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# Analysis of the effects of rice husk ash on the hydration of cementitious materials

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#### HIGHLIGHTS

• Propose a hydration model for rice husk ash (RHA) blended concrete.

- Simulate hydration of cement and pozzolanic reaction of RHA.
- Consider the effect of internal pores of RHA on capillary water content.
- Is valid for concrete with different water to binder ratios.
- Is valid for concrete with different RHA replacement ratios.

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#### ABSTRACT

Rice husk ash (RHA) is a highly reactive pozzolanic material produced by the controlled burning of rice husk, and it is widely used as a mineral admixture to produce high-performance concrete. The addition of the rice husk ash has complex effects (such as the cement dilution effect, the pozzolanic reaction, and the absorption and the release of mixing water) on cement hydration. Current models do not explain all these complex effects. This paper fills this gap by presenting an analytical model to simulate the hydration of the cement–RHA blends by considering both the cement hydration and the RHA reaction. The proposed model considers the influence of factors including the water to binder ratio, the RHA replacement ratio, the absorbed water in the RHA internal pores, the fineness of RHA (that is the mean particle size of the RHA), and the amorphous SiO<sub>2</sub> contents, on the hydration degree of the cement in the cement–RHA blends is improved due to the dilution effect. The calcium hydroxide contents in the cement–RHA blends using experimental data on the RHA replacement ratio. The proposed hydration model is verified by using experimental data on the RHA blended concrete with different water-to-binder ratios and different RHA substitution ratios.

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#### 1. Introduction

Rice husk ash (RHA), produced by the controlled burning of the rice husk, is used as a highly reactive pozzolanic material. RHA represents a significant improvement on the properties of both fresh and hardened concretes. The improvements in properties include the increased compressive and flexural strengths, the reduced permeability, the enhanced workability of concrete, and the reduced potential for efflorescence due to reduced calcium hydroxide [1].

There is abundance of experimental research on the physical and chemical properties of the concrete incorporating rice husk

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http://dx.doi.org/10.1016/j.conbuildmat.2015.12.086 0950-0618/© 2015 Elsevier Ltd. All rights reserved. ash. Feng et al. [2] find that the RHA replacement of cement increases the compressive strength of concrete, greatly decreases the average pore radius of the concrete, and decreases the amount of Ca(OH)<sub>2</sub> in concrete. Zhang et al. [3] propose that the rice husk ash is a highly reactive pozzolanic material that improves the microstructure of the interfacial transition zone (ITZ) between the cement paste and the aggregate in high-performance concrete. Yu et al. [4] find that for the RHA blended concrete, due to the formation of CSH gel and less portlandite, the properties of concrete (such as strength and resistance to acid attack, carbonation, and penetration) can be improved. Saraswathy and Song [5] found that the incorporation of the RHA up to a 30% replacement level reduces the chloride penetration, decreases permeability, and improves the strength and the corrosion resistance properties.

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Compared with the abundant experimental investigations on the properties of concrete incorporating the RHA [2–5], the research on theoretical models to predict the properties of the RHA blended concrete is quite limited. Feng et al. [6] studied the pozzolanic properties of the RHA by hydrochloric acid pretreatment. They propose that the kinetics of the reaction of the rice husk ash with lime is consistent with diffusion control and can be represented by the Jander diffusion equation. Using a blended cement hydration model, Nguyen [7] models the development of the microstructure of the cement–RHA paste and evaluates the contents of the calcium hydroxide in the hydrating cement–RHA paste with a water–binder ratio of 0.4.

On the other hand, some researchers propose hydration models for the blended concrete. Narmluk and Nawa [8–9] model the effect of the fly ash on the hydration kinetics of the cement in fly ash-cement with a low water-binder ratio at different curing temperatures. The modified shrinking-core model was used to quantify the reaction degree of the cement in the cement-fly ash blends. Breugel [10–11] modeled the hydration and the micro-structural development of the hardening concrete by considering factors including the particle size distribution of the cement, the chemical composition of the cement, the water-cement ratio and the actual reaction temperature. Papadakis et al. [12–13] propose a simplified scheme that describes the activity of the silica fume and the fly ash in terms of the chemical reactions. Furthermore, they evaluate the carbonation and chloride ingress through the stoichiometry of the chemical reactions and mathematical models.

Summarily, Narmluk and Nawa [8-9], Breugel [10-11], and Papadakis [12–13] show that the development of the properties of the blended concrete is related to both the cement hydration and the mineral admixtures reaction. The properties of the hardening concrete are related to the degree of hydration. On the other hand, Wang and Lee [14-15] proposed that the addition of the mineral admixtures mainly represents the dilution effect and the chemical effect on the cement hydration. The dilution effect is a consequence of the replacement of the cement by the mineral admixtures and it results in an increase in the water-cement ratio. The chemical effect is the pozzolanic reaction between the mineral admixtures and the calcium hydroxide. Additionally, the rice husk ash is a porous material, and water mixed with it will be absorbed into the porous structure of the RHA during the mixing process. Due to the self-desiccation of the hydrating cement-RHA blends, the absorbed water in the RHA will be released and will contribute to the hydration of the cement [1].

