Construction and Building Materials 64 (2014) 22–27

Contents lists available at ScienceDirect

Construction and Building Materials

journal homepage: www.elsevier.com/locate/conbuildmat

# Chemical reactivity of lightweight aggregate in cement paste

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## HIGHLIGHTS

- All the types of LWA showed chemical activity, which absorb $Ca^{2+}$ and $OH^-$, and released $Si^{4+}$ into cement solution.
- The TGA and XRD results showed that a pozzolanic reaction between cement paste and LWA can be expected.
- The chemical reactivity of LWA in cement paste is not significant, but it may also make some contribution to the dense ITZ.

## ARTICLE INFO

*Article history:*
Received 18 October 2013
Received in revised form 9 March 2014
Accepted 2 April 2014
Available online 23 April 2014

*Keywords:*
Lightweight aggregate
Cement paste
Pozzolanic interaction
Fly ash
Calcium hydroxide content

## ABSTRACT

Quantitative investigation on the chemical reactivity of lightweight aggregate (LWA) in cement paste is important to enrich our understanding of the role played by LWA in influencing the microstructure of the interfacial transition zone (ITZ). In this paper, three types of LWA (YT, NT and FT) were chosen, which are based on shale rock, expanded clay and sintered fly ash respectively, and the ion concentration, calcium hydroxide (CH) content and reaction degree of LWA in cement pastes at different curing ages were studied, the fly ash (FA) was also included for comparison. The results show that, all the types of LWA studied showed chemical activity, which absorb very significant amounts of $Ca^{2+}$ and $OH^-$, and released significant amounts of $Si^{4+}$ into cement solution. The CH reduction content of the paste also indicates that the LWA have pozzolanic reactivity, especially in the later period the pozzolanic reaction become the main part of the hydration, but the reaction degree of them is slightly lower than that of FA. In addition, with the increase of the water–binder ratio, the reaction degree of FA and LWA has some increase as well. Although LWA in the form of a powder is more reactive than in the form of a coarse aggregate, the XRD results also show that the CH peak height of the ITZ samples around different types of LWA have a small decrease, in contrast to the bulk cement paste. The results obtained confirm that the dense ITZ around LWA was not only characterized by the water absorbing and mechanical interlocking, the chemical reaction at the interface also has some contribution.



## 1. Introduction

Concrete is a composite material with aggregate embedded in a cement paste matrix, between aggregate and cement paste there exists the interfacial transition zone (ITZ). For normal weight concrete, the ITZ is known to be the weakest region in concrete, influencing both mechanical properties and durability of concrete [1–3]. However, in the case of lightweight aggregate concrete, the microstructure of the ITZ is denser [4–7], thus the durability of concrete can be improved. Some studies postulate that the mechanisms responsible for the formation of the ITZ are related to the water absorbing and releasing character of lightweight aggregate (LWA) in concrete, and then the local water–cement ratio in the ITZ can be reduced [8,9]. Furthermore, a mechanical interlocking between LWA and cement paste can also be observed. As it is often assumed that the LWA used in concrete making has less chemical reactivity and the interaction with cementitious matrix is mostly physical, thus the influence of LWA chemistry on the properties of the ITZ has been given little attention.

The most LWA used for structure are artificial ceramsite, which are mainly made of expanded clay or sintered fly ash, and manufactured at a high firing temperature. The main chemical compositions of LWA are $SiO_2$ and $Al_2O_3$, which may have the ability to produce components having bonding property as a result of their reaction with calcium hydroxide (CH) in presence of moisture. For example, a certain degree of pozzolanic reaction between cement paste and LWA has been observed [10]. It has also been found that in the ITZ of lightweight aggregate concrete, there exists the chemical process, which is associated with pozzolanic activity of the aggregate and deposition of CH in the pores in the shell of the aggregate, and can have an influence on the concrete strength

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http://dx.doi.org/10.1016/j.conbuildmat.2014.04.024

[11]. From the investigations have shown above, it can be seen that both of the physical and chemical interactions between LWA and cement paste are responsible for the high strength of the ITZ, which are advantageous to the performance of concrete. Many studies have shown that the water absorption and pre-wetting degree of LWA can influence the interface physical process [12,13]. However, more research is needed to understand fully the chemical process between LWA and cement paste, which depends on the mineralogical composition, amount of pozzolanic phases, and the curing age.

According to numerous researchers, the thickness of the ITZ is only 40–50 μm, thus it is difficult to study the chemical reaction between LWA and cement paste directly, which is limited by the sampling. In this paper, three different types of LWA (YT, NT and FT), which are based on shale rock, expanded clay and sintered fly ash respectively, were chosen for the study. To investigate the pozzolanic reaction of aggregate–cement paste, the change of ions in a cement solution with finely ground material of each LWA was measured. Furthermore, the content of CH and combined water in a cement paste with finely ground LWA as a cement replacement was tested, as well as the reaction degree of LWA. The fly ash (FA) was also included for comparison. The results obtained are expected to enrich our understanding of the role played by LWA in influencing the chemical process of the ITZ.

## 2. Experimental program

### 2.1. Materials

Cement paste specimens were prepared using Portland cement type I, which is produced by the pure cement clinker mixed with 5 wt.% of gypsum. Three types of LWA (Fig. 1), YT, NT and FT which are based on shale rock, expanded clay and sintered fly ash respectively, were chosen for the study. The sintering temperature for their production is in the range of 1100–1150 °C. Fly ash (FA) used in this study conforms to Class F. Particle size distribution of LWA and FA is illustrated in Fig. 2. It can be seen that FA and NT have the finest and FT has the coarsest particle size distribution, whereas the particle size distribution of YT is in the middle. The chemical compositions of the LWA and FA are listed in Table 1, and their mineralogical compositions are estimated by means of X-ray diffraction (XRD), as shown in Fig. 3. The main minerals identified for NT and YT are similar, mainly quartz and ringwoodite. The XRD pattern for FT is different from that of YT and NT, mainly because of completely different raw materials, and the main minerals for FT are mullite, quartz, hematite and anorthite.

