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EFFECTS OF COMPOSITION OF FLY ASH-BASED ALKALI-ACTIVATED MATERIALS ON COMPRESSIVE STRENGTH: A REVIEW

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A fly ash-based alkali-activated material (FAAM) is an important member of the alkali-activated geopolymeric family. The compressive strength is one of most important mechanical properties for FAAMs in its use as a construction material. The effects of the components of the precursors from fly ash, an alkali-activator, or additives on the compressive strengths of FAAMs have been reviewed. The SiO_2/Al_2O_3, Al_2O_3/Na_2O, *and water/solid (W/S) ratios are crucial in developing the compressive strengths of FAAMs. The strength map, as a function of the change in the* SiO_2/Al_2O_3 *and* SiO_2/Na_2O *ratios has been established. There is a critical value of the* SiO_2/Al_2O_3 *ratio at about 4.20 to 4.30, in which the increase or decrease of the compressive strengths at a constant* SiO_2/Al_2O_3 *ratio with various* Al_2O_3/Na_2O *ratios is found. The active CaO resource, originating from a ground blast-furnace slag (GBFS), ordinary Portland cement (OPC), or chemical agents, such as* $Ca(OH)_2$ *or CaO, is of benefit to the improvement in the strength of FAAMs: however, to avoid the dominant formation of C–A–S–H, N–C–A–S–H, and C–S–H gels instead of N–A–S–H and A–S–H gels, an optimal addition of 7.5 % to 10 % OPC and a 15 % to 20 % GBFS replacement is recommended by considering the setting time, workability, and strength development. The effects of* Fe_2O_3 *in the fly ash and the silica on the compressive strengths of the FAAMs are also generalised.*

INTRODUCTION

Alkali-activated materials (AAMs) are environmentally-friendly binders, which are fabricated by the reaction between alkali metal sources and a solid silicate, calcium silicate, or aluminosilicate powder. During recent years, much research has been undertaken and AAMs have undergone many scientific advances [1-3]. The precursor materials of binders have also been expanded upon [4, 5]. In fact, any materials could serve as precursors for the binders provided that they contain proper amounts of silica and alumina. Waste materials or industrial by-products, such as GBFS [6-10], red mud [1, 11], fly ash [12-15], lignite bottom ash [16], recycled glass [17], ceramic dust waste [18], slag waste from metallurgical industries [19], etc. have been used as precursors to synthesise binders [20-22]. Among these different types of waste, fly ash is by far the most interesting, because of its suitable chemical composition, easy availability, and ready availability in a fine powder form [14, 17, 23-27].

Fly ash is known as pulverised fuel ash generated through the combustion of coal powders. It contains amorphous silica and alumina, and some calcium compounds, magnesia and ferric oxide, which varies noticeably for different coal minerals and combustion processes [28]. Although fly ash has found many different options to be put into practice, only a small fraction of the fly ash has been managed efficiently [25, 28]. It is a better choice for fly ash to be used as a resource in the fabrication of binders by alkaline activation, because: (I) fly ash embraces the alumina-silicate chain essentially needed by the geopolymerisation; (II) there are less strict requirements for fly ash, compared to those used in OPC and (III) fly ash can be effectively used, in which, as much as 80 % or even 100 % of the fly ash can be added to fabricate the binders. Therefore, fly ash-based alkaline activated materials (FAAMs) have undergone a rapid upswing since they were reported several decades ago [23, 25, 29-31]. The curing processes, the ratios of SiO_2/Al_2O_3, SiO_2/Na_2O, Na_2O/Al_2O_3, and CaO/Al_2O_3, the content of the alkaline activator, and the various fly ashes, have been extensively studied: however, the effects of the different parameters on the properties of AFFMs are complex and it is difficult to predict which one is more important [32-35]. Besides, for any of the parameters, different results or even the opposite results can often be observed. Thus, it is necessary to explore the fundamentals of how the parameters determine the properties of FAAMs. Although there are some review papers in the literature regarding AAMs in recent years, in which, the structural techniques, the precursors and their characteristics, silicate waste, the effects of fibres, chemical admixtures and mineral admixtures have been reviewed [2, 17, 36-47], there are few papers mainly aimed at the development of fly ash-based AAMs [43, 46]. In this paper, the effects of the components on the compressive strengths of AAMs prepared by fly ash as a main resource material were reviewed, in which, the ranges of the components in the binders and their fundamental roles on FAAMs were generalised.

DISCUSSION

The role of the alkaline activator

Alkaline activation is the first step in the fabrication of FAAMs. By the alkaline activation, Si^{4+}, Al^{3+} and other ions are dissolved from the fly ash and passed into the solution, therefore, the role of the alkaline activator is primarily important. At NaOH contents of 5, 6, 7, 8 and 10 % relative to the binder, compressive strengths of 9, 21, 30, 31 and 50 MPa at 28 days were observed, respectively [48]. In another FAAM cured at 70 °C for 24 h, as the Na_2O content increased from 4 to 8 %, the strength at 28 days was increased by 629 % [49]. Ryu et al. [50] reported that the strength at 28 days at an Na_2O content of 7.8 % was improved by more than 1.7 times compared to that at 6.3 % in an FAAM mortar. Similar results were also obtained by some other authors [51-57].

