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Case Studies in Construction Materials



journal homepage: www.elsevier.com/locate/cscm

Factors affecting compressive strength and expansion due to alkali-silica reaction of fly ash-based alkaline activated mortar

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ARTICLE INFO

Keywords: Alkali activated material Compressive strength Expansion Fly ash

ABSTRACT

The development of environmentally friendly alkaline-activated materials (AAMs) holds promise, as AAMs can be derived from waste materials. This study aims to investigate the factors influencing (i) compressive strength and (ii) expansion due to alkali-silica reaction (ASR) in AAMs. These factors include alkaline concentration, heat curing conditions, fineness of fly ash, and the liquid alkaline-to-binder (L/B) ratio. The findings indicate that the higher concentrations of NaOH solution led to an increase in AAM compressive strength due to the enhanced dissolution and polymerization rates in a more alkaline environment. Heat curing stimulated chemical reactions and structure formation, while the reduced water content resulted in lower porosity and higher compressive strength in the hardened cement. Finer fly ash yielded greater compressive strength than coarser ash, as its smaller spherical particles contributed to denser and firmer structures. The presence of calcium minerals, from both Ordinary Portland Cement (OPC) and high-calcium fly ash, bolstered the strength of hardened products. Moreover, calcium minerals like CaO, Ca(OH)₂, and CaSO₄ were found to induce ASR expansion by promoting gel formation, leading to later-stage expansion in the hardened cement or concrete. However, finely milled fly ash as a precursor significantly reduced ASR expansion in AAMs, by approximately 40% compared to ordinary Portland cement. This study provides valuable insights for civil engineers for better understanding of AAM behavior and makes contributions to the safety and sustainability of cement and concrete systems.

1. Introduction

Most of the electricity nowadays is generated from coal-fired power stations. In 2022, there were over 2000 global coal power plants and the top five countries with the most operational plants are China, India, the United States, Japan, and Russia [1]. Fly ash constitutes one of the primary by-products arising from coal combustion within power plants. Within the context of Thailand's electricity landscape, coal-fired power stations persist as a prominent source of electrical energy production. Notably, the Mae Moh power plant, situated in Lampang province, plays a pivotal role in this regard, standing as the largest facility of its kind. Situated atop

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https://doi.org/10.1016/j.cscm.2023.e02595

Received 25 July 2023; Received in revised form 16 October 2023; Accepted 17 October 2023

Available online 18 October 2023

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an extensive lignite mineral deposit, the Mae Moh mine boasts a substantial reserve of lignite coal. Consequently, the Mae Moh power plant yields a cumulative electricity output exceeding 2200 Megawatts, concurrently generating a noteworthy volume of fly ash (FA), approximating 6000 tons daily. In addition, these power plants consistently produce pollutants, including sulfur oxides (SO_x), nitrogen oxides (NO_x), and fine particulate matter. Consequently, specialized treatment processes have been implemented to mitigate the emissions of these hazardous substances. For instance, flue gas desulfurization (FGD) systems have been deployed to eliminate sulfur compounds, while selective catalytic reduction (SCR) systems are utilized to reduce nitrogen compound emissions. These measures serve the critical purpose of curtailing the release of environmentally harmful toxins from the plants [2].

In the early stage of fly ash utilization, it was categorized as waste and landfilled. This practice gave rise to various issues, including the contamination of water and land by toxic substances, the dispersion of fine fly ash particles into nearby communities, and the imposition of substantial management expenses. Over 2.2 million tons of fly ash is produced by the Mae Moh power plant each year and increases annually [2,3]. Rather than just being dumped to landfill sites, many research studies have therefore been working on utilizing those massive amounts of fly ash during the past two decades [4]. Fly ash has found various applications in different domains, such as metal matrix composites (MMCs), thermal spray coating techniques employed for surface modification, the agricultural sector, polymer matrix composites, and, notably, the construction industry, where it is predominantly utilized as a geopolymer and alkaline-activated material [5–7].

