



Unveiling the Influence of Varied Alumina Sources on Fresh Properties of Ordinary Portland Cement Mortar and Concrete: A Comprehensive Review

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Abstract

The fresh properties of ordinary Portland cement (OPC) have garnered significant consideration due to their significant influence on the physical and mechanical behavior of OPC under normal and aggressive media. While numerous research articles and reviews have explored the influence of several organic and inorganic additives on the fresh properties of OPC, there is a limited focus on comprehensively understanding the impact of the chemical reaction mechanism of these additives on the early performance of OPC. Therefore, this study aims to examine the impact of alumina from different sources, such as supplementary cementitious materials (SCMs), calcium aluminate and calcium sulfoaluminate cement (CAC and CSAC, respectively), and chemical admixtures, on the hydration heat, setting times, and workability of PC. After analyzing the data published in the literature, it has been discovered that the impact of alumina on the fresh performance of OPC primarily relies on the reactivity of SCMs, superplasticizer (SP) content, and water demand. However, other alumina-rich materials demonstrate an increase in the initial flow and hydration rate, which is accompanied by workability loss and setting time shortening. The authors have also thoroughly discussed the reaction mechanism of each alumina source. Finally, the missing topics and proposed future directions for this research were identified. From the literature survey, it is recommended that the synergy between alumina and sulfate, as well as alumina and carbonate, is important in controlling the hydration reaction of Portland cement.

Keywords: Chemical admixture; Ettringite; Fineness; Hydration heat; Setting time; Workability.

Received: 20 December 2023; Revised: 19 January 2024; Accepted: 15 February 2024.

Article type: Review article.

1. Introduction

Portland cement (PC) is considered the greatest widely used cemented material in concrete making. It is synthesized by burning a mixture of clay and calcite in a rotary kiln at around 1460 °C, followed by cooling and grinding with 3-5% gypsum.^[1] Based on its chemical and phase composition, PC can be classified into several types, including ordinary Portland cement (OPC), sulfate resistance cement (SRC), calcium sulfoaluminate cement (CSAC), calcium aluminate cement (CAC), and belite cement (BC).^[2] The fresh properties of PC are of high importance as they strongly affect the physicomaterial properties and resistance of concrete

against aggressive attacks.^[3] This mainly refers to the plastic behavior of concrete/mortar in its plastic state before setting and hardening.^[4] The properties of fresh concrete strongly influence the construction process as they govern the ease of mixing, transportation, and placement of the concrete in the intended location.^[5] The hydration heat, flowability, and setting time are the main key parameters that determine the fresh performance of concrete. The fresh performance of OPC-concrete is strongly influenced by the fineness of the cement, the type and dosage of inorganic and organic chemical admixtures, the mixing water content, and the temperature.^[6-8] It is a widely known fact that OPC primarily consists of four main phases, namely tricalcium silicate (C₃S), dicalcium silicate (C₂S), tricalcium aluminate (C₃A), and tetracalcium ferro-aluminosilicate (C₄AF).^[9] C₃A is considered the main phase, which is primarily responsible for the setting process of OPC.^[2] As mentioned in previous studies, CAC and CSAC, with higher alumina content compared to PC, were found to exhibit higher heat of hydration, faster hydration rate, lower

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workability, and shorter setting time.^[10,11] Additionally, replacing OPC with CAC and CSAC strongly influenced its performance.^[12,13] The influence of supplementary cementitious materials (SCMs) rich in alumina, such as bauxite, fly ash (FA), metakaolin (MK), blast-furnace slag (BFS), and red mud (RM), on the fresh performance of OPC was also extensively studied.^[14–17] It was found that the fresh properties are mainly influenced by the chemical composition, physical properties of particles, mix design, and, especially, alumina and reactivity of SCMs.^[6,18–20] This indicates that the alumina within the OPC system demonstrates a cyclical role in the hydration performance of OPC concrete/mortar.

However, numerous inquiries persist regarding the effect of various alumina sources on the initial hydration and the fresh properties of PC. Understanding the role of alumina content and sources in the cementitious system can strongly address many issues in the fresh state of PC concrete. Without a comprehensive understanding of the impact of alumina content and sources on the fresh properties of PC-concrete, these challenges become more prevalent as the utilization of various alumina sources expands. Accordingly, this review focuses on the role of alumina sources, such as SCMs, CAC, CSAC, and chemical alumina-rich admixtures, on the heat of hydration, workability, and setting time of PC-mortar/concrete. This study also sheds more light on the relationship between the alumina source and the mechanisms of retardation and acceleration of the initial hydration rate of PC.

2. Impact of alumina-rich materials on the fresh performance

The fresh performance of OPC mainly depends on the source and content of alumina. Alumina can be derived from several aluminosilicates, such as MK, FA, BFS, RM, CAC, and CSAC. Alumina can also be derived from chemical admixtures like nano alumina (nA) and alum (aluminum sulfate: $\text{Al}_2(\text{SO}_4)_3$) – based accelerator. The role of alumina in the fresh properties primarily depends on its origin.

2.1 Impact of alumina sources on hydration heat of OPC

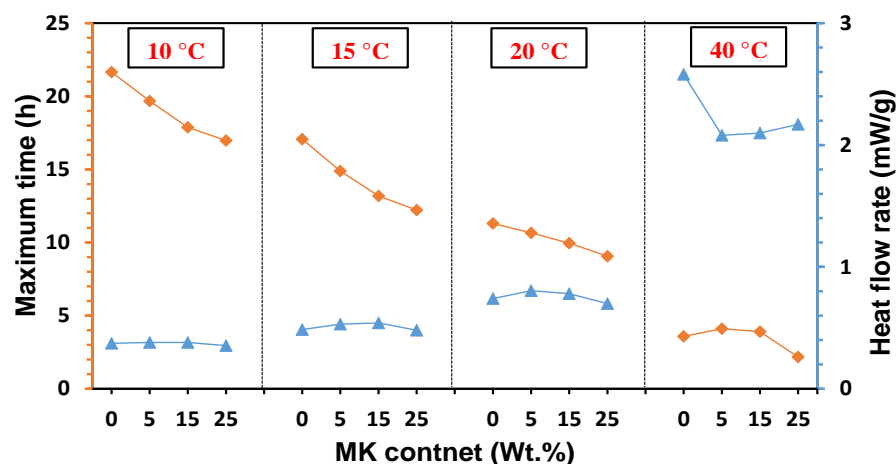


Fig. 1 Hydration heat of OPC-MK composites as a function of temperature. Reproduced with the permission from [37], Copyright 2023 Akadémiai Kiadó, Budapest, Hungary.

2.1.1 SCMs

The heat of hydration is one of the essential parameters that affect the fresh performance of OPC mortar/concrete.^[21,22] During the first three days of hydration, approximately 50% of the total heat is released, followed by 75% in seven days and 90% in 180 days.^[23] The heat of hydration curve of OPC shows three main peaks; the first one is associated with the wetting of the cement and the hydration of the C_3A phase, while the second and third stages correspond to the hydration of C_3S and the formation of CSH.^[24–27] The liberated heat from OPC hydration primarily depends on the cement composition, mixing water content, and admixtures incorporated.^[28,29]

SCMs such as MK, FA, and BFS are considered good sources of alumina, which strongly affects the heat flow rate of OPC.^[30,31] As reported by Jiang *et al.*,^[32] replacing OPC with desirable MK mass improves the hydration heat and shortens the time of occurrence. They found that incorporating 6 wt.% MK accelerated OPC cement hydration and significantly increased the cumulative heat. However, higher MK content up to 14 wt.% delayed the occurrence of the acceleration period and decreased the relevant heat of hydration peak. Other studies have also shown that the OPC-MK system released comparable or slightly higher heat of hydration than that of OPC.^[33,34] According to Tang *et al.*,^[35] the addition of 15 wt.% MK to limestone blended OPC caused a substantial acceleration in the rate of C_3S hydration. The peak related to the acceleration stage appeared after adding MK, which is primarily linked to the interaction of the dissolved alumina from MK with Portlandite, stimulating the creation of an expansive ettringite phase.^[36] Additionally, Zunino and Scrivener^[31] stated that increasing alumina content (by increasing MK content) increased the demand for sulfate and resulted in the shortening of the main hydration occurrence.

