



Influence of aggregate type on conversion and strength in calcium aluminate cement concrete



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ABSTRACT

Calcium aluminate cement (CAC) is gaining popularity in North America as a rapid repair material due to its ability to quickly gain strength, even at low curing temperatures. Use of CAC has been limited due to a lack of understanding of the process of conversion and the role of aggregates in CAC concrete. Conversion, which occurs only in 100% CAC systems, is a chemical process in which metastable hydrates convert into denser, stable hydrates. Presented is an examination of aggregate source impacts on this conversion process and converted CAC concrete strengths. Nine different concrete systems with fifteen varying aggregate sources were examined. Results indicated that carbonate limestone and siliceous limestone aggregate systems had significantly less strength reduction due to conversion compared to siliceous aggregate systems. Microstructural analysis of systems suggested that the carbonate limestone system had less porosity and better-formed aggregate/paste interfacial transition zones compared to the siliceous systems. Chemical analysis of the concrete pore solution indicated that the carbonate limestone system's pH and ionic concentrations of aluminum, sodium, and potassium were significantly higher than that of a siliceous system, indicating more dissolution of unhydrated cement in the carbonate limestone system. These studies are presented along with a proposed theory explaining the cause of the significant converted strength differences in CAC concrete systems made with limestone aggregates compared to siliceous aggregates.

1. Introduction

Calcium aluminate cement (CAC) concrete has been used in the construction industry for over one hundred years, first gaining popularity as a rapid strength gain material to produce gun emplacements during WWI in France [1]. Today the material has a wide range of uses including as a temperature resistant material in refractory applications [2,3], scour resistance in dam spillways and wearing surfaces [4], and acid attack resistance for industrial floors and sewage applications [4–7]. More recently there has been significant interest in North America concerning the use of CAC concrete as a rapid repair material [8,9]. Due to its ability to gain strength rapidly, even at low temperatures, CAC will continue to be an important material for use in construction and repairing deteriorating infrastructure.

Despite the fact that CAC hydration has been studied significantly, it is still not well understood within the general construction industry. Furthermore, much of the research into CAC systems has focused on cement pastes and mortars and as a result, the impact of aggregates on CAC hydration and concrete properties is not well understood. Limited

previous work has shown that aggregate mineralogy can significantly alter the strength development and hydration in CAC concrete [10,11]. Despite these findings, however, no systematic study of the impact of different aggregate types on CAC concrete has been done. Of particular concern is the impact that aggregates have on strength development and conversion, a chemical process which causes porosity to form in the system over time.

1.1. Hydration and conversion in CAC systems

The following section discusses hydration in standard grade CACs and applies only to pure CAC systems. Additions of other cementitious products will alter the hydration products and change the overall chemistry of the system. Monocalcium aluminate (CA) is the main unhydrated cement phase in CAC and makes up > 40% of the phase composition of standard grade CAC systems [12]. CA will hydrate to form four main phases: CAH_{10} , C_2AH_8 , C_3AH_6 , and AH_3 . These hydrates fall into two main categories, metastable (CAH_{10} , C_2AH_8) and stable hydrates (C_3AH_6 , and AH_3) [12–14]. Which hydrate is initially formed

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Table 1
Oxide composition for CAC.

Oxide (% wt)	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	Na ₂ O	K ₂ O	SO ₃	TiO ₂	Mn ₂ O ₃	P ₂ O ₅	SrO	LOSS
CAC	4.98	38.23	15.4	37.53	0.71	0.03	0.23	0.06	1.8	0.23	0.13	0.02	0.65

depends on the temperature history of the system. When cured isothermally at temperatures below 15 °C, CAH₁₀ is primarily formed during initial hydration. When cured isothermally at temperatures between 15 °C and 27 °C, both CAH₁₀ and C₂AH₈ are formed during initial hydration [12,13], however as the temperature approaches 27 °C, formation of neither hydrate is favored, slowing setting time at this temperature [15–19]. When cured isothermally at temperatures above 27 °C, C₂AH₈ is the primary hydrate formed during initial hydration along with AH₃, a stable gel hydrate. CAH₁₀ and C₂AH₈ are both metastable hydrates and will undergo transitions to form the stable hydrate C₃AH₆ along with additional AH₃ gel. The speed at which this transition reaction occurs is dependent on the temperature and moisture state of the system [12–14] as well as w/cm [13]. As temperatures increase above 27 °C, the conversion of C₂AH₈ to C₃AH₆ occurs with increasing rapidity, therefore, higher curing temperatures result in faster conversion from metastable to stable hydrates. However, it is important to note that this reaction is thermodynamically inevitable and will occur at all temperatures [12,14].