At present, fly ash has been used as mineral material to partially replace ordinary Portland cement (OPC) in cement and concrete materials. In terms of environmental concern, the usage of FA as an OPC replacement material can support industrial waste utilization and significantly reduce the amount of carbon dioxide (CO_2) emission for OPC production. The production process of OPC is widely recognized as a highly energy-intensive operation, characterized by the substantial release of greenhouse gases into the atmosphere. Approximately 7 % of global carbon dioxide (CO_2) emissions can be attributed to this clinker production process, making a significant and detrimental contribution to the phenomenon of global climate change. Consequently, there has been extensive research into the development of alternative low-carbon cementitious binders as a means of mitigating the emissions of greenhouse gases associated with cement production [4,8,9]. The use of FA as a partial replacement material of OPC has been confirmed to improve elementary properties of paste and mortar, such as normal consistency, setting time, workability, water requirements, durability, and long-term strength [4]. However, to be more environmentally friendly, the development of non-OPC cementitious materials has been introduced using the 'Alkaline Activated Material (AAM)' technique.

AAM is an alternative cementitious material synthesized by using up to 100 % aluminosilicate materials as a raw starting material, e.g., fly ash. The specific dosage of alkaline solutions (e.g., NaOH and Na₂SiO₃) is added to activate the reaction and form a cement slurry or paste. The AAM may either be left at ambient temperature or cured with heat in an oven to accelerate its polymerization [10, 11]. The hydrated products can be Calcium Silicate Hydrated (C-S-H) when high calcium content binder is used, or Calcium/Sodium Alumino-Silicate Hydrated (C,N-A-S-H) for medium-to-low calcium content binders [12]. The substitution of OPC with alumina-silicate waste confers several advantages, such as cost reduction and less environmental impact. It was found that a note-worthy reduction in carbon dioxide (CO₂) emissions of up to 9 % when utilizing alumina-silicate waste as a binder in comparison to traditional OPC. Additionally, the pursuit of energy efficiency could be further advanced through the implementation of heat curing processes, such as employing finely ground precursors, harnessing supplementary heat from environmental sources, or augmenting the calcium content within AAM mixtures, thereby enhancing environmental sustainability and practical feasibility [13]. The properties of well-cured AAM have also been proven to be in the same order or even better than those of OPC and can be produced commercially as construction blocks or precast reinforced-concrete products [12,14]. This approach aligns with a broader initiative to address both economic and environmental concerns associated with cementitious materials, making it pertinent for research and application within the field.

Alkali-silica reaction (ASR) is an expansion phenomenon in cement and concrete structures due to the amalgamation of alkaline substances and silica minerals in the mixtures [15]. There are three main factors relating to ASR viz. (i) alkalinity in the cementitious system, (ii) moisture or water content in the cementitious system, and (iii) the alkali-sensitive aggregates. With specific conditions, the silica gel could be formed and expanded under a high moisture environment. Internal pressure from those expansions may cause micro-cracks of cement and concrete structures. During this reaction, a gel forms and expands within the concrete, creating tiny cracks. These micro-cracks can result in significant deterioration of the concrete structures, leading to loss in strength, moisture penetration, and corrosion of steel reinforcement. This deterioration not only affects the functionality of the structures but also compromises their overall structural performance [16,17]. ASR has a slow process that can extend over years or even decades before significantly compromising the structural integrity of OPC-based concrete structures. In OPC-based concrete, the pozzolanic reaction consumes portlandite within the cementitious system, thereby reducing alkalinity and calcium content. Consequently, this reduction suppresses ASR degradation [18]. Long-term testing of OPC-based concrete has been conducted using methods such as the Concrete Prism Test (CPT) ASTM C1293 (with testing durations of 1-2 years) or the Modified Concrete Prism Test (MCPT) AASHTO T380 (with testing durations of 56-84 days) [19]. Furthermore, a comprehensive study spanning 53 years in the United States has assessed ASR and the performance of various cements employed in construction. The results underscore those additional factors, including cement fineness, sulfate content, and curing conditions, can influence concrete's behavior in the presence of ARS. It is especially critical to limit the mass fraction of equivalent alkalies in cement to less than 0.6 % for mitigating ASR [20].

