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Huang, Hao; Ye, Guang

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Use of rice husk ash for mitigating the autogenous shrinkage of cement pastes at low water cement ratio

Hao Huang, Guang Ye
Faculty of Civil Engineering and Geoscience, Delft University of Technology, Netherlands

It is well recognized that the high risk of early age micro-crack of HPC/UHPC is attributed to the large magnitude of early age autogenous shrinkage caused by self-desiccation in binder hydration. Over the years, several methods have been proposed to mitigate autogenous shrinkage based on internal curing theory, and a better internal curing agent is always being sought.

Rice husk ash (RHA), was recognized having potencial to be an internal curing agent. In this paper, the effect of RHA on mitigating the autogenous shrinkage has been evaluated by different mean particle sizes of RHA, and the internal RH change was measured at the same situation. The results show a high efficiency of RHA for internal curing purpose, to drastically reduce the autogenous shrinkage of cement pastes. Comparing to the cement paste without RHA, the internal RH results show that the internal RH in pastes with RHA is evidently higher at either early or later ages. These were addressed in order to explain the mechanism RHA mitigates the autogenous shrinkage of pastes.

Keywords: autogenous shrinkage, rice husk ash, internal RH.

1 Introduction

Ultra high performance concrete (UHPC) and high performance concrete (HPC) shows excellent performances on ductility, compressive strength, and durability [1]. However, UHPC/HPC can experience large shrinkage deformation. A very high autogenous shrinkage already found in the first one or two days after casting, which points to a considerable cracking potential at early ages [2-3]. Such early age cracking due to restrained autogenous shrinkage tends to negate the numerous advantageous properties of HPC and UHPC and significantly limits their prospective utilization in construction. The high autogenous shrinkage of these concrete is due to the low water binder ratio and high amount of silica fume used which causes a significant drop in internal relative humidity (RH) in the cement paste during hardening, and self-desiccation occurs in absence of an external source of water [4]. Autogenous shrinkage and self-desiccation of concrete are the phenomenon known since the year 1900 [5], but its practical importance has only been recognized in last decade. Though the actual mechanism of autogenous shrinkage is unclear [6] until now, while there is general agreement about the existence of a relationship between autogenous shrinkage and RH changes in hardening cement paste [7,8].

Seeking the ways to reduce or to limit the autogenous shrinkage of HPC/UHPC is becoming very important for engineering practice. Report [9] has shown that mitigation of autogenous shrinkage using external curing is not effective; due to the fact that the very dense microstructure of HPC/UHPC enables only very slow water penetrate into the interior of concrete members. Because of this, internal water curing is considered to be an effective solution to counteract autogenous shrinkage for the low w/b ratio cementitious system, thereby reducing the likelihood of early-age cracking. Over the years, several methods like using lightweight aggregate [10] and super absorbent polymer [11], have been proposed to mitigate autogenous shrinkage based on internal curing theory, and a better internal curing agent is always being sought.

Rice husks are a by-product of the rice paddy milling industry. On the average, each tone of rice husks, on completion of combustion, produces 200 kg of ash. The surface area of the incinerated ash ranges from 60 to 100 m^2/g when determined by nitrogen adsorption. The chemical analysis of the ashes produced by the incineration of rice husks show that the silica content ranges from 85 to 95 percent, alkalis (K_2O and Na_2O) from 1 to 2 percent, and unburned carbon from 3 to 18 percent [12].

The use of rice husk ash (RHA) in concrete is not new [13]. RHA is a mineral admixture for concrete and a lot of data [13-14] has been published about its influence on the mechanical properties and durability of normal concretes. But for the effect of the RHAs on autogenous shrinkage of cement pastes at low water cement ratio, the relevant research is scarce and vague. Tuan [15] found that the autogenous shrinkage of UHPC can be mitigated by RHA with certain particle size. However, the mechanism of this phenomenon is not revealed in the research. In this paper, the effect of different mean particle sizes of RHAs on autogenous shrinkage of cement pastes was studied. The internal RH change and compressive strength of cement pastes with and without RHAs addition, were measured to estimate the potential of RHA for being an internal curing agent.

