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Implications of pre-formed microcracking in relation to the theories of DEF mechanism

S.O. Ekolu [a],*, M.D.A. Thomas [b], R.D. Hooton [a]

[a] *Department of Civil Engineering, University of Toronto, 35 St. George Str, Toronto, ON, Canada M5S 1A4*
[b] *Department of Civil Engineering, University of New Brunswick, P.O. Box 4400, Fredericton, N.B., Canada E3B 5A3*

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Abstract

Microcracks were induced in cementitious systems by freeze-thaw action and by alkali–silica reaction. These mechanisms often co-exist with delayed ettringite formation in concretes. Mortars and concretes were subjected to a heat treatment cycle consisting of a pre-set period of 4 h at 23 °C followed by accelerated curing at 95 °C. To isolate the mechanical effects of induced microcracking, heat-cured specimens were subjected to varied prescribed damage induced by freeze-thaw or alkali–silica reaction prior to the onset of delayed ettringite formation. It was found that inducement of pre-formed microcracks led to an earlier onset of expansion due to delayed ettringite formation. Initially, microcracks enhanced ultimate expansion until a certain relatively high extent of microcracking was reached. Thereafter, ultimate expansion decreased with any further increase in microcracking. This report gives support to the paste expansion theory.

Keywords: Delayed ettringite formation; Microcracking; Steam-curing; Alkali–silica reaction; Freeze-thaw

1. Introduction

1.1. Inducement of microcracks in cementitious systems

In cementitious systems, the presence of microcracking weakens the microstructure of the paste matrix. As well, microcracks provide spaces for ettringite crystallization. Batic et al. [1] investigated the effects of mechanically induced microcracking using moist-cured concrete specimens of size 4 × 4 × 16 cm. The specimens were loaded to 80% of their flexural strength and subsequently stored in limewater. Ettringite formation was observed in pores and around the aggregate-paste interface. Fu and Beaudoin [2] investigated delayed ettringite formation (DEF) in concretes that had been initially subjected to thermally induced microcracking. The cycle consisted of heat curing of specimens at 95 °C followed by wetting in water at 23 °C for 6 h then oven dry-heating at 85 °C for one day. The 0.43 water–cement ratio (w/c) concretes were made using ASTM Type I, III, and V Portland cements. DEF expansion was observed in those specimens made using Type III cement. Results showed that microcracking can lead to increased deterioration due to DEF.

The Duggan test [3] has also been used to induce thermal microcracks in cementitious systems. It involves applying two wet-dry cycles, each cycle consisting of oven dry-heating at 82 °C for one day followed by wetting in distilled water at 21 °C for one day. Finally, specimens are oven dry-heated at 82 °C for three days before cooling and storage in water. Among other criticisms [4–7], the test is considered too severe and the observed expansion might induce DEF as it involves temperatures above the threshold of about 70 °C.

1.2. ASR and DEF co-existence

In cementitious systems where both alkali–silica reaction (ASR) and DEF mechanisms co-exist, Pettifer and Nixon [8] suggested that sulfate attack may promote ASR by increasing the alkalinity of the pore solution. But later studies [9] have shown that ASR occurs first followed by crystallization of ettringite in the cracks formed, and that ASR plays the main role of inducing cracks [10]. Also, ASR lowers the alkalinity of the pore solution thereby promoting ettringite formation [11].

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* Corresponding author. Now at the School of Civil and Environmental Engineering, University of the Witwatersrand, Private Bag 3, WITS 2050, Johannesburg, South Africa. Tel.: +27 11 717 7104/7121; fax: +27 11 3391762.
E-mail address: ekolus@civil.wits.ac.za (S.O. Ekolu).

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Table 1
Composition of CSA Type 30 Portland cement

	SiO_2	Al_2O_3	Fe_2O_3	CaO	SO_3	K_2O	Na_2O	Na_2Oe	LOI	Blaine (cm^2/g)
CSA Type 30 cement	20.38	5.45	2.06	63.41	4.82	1.21	0.10	0.90	0.91	5490

1.3. Freeze-thaw and delayed ettringite formation

The main influence of freeze-thaw (F/T) action on concrete is through the formation of F/T microcracks. This mechanical effect directly reduces the modulus of elasticity of concrete and is the basis for some standard F/T test methods [12]. Some studies [13,14] also appear to suggest that F/T conditions might support the formation of ettringite (AFt) at the expense of monosulphate (AFm) within a cementitious system.