There were differences of opinions as to the most effective amount leading to the maximum compressive strength [48, 53, 58]. Fernandez-Jimenez et al. [48] proposed that the addition of Na_2O with 14 % of fly ash led to the optimum mechanical properties. In another FAAM paste, the optimal Na_2O content was 10 % of fly ash by mass [59]. According to the results of Williamson et al. [53], however, the FAAM mortar prepared with 4.5 % NaOH exhibited the highest strength at 28 days. Thus, it is difficult to identify the effectiveness of different Na_2O contents. In fact, as alkaline activators are added to the fly ash, the alkaline content can change the SiO_2/Al_2O_3, Al_2O_3/Na_2O and SiO_2/Na_2O ratios, as well as the W/S ratio. The increase in the Na_2O or NaOH content decreases the SiO_2/Na_2O and Al_2O_3/Na_2O ratios. If the alkaline activators include water glass, the SiO_2/Al_2O_3 ratio also changes. It seems that it is more reasonable to expect the compressive strength to vary with the SiO_2/Al_2O_3, or SiO_2/Na_2O ratio than by the NaOH or Na_2O content only.

Table 1. The components of the FAAM pastes and their properties.

Binders (g)				Activator (to binders) (g)			Curing process		SiO_2/Al_2O_3	Al_2O_3/Na_2O	Compressive strength (MPa)	Ref.
Total	FA[1]	CE[2]	S[3]	$NaOH^{(s)}$	$Na_2O{\cdot}nSiO_2^{(s)}$	W[4]	T (°C)	t (h)				
100	100	–	–	8.09	–	15.0	RT	–	2.88	1.62	4.0	[78]
100	100	–	–	10	–	40	60	24	3.65	1.10	10	[122]
100	100	–	–	10.78	–	30	75	24	3.03	2.75	45	[51]
100	100	–	–	8.09	–	15.0	80	90d	2.88	1.62	26.2	[78]
100	100	–	–	6.16	–	28.8	85	24	3.64	3.15	70.4	[48]
100	100	–	–	4.03	–	36.0	85	20	3.64	1.93	64	[155]
100	100	–	–	-	14.25	20.8	80	90d	3.33	3.36	58.4	[78]
100	100	–	–	-	19.9	30	80	6	6.25	1.06	46	[110]
100	100	–	–	1.88	16.8	20.6	RT	–	4.03	2.11	11.2@	[110]
100	100	–	–	7.1	10.0	22.9	RT	–	4.70	1.15	59.3	[59]
100	100	–	–	4.92	9.43	15.6	65	24	3.95	2.61	62.6	[60]
100	100	–	–	10.78	21.6	41.0	75	24	3.79	2.75	52	[51]
100	100	–	–	10.32	11.6	36.9	70	24	6.48	0.41	44.8	[168]
100	100	–	–	7.0	0.88	32.1	85	24	3.69	2.69	91.6	[48]
100	100	–	–	4.08	0.64	35.2	85	20	3.72	1.82	46.0	[155]
100	100	–	–	4.92	9.43	15.6	85	24	3.95	2.61	68.7	[60]
100	100	–	–	9.95	8.76	34.40	–	–	3.21	2.28	70	[154]
100	80	–	20	–	14.2	20.8	RT	–	3.48	2.99	77.4	[78]
100	50	–	50	–	17.6	36.0	RT	–	5.03	1.14	67.5	[117]
100	50	–	50	10	–	40	60	24	3.65	1.10	45	[122]
100	70	30	–	5.8	15.1	27.8	RT	–	3.44	1.66	36.94	[87]

Note: [1]Fly ash, [2]OPC, [3]GBFS, [4]Water. @: 7-day strength

Palomo et al. [60] demonstrated an FAAM cured at 85 °C for 24 h. The compressive strength of the binder reached nearly 70 MPa, as the SiO_2/Al_2O_3 and Al_2O_3/Na_2O ratios were 3.95 and 2.62, respectively. As the SiO_2/Al_2O_3 and Al_2O_3/Na_2O ratios changed to 3.87 and 3.13, respectively, the compressive strength of the binder decreased to 54.5 MPa. Ryu et al. [61] also explored the effects of the SiO_2/Al_2O_3, and Al_2O_3/Na_2O ratios in an FAAM. At an SiO_2/Al_2O_3 ratio of 4.13, the compressive strength was seen to be reduced by the increase in the Al_2O_3/Na_2O ratio, and the compressive strength was 43.1 MPa at 28 days at an Al_2O_3/Na_2O ratio of 1.94.

Škvára and Bohuněk [62] found that the Al_2O_3/Na_2O ratio should be less than 2.32 to obtain a high-strength FAAM. Fernández-Jiménez et al. [63] concluded that, when the SiO_2/Al_2O_3 and Al_2O_3/Na_2O ratios were 3.50 and 1.50, respectively, the strength of the FAAM reached 80 MPa. In the sample with an Al_2O_3/Na_2O ratio of 1.29 and 0.93, an SiO_2/Al_2O_3 ratio of 3.19 and 3.68, strengths of 72 and 31 MPa were achieved, respectively. The optimal SiO_2/Al_2O_3 and Al_2O_3/Na_2O ratios were 3.21 and 2.88 in a binder designed to yield a high compressive strength [64]. Temuujin et al. [65] found that the binder with an SiO_2/Al_2O_3 ratio of 4.6 and an $Al_2O_3/Na_2O = 1.14$ exhibited the highest compressive strength. Songpiriyakij et al. [66] proposed that the optimal SiO_2/Al_2O_3 ratio was 15.9 to obtain the highest compressive strength in an FAAM. Kovalchuk et al. [67] obtained a result that showed the optimum SiO_2/Al_2O_3 ratio was between 3.5 and 4.0. In another FAAM mortar, the optimal SiO_2/Na_2O ratio was between 1.0 and 1.5, in which the compressive strengths exceeded 50 MPa [68].