2 Materials and Methods

2.1. Materials and mixture proportion

The materials used in this study were Portland cement (CEM I 52.5N), RHA, and a polycarboxylate-based superplasticizer with 30% solid content by weight. The RHA was produced by a drum incinerator which was introduced in [15]. The well-burned ash was grinded to different particle size by ball mill. The mean particle sizes of these RHAs were respectively 7, 9 and 12 μm. The physical properties of these RHAs were shown in Table 1. The chemical composition of cement and RHA are shown in Table 2.

Table 1 Physical property of different types of RHA

	RHA-7	**RHA-9**	**RHA-12**
Mean particle sizes, μm	7.09	8.94	12.08
Specific surface area (BET nitrogen absorption), m^2/g	25.2	30.1	28.8
Total Pore Volume (Mercury Intrusion Porosimetry), mL/g	0.9773	1.0310	0.9838

Table 2 Chemical composition of cement and RHA used in this study

Components	**Cement***	**RHA****
Chemical properties, % by weight		
CaO	64.00	1.72
SiO_2	24.00	88.86
Al_2O_3	5.00	0.30
Fe_2O_3	3.00	0.20
SO_3	2.40	0.60
Na_2O	0.30	-
K_2O	-	4.22
Loss on ignition (L.O.I)	1.30	2.51

**) Data provided from the company*

***) Chemical composition determined by X-Ray Fluorescence Spectrometry method*

Four types of cement mixtures were made. The mixture proportion is listed in Table 3. Due to the high surface area of RHAs, the workability of cement pastes is dramatically reduced with

RHA additions [15]. In this paper, cement pastes with RHAs is designed to have good workability by using proper amount of superplasticizer.

Table 3 Mix compositions of pastes used to study autogenous shrinkage

Mixture	w/b ratio (by weight)	Superplasticizer/binder ratio (by weight)	RHA (%, by weight)	The mean particle size of RHAs (μm)
REF 0.25	0.25	1.6%	0	
RHA-7	0.25	1.6%	20	7.09
RHA-9	0.25	1.6%	20	8.94
RHA-12	0.25	1.6%	20	12.08

2.2. Test method

Autogenous deformation tests: The autogenous deformation of mixtures was measured following ASTM C1698 standard developed by Jensen and Hansen [16], in which three sealed corrugated moulds of 440 mm (φ28.5 mm) were tested for each mixture. After mixing, the fresh paste was carefully filled into corrugated tube and sealed by plug and sealing glue. The specimens and test instrument were immersed in glycol in a box where the temperature was maintained at 20 ± 0.1 °C with the help of water bath. The autogenous shrinkage of specimens was recorded every 5 minutes by linear variable differential transformers (LVDTs) automatically. Three parallel samples were measured for each mixture. Figure 1 shows the autogenous shrinkage test system [17]. The deviation of autogenous deformation of three parallel samples in each test group is less than 50 micro strains.

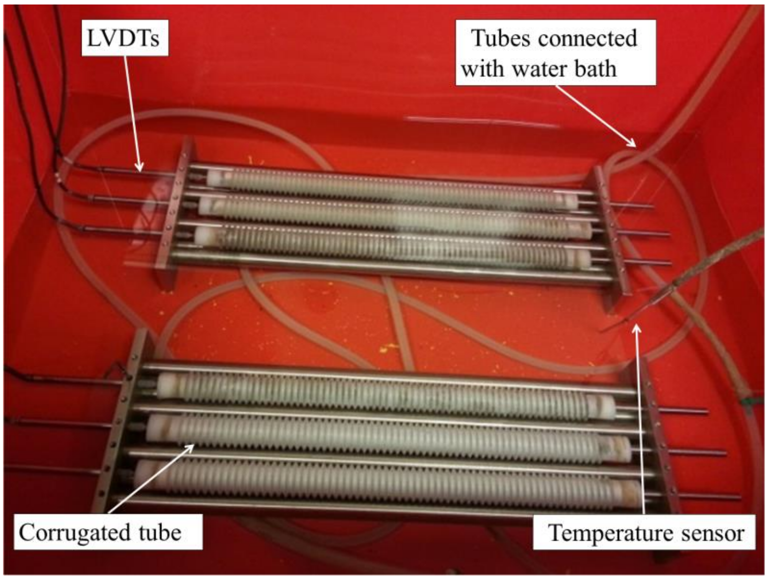

Figure 1: Autogenous deformation measurement. Specimens and test instrument were immersed in glycol which maintained the temperature at 20 ± 0.1 °C with the help of water bath. The autogenous deformation of specimens was recorded by LVDTs automatically.

