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Impact of the Hybrid-Aluminum Additive on the Hydration Kinetics of Portland Cement in Fiber-Reinforced Cement Composites

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Abstract

A hybrid-aluminum additive (HAA) synthesized from industrial wastes including aluminum dross and flue gas desulfurized (FGD) gypsum was used as an additive for fiber-reinforced cement composites (FRCC). The impact of this additive on hydration kinetics was observed by the temperature change over time for the various HAA mixtures with ordinary Portland cement (OPC), sand, cellulose fibers, polyvinyl alcohol (PVA) fibers, and water, based on the method described in ASTM C186-98. The results showed that the hydration kinetics of OPC in the FRCC was improved by using HAA. In addition, when the amount of HAA was at 3% of the OPC weight, the hydration reaction rate was improved by 41%. The HAA additive acted as an accelerating agent by shortening the setting time and enhancing the temperature of the hydration reaction. This suggests that the cement paste can set faster, reducing the cycle time in FRCC processing. Even though further addition of the HAA increased the reaction rate, the setting time of OPC was too short to form a green sheet for the actual production of FRCC on an industrial scale. In addition, the heat released during the test, representing by the temperature change of the sample, was too high which could have a negative impact on the finished FRCC products.

Keywords: Hydration, Cement, Additive, Circular economy, Recycling, Waste

1. Introduction

The FRCC are construction materials commonly used in the roofing, flooring, wall, and ceiling of a building (Figure 1). Generally, this material exhibits exceptional mechanical properties and durability. The basic raw materials for manufacturing FRCC are ordinary Portland cement (OPC), silica sand, fibers, additives, and water. The mixtures are shaped into the green sheets by manufacturing processes such as filter pressing, Hatschek process, extrusion, mold casting, flow-on process, and roll forming. After molding, the green sheets are cured either by air or an autoclave. Normally, the water-cement ratio in FRCC production can be as high as 0.3, which significantly affects the kinetics of the hydration reaction between OPC and water during the curing stage.

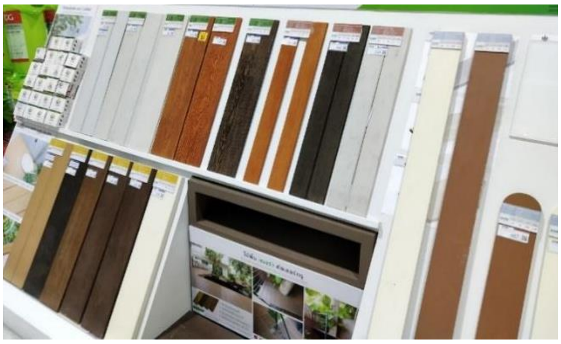

Figure 1. Various FRCC products with different applications available at a hardware store.

Cement additives are materials added to cement-based products to improve their performance and properties such as increasing hydration reaction rate and toughness, density, water absorption, and enhancements to the strength after curing. Therefore, an additive that can improve the reaction kinetics would be beneficial. Many researchers have studied the effect of additives on physical properties of cementitious materials (Chakartnarodom et al., 2020; Chakartnarodom Prakaypan, Ineure, Kongkajun, & Chuankrerkkul, 2018; Chakartnarodom, Wanpen, Prakaypan, Laitila, & Kongkajun, 2022; Khorami & Ganjian, 2011; Mohr, Nanko, & Kurtis, 2005; Pahuswanno, Chakartnarodom, Kongkajun, & Prakaypan, 2020; Sonprasarn, Chakartnarodom, Kongkajun, & Prakaypan, 2020). Previous work found that using polyurethane-based corn starch containing lithium perchlorate ($LiClO_4$) reduced the water absorption of fiber-reinforced cement composites (FRCC) due to the modified morphology of tobermorite, the crystalline calcium silicate hydrate (Chakartnarodom, Kongkajun, Chuankrerkkul, Ineure, & Prakaypan, 2019). Lagazzo, Vicini, Cattaneo, and Botter (2016) studied the use of fatty acid as a hydrophobic substance, these results showed enhanced water proofing of mortar. A study by Azarhomayun, Haji, Kioumarsi, and Shekarchi (2022) on aluminum powder and calcium stearate showed damp proofing of cement-based materials.

