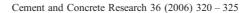


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Internal relative humidity distribution in high-performance cement paste due to moisture diffusion and self-desiccation

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Abstract

This investigation was carried out to study internal relative humidity (IRH) distribution of cement paste made with different water/cement ratios (w/c) and mineral admixtures in isothermal drying conditions. IRH changes in cement paste resulting from self-desiccation and moisture diffusion, respectively, at different ages were studied. The change laws of IRH in cement paste resulting from combining moisture diffusion with self-desiccation were discussed. The results indicate that IRH reduction of cement paste with w/c higher than 0.4 is mainly affected by moisture diffusion. However, IRH reduction of cement paste with w/c no higher than 0.4 is controlled by both moisture diffusion and self-desiccation. With the decrease of w/c, IRH reduction of cement paste resulting from self-desiccation increases, and IRH reduction resulting from moisture diffusion decreases at a given age. IRH decrement of cement paste incorporated with silica fume and ground blast-furnace slag is higher than that of control paste. w/c and the distance to the exposed surface play a significant role in IRH change resulting from moisture diffusion in isothermal drying condition. Change laws of IRH in cement paste with silica fume due to moisture diffusion considering self-desiccation are different from those in cement paste without silica fume.

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Keywords: Cement paste; Moisture diffusion; Humidity; Self-desiccation

1. Introduction

Moisture and relative humidity in concrete plays an important role in affecting its properties such as strength, shrinkage, creep and durability, etc. [1–3]. Moreover, moisture is one of the necessary conditions in which many kinds of deterioration processes of concrete occur [4], and the basis on which many deterioration mechanism and models are established [5]. Internal relative humidity (IRH) reduction of concrete structures in dry service environment will increase the risk of shrinkage and cracking. So, it is very important to predict IRH change and distribution in concrete structures for durability problems [6,7].

Moisture diffusion and self-desiccation are main factors that cause IRH reduction of cement paste in high-performance concrete. IRH change in normal concrete exposed to dry environment is mainly governed by moisture diffusion. However, IRH change in high performance concrete is affected not only by moisture diffusion but also by self-desiccation [8,9]. In general, self-desiccation means unhydrated cement in paste or concrete continues to hydrate accompanying the consumption of free water and consequently results in its IRH reduction. Self-desiccation has significant effects on IRH changes of paste in concrete with low w/c, especially at early ages [10,11]. It is of great significance to study IRH change and distribution in cement paste. How to distinguish IRH change in concrete resulting from moisture diffusion or self-desiccation is the key to predict and control IRH change of cement paste in high performance concrete.

This study is aimed at investigating IRH distribution in high performance cement paste in isothermal drying conditions and effect of moisture diffusion and self-desiccation on IRH distribution, respectively. Firstly, the IRH of uniaxial drying specimens of cement paste at 3, 11, 21 cm from drying surface in isothermal drying conditions, as well as IRH of sealed specimens of cement paste

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Table 1 Chemical components of cement and mineral admixtures

Raw materials	SiO ₂	Al ₂ O ₃	CaO	MgO	Fe ₂ O ₃	SO ₃	TiO ₂	K ₂ O	Na ₂ O	Specific area (m²/kg)	Density (g/cm ³)
Cement	21.4	5.6	63.3	1.9	2.7	3.4	0.4	0.7	0.2	351	3.10
SF	96.1	0.5	0.3	0.6	0.5	0.5	0	0.8	0.0	19600	1.91
GBFS	37.4	16.7	28.6	6.2	4.0	0.6	0.8	0.9	0.2	642	2.78

resulting from self-desiccation, was measured at different ages. And the effect of water to cement ratio (w/c) and mineral admixtures such as silica fume (SF), ground blastfurnace slag (GBFS) on IRH change of cement paste at different ages was studied. Then, IRH change laws of cement paste resulting from moisture diffusion were discussed and compared on the basis of the results measured in this paper and one presented in previous literatures.