A recent study by Leclou and Das^[37] investigated the hydration heat released by OPC-MK (at weight ratios of 100-0, 95-5, 85-15, 75-25) at various temperatures (10, 15, 20, and 40 °C). Regardless of the MK content, the results depicted in Fig. 1 demonstrate that raising the curing temperature has resulted in an increase in both the flow rate and the

acceleration of the hydration time. Additionally, the mixtures with 5 to 15 wt.% MK showed the highest flow rate, which occurred in a shorter time than that of the reference sample. MK provides the hydration medium with reactive alumina and silica, inducing the formation of calcium aluminosilicate through a pozzolanic reaction^[37,38] However, increasing the content up to 25 wt.% decreased the released heat significantly, which could be attributed to the dilution effect of MK.^[39]

The role of low and high alumina BFS in the heat of hydration of OPC was evaluated by Whittaker *et al.*^[40] They have individually replaced OPC with 40 wt.% of two types of BFS containing different alumina. Regardless of the variation in alumina content, incorporating BFS accelerated the heat flow rate of the C₃A phase but had no effect on the hydration time. Additionally, a new hydration peak related to the hydration of alumina inside the slag was detected when PC was replaced by BFS. The occurrence time of this peak shortened, and the cumulative heat increased with an increasing alumina content inside BFS. The same trend was observed by Moula *et al.*^[41] This could be ascribed to synergistic three reasonable reasons: firstly, increasing alumina inside BFS enhances the potential formation of ettringite and monocarboaluminate, which increases the heat flow rate.^[31,40,42] Secondly, the hydration of BFS itself is exothermic.^[43] Finally, BFS acts as a filling/nucleating effect, which promotes the cement hydration rate accompanied by increasing heat evolution.^[44]

The extent to which BFS influences the heat of hydration primarily relies on factors such as the composition and properties of the cement, as well as the water-to-powder (W/P) ratio and curing temperature. Yang *et al.*^[45] found that replacing high-ferrite PC with 20, 40, and 60 wt.% BFS at a constant curing temperature of 20 °C and W/P ratio of 0.50 decreased the heat evolution and elongated the occurrence time of peak II related to the formation of CSH and renewed ettringite formation.^[25] In contrast, replacing OPC with 20 wt.% BFS at the same conditions increased the heat flow rate of the second peak related to C₃S hydration and shifted it to a lower time (from 32.5 h to 26.5 h). Further incorporation of BFS up to 60 wt.% decreased the heat of hydration significantly while still accelerating the C₃S hydration.^[45] Regardless of the cement type, increasing the curing temperature up to 60 °C accelerated the occurrence of the main hydration peak, accompanied by an increase in the heat flow rate.^[45]

Replacing BFS with 30, 50, 75, and 100% red clay brick powder (RCBP) inside PC-BFS decreased the total hydration heat Zhao *et al.*^[46] Specifically, the OPC-BFS-RCBP composites at weight percentages of 70-30-0, 70-15-15, and 70-0-30 decreased the total heat of the control OPC, which was 234.2 J/g in 144 h, by 4.1%, 11.6%, and 21.3%, respectively. This means that RCBP has a higher retardation effect on the hydration of OPC, which is mainly due to its higher crystallinity compared to BFS. Nevertheless, the released hydration heat of PC-FA is much lower than that of the OPC-BFS system.^[18] Similar findings were detected by

other works.^[18,47] This indicates the low FA hydration reactivity, mainly caused by its spherical morphology, leading to the release of low reactive aluminosilicate content.^[48] The opposite result was highlighted by Wang *et al.*^[49] as the hydration of OPC-FA released cumulative heat higher than that of the OPC-BFS. This could be linked to the difference in the composition and surface area of BFS.^[50] Bauxite, as alumina-rich waste, was found to accelerate the hydration of BFS-blended OPC accompanied by an increase in the heat flow, mainly attributable to the formation of alumina-rich phases such as calcium aluminate hydrate and ettringite.^[15,51]

2.1.2. CAC and CSAC

Khan *et al.*^[10] proved that the content of alumina inside the PC played an important role in the hydration heat of the resultant fresh paste. They found that the neat CAC exhibited a higher heat flow rate and heat peak intensity at the initial stage of hydration than that of OPC, which is mainly linked to the hydration of aluminate phases accompanied by ettringite formation.^[52,53] However, the occurrence time of the induction period (10 h) was longer than that of OPC (6 h).^[10] Similarly, the use of CAC as a substitute for OPC resulted in a synergistic elongation and reduction in the hydration heat of OPC mortar.^[12,54] This delay in hydration reaction is mainly ascribed to the retardation of C₃S hydration.^[5] Pustovgar *et al.*^[55] proved that the retardation mechanism of C₃S is mainly performed by the adsorption of aluminate on the hydroxylated C₃S via a strong ionic bond with calcium. This is also the main reason why CAC released a total hydration heat lower than that of OPC.^[10] The same trend was observed when OPC was replaced by 5 to 30 wt.% CSAC.^[13,56–59] Trauchessec *et al.*^[60] found that the main reason behind the delay in the main hydration of OPC (C₃S hydration) was the fast initial hydration and the consumption of water through the creation of the alumina-containing phase, *i.e.*, ettringite phase. Other researchers stated that the delay in C₃S hydration is mainly attributed to the poison effect of aluminate on the CSH nucleation.^[61]

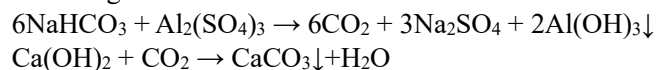
2.1.3. Chemical admixtures

The role of chemical admixture type and content on the heat of hydration of OPC is summarized in Table 1. Nano alumina (nA) is an oxide that exhibits amphoteric properties, meaning it is not soluble in water at normal conditions, but it demonstrates significant solubility in both acidic and alkaline environments.^[62] As is well-known, hydrated OPC exhibits high alkalinity (pH over 12.5). Accordingly, nA was found to play a vital role in the hydration heat of OPC. The isothermal calorimeter showed that adding nA increased the heat flow rate corresponding to C₃A hydration and the creation of ettringite.^[63] Adding 5 wt. % nA significantly increased the cumulative heat released during OPC hydration.^[22] This was explained by the role of nA in increasing the surface area of the cementitious system, leading to improved nucleation and growth of hydration products,^[64,65] Moreover, nA provides the

pore solution with reactive Al, which speeds up the C₃A hydration rate via increasing the intensity of the peak of hydration heat.^[66]

Adding alum (Al₂(SO₄)₃) also enhanced the early hydration rate, accompanied by an increase in the heat flow rate and accumulation of heat of hydration.^[67] Specifically, adding 6 wt.% (Al₂(SO₄)₃) induced the formation of an initial hydration peak, which was not observed in the control sample. Additionally, it increased the heat released during the acceleration stage in a short time (15 h) compared to that of the reference sample (30 h). They also stated that Al₂(SO₄)₃ exhibited high efficiency in the acceleration of cement paste during the first 12.5 h, while it represented a lower effect after this period. Tan *et al.*^[68] stated that Al₂(SO₄)₃ provides the cement matrix with dissolved alumina and sulfate, which interact with Ca(OH)₂ to yield ettringite. The latter acts as nucleating seeds for enhancing the CSH formation through the acceleration of C₃S hydration. The production of ettringite consumes much content of calcium, which in turn induces the hydration of C₃S to provide the system with dissolved calcium.

Ettringite not only accelerates the hydration of C₃S but also retards the crystallization of Ca(OH)₂. Liu *et al.*^[69] found that the combination of sodium bicarbonate (NaHCO₃) and Al₂(SO₄)₃ not only increased the heat released from cement hydration but also accelerated both initial and main hydration rates. Incorporating both NaHCO₃ and Al₂(SO₄)₃ induced the formation of CaCO₃ through the consumption of Ca(OH)₂ resulting from the cement hydration according to the following reaction:



CaCO₃ can act as a nucleating site to accelerate the rate of ettringite formation, which in turn increases the heat flow rate during the early hydration of cement.^[70,71]

The influence of Al₂(SO₄)₃-based accelerator on the rate of cement hydration depends mainly on its content and composition. Wang *et al.*^[5] found that incorporating an accelerator admixture composed of Al₂(SO₄)₃, magnesium sulfate, and di-ethanolamine significantly increased the heat flow rate and accelerated both the initial and main hydration

Table 1. Effect of chemical admixtures on the hydration heat of OPC.

