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Mix Design Effects on the Durability of Alkali-Activated Slag Concrete in a Hydrochloric Acid Environment

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Abstract: Because of its high strength, energy reduction, and low environmental impact, researchers have encouraged considering alkali-activated slag concrete (AASC) as a potential alternative to conventional concrete. In this study, the impact of mix design parameters on the durability of AASC, made with ground granulated blast furnace slag and activated with different alkaline solutions (NaOH, KOH, and Na₂SiO₃) immersed up to six months in a hydrochloric acid bath with pH = 3, has been investigated. A total of 13 mix designs were made in a way that, in addition to the type of alkaline solution, considered three other parameters, namely the molarity of alkaline solutions, the weight ratio of alkaline solutions to slag, and the weight ratio of alkaline solutions to sodium silicate. Visual inspections displayed that the AASC samples almost remained intact after exposure to an HCl acid solution with pH = 3 for up to 6 months, while the OPC sample experienced deleterious deterioration. The results clearly show that AASC outperformed OPC concrete when it comes to durability in an HCl acid solution. The strength reduction and weight loss of AASC compared with OPC concrete were approximately one-tenth and one-fifth, respectively. The AASC samples containing potassium hydroxide showed a higher strength reduction and weight loss in the HCl acid solution than the samples made with sodium hydroxide.

Keywords: alkali-activated concrete; slag; alkaline solution; hydrochloric acid



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1. Introduction

Ordinary Portland cement (OPC) concrete has adverse impacts on the environment because of its production, the release of huge quantities of CO₂, and consuming a large amount of energy [1]. Moreover, its durability is also a significant drawback, especially in acidic environments [2,3]. Thus, researchers have always been searching to compensate for OPC's shortcomings and to reduce its environmental impact. An array of materials have been suggested, one of which has extensive popularity is alkali-activated concrete [4–6]. In general, alkali-activated concrete is a mixture of an aluminosilicate precursor activated with alkaline solutions [7,8]. Metakaolin, ground granulated blast furnace slag (GGBFS), and fly ash are common aluminosilicate sources. Sodium hydroxide (NaOH), potassium hydroxide (KOH), and sodium silicate (Na₂SiO₃) are the mainly used alkaline solutions, where sodium silicate is in solution with NaOH and KOH. Ground granulated blast furnace slag is a solid waste of steel production reported to have an acceptable performance in alkali-activated concrete as an aluminosilicate source [9]. Alkali-activated slag concrete (AASC) is an environmentally friendly product characterized by high percentages of alumina and silica activated with alkaline solutions, which provides a possibility for the production of cement-free concrete. AASC with relatively large amounts of silicon oxide and aluminum oxide is considered a potential alternative to OPC concrete for non-structural concrete and reinforced concrete structures [4,10,11].

It is reported that AASC is environmentally friendly because of its lower greenhouse emissions compared with OPC concrete, but it is also economically sustainable owing to its lower energy consumption [12–14]. Because of its high final strength, energy reduction, and low environmental impacts, researchers have encouraged considering AASC as a potential alternative for conventional concrete, especially in the precast concrete industry [15,16], while high shrinkage (3.3 times the OPC drying shrinkage as reported in [17]) and carbonation are two major imperfections of AASC compared with conventional concrete. AASC offers favorable properties, some of which include durability in aggressive and acidic environments [4,17]. Numerous studies have been performed to investigate the diverse characteristics of AASC, such as the mechanical properties [7,18–20], shrinkage [21,22], carbonation [23,24], and elevated temperature endurance [25]. The main hydration product of AASC is calcium aluminum silicate hydrate (C-A-S-H) gel, with a quite quick setting time and a low Ca/Si ratio compared with OPC concrete [13,20].

Cities in the developing world face outstanding economic and human losses caused by man-made or natural hazards, and the amount of loss is affected by the quality of the preventive measures and emergency management [26,27]. Concrete can be subjected to an acid attack in diverse circumstances. Conditions such as acid rain, acid river, sewerage, and chemical facility are examples of acidic environments that can bring about irreparable damage to concrete structures [28]. A few studies have been carried out to investigate the durability of AASC in acetic acid, sulfuric acid, nitric acid, and phosphoric acid environments [29–32]. Acid attack is a vital issue regarding concrete performance that adversely affects concrete durability. AASC is reported to have superior durability in acidic environments compared with conventional concrete. The main reasons for the high durability are the low permeability and less calcium in its composition compared with OPC concrete [29,32].

In an actual environment, the acid attack is not a pure acid, but often mixed acids. For example, in acid rain, the acid attack on concrete contains a mixture of sulfuric acid, nitric acid, and hydrochloric acid [33]. OPC concrete is alkaline and is susceptible to acidic environments [3]. The equilibrium of the cement matrix is quickly disturbed when the pH of the concrete pore solution drops. At pH values lower than 12.6, the first hydration product that dissolves is calcium hydroxide (portlandite), which converts to a calcium salt of the acid, hydrogels of silicium, aluminum, and ferric oxide [34]. The corrosion rate of concrete after an acid attack is a complex process governed by the combination of the dissolution, precipitation, and transport processes, as determined by the cement chemical composition, paste matrix reactivity, aggregate reactivity, aggregate grading, and concrete composition [35].

Hydrochloric (HCl) acid is a strong acid categorized as a mineral acid found in municipal sewage, acid rain, and industrial effluent. HCl acid is utilized in the metal industry as well as in the manufacture of fertilizers, dyes, and pigments, and the wastewater from these industries can be very aggressive for concrete structures [36]. When OPC paste comes in contact with HCl acid, calcium chloride (CaCl_2) is produced [28]. CaCl_2 is a highly soluble salt that can be leached out and causes concrete deterioration [37]. It is reported that the most important factor governing the corrosion rate in an acid attack on cement-based materials is calcium salt solubility, and the second factor is acid strength [38]. HCl acid is considered an aggressive acid because of its highly soluble calcium salts and dissociation ability [39]. In addition, the formation of Friedel's salt, which is an expansive product, accelerates concrete deterioration [34].

