- Technical Paper -

AN EXPERIMENTAL STUDY OF SHRINKAGE BEHAVIOR OF BULK HYDROPHOBIC MORTAR

Yao LUAN*1, Shingo ASAMOTO*2 and Taiju YONEDA*3

ABSTRACT

Bulk hydrophobic mortar, made by adding hydrophobic chemicals in mix proportion, has been proven with good water penetration resistance even if crack exists. In this study, autogenous shrinkage and drying shrinkage of bulk hydrophobic mortar with different w/c ratios were investigated experimentally. Other properties, such as setting time, water loss, strength and Young's modulus were also measured. It is found that hydrophobic mortars were likely to have lower autogenous shrinkage and drying shrinkage. The lower shrinkage was discussed from a viewpoint of capillary tension.

Keywords: hydrophobic mortar, autogenous shrinkage, drying shrinkage, contact angle, setting time

1. INTRODUCTION

Durability problems of concrete structures, such as alkali-silica reaction, freeze-thaw damage, chloride attack and corrosion of steel in concrete, are causing attention all over the world. Those deterioration phenomena are closely related to water ingressions into concrete, so water tightness of concrete, which indicates the resistance to water penetration, is very important from the viewpoint of durability. Usually, lower water-to-cement (w/c) ratio or surface coating can improve the water tightness, but both of them have flaws. Lower w/c ratio increases the amount of cement so produces more hydration heat, causing thermal cracks. Concrete coated at the surface, despite the water repellent layer, may lose its water tightness significantly if crack occurs, because crack becomes a convenient channel for water and ion penetration. As a result, deterioration may be accelerated greatly.

An idea way, from maintenance viewpoint, is that cracked concrete could keep its water tightness even without repairs. Towards this direction, recently a concept of hydrophobic concrete or mortar is attracting the attentions of researchers. Manufactured by mixing hydrophobic agents as admixtures, not only the surface but also the bulk concrete tends to be water repellent. Compared with surface coating, hydrophobic concrete saves the labor and time of treatment. More importantly, it shows a potential that, even if the crack occurs, the overall water tightness of concrete can be preserved, because the crack surface is also water repellent (Fig. 1). It is advantageous in both reducing maintenance cost and increasing the durability of concrete, having a broad perspective of practical use.

Past researches have indicated the excellent water tightness of hydrophobic concrete and mortar [1-3]. In our group, such mortars were made, and we call it bulk hydrophobic mortar. Our previous experiment showed that even if crack occurred, the bulk hydrophobic mortar still had relatively good water tightness due to the internal water repellency [4]. Therefore, it is promising to be used in practice to improve the durability of structure. However, besides the water tightness, other properties of this mortar, particularly the shrinkage behavior, are still unknown. Despite the bulk hydrophobicity, shrinkage induced cracking is still a concern to other properties, such as load capacity or carbonation. In this study, the objective is to investigate autogenous shrinkage and drying shrinkage of the bulk hydrophobic mortar experimentally. Other properties related to shrinkage, such as water loss, strength and Young's modulus were also measured and discussed.

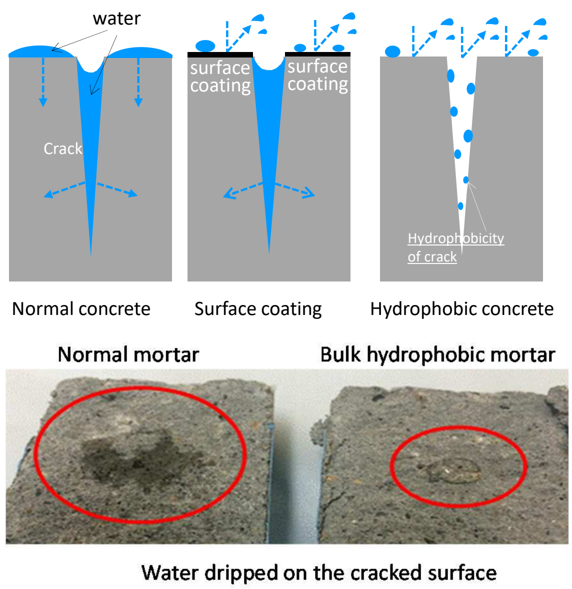


Fig. 1 Advantage of hydrophobic concrete: water repellency under cracked condition