2. Experiment

2.1. Raw materials and paste composition

In this experiment, 52.5 ordinary Portland cement according to Chinese standards with compressive strength of 63.7 MPa at 28 days was used. The mineral admixtures of SF and GBFS are also used. Table 1 gives chemical components and physical properties of cement, SF and GBFS. In order to make paste with expected fluidity, a naphthalene–formaldehyde condensate type high-range water-reducing admixture (HRWA) was also employed. Mix proportions of high performance cement pastes with different w/c and with mineral admixtures of SF and/or GBFS are given in Table 2.

2.2. Specimen preparation and IRH measurement method

IRH of all cement paste at different ages was measured using a YIDA NSWB150 probe and digital indicator made in China. The dimensions of specimens for uniaxial drying experiment were $10 \times 10 \times 30$ cm. The dimension of drying surface area of specimens was 10×10 cm with a length of 30 cm, shown in Fig. 1. Except the drying surface, the other five surfaces were sealed with the epoxy resin sealant with the thickness of 2 mm which would guarantee moisture diffusion conducts uniaxially during drying process. For

Table 2 Mix proportions of cement paste with different w/c and mineral admixtures

No.	w/c	HWRA (%)	Cementitious materials (%)			
			С	SF	GBFS	
H1	0.5	0	100	0	0	
H2	0.4	0	100	0	0	
Н3	0.3	0.5	100	0	0	
H4	0.2	1.0	100	0	0	
H5	0.3	0.5	90	10	0	
Н6	0.3	0.5	70	10	20	

uniaxial drying experiment, holes in paste specimens for RH probes were preformed at the distances of 3, 11, and 21 cm from the exposed surface when specimens of cement paste was molded. At the age of 1 day, moulds were removed from specimens. And initial IRH was measured. Then specimens were moved to a constant-temperature and constant-humidity container for uniaxial drying experiment. The drying environment in the container is controlled at a constant temperature of (20±2) $^{\circ}$ C and a constant relative humidity of $(50\pm2)\%$. The relative humidity is also controlled by saturated K₂CO₃ solution. At stated ages, RH probe was inserted and sealed in specimens with a rubber ring, and the read in indicator was recorded when relative humidity equilibrium was reached. IRH at the distance of 3, 11, and 21 cm from the exposed surface was measured.

The dimensions of the specimen for self-desiccation experiment were $10 \times 10 \times 10$ cm. All six surfaces of the specimens were sealed with the sealant. For self-desiccation experiment, the probe hole was located in the center of the specimens. The details of geometric size and probe location of specimens are shown in Fig. 2. The specific RH testing method, calibration and accuracy of measured RH are described in the literature [12].

3. Results and discussion

3.1. IRH distribution of cement paste in isothermal drying conditions

IRH distributions in cement paste at 3, 11, and 21 cm from the exposed surface in isothermal drying experiment are shown in Fig. 3a, b, and c, respectively, which are mainly caused by both uniaxial moisture diffusion and self-desiccation. From the results given in Fig. 3a, one can seen that, for a given w/c ratio, IRH of cement paste decreases rapidly at early ages and then decreases slowly at later ages. At the distance of 3 cm from the exposed surface, although

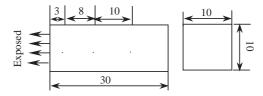


Fig. 1. Dimensions and RH probe locations of specimens for uniaxial isothermal drying experiment (unit: cm).

