Arid and tropical climates are prone to the occurrence of drying shrinkage cracking at early ages. Shrinkage reducing agents (SRAs) are often used to mitigate drying shrinkage, whose key action on the reduction of early-age shrinkage was seen to be a reduction of capillary tension. However, the roles of SRAs on reduction of shrinkage that have been reported, throw doubt on the idea that SRAs work solely via capillary tension. In order to clarify the mechanisms of early-age shrinkage in the presence of SRAs, the effects of fatty alcohol-based SRAs on shrinkage strain, capillary pore pressure and crack area of cement-based mortars at early ages were investigated. Our results demonstrated clearly that capillary shrinkage in the presence of SRAs, the effects of fatty alcohol-based SRAs on shrinkage strain, capillary pore pressure and crack area of cement-based mortars at early ages were investigated. Our results demonstrated clearly that capillary tension does not directly correlate with shrinkage strain at early ages in the presence of SRAs, even under the same profiles of evaporation and hydration. We interpreted the actions of SRAs on early-age shrinkage as follows: SRA micelles adsorb onto hydrates and/or exist in pores, and form the hydrate matrix with incorporated SRA micelles. The matrix could mitigate the capillary tension as stress absorbers, and thereby shrinkage and crack area are reduced. The concept of the matrix could be experimentally demonstrated by the changes in shear creep.

1. Introduction

Shrinkage begins due to rapid hydration and evaporation as soon as water is mixed in cement-based mortars and concretes. Shrinkage can be distinguished as early-age and long-term behavior, where “early-age” is limited to the 24 h after mixing and “long-term” refers to the time beyond. Arid and tropical climates, e.g., climates in South Asia and Southeast Asia, are more prone to the occurrence of drying shrinkage cracking at early ages. Furthermore, the use of fine inorganic admixtures such as limestone powder, which is commonly utilized in South Asia and Southeast Asia, could increase the risk of early-age shrinkage cracking due to drying (Uno 1998; Mayuran et al. 2017). Cracks provoked by the early-age shrinkage may exceed a few mm in width and may run through the whole depth of structures (Slowik et al. 2009; Slowik and Ju 2011), and lead to durability concerns in concrete structures. Although standard specifications for hot weather concreting are applied to avoid the early-age shrinkage cracking due to drying, such as the ACI guideline 305 R-10, experimental investigations suggest that the standard specifications are inadequate to avoid the early-age shrinkage cracking because local raw materials and climate conditions differ in arid and tropical countries (Almusallam et al. 1998).

The action of capillary pressure in fresh mortars and concretes is assumed to be the main cause of plastic shrinkage, which occurs due to water loss in hardening mortars and concretes (Powers and Brownyard 1947; Wittmann 1976). Shrinkage reducing agents (SRAs) are often utilized to mitigate drying shrinkage. Reduction of capillary tension was seen to be a key action of SRAs in reduction of shrinkage (e.g., Tomita 1991; Ai and Jung 1997; Bentz et al. 2001). Although drying shrinkage can be reduced up to about 60% by the dose of SRAs at ambient temperatures (e.g., Saliba et al. 2011; Holt and Leivo 2004), our previous experiments show that some SRAs, which can reduce drying shrinkage enough at ambient temperatures, are not effective under high temperature conditions where water evaporates quickly (Takahashi et al. 2018). Recently, the roles of SRAs on reduction of shrinkage for hardened cement pastes have been reported, which throw doubt on the idea that SRAs work solely via a capillary tension mechanism (e.g., Maruyama et al. 2016, 2018; Gartner et al. 2017; Weiss et al. 2008; Sant et al. 2011; Fukushima et al. 2010).

Maruyama et al. (2018) reported that SRAs prevent the agglomeration of calcium silicate hydrate (C-S-H) due to drying and/or by changing the amount of outer C-S-H and therefore drying shrinkage is reduced. On the basis of this report, a dominant influencing factor on early-age shrinkage could be capillary tension because of the small amount of precipitated C-S-H and less water evaporation from C-S-H within 24 h after casting. However, the authors have reported that some types of...
fatty alcohol-based SRAs, whose surface tension was 30 to 35 mN/m, decrease capillary pore pressure but reduce the early-age shrinkage of mortars slightly (Takahashi et al. 2016). Thus, mechanisms of reduction of shrinkage in the presence of SRAs, especially at early ages, have not been fully understood.