Internal RH measurements: The internal RH inside the cement paste was monitored by two Rotronic hygroscopic DT station equipped with HC2-AW measuring cells, which is the same test method used by Jensen [18]. In practice, the internal RH of the mixtures always reached 100% within 24 h after mixing, even the temperature deviation of test environment was less than 0.1 ^{o}C and the instrument was calibrated. That's because in the high RH environment, water vapor is very easy to condense (RH=100%) on the surface of colder object (humidity sensor) [14].

In order to avoid condensation on the humidity sensor, a test procedure of internal RH measurement was developed and shown in Figure 2 [17]. The specimen was sealed in sample holder by a sealing plug and put in test room with temperature of 20 °C for 0.5 hour to achieve temperature equilibrium after mixing (Figure 2 b). The sample holder was kept in very tight sealed condition in order to prevent moisture loss. Considering the temperature of water vapor from cement pastes is a little higher than pastes due to hydration heat of cement, the temperature of water bath is set to 19.5 ± 0.1 °C for lowering the temperature of cement pastes. Afterwards the sealed sample and sample holder were put in the water bath for at least 0.5 hour to achieve temperature equilibrium (Figure 2 c). Then the sealing plug was replaced by humidity sensor to start the test (Figure 2 d). The humidity sensor was kept in a climate chamber at a slight higher temperature (for example, 20.3 °C) prior to the test for avoiding vapor condensation. The total time for the preparation before test is about 1 hour. The internal RH changes can be captured since the first hour after mixing, which is very hard to be obtained at so early ages before. The internal RH in the specimen and the temperature of water vapor were continuously measured by every 3 minutes for a period of 1 week after mixing. The RH sensors were calibrated by three saturated salt solutions in the range of 65-95% RH before and after every test. According to the calibration, the maximum measurement error of the RH sensors was ± 0.5%.

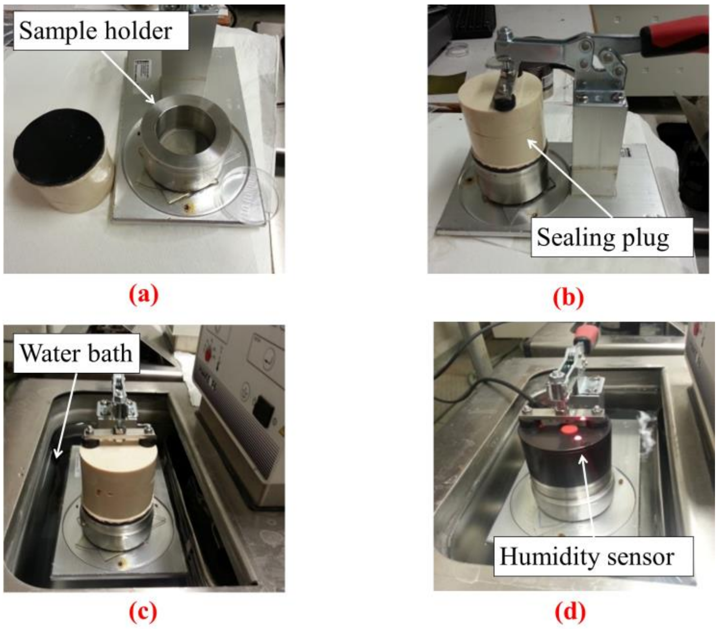

Figure 2: Internal relative humidity test. (a) Put the cement pastes into sample holder after mixing; (b) Specimen was sealed in sample holder and cured in a room with temperature of 20 ° C for 0.5 hour; (c) the sealed specimen and sample holder were moved in the water bath where the temperature was kept at 20 ± 0.1 ° C for at least 0.5 hour; (d) Humidity sensor (with slight higher temperature (20.3 ° C)) was placed to start the test

Compressive strength measurement: The compressive strength of cement mixtures was measured at the age of 1d, 3d and 7d. The size of specimen is 40 × 40× 40 mm. Three parallel samples were measured for each test. The deviation of compressive strength of three parallel samples in each test group is less than 4%.