Tables 1-3 summarise the typical FAAM pastes, mortars, and concretes with different curing processes, SiO_2/Al_2O_3 and Al_2O_3/Na_2O ratios, in which, the possible maximal strength obtained at various fly ash systems is emphasised. The compressive strengths of the FAAMs are distinctly different in various fly ash systems as well as the SiO_2/Al_2O_3 and Al_2O_3/Na_2O ratios. It is difficult to identify which aspect plays the primary role. Details of the compressive strength versus the changing SiO_2/Al_2O_3 and Al_2O_3/Na_2O ratios are shown in Figures 1

Table 2. The components of the FAAM mortars and their properties.

Binders (g)				Sand (g)	Activator (g)			Curing process		SiO_2/Al_2O_3	Al_2O_3/Na_2O	Compressive strength (MPa)	Ref.
Total	FA[1]	CE[2]	S[3]		$NaOH^{(s)}$	$Na_2O\cdot nSiO_2^{(s)}$	W[4]	T/°C	t/h				
100	100	–	–	212.8	14	–	31.5	40	72	2.79	1.59	28	[68]
100	100	–	–	50	10	–	30	85	20	3.19	2.15	41	[63]
1600	1600	–	–	2400	220.8	–	739.2	–	–	3.76	1.33	13.8	[50]
100	100	–	–	212.8	–	29.1	37.0	40	72	3.84	1.49	58	[68]
100	100	–	–	200	8.07	5.67	31.3	60	24	2.90	2.29	45	[125]
100	100	–	–	212.8	3.2	26.6	36.3	40	72	3.84	1.49	58	[68]
24.1	24.1	–	–	65.0	0.45	4.0	4.45	RT	–	4.00	2.08	66.1 ± 3.3	[110]
1600	1600	–	–	2400	79.2	160	720.8	60	24	4.13	1.94	43.1	[50]
1200	1200	–	–	1800	92.9	150.5	324	80	24	4.43	1.62	65	[57]
100	100	–	–	200	8.09	15.7	43.2	65	48	4.13	1.30	38.3	[153]
306	306	–	–	1362	–	148	227	RT	–	4.58	0.70	21.29[&]	[152]
730	730	–	–	1178	32.9	86.6	172.5	RT	–	3.89	2.45	27.0	[139]
100	100	–	–	150	7.04	21.4	41.6	RT	–	5.67	0.72	36	[147]
100	100	–	–	275	7.25	7.71	25.0	RT	–	2.77	1.12	45.0	[140]
577	577	–	–	1348	33..3	52.1	254.6	RT	–	3.99	1.83	60	[55]
342	342	–	–	823.4	35.3	39.8	138.8	70	24	6.48	0.41	46.2	[168]
100	100	–	–	50	8.3	–	24.7	85	20	3.5	2.50	40	[21]
730	694	36	–	1141.5	37.1	97.3	194.0	RT	–	4.04	2.09	62.0	[139]
100	85	15	–	275	7.25	7.71	25.0	RT	–	2.86	1.01	57.8	[140]
100	70	30[$]	–	150	23.8	10.8	44.8	RT	–	6.10	0.56	45	[147]
100	90	–	10	200	8.07	5.67	31.3	60	24	2.98	2.15	41	[125]
579	463	–	116	1348	33.6	52.1	262.3	RT	–	4.13	1.64	60	[55]
450	225	–	225	1350	59.2	102	288.8	60	48	6.72	0.58	16.0	[112]
100	50	–	50	160	6.0	18.9	35.1	RT	–	4.76	1.00	108	[84]
100	80	–	20	160	6.0	18.9	35.1	RT	–	4.52	1.16	78	[84]

Note: [1]Fly ash, [2]OPC, [3]GBFS, [4]Water. $: CCR; &: 91-day strength and 2. From Figure 1, it can be seen that the changes in strength with the Al_2O_3/Na_2O and SiO_2/Al_2O_3 ratios are such that, although the absolute values of compressive strength vary in various fly ash systems, the relative values of the compressive strength in the same system exhibit a regular change with the changing Al_2O_3/Na_2O and SiO_2/Al_2O_3 ratios. There is a critical value of the SiO_2/Al_2O_3 ratio at about 4.20 to 4.30. The compressive strength is reduced with an increase in the Al_2O_3/Na_2O ratio when the SiO_2/Al_2O_3 ratio is above the critical value; while, the strength is increased with an increase in the Al_2O_3/Na_2O ratio as the SiO_2/Al_2O_3 ratio is below it.

From the data in Figure 2, it can be seen that the ranges of the Al_2O_3/Na_2O and SiO_2/Na_2O ratios are rather wide in the FAAM systems studied. The strength of the FAAMs varies from several MPa to more than 100 MPa. The SiO_2/Al_2O_3 ratio is between 2.0 and 8.0, and the Al_2O_3/Na_2O ratio is between 0.5 and 7.0. When the Al_2O_3/Na_2O ratio is larger than 4.0 or smaller than 0.5, the compressive strengths of the binders are usually lower than 30 MPa, regardless of the SiO_2/Al_2O_3 ratio. A high strength (above 42.5 MPa) is achieved when the SiO_2/Al_2O_3 and Al_2O_3/Na_2O ratios are 2.5 to 6.0, and 0.7 to 3.5, respectively; however, it seems that the

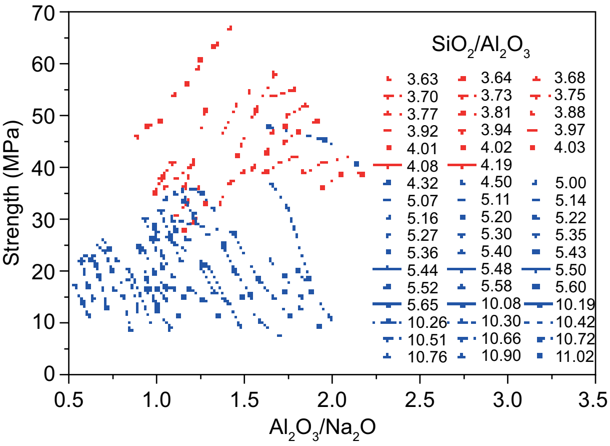

Figure 1. Compressive strength changes with the ratio of Al_2O_3/Na_2O at various SiO_2/Al_2O_3 ratios.