This study comprehensively investigates the dimensional stability, capillary pore pressure, cracking behavior, hydration kinetics, evaporation rate, pore size distribution by mercury intrusion porosimetry (MIP), weight loss by thermal gravimetry (TG), shear modulus and shear creep by rheometer under high temperature conditions in early age cement-based pastes and mortars with different types and dosages of the fatty alcohol-based SRAs, and explains the corresponding mechanisms.

2. Experimental protocol

2.1 Materials and mix proportions
The mortar mixes used in the experiments contain ordinary Portland cement (OPC), siliceous sand with a maximum grain size of 0.8 mm, polycarboxylic ether superplasticizer (SP), an acrylic viscosity-modifying agent (VMA), SRAs and de-foamer (DF) in the proportions shown in Table 1. Mix proportions of the experimental pastes are shown in Table 2. Organic-based retarder was added to control setting time of pastes at the same level with the mortars. The mortar mixes were used for the tests of flow values, air contents, hydration kinetics, dimensional stability, capillary pore pressure, weight change and cracking areas. The paste mixes were used for the tests of flow values, air contents, setting time, pore size distribution by MIP, weight loss by TG, shear modulus and shear creep by rheometer. The dry mortar mixes and paste mixes were pre-mixed prior to testing and subsequently mixed with water for 2 min at 700 rpm using a mechanical mixer. SRAs are fatty alcohol-based agents to which different ratios of ethylene oxide/propylene oxide are added. Cloud point and hydrophilic-lipophilic balance (HLB) of the SRAs in 1% solution and surface tension of the SRAs in 1% and 3% solution are listed in Table 3. Less changes in surface tension between 1% and 3% occurred as the concentrations were higher than the critical micelle concentration of the SRAs.

Table 1 Proportions of the experimental mortars (unit; g).

| Mortars | OPC  | Sand  | SP   | VMA  | SRA-1 | SRA-2 | DF   | Water |
|---------|------|-------|------|------|-------|-------|------|-------|
| A       | 430  | 570   | 1.00 | 1.00 | 0.00  | 0.32  | 180  |
| B       | 430  | 570   | 1.00 | 1.00 | 4.30  | 0.00  | 0.97 | 180   |
| C       | 430  | 570   | 1.00 | 1.00 | 0.00  | 4.30  | 0.32 | 180   |
| D       | 430  | 570   | 1.00 | 1.00 | 12.90 | 0.00  | 0.32 | 180   |

Table 2 Proportions of the experimental pastes (unit; g).

| Pastes | OPC  | SP   | VMA  | Retarder | SRA-1 | SRA-2 | DF   | Water |
|--------|------|------|------|----------|-------|-------|------|-------|
| A      | 430  | 1.00 | 1.00 | 2.00     | 0.00  | 0.00  | 0.32 | 180   |
| B      | 430  | 1.00 | 1.00 | 2.00     | 4.30  | 0.00  | 0.97 | 180   |
| C      | 430  | 1.00 | 1.00 | 2.00     | 0.00  | 4.30  | 0.32 | 180   |
| D      | 430  | 1.00 | 1.00 | 2.00     | 12.90 | 0.00  | 0.32 | 180   |

Table 3 Characteristics of the SRAs.

| SRA-1 | Concentration (wt,%aq) | 1% | 3% |
|-------|-----------------------|----|----|
|       | Concentration (wt,%aq) | 1.0 | 10.0 | 0.1 | 1.0 | 10.0 |
| Solvent | Water | Water with 5 wt.% of OPC |
| Micelle size (nm) | 10.5 | 12.6 | 11.5 | 11.3 | 11.0 |
| SRA-2 | Concentration (wt,%aq) | 1.0 | 1.0 |
|       | Concentration (wt,%aq) | 1.0 | 1.0 |
| Solvent | Water | Water with 5 wt.% of OPC |
| Micelle size (nm) | 730 | 832 | 702 | 893 | 762 |

Table 4 Micelle size of the SRAs with different solvents and concentration at 40°C.

2.2 Testing methods
Tropical concreting is normally conducted at the average temperature of 30°C, and occasionally at about 35°C (Tim 2014). In order to simulate the tropical climates, all tests were performed at a constant temperature of 35°C and a humidity of 65% RH. Variation of the temperature and humidity was ±1°C and ±5% RH, respectively. Every test was repeated more than three times under the same conditions.