Reference	Type of chemical admixture	Content of chemical admixture, Wt. %	Observation
Li <i>et al.</i> ^[63]		2, 4, and 6	The addition of nA not only led to an increase in heat flow rate but also accelerated the onset of the second peak in heat flow, which is associated with the hydration of C ₃ A.
Zhou <i>et al.</i> ^[22]	nA	5	
Tan <i>et al.</i> ^[68]		4	Al ₂ (SO ₄) ₃ enhances ettringite formation by providing the cement matrix with dissolved aluminate and sulfate. This, in turn, led to an increase in the heat flow rate of the first hydration peak and accelerated the hydration of C ₃ A.
Liu <i>et al.</i> ^[67]	Al ₂ (SO ₄) ₃	6	This accelerator increases the heat flow through its interaction with Portlandite released from OPC to yield CaF ₂ and MgF ₂ nucleating agents. Additionally, the aluminate contained within the accelerator enhances the formation of ettringite, which acts as a seed for CSH formation. This overall process is associated with the acceleration of C ₃ S hydration.
Yang <i>et al.</i> ^[72]	Al ₂ (SO ₄) ₃ -dissolved mother liquid composed of 53.6% water, 25% magnesium fluosilicate, 2.4% organic acid, 8% alcohol amine, 5% urea, and 6% nanosilica	7 and 9	The addition of this accelerator resulted in a significant increase in the heat flow rate of OPC. The accelerator agent also accelerated the hydration of C ₃ S by forming CaCO ₃ , which acts as a nucleating site for enhancing ettringite formation. Notably, the sample with a 3 wt.% addition of this admixture exhibited the highest cumulative heat.
Liu <i>et al.</i> ^[69]	Al ₂ (SO ₄) ₃ -NaHCO ₃ (1:6 molar ratio)	1, 3, and 5	Increased the heat flow rate of the first hydration peak and promoted the hydration of the C ₄ AF phase at early ages.
(Wu <i>et al.</i> ^[47]	Al ₂ (SO ₄) ₃ -dissolved in Tri-isopropylamine and Tri-ethanolamine	7	Increased the heat of hydration of the first and second peaks, which is associated with the acceleration of C ₃ A and C ₃ S hydration.
Wang <i>et al.</i> ^[5]	Al ₂ (SO ₄) ₃ -MgSO ₄ dissolved in di-ethanolamine and phosphoric acid	2, 4, 6, 8, and 10	

stages. Additionally, the final width at half maximum of peaks associated with the main hydration stage decreased with the incorporation of the $\text{Al}_2(\text{SO}_4)_3$ accelerator at levels ranging from 2 to 10 wt.%. The same trend was observed when $\text{Al}_2(\text{SO}_4)_3$ and alkanolamine were used as accelerators for cement hydration.^[47] The use of an $\text{Al}_2(\text{SO}_4)_3$ dissolved in a mother liquid composed of magnesium fluorosilicate and 2-amino-2-methyl-1-propanol was also found to significantly accelerate the initial and main hydration periods and increase the heat flow rate at both stages.^[72]

The formulated accelerator enhances the initial hydration of cement through the induction of the creation of monosulfate and ettringite. It also accelerates the hydration of C_3S through three synergistic mechanisms: Firstly, decreasing the pH of the hydration medium, promoting the dissolution of calcium ions and hydrosilicate on the surface of C_3S , which results in the release of CSH coat on the surface of C_3S . Secondly, providing the cement system with dissolved magnesia and fluoride, which reacts with dissolved calcium from cement and magnesium from the accelerator to yield crystalline CaF_2 and MgF_2 nuclei with high efficiency, thus reducing the nucleation barrier of portlandite and accelerating the C_3S hydration.^[72]

Overall, the OPC hydration rate depends mainly on the source of Al-rich materials (see Fig. 2). Replacing OPC with a desirable content of MK, BFS, and FA increased hydration heat and shortened the occurrence time of the main hydration peak. However, a further addition of these materials decreased the heat flow rate significantly while still accelerating the rate of C_3S hydration in special cases. The impact of SCMs in enhancing the OPC hydration rate can be ordered as follows: $\text{MK} > \text{BFS} > \text{FA} > \text{RCBP}$. In some cases, FA showed higher reactivity than BFS, indicating the importance of the physical properties and chemical composition of SCMs on OPC hydration rate. Apart from their content and curing conditions, CAC and CSAC increased the initial hydration heat flow rate;

however, they exhibited a retardation effect on C_3S hydration. As a chemical admixture, nA increased the flow rate of both the first and second hydration peaks while also accelerating the C_3A hydration. $\text{Al}_2(\text{SiO}_4)_3$ and $\text{Al}_2(\text{SiO}_4)_3$ -based accelerators provide the pore solution with dissolved alumina and sulfate, which in turn increases the OPC hydration heat at the initial and main stages and shortens the occurrence time of the second peak associated with the heat flow rate of C_3S hydration.

2. 2. Effect of alumina sources on the workability of OPC concrete/mortar

In this section, particular attention is given to studying the impact of alumina from various sources (SCMs, CAC, and CASC, and chemical admixtures) on the workability of the fresh OPC-based cementitious materials.

2.2.1. SCMs

Generally, the substitution of OPC with MK resulted in a decrease in the flow rate of the fresh mortar.^[14,73,74] Apart from the high surface area of MK, the alumina content within MK played a crucial role in determining the workability of the fresh mortar. Ferreiro *et al.*^[6] noted that an increase in the alumina content negatively impacted the fluidity of the fresh mortar, even with the addition of superplasticizer (SP). This can be explained by considering that MK with high alumina content is similar to 1:1 calcined clay, where a layer of tetrahedral silica is interconnected with another layer of octahedral alumina, resulting in an open and long-range ordered structure.^[75] This type of MK has a strong affinity to adsorb SP within the open structure, reducing the efficacy of SP in compensating for the loss of workability despite providing a good dispersing effect. On the other hand, the flow rate of cement mortar containing 2:1 MK with low alumina

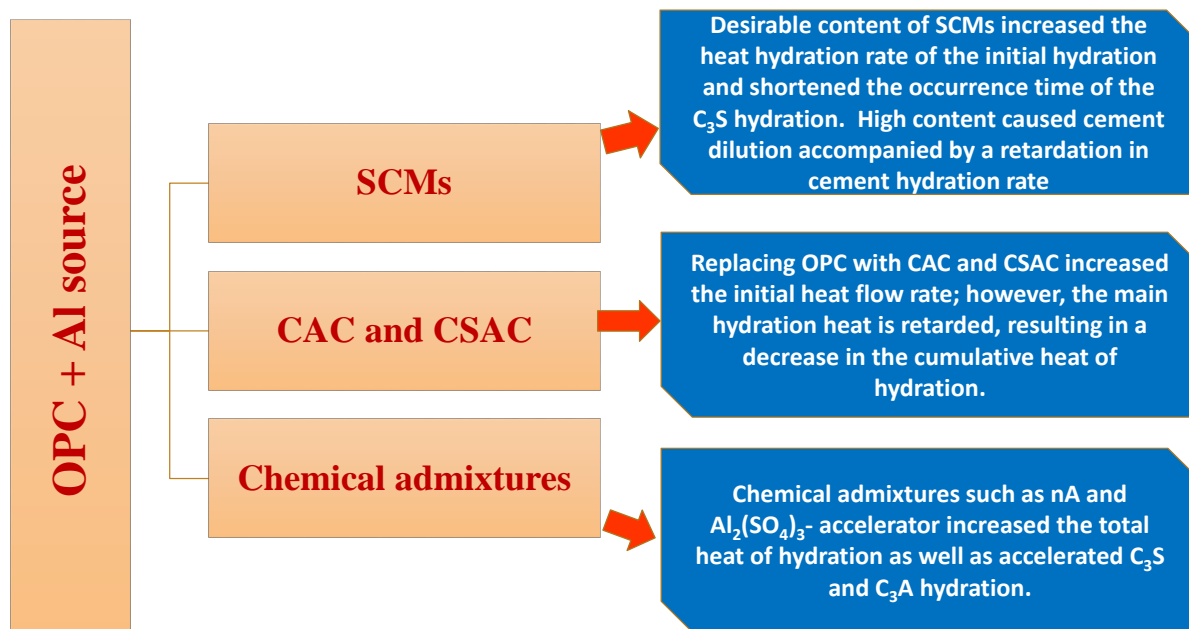


Fig. 2 Flow diagram summarizes the impact of aluminate sources on the hydration heat of OPC.

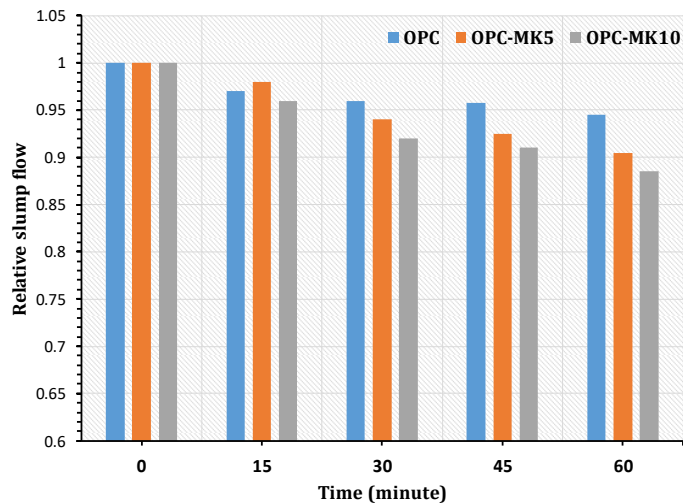


Fig. 3 Impact of MK content on the relative flow of 3D printed concrete at different times.

content significantly increased with the addition of SP.^[6] This effect can be attributed to the formation of a multilayered structure within low alumina MK, which allows for the adsorption of SP in the grain boundaries of aluminosilicate, blocking the interlayer of the pseudo-laminar structure.^[76] This reduces the water demand and greatly enhances the effectiveness of any SP addition in providing an excellent dispersion effect. In general, the presence of alumina in MK plays a crucial role in influencing the organization of aluminosilicate, which directly impacts the effectiveness of SP in obstructing interlayer spaces, reducing water requirements, enhancing dispersion efficiency, and ultimately improving workability.

