Journal of Metals, Materials and Minerals, Vol. 33, No. 2, pp. 75-80, 2023

The influence of water-cement ratios and alumino-silicate based accelerator on the properties of fiber-reinforced cement composites

Parinya CHAKARTNARODOM[1], Passakorn SONPRASARN[1], Sureerat POLSILAPA[1], Nuntaporn KONGKAJUN[2,*], Edward A. LAITILA[3], and Wichit PRAKAYPAN[4]

[1] *Department of Materials Engineering, Faculty of Engineering, Kasetsart University, Chatuchak, Bangkok, 10900, Thailand*
[2] *Department of Materials and Textile Technology, Faculty of Science and Technology, Thammasat University, Klong Luang, Pathumthani 12121, Thailand*
[3] *Department of Materials Science and Engineering, Michigan Technological University, Houghton, MI 49931, USA*
[4] *Source Runner Enterprise Company Limited, Bangkhen, Bangkok, 10220, Thailand*

***Corresponding author e-mail:** n-kongkj@tu.ac.th

Received date:
16 February 2023
Revised date
8 May 2023
Accepted date:
16 May 2023

Keywords:
Fiber cement;
Air curing;
Autoclave curing;
Chemical admixture;
Water-cement ratio

Abstract

Due to more strict environmental protection and greenhouse gas reduction, it is very important for all industries to appropriately manage their energy consumption. Fiber- reinforced cement composites are the popular building materials which consume enormous energy to intensify its chemical reaction during the autoclave steam curing process. Utilization of chemical admixture to replace the conventional energy-driven autoclave steam curing process will support the fiber- reinforced cement composites industry to develop sustainable building materials. In this research, typical and mechanical properties of the air-cured fiber- reinforced cement composites incorporated with alumino-silicate based accelerator were investigated. The results show an excellent correlation between the water-cement ratio of the mix design and the mechanical strength which is the optimum water-cement ratio for this FRCC are 0.53. Moreover, the properties of fiber- reinforced cement composites cured by either the autoclave steam curing process or air-curing process are comparable.

1. Introduction

Fiber-reinforced cement composites (FRCC) or fiber cement is a popular building material. Its market is expected to grow with the growth rate of 5.3% between 2021 and 2030 due to the rapid increase in usage of FRCC for internal and external residential applications [1]. This is because the superior characteristics of FRCC such as high strength, fire-resistant, durability, and stability. Ordinary Portland cement (OPC), silica sand, gypsum and cellulose fiber are the major raw materials for FRCC production. After forming process, the green sheets of FRCC must be cured by the autoclave steam curing (ASC) process which consumes massive energy to accelerate the hydration reaction of OPC. The ASC process usually takes about 24 h (1 day) by using the curing temperature between 160°C to 180°C and pressure between 8 bar to 12 bar. [2-8].

Due to elevated environmental concerns, reduction of the energy consumption in the FRCC curing process is a high-impact approach to reduce the greenhouse gas emission from fiber-cement industry and the carbon footprint of FRCC. Using chemical admixture is an approach to enhance the chemical reaction and the early strength development of FRCC at atmospheric temperatures. The strength development of cement-based materials depends on calcium silicate hydrate (C-S-H) phase generated from the hydration reaction [8-11]. Chemical admixture may be added to concrete and mortar to enhance the hydration rate and their strength, especially the early strength. An example of chemical admixture is the chemical accelerator such as alumino-silicate based accelerator (ASBA). When ASBA is added to the concrete and mortar, it will increase the rate of hydration of cement, reduce the setting time of concrete, and increases the rate of hardening or strength development [12]. From our prior work [13], using ASBA also reduced the energy consumption in the curing process resulting in the carbon-footprint reduction in FRCC.

Therefore, this work aims to develop the FRCC having the mechanical strength after 1 day of the curing period under atmospheric pressure be comparable to the strength of FRCC cured by ASC process. The experiment was carried out by adjusting the FRCC mix design and the application of ASBA to improve the early strength development. The characterization for mechanical and physical properties was carried out based on the industrial standard.

2. Experimental

2.1 Materials

To produce FRCC samples, the raw materials were OPC, silica sand, gypsum, and cellulose fibers. X-ray fluorescence spectrometer (XRF, Panalytical-Miniplal 4) was used to determine the chemical composition of raw materials as shown in Table 1. The particle size distribution of raw materials was measured by laser particle size analyzer (HELOS/BR with wet dispersing unit QUIXEL). The results of particle size analysis of raw materials are shown in Figure 1. ASBA was used as a chemical admixture. ASBA was prepared by the heat treatment of agro-waste ash and recycle concrete mixture. ASBA was produced under the license of Shera Public Company Limited.