(1) Flow values, air contents and setting time
Flow values were measured using a mini-slump cone of 50 mm in diameter and 100 mm in height, based on the European Standard EN 12706. Air content and setting time were measured based on the Japanese Standards JIS A 5308 and JIS R 5201, respectively.
(2) Hydration kinetics
A calorimetric test as an index of hydration kinetics was carried out using an isothermal calorimetric test of a triple type. Mortars were mixed and subsequently poured into sample cells where the temperature was controlled at 35°C. The rate of heat evolution was monitored for 12 h.

(3) Dimensional stability, capillary pore pressure and weight change
Figure 1 (top) shows the apparatus used for the measurement of dimensional stability, capillary pore pressure and weight change. Shrinkage/expansion strain was measured using a contactless laser sensor. The laser sensor was set to monitor the horizontal movement of an end rod embedded in the casted mortar. Mortars were cast in a 25 cm long stainless steel mold with a rectangular cross section of 30 mm by 40 mm. Strain was measured continually during the 12 h immediately after casting, with and without a sealed cover over the mortar.

Capillary pore pressure was monitored with a pressure transducer connected to a de-aired water-filled system with a 50 mm needle having an external/internal diameter of 0.7/0.4 mm as shown in Fig. 1 (bottom). Needles were inserted at a depth of 20 mm (middle) and 35 mm (bottom) from the mortar surface. Capillary pore pressure increases until a breakthrough point at which air penetrates locally into the pore structure accompanied by a local pressure breakdown (Slowik et al. 2008). Capillary pore pressure was measured continually after casting with and without a sealed cover over the mortar until the breakthrough point. The weight change of mortars was measured using an electric balance in order to investigate the evaporation rate from the mortar surface. Shrinkage strain, negative capillary pore pressure and weight loss were considered negative values.

(4) Cracking areas
A schematic view of the apparatus used for the cracking test is shown in Fig. 2. A steel plate as a crack inducer was placed inside the plastic mold. Testing was conducted with and without a fan. In the case with the fan, specimens were fanned at 18 km/h by using a circulator to enhance the cracking behavior. The rates of evaporation derived from the evaporation nomograph in ACI 305 R-96 under the fanned and unfanned conditions were 0.80 kg/m²/h and 0.15 kg/m²/h respectively. Cracking areas were analyzed by using an image-processing program ImageJ and were monitored every hour during the 12 h after casting of mortars.

Fig. 1 Apparatus for the measurement of strain, capillary pore pressure and weight change (top) and schematic illustration of pore pressure transducer (bottom).

Fig. 2 Mold and steel plate as a crack inducer for cracking test (unit: mm).
(5) Pore size distribution
The pore size distribution was determined via the MIP method using AutoPore IV 9500 (Micrometrics Instrument Corporation) porosimeter. The contact angle was set at 140°. After paste specimens were hydrated for 24 h with a sealed cover, they were cut into 5-mm cubes and then dried with organic solvents (acetone and chloroform) to stop cement hydration as well as to wash out the residual SRA micelles in the hardened cement paste, followed by D-drying pretreatment. The paste specimens were firstly immersed into acetone for 15 min and subsequently immersed into chloroform for 15 min.

The weight of SRAs extracted from the organic solvents was measured by filtering SRAs from the organic solvents and then drying the filtrated substance containing SRAs (cement pastes contain only SRAs in this measurement). As a result, 75±3% by weight of SRAs dosed in the pastes was extracted from all paste specimens.

(6) Weight loss using TG
The amount of portlandite was measured by TG using a differential thermogravimetric analyzer SSC 5200 (SII Co.) with a resolution of 0.01 mg. A type R thermocouple (Pt-13% Rh/Pt) was used to take temperature measurements in this instrument. The experimental conditions involved were as follows; a nitrogen gas dynamic atmosphere of 100 ml min⁻¹, a heating rate of 10°C min⁻¹ and a platinum top-opened crucible. Alumina powder was used as the reference material. The specimens were heated in the range of 20 to 1000°C at a constant rate. The amount of hydrates was estimated from the weight loss measured in the TG curve between 20 and 430°C of the corresponding TG peak, which represents dehydroxylation of portlandite between 360 and 430°C. Pastes A, B, C and D hydrated for 12 h with a sealed cover were crushed and then dried with acetone, followed by D-drying pretreatment.

