

INFLUENCE OF PASTE QUALITY CONSTITUENTS ON DRYING SHRINKAGE

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MASTER OF SCIENCE

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ABSTRACT

The paste system of concrete materials often holds the “DNA” to understanding their potential mechanical properties. As drying shrinkage and related cracking remains a top concern for the service life of concrete, this paper explores the effect of paste quality on drying shrinkage. The semi-adiabatic calorimetry, restrained, and unrestrained shrinkage of concrete made with 15 different cementitious materials were studied. Cementitious blends included fly ash, slag, silica fume, metakaolin, zeolite and limestone as paste volume was kept constant across mixes. Results showed that the rate at which the paste system allowed free water to dissipate at early ages may have dictated the extent of drying shrinkage. Higher cement replacement with limestone improved the tensile capacity of concrete which resulted in better drying shrinkage cracking resistance. High tricalcium aluminate and alkali contents in cements were found to be drivers of early age shrinkage and potential cracking at later stages in concrete.

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DEDICATION

The study is dedicated to everyone at Strata Corporation and Terracon who in the mist of the COVID pandemic risked their lives to assist with the research.

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LIST OF ABBREVIATIONS

- SCMSupplementary Cementitious Materials
- PLCPortland Lime Cement
- RH.....Relative Humidity
- HPCMHigh Performance Concrete Material
- C-S-HCalcium Silicate Hydrates

LIST OF SYMBOLS

$^{\circ}\text{C}$Celsius

$^{\circ}\text{F}$Fahrenheit

μeMicron

1. INTRODUCTION

Concrete remains one of the most widely used construction materials because of its strength, durability, and low maintenance costs. It consists of primarily cementitious paste and aggregates. Fresh cement paste is typically a plastic network of particles of cement in water, however, when hardened, consists of hydrates of various compounds, crystals of CaOH, unhydrated cement and the residue of the water-filled spaces from the fresh paste called capillary pores[1]. Shrinkage primarily occurs because of loss of water from these capillary pores, either through self-desiccation (autogenous shrinkage) when the cementitious material hydrates or drying (drying shrinkage).

Although, several forms of volume changes exist[2], a significant portion of the total deformations in concrete can be attributed to drying shrinkage[3]. As concrete shrinks, any form of restraint (internal or external) causes the development of tensile stresses. It is also possible that tensile stresses can develop because of uneven drying in layers of a concrete element. Shrinkage cracking occurs when tensile stresses exceeds the tensile strength of the concrete. The cracks allow for the ingress of deleterious ions into the concrete which results in the loss of durability and serviceability of structures [4-6].

Hydrated cement paste is considered the ingredient in concrete that is responsible for its properties such as strength and shrinkage. The paste system is subject to moisture-related movement whereas aggregates tend to be dimensionally stable. Therefore, understanding the shrinkage behavior of the cement paste itself should be the key to minimizing shrinkage within the concrete material. Taylor and Wang[7] discussed the drying shrinkage factors that affect the paste system under paste quantity and paste quality. Drying shrinkage almost linearly increases with increasing paste volume in a concrete mixture [8-14]. Since the drying shrinkage and drying

shrinkage cracking potential of most cementitious materials are studied in isolation, this does not lend itself to easy comparison due to differing paste quantities. This has led to a barrage of conflicting reports on the actual effect of cement compositions and supplementary cementitious materials (SCM) on drying shrinkage and related cracking[7, 15-17]. In this regard, there appears to be some research gaps in the way cements and cementitious combinations influence the drying shrinkage cracking potential of concrete elements.

Existing literature suggests that cement fineness and cement composition (mainly C_3A , alkali content) could have some effect on shrinkage[18-25]. Portland Cement Association[26] reported that; when SCMs are used in low to moderate amounts, the drying shrinkage effect are likely to be generally small and insignificant. As such, it will be of value to study the actual drying shrinkage impact of the various commercially available SCMs like fly ash, slag, silica fume and metakaolin at moderate to high dosages. Additionally, the use of limestone as a partial replacement in Portland Lime Cement (PLC) has been reported to provide a good particle size distribution and improve the durability of concrete. Studies have shown similar or lower drying shrinkage with the addition of limestone[27, 28], however its effect on drying shrinkage cracking requires further study. Also, recent reports indicate that natural zeolites, as a cement replacement, possesses internal curing properties which could be beneficial to drying shrinkage of concrete[29, 30].