3 Results and discussion

The internal RH changes of four mixtures in 7 days after mixing were shown in Figure 3. It can be found that the internal RH of mixtures with RHA is higher than the mixture without RHA in 7 days after 2.5 days of curing.

The free strain measured on cement mixtures after casting were shown in Figure 4. The strain is zeroed at final set. The ASTM 1698 proposes to use final setting time determined by Vicat apparatus as the “time-zero” of autogenous shrinkage. However, due to the relative arbitrariness of the penetration method, some researchers [19-22] query the reliability of using final setting time as “time-zero” for autogenous shrinkage. They believe that the penetration method does not precisely correspond to “time-zero”. As discussed in [17], when the “stable” solid skeleton is formed in hardening cement paste, empty pores could form inside the pastes due to chemical shrinkage and air-water menisci occur. When the air-water menisci form, the RH of pore solution begins to decrease. At the same time, the capillary tension come from the air-water menisci starts to act on the solid skeleton, and results in the autogenous shrinkage. With the water consumption by hydration, the Kelvin radius of menisci continuously decreases (Kelvin’s law); the solid skeleton sustains more and more capillary tension. This will lead to a considerable crack potential when the specimen is restrained. The formation of “stable” solid skeleton is a precondition for the RH drop, but it doesn’t mean that the RH must decrease right after the formation of “stable” solid skeleton. It’s more reasonable to link the “time-zero” to the start of self-desiccation, i.e., use the start time of internal RH drop as the “time-zero” of autogenous shrinkage for estimating crack potential. Table 4 shows the final setting time and “time-zero” (start time of self-desiccation) for each mixtures. Figure 5 shows the autogenous shrinkage of cement mixtures with this “time-zero”.

Table 4 Final setting time and “time-zero” of autogenous shrinkage for each mixtures

Mixture	**Final setting time (h)**	**“Time-zero” (start time of self-desiccation) (h)**
REF 0.25	17.28	21.97
RHA-7	11.23	24.98
RHA-9	8.82	21.03
RHA-12	7.22	20.43