(7) Shear modulus and specific shear creep
Shear modulus and specific shear creep of pastes at very early periods (before setting) were investigated by using a physical modular compact rheometer with a concentric-cylinder system. After placing the pastes into the container of the rheometer, the hardening pastes at ages of 5 h were subjected to a controlled shear strain where the shear strain was increased logarithmically from 10⁻¹% to 10⁻⁵% over 120 s to obtain shear stress-shear strain curves, the slope of which represents the shear modulus. In order to obtain creep behavior of hardening pastes at ages of 5 h, shear strain response to the applied constant shear stress of 500, 3000 and 14 000 Pa was monitored over 600 s. Offset shear strain, subtracting the initial strain from the measured strain and dividing by the applied constant stress, was defined as specific shear creep for early periods.

3. Results
3.1 Flow values, air contents and setting times
Flow values, air contents and setting times of mortars and pastes are listed in Table 5. Flow values increased by less than 10% with the dosage of SRAs. Air contents and setting times changed little whether or not SRAs were added. The difference of setting time between mortars and pastes was negligibly small (max. 10 min).

3.2 Hydration kinetics
Figure 3 demonstrates the heat flows of mortars as a function of time. The curves changed little, irrespective of whether SRAs were added and/or SRA dosage was increased.

3.3 Dimensional stability
In Figs. 4 and 5, shrinkage/expansion strains of mortars under uncovered (drying) and sealed (autogenous) conditions are plotted as a function of time. Autogenous shrinkage can be observed in mortars produced under sealed conditions. Both drying shrinkage and autogenous shrinkage can be observed in mortars produced under uncovered conditions. Compared to sealed mortars, shrinkage of uncovered mortars started earlier and was approximately 3 to 5 times higher at the end of measurement. Shrinkage strain was reduced with the addition
of SRA-1 (mortar B), but reduced only slightly with the addition of SRA-2 (mortar C). Reducing effects on shrinkage became more pronounced with a higher SRA dosage (mortar D) under both sealed and uncovered conditions. SRAs did not change the onset of shrinkage, i.e., shrinkage consistently began under the uncovered conditions after 2 h and under sealed conditions after 6 h.

### 3.4 Capillary pore pressure

Figures 6 and 7 show capillary pore pressure of uncovered mortars measured at the middle and bottom as a function of time, respectively. Figure 8 indicates capillary pore pressure of sealed mortars measured at the middle as a function of time. Capillary pore pressure at any given time (until the breakthrough point) was decreased by the addition of SRAs under both sealed and uncovered conditions. Table 6 shows the observed pressure values at 5 h for the uncovered condition and the reduction as compared to the mortar without SRA (mortar A) is given in percent. Changes in capillary pore pressure were not affected by the type of SRAs (mortars B and C) and the increases in the dosage of SRA-1 (mortar D) under both sealed and uncovered conditions. Compared to the data measured at the bottom of the mortar specimen, capillary pore pressure at the middle was generated earlier (middle: 2 h, bottom: 4 h). Table 6 shows that measurement depth did not affect the reducing effects of the SRAs on capillary pore pressure.

### 3.5 Weight change

The weight loss caused by water evaporation from mortars as a function of time is plotted in Fig. 9. Curves of weight change can be divided into 3 stages. At the first
stage, which corresponds to the initial 2 h before the generation of pore pressure, evaporation proceeds from the bleeding of water at the mortar surface. The thickness of the bleeding water layer was too thin to be measured in this study. SRAs dose did not affect the weight change during the initial 2 h, as the difference in water activity between water with and without SRAs is negligibly small (Lura et al. 2007). Once the bleeding water at the mortar surface is consumed by evaporation, capillary pore pressure is generated and pore fluid is brought to the mortar surface and evaporates. During this second stage from 2 to 5 h, there was little difference between mortars A, B, C and D, although capillary pore pressure was already generated, as shown in Figs. 6 and 7. At the third stage, i.e., after 5 h when hydrate formation started significantly as observed by calorimetry as shown in Fig. 3, the weight loss was reduced by the dosage of SRAs and by increasing the SRA dose.

3.6 Crack areas
Figures 10 and 11 represent crack areas of mortars A, B, C and D without and with the fan as a function of time, respectively. SRA-2 decreased crack areas slightly (mortar C), and SRA-1 decreased crack areas drastically (mortars B and D) under both conditions. Although the air current generated by the fan enhanced the formation of cracks (earlier start and larger area for mortar B), cracks were not generated in mortar D and the effect of SRA dosage was not clearly detected in this study.