### 2.2. Mixture proportions

The LWA was ground into a fine powder with the same fineness as fly ash, and the specific surface area for YT, NT, FT and FA are 375, 382, 356 and 398 $m^2/kg$ (BET method), respectively. The mixes with FA and finely ground LWA as a cement replacement were produced with the mixture proportions as shown in Table 2.

### 2.3. Specimens preparation and testing

#### 2.3.1. Chemical interaction test

This test using the method described by Tasong [14]. First, cement-deionized suspensions (1 part of cement and 4 part of deionized water by weight) were mixed in 500 ml polythene bottles, using a mechanical tumbler. After about 3 h, the solid phases were filled off through a 0.45-μm membrane filter, and the cement solution containing $Ca^{2+}$, $K^+$, $Na^+$, $Al^{3+}$, $Si^{4+}$, $OH^-$ and other minor ions was obtained. Then 25 g of finely ground LWA was suspended in (a) 150 mL of cement solution, and (b) 150 mL of deionized water, under 70 °C condition for various time periods (i.e., 1, 3, 7, and 28 days). At last, the extracts were taken from the suspensions and analyzed for metal ions, sulfur, and hydroxyl ions using Inductive Coupled Plasma (ICP) methods and titration. The test process is shown in Fig. 1. Comparison of the results with those for the original cement solution (which serves as the control) may indicate a potential LWA–cement chemical interaction.

#### 2.3.2. Content of CH and combined water test

Fresh pastes were mixed with the proportions as shown in Table 2, and filled into plastic centrifugal tubes to prevent moisture loss and carbonation. Then the pastes in centrifugal tubes were cured at 70 ± 1 °C to accelerate the pozzolanic reaction. At 3, 7 and 28 days of age, the paste samples were soaked in acetone in order to stop hydration and ground into a fine powder.

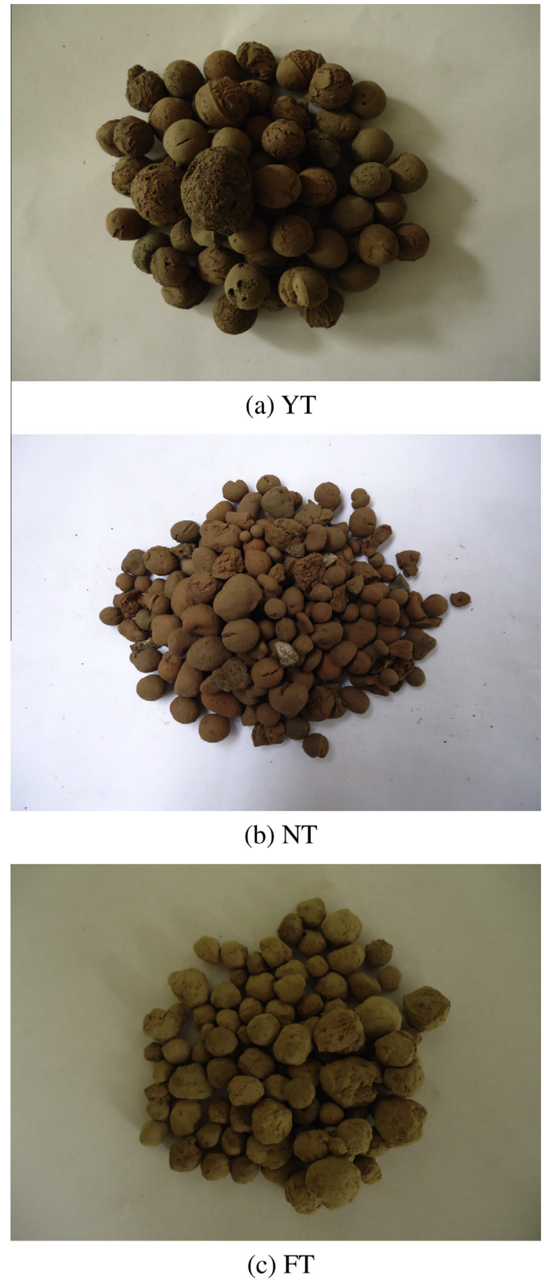


**Fig. 1.** LWA used in this study.

In order to study the pozzolanic activity of LWA and FA, the cement paste with finely ground LWA (FA) as cement replacement was monitored by Thermogravimetric Analysis (TGA) conducted at various ages, and the content of CH and combined water was calculated. For each measurement a sample of 150 mg was heated in a $N_2$ atmosphere at a rate of 10 °C/min.

#### 2.3.3. Reaction degree of LWA and FA test

The reaction degree of LWA and FA in cement paste samples were tested according to the hydrochloric acid selective dissolution method of Chinese (GB/T12960-2007) standards. First, one part hydrochloric acid was mixed with two parts deionized water. Then this solution was used to dissolve pasted samples at 40 ± 2 °C. After extracting the filtrates, the residues were dried to constant weight. At last, by deducting the part of LWA and FA dissolved in hydrochloric acid and the part of cement undissolved in hydrochloric acid, the reaction degree of LWA and FA can be calculated.

#### 2.3.4. Mineral compositions of the ITZ test

The saturated LWA and cement paste with a water–cement ratio of 0.3 were mixed and molded. After 24 h, the specimens were demolded and cured for 28 days, at 90% of relative humidity and approximately 20 °C constant temperature. Then