequently, the capillary tension in the pore structure and shrinkage decreases during the drying process. Chemical admixtures such as malic acid and sodium chloride are also used as retardants. The amount of admixtures in different types of cements is less than 5 wt. % [23,24].

4. MIMING PROTOCOL OF AASC

Several different mixing protocols were proposed in the literatures. Generally, slag is ground with gypsum. The amount of gypsum was reported to be 2-3 wt. % of the slag. The specific surface area of the slag was suggested to be between 250 and 700 m^2/kg . The water-to-binder ratio (W/b) in AASC is usually set between 0.3 and 0.5 [25]. Activators are either directly mixed with the slag or first dissolved in water and then mixed with it. The corrosive properties of the activators and high pH of the material should be considered in the mix of AASC. The slag-to-aggregate ratio is usually either 1:2 or 1:3 [19-25]. The admixtures are then directly added during mixing [23]. The common aggregates in PC concrete are used to produce AASCC. It should be noted that AASC is generally formed by casting.

5. HYDRATION REACTION MECHANISM AND PRODUCTS

The slag composition, activator type, and pH are the main parameters that control the hydration products of AASC.

Wang et al. [26] reported that the main hydration product was C-S-H with low C/S ratio and different crystallinity. In addition, the XRD analysis conducted by Puertas et al. confirmed the existence of hydrotalcite ($\text{Mg}_6\text{Al}_2\text{CO}_3(\text{OH})_{16}\cdot 4\text{H}_2\text{O}$), calcite (CaCO_3), and CSH as

hydration products in the slag activated with NaOH [27].

Hydration of slag is a multipart process includes initial degradation of slag and densification of hydration products. The hydration of slag initiate by ion exchange of H^+ in water for Ca^{+2} or Na^+ . Al-O-Si bonds hydrolyze and then the depolymerized slag network breakdown. So Si and Al release as $\text{Al}(\text{OH})_4^-$ and $(\text{H}_3\text{SiO}_4)^-$ and $(\text{H}_2\text{SiO}_4)^{2-}$ ions in water and finally precipitate on the surface of slag particles with increasing the concentration of them [28].

Krizan et al. found that the hydration reaction rate was correlated to the modulus of Na_2O and silica (M_s) in sodium silicate-activated slag. In summary, hydration reaction initiates with the degradation of Ca-O, Mg-O, Si-O-Si, Al-O-Al, and Al-O-Si bands in the slag and then, a Si-Al layer precipitates on the surface layer of the slag particles [29].

An investigation by Wang et al. confirmed that during the hydration of AAS, dissolution and precipitation were the predominate mechanisms in the initial stage of the reaction that continued with the solid-state mechanism. [26]. Mozgawa et al. concluded that Si^{+4} of the sorosilicate $[\text{Si}_2\text{O}_7]^{-6}$ units in the glassy phase were partially substituted by Al^{+3} during the AAS hydration. The ^{29}Si MAS-NMR spectra confirms this claim. According to the ^{27}Al MAS-NMR spectra, Al atoms can occupy tetrahedral and octahedral coordination in the hydration process determined by the activator type and factors of hydration process [30].

6. THE HEAT OF HYDRATION

The hydration heat in the AAS is usually about three times lower than that in the OPC [8,31], depending on type and dosage of the used activator. For example, Krizan et al. observed that the hydration heat of the AAS increased upon increasing the dose and modulus of sodium silicate [29]. It was also reported that using Lime and gypsum as the activator would increase the hydration heat more than that in the slag activated with lime only, mainly due to the formation of ettringite phase and its transformation to monosulfoaluminate phase. However, the hydration heat of OPC was higher than that of AAS [32]. As mentioned earlier, AASC hydration products have a lower Ca/Si ratio, hence they need less hydration energy than OPC [33].

7. PROPERTIES OF AASCC

7.1. Mechanical Properties of AASCC

The compressive strength of AASCC is its most significant mechanical property. Several parameters including the composition and specific surface area of the slag, type and amount of alkaline activator, water-to-slag ratio, and curing conditions of the concrete affect the

mechanical strength of AASC [34-42].