It is interesting to note that by increasing the amount of SP, the initial workability of concrete with a 1:1 MK (metakaolin) ratio can be enhanced, leading to a comparable flow rate as that of a plain OPC concrete.^[77–79] However, OPC-MK mixtures exhibited a higher rate of flowability reduction over time compared to the reference sample. Fig. 3 (adapted from the previous work published by Duan *et al.*^[80] illustrates that OPC and OPC-MK mixtures initially had similar relative slump flow. However, as time progressed, a significant decrease in relative slump flow was observed, particularly in fresh samples containing MK (5–10 wt.%). Nevertheless, the rapid loss of flowability caused by the addition of MK can be advantageously utilized to enhance the buildability of 3D-printed concrete.^[80,81]

The replacement of a portion or the entire MK with FA in MK-blended cement concrete was observed to slow down the decrease in workability of the fresh concrete over time. (Fig. 4). Increasing the content of FA also decreased the water demand at a given initial workability (Table 2).^[82] The spherical-shaped particles of FA function like small ball bearings, creating a lubricating effect.^[83,84] This results in a retardation of PC hydration through a slow rate of aluminosilicate dissolution^[48]. As stated by Ferreiro *et al.*,^[6] the rate of alumina dissolution plays an important role in the

workability of fresh concrete. Increasing alumina reactivity induces the formation of ettringite and monosulfate, accompanied by an acceleration in the rate and heat of hydration, as well as a reduction in workability.^[85] Accordingly, there is a direct relationship between the hydration rate and the workability of fresh concrete. The lower the hydration rate, the higher the workability.^[86–89] It can be argued that, apart from having a lower alumina content compared to MK, the reduced reactivity and dissolution rate of FA contribute to the delay in the formation of ettringite and monosulfate. As a result, this leads to a slower hydration rate and an enhancement in workability.

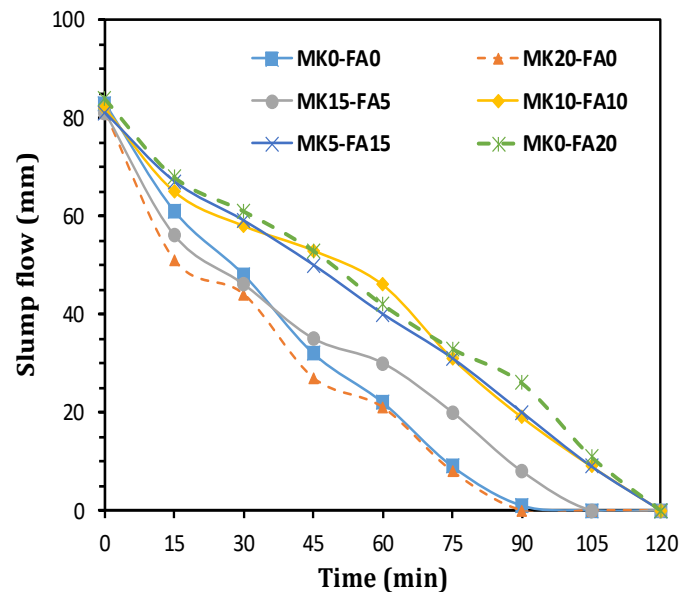


Fig. 4 Effect of FA-MK content on the flowability of the fresh OPC concrete. Reproduced with the permission from [82], 2017 Elsevier Ltd.

Table 2. The role of FA in the water demand of MK-blended cement concrete. Reproduced with the permission from [82], 2017 Elsevier Ltd.

MK: FA content wt.%	Water (kg/m ³)
0:0	208
20:0	225
15:5	221
10:10	208
5:15	199
0:20	189

As shown in Fig. 5, the alumina content within BFS does not have any effect on the workability results of OPC-BFS mortar/concrete when the water and SP contents are kept constant. Interestingly, replacing OPC with BFS actually improves the workability of cement mortar and concrete, despite the higher surface area of BFS compared to OPC.^[90–92] This improvement can be ascribed to the lower particle size of BFS, which allows for better filling of air voids between cement grains and enhances the dispersion of agglomerated cement powder, leading to the release of more free water.^[93–95]

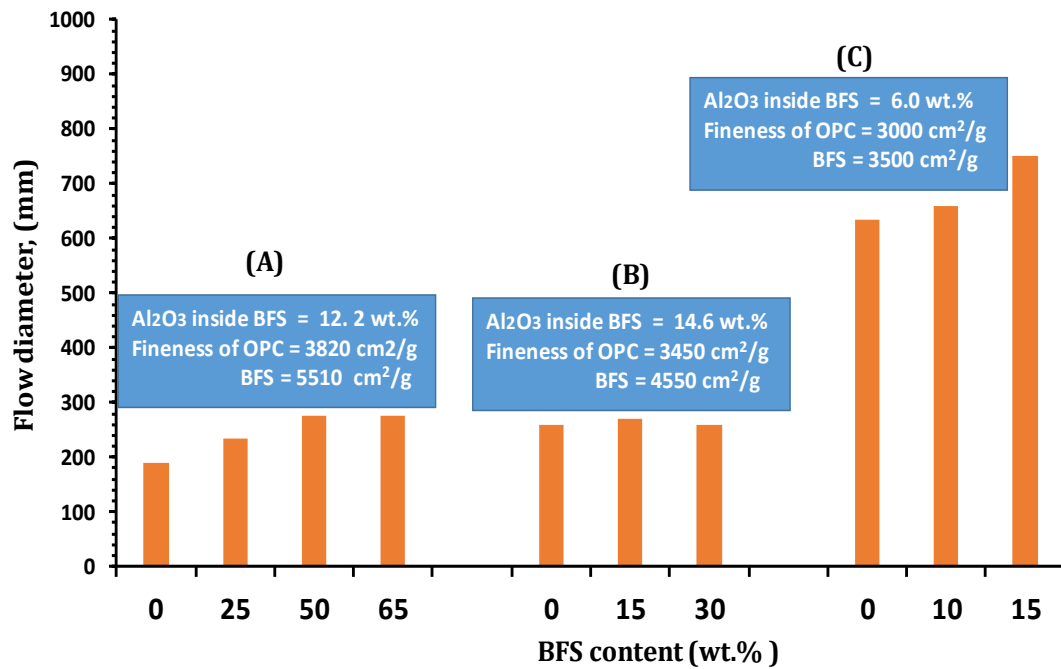


Fig. 5 Effect of Al content inside BFS on the workability of cement mortar (A: Reproduced with the permission from [90], Copyright 2018 Elsevier Ltd and concrete (B: Reproduced with the permission from [91], Copyright 2012 Elsevier Ltd.; C: Reproduced with the permission from [92], Copyright 2009 Elsevier Ltd.