*1 Assistant Prof., Dept. of Civil and Environmental Engineering, Saitama University, Dr. E., JCI Member
*2 Associate Prof., Dept. of Civil and Environmental Engineering, Saitama University, Dr. E., JCI Member
*3 Engineering Researcher, Technical Institute, Maeda Corporation, Dr. E., JCI Member

2. EXPERIMENT PROGRAMS

2.1 Materials, mix proportion and mixing method

Two types of hydrophobic agent (HPA), named A and K, were added, respectively. They are both in the form of liquid with low viscosity. A is a commercial hydrophobic solution with silicon based chemical, and K is alkyl-modified silicone polymer. They have been used in the bulk hydrophobic mortar in the authors' previous research [4]. Using ordinary Portland cement and sand, mortar specimens with w/c 0.35 and 0.50 were prepared. For the groups of w/c 0.35, superplasticizer was added to increase workability. Experiment groups are shown in Table 1. Ref. represents the reference mortar without hydrophobic agent. Following previous methods, two types of agent mixing were conducted. One was to directly mix the hydrophobic agent A or K with water, cement and sand (abbreviated as directly mixing). The other was abbreviated as sand spraying, which was to spray the hydrophobic agent K onto the surface of sand, and after standing for 24 hours the sand was mixed with water and cement. Those groups are 0.35-K-S and 0.5-K-S. Sand fraction of the mortar was 0.50.

Table 1 Experiment groups of shrinkage

Group Name	w/c	HPA addition ratio	Group Name	w/c	HPA addition ratio
0.35-Ref	0.35	0	0.5-Ref	0.5	0
0.35-A-1		1% A*	0.5-A-1		1% A*
0.35-K-1		1% K*	0.5-K-1		1% K*
0.35-K-3		3% K*	0.5-K-3		3% K*
0.35-K-S		10ml K/ 200g Sand**	0.5-K-S		10ml K/ 200g Sand**

* Directly mixing method (by weight of cement);
**Sand spraying method

2.2 Initial and final setting times

The times of initial and final setting were measured experimentally, according to the JIS A 1147. Corresponding to each group of autogenous shrinkage, the same batch of fresh mortar was used. An iron cylinder bucket with both the diameter and height 150 mm was selected as container. A loading apparatus capable of measuring the penetration force with an accuracy of 10 N and a capacity of 1000 N was used (Fig. 2). The loading apparatus had a set of penetration needles with the diameter of 11.4, 8.2, 5.9 and 4.1 mm.

The mortar was filled into the bucket to a depth of 140 mm, and sealed with plastic sheet. Then, the bucket was kept into environment room (21±1°). Pushing the handle, a needle of diameter 11.4 mm was inserted into the mortar at two hours after the initial time. Then, the needle was inserted every 30 minutes until the penetration force appeared. The time and the penetration force was recorded. Then, the insertion was carried out every 30 minutes to 1 hour with appropriate needle, recording the time and force. This operation was repeated until the penetration force reached 400 N with the needle of diameter 4.1 mm. Using the penetration force and radius of the needles, the pressure was calculated.

Fig. 2 Loading apparatus for setting time

2.3 Autogenous shrinkage and drying shrinkage

Prismatic specimens with the size of 40 × 40 × 160 mm were made. There were three specimens in each group. Shown in Fig. 3, a type of embedded strain gauge was adopted. The strain gauge were fixed in the center of the mold with strings before mixing. Deformation was automatically recoded through with data logger. In order to eliminate frictional resistance, 1 mm thick Teflon sheets was attached on the bottom and other surfaces of mold. To avoid water and vapor leakage soon after casting, waterproof cream was smeared in each gap or void. The measurement was started immediately after casting. After 24 hours, all of the specimens were demolded carefully without disturbing the measurement, and wrapped with an inner layer of plastic and an outer layer of aluminum tape tightly. The specimens were kept sealing until the age of 7 days to prevent from water evaporation. During the whole process, specimens were kept in an environment room with ambient temperature 21±1°, and autogenous shrinkage was measured continuously. Weight of the specimens were checked to confirm no water exchange with ambience.

Fig. 3 Embedded strain gauge in mold

At the age of 7 days, sealing was removed, and all of the specimens were kept under the condition of temperature 21±1°C and relative humidity 60%. Drying shrinkage measurement with the gauges was carried out for two weeks. In addition, as a comparison, we also measured the drying shrinkage of some groups using another method. Contact tips were attached at the surface of the specimen before drying. Their distance was measured using a portable length meter. Weight of the specimen was periodically measured.

