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Investigation of incorporating cinnamaldehyde into Lightweight Aggregate for potential corrosion reduction in cementitious materials

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ABSTRACT

Each year, corrosion of concrete results in billions of dollars' worth in damage. Cinnamaldehyde, a bioactive agent, can mitigate the corrosion of metals and potentially protect rebar within concrete. However, it cannot be incorporated into concrete during mixing since it negatively effects the hydration reaction between cement and water. To avoid these undesirable effects while keeping anti-corrosive properties in a cementitious mixture, an innovative approach through the use of Lightweight Aggregate (LWA) was taken. The experimental cinnamaldehyde-LWA mortar showed reduced compressive strength, heat evolution, and rebar pullout bond stress, but promising results regarding chloride threshold level and sorptivity.

1. Introduction

Concrete can prematurely deteriorate due to the corrosion of reinforcing steel. This is particularly critical since concrete is a staple building material worldwide of which nearly five billion tonnes is produced annually – approximately 1 tonne (1.102 tons) of concrete per person each year [1]. When aggressive media such as chloride ions from deicing salts or seawater diffuse through concrete, they can depassivate reinforcing steel and result in the production of expansive products, mainly iron oxides (rust). This buildup of rust can crack the concrete and lead to spalling [2]. Although there are numerous mitigation methods on the market today (e.g. epoxy coated rebar, waterproofing membranes, cathodic protection, etc.), $100 billion is still expended annually on corrosion related damage [3–5]. Therefore, novel methods for corrosion prevention are necessary.

This experimental program takes an innovative approach to corrosion mitigation. Cinnamaldehyde, the essential oil of cinnamon and a derivative of cinnamon bark, is a natural bioactive agent. This bioactive agent is commonly used in food for flavoring and in the medical field as a way to help those with diabetes [6]. However, it has also been shown to mitigate the corrosion of steel by forming a protective film on the surface of metals [7–9]. One drawback is that when cinnamaldehyde is included in a cementitious matrix it can coat cement particles and prevent the hydration of the cement, therefore causing a reduction of its compressive strength [10,11]. One way that cinnamaldehyde can be incorporated without interfering with the properties of concrete is by encapsulation in Lightweight Aggregate (LWA) [12,13]. Conventionally, LWA is presoaked with water and used to prevent early age cracking, a method known as 'internal curing' [14]. Over time, hydration of the cement causes an internal drop in humidity, causing the liquid absorbed by the LWA to be released in order to attain pressure equilibrium. Thus, by soaking the LWA in cinnamaldehyde, the cinnamaldehyde will be encapsulated within the LWA allowing it to be transported into the cementitious matrix and can be released once the early-age properties have developed [14]. After release, the cinnamaldehyde can diffuse towards the reinforcing steel and coat the steel to protect it against corrosion.

This research program further investigates a previous study by providing other considerations as well as additional experimental research [25]. This study investigates the potential impacts that cinnamaldehyde has on the cementitious matrix, and the efficiency of LWA as an encapsulation method for corrosion prevention agents. In doing so, six tests were carried out: compressive strength, to determine the mechanical effects of cinnamaldehyde incorporation; isothermal calorimetry, to examine hydration

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Table 1
Mix proportioning.

Mix number		Mix 1		Mix 2		Mix 3	
		Volume (cm^3)	Mass (g)	Volume (cm^3)	Mass (g)	Volume (cm^3)	Mass (g)
Sand (Total)		1375.0	3588.8	1046.7	2731.9	1046.7	2731.9
LWA (Total)		N/A[a]	N/A	328.3	492.5	328.3	492.5
#8	Sand	412.5	1076.6	314.0	819.6	314.0	819.6
	LWA	N/A	N/A	98.5	147.7	98.5	147.7
#16	Sand	343.8	897.2	261.7	683.0	261.7	683.0
	LWA	N/A	N/A	82.1	123.1	82.1	123.1
#30	Sand	275.0	717.8	209.3	546.4	209.3	546.4
	LWA	N/A	N/A	65.7	98.5	65.7	98.5
#50	Sand	206.3	538.3	157.0	409.8	157.0	409.8
	LWA	N/A	N/A	49.2	73.9	49.2	73.9
#100	Sand	137.5	358.9	104.7	273.2	104.7	273.2
	LWA	N/A	N/A	32.8	49.2	32.8	49.2
Mixing water		585.4	585.4	585.4	585.4	585.4	585.4
Cement		464.6	1463.5	464.6	1463.5	464.6	1463.5
Internal water		N/A	N/A	86.2	86.2	N/A	N/A
CA[b]		N/A	N/A	N/A	N/A	86.2	90.5

[a] Not Applicable.
[b] Cinnamaldehyde.

kinetics; sorptivity and diffusion of sodium chloride (NaCl) tests to determine transport properties; measurement of chloride threshold level; and rebar pullout.