2.2.2. CAC and CSAC

As mentioned earlier,^[10,11,52] CAC and CSAC exhibit a higher hydration rate compared to OPC within the first hour of hydration, resulting in rapid initial setting. This rapid setting can lead to a significant loss in workability during this short period. The primary explanation for this outcome is the involvement of alumina in the accelerated development of calcium aluminate hydrate and ettringite phases.^[96,97] Only a few studies have focused on the workability of CAC and CSAC, which may be attributed to their fast initial setting and associated challenges with workability.^[98] Furthermore, these types of cement are primarily used in the fabrication of rapid-hardening concrete, which is widely employed in repairing highway pavements and airport roadways,^[99] as well as in specific applications such as ceiling and soil stabilization.^[100]

According to a study conducted by Idrees *et al.*,^[101] it was observed that CAC demonstrates a small flow diameter. The addition of SP was observed to have a marginal improvement in workability. Fig. 6 (adapted from the paper published by Huang *et al.*^[102]) illustrates that OPC paste demonstrates a higher spread diameter compared to neat CSAC paste. Replacing OPC with up to 80 wt.% of CSAC content results in a slight decrease in the flow diameter. However, further replacement of OPC with CSAC results in a great loss in the flow rate of the fresh paste. A similar trend was observed when CSAC was used as a replacement for BFS-blended cement, as the rate of workability loss accelerated with increasing CSAC contents.^[103] The primary reason behind the rapid workability loss in CSAC is the increased heat evolution resulting from the rapid ettringite formation rate, which is consistent with previous reports.^[22,58]

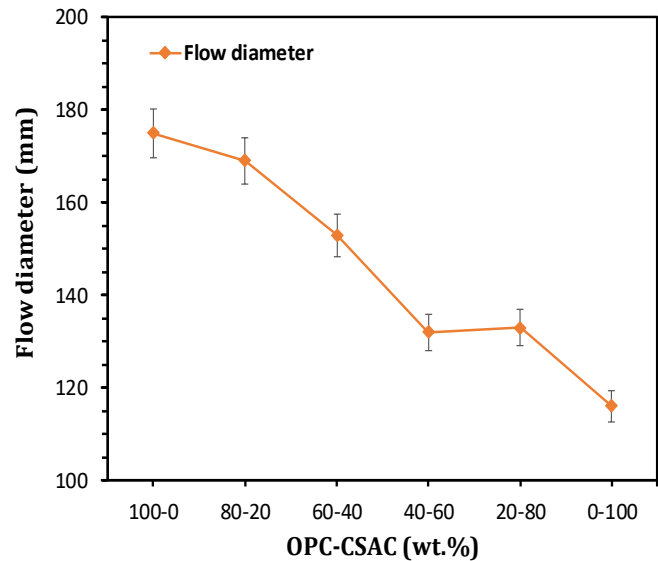


Fig. 6 Effect of CSAC on the flowability of OPC. Reproduced with the permission form [102], Copyright 2021 Elsevier Ltd.

2.2.3. Chemical admixtures

Nano alumina (nA) is considered one of the most reactive alumina sources, which strongly affects the performance of fresh OPC products.^[104] As shown in Table 3, increasing the dosage of nA leads to a decrease in the flowability of the fresh samples. The variation in OPC products (mortar or concrete), the surface area of nA, and the W/P ratio do not affect the trend of workability loss. However, increasing the mixing water can mitigate the workability loss of the fresh samples. There are two main synergistic reasons behind the workability loss, involving the physical and chemical properties of nA. From a

Table 3. The role of nA in the workability of OPC mortar/concrete (n.m. = not measured).

Reference	nA content, Wt. %	nA surface area, m ² /g	W/C ratio	Type of OPC-product	Observation
Nazari and Riahi ^[108]	0.0, 0.5, 1.0, 1.5, 2.0	163 ± 20	0.40		
Rad <i>et al.</i> , ^[109]	0.0, 2.0, 3.0	200	0.38	Concrete	Increasing the content of nA significantly decreased the flowability.
Behfarnia and Salemi ^[110]	0.0, 1.0, 2.0, 3.0	200	0.48		
Khorasani <i>et al.</i> , ^[111]		n.m.	0.33		
Ahmad ^[112]		100	0.44		
Gowda <i>et al.</i> , ^[113]	0.0, 1.0, 3.0, 5.0	n.m.	0.79	Mortar	
Joshaghani <i>et al.</i> , ^[114]	0.0, 3.0, 5.0	60 ± 5	0.32-0.40	Concrete	The flowability decreased with increasing nA dosage at different W/C ratios. However, the workability loss percentage decreased with increasing the mixing water content.

chemical standpoint, the high reactivity of nA accelerates the rate of hydration of C₃A by introducing reactive alumina into the cement matrix. This leads to the acceleration of the ettringite phase and a subsequent increase in the heat of hydration.^[66,105] Physically, nA with high reactivity increases the surface area of the cement matrix and acts as a nucleating and filling material, which, in turn, accelerates the formation of hydration products.^[64,65]

Like CSAC, Al₂(SO₄)₃ exhibited high efficiency on the acceleration of OPC hydration rate, accompanied by increasing heat evolution and accelerating the setting of the fresh OPC.^[106] As mentioned by Liu *et al.*,^[67] the bad workability of the fresh OPC paste was observed with adding 6% Al₂(SO₃)₃. The early formation of the ettringite phase is the main reason behind accelerating the hydration of OPC^[68] and, consequently, workability loss. As stated by Bhattacharjee and Santhanam,^[107] the flowability of concrete decreased significantly with increasing Al₂(SO₄)₃ content. Fig. 7 demonstrates that adding 1, 2, and 3 wt.% Al₂(SO₄)₃

decreased the initial flowability of OPC concrete from 80% to 55%, 50%, and 40%, respectively. Additionally, the workability loss with time increased as Al₂(SO₄)₃ increased. Specifically, after 30 minutes, increasing Al₂(SO₄)₃ from 0 to 3 wt.% increased the flowability loss percentages from 16.3 to 62.5%. Although the known role of Al₂(SO₄)₃ is to decrease workability, it can be beneficially used for enhancing the buildability of 3D printable concrete.^[107]

The role of different alumina sources in the workability of OPC mortar/concrete is illustrated in Fig. 8. The alumina content within MK has a significant influence on the workability of OPC-mortar and concrete. Increasing the alumina content inside MK increases the demand for SP and water, resulting in a significant loss of workability in fresh OPC concrete. Incorporating FA into the OPC-MK blend leads to an improvement in the workability of the fresh samples, mainly due to the lubrication effect of FA. Replacing MK with FA reduces the alumina content and decreases the demand for water and SP. On the other hand, the variation of alumina

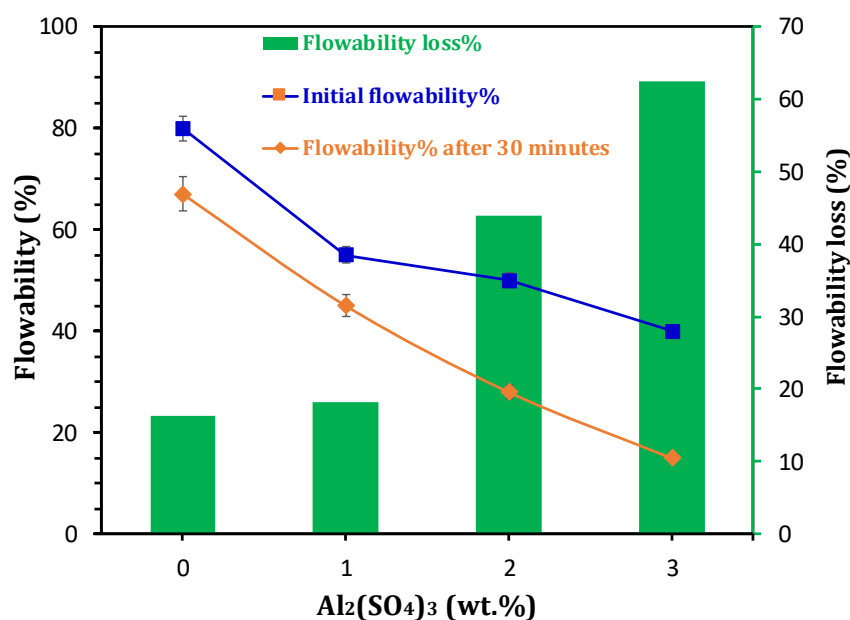


Fig. 7 Impact of Al₂(SO₄)₃ on the workability of OPC concrete. Reproduced with the permission form [107], Copyright 2021 Elsevier Ltd.