A few studies have investigated the durability of AASC in HCl acid attack [40,41]. It has been reported that AASC has a better durability compared with conventional concrete in aggressive environments [42]. Thunuguntla et al. investigated the effect of mix design parameters on the mechanical and durability properties of AASC in three different acidic environments, including hydrochloric acid solution, and NaOH was considered the only alkaline solution. Eight different mix designs were formulated for the tests. The results showed that the NaOH concentration was found to be the most influential parameter on the

mechanical strength and durability characteristics of AASC [41]. Molarity and type of alkaline solutions are two vital elements in the mix design of AASC [43]. There is no denying that variability in the chemical composition of slag may impact its durability, and concretes prepared using various slags may have different resistance in aggressive media [44].

In the literature, a limited number of studies focused on a systematic approach for considering the effect of the mix design parameters of AASC on its performance and durability in acidic environments. Concrete durability is a crucial property, as concrete structures are required to sustain and endure throughout the entire service life acceptably. In the vast majority of studies, the acid resistance of alkali-activated concrete with a fixed mix design was studied. To fill this knowledge gap, an experimental study is required to investigate the effect of the diverse mix design parameters on the durability and mechanical properties of AASC exposed to an HCl acid solution. Thus, in this paper, the impact of mix design parameters on the durability of AASC samples, made with GGBFS and activated with different alkaline solutions (NaOH, KOH, and Na_2SiO_3) immersed up to six months in a hydrochloric acid bath with $\text{pH} = 3$, has been investigated. As well as the type of alkaline solution, three other parameters, including the molarity of alkaline solutions, the weight ratio of alkaline solutions to slag, the weight ratio of alkaline solutions to sodium silicate, were considered. OPC concrete samples were also made as a control mixture. The XRF test measured the chemical compositions of the slag and Portland cement. For all AASC and OPC concrete specimens, compressive strength reduction and weight loss were monitored at predetermined intervals.

2. Materials and Methods

In this study, slag as a silica-aluminate precursor was used according to ASTM C989M. The chemical compositions of GGBFS and Portland cement, based on the XRF test, are presented in Table 1. Additionally, GGBFS specific gravity and Blaine fineness were equal to 2.85 gr/cm^3 and $400 \text{ m}^2/\text{kg}$, respectively. The weight ratio of $\text{Al}_2\text{O}_3/\text{SiO}_2$ and CaO/SiO_2 , basicity coefficient ($K_b = (\text{CaO} + \text{MgO})/(\text{SiO}_2 + \text{Al}_2\text{O}_3)$), and hydration modulus ($(\text{CaO} + \text{MgO} + \text{Al}_2\text{O}_3)/\text{SiO}_2$) were equal to 0.447, 1.079, 0.916, and 1.77, respectively.

Table 1. Chemical compositions of GGBFS and Portland cement based on XRF test (wt.%).

	CaO	SiO ₂	Al ₂ O ₃	MgO	TiO ₂	MnO	S	K ₂ O	Fe ₂ O ₃	Na ₂ O	SO ₃	L.O.I *
GGBFS	36.52	38.35	10.88	8.77	1.48	1.25	1.21	0.93	0.52	0.49	-	0.26
Cement	63.50	21.50	5.10	2.30	-	-	-	0.93	3.80	-	2.00	0.70

* Loss of Ignition.

Alkaline solutions were added to the binder in liquid form. The alkaline solutions were sodium hydroxide (NaOH), potassium hydroxide (KOH), and sodium silicate (Na_2SiO_3), which had a $\text{SiO}_2/\text{Na}_2\text{O}$ ratio equal to 2.5 ($\text{Na}_2\text{O} = 14\%$, $\text{SiO}_2 = 35\%$, and water = 51%). The hydration process of alkali-activated slag is influenced by the sodium content and silica module [45]. It should be stated that water glass (NaOH) and potassium hydroxide (KOH) flakes were dissolved in water to make the solutions with a needed molarity based on the mix design parameters. The mixing process was carried out using a mixer with 60 L volume. Note that the preparation of the alkaline solution released heat, however, these solutions were not added to the mixture right away after preparation. After preparation, it took at least 5 min before being added to the final mixture. As a result, the temperature almost reached the ambient temperature.

The physical properties of aggregates are presented in Table 2. The fine aggregates were crushed sand. Sand equality and water absorption were measured in accordance with ASTM D2419 and ASTM C128, respectively. Gravel (Crushed stone) with a maximum aggregate size (MSA) of 19.5 mm was used as the coarse aggregates. The saturated surface dry specific gravity and water absorption were measured as per ASTM C127. It should be

noted that limestone aggregates were used because they are widely used in a majority of projects in Iran. The grading of aggregates met the requirements of ASTM C33.

Table 2. Physical properties of aggregates.

Type of Aggregate	Fineness Module	Sand Equality	SSD Specific Gravity (gr/cm ³)	Water Absorption (%)
Fine	2.99	77	2.47	2.06
Course	-	-	2.59	0.76

The AASC mix proportions were selected based on previous studies [14,15]. As mentioned before, the selected parameters included the alkaline solution type, the molarity of alkaline solutions, the weight ratio of alkaline solutions to slag, and the weight ratio of alkaline solutions to sodium silicate. A total of 13 mix designs were made in a way that four selected parameters are considered, as shown in Table 3. It should be noted that when a parameter (for example, NaOH molarity) was changed, the other mix design parameters (such as NaOH/Na₂SiO₃, alkaline solution/slag, and alkaline solution type) remained constant.

Table 3. AASC mix designs.