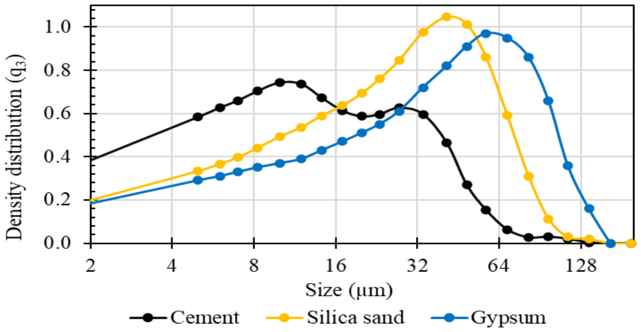

Figure 1. Particle size distribution of raw materials.

2.2 Sample preparation and mixed designs

FRCC samples were prepared using filter pressing process. The tools used for the sample preparation are shown in Figure 2. The mixed designs are shown in Table 2 and Figure 3. The sample preparation process was divided into two parts. The first part is the mixing of raw materials. The second part was the green sample forming.

For mixing process, the cellulose fibers were dispersed in the water using an overhead high-speed mixer (Figure 2(a)) for 15 min. After that, gypsum, sand, and ASBA were added, and mixed for 5 min. Then, OPC was added and mixed continuously for 5 min until the mixture became homogeneous. As shown on Figure 3, the weight percentage of water is the same in all mixed designs. even though w/c ratios were different.

To form a green sample, the mixture was poured into the mold as shown in Figure 2(b) and then pressed by the filter pressing machine (Figure 2(c)) using the applied pressure at 1 MPa. The green FRCC sample size is 8 mm × 210 mm × 7 mm as shown in Figure 2(d).

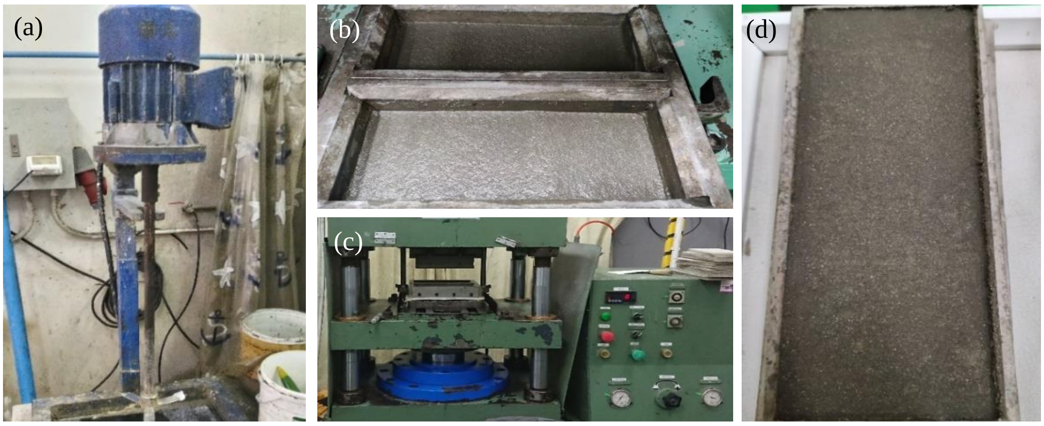

Figure 2. (a) an overhead high-speed mixer, (b) filled mold for sample forming, (c) filter pressing machine, and (d) green FRCC sample.

Table 1. chemical composition of raw materials.

Materials	Composition (wt%)							
	MgO	Al_2O_3	SiO_2	K_2O	CaO	Fe_2O_3	SO_3	Others
OPC	1.09	5.98	19.9	2.74	63.72	3.17	0.35	3.05
Sand	0.01	2.06	96.04	0.25	0.12	0.98	0.16	0.38
Gypsum	0.21	0.14	0.15	0.05	24.58	0.11	73.01	1.75

Table 2. Mix designs of FRCC used in this research.