1.1. Objectives and Dissertation Organization

This thesis aims to explore the net influence of paste quality on the drying shrinkage properties of concrete elements, taking into consideration the effect the binder fineness, physical and chemical composition, and SCMs as the main constituents. To do this, the drying shrinkage behavior of fifteen (15) different cements and cementitious combinations are investigated,

keeping paste content, water content and other external factors constant. The thesis is organized into six chapters.

Chapter 1 sets up the overall premise of the research work whereas Chapter 2 reviews the existing literature that underline concepts, methodologies and materials explored in the various studies.

Chapter 3 discusses the experimental program used for the study; taking into account the material selection process, specimen preparation and testing regime used. The results from the study are presented and discussed in Chapter 4.

The outcome of all the studies is summarized in chapter 5, highlighting the potential for future research, and recommending ideas for further improvement of concepts.

2. LITERATURE REVIEW

2.1. Factors Affecting Shrinkage

Volume changes in concrete can occur under different conditions and at different ages. Plastic shrinkage, drying shrinkage, and chemical shrinkage (autogenous shrinkage) are some of the known causes of volume changes in concrete, but the emphasis of this research is on drying shrinkage. Drying shrinkage is due to the loss of water from capillary pores, normally by evaporation. At a relative humidity (RH) between 45% and 95%, free water in the pores is readily extracted from the system. When the RH in the pores drops below about 45% then water adsorbed to the walls of the pores starts to be removed, allowing pores to close and solid particles to move closer together[31].

Typically, water exists in hydrated cementitious materials as either free water or bound water. Free water refers to the part of the mixing water that has not reacted with the cement phases whereas bound water is chemically combined in the solid phases or physically bound to the solid surfaces. Free water presents the most concern in terms of drying shrinkage because it is the loss of free water from the capillary pores that primarily causes shrinkage of the concrete specimen. Lee and Zollinger[32] asserted that quantitative estimate of free water could be helpful to in evaluating the quality and other related properties of hardening concrete. As such, the influence of paste quality on free water is paramount to understanding the behavior of cementitious materials under drying shrinkage.

In practice, the degree of shrinkage of a concrete specimen is affected by several interrelated factors. Taylor and Wang[7] summarized these factors in Table 2.1 and categorized the bulk of the factors under, the paste quantity and the quality of the paste. Primarily, the concrete's paste system is subject to moisture-related movement and aggregates tend to be

dimensionally stable. Drying shrinkage almost linearly increases with higher paste quantity/volume in a concrete mixture[8-14]. The quality of includes such as composition and amounts of cementitious materials, w/cm, and air void system. This review discusses current literature on the effects of different paste quality constituents on drying shrinkage.

2.2. Influence Cement Chemistry on Drying Shrinkage

Reports suggests that the composition of cement can affect drying shrinkage[21]. Observations indicate that higher C₃A and alkali content increase shrinkage [33-35]. Li et al [23] confirmed that the increase in the alkali content of cement increased the drying shrinkage of High Performance Concrete Materials (HPCM) with or without Fly Ash

Also, the sulfate content in cement affects the drying shrinkage of concrete[22, 24]. Pickett[36] observed that the effect of C₃A and alkali content on shrinkage is influenced by the gypsum content of the cement, i.e. shrinkage of cements of the same C₃A content differs for different gypsum contents. Therefore, it may be possible to reduce the drying shrinkage by increasing the gypsum content in cement[25]. Some specifications in Australia specify limits on the chemical composition of cement as a means to control shrinkage of concretes in field applications. These specifications require a maximum C₃A content of 7% and a minimum SO₃ content of 1.8% [21].