Microcracking within the cement paste matrix weakens concrete by reducing its tensile strength [15]. The tensile strength property is important for resisting expansive pressures [16]. But several factors including the microcracking patterns, size and number of cracks, crack locations, the degree of microcracking, and the method of crack inducement are bound to contribute to the influence of microcracks on DEF expansion. In this investigation, microcracking in mortars and concretes was induced by ASR and freeze-thaw damage mechanisms prior to the onset of DEF. These microcracks were isolated and their mechanical effects on DEF expansion were then examined.

2. Experimental

Mortar mixtures of 1:2.25:0.47 cement to sand to water were prepared using crushed Placitas aggregate. Specimens consisting of 25 × 25 × 285 mm mortar bars were then made for the experiment involving ASR-induced microcracking. The Placitas aggregate (NM) was of gravel rock type known to contain reactive volcanic glass [17]. The NM mortar specimens identified using the notations NM/95/nalw/14d, NM/95/nalw/2m, NM/95/nalw/6m were steam-cured at 95 °C and kept in NaOH at 38 °C for 14 days, 2 months, 6 months respectively (see Figs. 2 and 4).

At the end of these prescribed periods, the specimens were removed from NaOH then stored in saturated limewater at room temperature. Concrete mixtures of 0.45 w/c were made using non-reactive limestone aggregate and used to prepare 75 × 75 × 285 mm concrete prisms for the experiment involving F/T induced microcracking. The prisms were steam-cured at 95 °C then subjected to F/T cycles until the original elastic modulus (Ed) was reduced to 70% Ed for specimens identified as 95 °C-70% Ed, and to 10% Ed for specimens 95 °C-10% Ed (see Fig. 7). The prisms were then stored in water at room temperature. Control specimens consisting of mortars bars and concrete prisms were not heat treated, but were cured at 23 °C and 95% RH in a fog room. Similar notations as for heat-cured specimens are used for control specimens, replacing 95 °C with 23 °C. The steam-curing cycle applied consisted of a 4 h pre-set period, temperature rise at 20 K/h, then the maximum temperature of 95 °C was maintained for a specified period of time before cooling back to 23 °C. The maximum temperature was held for 9 h in accordance with the conventional 18-hour curing cycle for concretes [12], and for 12 h according to the Kelham method used for heat curing of mortars [18]. No air entrainment or admixtures were used in the mixtures. Table 1 shows composition of the cement used.

3. Results

3.1. ASR-induced microcracking

The mechanical effects of ASR-induced microcracking on DEF were isolated to get insight into the role of microcracks in field concretes where both ASR and DEF damage mechanisms may co-exist. It is known [11] that ASR has a chemical effect of promoting ettringite crystallization as a result of the consumption of pore solution alkalis. To isolate the mechanical effects, ASR damage in mortars was left to occur for 14 days, 2 months, and 6 months in NaOH at 38 °C. At the prescribed periods, specimens were removed from NaOH, cooled to room temperature and immediately stored in limewater. Pore solution alkalis are known to hinder delayed ettringite formation [11]. Hence upon transfer of the mortars from NaOH to limewater, the alkalis in mortars leach out quite rapidly, promoting ettringite crystallization for DEF. This method used to isolate microcracking due to ASR is illustrated in Fig. 1. Soon after the transfer of specimens from NaOH into limewater, ASR is expected to stop as pore solution alkalis leach out. Consequently, there is a brief period of little or no expansion characterized by a plateau prior to the onset of DEF expansion. This region is important as it indicates, (1) whether ASR actually stopped upon transfer of specimens from NaOH to limewater, (2) whether no DEF occurred in conjunction with ASR during storage in NaOH and,

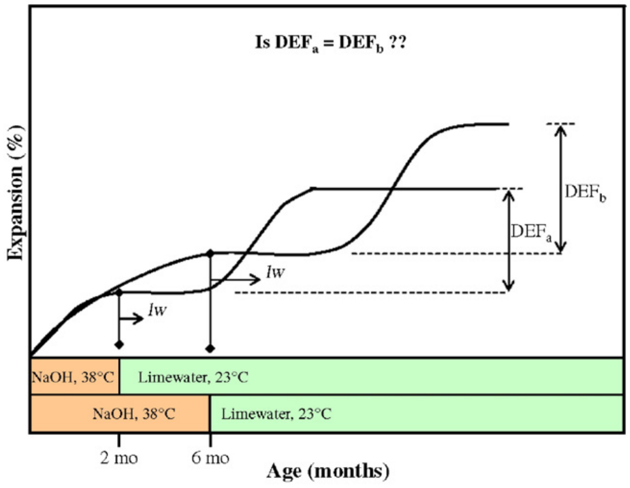

Fig. 1. Schematic of the methodology used to isolate mechanical effects of ASR microcracks on DEF (DEF — delayed ettringite formation, lw — limewater, mo — months).