AASCC is characterized by high initial strength. The initial compressive strength of this concrete is 60 MPa, and its one-year strength can reach more than 100 MPa. The higher rate of hydration reactions at higher pHs, which is the cause of high strength in the first days of concrete preparation, and homogenous and dense interface between the paste and aggregate are the two vital factors that contribute to the high mechanical strength of AASCC [34].

AAS concrete is susceptible to curing conditions. Bakharev et al. [37] reported that the initial strength of the Room-Temperature (RT) cured samples are less than those cured at higher temperatures. However, the later strength of the RT cured samples is clearly higher than that of the others (Figure 3) resulting from the inhomogeneity of the matrix microstructure due to the higher hydration reaction rate than the diffusion rate at higher-temperature curing. In other words, the hydration products precipitated around the slag particles, make barriers to diffusion, and form an inhomogeneous microstructure [37].

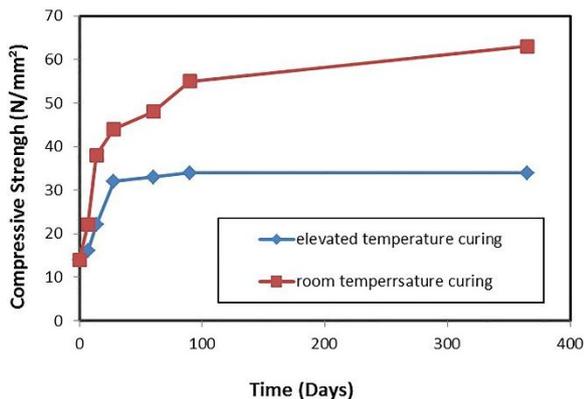


Figure 3. Compressive strength of AASC versus curing temperature. Lines are shown as guides for the eyes

Collins et al. [40] found that the compressive strength of AASC samples in a water bath increased up to 400 days, while the maximum strength of the sealed specimens was obtained in 91 days. In addition, the samples exposed to air were more susceptible to a decrease in their strength over time mainly due to the presence of microcracks in the matrix. Figure 4 shows the compressive strength changes in different curing conditions.

According to the literature, the effect of alkali activator on the strength of AASC has not been systematically investigated yet. Chi showed that the amount of Na_2O in the activator could affect the strength of AASC. In addition, the liquid sodium silicate was characterized by a higher initial strength than that of its powder counterpart [35,40].

In another research, replacing 20 % slag with the

recycled concrete fines improved the compressive strength of AAS concrete blocks up to about 13 % [43].

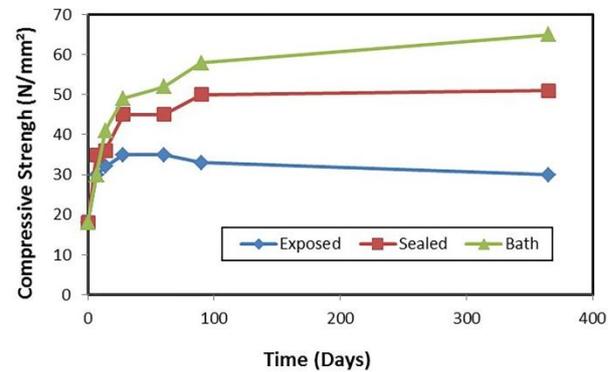


Figure 4. Compressive strength of AASC versus curing condition. Lines are shown as guides for the eyes

7.2. High-Temperature Resistance of AAC

Several physical and chemical mechanisms can decrease the strength of PC cement at high temperatures [44].

Variances in the coefficient of the thermal expansion between the aggregate and mortar cause an internal stress in the concrete and micro cracks. In addition, decomposition of calcium hydroxide ($\text{Ca}(\text{OH})_2$) in PC at temperatures above 400°C would decrease its strength. Moreover, scaling of OPCC at higher temperatures plays a key role in strength deterioration [45,46].

A number of studies have evaluated the performance of AASC at high temperature, considering the significance of this parameter in the concrete. The compressive strength of alkali-activated slag mortar at temperatures above 400°C is the same of that of PC mortar. In this case, dehydration of C-S-H gel is the main reason for strength reduction [47]. Approximately, 65-85 % loss in the strength of AAS mortars was reported at temperatures above 800°C [47-49].