Mix Code	Alkaline Solution (Kg/m ³)	Slag (Kg/m ³)	The Weight Ratio of NaOH (KOH) to Na ₂ SiO ₃	The Weight Ratio of Alkaline Solution to Slag	Molarity	Type of Alkaline Solution
N6041	158	394	1	0.4	6	NaOH
N6043	158	394	3	0.4	6	NaOH
N10041	158	394	1	0.4	10	NaOH
N10043	158	394	3	0.4	10	NaOH
N14041	158	394	1	0.4	14	NaOH
N14043	158	394	3	0.4	14	NaOH
N60404	158	394	0.4	0.4	6	NaOH
N100404	158	394	0.4	0.4	10	NaOH
N6061	207	345	1	0.6	6	NaOH
N10063	207	345	3	0.6	10	NaOH
K6041	158	394	1	0.4	6	KOH
K6043	158	394	3	0.4	6	KOH
K10043	158	394	3	0.4	10	KOH

To evaluate the acid resistance of the AASC mixes, samples were exposed to an HCl acid bath with pH = 3. The pH was selected based on previous research [29,30,44,46]. The ratio of water to solid materials was 0.50, and the weight percentage of the aggregates in the mixture was 77%. To keep the workability of all of the AASC samples the same, a naphthalene-based superplasticizer was used [14,15].

At first, the aggregates and slag were mixed for 3 min, then the alkaline solutions were added. In the end, water and the admixture were added, and the mixture thoroughly mixed for 5 min, followed by a rest for 1 min, and remixing for 3 min. Cube molds (100 × 100 × 100 mm) were used to prepare the AASC samples. The prepared samples were covered with plastic sheets to minimize water evaporation. They were kept at ambient temperature (23 ± 2 Celsius) for 24 h, and were then taken out of molds and placed in water baths for two weeks before immersion in acid solutions. The reason for selecting this time is that, based on the compressive results, a majority of AASC specimens gained nearly 80% of 180 days compressive strength in the first 14 days, as discussed in the Results and Discussion section. To have a constant pH level in acid baths, the pH of the container was measured continuously with a pH meter twice a week, and the HCl acid was added as needed. Three replicate cubes from each mix design at predetermined interval days (28, 90, 120, and 180 days) were used for the compressive strength tests and weight loss measurements.

The compressive strength of the OPC concrete cubic samples (100 × 100 × 100 mm), as a control mix, was 60 MPa at 28 days, and its water to cement ratio was 0.28. Furthermore,

the same aggregates that were used in the AASC mixtures were also used in the OPC samples. For mixing, first aggregate and cement were mixed for 3 min and then water was gradually added to the mixture. Moreover, a polycarboxylate superplasticizer was used to enhance the workability. After casting, OPC concrete specimens were kept at ambient temperature (23 ± 2 Celsius) for 24 h, and then were taken out of molds and cured in water for up to 28 days. Then, they were transferred to the HCl acid bath.

Because of the deficiency of a standard procedure for evaluating acid attacks on concrete [29], the resistance to acid attacks on AASC and conventional concrete was tested by immersion in an HCl acid bath as presented. Deterioration of specimens was tested by compressive strength reduction and weight loss at predetermined intervals (28, 90, 120, and 180 days after immersion in HCl acid bath) based on the procedure used by [29,44,46]. A reduction in strength or weight loss was calculated using following equation $((A-B)/A)$; where A is the average compressive strength or weight of three cubic samples cured in water, and B is the average compressive strength or weight of three cubic samples cured in an HCl acid bath.

The process was as follows: after casting, the AASC samples were kept at ambient temperature (23 ± 2 Celsius) for 24 h and were taken out of the cube molds and cured in water for 14 days. Then, the specimens were taken out of the water, were kept at ambient temperature for three hours, and were sandpapered on all six sides before weighing. Afterward, they were immersed in HCl acid baths until the experiment time (90, 120, and 180 days). The samples were weighed again after 3 h and after being sandpapered on all six sides. The weight difference before and after immersion in the HCl acid solutions was calculated as the weight loss. The compressive strength of the cubic samples was measured according to EN 12390-3, and three cubes were tested at predetermined intervals. The companion specimens, cured in potable water, were also tested to find the strength reduction.

3. Results and Discussion

In the following paragraphs, firstly, the compressive strength development of the AASC cured in water is discussed. Then, the impacts of four mix design parameters on the acid resistance of AASC exposed to an HCl acid solution are exploited. Finally, a comparison between the results of this study and the reference OPC concrete is discussed.

3.1. Compressive Strength of AASC Samples before Acid Exposure

The compressive strength results of the OPC concrete and AASC specimens cured in water for up to 180 days are shown in Figure 1. After 14 days of curing in water, most AASC samples showed a high compressive strength, where more than 44 MPa is considerable. This agrees well with previous findings [47], as reposted by [48], that AASC has a rapid development of compressive strength at early ages [49]. Gruskovnjak et al. reported that a faster strength development of ASSC compared with OPC is owed to a thin protective layer that covers the unhydrated slag grain systems. In the OPC concrete, this layer is considerably thicker, causing slower compressive strength growth [50]. Moreover, different hydration products in the two systems can also be considered as another main reason for the different strength development rates [12,13].

The results show that the mix design contains 10 M KOH, the weight ratio of the slag to alkaline solution equals 0.4, and the weight ratio of KOH to Na_2SiO_3 equals 3 (Mix code: K10043), which gained 86% of its 180 days compressive strength after 14 days. All three mix designs made with KOH gained more than 60 MPa compressive strength after 14 days of curing in water. Reddy et al. observed the same results [51]. It can be argued that KOH is a stronger alkaline solution than NaOH, and when it comes in contact with slag, a very rapid reaction happens, leading to gaining a considerable percentage of compressive strength at early ages.