In this paper, a numerical model is proposed to simulate the hydration of the RHA–cement blends by considering the dilution effect, the chemical effect, the absorption of water mixed into the porous structure of RHA, and the release of absorbed water during the hydration process. The properties of the hardening concrete are predicted from the contribution of the cement hydration and the pozzolanic reaction.

#### 2. Hydration model of Portland cement

The shrinking-core model, originally developed by Tomosawa [16] and modified by Park [17] and Maruyama [18], is used in this study to simulate the development of the cement hydration. The model considers the rates of formation and destruction in an initial impermeable layer, the activated chemical reaction process, and the following diffusion-controlled process. This model is expressed as a single equation consisting of three coefficients:  $k_d$  representing the reaction coefficient in the induction period;  $D_e$  representing the effective diffusion coefficient of the water through the C–S–H gel; and  $k_r$  representing a coefficient of the reaction rate of the cement as shown in Eqs. (1) to (3) given below:

$$\frac{d\alpha}{dt} = \frac{3(S_{\rm w}/S_0)\rho_{\rm w}C_{\rm w-free}}{(\nu + w_{\rm g})r_0\rho_{\rm c}} \frac{1}{\left(\frac{1}{k_{\rm c}} - \frac{r_0}{D_{\rm c}}\right) + \frac{r_0}{D_{\rm c}}\left(1 - \alpha\right)^{\frac{-1}{3}} + \frac{1}{k_{\rm c}}\left(1 - \alpha\right)^{\frac{-2}{3}}}$$
(1)

$$D_{\rm e} = D_{\rm e0} \ln \left(\frac{1}{\alpha}\right) \tag{2}$$

$$k_{\rm d} = \frac{B}{\alpha^{1.5}} + C\alpha^3 \tag{3}$$

where  $\alpha$  is the degree of cement hydration; v is the stoichiometric ratio by mass of water to cement (= 0.25);  $w_g$  is the physically bound water in C–S–H gel (= 0.15);  $\rho_w$  is the density of water;  $C_{w-free}$  is the amount of water at the exterior of the C–S–H gel;  $r_0$ is the radius of the unhydrated cement particles ( $r_0 = 3/(S\rho_c)$ , the terms *S* and  $\rho_c$  stand for the Blaine surface area and density of the cement, respectively;  $S_w$  is the effective surface area of the cement particles in contact with the water and  $S_0$  is the total surface area if the surface area develops unconstrained;  $D_{e0}$  is the initial value of the effective diffusion coefficient; *B* controls the rate of the initial shell formation and *C* controls the rate of the initial shell decay.

The amount of water in the capillary pores  $C_{w-free}$  is expressed as a function of the degree of hydration in the previous step as shown in Eq. (4):

$$C_{\text{w-free}} = \left(\frac{W_0 - 0.4 * \alpha * C_0}{W_0}\right)^r \tag{4}$$

where  $C_0$  and  $W_0$  are the mass fractions of the cement and the water in the mix proportion, and *r* is an empirical parameter representing the accessibility of water into an inner anhydrous part through an outer hard shell of the cement particles [19–20].

The effect of the temperature on the reaction coefficients is assumed to follow Arrhenius's law [16–18]. By using the proposed Portland cement hydration model, Tomosawa [16] evaluated the heat evolution rate, the chemically bound water, and the compressive strength of the hardening concrete. Park et al. [17] predicted the temperature distribution in the high-strength concrete using this hydration model. A good correlation was found between the analytical results and experimental results.

#### 3. Hydration model for cement blended with rice husk ash

### 3.1. The amount of calcium hydroxide (CH) during the hydration process

The RHA is normally obtained by a controlled combustion of the rice husks and retains the porous structure of the husk. It consists essentially of silica in the amorphous form. The mean particle size of the RHA ranges from 5 to 10  $\mu$ m, which is much larger than that of the silica fume, but also the RHA has a very large surface area originating from the porous structure of its particles [4]. In fact, increasing the fineness of a pozzolanic material also increases its reactivity. It is expected that the RHA when pulverized yields finer particles producing the size distribution required for a higher reactivity.

The RHA is a highly pozzolanic material with a large amount of amorphous SiO<sub>2</sub>. Using the stoichiometry of the reaction of the amorphous silica proposed by Bentz et al. [21], the amounts of the calcium hydroxide in the cement–rice husk ash blends during hydration can be determined with the following equation:

$$CH = RCH_{CE} * C_0 * \alpha - 1.36 \times \alpha_{RHA} \times m_{RHA0} \times \gamma_s$$
(5)

where RCH<sub>CE</sub> is the mass of the produced calcium hydroxide from 1 gram hydrated cement;  $\alpha_{\text{RHA}}$  is the degree of hydration of the glass (active) phase of the rice husk ash;  $m_{\text{RHA0}}$  is the rice husk ash mass in the mixing proportion;  $\gamma_s$  is the mass percentage of the glass sil-

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