3.7 Pore size distribution
Figure 12 shows the differences in the pore size distribution among pastes A, B, C and D hydrated for 24 h. Compared to paste A, pores of 40 to 300 nm in volume increased with the addition of SRAs. However, differences in heat release rate (Fig. 3), air contents (Table 5) and the precipitated hydrates shown later in 3.8 (Table 7) among the specimens A, B, C and D were negligibly

### Table 6 Reduction of capillary pore pressure measured at middle (Fig. 6) and bottom (Fig. 7) of mortar specimens after 5 h elapsed using SRAs under uncovered conditions.

|                  | A     | B     | C     | D     |
|------------------|-------|-------|-------|-------|
| At middle        | –37.9 | –26.3 | –24.6 | –28.6 |
| At bottom        | –10.1 | –6.7  | –7.2  | –6.3  |
| Reduction (%)    |       |       |       |       |
| At middle        | -     | 34    | 35    | 25    |
| At bottom        | -     | 29    | 38    |       |

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**Fig. 9** Weight changes of mortars A, B, C and D as a function of time.

**Fig. 10** Cracking areas of mortars A, B, C and D under unfanned conditions as a function of time.

**Fig. 11** Cracking areas of mortars A, B, C and D under fanned conditions as a function of time.

**Fig. 12** Incremental pore volume of pastes A, B, C and D hydrated for 24 h.
small, which should result in comparable pore size distributions.
Specifically, the peak of pore volume between 40 to 100 nm increased with the use of SRA-1, and the peak was higher as the dosage of SRA-1 increased. SRA-1 has a micelle size of approx. 11 to 12 nm (cf. Table 4) in pore solution and could easily be incorporated in the main pore sizes observed by MIP. These increases of pore volume caused by the use of SRAs could possibly provide the evidence for an altered pore structure after residual SRA micelles were washed out from the pores by immersion into organic solvents due to sample preparation.

Pore volume of less than 30 nm decreased with the addition of SRAs. As SRAs adsorb onto the surface of hydrates (Gartner et al. 2017) and partially plug small pores (Fukushima et al. 2010), organic solvents might not be able to infiltrate the pore, which results in the decrease of pore volume of less than 30 nm.

3.8 Weight loss using TG
Table 7 lists the weight loss of pastes A, B, C and D occurring between 20 to 430ºC of the TG curves. There was little difference in the amount of initial hydrates and portlandite among the pastes A, B, C and D hydrated for 12 h.

3.9 Shear modulus and specific shear creep
Figure 13 shows shear stress as a function of the applied shear strain in pastes A, B, C and D hydrated for 5 h. Shear stress increased linearly in all pastes with the increase in the applied shear strain. The modulus of shear decreased with the dosage of SRAs and the increase of SRA dosage. Figures 14 to 16 demonstrate specific shear creep response to the applied constant shear stress of 500, 3000 and 14 000 Pa, respectively. In all cases, specific shear creep increased and then leveled off, and the creep at any given time increased in A, C, B and D in that order. Shear creep response of paste D at 14 000 Pa was plotted during the initial 30 s as the specimen started to flow at 30 s after adding the shear stress.

4. Discussion
4.1 Relationship between capillary tension and early-age shrinkage in the presence of SRAs
According to past reports (e.g., Powers and Brownyard 1947; Wittmann 1976; Lura et al. 2007; Weiss et al. 2008), a dominant influencing factor on early-age shrinkage is seen to be capillary tension. Earlier studies of the authors with conventional cement mortars without SRA showed an increase in shrinkage strain at the time when capillary pore pressure developed (Takahashi et al. 2016).