Guerrieri et al. [50] observed that the decrease in the strength resulted from the differences in the thermal expansion coefficient of the aggregate and slag at high temperatures.

7.3. Chemical Properties of AASCC

7.3.1. Water Absorption and Chloride Diffusion

The permeability of concrete is assessed by water absorption which is lower in AASC than in PC thanks to the dense microstructure of AASC. Water absorption is directly associated to the chloride diffusion [51]. According to the findings, using porous recycled aggregates instead of natural aggregates in concrete would cause higher water absorption and chloride diffusion. Water absorption in AASCC also increases upon increasing the specific surface area of the recycled aggregates [52,53].

7.3.2. Sulphate and Acid Attack

Cement resistance against the sulfate and acid attacks is one of the significant aspects in the durability of different types of concrete that should be taken into consideration. Both properties of the pores and type of acid corrosion products affect the resistance of a concrete against acid corrosion.

As reported, AASCC exhibited higher resistance to sulphate attack than PC concrete after one-year exposure to 10 % sodium sulphate. Nevertheless, the strength decreased when the samples were exposed to 10 % magnesium sulfate in the same time period [51].

Numerous researches have been conducted on the resistance of AASC in acid environments. For instance, Mithum et al. [51] observed a considerable decrease in the mass of AASCC after 120 days of exposure to H_2SO_4 solution, which was attributed to a decrease in the concrete strength mainly due to the slag dissolution which created more paths for the acid to move along the AASC, hence further dissolution [53].

C. Shi et al. [54] also observed that the PC corrosion in acetic acid solution with pH = 5 after one year was much more than the AASC corrosion due to its more porous structure of the corrode layer. They concluded that acid resistance depended on hydration products and protective layer.

7.3.3. Carbon Diffusion

Carbonation of AAS concrete in CO_2 atmosphere or solution starts from its surface and gradually diffuses into the concrete. The carbonation rate is determined by the rate at which HCO_3^- and CO_2 diffuse into the concrete. This process follows Fick's law. In other words, the carbonation rate (increasing the depth of carbon with time) is proportional to the square root of time.

When the concrete is exposed to CO_2 , the C-S-H phase reacts to produce $CaCO_3$ and calcium-free C-S-H and aluminosilicate gel. Given that the amount of Ca in AASC is low, there are not enough $CaCO_3$ precipitations. In addition, the porosity of the substrate increases owing to the carbonate reaction. The entry of HCO_3^- and CO_2 into the interior increases and as a result, the reaction proceeds faster. Therefore, AASC is more susceptible to carbon diffusion than the ordinary PC [55].

7.4. Shrinkage of AASCC

Generally, AASCC is characterized by finer pores than OPCC, hence higher shrinkage of AASCC. According to the study conducted by Collins and Sanjayan [56], more meso pores in AASCC microstructure and higher capillary stress can cause higher shrinkage in alkali-activated cement pastes. Another research [32,57] confirmed this theory. However, the different composition of the reaction product (C-S-A-H and C-S-H) was the other cause of further shrinkage of AASCC [29].

Numerous studies have focused on reducing shrinkage

in the AASC. To this end, they employed several methods such as application of suitable admixtures [58], fibers [59,60], heat treatment [37], expanding admixture [61], and low-temperature preparation technology [62]. In general, the type and quantity of the activator, surface area of the slag, and curing condition are the factors affecting the shrinkage of AASC. T. Bakharev et al. [35] found that the highest shrinkage belonged to the sodium-silicate-activated sample.

According to the findings, the slag activated with NaOH and sodium silicate exhibited three to six times more shrinkage than that of PC [57]. Figure 5 illustrates the effect of different activators on the AASC drying shrinkage. As observed, the shrinkage attributed to sodium carbonate was less than/ similar to that of OPC mortar. Ye and Radlińska [63] made a comparison of the obtained results.