Accelerating additives, chemical admixtures introduced to cement mixtures, allow cement paste to set faster by increasing the rate of hydration and promoting early strength development. In FRCC processing, sufficient early strength is needed to enable earlier removal of formwork thus reducing the cycle time. The study of aluminum sulfate as an alkali-free accelerator for cement, Chen and Sun (2018), showed this shortened setting time, enhancing the degree of hydration and strength of cement paste due to the formation of a tight mesh structure in the ettringite phase.

Up to now, the generation of industrial waste has significantly increased worldwide. Due to environmental concerns, the recycling of industrial wastes as cement additives and/or as the raw material to reduce the usage of non-renewable raw materials has been extensively studied. Utilization of brick waste and soft sludge from the factory of FRCC in soil-cement brick production was studied by Kongkajun et al. (2020). The results showed improved strength and a reduction of the density and thermal conductivity of soil-cement bricks, in addition to the reduction of the usage of natural raw materials such as laterite. According to Sonprasarn, Chakartnarodom, Ineure, and Prakaypan (2019), Sonprasarn et al. (2020), Pahuswanno Chakartnarodom, Ineure, and Prakaypan (2019), and Pahuswanno et al. (2020), pretreated coal-fire bottom ash and FRCC waste could be used to enhance the hydration rate of Portland cement, the mechanical properties such as flexural strength and toughness of the FRCC product, and also reduce the natural raw materials used in FRCC production. Replacing Portland cement with paper sludge ash (PSA) by 12% reduced water absorption by 84% without a change in strength of materials (Wong, Barakat, Alhilali, Saleh, & Cheeseman, 2015). Pretreated crumb rubber was used as an additive to develop a hydrophobic rubberized cement paste (Chen, Shen, & Lee, 2021). The compressive and flexural strength of the concrete were maximized when 8% of short textile waste fibers containing cotton and polyester were added (Sadrolodabaee, Claramunt, Ardanuy & de la Fuente, 2021). Previous work by Li, Zhou, Zeng, Liu and Zhang (2019) added modified fenton paper sludge ash, which increased the bending and compressive strength after curing for 28 days.

At present, various kinds of industrial waste such as alumina waste and flue gas desulfurized gypsum (FGD) gypsum are blended during concrete production. Aluminum dross, a by-product of the aluminum melting process, consists mainly of alumina. Previous work by Mailar et al. (2016) showed that the utilization of recycled aluminum dross in concrete production yielded superior mechanical and durability properties. The FGD gypsum is a by-product generated from flue gas desulfurization process in coal-fired power plants. Yao, Wang, Liu, Yao and Wu (2019) investigated the utilization of fly ash, FGD gypsum, and carbide slag in a cementitious material to create a lightweight porous concrete. The results revealed that the synergistic effects of the various wastes improved the mechanical properties.

Waste recycling in cement and concrete products is an approach supporting circular and green economies. Thus, this work studied the effect of hybrid-aluminum additive (HAA) synthesized by both industrial wastes, aluminum dross and FGD gypsum, on the kinetics of the hydration reaction in FRCC.

2. Materials and Methods

The hydration reaction between water and the OPC components is an exothermic chemical reaction as shown below (Askeland, Fulay, & Wright, 2010).

$$3CaO \cdot Al_2O_3 + 6H_2O \rightarrow Ca_3Al_2(OH)_{12} + \text{Heat} \quad (1)$$

$$3CaO + SiO_2 + (x+1)H_2O \rightarrow Ca_2SiO_4 \cdot xH_2O + Ca(OH)_2 + \text{Heat} \quad (2)$$

In this work, heat evolution from the exothermic reaction of hydration was analyzed based on the ASTM C186 (1998) standard. Heat of hydration measurements are important for assessing the rate of cement hydration or the potential temperature rise in cementitious materials.