However, the study of ASR in AAM mortar or concrete is scarce and much less than those of OPC-based materials. Although many recent studies found that FA can minimize ASR expansion, more studies on other parameters are still undiscovered, especially for the chemical compositions of raw starting materials [17]. This study is to investigate the mixture design and effect of various combinations of precursors of AAM on both compressive strength (ASTM C109-cubic specimen) and ASR expansion (ASTM C1260 and C490-prism specimen) respectively. The two primary raw materials are high calcium fly ash and typical type I ordinary Portland Cement (OPC). It

is noted that a limited range of alkaline activator (NaOH) concentrations, ranging from 2 to 6 molar, were employed to effectively optimize the utilization of the alkaline activator while ensuring the attainment of acceptable engineering properties for construction applications. The utilization of lower concentrations of NaOH can yield a direct reduction in the environmental impact, specifically in terms of the overall global warming potential (GWP) and offers a practical means of achieving cost savings associated with AAM [21, 22]. Overall, this information is vital for civil engineers to understand the behavior of the ASR behavior of AAM and construct more safe and sustainable cement and concrete structures, in response to the United Nations' Sustainable Development Goal (SDG) 12: Responsible consumption and production.

2. Materials

2.1. Raw starting materials

Ordinary Portland Cement (OPC) type I for general purpose was purchased from a local distributor. High calcium fly ash was obtained from the Mae Moh coal-fired power plant, Lampang, Thailand. The experimental series used two types of fly ashes which are fine milled-high calcium (FM) and original high calcium (OM) fly ashes. The FM had less than 5 % retained on sieve no.325 by wet sieve analysis (ASTM C430) [23]. The specific gravity (ASTM C188) [24] and percent retained on sieve no. 325 are as presented in Table 1.

From X-ray fluorescence (XRF) analysis, the chemical compositions are as shown in Table 2. The alkaline content (Na₂O equivalent=Na₂O+0.658 K₂O) of the OPC was 0.6, which conformed to the ASTM C150 [25]. The alkaline content of fly ash was 3.7, while the loss on ignition (LOI) was less than 6 % for the ASTM C618 standard [26].

2.2. Alkaline activator and admixture

A laboratory-grade micro-pearl sodium hydroxide (NaOH, SH) with 99% purity was used. The NaOH solution was prepared by dissolving NaOH solid in distilled water to reach the targeted concentrations of 2, 4, and 6 molars (M). A high range water-reducer (superplasticizer) type F was used to maintain the workability of the fresh mixtures.

2.3. Aggregates

River sand (S) with a standard gradation was used as a fine aggregate in the control mixtures. The dry density of sand was 1660 kg/m³. The specific gravity of sand was 2.53, while the water absorption was 1.63 % with the fineness modulus of 1.82 by applying the ASTM C128 standard [27]. The sand was oven-dried at 110 °C for 24 h in the oven before use. Crushed limestone (CL) was obtained from a local supplier. It was rinsed through the water to remove all dust and residues before oven-drying at 110 °C for 24 h. CL was then ground in the impact-grinding machine for 30 min to achieve the standard gradation (ASTM C1260) [28] as a fine aggregate for ASR testing, then called 'Alkaline-Aggregate Reactivity (AAR)'. The size distribution and gradation are presented in Fig. 1 and Table 3, respectively.

3. Mixture designation and testing methodology

3.1. Mixture designation and sample preparation for compressive strength test

The proportion of binder-to-fine aggregate (B:S) was 1:2.75. The ratio of liquid alkaline-to-binder (L/B) varied from 0.40 to 0.50 and 0.60. The type F superplasticizer was also used to maintain the flow at 110 ± 5 %. Various concentrations of sodium hydroxide solution, i.e., 2, 4, and 6 molars (M), were applied as the main factors activating the alkaline reaction of the mortars.