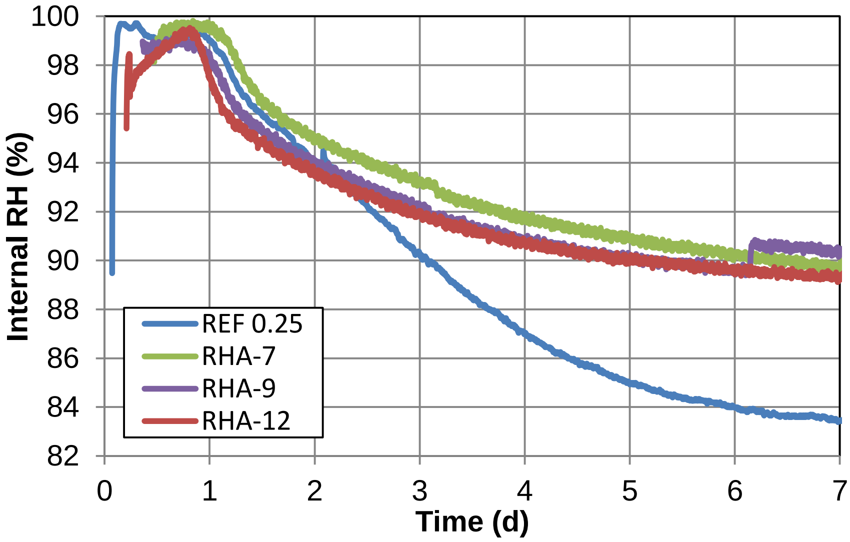

Figure 3: Internal RH change of different cement mixtures in 7 days

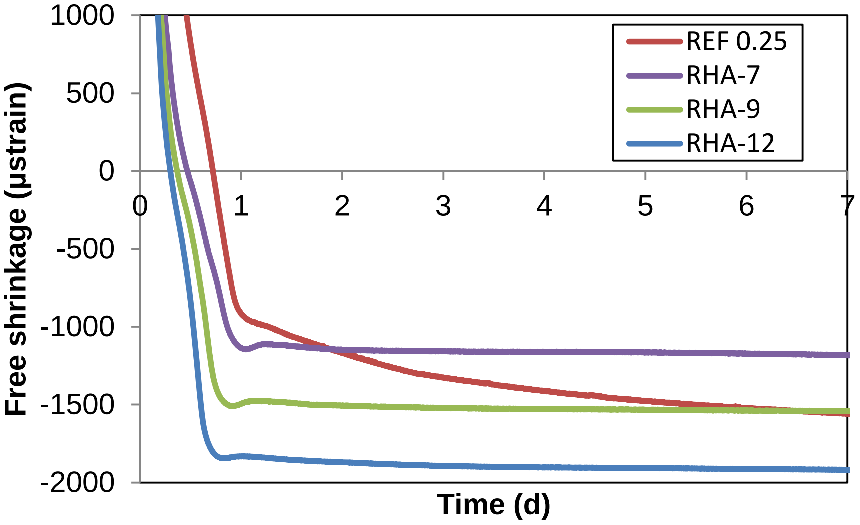

Figure 4: Free strain measured on different cement mixtures after casting. Strain is zeroed at final set.

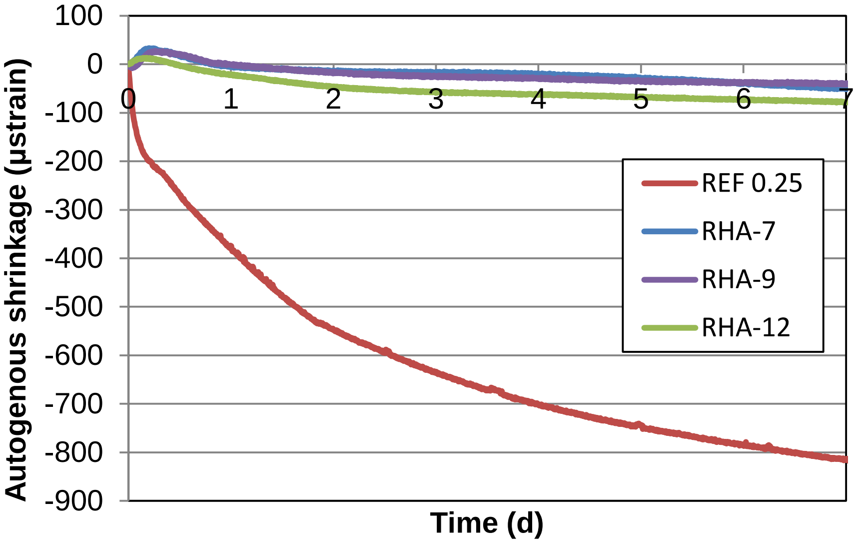

Figure 5: Autogenous shrinkage of different cement mixtures with "time-zero" (the start time of self-desiccation).

In Figure 5, it is shown that the autogenous shrinkage of cement mixtures with RHAs is clearly less than the autogenous shrinkage of cement mixture without RHA. The autogenous shrinkage of cement pastes is dramatically reduced to be less than 100 µstrain by RHA addition. The different particle sizes of RHAs show the slightly different efficiency on mitigating the autogenous shrinkage.

Figure 6 shows the compressive strength of different cement mixtures at the age of 1d, 3d and 7d. Due to the same addition amount of superplasticizer in each mixtures, and without the absorption effect of RHA, the mixture REF 0.25 has the longest final setting time (17.28h). The delayed coagulation effect of superplasticizer made the hydration time of mixture REF 0.25 is less than other mixtures with RHAs. This is the reason why the compressive strength of mixture REF 0.25 at 1d is distinctly lower than others. For the later ages, the experimental results show the positive effect of RHA on increasing the compressive strength of cement mixtures.