The dry mixtures consisting of OPC, cellulose fibers, polyvinyl alcohol (PVA) fibers and sand were prepared based on the compositions shown in Table 1. To form the slurry, a water to cement ratio (w/c ratio) of 0.3 was used. The control formula is signified by REF in Table 1, a common production formula of fiber-cement composites used in wallboard applications. The HAA was synthesized from aluminum dross and FGD gypsum in a H_2SO_4 solution with Al^{3+}:SO_4^{2-} in a ratio of 1:1.5. The HAA was initiated and developed under a technology licensing agreement with Shera Public Company limited. The HAA was added to the dry mixture based on the amount of OPC in the mixture. The amounts of HAA added varied from 0 to 5 wt.% of total OPC weight in the dry mixture.

Instrumentation for studying the heat evolution from hydration reaction of the Portland cement and water composed of an insulated chamber, thermocouple, and temperature data logger, shown in Figure 2. The cement paste was kept in an insulated chamber. The effect of the HAA additive on the hydration reaction of OPC and water is determined by observing the temperature change within the containers over time. The temperature-data logger collected the temperature measured by the thermocouple and was recorded every 30 sec, for a total measurement time of 16 h.

Table 1. Formulations for the dry mixtures.

Raw Material	**Formula Composition, wt%.**					
	REF	**HAA1**	**HAA2**	**HAA3**	**HAA4**	**HAA5**
OPC type I	70	70	70	70	70	70
Sand	25	25	25	25	25	25
Cellulose fibers	4	4	4	4	4	4
PVA fibers	1	1	1	1	1	1
HAA (% of OPC weight added to the mixture)	0	1	2	3	4	5

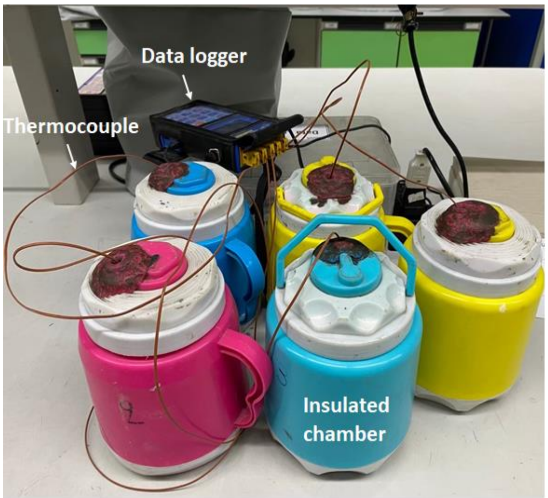

Figure 2. Schematic drawing of the instrument used for the heat of hydration test (Pahuswanno, et al., 2019; Sonprasarn et al., 2019).

3. Results and Discussion

The temperature profiles from the heat evolution of the hydration reactions are plotted in Figure 3. The temperatures initially increased with time, the cement paste was hydrated when reaching a maximum temperature (T_{max}) at the setting time (t_{max}), after which the temperature decreased significantly. When the amount of the HAA was increased from 1 to 5 wt.% of the OPC in the dry mixture, the temperature profile for all formulas using the HAA systematically shifted to shorter setting times (t_{max}) with higher maximum temperatures (T_{max}) compared to the control REF formula.

The average rate (R) of temperature change can be calculated from the difference of the maximum temperature (T_{max}) and the initial temperature (T_o), divided by the time to reach maximum temperature (t_{max}), as shown in the equation below (Sonprasarn et al., 2019).

$$R = \frac{T_{max} - T_o}{t_{max}} \qquad (3)$$

The data T_{max}, $T_{initial}$, and t_{max} from Figure 3 are listed in Table 2 for each formula. The value R_i is the average rate of the temperature change for the formulas HAA1, HAA2, HAA3, HAA4 and HAA5, while R_{REF} is the average rate of the temperature change for the control REF formula. The average rate of the temperature change R_{REF}, R_1, R_2, R_3, R_4 and R_5 are calculated by Equation (3). The R_i/R_{REF} ratio represents the degree of the hydration reaction, with a value greater than 1 an enhancement.