The samples were cured in ambient conditions, except for the two mixtures (6OM-0M60 and 6FM-0M60), which were placed in the oven for heat curing at 60 °C for 24 h. Then, they were left at ambient temperature until reaching their testing ages. The mixture designations are presented in Table 4.

3.2. Mixture designation for ASR test

Table 1

The expansion determination for the mortars' ASR test was carried out with a $2.5 \times 2.5 \times 28.5$ cm³ control prism. The proportion of binder-to-fine aggregate (B:S) was 1:2.25. To maintain the workability, the water-to-binder (W/B) ratio of the control mix (CT) was set to 0.47, while the ASR test was initially done with L/B ratio of 0.47 with 6 M NaOH activator. After water-curing at 80 °C for 24 h,

Specific gravity and amount retained on sieve no. 325 of the binders.

Material	Specific gravity	Retained on sieve no.325 (%)
OPC	3.15	-
FM (Milled)	2.39	1.05
OM (Original)	2.26	28.70

Table 2

Chemical compositions (oxides) of the binders by XRF analysis (%).

Material	SiO_2	Al_2O_3	Fe ₂ O ₃	CaO	SO_3	MgO	Na ₂ O	K ₂ O	Na ₂ O _{eq}	LOI
OPC	20.9	4.8	3.4	65.4	2.7	1.3	0.3	0.4	0.6	1.0
FM	27 9	14 4	15.6	27 9	7 1	2.2	1.9	2.8	3.7	0.2



Fig. 1. Size distribution of standard sieve for crushed-limestone ASR testing.

Table 3Gradation of standard sieve for ASR testing.

Sieve size		Mass (%)
Passing	Retained on	
4.75 mm (No.4)	2.36 mm (No.8)	10
2.36 mm (No.8)	1.18 mm (No.16)	25
1.18 mm (No.16)	600 μm (No.30)	25
600 μm (No.30)	300 µm (No.50)	25
300 μm (No.50)	150 µm (No.100)	15

 Table 4

 Mixture designations for compressive strength test of fly ash mortars.

Mixes	Mixture p	proportion (g.)			L/B	Curing Type	Flow (%)	
	OM	FM	Sand	NaOH (g.) / M	SP*			
60M-0 M	100	-	275	0 / 0 M	-	0.6	Ambient	115
60M-2 M	100	-	275	4.8 / 2 M	1.0	0.6	Ambient	114
60M-4 M	100	-	275	9.6 / 4 M	1.5	0.6	Ambient	107
60M-6 M	100	-	275	14.4 / 6 M	3.0	0.6	Ambient	105
60M-0M60	100	-	275	0	-	0.6	60 °C	115
6FM-0 M	-	100	275	0	-	0.6	Ambient	108
6FM-0M60	-	100	275	0	-	0.6	60 °C	108
6FM-6 M	-	100	275	14.4 / 6 M	4.5	0.6	Ambient	108
5FM-6 M	-	100	275	14.4 / 6 M	6.0	0.5	Ambient	105
4FM-6 M	-	100	275	14.4 / 6 M	10.0	0.4	Ambient	105
4FM-6M60	-	100	275	14.4 / 6 M	10.0	0.4	60 °C	105

Note: *SP is a 50% w/w superplasticizer

Example of mixture's name:

6OM-6 M refers to L/B= 0.6, high calcium fly ash - 6 Molar NaOH, ambient curing

6OM-0M60 refers to L/B= 0.6, high calcium fly ash - 0 Molar NaOH, 60 °C curing

6FM-0 M refers to L/B= 0.6, fine milled-high calcium fly ash - 0 Molar NaOH, ambient curing

4FM-6 M refers to L/B= 0.4, fine milled-high calcium fly ash - 6 Molar NaOH, ambient curing

the samples were then continuously soaked in 1 Molar sodium hydroxide (NaOH) solution at 80 °C until the testing age. The mixture designations are as presented in Table 5.