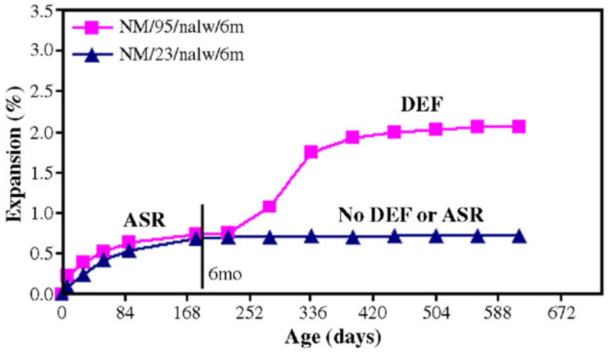

Fig. 2. DEF hindered during storage of heat or moist cured mortars in 1M NaOH solution (NM — Placitas, nalw/6m — kept in NaOH for 6 months then stored in limewater (lw)).

(3) the relative time to the onset of DEF expansion depending on the degree of microcracking developed during ASR.

In this investigation, it was considered that prior to the removal of specimens from NaOH, no DEF will have occurred. In Fig. 2, it can be seen that both the heat-cured and non-heat cured specimens showed similar ASR expansion and no DEF during the 6 months period when the specimens were kept immersed in NaOH. Upon transfer of specimens to limewater, the heat-cured specimens showed DEF expansion while the corresponding non-heat cured specimens did not show DEF or further ASR expansion. Also, it is apparent in Fig. 3 that ASR stopped soon after the transfer of specimens from NaOH solution to limewater irrespective of the length of specimen storage in NaOH. These data suggest that alkalis leached out of mortars quickly, stopping ASR.

Fig. 4 shows data of the mechanical influence of varied degrees of ASR-induced microcracking on DEF expansion. It is expected that the longer the specimens were kept in NaOH at 38 °C to allow ASR to proceed, the greater the extent of microcracking developed as monitored by ASR expansion. As a control, some specimens were not kept in NaOH but were instead stored in limewater at room temperature right after heat treatment.

3.1.1. Onset of delayed ettringite formation

It can be seen in Fig. 4 that the shorter the period of specimen storage in NaOH, the earlier DEF expansion commenced. In Table 2, the various periods of specimen storage in NaOH are given along with the corresponding observed time taken for DEF expansion to commence. The specimens that were kept longer in NaOH also took longer times to show DEF expansion. The major factors here that influence the onset of DEF are the reserve of alkalis in the specimens and the degree of microcracking developed during specimen storage in NaOH. It is possible that those specimens that were kept longer in NaOH also accumulated larger reserves of alkalis. Upon transfer of specimens from NaOH to limewater, the alkalis would be leached more quickly from those specimens which spent a shorter time in the NaOH solution, therefore promoting an earlier onset of DEF. In this analysis, however, it has been assumed that the period taken to leach out the alkalis was the same for all specimens. This is considered a reasonable assumption since NaOH and KOH alkalis characteristically leach out rapidly as discussed in Section 3.1, and also reported in a study by Famy [19] on heat-cured mortars.

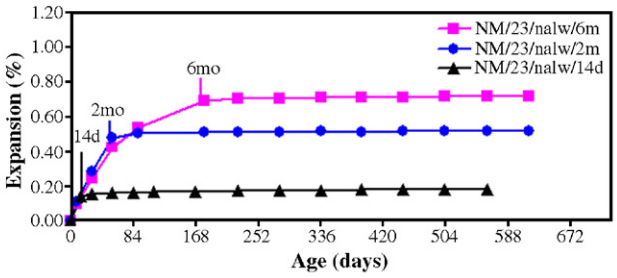

Fig. 3. ASR stopped immediately upon transfer of moist cured mortars from NaOH to limewater (NM — Placitas, nalw/6m — kept in NaOH for 6 months then stored in limewater (lw)).