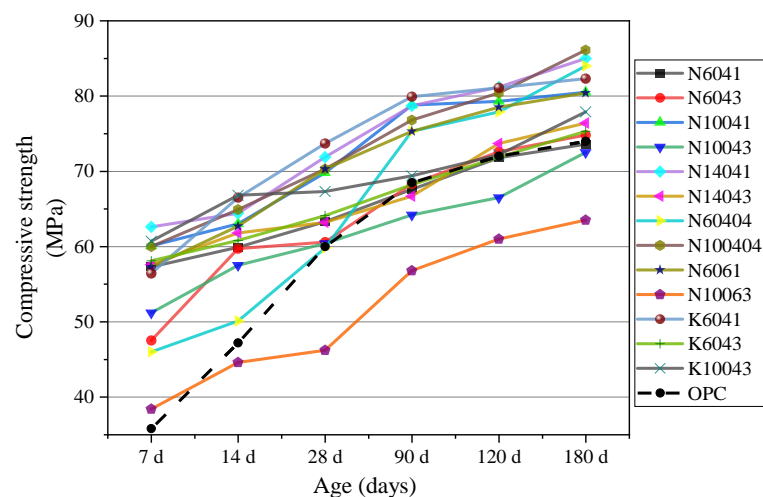


Figure 1. Compressive strength of AASC samples ($100 \times 100 \times 100$ mm) cured in water before exposure to HCl acid.

At 28 days, the lowest and highest compressive strength were 46.2 and 73.7 MPa related to N10063 and K6041, respectively. According to the literature, when the NaOH or KOH molarities are increased and the other parameters are the same, a higher compressive strength is achieved for all ages [52]. This can be related to the higher alkalinity of the AASC brought about by the increase in molarity, leading to the formation of more hydration products [49,53]. Fang et al. reported that the increase of NaOH from 10 to 12 led to a 23% increase in compressive strength [54]. The concentration of alkaline solutions increased the reaction rates and thus led to a higher compressive strength. Aliabdo et al. used 10 M, 12 M, and 14 M NaOH solutions, and reported an increase in molarity that significantly impacted the compressive strength at the age of 28 days [7].

3.2. Effect of Mix Design Parameters on Performance in Acid Attack

The HCl acid attack mechanism on AASC is a reaction between calcium-based compounds in the concrete paste and a solution of hydrogen chloride. This reaction initiates the formation of highly soluble calcium salt (CaCl_2). Visual inspections show that AASC samples were almost intact after exposure to HCl acid solution for up to 6 months, while the OPC sample experienced a deleterious deterioration, as shown in Figure 2. Although this comparison proved that AASC is superior to OPC in resisting HCl acid attack, other conducted tests also confirmed this observation, as discussed in the following sections.

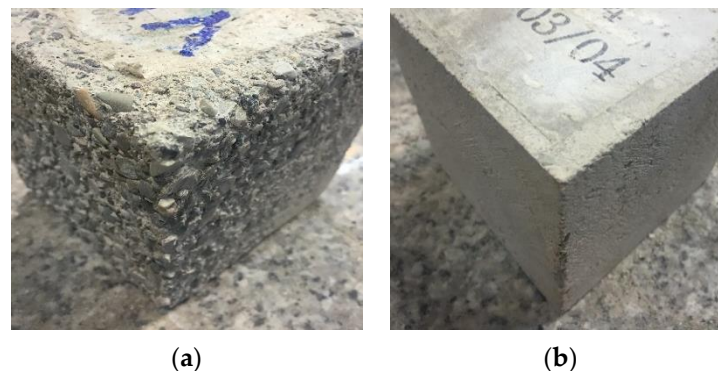


Figure 2. (a) OPC and (b) AASC samples ($100 \times 100 \times 100$ mm) appearance after six months of immersion in an HCl solution with pH = 3.

3.2.1. Type of Alkaline Activator

Numerous studies investigated AASC durability [29,31,44,55] and the correlation between the durability and microstructural characteristics [56] containing NaOH as an alkaline activator. Potassium hydroxide (KOH) has always been a potential alternative for sodium hydroxide (NaOH) in alkali-activated concrete [12]. In this study, two alkaline solutions were used, caustic soda (NaOH) and potassium hydroxide. KOH is a stronger alkaline solution than NaOH [57], and its price is at least two times greater than NaOH. It is reported that alkali-activated materials activated by KOH have acceptable mechanical and durability properties [46,58–60]. As shown in Figure 3, replacing KOH with caustic soda when other mix design parameters remained the same resulted in more strength reduction, especially after 120 and 180 days of exposure to the acid solution.

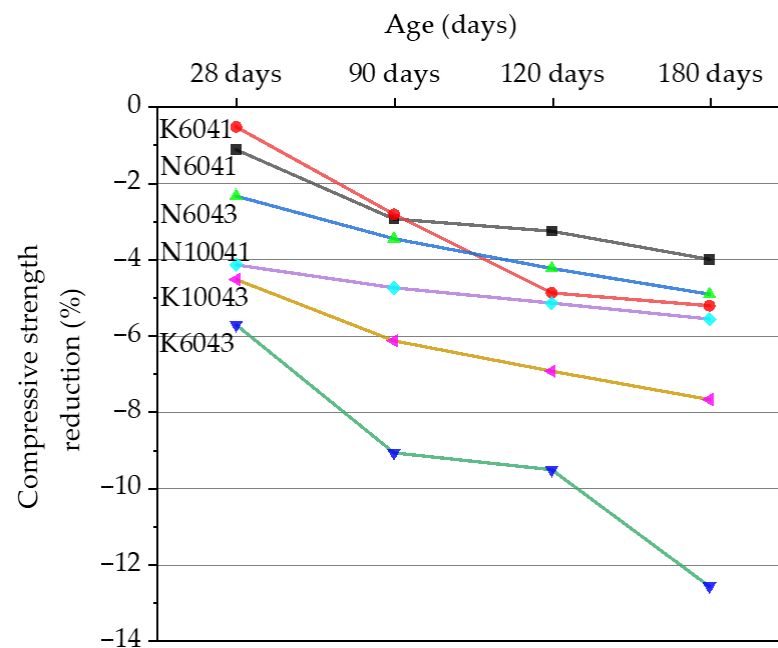


Figure 3. Compressive strength reduction of AASC with different alkaline activators immersed in an HCl acid solution.