3.3. Analytical techniques

The compressive strength of the alkali-activated mortars was examined by a Universal Testing Machine (UTM) following the ASTM C109 [29]. It is noted that the suitable workability of the mortar was first carried out by using a flow test. The $5 \times 5 \times 5$ cm³ cubic samples were neatly prepared with 165 specimens in total. After demolding in the next 24 h, the samples were water-cured at ambient temperature until reaching their testing ages at 7-, 14-, 28-, 60-, and 90-days.

The expansion determination due to the alkali-silica reaction of the mortars was modified from specimen's size of the ASTM C1260

Table 5

Mixture (designations	for ASR	test of	high	calcium	fly	ash mortars.

Mixes	Mixture pr	roportion (g.)	Ratio	Curing type			
	OM	FM	OPC	AAR*	NaOH (g.) / M		
СТ	-	-	100	225	0 / 0 M	0.47 (W/B)	Ambient
OM-6 M	100	-	-	225	14.4 / 6 M	0.47 (L/B)	Ambient
FM-6 M	-	100	-	225	14.4 / 6 M	0.47 (L/B)	Ambient
FM-6M60	-	100	-	225	14.4 / 6 M	0.47 (L/B)	0° C

Note: *AAR is Alkaline-Aggregate Reactivity

Example of mixture's name:

CT refers to OPC mortar with W/B=0.47, ambient curing

OM-6 M refers to L/B= 0.47, high calcium fly ash - 6 Molar NaOH, ambient curing

FM-6M60 refers to L/B= 0.47, fine milled-high calcium fly ash - 6 Molar NaOH, 60 °C curing

and C490 [28,30]. The $2.5 \times 2.5 \times 28.5 \text{ cm}^3$ control prisms were demolded and measured after 24 h curing in 80 °C-water baths. That sample was then soaked in 1 Molar sodium hydroxide (NaOH) solution at 80 °C until the testing age of 28 days. The average percentage of expansion was calculated in the following Eq. 1. Fig. 2 shows the overall schematic diagram of the experimental program in this study with 24 specimens in total.

$$L = \frac{Lx - Li}{Lg} \times 100$$

Where L =length change (%).

 $L_x =$ Length at the testing age (in.).

 $L_{i}=\mbox{Length}$ at the initial stage (in.).

 $L_g = Gauge length (in.).$

4. Results and discussion

4.1. Compressive strength of high calcium fly ash-based alkali-activated mortar

The compressive strength of high calcium fly ash-based alkali-activated mortar in various ages viz. 7, 14, 28, 60, and 90 days are presented in Table 6. The effects of NaOH solution concentration, heat curing, fly ash fineness, and L/B ratio are reported and discussed, respectively.

4.1.1. Effect of NaOH solution concentration

This test used original high calcium fly ash (OM) to prepare the mixtures with a L/B ratio of 0.60. Various sodium hydroxide concentrations were 0, 2, 4, and 6 molars (M). It is important to highlight that, in this context, a specific range of alkaline activator (NaOH) concentrations, spanning from 2 to 6 molar, has been carefully selected. This range has been chosen to effectively optimize the use of the alkaline activator, while also ensuring that the resulting construction materials exhibit acceptable engineering properties. After demolding, the samples were left in room conditions until the testing age. It can be seen in Fig. 3 that the compressive strength was developed by the time from 7 days- to 90 days-age for all mixtures.



Fig. 2. Schematic diagram of the experimental program.

(1)

Table 6

Compressive strength of the samples in various ages.