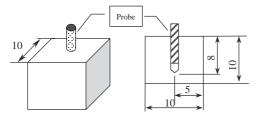


Fig. 2. Dimensions and RH probe location of specimens for self-desiccation experiment (Unit: cm).

it is generally believed that IRH of cement paste resulting from moisture diffusion increases rapidly with the increase of w/c, IRH decrement (herein, IRH decrement means the difference of IRH at a certain age and IRH at the age of initial testing) of cement paste with w/c no higher than 0.4 increases as w/c decreases at same ages, particularly at early ages. The effect of self-desiccation of cement paste with w/ c no higher than 0.4 is responsible for that (in Fig. 3). IRH

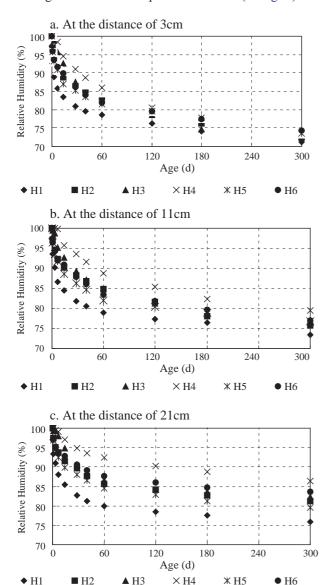


Fig. 3. IRH distribution of cement paste in isothermal drying conditions.

H6

of cement paste incorporated with SF and/or GBFS mineral admixtures decreases more rapidly than that of cement paste without mineral admixtures. As shown in Fig. 3, it indicates that IRH decrease in cement paste is dependent on moisture diffusion as well as self-desiccation, especially for high performance cement paste with w/c no higher than 0.4.

Given a certain w/c ratio, the trends of IRH distribution of cement paste at the distances of 3, 11, and 21 cm are similar(Fig. 3). At the same age, IRH value of cement paste is closely related to the distance from the exposed surface. Its IRH decrement at the distance of 3 cm is higher than those at the distances of 11 and 21 cm. IRH differences of cement paste with w/c higher than 0.4 between different distances are far higher than those of cement paste with w/c no higher than 0.4. It also indicates that IRH decrement of cement paste resulting from moisture diffusion increases with the increase of w/c.

3.2. IRH distribution of cement paste resulting from self-desiccation

IRH distribution of sealed cement paste resulting from self-desiccation is shown in Fig. 4. As seen from Fig. 4a, IRH of cement paste with w/c at 0.2 decreases to 79% at 300 days. However, IRH of cement paste with w/c at 0.5 only decreases down to 94%. It indicates that the IRH decrement of cement paste resulting from self-desiccation increases with the decrease of w/c, especially at early ages. The lower w/c is, the higher the self-desiccation effect on IRH decrement of cement paste. From Fig. 4b, it can be observed that IRH decrement of cement paste incorporated

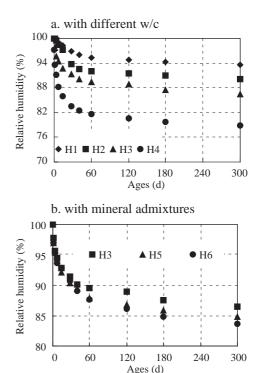


Fig. 4. IRH distribution of cement paste resulting from self-desiccation.

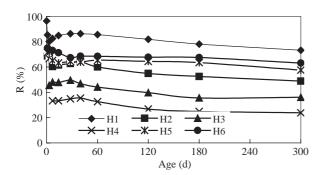


Fig. 5. R change curves with ages and different w/c ratios at the distance of 3 cm to the exposed surface.

with 10% SF and with 20% GBFS is higher than that of control paste at the same ages. This may be the reason that pozzolanic effect of SF and GBFS mineral admixtures accelerates cement hydration at early ages and so moisture in capillary pores of cement paste was consumed greatly. Consequently, IRH decrement of cement paste incorporated with SF and GBFS increases.

Proportion of IRH decrement of cement paste resulting from self-desiccation to total IRH decrement at a certain age is calculated as Eq. (1).

$$R = \frac{h_0 - h_s}{h_0 - h} \times 100 \tag{1}$$

Here, R is the percent ratio of IRH decrement of cement paste resulting from self-desiccation to total IRH decrement of cement paste in isothermal drying conditions at a certain age, h represents IRH of cement paste, h_s represents IRH of sealed cement paste resulting from self-desiccation and h_0 represents the initial IRH of cement paste in isothermal drying conditions.