Based on the results, the AASC samples made with KOH immersed in HCl acid for six months showed at least a 30% increase in strength reduction compared with the specimens made with NaOH. The obtained results showed that a progressive compressive strength reduction was observed with an increase in exposure time for all of the mixes. Figure 3 shows that N6043 experienced roughly a 5% strength reduction after 180 days of immersion in HCl acid. However, by replacing KOH (K6043), the strength reduction was about 13% at the same age. The cause of the unsatisfying performance lies in how KOH reacts to slag [61]. When potassium hydroxide reacts with slag, the reaction is rapid, creating a structure with countless tiny unfilled holes and a less homogeneous microstructure [61]. When this porous structure exposes aggressive environments such as an acid solution, the acid expands in the pores, causing concrete deterioration and strength reduction [62].

Thus, because of the undesirable compressive strength and costly prices of KOH compared with NaOH, its application does not have technical justification or financial benefits. A limited number of studies have been conducted on the effect of acid attack on alkali-activated slag concrete made with KOH. Bakharev reported that alkali-activated concrete made with fly ash and activated by KOH resulted in a larger pore diameter, which deleteriously impacted the concrete durability in the sulfuric acid solution [46]. The compressive strength reduction of the reference OPC concrete is reported in Section 4, and a comparison between the OPC and AASC results is also discussed.

Figure 4 shows the effect of the four selected mix design parameters on the weight loss of AASC samples immersed in HCl solutions for up to 180 days. Figure 4a indicates that when KOH replaced NaOH, weight loss increased approximately 67% after 180 days of immersion in an HCl acid bath. Hence, the replacement of KOH caused adverse effects on the durability of AASC. The results indicated that KOH replacement increased the weight loss of the AASC specimens by 100%, 123%, and 67% on days 90, 120, and 180, respectively. To explain this observation, as discussed before, it can be argued that KOH rapidly reacts with slag and creates a less homogeneous microstructure with countless tiny unfilled holes. This weak microstructure allows acid agents to penetrate the AASC structure and deteriorate bonds [61]. Hence, it seems unsatisfying to replace NaOH as a common activator with a reasonable price with KOH when the durability of AASC matters. The effect of other parameters (as shown in Figure 4) will be discussed in the corresponding section.

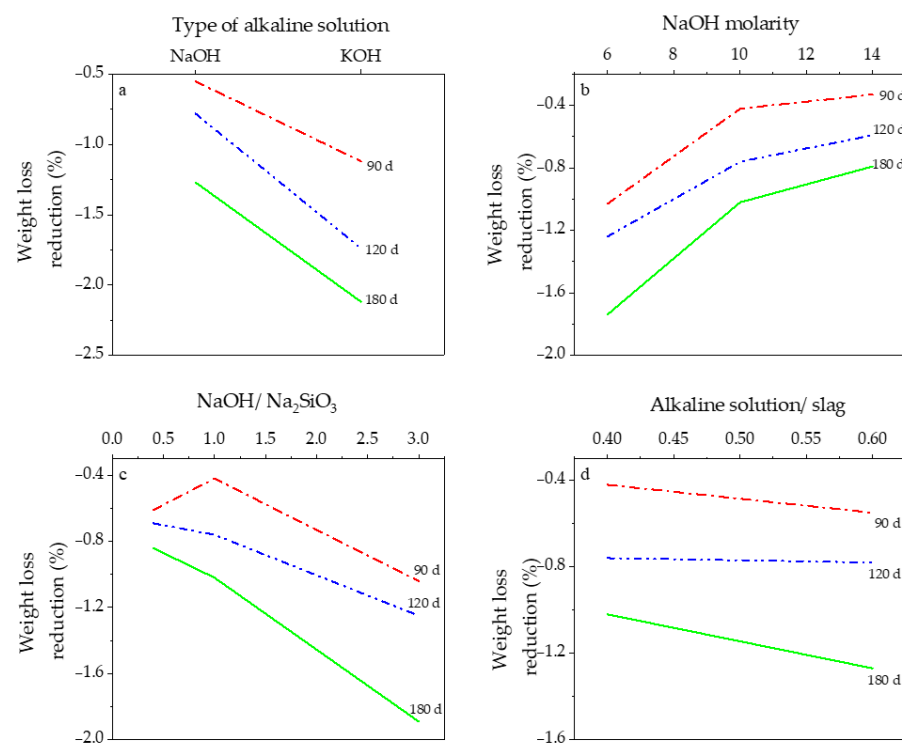


Figure 4. The effect of mix design parameters on the weight loss of AASC samples immersed in an HCl acid solution (a) Type of alkaline solution, (b) NaOH molarity, (c) NaOH/Na₂SiO₃, and (d) weight ratio of the alkaline solution to slag.

3.2.2. Sodium Hydroxide Molarity

One of the principal mix design parameters that can tremendously affect the alkali-activated slag concrete characteristics is the molarity of the alkaline solutions [41]. Figure 5 shows the compressive strength reduction of the AASC samples with different molarities immersed in an HCl acid solution for up to six months. In Figure 5a, the weight ratio of NaOH/Na₂SiO₃ equals 1, and in Figure 5b, the ratio is 3. Regardless of the NaOH to Na₂SiO₃ ratio, an increase in the NaOH concentration led to more strength reduction at all ages. Therefore, AASC samples containing less molarity of caustic soda had a superior performance in the HCl acid environment. Therefore, although a higher NaOH molarity led to a higher compressive strength for the AASC cured in water, the acid resistance of the AASC improved when the lower molarity of NaOH was used. Thunuguntla investigated the durability of AASC using 1 M and 8 M NaOH in contact with HCl, H₂SO₄, and HNO₃ for up to 56 days. The loss of compressive strength for the specimens immersed in HCl

acid was about 1 to 4% that is very little. He concluded that AASC provides outstanding resistance to all of the acid solutions used here compared with OPC [41].