Mixes	Compressive strength (MPa)								
	7 d	14 d	28 d	60 d	90 d				
60M-0 M	2.14	3.88	4.80	7.32	8.08				
60M-2 M	2.39	4.12	5.05	9.26	10.21				
60M-4 M	2.70	4.88	7.28	10.77	11.98				
60M-6 M	3.16	7.06	9.33	11.85	12.63				
60M-0M60	2.68	4.61	5.30	8.38	9.25				
6FM-0 M	3.42	5.19	6.13	9.13	10.08				
6FM-0M60	4.54	5.85	7.06	9.82	10.83				
6FM-6 M	6.85	9.34	10.65	12.58	14.84				
5FM-6 M	7.23	9.80	11.06	12.79	15.98				
4FM-6 M	7.81	11.01	11.87	14.10	16.89				
4FM-6M60	8.09	11.64	13.38	15.38	18.14				

The high calcium fly ash-based alkali-activated mortar with a higher concentration of NaOH activator (e.g., 6 M) achieved higher strength than that of both lower concentrations (i.e., 4 M and 2 M) and none-NaOH mixture (i.e., 0 M). In addition, it is seen that the compressive strength increased dramatically in the first 7 and 14 days-age. Then, the strength continued to rise slowly from 28 to 90 days of testing age. The results were similar to previous studies, which described that some additional alkaline-activated products were formed by more dissolution and polymerization rates in such a higher alkaline environment [31]. Therefore, in this study, the higher NaOH concentrations used in AAM can provide higher strength than the lower ones. In addition, the optimal NaOH concentrations for each raw material scenario need to be defined as a higher concentration may lead to a coagulated structure with less workability and much more costly for production [32].

4.1.2. Effect of heat curing on the compressive strength

Numerous research investigations have diligently explored ambient curing conditions in AAM. For instance, some studies have explored the utilization of high-calcium content minerals as precursors, such as high calcium fly ash, furnace slag, and ordinary Portland cement (OPC), or have even examined alternative heat sources for the curing environment [33,34]. However, the focus of this particular study is the utilization of fly ash as the primary precursor due to its abundant availability as a by-product in Thailand.

The effect of heat curing on the compressive strength was performed on high calcium-milled fly ash (FM) and original high calcium fly ash (OM). The L/B ratio was 0.60, without using any superplasticizer and NaOH solution. In a previous study, a range of different curing temperatures, spanning from 10° to 80 °C, were investigated to observe their impact on the strength of AAM. The results ultimately revealed that a temperature of approximately 60 °C yielded the highest compressive strength while maintaining reasonable energy consumption, meeting the required standards for practical use. As a result, for the heat-curing mixtures in this current study, an oven curing temperature of 60 °C for a duration of 24 h was selected as per references [35,36]. It can be seen that the heat curing mixtures (6OM-0M60 and 6FM-0M60) gained higher strength than that of the ambient temperature curing (6OM-0 M and 6FM-0 M) (Fig. 4). The presence of calcium minerals in high calcium fly ash acted as a parent formation of calcium silicate hydrated product (C-S-H), which provided the strength of the hardened cementitious material. Alternatively, calcium hydroxide (Ca(OH)₂) can also be formed and provide an alkaline environment for the pozzolanic reaction of the rest silica (Si) and alumina (Al), offering the secondary C-S-H and C-A-S-H products [12,14]. A suitable heat curing regime could stimulate those formations as part of the alkaline activated reaction without any additional NaOH solution, increasing both early strength (7 days) and later ages [35,36]. It is worth noting that high-fineness fly ash (FM) evidently achieved higher compressive strength than low-fineness (OM) for all cases, around 20 % for the



Fig. 3. Compressive strength of high calcium fly ash-based alkali-activated mortar in various NaOH concentrations.



Fig. 4. Compressive strength of high calcium fly ash-based alkali-activated mortar in various heat curing regimes.

early age and around 10% for the later period.