Formula	Materials (wt%)				ASBA (% of OPC weight)	Water cement ratio (w/c)
	OPC	Sand	Gypsum	Cellulose fibers		
R-ASC	35.0	35.0	25.0	5.0	-	1.14
R-AC	35.0	35.0	25.0	5.0	-	1.14
M1-1	35.0	55.0	5.0	5.0	-	1.14
M1-2	35.0	55.0	5.0	5.0	3.0	1.14
M2-1	45.0	45.0	5.0	5.0	-	0.89
M2-2	45.0	45.0	5.0	5.0	3.0	0.89
M3-1	55.0	35.0	5.0	5.0	-	0.73
M3-2	55.0	35.0	5.0	5.0	3.0	0.73
M4-1	65.0	25.0	5.0	5.0	-	0.62
M4-2	65.0	25.0	5.0	5.0	3.0	0.62
M5-1	75.0	15.0	5.0	5.0	-	0.53
M5-2	75.0	15.0	5.0	5.0	3.0	0.53

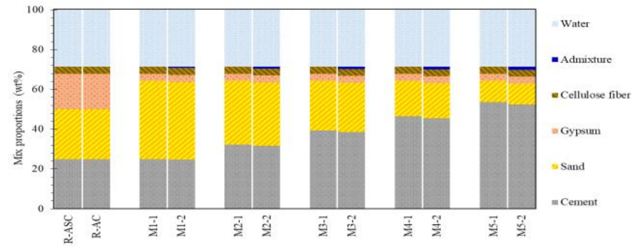

Figure 3. Mix designs in 100% of total weight, including water.

For curing process, from Table 2, the green samples were cured by:

(a) the ASC process for 24 h at a temperature between 160°C to 180°C and a pressure between 8 bar to 12 bar for R-ASC formula, and

(b) the air curing process under atmospheric pressure, at room temperature (about 27°C) and the relative humility between 50% to 60% for 24 h for R-AC and M-series formulas.

2.3 Sample characterization

After curing, the mechanical properties including modulus of rupture (referred as MOR) and modulus of elasticity (referred as MOE), were analyzed by Instron 3300-series universal testing machine (UTM) and based on the methods described in ASTM C1185 standard [14]. Ten samples of the same formula were used for each test. The physical properties including bulk density and water absorption of the samples were determined by Archimedes method as mentioned in ASTM C948-81 [15] using the equations as shown below

$$\text{Bulk density} = \frac{W_d}{W_w - W_s} \quad (1)$$

$$\%\text{Water absorption} = \frac{W_w - W_d}{W_d} \times 100 \quad (2)$$

where W_d is the weight of the dry sample after 24 h. of drying at 100°C, W_s is the weight of sample immersed in the water, and W_w is the weight of the sample just removed from water after 24 h of immersion.

In addition, the scanning electron microscopy (SEM: Philips XL30) was utilized to examine the microstructure of samples.

3. Results and discussion

Figure 4(a) and Figure(b) show the mechanical properties including MOR and MOE of the samples. The minimum requirements on MOR and MOE of FRCC by the industry are shown with the dash lines [13,16]. The samples from formula R-ASC are the control samples cured by ASC process. The samples from formula R-AC-28D and R-AC-1D are also the control samples cured under atmospheric pressure at room temperature for 28 days (about 4 weeks) and 1 day, respectively.

From Figure 4, the average MOR and MOE of the R-ASC and R-Air 28D samples passed the industry requirement, while those properties of R-Air 1D samples did not. Normally, mechanical strength development of the air-cured cement-based materials reaches 95% of their full strength in 28 days (about 4 weeks) [17]. However, the average MOR of R-Air 28D samples are significantly lower than that of R-ASC samples.

Typically, using lower water-cement (w/c) ratio in the mix design increases the mechanical strength of cement-based materials [11,17-22]. From Table 2, w/c ratio reduces from 1.14 in formula M1-1 to 0.53 in formula M5-1. This was achieved by keeping the amount of water constant while the OPC content increased as shown on Figure 3 resulting in the improved MOR and MOE of the samples from formula M1-1, M2-1, M3-1, M4-1, and M5-1 as shown on Figure 4.