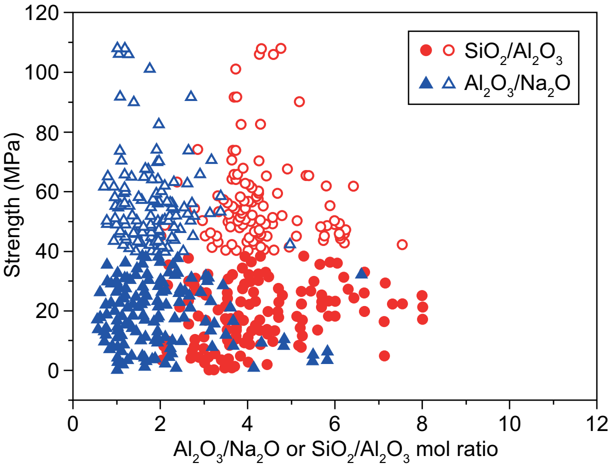

Figure 2. The strength map with the changes of SiO_2/Al_2O_3 and Al_2O_3/Na_2O ratios.

Table 3. The components of the FAAM concretes and their properties.

Binders (g)					Aggregates (g)		Activator (g)			A/S^{10}	Super plastciser (g)	Curing processes		Compressive strength/MPa	Ref.
Total	FA^1	CE^2	S^3	SF^4	F^5	C^6	A$^{(s)7}$	WG$^{(S)8}$	W^9			T (°C)	t (h)		
500	500	–	–	–	575	1150	25.4	70.9	128.7	0.22	6	70	48	51.63	[162]
400	400	–	–	–	554	1293	17.8	45.2	95.0	0.21	4	100	72	42.5	[163]
468	468	–	–	–	1043	828	52.8	–	157.8	0.30	–	85	20	50	[113]
400	400	–	–	–	651	1209	18.1	47.4	94.5	0.20	–	RT	–	27	[139]
400	400	–	–	–	651	1209	18.0	47.4	94.5	0.20	–	RT	–	27	[139]
440	440	0	0	0	723	1085	22.4	50.4	103.2	0.20	–	60	48	35.11	[161]
202	202	–	–	–	890	1373	6.1	46.0	75.0	0.29	–	60	24	56.4	[82]
100	100	–	–	–	–	–	4.0	11.0	20.0	0.17	–	60	24	67.6$^@$	[99]
381	293	–	88	–	760	1005	20.0	28.0	164.0	0.38	–	RT	–	37.4	[89]
400	280	–	120	–	652	1210	16	49.2	94.8	0.27	4	RT	–	59.3	[3]
381	293	–	88	–	760	1005	28.4	28.0	158.6	0.42	–	RT	–	37.4	[89]
400	360	40	–	–	651	1209	18.0	47.4	94.5	0.20	–	RT	–	35	[139]
515	500	–	–	15*	575	1150	25.4	70.9	128.7	0.21	6	70	48	48.4	[162]
466	440	0	–	26*	723	1085	22.4	50.4	103.2	0.19	–	RT	–	46.43	[161]
400	320	–	–	80	554	1293	17.8	45.2	95.0	0.21	–	100	72	65.0	[163]
400	240	–	–	160	554	1293	17.8	45.2	95.0	0.21	–	100	72	72.5	[163]

*Note: ^{1}Fly ash; ^{2}OPC; ^{3}GBFS; ^{4}Silica fume; ^{5}Fine aggregate; ^{6}Coarse aggregate; ^{7}NaOH (solid contents); ^{8}Waterglass (solid content); ^{9}Water, ^{10}Ratio of water to solids (sum of the mass of fly ash, sodium hydroxide, and sodium silicate solids); *nano-silica, @: 7-day strength, \$: 90-day strength*

strength is not directly related to the SiO_2/Al_2O_3 and Al_2O_3/Na_2O ratios. In some binders, a low strength is obtained, however, high strengths are acquired in other binders with similar SiO_2/Al_2O_3 and Al_2O_3/Na_2O ratios. Therefore, it is not enough to determine the strength by a simple combination of the SiO_2/Al_2O_3 and Al_2O_3/Na_2O ratios.

It could be seen that there are threshold values for the SiO_2 and Na_2O contents in the alkaline solution to guarantee the system has a high pH and a high level of soluble Al^{3+} and/or Si^{4+} ions [48]. A weak chemical reaction would occur [69] in a low-alkalinity solution due to the very low pH, leading to little strength development [48]. A sufficient concentration of the alkaline solution, which is effectively capable of dissolving fly ash particles and forming the gel phases, is necessary to make a binder reach a high compressive strength [48, 70-73]. If the alkalinity was low enough, binders with a relatively low strength of less than 4.0 MPa were obtained, even when cured at high temperatures [74, 75].

Based on an ion discharge balance, if the reactive Al_2O_3 content is 1.0 mol in the fly ash, it requires 1.0 mol Na_2O to balance the complete reaction of the fly ash; however, the optimal dosage of alkali in practice did not agree with the ideal values as published [76], as shown in Figure 2. The unreacted particles of the fly ash and the crystallinity were always sufficient to weaken the sample [77]. Therefore, in order to reduce the risk of efflorescence and carbonation at the surface of the binder, the extent of the reaction of the glassy phases in the fly ash should be considered [10, 78, 79]. The extent of the reaction of the amorphous phases in the fly ash was estimated at 30 % to 35 % [78]. Therefore, as low an alkaline content as possible is recommended by stoichiometrically matching it to the expected degree of the reaction [79].