2.4 Strength and Young's modulus

Cylindrical specimens of diameter 50 mm and height 100 mm were prepared for compressive strength and Young's modulus. Those specimens were kept sealed under 21±1°C, and the measurement was carried out at the ages of 7 days and 28 days, respectively. A compressometer, measuring the axial strain on the specimen placed inside, was used for Young's modulus.

2.5 Contact angle

The hydrophobicity of mortar was evaluated by measuring contact angle. After polishing the mortar surface, a droplet of water was dripped. Using the digital microscope, the tangent line of the droplet at the liquid-solid surface was drawn, and the contact angle was measured (Fig. 4). Generally the higher contact angle indicates higher water repellency.

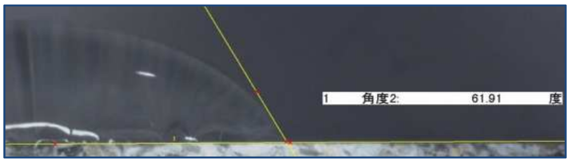


Fig. 4 Measurement of the contact angle

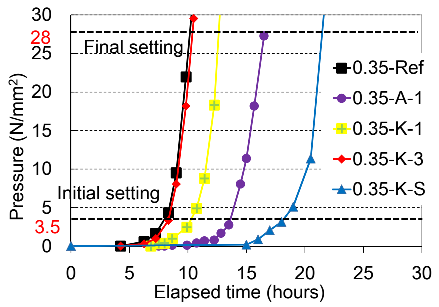
3. RESULTS AND DISCUSSIONS

3.1 Times of initial and final setting

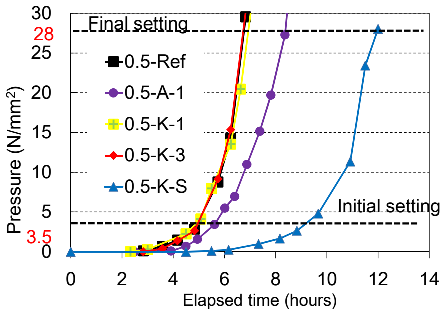
The results of setting time, which is the relationship between the penetration pressure and elapsed time, are shown in Fig. 5. The initial setting time is defined as that when the pressure arrives at 3.5 N/mm^2, whereas the final setting time is that when arriving at 28 N/mm^2. For w/c 0.35, it can be found that the reference one had an initial setting time of around 8 hours, and a final setting time of 10 hours. The case of 0.35-K-3 shows approximately the same setting times. For other cases of direct mixing, the times of initial and final setting were postponed by several hours. The case of sand spraying, 0.35-K-S, shows the largest retardation in the setting. The initial setting time is 17 hours, and the final setting time is 22 hours. For w/c 0.35, the setting time of each group is longer than that of the corresponding case with w/c 0.50. This may be due to their addition of superplasticizer.

As to w/c 0.5, the reference one took 5 hours to the initial setting, and more than 6 hours to the final setting. The cases of 0.5-K-3 and 0.5-K-1 show similar setting times as the reference one. For the case of A-1, the setting times were postponed slightly, with the initial time less than 6 hours and the final time 8 hours. The sand spraying one shows the most postponed setting, which are more than 9 hours for initial setting and right 12 hours for the final setting.

Combining the results of w/c 0.35 and 0.5, it can be found that the influence of hydrophobic agent K on the setting times, when directly mixing it, is not significant. This implies a relatively good compatibility with cement at the early stage. The hydration may be retarded only slightly at this stage. With regarded to the agent A, it retarded the hydration a little bit more. An interesting point lies in the sand spraying cases, in which the setting times were much more postponed. It is difficult to attribute it to the retardation of cement hydration, because the agent was sprayed only on the sand and dried before mixing. A reasonable explanation may be lies on the interface between cement paste and sand. Due to relatively hydrophobic surface, the precipitation of hydration product in adjacent area of sand particles becomes difficult, so the development of bonding is retarded. Even if cement hydration and the hardening of pure paste is assumed to keep the same pace as the reference one, the hydrophobic mortar, as an assembly of sand and paste, tends to be much softer, showing postponed setting. In previous study, the authors have discussed this influence on the interfacial transition zone (ITZ) and its attribution to the lower strength of the hydrophobic mortar [4]. From the autogenous shrinkage test, it reveals that the effect of weaker ITZ at very early age lies also in the setting process of the hydrophobic mortar.