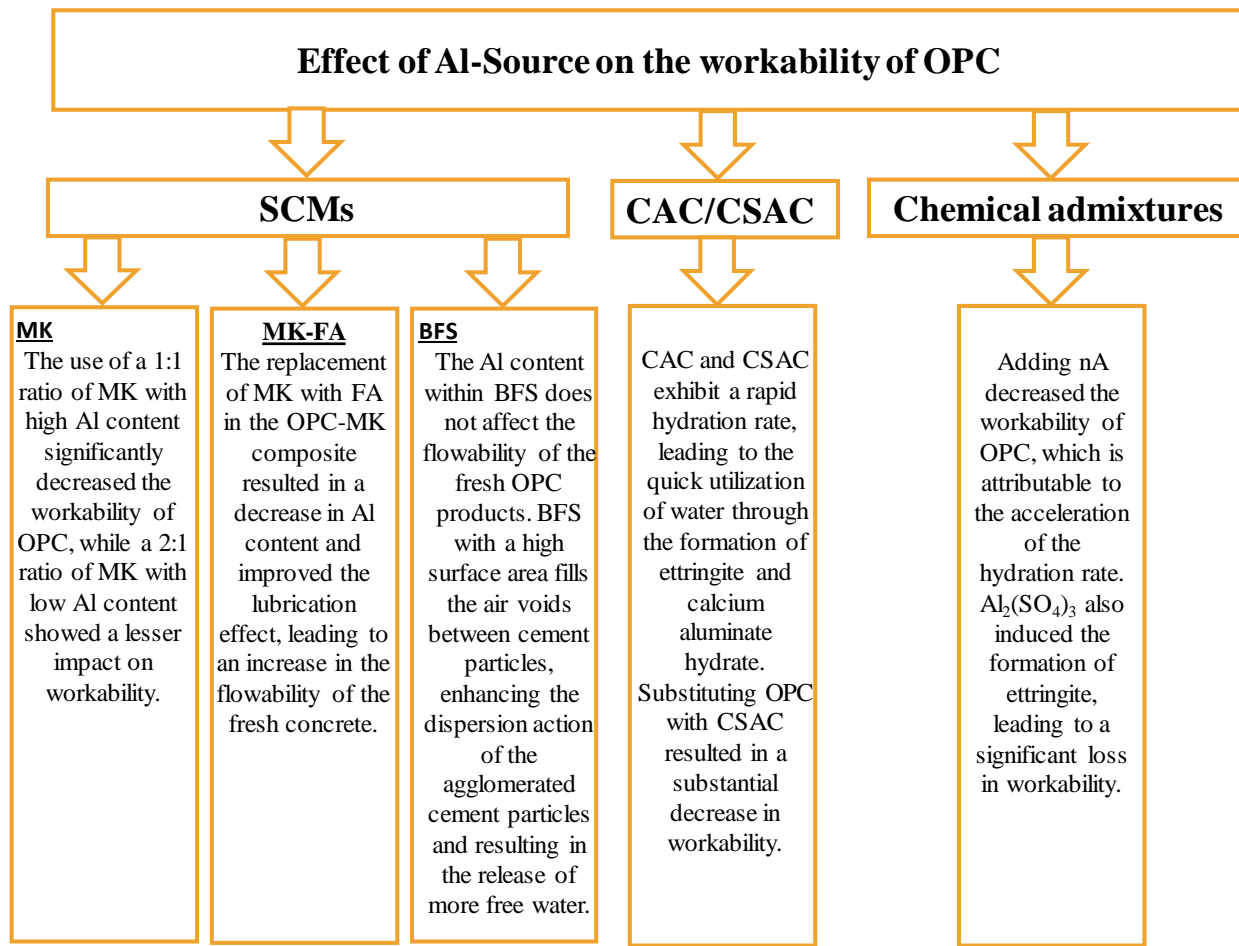


Fig. 8 Flow chart summarizes the impact of alumina sources on the workability of OPC-mortar/concrete.

content within BFS has no effect on the workability of the fresh OPC sample. In addition to the alumina content, embedding BFS with a high surface area helps fill the air voids within the OPC particles, leading to the release of free water into the cement matrix. The individual replacement of OPC with CAC, CSAC, and $Al_2(SO_4)_3$ causes a notable decrease in the initial workability, primarily attributed to the acceleration of early hydration through the formation of Al-rich phases, such as ettringite and calcium aluminate hydrate.

2.3 Effect of alumina sources on setting time of OPC

2.3.1 SCMs

As mentioned earlier,^[73] the setting time of MK-blended cement mainly depends on several factors, including the fineness of both MK and OPC, the type of cement, the SP content, and the reactivity of MK. The alumina content is regarded as one of the most important parameters that affect the setting time of OPC (Fig. 9). An increase in the initial setting time of the fresh cement mortar was recorded when OPC was replaced by different contents (up to 5 to 30 wt.%) of high-grade MK with an alumina content of 40.40 wt.%.^[115,116] In contrast, Subaşı and Emiroğlu^[117] stated that incorporating a desirable content (up to 15 wt.%) of low-grade MK with an alumina content of 11.10 wt.% significantly reduced the setting time. Nevertheless, increasing the fineness

of low-grade MK could cause an elongation in the setting time, but it is still shorter than that of high-grade MK.^[116] It can be said that the SP demand is the critical parameter that affects the setting time. As stated by Ferreiro *et al.*,^[6] MK with high alumina content exhibited an open aluminosilicate structure with a high affinity to absorb a high SP content, resulting in an increase in SP demand to attain sufficient workability. Although increasing the SP content enhanced the flowability of the cement mortar blended with MK, it strongly retards its setting process.^[73] Irrespective of its grade, the presence of a significant amount of MK in the mixture weakens and delays the initial hydration process, resulting in prolonged setting time. This delay is attributed to the dilution of the cement content.^[115]

On the other hand, the desirable content of BFS decreased the setting time of OPC, while a high content (higher than 20 wt.%) showed a retardation effect on the setting process.^[119-121] Nevertheless, increasing the fineness of BFS enhanced its reactivity, accompanied by a significant reduction in setting time, even with a higher BFS content.^[7] The dissolved alumina from BFS actually accelerates the initial hydration of OPC (C_3A), which is associated with the formation of alumina-containing binders (ettringite and monocarboaluminate). This finding is the main reason behind the shortened setting time.^[40,42] Unlike MK, BFS has low water and SP demand,

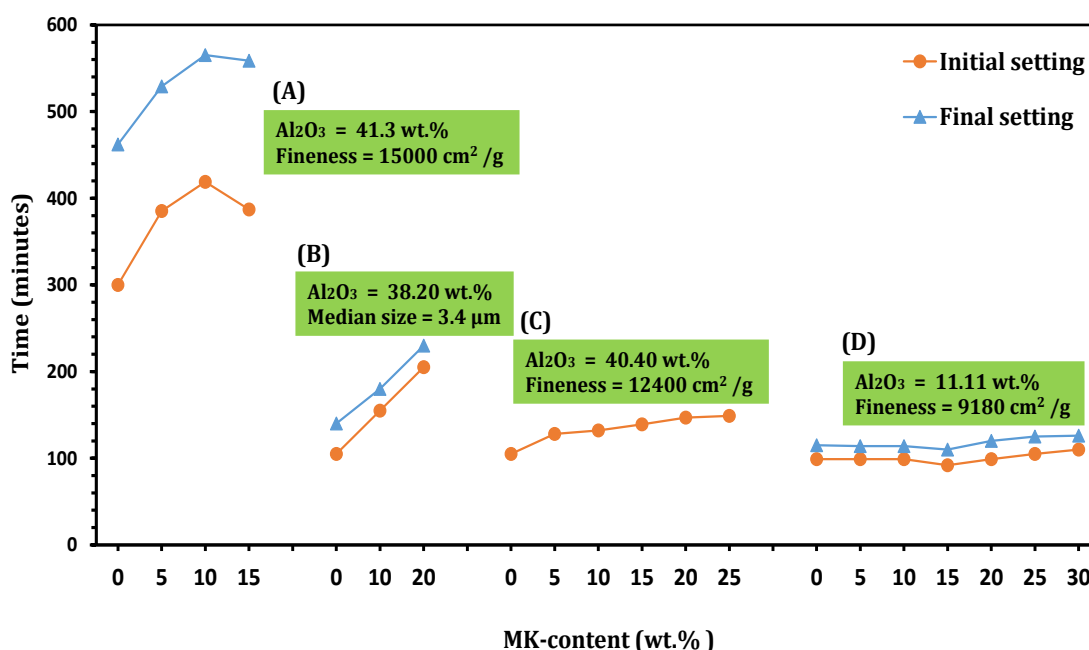


Fig. 9 Setting time of PC-MK at different weight ratios as a function of alumina content (A: Reproduced with the permission from [118], Copyright; B: Reproduced with the permission from [116], Copyright 2004 Elsevier Ltd; C: Reproduced with the permission from [115], Copyright 2018 Elsevier Ltd; D Reproduced with the permission from [117], Copyright 2015 Elsevier Ltd.).

which could be another reason for its acceleration effect.^[41] Additionally, BFS does not significantly affect the content of Ca inside the cementitious system, which is responsible for the setting at ambient curing^[7]

An elongation in setting time was observed when OPC was replaced by FA. As stated by Fu *et al.*,^[122] incorporating 65, 54, and 41 wt.% FA as a substitute for OPC has resulted in an elongation in both initial and final setting times. Kocak and Nas^[123] found that the water demand to attain a workable paste and setting time increases with increasing FA content up to 25 wt.%. The low initial reactivity and the dilution of cement content are the main reasons behind the elongation of setting time.^[124] The reduced initial reactivity of FA leads to a decline in dissolved alumina, attributed to the FA-spherical shape.^[48] This decline is responsible for the retardation rate of the C₃A hydration.^[31] The dissolution rate of alumina is also delayed by increasing the crystallinity of SCMs. As reviewed by Patangia *et al.*,^[17] replacing PC with crystalline red mud led to an increase in the setting time of the resultant fresh paste.