In fact, only the ions available in the fly ash could interact with the alkali activators. Therefore, the amount of dissolved Al^{3+} and/or Si^{4+} ions and the Si/Al ratio in the precursor are the factors dominating the geopolymerisation [67]. This point has been widely recognised [28, 30, 44, 57, 60, 68, 80-86]. The reactive SiO_2 and Al_2O_3 phases, not that of the original prime material, are the most important [50, 48, 85]. Unfortunately, many current reports are insufficient in terms of the details of the reactive phases. Despite this, however, we estimated the role of the reactive components through the limited data. Figure 3 illustrates the effects of the SiO_2/Al_2O_3 and Al_2O_3/Na_2O ratios, as calculated from the reactive phases, on the compressive strengths of the FAAMs. The data are less scattered and the effect ranges are clearer, compared with those in Figure 2. The Al_2O_3/Na_2O ratio is in the range of 0.5 to 2.25, and the SiO_2/Al_2O_3 ratio has several ranges, most of which are 3.5 to 6.0, 6.8 to 8.6, or 10.0 to 11.2, respectively. Among these, the ranges, in which, a possible strength of more than 40 MPa is achieved, are 3.5 to 4.2 and 6.8 to 8.6 for the SiO_2/Al_2O_3 ratio and 0.6 to 2.10 for the Al_2O_3/Na_2O ratio. As the SiO_2/Al_2O_3 ratio exceeded 8.6, a lower strength was observed, regardless of the Al_2O_3/Na_2O ratio.

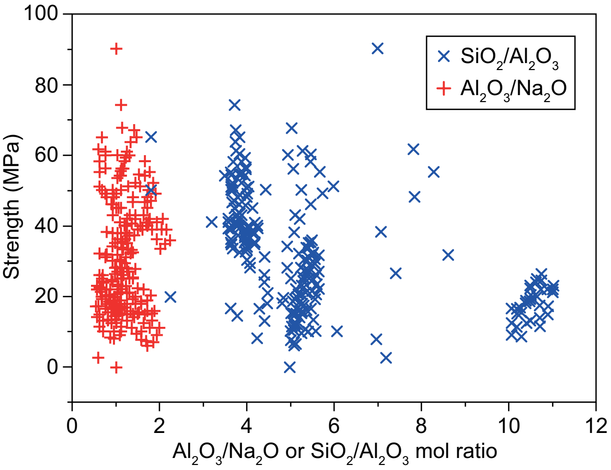

Figure 3. Compressive strength map with the changes of SiO_2/Al_2O_3 and Al_2O_3/Na_2O ratios.

From the above results, it could be seen that the effects of the SiO_2, Al_2O_3, Na_2O contents, and their ratios on the FAAMs are complicated. In fact, the situation is more complicated; so, at first, the alkaline activator has various types [87, 88]. There are many options for the choice of the alkaline activator, such as sodium hydroxide, sodium carbonate, sodium silicate, potassium hydroxide, water glass, etc. Also, the combination of sodium hydroxide and water glass was often used in the FAAMs: strengths of 13.1 MPa and 13.8 MPa were obtained as Na_2O was only added via NaOH or the water glass solution, these were much lower than those activated by the mutual addition of NaOH and water glass solutions [50]. Similar results were also reported elsewhere [60]. As only the NaOH solution (as the alkali activator) was added, a sample with a compressive strength of about 20 MPa was acquired; while the sample with more than 60 MPa could be achieved as the mixture of NaOH and the water glass solution with similar Na_2O contents was added. Second, there was a critical value in the content of the alkaline solution [51, 53, 89, 90]. A too high of an alkaline content in the FAAMs had the opposite effect on the development of the strength [51, 52, 60, 61, 91-93]. The reduction in strength was thought to result from excess silicates hindering the formation of the hardened binders or an increase in the coagulation of silica [90, 94, 95].

The role of water

Water not only dissolves fly ash, but participates in the geopolymerisation of FAAMs [48, 96, 97]. The water content plays an important role in determining the properties of the binders [65, 73]. In an FAAM paste made from “one-part binders”, the compressive strength was 65 MPa at 21 days at a W/S ratio of 0.34; while as the ratio increased to 0.392, the compressive strength falls to 50 MPa under the same curing process [45]. Another report also confirmed this result [98]. At a W/S ratio of 0.47, the compressive strength was 50 MPa. As the ratios were changed to 0.42, 0.31, and 0.26, the strength reached 63.5, 82.5 and 96.0 MPa. It was also found that an increased W/S ratio reduced the strength of FAAM concretes. At a W/S ratio of 0.174, the compressive strength at 7 days was 67.6 MPa in the sample cured for 24 h at 60 °C. By adding extra water, at W/S ratios of 0.197, and 0.220, the compressive strength of the samples decreased to 60 MPa and 45 MPa, respectively [99]. In another FAAM concrete fabricated from "one-part binders", at a W/S ratio of 0.47, the compressive strength was 50 MPa; while at the ratios 0.42, 0.31, and 0.26, the strength increased to 63.5, 82.5 and 96.0 MPa, respectively [98].

Lee et al. [100] investigated the difference in the compressive strengths of sealed and unsealed FAAM pastes. The average compressive strength at 28 days of the sealed binders surpassed 130 MPa, which was 30.8 % higher than that of the unsealed samples. It was clear that the sealed water promoted an increase in strength in the binders. The authors attributed this to a full reaction rather than carbonisation or the presence of zeolites. Water is consumed during the hydrolysis/dissolution of the source material and then regenerated by polycondensation processes and is then enclosed in the pores of the hardened binders. Water is presumed to accelerate the polycondensation rate [101]. In particular, the sealing was more important in "one-part binders", because water loss would be promoted if the binders were cured in an unsealed state [45].