The R_i/R_{REF} ratios for all were greater than unity with the utilization of the HAA, Table 2. Moreover, the R_i/R_{REF} ratios increased when the amount of the HAA increased. Plotting these ratios for each HAA formula, Figure 4, showed increasing the HAA content linearly improved the kinetics of the hydration reaction of the mixture due to the positive slope with a R^2 value of 0.9933. Thus, the HAA acted as an accelerating additive in the cement mixtures by shortening the setting time and enhancing the degree of the hydration reaction. This result corresponded to previous work by Chen and Sun (2018), that showed that aluminum sulfate could shorten setting time and increase the early strength due to the formation of a tight mesh structure in the ettringite phase. The Al^{3+} and SO_4^{2-} ions in the HAA additive could react with tricalcium aluminate ($3CaO \cdot Al_2O_3$, C_3A) and gypsum phase ($CaSO_4 \cdot 2H_2O$) to produce a large number of ettringite ($Ca_3Al_2[SO_4]_3 \cdot [OH]_{12} \cdot 25H_2O$) crystals which promotes a shortened setting time for cement pastes.

Table 2. Data based on the temperature profiles in Figure 3, $T_{initial}$, T_{max}, t_{max}, R, and R_i/R_{REF}.

	REF	**HAA1**	**HAA2**	**HAA3**	**HAA4**	**HAA5**
T_o (°C)	28.6	27.7	28.6	30.9	34.4	33.9
T_{max} (°C)	66	69	69.2	69.2	72.1	75.7
Setting time, t_{max} (h)	6.7	6	5.21	4.57	4.15	4.05
$R = \frac{T_{max}-T_o}{t_{max}}$, (°C /h)	R_{REF} =5.58	R_1 =6.88	R_2 =7.79	R_3 =8.38	R_4 =9.08	R_5 =10.32
$\frac{R_i}{R_{REF}}$	1	1.15	1.31	1.41	1.53	1.73

However, when adding amounts of the HAA greater than 3%, the setting time was too rapid to shape the green samples and the maximum temperatures were very high (above 70°C) which could have a negative impact on the finished FRCC product. It is well known that the rate and amount of heat generation due to hydration are important. Non-uniform thermal expansion/contraction due to a very large amount of heat can create undesirable stresses in cement. This may cause detrimental cracking, reducing its strength. Thus, the amount of 3% for the HAA is selected for further study.

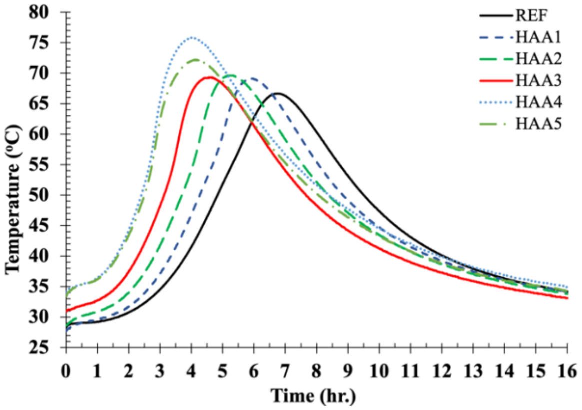

Figure 3. Temperature profile of the heat of hydration reaction of the mixtures.

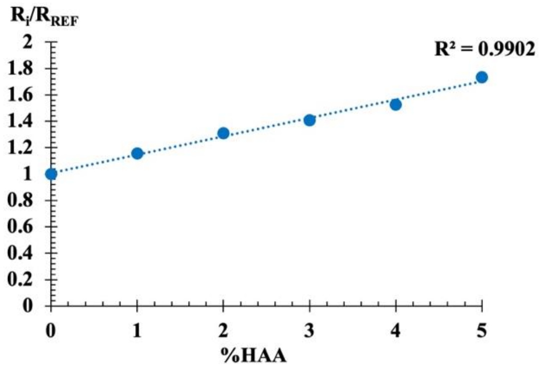

Figure 4. Effect of the % of HAA on the relative rate of temperature change R_i /R_{REF}.

4. Conclusions

This work studied the effect of the HAA synthesized by industrial wastes including aluminum dross and FGD gypsum on the kinetics of the hydration reaction of OPC in FRCC. It is found that the HAA systematically improved the hydration kinetics. However, the most suitable amount of HAA is 3% of the OPC weight in the dry mixture as higher amounts the setting time is too rapid with very high maximum temperatures. At this amount of the HAA the hydration reaction rate is significantly improved by 41%. This can dramatically alter setting times and thus cycle times reducing production times.

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Conflict of 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.

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