(a) w/c = 0.35



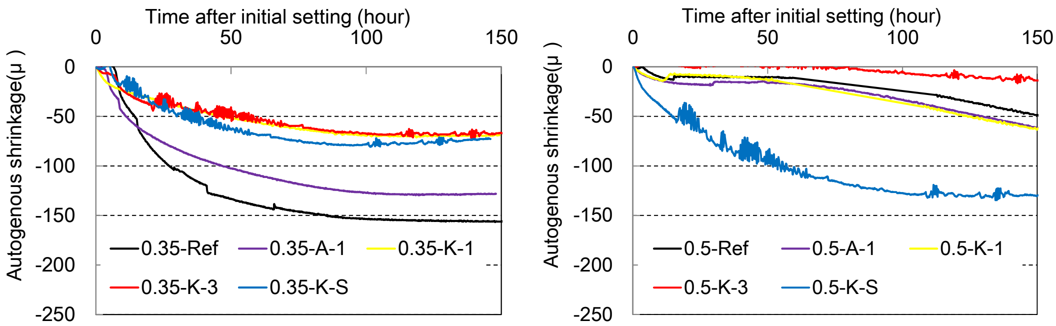
(b) w/c = 0.5

Fig. 5 Setting time

3.2 Autogenous shrinkage

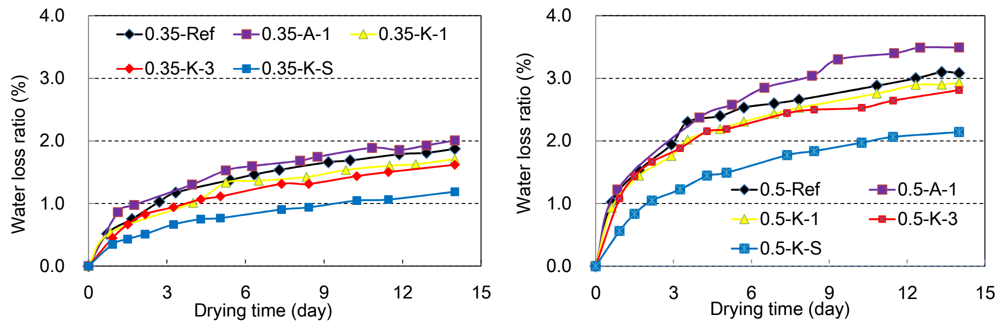
Initial setting is regarded approximately as the turning point of concrete from liquid to skeleton formation, and pore structure starts to form. From initial setting time, concrete starts to harden, and strength and stiffness are developed. Hence, only deformation after initial setting is related to crack occurrence. JCI technical committee defines the autogenous shrinkage as the volume reduction after initial setting [5]. Accordingly, in this study, the initial setting time was set as zero point, and the shrinkage value before initial setting time is deducted. The results of autogenous shrinkage from initial setting time are shown in Fig. 6.

It can be seen that the autogenous shrinkage in the cases of w/c 0.35 is much larger than the corresponding one of w/c 0.5. The only exception is 0.5-K-S. It shows an abnormally large shrinkage. It was found that 0.5-K-S is easy to bleed after standing for a while, so the abnormal shrinkage may be related to that. The reason needs to be inspected and verified again in the future. Anyway, the results reveal that, for low w/c, hydrophobic mortar tends to have smaller autogenous shrinkage, particularly when K is mixed or sprayed.