2. Materials and methods

[1]Three mortar mixes were produced and tested. Mix 1 served as the control and was composed of local sand, ASTM C150 Type I/II cement, and water. The two other mixes (Mix 2 and Mix 3) were experimental and consisted of the same components as Mix 1, but included a partial replacement of the sand with presoaked LWA. Mix 2 contained water-LWA and Mix 3 contained cinnamaldehyde-LWA. The concentration of cinnamaldehyde (C_9H_8O) was provided by the manufacturer as 132.16 g/mol. Each mix had a water:cement ratio of 0.4 (not including the water stored within the LWA), and a 55 vol% of aggregate as shown in Table 1. It should be mentioned that these are arbitrary percentages in the acceptable and conventional range for mortar mix proportioning; and since they are the same for all the mixes, they do not affect the results. LWA was soaked in water for at least 24 h in a sealed container. After 24 h, the presoaked LWA was then incorporated into Mix 2 in a Saturated Surface Dry (SSD) state. The same method was used for encapsulating cinnamaldehyde in the LWA for Mix 3.

The LWA was commercially available expanded shale (Northeast Solite Corporation) with an absorption capacity of water of 17.5 wt% (as determined by the manufacturer). Mixes that included LWA involved a partial replacement of the local sand with LWA on a volumetric basis in order to retain the same particle size distribution as the sand. The amount of LWA needed for the mixes was calculated by Ref. [14]:

$$M_{LWA} = \frac{C_f \times CS \times \alpha_{max}}{S \times \Phi_{LWA}} \quad (1)$$

where M_{LWA} is the mass of dry LWA per unit volume of mortar (kg/m^3); C_f is the cement factor of the mortar (kg/m^3); CS is the chemical shrinkage of the cement (g of water/g of cement) (determined using ASTM C1608 [15] – Fig. 1); α_{max} is maximum degree expected of hydration of cement (dimensionless); S is the saturation degree of LWA (dimensionless); and Φ_{LWA} is the absorption capacity of the LWA (g water/g dry LWA) (Table 2).

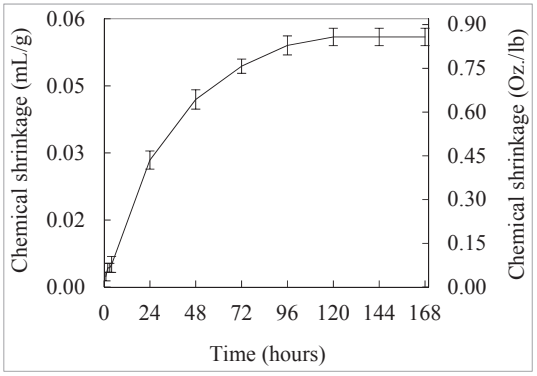

Fig. 1. Chemical shrinkage.

In order to investigate the chemistry of the mortars, Fourier Transformation Infrared Spectroscopy (FTIR) and X-ray Diffraction (XRD) were used for the identification of chemical species. The microstructures of the mortars, focusing on the Interfacial Transition Zone (ITZ), were further examined by Scanning Electron Microscopy (SEM).

The compressive strengths of all mixes were determined in accordance with ASTM C109 [16]. Once mixed, the mortar (at least three samples per mix) was immediately placed and tamped into 2 in. (50.8 mm) cube molds, sealed in a plastic bag, and stored in a fog room. Samples were demolded after 24 h, placed back into the fog room to cure, and tested at ages of 3, 7, 28, and 91 d.

Isothermal calorimetry was used to assess the hydration kinetics of the mortars. The heat flows of the mortars (two samples of each mix) were evaluated during the initial 24 h of hydration. Once the mortars were mixed, the appropriate amount was immediately placed in an ampoule and sealed. Heat flow was normalized by cement content.

Sorptivity tests were conducted following ATSM C1585 [17]. Mortar was placed into 4 × 2 in. (101.6 × 50.8 mm) cylinders, stored in a fog room, demolded after 24 h and placed back in the fog room to cure for 28 d (three samples per mix). On day 28, the samples

[1] Certain commercial equipment, instruments, or materials are identified in this report in order to specify the experimental procedure adequately. Such identification is not intended to imply recommendation or endorsement, nor is it intended to imply that the materials or equipment identified are necessarily the best available for the purpose.