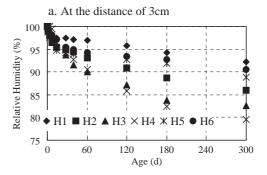
As the calculated results of cement paste at 3 cm distance to the exposed surface are shown in Fig. 5, obviously, in the one hand, the proportion R decreases gradually with the increase of age for a given w/c ratio. It indicates that selfdesiccation effect on IRH decreases gradually with age change. On the other hand, with the decrease of w/c, the proportion of IRH decrement in cement paste resulting from self-desiccation increases gradually, and the proportion of IRH decrement resulting from moisture diffusion decreases. It lies in the fact that, the lower w/c of cement paste is, the lower free water content of it has. Limited free moisture is consumed quickly when unhydrated cement in cement paste hydrates continuously at early ages. Consequently, IRH decrement of cement paste with low w/c resulting from selfdesiccation increases [11]. Of course, sizes of capillary pores in pastes with low w/c becomes smaller, which is responsible for it as well. Meanwhile, moisture diffusion in cement paste with low w/c becomes more and more difficult due to its compact microstructure and low porosity. It could be concluded that, for cement paste with w/c higher than 0.4, its IRH decrement is mainly affected by moisture diffusion, but for cement paste with w/c no higher than 0.4,

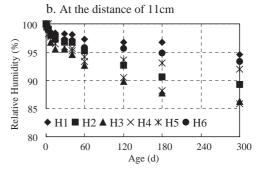
its IRH decrement is dependent not only on moisture diffusion but also on self-desiccation. However, since the point of w/c=0.4 is just selected in the experimental program and no more other adjacent points is investigated, the analysis is based on it. In fact, effect of moisture diffusion and self-desiccation on IRH decrement with w/c change is a gradual change process. There is no distinct boundary for which weighs high or low.

It also is concluded that R of cement paste at 11 and 21 cm distance to the exposed surface is higher than that at 3 cm distance at a given age for a given w/c since moisture diffusion of cement paste at long distance becomes more difficult.

3.3. IRH distribution of cement paste resulting from moisture diffusion

Fig. 6 shows calculated IRH distribution in cement paste with ages and at 3, 11, and 21 cm from the exposed surface resulting from moisture diffusion. These values of Fig. 6





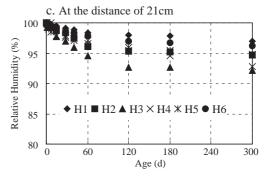


Fig. 6. Calculated IRH of cement paste resulting from moisture diffusion considering self-desiccation.

were calculated through adding IRH decrement of cement paste resulting from self-desiccation presented in Fig. 4 to the results presented in Fig. 3. The calculated Eq. (2) is described as follows:

$$h_{\rm m} = h + (h_0 - h_{\rm s}) = h + h_0 - h_{\rm s} \tag{2}$$

Here, $h_{\rm m}$ represents IRH of cement paste resulting from moisture diffusion considering self-desiccation.

As seen clearly from Fig. 6, w/c also plays a significant role in moisture diffusion and so does the distance to the exposed surface. IRH decrement of cement paste resulting from moisture diffusion decreases with the decrease of w/c. The rate of moisture diffusion in cement paste with w/c higher than 0.4 is much higher than that of cement paste with w/c no higher than 0.4, which results in rapid reduction of IRH in cement paste with w/c higher than 0.4. As known before (13), the nearer the distance from exposed surface is, the faster the IRH reduction in cement paste. Given the same w/c, IRH decrement of cement paste incorporated with 10% SF and 20% GBFS resulting from moisture diffusion at different distances is the smallest of all pastes. It mainly lies in that mineral admixtures make the microstructure of cement paste denser, and porosity in pastes changes as well; thus its diffusion coefficient becomes smaller. These results are in accordance with results presented in other literature [2,8]. It follows that, when coefficient of moisture diffusion in cement paste with w/c no higher than 0.4 is needed to calculate and simulate, effect of self-desiccation should be considered.