4.1.3. Effect of fly ash fineness on the compressive strength

The effects of fly ash fineness on the compressive strength were performed on both original high calcium fly ash (OM) and high calcium-milled fly ash (FM). The L/B ratio was set to 0.60, while none (0 M) NaOH and 6 M NaOH concentrations were prepared and cured in ambient conditions. With the same dosage of alkaline NaOH (0 M and 6 M), FM mixtures achieved higher compressive strength than OM mixtures (Fig. 5). High fineness material, which refers to high surface areas, can quickly be reacted with water and alkaline solution in the system [37]. Moreover, those small spherical particles could also fill-up most voids and cavities in the cement matrix [38]. It is also observed that the usage of NaOH as an alkaline activated mixture provided higher strength than non-NaOH mixtures, especially for the early age strength at seven days old.

4.1.4. Effect of L/B ratio on the compressive strength

High calcium-milled fly ash (FM) was used to prepare the mixtures with a sodium hydroxide concentration of 6 M. Various L/B ratios in the mixtures were 0.40, 0.50, and 0.60. The samples were left in room conditions after demolding until the testing age. In Fig. 6, the lower amount of L/B ratios provided greater compressive strength than the higher ones. The reasons are that less amount of water in the system led to less porosity of the hardened cement and the reduction of microcracks between cement paste and the fine aggregates [14,39]. However, the workability of the mixtures may need to be carefully considered for practical usage.

4.2. Expansion due to alkali-silica reaction (ASR) of the alkaline activated mortars

To examine the ASR of high calcium fly ash-based alkaline activated mortars, an expansion test was performed on high calciummilled fly ash (FM) and original high calcium fly ash (OM). In general, the testing durations for concrete specimens encompassed a range of 56–84 days, following the protocol outlined in the Moisture Curing Period Test (MCPT) per AASHTO T380, or extended to 1–2 years as prescribed by the Concrete Prism Test (CPT) in accordance with ASTM C1293 standards. Conversely, the accelerated mortar



Fig. 5. Compressive strength of high calcium fly ash-based alkali-activated mortar in different fineness of fly ash.



Fig. 6. Compressive strength of high calcium fly ash-based alkali-activated mortar in various L/B ratios.

tests involving immersion in a sodium hydroxide (NaOH) solution, as specified by ASTM C1260, required a considerably shorter testing period of only 16 days. Nevertheless, in order to monitor expansion characteristics over an extended timeframe, a 28-day testing duration was also conducted. It should be noted that the testing of ASR expansion in these AAM mixtures was conducted with modifications to the ASTM C1260 standard.

The W/B ratio of 0.47 for OPC mortar, and L/B ratio of 0.47 of 6 M NaOH solution for AAM were investigated. OPC mortar (CT) was prepared as a control ASR expansion mixture. While another mixture of FM-based mortar was set for a heat curing approach at 60 °C for 24 h. CT had the expansions of 0.126 % at 14 days, which were higher than the limitation for innocuous aggregate in mortar due to ASR (0.10 %). After that, the expansion of CT continually increased up to 0.211 % at 28 days. This proved that the aggregate used in the mixture is highly reactive for ASR. CT had the most significant expansion due to the presence of calcium oxide (CaO), Ca(OH)₂, and CaSO₄ in the system [40]. More minor expansions were observed in the mixtures with fly ash as the pozzolanic reaction could reduce the amount of Ca(OH)₂ [41].

High-fineness fly ash particles (FM) can easily and efficiently fill the voids of cementitious structures, leading to an increase in density and strength with low expansion [37,38]. It can be observed in Fig. 7 that both FM-6M60 and FM-6 M mixtures had less ASR expansion than the CT and OM-6 M since the first day of observation. The heat curing may induce more penetration of NaOH solution through the samples and cause additional unexpected expansions [42]. The expansion due to ASR of the heat-cured mixture (FM-6M60) slightly increased from the ambient curing mixture (FM-6 M). However, all the 28 days-age samples had the expansion results in the innocuous and deleterious zone (0.10–0.20 %).