Accordingly, the relationship between capillary tension and early-age shrinkage in the presence of fatty alcohol-based SRAs is discussed. Table 8 shows the absolute, observed values for shrinkage strain and capillary pore pressure at 5 h in uncovered conditions. Reduction (shown as a positive value) of shrinkage strain and capillary pore pressure as compared to the mortar without SRA (mortar A) is given in percent. Figure 3 indicates that the 5 hours correspond to the onset of the acceleration period of hydration. Compared to mortar without SRA (mortar A), capillary pore pressure was decreased ca. 30% by the dosage of SRAs due to lowered
surface tension of water, as shown in Table 3. Types of SRAs as well as the amount of SRA did not influence the changes in capillary pore pressure due to less change in surface tension between SRA-1 and SRA-2, and between 1% and 3% of SRA-1 solution. However, compared to the mortar without SRA, shrinkage strain was decreased 58% by the dosage of SRA-1 (mortar B), which was twice higher than SRA-2 (mortar C), and it was decreased 76% by increasing the dosage of SRA-1 (mortar D). During the first 5 hours after casting, the hydration rate, setting time and evaporation rate did not differ among mortars A, B, C and D, as shown in Table 5 and in Figs. 3 and 9. These results demonstrate clearly that capillary tension does not directly correlate with shrinkage strain at early ages even under the same profiles of evaporation and hydration, and that the actions of SRAs on reduction of shrinkage at early ages are not explicable solely by capillary tension. Figures 5 and 8 also demonstrate that the reduction of capillary tension does not directly correlate with the decrease in early-age shrinkage under sealed (autogenous) conditions.

For times beyond 5 h after casting, shrinkage strain needs to be discussed by taking into account water evaporation in addition to capillary tension because weight changes differed among mortars A, B, C and D. Liquid water supply to menisci in cement paste/mortar matrix affects water vapor diffusion from menisci, which is a driving force of shrinkage. In this discussion, menisci means the menisci formed in the pores at the vicinity of the specimen surface or the drying front as internal humidity of the specimen is over 99% RH at early ages. Water mainly moves through pores and flow paths as a liquid phase (not a vapor phase) at such high humidity (Bažant and Najjar 1971; Maruyama et al. 2006). Darcy’s law describes that upward movement of liquid water is reduced as the upward flow paths of pores become narrow and/or blocked. If the flow paths become narrow and/or blocked by SRA micelles clogging, the meso-pores form upon hydration and subsequent internal and external drying (Aono et al. 2007), liquid water supply as well as water vapor diffusion is reduced by the addition of SRAs, and shrinkage strain is decreased. This action of SRAs based on Darcy’s effect might become pronounced after the onset of the acceleration period because many menisci start to form at the surface at this time period (Mora-Ruacho et al. 2009). Figure 9 shows that there was less difference in evaporation among mortars A, B, C and D during the dormant period and therefore both SRA-1 and SRA-2 could exist in larger/thicker pores and flow paths. Flow paths where liquid water moves and menisci where water evaporates could shift to a microscopic size at the next time frame (5 to 24 h). Pores and flow paths of tens to hundreds nm increase/decrease in OPC pastes during the time periods from final set to 24 h (Takahashi 2015). Such microscopic pores and flow paths could be impregnated with the finer 10 nm micelles of SRA-1 rather than the agglomerated larger 700 nm micelles of SRA-2, thereby reducing water vapor diffusion from the forming meso-pores in mortars B and D with additions of SRA-1.

In Fig. 12, pore volume of 40 to 100 nm was increased in pastes B and D whose pores and flow paths could be clogged by SRA micelles. The increase of pore volume with the addition of SRAs could imply the increase of residual SRA micelles in the pores of 40 to 100 nm.

4.2 Actions of SRAs on early-age shrinkage besides capillary tension mechanism

Changes in the solubility of portlandite caused by the dosage of SRAs could affect dimensional stability of hardened cement pastes. According to Maruyama et al.
(2016), crystal growth of portlandite becomes stagnant in the presence of alcohol-based SRAs and portlandite precipitates as a fine and rigid clumping structure in or near the outer C-S-H, which effectively resist drying shrinkage. Sant et al. (2011) reported that the presence of SRAs increases the portlandite oversaturation level in pore solution at early ages, which can result in higher crystallization stresses that could lead to an initial expansion. However, it is unlikely that fatty alcohol-based SRAs used in this study changed the solubility of portlandite because less change in the amount of portlandite and initial hydrates and heat release rate was observed by the dosage of SRAs, as shown in Table 7 and Fig. 3.