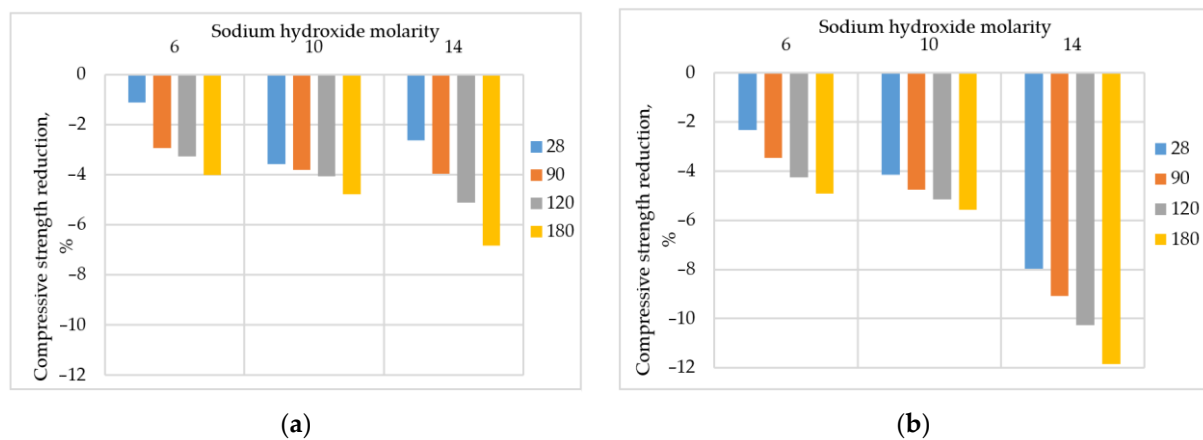


Figure 5. Compressive strength reduction of AASC with different NaOH molarities in an HCl acid solution for (a) NaOH/Na₂SiO₃ = 1 and (b) NaOH/Na₂SiO₃ = 3.

Figure 5a,b shows that the NaOH/Na₂SiO₃ ratio directly affects the influent of NaOH molarity. In Figure 5a, the AASC samples exposed to HCl acid for 180 days lost around 7% of their compressive strength, but the strength reduction was approximately 12% in Figure 5b. The underlying reason is that the sodium hydroxide amount was higher, and in the NaOH/Na₂SiO₃ ratio it equaled 3. The higher sodium hydroxide molarity increased the reaction rate between the slag and alkaline solutions, creating AASC with irregular microstructures and causing more deterioration in acid attacks. After 180 days of exposure to the acid, when the sodium hydroxide molarity changed from 6 to 14, the strength reduction increased by approximately 71% (Figure 5a) and 140% (Figure 5b).

Figure 4b shows the weight loss reduction of the AASC samples with NaOH concentrations of 6 M, 10 M, and 14 M. The results indicated that an increase in NaOH molarity led to a decrease in weight loss. The main reason for this is that when the NaOH molarity increased from 6 to 14, the alkalinity of the concrete increased. It should be noted that AASC samples made with 14 M NaOH had a dark green color compared with the specimens made with a lower molarity, which had a pale green color. The mix design, which had the highest concentration (14 M), experienced 55% less weight loss than the AASC samples with the lowest molarity (6 M) after six months of exposure to the HCl acid solution.

3.2.3. Weight Ratio of Sodium Hydroxide to Sodium Silicate

In a majority of investigations, two combined alkaline solutions were used. Sodium hydroxide and sodium silicate are the two most used alkaline activators in the literature [44,52,63]. Thus, the weight ratio of these two alkaline solutions can be an influential parameter when AASC mix design is to be selected. In this study, three ratios, 0.4, 1, and 3, were used. The effect of the ratio of NaOH/Na₂SiO₃ on the strength reduction was evaluated when the other parameters were the same. Figure 6a,b shows the strength reduction when the NaOH molarities were 6 and 10, respectively. Regardless of the concentration of the sodium hydroxide solution, the highest strength reduction occurred when the NaOH/Na₂SiO₃ ratio equaled 3 at all ages. In addition, when the ratio increased from 1 to 3, strength reduction at 180 days rose approximately 23% and 16% for 6 M and 10 M NaOH, respectively. To explain this phenomenon, it can be said that the lower NaOH/Na₂SiO₃ ratio means the more sodium silicate in the paste, which is a great source of Si, enhancing the production of the C-S-H (or C-A-S-H) gel and providing a denser AASC. Therefore, when the ratio was one, the AASC samples showed a better performance after acid attacks in the long run.

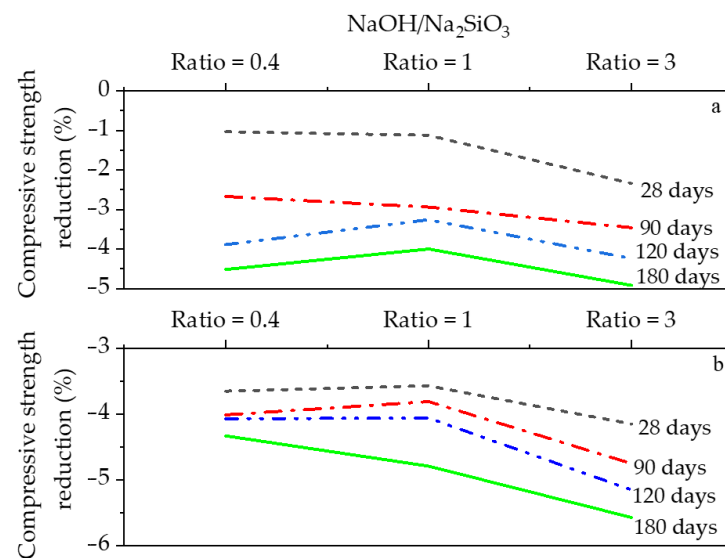


Figure 6. Compressive strength reduction of AASC with different NaOH/Na₂SiO₃ ratios for (a) 6 M NaOH and (b) 10 M NaOH.