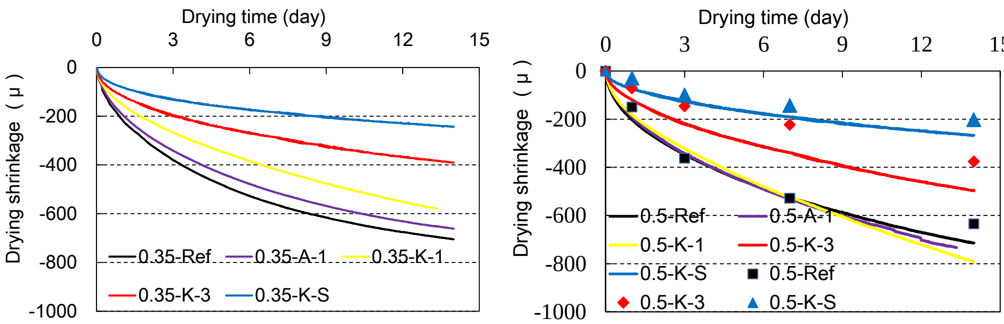
(a) w/c = 0.35 (b) w/c = 0.5

Fig. 6 Autogenous shrinkage



(a) w/c = 0.35 (b) w/c = 0.5

Fig. 7 Water loss during drying



(a) w/c = 0.35 (b) w/c = 0.5

Fig. 8 Drying shrinkage

3.3 Water loss and drying shrinkage

The results of water loss are shown in Fig. 7. The cases with w/c 0.35 generally show smaller water loss than the corresponding one with w/c 0.5. Moreover, for both the two w/c ratios, the hydrophobic mortars directly mixed with agent K, 1% and 3%, show slightly lower water loss than the reference one. In contrast, the case of 0.5-A-1 shows slightly larger value. The reduction in water loss is most significant in the sand spraying case than in the directly mixing case. For both w/c ratios 0.35 and 0.5, their water loss ratio are approximately 50% of the value of the reference one.

The results of drying shrinkage are shown in Fig. 8. For w/c 0.5, some groups were also measured with contact chips. Those data are expressed by dots in the figure as a comparison. They appears in good consistence with those measured by embedded strain gauge. For the reference ones, the shrinkage values are 600-800 micros after 14-day drying. Most of the shrinkage in the cases of the hydrophobic mortar are smaller than the reference ones. The exceptions are only 0.5-A-1 and 0.5-K-1. They are almost as same as the reference one. Significantly smaller values in drying shrinkage are found in the cases of K-3 and K-S. Most significantly, for sand spraying cases with K (i.e. K-S), the drying shrinkage is only one third of that of the normal ones, for both w/c 0.35 and 0.5.

3.4 Contact angle

The results of contact angle are shown in Fig. 9. The contact angles of the reference mortar are 7˚ and 11˚, respectively for w/c 0.35 and 0.5. For A-1%, the contact angles were 10˚ and 14˚, indicating little hydrophobic result. In the past experiment, it has been found that A is not so effective as K, especially when less addition ratio such as 1% is used. The contact angles of groups of K are much larger, which are 41˚ and 58˚ for K-1%, 56˚ and 91˚ for K-3%, and 80˚ and 99˚ for sand spraying cases, respectively for w/c 0.35 and 0.5. Water tended to be spherical shape at the surface of mortar, in the cases of high contact angle. Those results are consistent with the past experiment.

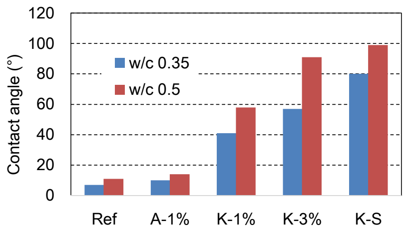


Fig. 9 Contact angle

3.5 Strength and Young's modulus

The results of compressive strength and Young' modulus of w/c 0.5 are shown in Fig. 10 and Fig. 11. It can be found that, compared with the normal one which has a respective strength of 48.4 MPa at 7 days and 65.0 MPa at 28 days, all of the hydrophobic mortar showed lower strength. For sand spraying cases, they had lowest strength, only about half in the value. For agent K, strength decreased with the addition ratio increasing. For Young's modulus, the tendency of decreasing can also be observed, but the extent is quite little compared with strength.

The tendency of decreasing in strength agrees with the previous experiment, and the reasons have been explained [4]. One of the reason is attributed to the increased air content, due to addition of hydrophobic agent. In addition, for the case of sand spraying, the ITZ between cement paste and sand is further weakened. The third reason is that hydrophobic agent may inhibit hydration process of cement even the extent is not large.