Aggregation and dispersion of C-S-H units during dry and wet cycles cause long-term shrinkage and expansion of hardened cement pastes. C-S-H units become irreversible and agglomerated as a cluster at high temperature and low humidity, and the coarsened pore structure leads to an increase of long-term shrinkage (Fukushima et al. 2010). Gartner et al. (2017) hypothesized that alkoxide groups of the organic molecules of SRAs can partially replace hydroxide groups of cement hydrates and the molecules adsorb onto C-S-H surfaces. The molecules adsorbed on the surface prevent the formation of hydrogen bridges between silicate groups brought into close proximity by capillary forces during drying. Maruyama et al. (2018) and Fukushima et al. (2010) reported that SRA micelles remaining in small meso-pores could prevent the agglomeration of C-S-H due to drying. In other words, a hydrate matrix with incorporated SRA micelles is formed and could mitigate capillary pore pressure as stress absorbers and/or stress reducers, and therefore shrinkage strain is reduced. According to earlier studies, (e.g., Fukushima et al. 2010; Gartner et al. 2017; Maruyama et al. 2018), this action of SRAs mainly affects the long-term shrinkage while the effects of SRAs on the early-age shrinkage have not been discussed.

When we refer to the above action of SRAs, our results demonstrating that capillary tension does not directly correlate with early-age shrinkage in the presence of SRAs, and SRA type and dosage affect the reduction of shrinkage, can be explained as follows. Finer and nanoscale micelles of SRA-1 could easily absorb onto hydrate surface and/or could exist in capillary pores and small meso-pores. Thereby, the reduction of shrinkage strain in the mortar with SRA-1 was higher than SRA-2 even under the same capillary pressure condition with almost same surface tension. With increasing dosage of SRAs, the adsorption of SRA micelles onto hydrate surface and/or the clogging of capillary pores and small meso-pores by SRA micelles could become pronounced. Consequently, shrinkage was decreased with increasing dosage of SRAs even though capillary pore pressure changed little. An increase in the pore volume at 40 to 100 nm, related to the dose of SRAs (Fig. 12), could possibly provide the evidence of residual SRA micelles in pores. As pores in the range of tens to hundreds nm increase/decrease in OPC pastes during the time period from final set to 24 h (Takahashi 2015), the residual SRA micelles could act as stress absorbers and/or stress reducers.

Furthermore, the matrix could mitigate cracking when shrinkage strain occurs. Figures 4 and 10 indicate that the reducing effect of SRAs on cracking area followed the same pattern as the reducing effect on shrinkage strain, i.e., cracks were not observed in the mortar with SRA-1 (mortar B) but in the mortar with SRA-2 (mortar C) where cracks were observed at the same magnitude as in the mortar without SRAs (mortar A).

Changes in specific shear creep could experimentally demonstrate the presence of the hydrate matrix with incorporated SRA micelles. Figures 14 to 16 indicate that specific shear creep is higher in paste B (SRA-1) than in paste A (without SRA) and paste C (SRA-2) at the applied shear stress of 500, 300 and 14 000 Pa. As mentioned above, the SRA with finner micelles (SRA-1) could easily form a hydrate matrix with incorporated SRA micelles. When shear stress is applied to the matrix, the micelles could act as lubricant between cement particles/hydrates and increases the creep. An increase in specific shear creep provoked by increasing the dosage of SRAs (paste D) could indicate the enhancement of the slip of the micelle themselves due to a volumetric increase of the incorporated fine micelles in the hydrate matrix. Thus, results of shear creep measurement have correlation with the reducing effects of SRAs. Designing SRAs by utilizing the shear creep measurement could be useful when customized SRAs appropriate to local uses and climate conditions are required. Further investigation using variety of SRAs is necessary in order to discuss more precisely the changes in shear creep and the corresponding relationship with shrinkage and cracking.

5. Conclusions

This study examined the effects of fatty alcohol-based SRAs on shrinkage strain, capillary pore pressure and crack area of cement-based mortars at early ages under high temperature conditions. Although a dominant factor on early-age shrinkage was seen to be capillary tension, our results identified less direct correlation between early-age shrinkage and capillary tension under the same profiles of evaporation and hydration. In order to interpret the actions of SRAs on early-age shrinkage, we hypothesized that SRA micelles adsorb onto hydrates and/or exist in small pores, and form the hydrate matrix with incorporated SRA micelles. The matrix could mitigate the capillary tension as stress absorbers and/or stress reducers, and thereby shrinkage strain and crack area are reduced.

The concept of the matrix with incorporated SRA micelles could be experimentally demonstrated by the changes in specific shear creep of pastes with SRAs - when shear stress has occurred in the mortar specimen with fine SRA micelles, the micelles themselves could slip in the matrix and increases the creep.
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