The mechanical strength of cement-based materials can further increase by using the accelerator admixture. In this work, ASBA has a high amount of SiO_2 and Al_2O_3 that could promote C-S-H formation from pozzolanic effect [23,24] when ASBA reacts with the $Ca(OH)_2$ to produce calcium silicate hydrate and calcium aluminate hydrate as shown in Equation (3) and (Equation (4) [13]:

$$Ca(OH)_2 + SiO_2 + H_2O \rightarrow xCaO \cdot ySiO_2 \cdot zH_2O \quad (3)$$

$$Ca(OH)_2 + Al_2O_3 + H_2O \rightarrow xCaO \cdot yAl_2O_3 \cdot zH_2O \quad (4)$$

Compared with the samples from formula M1-1 to M5-1, as shown on Figure 4, MOR and MOE of the samples from formula M1-2 to M5-2 are further improved when ASBA was added. Therefore, combination of the water to cement ratio reduction, and addition of ASBA improved MOR and MOE of the air-cured samples after 1 day of curing. Moreover, MOR and MOE of the samples from formula M5-2 are highest, and higher than that of R-AC28D samples, while they are comparable to that of R-ASC samples.

The physical properties including bulk density, and water absorption of the samples are shown in Figure 5(a) and Figure(b). Obviously, as the w/c ratio decreases, the density of samples increases, and their water absorption decreases. Moreover, the air-cured samples from formula M2-1 to M5-2 have higher density and lower water absorption than that of R-ASC samples. Again, the samples from formula M5-2 have highest density, and lowest water absorption. Low water absorption is beneficial for outdoor applications.

Microstructure of the samples from formula R-ASC, R-AC-1D, R-AC-28D, and M5-2 are shown in Figure 6(a-d), respectively. For formula R-ASC, the plate-shape tobermorite, the crystalline phase of C-S-H, is observed. Tobermorite is commonly found in the FRCC cured by ASC process. Obviously, this phase is only found in formula R-ASC. From Figure 6(a), beside tobermorite, some voids in the microstructure are revealed on the figure.

For formula R-AC-1D and R-AC-28D, only C-S-H gel and voids are observed. In Figure 6(c) for R-AC-28D samples, fiber-shape C-S-H is found with the voids in the cement matrix. For M5-2 samples, in Figure 6(d), both ettringite needle and fiber-shape C-S-H [25] are shown in the figure without the voids making M5-2 samples have better mechanical and physical properties than other samples.

Therefore, the relationship between mechanical properties, physical properties, and microstructure is examined. It is found that reducing the water-to-cement ratio can decrease voids in the cement matrix, and the high OPC content of the M5-2 sample can improve the C-S-H product. Ettringite needles can also fill the voids in the matrix, providing high bulk density and low water absorption. Additionally, the addition of ASBA in the mixture promotes the nucleation of C-S-H gel to a fiber-shaped form at ambient pressure during the 1-day curing period. This results in increased mechanical properties and bulk density, as well as reduced water absorption of FRCC.

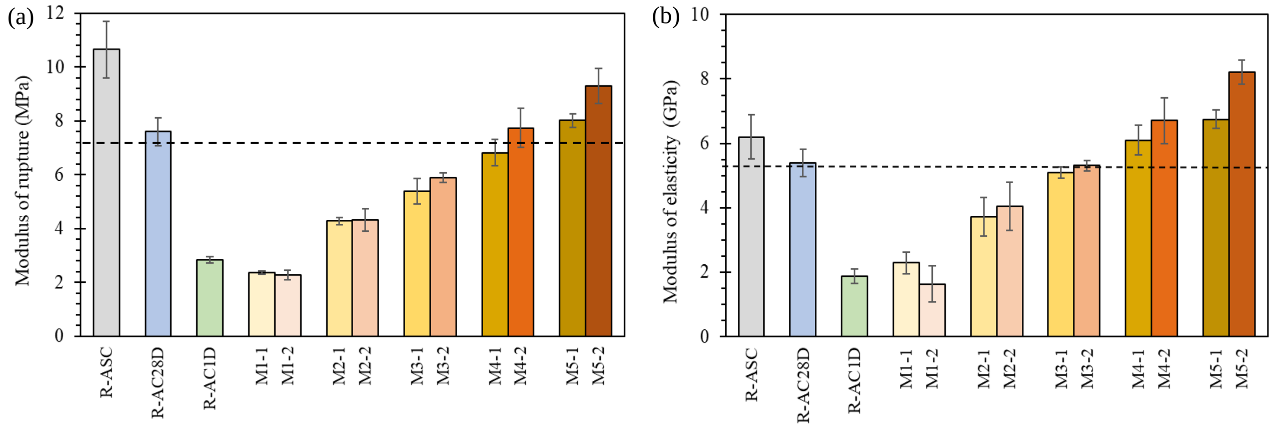

Figure 4. Mechanical properties of FRCC sample, consist of MOR and MOE.