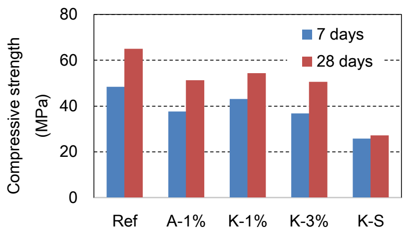


Fig. 10 Compressive strength

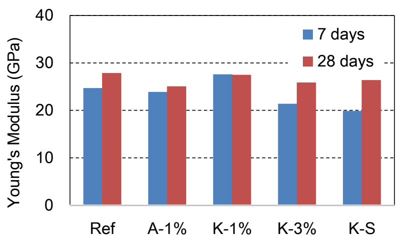


Fig. 11 Young's modulus

3.6 Discussions

Based on the above experiment results, it can be concluded that, regardless of few exceptional cases which needed to be inspected again, in general, the hydrophobic mortar made with agent K tends to show smaller values in both autogenous shrinkage and drying shrinkage, as well as water loss, if compared with the reference one. This tendency is most evident in the case of sand spraying. The agent A, in this experiment, showed little effect on the hydrophobicity, so shrinkage is roughly at similar level as that of the reference one.

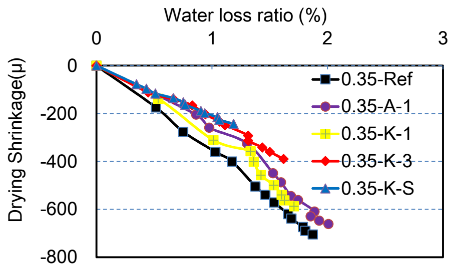
Shrinkage behavior is related to both the driving force due to water loss or self-desiccation, and the resistance due to mortar stiffness. From the result of Young's modulus, the resistance of hydrophobic mortar is only slightly lower than the reference one. Hence, the main discussion should focus on the driving force.

Firstly, referring to the results of water loss, it can be deduced that the lower drying shrinkage may be partially from the less water loss. Especially, for the case of sand spraying, water loss ratio is only a half of that of the reference one. Less drying implies relatively smaller drying shrinkage, if assuming the shrinkage mechanism consistent. There seems little experimental data about the water loss of bulk hydrophobic mortar in past researches, but for concrete or mortar coated with silane-based hydrophobic agent at the surface, some researches did report that water loss was reduced compared with normal one [6-7]. In such a case of surface coating by silane-based agent, the agent was usually impregnated several millimeters into the surface. Therefore, this question goes to the water loss, i.e. why does hydrophobic mortar have lower water loss? The authors attempted to explain it from microscale viewpoint tentatively. It can be imagined that bulks of water existing in the pore or ITZ become less continuous or more discrete, because of the water repellency on the gel or sand surfaces. Water tends to contact or curl into small and round droplet, and those droplets tend to be separated each other. This separation or discontinuity, may result in the difficulty of liquid water in transferring under drying caused force. Furthermore, the curled water droplets have smaller surface area than a spreading water layer, becoming also difficult to evaporate. Experimental study shows that droplet on a surface had a lower evaporation rate if contact angle increased [8]. From either the viewpoint of vapor or liquid, it can be inferred that water transfer coefficient decreases, resulting in a lower water loss ratio. In addition, the time of measurement in this study was still short. Water loss in a longer time scale needs to be investigated in the future.

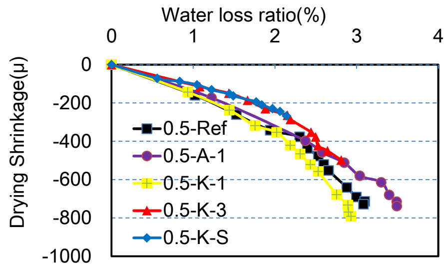
Fig. 12 shows the relationship between water loss and drying shrinkage. With the same water loss, all of the hydrophobic mortar show lower drying shrinkage, with only an exception 0.5-K-1. This can be tentatively explained based on the theory of capillary tension. Fig. 13 shows a tube with a radius r. Due to surface tension, a capillary meniscus is formed and pressure difference is generated across the vapor-liquid interface. According to the equilibrium, the following equation exists among the negative pressure P_l in water, contact angle θ and surface tension of water-air interface γ

$$P_l = \frac{2\gamma\cos\theta}{r} \tag{1}$$

Shrinkage force caused by capillary tension can be regarded to be proportional to P_l. According to the equation, the shrinkage force decreases with increasing the contact angle. This can explain the tendency that hydrophobic mortar have smaller drying shrinkage when under the same water loss. This can also explain the reduced autogenous shrinkage in Fig. 6 (a), if all of



(a) w/c = 0.35



(b) w/c = 0.5

Fig. 12 Relationship between water loss and drying shrinkage

the mortars have similar hydration degree at early stage. Additionally, for the directly mixing cases, hydrophobic agent retards the hydration process in some extent. This may be another reason for the decreased autogenous shrinkage.