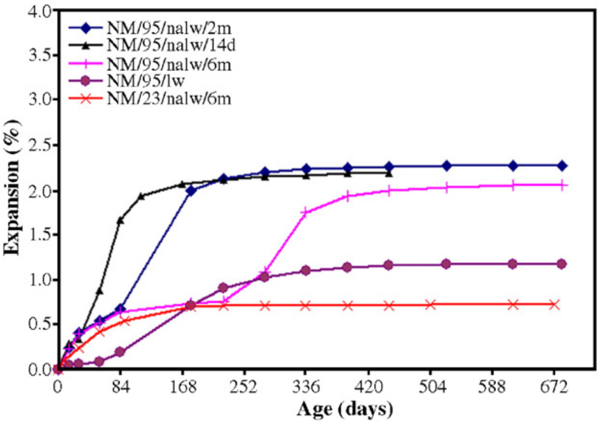

Fig. 4. Mechanical effects of pre-formed ASR microcracking of varying levels on delayed ettringite formation. The varied levels of ASR microcracking were achieved by keeping specimens in NaOH at 38 °C for 14 days, 2 and 6 months prior to storage in limewater (NM — Placitas, nalw/2m — kept in NaOH for 2 months then stored in limewater (lw)).

One of the theories that has been advanced, the *crystal pressure theory* [9,20–24] advocates that the crystallization of late ettringite in pre-formed cracks generates the required pressure that eventually causes expansion. If this is true then

Table 2
Length of storage in NaOH versus the time taken for onset of expansion due to delayed ettringite formation

Length of ASR expansion in NaOH at 38 °C prior to storage in limewater	Time between removal from NaOH to DEF expansion of 0.10%	Observations
0 days	56 days	
14 days	7 days	No plateau between the consecutive readings. Steep gradient.
2 months	21 days	No plateau between the consecutive readings. Less steep gradient.
6 months	>56 days	A plateau of at least 56 days before onset of DEF expansion.

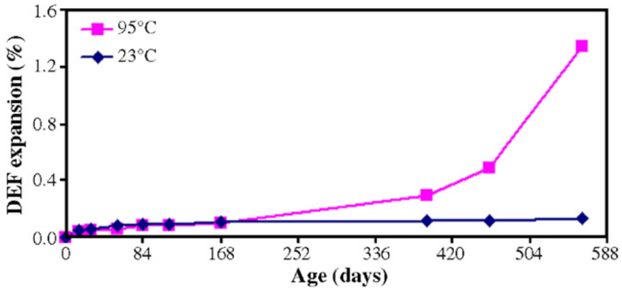

Fig. 5. Expansion of pastes of 0.47 water–cement ratio, heat cured at 95 °C and stored in limewater at 23 °C.

an increased size and/or number of pre-formed cracks must extend the time to the onset of DEF. In accordance with the theory, the existing cracks have to be initially filled with ettringite before expansion is observed. Consequently, highly cracked mortars should take longer to develop expansion. In other words, pre-formed microcracks will tend to extend the time required for expansion to commence but the ettringite formation process itself will have started before the observation of expansion. In this investigation, it was observed that those specimens that had a lower degree of cracking showed DEF expansion sooner. But interestingly, the specimens that had not been subjected to microcracking took longer to develop DEF expansion than the pre-cracked specimens. Hence these results may not justify the crystal pressure theory. Also, cement pastes having no aggregate-paste gaps or microcracks still exhibit DEF expansion as reported in the literature [25,26] and also shown in Fig. 5.

3.1.2. Extent of DEF expansion

The ultimate DEF expansion as influenced by varied levels of ASR expansion, representing different extents of ASR-induced microcracking, was analyzed as given in Fig. 6. The data were obtained as illustrated in Fig. 1 on the assumption that expansion due to DEF commenced only after ASR had been stopped by leaching out of alkalis. It can be seen that increase in ASR expansion generally led to an increase in ultimate DEF expansion. But this effect was more pronounced for a certain range of ASR expansion. The extent of microcracking corresponding to ASR expansion of 0.23% to 0.50% gave the highest level of DEF expansion.

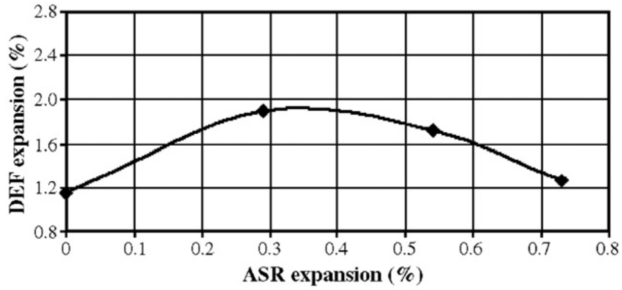

Fig. 6. Effect of pre-formed ASR microcracks on the extent of expansion due to delayed ettringite formation.