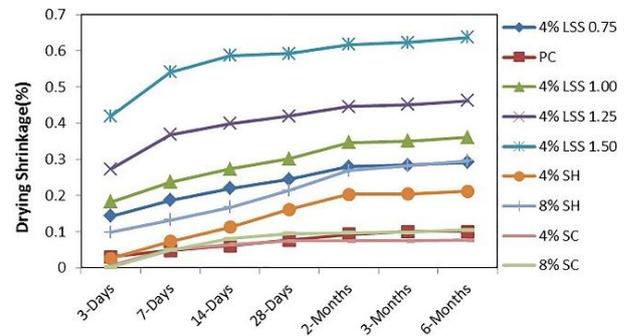


Figure 5. Effect of different activators on drying shrinkage of the slag

According to Figure 5, using 9 % Na_2SiO_3 and 10 % OPC simultaneously yields the desired shrinkage with OPC [64]. The slag activated with lime and gypsum exhibited lower shrinkage than that activated with lime only [32]. This dissimilarity is due to the difference in volume of smaller pore and nature of hydration products in the concrete.

8. CHALLENGES IN AASCC

Regardless of numerous advantages of AASC, its application is still quite challenging.

- **Slag Composition:**
The slag compositions differ depending on the raw materials and melting procedure. The variation in the slag compositions causes a different behavior of activation. In fact, one condition cannot be generalized to all slags. Therefore, it is suggested that a small-scale test be done for each slag to obtain the best activation mode.
- **Chemical Activators:**
The activators that are currently used for AASCC production are viscous, expensive, and corrosives.

In order to increase the application of this cement, safe and environmentally-friendly green activators characterized by the same mechanical properties of the chemical activators should be used. More researches on the efficiency of different green activators are also available.

- **Admixtures:**
To the best of the author's knowledge about PC, most of the admixtures are used in AASCC production. Given that AASC has different chemical and behavioral characteristics, development of admixtures that are specifically suitable for alkali activated slag gains significance. Therefore, the contradictory results observed in AASC will diminish, thus making it a good candidate to compete with PC.
- **Low workability and setting time:**
This restriction of alkali-activated cement depends on the type of activator. In fact, obtaining the desirable mechanical properties and suitable workability is made feasible only by using an appropriate activator.
- **High Shrinkage:**
Different studies proposed numerous methods to reduce the shrinkage of AASC. However, all of these methods reduce the shrinkage to some extent, and some of these methods affected other properties of the AASC that was uncalled for.
- **Lack of standard:**
There are a few standards related to AASC. However, the PC-related standards are currently used for AASC as well. This restriction prevents stockholders from accepting the AASC. Therefore, there is a need for developing appropriate rules and standards to improve the emerging market of AASC products.
- **Large-Scale Applications:**
Large-scale PC has been used for many years. However, not many applications have been reported for AASC. As a result, consumers doubt whether it is still the best option.

Although slag has been widely used in the preparation of alkali-activated concrete, its production becomes more difficult more than ever. Apparently, using slag in combination with other recycled materials can be promising. There are appreciated researches in this field. For Example, recycled concrete/brick powder may be a high value-added alternative [65-67].

9. APPLICATION OF AASC

In recent years, thorough information on the chemistry, slag and alkali activator reaction products, mechanical properties, and durability of AAS cement has been obtained. However, its application as a product has not been fully investigated yet. The quick setting of this type

of cement makes it a good candidate as a repair material with excellent durability. The excellent resistance of this cement against the diffusion of chlorides and freezing encourages its applications in cold climates and salty environments. Owing to its numerous advantages, AASC have wide applications in manufacturing products such as prefabricated cement blocks with different sizes, corrosion and heat resistant concretes, and mortars. In addition, this cement is innovatively used in the immobilization of radioactive waste and heavy metals [68-70].

10. CONCLUSION

The present study made an attempt to present a comprehensible overview of AASC. Compared to PC, AASC enjoys several advantages such as higher mechanical strength, lower hydration heat, higher resistance to chemical attacks, and carbon diffusion. However, it also has some drawbacks namely rapid setting and lower workability, higher shrinkage, and consequent cracking. According to the findings, different factors could affect the properties of AASC among which, water-to-slag ratio, mixing protocol, curing conditions (temperature and humidity), dosage and type of activator, different admixture, and slag composition are the most important ones that should be optimized according to the desired properties.

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