The conversion reaction has significant impacts on porosity and strength, in the CAC system. Long-term the strength is based on the stable hydrates C₃AH₆ and AH₃, whereas early-age strength is dependent on the metastable hydrates CAH₁₀ and C₂AH₈. The rapid strength gain observed in calcium aluminate cements is a byproduct of the rapid precipitation through solution observed during hydration of CAC cement. Lamour et al. observed through transmission soft X-ray microscopy that as the induction period ends, there is a rapid formation of metastable hydrates resulting in a significant binding of water and filling of space [13,20]. The rapid space filling due to the formation of the metastable hydrates provides high early strength in these systems [13]. The strength developed from the metastable hydrates is transitory, however, as conversion occurs strength loss is observed in CAC systems. Converted CA hydrates (density (g/cm³): C₃AH₆ = 2.52 AH₃ = 2.4) are denser than unconverted hydrates (density (g/cm³): CAH₁₀ = 1.72, density of C₂AH₈ = 1.95) [21,22]. As the hydrates densify during conversion, they release water into the system and porosity is formed. The formation of porosity in the system results in a significant loss of strength. After conversion occurs, and a minimum strength has been reached, the water released by conversion can continue to hydrate unhydrated cement grains resulting in continued long-term strength gain [13,22]. After conversion occurs, long-term strength gain will be stable because once C₃AH₆ has nucleated, direct formation of C₃AH₆ from CA becomes favorable [23,24].

Table 2
Coarse aggregate description.

Aggregate name	Absorption capacity (%)	G _{sb, OD}	Source	Mineralogical description	Particle shape and texture
S_1_Coarse	2.58	2.44	Oregon, USA	Siliceous	Smooth, rounded
S_2_Coarse	1.71	2.66	Proprietary	Siliceous	Smooth, rounded
S_3_Coarse	1.31	2.56	Texas, USA	Siliceous	Smooth, rounded
S_4_Coarse	0.73	2.62	Wyoming, USA	Siliceous	Smooth, rounded
S_5_Coarse	1.55	2.55	New Mexico, USA	Siliceous	Smooth, rounded
S_6_Coarse	0.83	2.72	New Brunswick, Canada	Siliceous	Smooth, rounded
CL_1_Coarse	2.73	2.49	Texas, USA	Carbonate limestone	Coarse, angular
CL_2_Coarse	0.67	2.68	Washington, USA	Carbonate limestone	Coarse, angular
SL_1_Coarse	0.96	2.64	Ontario, Canada	Siliceous limestone	Coarse, angular

Note: In the naming convention S is “siliceous,” CL is “carbonate limestone,” SL is “siliceous limestone,” and “coarse” refers to the coarse aggregate portion of this aggregate source.

1.2. Impact of aggregate mineralogy on CAC strength and conversion

Previous work has observed that aggregate mineralogy can have a significant impact on concrete strength and conversion in CAC systems. Cussino and Negro showed that specimens containing limestone aggregates steadily gained strength over a 5-year period, even when similar specimens made with siliceous aggregates lost strength due to conversion [10]. Similar findings were observed by Lamour et al. [11].

Cussino and Negro's preliminary analysis observed the formation of calcium monocarboaluminate in the samples containing finely ground limestone (FGLS) (15 μm to 150 μm particle size diameter) as opposed to limestone aggregates (150 μm to 4.75 mm particle size diameter for fine aggregates, 4.75 mm and greater particle size diameter for coarse aggregates) [10,25]. Additional work has confirmed that the use of FGLS as a partial replacement for CAC results in the formation of calcium monocarboaluminate [10,11,26–34]. However, using a high surface area material such as FGLS to explain how a larger limestone aggregate, with significantly less surface area on which topochemical reactions may occur may not provide an accurate answer to the results discussed above. Cussino and Negro [10] and Lamour et al. [11] assumed that analogous reactions were occurring in the systems containing limestone aggregates, however they did not confirm the existence of calcium monocarboaluminates in the concrete systems containing 100% CAC. Additionally, the literature discussed above did not clearly distinguish the mineralogy of the limestone aggregates.

The work presented in this paper seeks to address the gaps in literature concerning the impact of aggregate source on CAC concrete; and to address calls for mechanistic work examining the impact of limestone aggregate on CAC conversion and strength.

2. Materials

2.1. Cement

A standard grade CAC was used for all concrete and mortar mixtures examined in this study. The oxide composition of the CAC used in this study is presented below in Table 1.

2.2. Aggregates

A wide range of aggregates were used through this study in concrete and mortar mixtures. Nine coarse aggregates and six fine aggregates from around North America were used. Absorption capacities and specific gravities (G_s) were measured according to ASTM C127 for

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