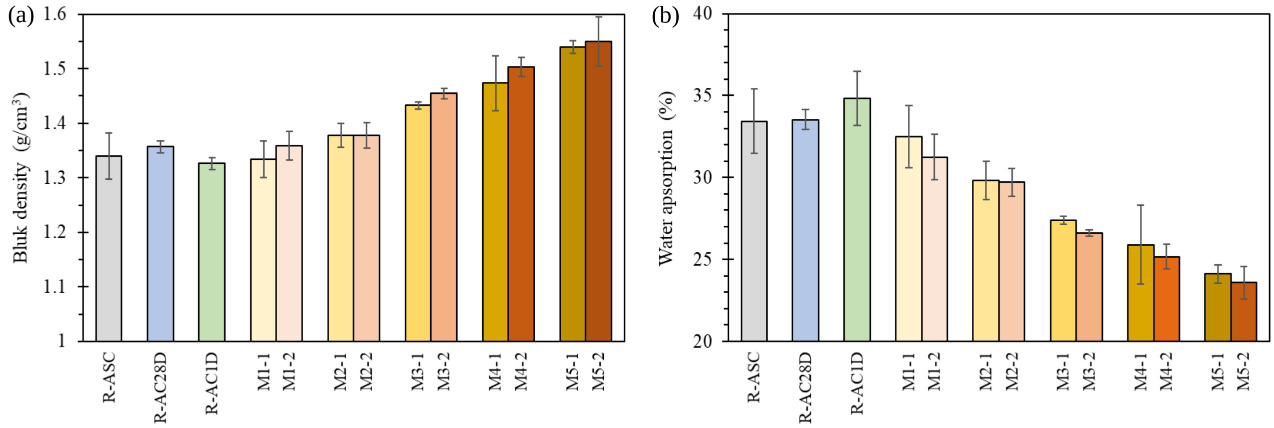

Figure 5. Physical properties of FRCC sample, consist of density and water absorption.

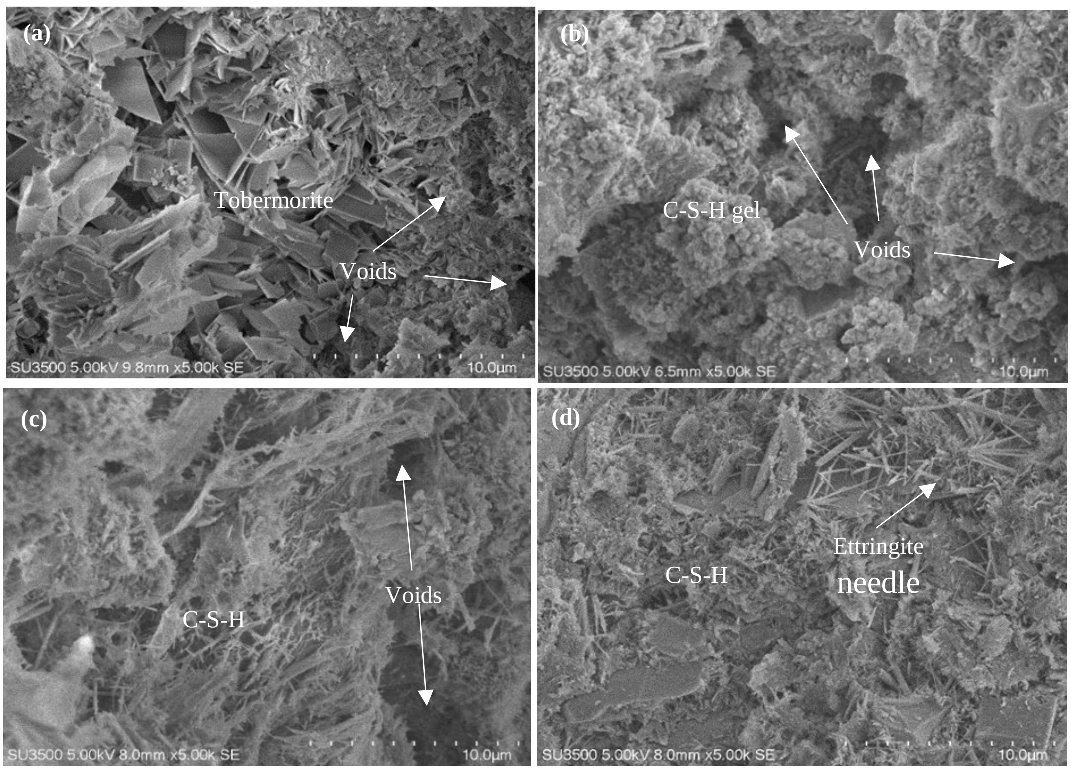

Figure 6. SEM image of FRCC sample from formula (a) R-ASC, (b) R-AC-1D, (c) R-AC-28D, and (d) M5-2.