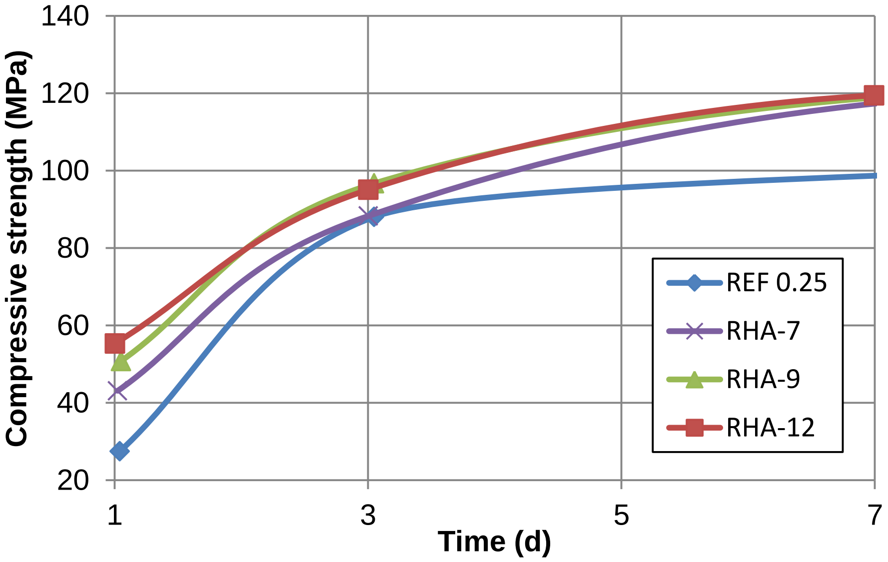

Figure 6: Compressive strength of different cement mixtures at the age of 1d, 3d and 7d.

Before the discussion on the mechanism of mitigating the autogenous shrinkage by RHAs, the formation mechanism of autogenous shrinkage need to be described. In the sealed condition when cement is mixed with water, the cement paste is in fluid state, the chemical shrinkage from cement hydration has been fully transformed into external volume change [**Error! Bookmark not defined.**]. The volume change of cement pastes caused by chemical shrinkage does not lead to a cracking potential at this stage [23-24]. With the increase of cement hydration, a “stable” solid skeleton is formed in the hardening paste. Since then, the chemical shrinkage cannot totally transform into external volume change, empty pores are formed inside the pastes and air-water menisci occur [25]. With water consumption by cement hydration, bigger pores inside the solid skeleton empty first [7]. Simultaneously, a drop in the RH occurs, i.e., self-desiccation happens. The internal RH drop will increase the main driving force for autogenous shrinkage in high RH, the capillary pressure. The lower internal RH, the bigger capillary pressure, according to Kelvin equation. These pressures act on the cement matrix, and force it to shrink. The stiffness of cement matrix represents the resistance ability to shrinkage. The autogenous shrinkage is the result from interactions between driving force and resistance ability.

For the cement pastes with RHA addition, on the one hand, the higher internal RH means the smaller driving force for autogenous shrinkage; on the other hand, the higher compressive strength means the higher stiffness for resisting the autogenous shrinkage. With the combined action of these two factors, the autogenous shrinkage of cement pastes with RHA should be less than the one without RHA, which is also found in experiment (Figure 5).

Due to high specific surface area and porous structure (Table 1), RHA is suspected to act as water reservoir to absorb water during the mixing. The absorbed water then is released to restrain the decrease of internal RH caused by cement hydration at early age. This internal curing effect from RHA explains the phenomenon that the cement mixtures with RHA have higher internal RH. Because of different total pore volumes and pore structures (Table 1), different particle sizes of RHAs have different efficiency on internal curing.

The improvement of the compressive strength of cement paste by using RHA may be explained by the enhancement of the packing density of granular mixtures, and the internal water curing of RHA. The hydration degree of cement at low water cement ratio is relatively low. In this situation, the filler effect of mineral admixtures, such as SF which was found by Detwiler and Mehta [26], is primarily responsible for the strength improvement. In this respect, the substitution of cement by RHA with a smaller particle size will improve the packing density of the granular mixture and thus increase the resulting compressive strength of cement pastes. RHA absorbs a certain amount of water during mixing. This absorbed water, as discussed above, will release to the cement matrix and promote the hydration of cement at later ages. This effect will reduce the crack potential and increase the compressive strength of cement paste.

4 Conclusions

The conclusions can be drawn as below:

- RHA has considerable influence on autogenous deformation and the internal relative humidity change of cement paste. RHA shows the same function like internal curing agent, markedly decreases the autogenous shrinkage as well as increase autogenous relative humidity at the early age of cement paste.
- Particle size and pore structure of RHA affects the efficiency of internal curing.
- RHA shows the positive effect on increasing the compressive strength of cement mixtures in later ages.

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