2.3.2 CAC/CSAC

The setting time of OPC strongly depends on the content of CAC.^[12] As shown in Fig. 10, replacing OPC with CAC up to 60 wt.% significantly decreased the initial and final setting times. Specifically, the control sample showed initial and final setting times of 200 and 319 min, respectively. The sample with 20 wt.% CAC recorded an initial and final setting time of 15 and 37 min, respectively. Flash setting was achieved when OPC was replaced by 30–60 wt.% CAC. A further increase in the CAC content up to 100 wt.% elongates the setting time again but is still shorter than that of the OPC paste. The main reason behind this result is the interaction between the

dissolved alumina in CAC and the liberated sulfate from OPC, which yields the ettringite phase associated with heat evolution^[54,125] However, the CAC content required to accelerate the hydration rate of OPC mainly depends on the solubility of the sulfate-containing phase, where OPC with hemihydrate requires a higher CAC content than that with dehydrate.^[12]

As an alumina-rich binder, CSAC demonstrates rapid initial and final setting times. Several authors have demonstrated that the initial and final setting times of CSAC vary between 5–30 minutes and 11–50 minutes, respectively.^[98,126–130] CSAC has been found to record shorter setting times than OPC, which is associated with the fast formation of ettringite.^[131] On the other hand, Fig. 11 shows that replacing OPC with different contents of CSAC causes a significant decrease in setting time.^[132] The ratios of alumina to sulfur and calcium to sulfur strongly affect the setting and hardening process of OPC-CSAC within the first 24 hours. Lee *et al.*^[133] state that the optimal alumina to sulfur and calcium to sulfur ratios are 1.23 and 1.9, respectively, which can be achieved by replacing OPC with 17 wt.% CSAC. These oxides strongly influence the formation rate of ettringite and, consequently, the initial hydration of OPC-CSAC.

2.3.3. Chemical admixtures

Nano alumina (nA) is considered one of the reactive nanomaterials that accelerate both initial and final setting times, as reviewed by Kumar *et al.*^[134] Irrespective of the content of SP, water-to-cement (W/C) ratio, and the type of mixing water, nA has been observed to have a notable impact on reducing the setting time. Nazari and Riahi^[108] demonstrated that when incorporating nA in the presence of

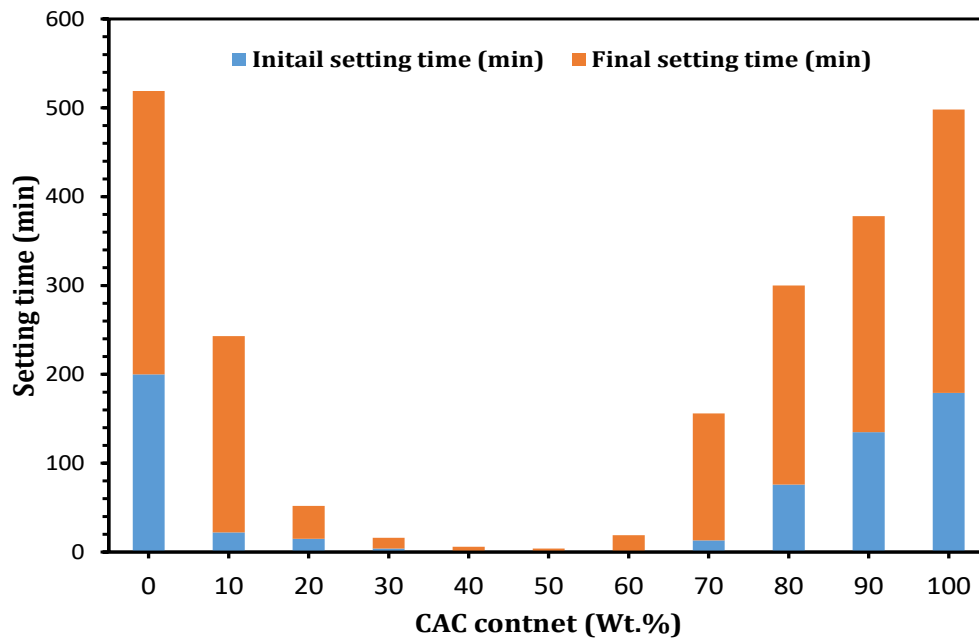


Fig. 10 Setting time of OPC-paste containing different CAC masses. Reproduced with the permission from [12].

3.5 wt.% SP and a W/C ratio of 0.40, a notable shortening of initial and final setting times was observed. Notably, the nA-modified samples mixed with lime water recorded the shortest setting time than those of mixing with tap water. In another study by Yusuf,^[135] it was found that the addition of different nA contents (1, 2, and 3 wt.%) to a PC-SF blend in the presence of 1 wt.% SP and a W/C ratio of 0.45 also significantly decreased the setting time. The addition of nA was found to increase the hydration rate of PC, leading to the formation of binding phases, particularly at the early stages of hydration. Furthermore, as shown in Fig. 12, the addition of 5 wt.% nA increased the content of chemically bound water, which is associated with the acceleration of calcium aluminate

hydrate and ettringite formation.^[22]

The same trend was observed when $Al_2(SO_4)_3$ was incorporated into the OPC. Yang *et al.*^[47] stated that a gradual shortening of the final and initial setting times of different types of PC was recorded with the incorporation of 6, 7, 8, and 9 wt.% alkali-free $Al_2(SO_4)_3$ accelerator. All samples recorded an initial setting time below 5 minutes, which enables the potential application of this mixture as spraying concrete. These outcomes are consistent with the work conducted by Wang *et al.*,^[5] Lv *et al.*^[63] and Zhang *et al.*^[136] $Al_2(SO_4)_3$ provides the system with dissolved aluminate and sulfate groups, which induce the formation of calcium sulfoaluminate hydrate, especially the ettringite phase.^[5]

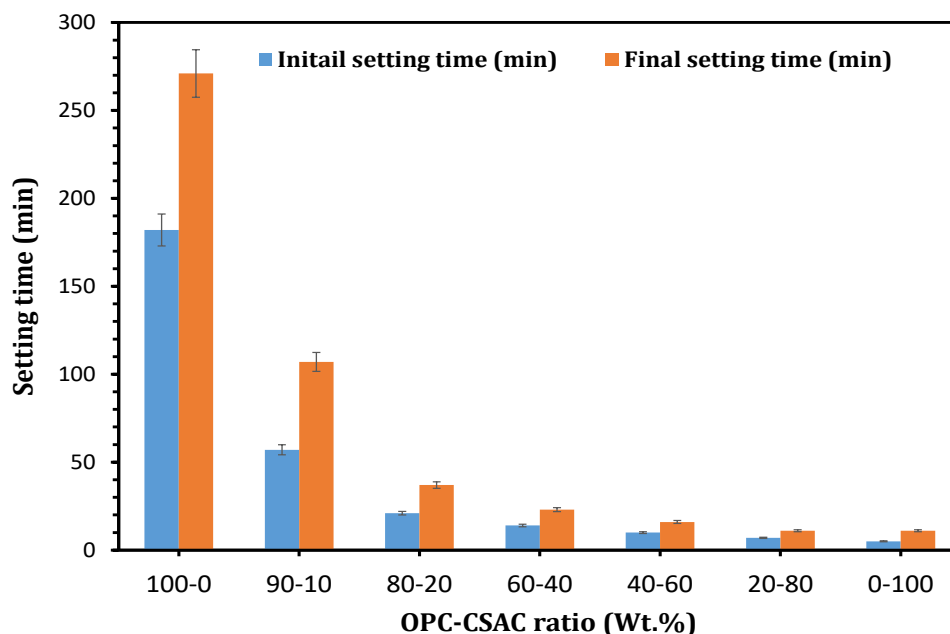


Fig. 11 Setting time of OPC-paste containing different masses of CSAC. Reproduced with the permission from [132], 2021, Higher Education Press.