When an insufficient amount of water is added, a low strength would be obtained due to the poor fluidity causing an uncompacted binder to remain in the mix. When an excessive amount of water is added, there is a sufficient mobility of ions in the mixtures for the solid to dissolve, resulting in a low level of unreacted material. Meanwhile, the addition of water at high levels can increase the porosity in the final binders [77], leading to a decrease in the strength of the binders [102, 103]. Therefore, the optimum W/S ratio is necessary to attain a high strength.

The role of calcium-based additives

The effects of calcium additives were extensively elucidated upon [55, 80, 104-112]. As these are added, Ca^{2+} ions could function as a network modifier to balance the negatively charged Si–O–Al– tetrahedron, leading to a more reactive fly ash. Meanwhile, it increases the CaO/Al_2O_3 and CaO/SiO_2 ratios in the binder resource, which induces the formation of C–A–S–H and N–A–S–H gels [112-116], resulting in a different chemical process and different chemical products [35, 118, 119]. GBFS is one of the most commonly used precursors to produce a calcium resource in FAAMs. GBFS has been used to fabricate AAMs as the main raw material for over a century [120] and GBFS-based AAMs have good mechanical and physical properties [10, 121, 122]. The addition of GBFS was found to be able to alter the geopolymerisation behaviour of fly ash to generate high strengths at ambient temperatures [62, 89, 122-125]. When the GBFS replacement of fly ash increased from 10 to 30 %, the compressive strengths of FAAM concretes at 28 days increased from 21.90 to 56.43 MPa, respectively [3]. In another FAAM concrete, as the replacement ratios of GBFS for fly ash were 10, 15 and 20 %, the compressive strengths at 28 days were 15.5, 23.0 and 29.9 MPa, respectively. If the optimal activator was added, binders with a compressive strength of 37.4 ± 4.04 MPa at 28 days could be obtained when the fly ash was replaced by 20 % GBFS [89]. Khan et al. [83] investigated an FAAM mortar: given a GBFS replacement of fly ash of 30 %, the compressive strength at 28 days was about 78 MPa, which was ca. 40 % better compared to that of the binder without the GBFS replacement. The compressive strengths of 108 MPa and 106 MPa at 28 days were achieved in the sample with 50 and 40 % GBFS replacements, respectively. A high strength of more than 170 MPa was reported by Škvára et al. [62] in a sample with a 50% GBFS replacement. Neupane [98] found an FAAM concrete with a 30 and 40 % GBFS replacement by "one-part binders", in which powdered activators were employed. Concretes with compressive strengths of 58 and 96 MPa at 28 days were obtained using a binder with 30 and 40 % GBFS replacement, respectively. It could be concluded that the addition of GBFS in fly ash increased the strength of the FAAMs.

The replacement of GBFS for fly ash might accelerate the fly ash dissolution [82] and induce the formation of a calcium-silicate-hydrate (C–S–H) gel [89, 122, 126-129], and a calcium-alumino-silicate-hydrate (C–A–S–H) gel [26, 119, 122, 126, 128, 130-133], besides an alumino-silicate-hydrate (A–S–H) gel. As the level of GBFS replacement was increased from 10 to 20 % in the binder, the main reaction product was N–C–A–S–H rather than N–A–S–H. After the amount of GBFS was increased to 30 %, C–S–H gels were formed as the main reaction products [134]. Meanwhile, the two phases (C–S–H and aluminosilicate) were not only combined, but they also interacted chemically [135, 136]. These factors promoted the strength development of the FAAMs with the GBFS replacement. In addition, as more GBFS was added, the formation of bonding hydrates, in particular the crystallinity of the calcium-based materials, which was similar to zeolites, was also responsible for the promotion of the beneficial mechanical properties in the FAAMs [35, 62, 106].

OPC is another calcium resource often adopted in FAAMs. It was proved that the replacement of fly ash with OPC could effectively improve the strength of FAAMs from the early age to the final days [137-140]. In an FAAM activated by a modified alkaline activator, by adding a silica fume to the sodium hydroxide solution, as the 15 % OPC replaced the fly ash, the early compressive strength at 1 day was ca. 5.0 MPa, which was improved by more than 50 % compared with that of the sample without the OPC. The compressive strength of a sample with 15 % OPC replacement reached 64.3 MPa at 28 days. As a comparison, the compressive strengths at 28 days were 27.2, 53.3, and 57.4 MPa in samples with 0, 5 and 10 % OPC replacement, which were 42.3, 83.2, and 89.3 % of that with 15 % OPC, respectively [75]. In another FAAM with naphthalene added to the alkaline solution, the strength at 28 days was 44.73 MPa in a binder without the OPC and increased to 54.76 and 64.46 MPa as the 10 and 20 % OPC were replaced by fly ash [141]. Similar results were observed elsewhere [87, 139, 140]. The replacement of OPC with fly ash ranged from 5 to 15 % and the compressive strengths of the binders increased by 15 % to 45 %.

As the OPC partially replaced the fly ash, heat is released during the hydration reaction of the OPC. This heat will accelerate the alkali activation reactions in FAAMs [87]. Moreover, the presence of calcium hydroxide resulting from the hydration of cement is believed to precipitate a C–S–H gel and calcium-riched alumino-silicate-hydrate (C–A–S–H) gels [121, 139, 143]. In addition, calcium provides extra nucleation sites [144, 145], which can also contribute to the alkali activation reaction of FAAMs.