It should be noted that when it comes to workability, the lowest ratio (NaOH/Na₂SiO₃ = 0.4) adversely affects the workability of the fresh concrete. Concrete workability is an indispensable part of concrete production and implementation. It should be taken into consideration when the mix design parameters are selected. The impact of the ratio of NaOH/Na₂SiO₃ on AASC weight loss is shown in Figure 4c. It can be concluded from Figure 4c that an increase in the ratio of NaOH/Na₂SiO₃ from 0.4 to 3 caused the escalation of weight loss. After six months of immersion of AASC samples in the HCl acid solution, weight loss for NaOH/Na₂SiO₃ = 3 became more than two times greater than the specimens with NaOH/Na₂SiO₃ = 0.4. The principal reason is that the higher ratio of NaOH/Si₂O₃ decreased the sodium silicate content, which is an excellent source of Si in the concrete paste. Si is an essential constituent for the production of C-(A)-S-H gel in alkali-activated slag concrete.

3.2.4. Weight Ratio of Alkaline Solution to Slag

In the OPC concrete, the water to cement ratio is a critical parameter, especially when it comes to durability. The weight ratio of the alkaline solution to slag has a critical role in the properties and performance of the alkali-activated materials [41]. Limited studies have evaluated the impacts of this essential parameter on AASC durability. In most studies, a ratio of alkaline solution to slag was selected in a range of 0.4 to 0.6 [29,41,44,63,64]. When the ratio increased from 0.4 to 0.6, the quantity of alkaline solution in the concrete increased, and a lower amount of slag existed in the mixture as paste. In this study, two weight ratios of the alkaline solutions to slag were used, 0.4 and 0.6.

Figure 7 shows the impact of the weight ratio of the alkaline solution to slag on the strength reduction of AASC specimens exposed to HCl acid solutions for up to 6 months. Figure 7a shows that altering the weight ratio of the alkaline solution to slag from 0.4 to 0.6 led to nearly 10% more strength reduction for the AASC specimens subjected to HCl acid solution for six months. Likewise, for 10 M NaOH and NaOH/Na₂SiO₃ = 3 (Figure 7b), more alkaline solution caused nearly 29% more strength reduction. In Figure 7a,b, the AASC samples with a ratio of 0.4 had the lowest strength reduction at all ages. The fundamental reason is that the higher ratio of alkaline solution to slag reduced the adequate amount of slag and paste in the mixture. A definite amount of alkaline solutions is needed to activate the entire volume of slag in the mixture, thus up to that volume, the presence of an alkaline solution enhanced the mechanical properties of AASC [64]. The same results are observed in [41], where an increase in paste content led to an enhancement of acid attack for AASC mixes.

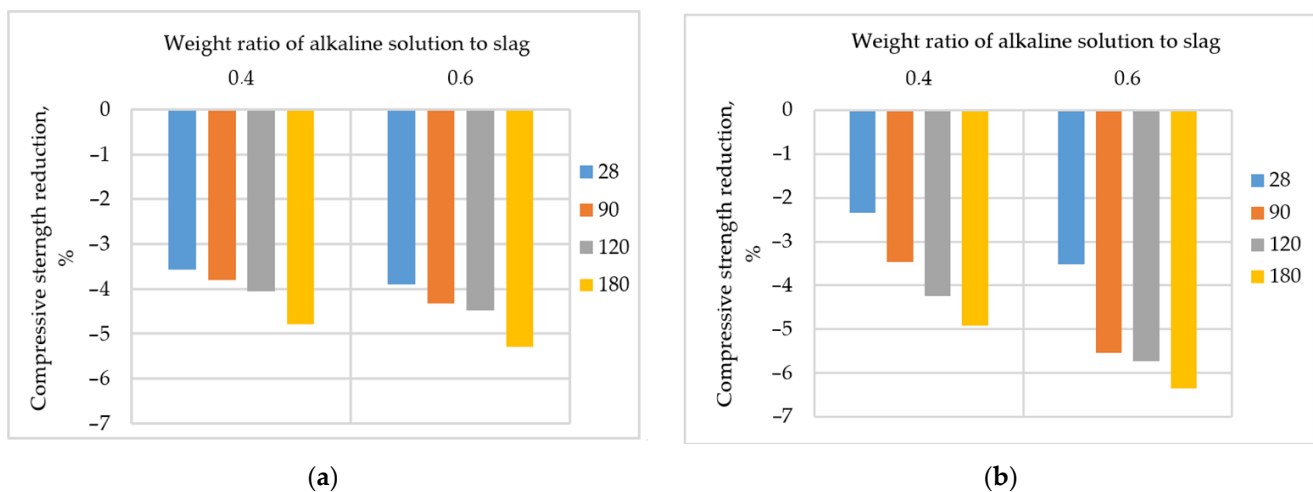


Figure 7. Compressive strength reduction of AASC with different alkaline solution to slag ratios for (a) NaOH molarity = 6, NaOH/Na₂SiO₃ = 1 and (b) NaOH molarity = 10, NaOH/Na₂SiO₃ = 3.

In addition, the alkaline solutions consisted of a considerable amount of water. Hence, when the ratio of the alkaline solution to slag increased, extra alkaline solutions created countless capillary pores, which could develop a poor structure, inferior performance, and deterioration in acid attacks [64]. The findings of this study pointed out that more alkaline solutions initially, with a ratio of 0.6, resulted in a lower resistance in acid attack, and a fewer alkaline solutions, with a ratio of 0.4, led to a better performance of AASC after exposure to the acid.