4. Conclusions

This work's aim was to develop fiber reinforced cement composites having mechanical strength after 1 day of curing under atmospheric pressure and room temperature be comparable to the strength of FRCC cured by autoclave steam curing process. The green samples were produced by filter pressing process and then cured under the atmospheric pressure, at room temperature (about 27°C) and the relative humility between 50% to 60% for 24 h. The properties of these samples were compared with the properties of control samples cured by ASC process.

The results from experiment showed that lowering water cement ratio from 1.14 to 0.53 and using alumino-silicate complex as the chemical admixture in the mix design improved the modulus of rupture (MOR), modulus of elasticity (MOE), and bulk density, and reduced the water absorption of FRCC samples. After 1 day of air curing, the samples from formula M5-2 (water-cement ratio of 0.53 with ASBA) had highest modulus of rupture (MOR), highest modulus of elasticity (MOE), highest bulk density, and lowest water absorption. Compared with the control samples cured by the ASC process, MOR and MOE of the samples from formula M5-2 after 1-day air curing were comparable to that of the control samples. For physical properties, bulk density of the sample from formula M5-2 was higher than that of the control sample while water absorption was lower.

Acknowledgements

This work was supported by Faculty of Engineering, Kasetsart University [Grant No. 63/02/D. Eng], and Shera Public Company Limited.

References

[1] P. S. Intelligence. "Fiber cement market report: By raw material (portland cement, silica, cellulosic fiber), application (siding, roofing, cladding, molding and trimming), end user (residential, non-residential) – global industry analysis and growth forecast to 2030." https://www.psmarketresearch.com/ market-analysis/fiber-cement-market (accessed May 2022).

[2] P. Chakartnarodom, N. Kongkajun, N. Chuankrerkkul, P. Ineure, and A. W. Prakaypan, "Reducing water absorption of fiber-cement composites for exterior applications by crystal modification method," *Journal of Metals, Materials and Minerals*, Article vol. 29, no. 4, pp. 90-98, 2019.

[3] P. Chakartnarodom, W. Prakaypan, P. Ineure, N. Kongkajun, and N. Chuankrerkkul. *Feasibility study of using basalt fibers as the reinforcement phase in fiber-cement products, Key Engineering Materials*, vol. 766 KEM, pp. 252-257, 2018.

[4] P. Pahuswanno, P. Chakartnarodom, N. Kongkajun, and W. Prakaypan. *Feasibility study of using modified recycled fiber-cement for the production of high performance fiber-cement composites, Solid State Phenomena*, vol. 302 SSP, pp. 93-99, 2020.

[5] P. Sonprasarn, P. Chakartnarodom, N. Kongkajun, and W. Prakaypan. *Microstructure and mechanical performance of fiber-reinforced cement composites made with nucleating-agent activated coal-fired power plant bottom ash, Solid State Phenomena*, vol. 302 SSP, pp. 85-92, 2020.

[6] P. Chakartnarodom, W. Prakaypan, P. Ineure, N. Chuankrerkkul, E. A. Laitila, and N. Kongkajun, "Properties and performance of the basalt-fiber reinforced texture roof tiles," *Case Studies in Construction Materials*, vol. 13, p. e00444, 2020.

[7] B. Kottititum, Q. T. Phung, N. Maes, W. Prakaypan, and T. Srinophakun, "Early age carbonation of fiber-cement composites under real processing conditions: A parametric investigation," *Applied Sciences*, vol. 8, no. 2, 2018.

[8] P. Chakartnarodom, S. Wanpen, W. Prakaypan, E. A. Laitila, and N. Kongkajun, "Development of high-performance fiber cement: A case study in the integration of circular economy in product design," *Sustainability*, vol. 14, no. 19, 2022.

[9] L. Wang, H. Q. Yang, S. H. Zhou, E. Chen, and S. W. Tang, "Hydration, mechanical property and C-S-H structure of early-strength low-heat cement-based materials," *Materials Letters*, vol. 217, pp. 151-154, 2018.