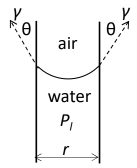


Fig. 13 Capillary tension in a tube

Interestingly, concrete with shrinkage reducing agent (SRA) usually shows higher water loss than normal concrete [9]. It seems different from the case of hydrophobic concrete. The reason for higher water loss is often attributed to lower surface tension due to shrinkage reducing agent, leading to a decreased Kelvin' radius in pores during drying. It implies that concrete is less capable of retaining water. On the other hand, lower surface tension leads to lower negative pressure. Comprehensively, the shrinkage driving force deceases. At present, there is no unified mechanism for shrinkage. The role of SRA, except the above mention, is still under discussion. Further study needs to be carried out.

4. CONCLUSIONS

(1) The times of initial setting and final setting were largely postponed, when mortar was made by sand spraying method with agent K. This may be due to the retardation of bonding formation between sand and cement paste at the early stage.

(2) The autogenous shrinkage of hydrophobic mortar is usually smaller than that of the normal mortar, in the case of low w/c ratio.

(3) For the hydrophobic mortar, water loss and drying shrinkage tended to be smaller than those of the normal mortar.

(4) The shrinkage mechanism was discussed. The authors hypothesize that water repellency in the pore or ITZ decrease water and vapor transfer coefficient, which may be the reason of less water loss for hydrophobic mortar. This may be also a reason for the decreasing drying shrinkage partially. In addition, the increased contact angle may cause the decreasing of the capillary tension force, which may be another reason for the decreased shrinkage. The above hypothesis needs to be validated in the future by experiment.

ACKNOWLEDGEMENT

This work was supported by JSPS KAKENHI Grant Number 16K18130. The authors would like to express the gratitude to their support.

REFERENCES

[1] Moriconi, G. and Tittarelli, F., "Use of Hydrophobic Agents as Concrete Admixtures," ACI Special Publication, Vol. 212, 2003, pp.279-288.

[2] Kubo, Y., Tamai, Y., Kurihara, S. and Miyagawa, T., "Durability of Hydrophobic Effect of Concrete Impregnated by Silane," Proceeding of JCI Annual Conference, Vol. 23, No.1, 2001, pp.421-426. (In Japanese)

[3] Ferrara, L. and Pattarini, A., "Siloxanes in Concrete: From Manual Application of Water-proofing Treatment to Mix-Design Addition for Concrete Hydrophobicity," Proceedings of 8th International Conference on Concrete under Severe Conditions – Environment and Loading CONSEC2016, Sep. 12-14, 2016, Lecco, Italy, pp. 263-268.

[4] Furuta, H., Luan, Y., Asamoto, S. and Yoneda Y., "A Study of Properties of Mortar Mixed with Hydrophobic Agents: Water Absorption, Strength and Drying Shrinkage", Proceeding of JCI Annual Conference, Vol. 38, No.1, 2016, pp. 693-698. (In Japanese)

[5] Committee Report, Technical Committee on Autogenous Shrinkage of Concrete, Japan Concrete Institute, 1998.

[6] Tanaka, H. and Miyagawa, T., "A Study of Concrete Applied with Urea, Limestone Aggregate and High Concentration Silane-based Surface Hydrophobic Agent", Proceeding of JCI Annual Conference, Vo. 34, No.1, 2012, pp. 568-573. (In Japanese)

[7] Suzuki, Y., Kobayashi, K., and Masuda, T., "Fundamental Study of Surface Hydrophobic Agent for High Durability Structure", JR East Technical review, No. 27, pp. 36-41, 2009. (In Japanese)

[8] Chandra, S., Marzo, M., Qiao, Y. M., and Tartarini, P., "Effect of Liquid-Solid Contact Angle on Droplet Evaporation," Fire Safety Journal, 1997, pp. 141-158.

[9] Kojima, T., "Handbook of Concrete Admixture", the Society of Materials Science, Japan, 2004. (In Japanese)