Figure 4d shows the weight loss percentage under the influence of the weight ratio of alkaline solutions to slag. The ratio did not significantly affect the weight loss after 90 and 120 days of immersion of the AASC samples in HCl acid. When the ratio of alkaline solutions to slag changed from 0.4 to 0.6 after 180 days, the weight loss reduction increased by about 20%. As discussed above, when the weight ratio of the alkaline solutions to slag increased, an excessive amount of alkaline solutions consisting of a substantial amount of water in the AASC mixture would be available, which could negatively affect the performance of AASC for a long time. As reported in [41], the weight loss of AASC tended to decrease when the solution to binder ratio increased. The main reason is that more paste in the structure reduced the voids and ingress of acid agents.

4. Comparison of OPC Concrete and AASC in HCl Acid Solution

In this section, the performance of OPC concrete is presented and compared with AASC in an HCl acid solution. As stated before, alkali-activated materials show acceptable mechanical properties and durability in acidic media [65,66]. Nevertheless, when acid attacks OPC concrete, calcium components, especially calcium hydroxide, hydrated calcium silicate, and hydrated calcium aluminate, convert into calcium salts, leading to deterioration and cracking [3].

As presented in Figure 8a, the strength reduction of OPC concrete after six months of immersion in HCl acid was approximately 38%. However, the minimum and maximum strength reductions of AASC samples with the same exposure time to HCl acid for all of the mix designs were 4% and 12.6%, respectively. In other words, the minimum and maximum AASC strength reductions were one-tenth and one-third of OPC concrete, respectively. This means a well-designed alkali-activated slag concrete has a superior performance when it comes to strength reduction. Accordingly, a concrete structure or element erected by OPC concrete that deteriorated after two years due to acid attacks would acceptably work for at least 20 years, if built by AASC with well-selected mix design parameters.

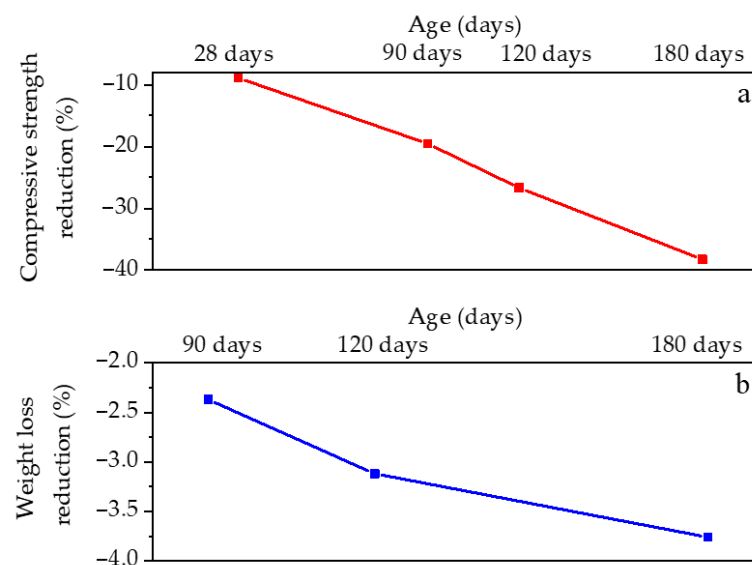


Figure 8. (a) Compressive strength and (b) weight loss reduction of OPC concrete in an HCl acid solution.

The weight loss of the OPC concrete was 3.76% after 180 days of immersion in an HCl acid solution, as shown in Figure 8b. Nonetheless, the minimum weight loss for the AASC samples was 0.79%. This means that the weight loss of AASC was nearly one-fifth of the OPC concrete. The better performance of AASC could be attributed to several factors. The main factor is the different structures between the two concretes. It is reported that AASC has a calcium (aluminato) silicate (C-(A)-S-H) hydrate structure similar to the OPC hydration products, but with a lower ratio of Ca/Si compared with OPC concrete [49]. The chemical and physical characteristics of slag (Na₂O content in NaOH and silicate module (SiO₂/Na₂O) in Na₂SiO₃) are other factors that can negatively or positively impact the durability of AASC [49]. In this study, the amount of CaO in the slag (Table 1) was about half of the CaO amount in the Portland cement. Hence, AASC samples have lower calcium components in their composition, leading to a better performance in acid attacks.

The obtained results of the weight loss reduction test are in agreement with the literature. Torgal et al. reported a 2.6% average weight loss for AASC specimens after exposure to three acid solutions (HCl, H₂SO₄, and HNO₃) for 28 days, which was less than half of the observed weight loss for OPC concrete [67]. Bakharev stated 1.15% and 12.43% weight loss for alkali active concrete made with fly ash activated by NaOH and Na₂SiO₃ after 60 days of exposure to acetic acid and sulfuric acid solutions, respectively [46]. Munn et al. investigated the weight loss for OPC and alkali-activated concrete made with fly ash and activated by NaOH and Na₂SiO₃. He reported a 40.9% weight loss for OPC concrete after a month of exposure to a 10% sulfuric acid solution, and 5.2% weight loss for alkali-activated concrete after two months of exposure to 10% sulfuric acid [68].

5. Conclusions

This study investigated the effects of four mix design parameters on the durability of alkali-activated slag concrete in an HCl acid solution. Having a clear understanding of the mix design parameters of alkali-activated slag concrete when there is no standard design procedure method can help researchers develop and design AASC efficiently. The results showed that AASC outperforms OPC concrete when it comes to durability in an HCl acid solution. The AASC samples contained potassium hydroxide as an alkaline activator, and showed a higher strength reduction and weight loss in the HCl acid solution than the samples made with sodium hydroxide. The lower molarity of the sodium hydroxide caused a superior performance of AASC in acid attacks. Moreover, the AASC samples with NaOH/Na₂SiO₃ = 1 compared with the samples with a ratio equal to three had an acceptable performance after acid attacks. While it might seem that the more alkaline solution would bring about a superior performance in an HCl solution, the results pointed out that

when the weight ratio of the alkali solution to slag increased from 0.4 to 0.6, the strength reduction and weight loss increased. Furthermore, for a well-designed mixture, strength reduction and weight loss of AASC compared with OPC concrete were approximately one-tenth and one-fifth, respectively.

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