The surfaces and lengths transformation after ASR expansion are shown in Fig. 8. The control CT had a porous structure that easily allows the NaOH solution to penetrate, causing a slight change in color with a bit of bending in the middle of the specimen. The sample with average fly ash particle size (OM-6 M) had no visible cracks or damages, but some salts and silica gels were found on the outer surfaces. Some damages were observed on the top surface of the heat-cured mixture (FM-6M60). However, no changes or deteriorations were found on FM-6 M. Similar findings have been observed in prior research endeavors aimed at assessing the potential for ASR within AAM systems. It was noted that the reactive aluminum, derived from precursors, undergoes dissolution, ultimately forming a protective alumino-silicate gel layer on the surface of precursor grains. This gel layer effectively inhibits the further dissolution of silica and the subsequent development of ASR gel. Additionally, high-fineness fly ash particles exhibit an advantageous characteristic in efficiently occupying void spaces within cementitious structures. This phenomenon contributes to heightened density and strength while minimizing expansion tendencies [18,43].

5. Conclusions

Influencing factors of alkaline concentration, heat curing condition, fineness of fly ash and liquid alkaline-to-binder (L/B) ratio of the mixture were examined on fly ash-based alkaline activated mortars (AAM). Main conclusions can be drawn as follows:

- 1) The compressive strength of AAM increased when the concentration of NaOH solution increased. A high alkaline environment resulted in a higher dissolution and polymerization rates leading to an extra strong alkaline-activated product. The finer fly ash achieved higher compressive strength than coarser fly ash, as the smaller spherical particles acted as a filler to fill up most tiny cavities in the cement matrix, allowing more compact and firm structures.
- 2) Heat curing at 60 °C for 24 h in the oven could stimulate the formation of the cement structure, especially for the alkaline-activated reaction. The alkaline activated cement had a similar characteristic as ordinary OPC mortar, where less amount of water in the system led to less porosity of the hardened cement, providing greater compressive strength. However, the compensation of evaporated water content needs to be considered if a heat curing regime is applied and the workability in the practical usage.



Fig. 7. Expansion due to alkali-silica reaction (ASR) of the high calcium fly ash based-alkaline activated mortars.





- 3) Finer particles led to a low ASR expansion as the voids in cementitious structures were efficiently filled. However, the heat curing might cause additional unexpected ASR expansions due to more penetration of NaOH solution through the samples. Therefore, the optimal combination for the blended mortars might be considered in compressive strength and any possible ASR expansion.
- 4) The utilization of reduced concentrations of the alkaline activator (NaOH) serves the dual purpose of optimizing engineering properties and mitigating environmental impact, thereby aligning with the principles set forth in Sustainable Development Goal (SDG) 12: responsible consumption and production. This research carries significant value for civil engineers who aspire to develop cement and concrete formulations that are not only safer but also more ecologically sustainable for use in construction materials.

At this stage, the results have proven that AAMs have the potential to be used as an alternative cementitious material. For future research, the formation of internal compounds in the AAM, likely using the Fourier-transform infrared spectroscopy (FTIR) technique, X-ray diffraction analysis, and the determination of the morphology using the scanning electron microscope (SEM) technique should be performed.

CRediT authorship contribution statement

Artith Wongpaun: Investigation, Formal analysis. Weerachart Tangchirapat: Conceptualization, Methodology, Writing-Review & Editing, Supervision, Visualization, Investigation, Formal analysis. Teewara Suwan: Writing-Original Draft, Writing-Review & Editing, Visualization, Investigation, Formal analysis. Mizi Fan: Writing-Review & Editing, Visualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgments

The first and second authors would like to thank Thailand Science Research and Innovation (TSRI) under Fundamental Fund 2023 (Project: Advanced Construction Towards Thailand 4.0) for providing financial support during the research. The corresponding author would also like to express gratitude to the Department of Civil Engineering, Faculty of Engineering, Chiang Mai University (CMU) who partially supported this research work.

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