Calcium carbide residue (CCR), a by-product of acetylene fabrication through the hydrolysis of calcium carbide, was also found to introduce calcium into FAAMs [116, 146]. It was found that the incorporation of CCR increased the compressive strengths of FAAM mortars. The compressive strength of FAAM (high calcium fly ash) at 28 days without the CCR was 35.6 MPa. As the CCR replacements for fly ash were 10, 20 and 30 %, the compressive strengths at 28 days increased to 41.8, 44.4, and 45.8 MPa, which were 17.4, 24.7 and 28.6 % higher than that of the FAAM without CCR, respectively [147]. It was implied that the additional formation of a C–S–H gel co-existed with N–A–S–H gels was formed due to the reaction of $Ca(OH)_2$ from the CCR with Si^{4+} and Al^{3+} from the fly ash [115,116]. It was also found that the replacement CCR for fly ash decreased the initial and final, setting times [147].

The calcium sources could be introduced by directly adding chemical agents containing calcium [148,149]. Zhao et al. [149] introduced $Ca(OH)_2$ to an FAAM. When ca. 8.9 % $Ca(OH)_2$ relative to the mass of the fly ash was added, the compressive strength of the binder at 28 days was 9.4 MPa, which is about seven times higher than that without the calcium additives. In another FAAM activated by an Na_2SiO_3 solution, a compressive strength of 22.9 MPa at 28 days was achieved in the binder with the 5.5 % $Ca(OH)_2$ additive, which is 5.6 times higher than that obtained with no-calcium additives. The addition of $Ca(OH)_2$ induced the formation of the gel product C–N–A–S–H, which contributed to the higher strength of the binders. Kim et al. [150] indicated that the addition of CaO was more effective than $Ca(OH)_2$. A sample with more than 40 MPa was achieved as 20 % CaO and 5 to 10 % $CaCl_2$ was added to an FAAM paste. The incorporation of $CaCl_2$ in the CaO-activated FAAM could induce the dissolution of the fly ash, leading to the formation of more C–S–H gels: however, the excessive addition of $CaCl_2$ was detrimental to the strength development due to the formation of calcium oxychloride [151].

From the above, it is concluded that the addition of calcium in FAAMs leads to a higher strength, whether the calcium resource came from GBFS, OPC, or other calcium-rich materials. Thus, it is not enough to only consider the effects of the SiO_2/Al_2O_3 and Al_2O_3/Na_2O ratios on the compressive strength in the calcium-rich FAAMs. Yang et al. [152] introduced an index – the alkali quality coefficient (QA) – combining the components of an activator and the source materials. The QA was proposed as $Na_2O/SiO_2 \cdot Al_2O_3/SiO_2 \cdot CaO/B$, where B was total mass of the solids in the binders. They thought that the QA could be used to evaluate the properties of AAMs more effectively than each single parameter. We collated the data from high calcium-content FAAM systems, based on the equation for QA and the strength map is demonstrated in Figure 4: no direct relationship with the compressive strength can be seen, indicating that the QA introduced by Yang et al. [152] has its limitations in anticipating the strength of FAAMs. As seen in Figures 1 and 2, the compressive strengths of the binders did not keep increasing with an increase in $Na_2O/SiO_2 \times Al_2O_3/SiO_2$. Meanwhile, due to the large differences among the fly ash systems adopted by various authors, the final strength of the FAAMs caused by the effects of the fly ash also varied. Assuming the increased strength in a binder was caused by the active CaO introduced by the calcium-rich resources, we re-calculated the

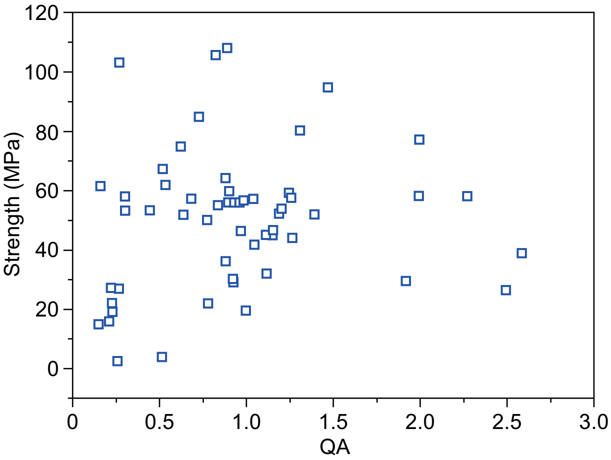

Figure 4. Compressive strength map with the QA changes.

compressive strength by eliminating the effects of the fly ash itself and only considered the active calcium and its promotion of the compressive strength (the active CaO replaced the total CaO and the increased strengths replaced the absolute strengths). The results are shown in Figure 5: the extra-strength developed by introducing active CaO was in line with the CaO content. With an increase in the active CaO content, the strength of the binders increased in an overall trend, except for the data on the bottom right corner in Figure 5 (it was found that the SiO_2/Al_2O_3 ratio was relatively large in these cases, at 6.7 and 7.2, respectively). Returning to the effects of the SiO_2/Al_2O_3 ratio, as shown in Figures 2 and 3, it can be seen that a too large an SiO_2/Al_2O_3 ratio has an opposite effect on the strength, therefore, it could be understood why the low-strength areas in Figure 5 could have arisen.