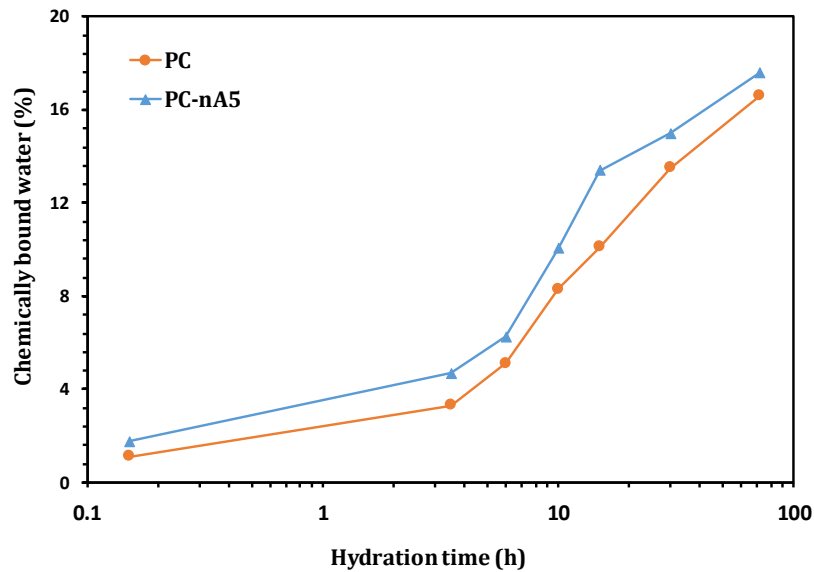


Fig. 12 Effect of nA on the chemically combined water content during the initial hydration time. Reproduced with the permission from [22], Copyright 2022 Elsevier B.V.

The role of different alumina sources in the setting time of PC is summarized in Fig. 13. The alumina content has a strong impact on the internal structure of MK, leading to differences in the ordering of the aluminosilicate group. Increasing the alumina content results in the formation of an open aluminosilicate skeleton with high demand for SP and water. This significantly prolongs the setting time of PC-MK blends. Another factor contributing to the retardation of the PC-MK hydration rate is the dilution of cement content caused by replacing PC with a high content of MK. In contrast,

replacing PC with a desirable content (up to 15 wt.%) of BFS induces the growth of ettringite and monocarboaluminate phases, leading to a shortened setting time. However, replacing a PC with a higher content of BFS exhibits a retardation effect. When PC is replaced by alumina-rich FA and red mud, a significant elongation in setting time is observed due to their low solubility and pozzolanic reactivity.

3. Missing hotspot points

Several missing perspectives for future work can be

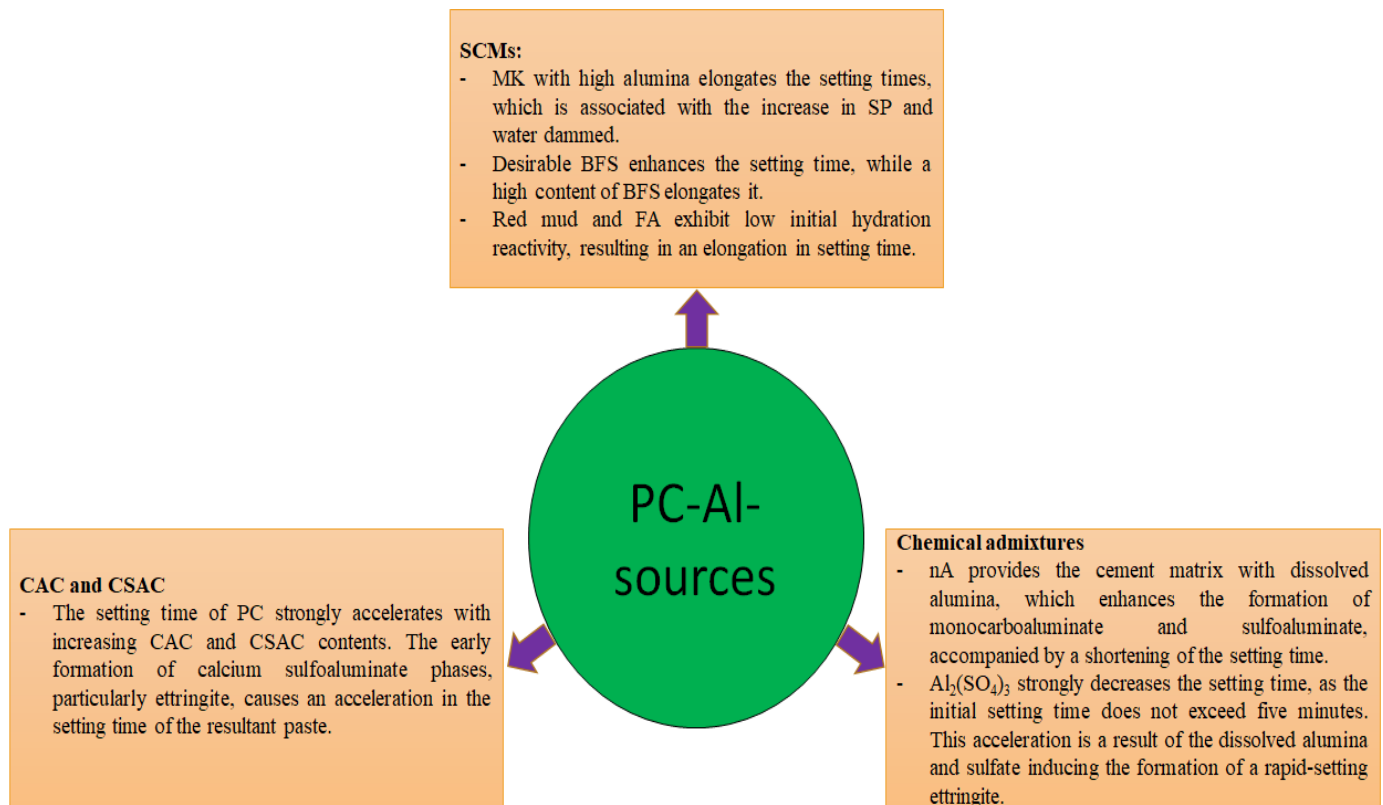


Fig. 13 Flow chart summarizes setting time of OPC mixed with different sources of alumina.

mentioned as follows:

- Investigating the impact of mixing BC with alumina resources, particularly CAC/CSAC and chemical admixtures, on the fresh and hardened properties of BC. This research can provide insights into how these additives can enhance the performance of BC.
- Exploring the formulation of limestone calcined clay cement (LC3) with CSAC as the sole binder. This approach can lead to the development of a new type of LC3 that exhibits higher early strength compared to the conventional calcined clay-limestone-OPC composite.
- Evaluating the impact of varying solubility levels in different alumina resources on the mechanical properties, drying shrinkage, and long-term durability of OPC. This investigation can shed light on how the solubility of alumina resources affects the performance and sustainability of PC.

4. Conclusions

This survey provides an intensive and comprehensive summary of the role of alumina from various resources in influencing the fresh properties of OPC mortar and concrete. Several findings can be pointed out as follows:

- The dissolved alumina from SCMs enhances the hydration of C_3A , leading to an increase in heat generation. Among the various SCMs, a specific quantity of metakaolin (MK) has demonstrated the strongest ability to accelerate the early hydration rate of Portland cement (PC). However, it is important to note that an increase in alumina content in MK also results in a higher requirement for water in the OPC-MK to SP (superplasticizer) ratio.
- OPC blended with low-grade MK (with low alumina content) exhibits better workability compared to that blended with high-alumina MK at the mixing parameters. Alumina derived from BFS and FA does not affect the workability of OPC. However, the physical properties of these materials play a significant role in improving workability
- The setting time is strongly influenced by the fineness and content of MK, as well as the demand for SP. Increasing Al inside MK results in an increase in SP demand accompanied by an elongation in setting time. It has been observed that increasing the fineness and content of MK also leads to an elongation of the setting time of OPC. Unlike MK, increasing the fineness of BFS enhanced its reactivity, accompanied by a significant reduction in setting time, even with a higher BFS content. BFS has low water and SP demand, which could be the reason for its acceleration effect. Additionally, BFS does not significantly affect the content of Ca inside the cementitious system, which is responsible for the setting at ambient curing. Although FA contains a high aluminum content and requires a lower demand for SP compared to MK, its spherical shape significantly retards the hydration reaction and prolongs the setting time of OPC.
- The reactivity of alumina-rich materials is a critical parameter that influences the fresh properties of OPC. Incorporating highly reactive CAC and CSAC accelerates

C_3A hydration but negatively affects C_3S hydration. Substituting OPC with CAC and CASC leads to a significant decrease in workability and setting time. Similarly, the use of nA accelerates early hydration, causes a loss in workability, and shortens setting time. $Al_2(SO_4)_3$ also exhibits an acceleration effect, but at a higher rate. The formation of alumina-rich binders such as ettringite, calcium aluminate hydrate, and monocarboaluminate is the primary reason behind the speeding-up effect of different alumina resources on the hydration of OPC.

- Understanding the role of aluminum from various sources in the performance of OPC could help resolve certain issues related to the resultant building products and facilitate the formulation of diverse composites for different construction applications.

Acknowledgments

His Majesty Trust Fund (HMTF) supported this work under the project “SR/ENG/CAED/21/01”. The authors are thankful for the financial support provided.

Conflict of Interest

There is no conflict of interest.

Supporting Information

Not applicable.

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