The *paste expansion theory* [19,26–28] argues that the expansive pressure responsible for DEF is developed within the paste matrix resulting in paste movement that will in turn cause microcracks to occur at the aggregate/paste interface and within the paste. Ettringite then eventually crystallizes in these microcracks. Hence, visible ettringite crystallization is viewed as a consequence rather than cause of expansion. The credibility of this theory is based on the understanding that the formation of late ettringite itself takes place within the paste matrix when the SO_4^{2-} desorbed from C–S–H encounter the presence of monosulphate in confined spaces [19]. It follows that the integrity of the paste matrix must be of a significant influence on the observed expansion as weaker pastes more easily succumb to expansive pressure.

As already seen, ASR-induced microcracking generally led to an increase in DEF expansion. This can be explained by weakening of the paste matrix as a result of the induced microcracks, lowering the ability of the paste to intrinsically resist expansive pressures during delayed ettringite formation. The mortars that were not microcracked showed lower DEF expansion than the pre-cracked mortars. This is presumably due to the sound structural integrity of the paste matrices for uncracked specimens. Although the elastic modulus in mortars was not monitored, it is known that the modulus reduces with increase in microcracking [24] which in this experiment was provided by ASR expansion.

In summary, the mortars that were not microcracked took longer to show DEF expansion and also had lower ultimate DEF expansion compared to the microcracked mortars. The effect of microcracking on DEF expansion was directly influenced by the extent of induced microcracking. The lower the extent of ASR-induced microcracking, the shorter the time to DEF onset. Pre-formed microcracks also initially increased the ultimate DEF expansion up to a certain extent of microcracking, represented by 0.23% to 0.50% ASR expansion. Beyond this critical range, any further increase in microcracking showed a corresponding decrease in ultimate DEF expansion.

3.2. Freeze-thaw induced microcracking

The effects of microcracks on DEF were also investigated using F/T-induced microcracking in concretes, as discussed in the experimental details given in Section 2. As seen in Fig. 7, F/T-induced microcracks gave a significant enhancement to

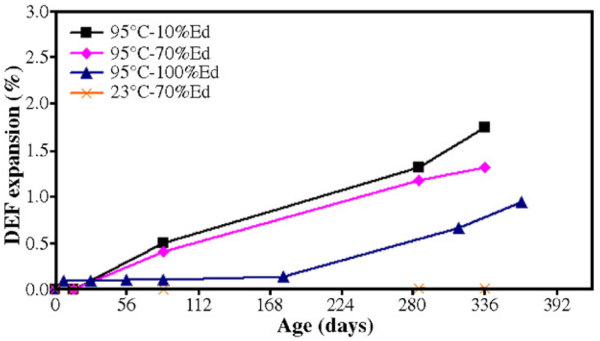

Fig. 7. Effects of the pre-formed freeze-thaw microcracks on expansion due to delayed ettringite formation (Ed — elastic modulus).

DEF expansion. The damaged heat-cured concretes not only developed DEF earlier but they also showed higher ultimate DEF expansion in comparison to their undamaged counterparts. At the age of one year, DEF expansion values were 1.75%, 1.32%, and 0.93% for the heat-cured concretes whose moduli were 10%, 70%, and 100% of the initial elastic moduli respectively. No DEF expansion was observed in non-heat cured concretes despite being subjected to pre-formed F/T microcracking. It was not possible to determine whether decrease in DEF expansion due to microcracking beyond the critical level (which gives maximum DEF expansion as observed in mortars subjected to ASR-induced microcracking), could also occur in concretes under F/T-induced microcracking. This experiment was a limited investigation with only two F/T-induced damage levels (10% Ed and 70% Ed) used. To observe the critical level effect in concretes, if any, would require more F/T damage levels than used in this limited experiment.

4. Conclusions

The influences of other damage mechanisms that often co-exist with delayed ettringite formation in cementitious systems were investigated with emphasis on the effects of ASR and freeze-thaw induced microcracking.

1. The presence of microcracking in DEF expansive cementitious systems promotes both an earlier onset of delayed ettringite formation and a greater related ultimate damage.
2. There appears to be a critical extent of ASR microcracking in mortars that gives maximum ultimate expansion due to delayed ettringite formation. Beyond this critical level, any further increase in microcracking lowers the ultimate expansion.
3. The integrity of the paste matrix has a significant influence on the ability of a cementitious system to resist expansion. The analysis gives credence to the paste expansion theory of delayed ettringite formation.

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