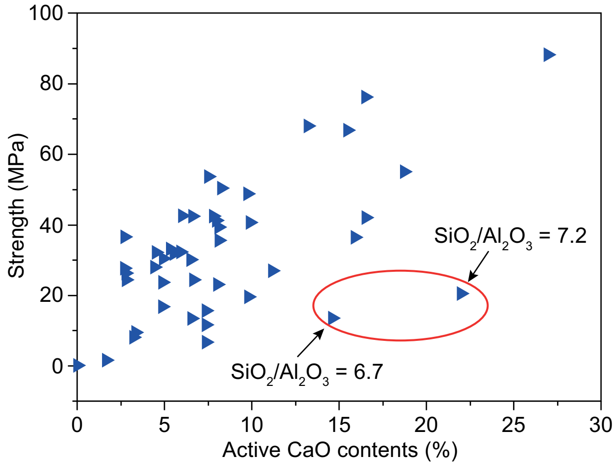

Figure 5. Compressive strength map with the active CaO contents.

The addition of calcium to the fly ash enhances the compressive strengths of FAAMs, implying that it induces the formation of a binder gel, besides this, it balances the ionic charges as a network modifier. However, some opposing views have been proposed: it seems that the calcium source from the fly ash was less effective in increasing the strength [153, 154]. The addition of calcium could not influence the mechanical strength, and the addition of too much calcium would even reduce it [83, 155]. The compressive strength of the binder with fly ash only at 90 days was 55.3 MPa, but fell to 25.4 MPa when 50 % of the fly ash was replaced by GBFS [32, 156]. Similar results were also observed in samples with the OPC replacement and the addition of $Ca(OH)_2$ [148, 157]. The excessive addition of Ca^{2+} ions to the binders might change the N–A–S–H and C–N–A–S–H gels to a C–S–H gel, decreasing the compressive strength at 28 days [35, 83, 106, 157-159]. Lee et al. [89] proposed that a calcium addition by GBFS induced a crack evolution, leading to a decreased compressive strength. The optimal addition of 7.5 to 10 % OPC and 15 to 20 % GBFS replacement was recommended by considering the setting time, workability, and strength development [32, 75, 129].

The role of silica

The use of silica in FAAMs was found to enhance the strength at ambient temperatures due to the promotion of the dissolution of Si^{4+} and/or Al^{3+} ions and the transformation of the crystalline phases in the binders [20, 58, 75, 160-164]. In an FAAM concrete, the compressive strength was about 70 MPa at 28 days in samples with 40 % silica fume as a replacement for the fly ash, which increased by 64.7 % when compared with that of the samples without the silica fume, i.e., ca. 42.5 MPa [164]. Rakngan et al. [76] reported that the effect of the silica fume was more effective as it was dissolved in the alkaline solution more than was replaced by fly ash. As 4.7 % silica fume by mass of the fly ash was added to the 4 M NaOH solution, the compressive strength of the binder at 28 days was 40 MPa, which was 61.2 % higher than that without the silica fume. Phoo-ngernkham et al. [165] reported that adding nano-silica to a high-calcium fly ash induced the formation of additional C–S–H or C–A–S–H and N–A–S–H gels in the binders, increasing the strength of the FAAMs. Criado et al. [166] concluded that the addition of a soluble silica modified the reaction kinetics and prompted the formation of more Si-rich gels. Adak et al. [161,164] reported that a colloidal nano-silica increased the strength. The compressive strength was 46.43 MPa in the FAAM concrete with a 6 % nano-silica, some 11.32 MPa higher than that of the sample without the additives. The FAAM paste with a 6 % nano-silica addition reached a compressive strength of ca. 48 MPa, which was 27 % higher than that of the sample without the nano-silica. Another report details a 28-day compressive strength of an AFFM paste containing 1 % nano-silica of 31.8 MPa, in comparison, the compressive strength of the paste without the nano-silica was 29.6 MPa [163].

On the contrary, contradictory results show the opposite effects of the nano-silicas: Okoye et al. [163] observed that, for the addition of a nano-silica of above 2 %, an adverse effect began on the strength of the binder. The compressive strengths at 28 days decreased to 31.4 and 29.7 MPa, as a 2 % and 3 % nano-silica by mass were added. Çevik et al. [162] also observed that the compressive strength decreased as a 3 % nano-silica was added, compared with that of the samples without the nano-silica. The authors attributed this to the unreacted nano-silica particles, which caused excessive self-dehydration and cracks in the binders.

The role of iron

To date, the role played by iron oxide in FAAMs is rarely investigated and generally accepted agreements about the effects of iron oxide are not recognised [44]. Fernandez-Jimenez and Palomo [155] claimed that the addition of iron had no influence on the mechanical strength because they did not find iron-containing phases in the main reaction products. The low iron content in fly ash positively influenced the reactivity of the fly ash [56, 57]; however, in calcium-rich FAAM structures, calcium silicate hydrate gels containing iron were found [57]. It seemed that iron played a positive role in the formation of FAAMs [57] but, the interaction between the alkali activator and the iron in fly ash and the final location and coordination in the FAAM structure were still not fully understood [44]. However, most of the fly ashes used as FAAM resources contain iron, and some are iron rich [44, 139, 147, 168, 169], making it necessary to investigate the effect of the iron and its reaction mechanism.

CONCLUSION

The components of binder resources have significant effects on the compressive strengths of FAAMs. The optimal SiO_2/Al_2O_3 and Al_2O_3/Na_2O ratios vary with the differences in the fly ash used. The appropriate amounts of the components of the binders are summarised, in which, the ranges of the SiO_2/Al_2O_3 and Al_2O_3/Na_2O ratios are 1.5 to 3.5 and 0.7 to 1.4, respectively. An active CaO addition proved to be an effective option to enhance the strength. In general, the strength of the binders increases with a modified QA, however, the effect of the active CaO is subject to interference from those of the SiO_2/Al_2O_3 and $Al_2O_3/$ $/Na_2O$ ratios. The roles of iron and its possible reaction in FAAMs are not yet fully understood. Considering there is iron present in almost all fly ashes, and some fly ashes are iron rich, the effect of the iron content on FAAMs warrants further investigation.

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