3.4. Analysis of Parrott IRH change laws of cement paste in the isothermal drying conditions

A formula for the prediction of moisture profiles throughout drying concrete was proposed by Parrott [13]:

RH = RHA + (100 - RHA) ×
$$f(t)$$

 $f(t) = 1/(1 + t/b)$
 $b = d^{X}(Y - e)(w - Z)/W$
(3)

Where RH is the predicted relative humidity (%), RHA is the ambient relative humidity(%), t is the drying age (days), d is the depth from drying surface (mm), e is the amount of OPC replaced by SF or GBFS (%), and w is the water/cementitious materials ratio. W, X, Y and Z are constants. Parrott then assigned values to the four parameters, such that W=8, X=1.35, Y=70 and Z=0.19, and D.B. McDonald and H. Roper suggested as W=11.4, X=0.85, Y=128.00 and Z=0.03 [14].

Eq. (3) was used to predict IRH of cement paste in the isothermal drying conditions in this program. The calculated results indicate that the predicted RH using Eq. (3) either with the parameters' value assigned by Parrott or D.B. McDonald and H. Roper is different from the measured RH presented in Fig. 3. It could be found that Eq. (3) is not suitable for predicting RH of cement paste with low w/c and mineral admixtures at different distances resulting from

moisture diffusion. The difference between predicted RH and measured RH mainly comes from several aspects as follows. As stated in previous literatures, Eq. (3) is just suitable for concrete or paste with high w/c that has higher porosity and more free water than paste with low w/c and consequently the effect of self-desiccation on its IRH change is little. Therefore, it easily concluded that IRH predicted by Eq. (3) would decrease as w/c decreases, which means IRH reduction of cement paste causing by moisture diffusion would increase with the increase of w/c. However, IRH reduction of cement paste with w/c no higher than 0.4 increases with the decrease of w/c as seen in Fig. 3. The reasons may lie in that IRH decrement of paste with w/c no higher than 0.4 resulting from self-desiccation predominates in IRH change of paste and effect of moisture diffusion on IRH change becomes minor, especially at long distance from the exposed surface. It indicates that when IRH change laws of paste with w/c no higher than 0.4 resulting from moisture diffusion is predicted or calculated, self-desiccation shall be taken into consideration and new IRH laws of cement paste with w/c lower than 0.4 shall be generalized and established.

4. Conclusions

Based on the results obtained from the present experimental investigation, the following conclusions can be obtained:

- 1. Effect of moisture diffusion and self-desiccation on IRH decrement with w/c change is a gradual change process. There is no distinct boundary for which weighs high. However, for the selected points of w/c parameter in the program, for cement paste with w/c higher than 0.4, its IRH change mainly affected by moisture diffusion, but for cement paste with w/c no higher than 0.4, its IRH change is controlled by both moisture diffusion and self-desiccation in isothermal drying condition.
- 2. With the decrease of w/c, IRH decrement of cement paste resulting from self-desiccation increases, and IRH decrement resulting from moisture diffusion decreases at a given age. The addition of SF and GBFS into paste makes IRH decrement resulting from self-desiccation increase and IRH decrement resulting from moisture diffusion decrease at different ages.
- 3. w/c and the distance to the exposed surface play a significant role in IRH change resulting from moisture diffusion in isothermal drying condition. IRH decrement of cement paste resulting from moisture diffusion decreases with the decrease of w/c, and the nearer the distance from exposed surface, the faster the IRH reduction in cement paste.
- 4. Parrott change law of IRH in cement paste with w/c no higher than 0.4 and mineral admixtures due to moisture diffusion considering self-desiccation are different from

those in cement paste with higher w/c ratios, and new IRH laws of cement paste with w/c no higher than 0.4 shall be established.

Acknowledgments

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