[10] Z. Zhang, Y. Yan, Z. Qu, and G. Geng, "Endowing strength to calcium silicate hydrate (C-S-H) powder by high pressure mechanical compaction," *Cement and Concrete Research*, vol. 159, p. 106858, 2022.

[11] X. Ma, Z. Mei, and P. Ma, "Influence of water to cement ratio on mechanical performance of concrete canvas reinforced with warp-knitted spacer fabric," *Geotextiles and Geomembranes*, vol. 50, no. 4, pp. 708-719, 2022.

[12] N. P. Mailvaganam, M. R. Rixom, D. P. Manson, and C. Gonzales, (1999). Chemical Admixtures for Concrete (3rd ed.). CRC Press. https://doi.org/10.4324/9780203017241

[13] P. Sonprasarn, W. Prakaypan, S. Polsilapa, N. Kongkajun, E. A. Laitila, N. Chuankrerkkul, and P. Chakartnarodom, "The Influence of different crystal modifiers on ultra-low embodied energy curing fiber-reinforced cement composites," *Journal of Metals, Materials and Minerals*, vol. 32, no. 3, pp. 93-100, 2022.

[14] *ASTM C1185 Standard Test Methods for Sampling and Testing Non-Asbestos Fiber-Cement Flat Sheet, Roofing and Siding Shingles, and Clapboards*, ASTM International, West Conshohocken, PA, USA, 2016.

[15] *ASTM C948-81 Standard Test Method for Dry and Wet Bulk Density, Water Absorption, and Apparent Porosity of Thin Sections of Glass-Fiber Reinforced Concrete*, ASTM International, West Conshohocken, PA, USA, 2016.

[16] P. Sonprasarn, P. Chakartnarodom, N. Kongkajun, and W. Prakaypan, "Microstructure and mechanical performance of fiber-reinforced cement composites made with nucleating-agent activated coal-fired power plant bottom ash," *Solid State Phenomena*, vol. 302, pp. 85-92, 2020.

[17] B. Kondraivendhan, and B. Bhattacharjee, "Strength and w/c ratio relationship of cement based materials through pore features," *Materials Characterization*, vol. 122, pp. 124-129, 2016.

[18] S. B. Singh, P. Munjal, and N. Thammishetti, "Role of water/ cement ratio on strength development of cement mortar," *Journal of Building Engineering*, vol. 4, pp. 94-100, 2015.

[19] Z. Lu, J. Lu, Z. Liu, Z. Sun, and D. Stephan, "Influence of water to cement ratio on the compatibility of polycarboxylate superplasticizer with Portland cement," *Construction and Building Materials,* vol. 341, p. 127846, 2022.

[20] P. C. Hewlett, H. Justnes, and R. M. Edmeades, "14 - cement and concrete admixtures," in *Lea's Chemistry of Cement and Concrete (Fifth Edition)*, P. C. Hewlett and M. Liska Eds.: Butterworth-Heinemann, 2019, pp. 641-698.

[21] P.-C. Aïtcin, "17 - The Influence of the water/cement ratio on the sustainability of concrete," in *Lea's Chemistry of Cement and Concrete (Fifth Edition)*, P. C. Hewlett and M. Liska Eds.: Butterworth-Heinemann, 2019, pp. 807-826.

[22] P. Wang, L.-y.-w. Ke, H.-l. Wu, and C. K. Y. Leung, "Effects of water-to-cement ratio on the performance of concrete and embedded GFRP reinforcement," *Construction and Building Materials*, vol. 351, p. 128833, 2022.

[23] Y. K. Cho, S. H. Jung, and Y. C. Choi, "Effects of chemical composition of fly ash on compressive strength of fly ash cement mortar," *Construction and Building Materials,* vol. 204, pp. 255-264, 2019.

[24] P. Sonprasarn, P. Chakartnarodo, P. Ineure, and W. Prakaypan, "Effects of the chemical treatment on coal-fired bottom ash for the utilization in fiber-reinforced cement composites," *Journal of Metals, Materials and Minerals,* vol. 29, no. 4, 2019.

[25] J. Du, W. Meng, K. H. Khayat, Y. Bao, P. Guo, Z. Lyu, A. A. Obeidah, H. Nassif, and H. Wang, "New development of ultra-high-performance concrete (UHPC)," *Composites Part B: Engineering*, vol. 224, p. 109220, 2021.