Table 2.1. Factors Affecting Shrinkage, Adopted from Taylor and Wang[7]

Categories	Factors	Specific factors	General findings and conclusions on shrinkage
Paste quality	Cement characteristics	Chemistry	Higher sulfate and gypsum content may reduce shrinkage
		Fineness	Through change of the rate of hydration and water demand, finer cement leads to higher shrinkage
	SCM	Fly ash	Class F fly ash potentially decreases shrinkage; Class C fly ash increases shrinkage with increasing dosage
		Slag cement	Conflicting results, depending on paste content; overall, comparable to ordinary PCC mixtures
		Silica fume	Conflicting results: depending on w/cm, curing period, and paste content; limited replacement may reduce long-term shrinkage
		Ternary mixtures	Normally, adverse effect can be diminished (i.e., PC+slag cement+silica fume) or even reduced (i.e., PC + slag cement + F fly ash)
	Chemical admixtures	Superplasticizers	May increase concrete shrinkage with a given w/cm ratio and cement content; dependent on admixture chemistry
	Air content		No significant effect on the magnitude of drying shrinkage if it is less than 8%
Paste quantity	w/cm		Influence is relatively small with a constant paste content for w/cm > 0.40
	Paste content		For a given w/cm ratio, shrinkage linearly increases with increasing paste content/volume
Other factors	Aggregate	Type	Influenced by mixing water demand and the stiffness of the aggregate
		Gradation	Indirect influence through the change of water demand and paste content; increased size results in decreased paste content, so decreased shrinkage
		Fines	Most fines increase shrinkage
	Curing	Duration	Increasing curing period reduces the amount of unhydrated cement, resulting in reduced overall shrinkage
	Environment	Relative humidity	Higher relative humidity leads to lower shrinkage
		Temperature	Higher relative humidity leads to lower shrinkage
		Wind	Similar to temperature effect
	Construction		Improper addition of water during finishing, and poor curing will increase shrinkage
	Geometry		Increasing the volume-to-surface area ratio decreases shrinkage

2.3. Influence of Cement Fineness

The fineness of cement affects the heat released and the rate of hydration of portland cement which invariably leads to more rapid strength gain in concrete. However, Brewer and Burrows' [18] work concluded that that coarse-ground cement produces more durable concrete than fine-ground cement. Bennett and Loat[37] explained that finer cements lead to a finer pore structure leads to a higher early age shrinkage. Generally, the higher the cement fineness, the higher water demand which in turn results in increased shrinkage. It has been reported that the greater water requirement of the finest cement resulted in about 200×10^{-6} greater strain at the same workability level [37]. The ACI Committee 209 [38] also concluded that finely ground cements result in greater shrinkage than coarser ground cements.

2.4. Influence of SCM's and Mineral Admixtures

SCM's are material used with portland or in blended cement to improve the desirable properties of concrete by way of pozzolanic activity. According to ACI 211 commentary[39], the proportioning of SCMs in concrete mix is predicated on chemical activity, water demand, density (or specific gravity), dosage rate of admixtures, the effect on other critical properties, and the amount of cementitious materials and cement needed to meet the requirements for the particular concrete. However, in terms of shrinkage, there seem to be conflicting findings on effect of proportioned SCMs, although one might expect an increase in drying shrinkage due to the increased proportion of C-S-H and the finer pore structure[40]. Nonetheless, reviewed literature seems to indicate that the contribution of SCMs to shrinkage may be affected by the dosage, the fineness, water demand of the SCM, and the overall paste content of the mix. The existing literature on SCMs used in this research are discussed as follows.

2.4.1. Fly Ash

Fly ash, a by-product of coal combustion, is widely used as a cementitious and pozzolanic ingredient in portland cement. The use of fly ash in concrete is regulated by ASTM C618[41] and can be categorized as either Class C or F Fly ash. ACI 211 commentary[39] recommends a dosage of 15 to 25 percent for Class F fly ash , and 15 to 35 percent for Class C fly ash. However, in terms of drying shrinkage, ACI 232 commentary[15] reported that in those cases where the addition of fly ash increases the paste volume, drying shrinkage may be increased slightly if the water content remains constant. If there is a water-content reduction, shrinkage should be about the same as concrete without fly ash.

Atis[42] the studied the replacement of ordinary cement by 50% and 70% Class F fly ash by mass. In order to maintain the same workability, the actual w/cm ratios for the fly ash mixtures were less than those for the control mixtures. Also, the paste volume in fly ash mixes was a little higher (27.3% in 50% fly ash mixture and 26.9% in 70% fly ash mixture compared to 25.9% in the control mixture) due to varied specific gravity. After curing in the same condition, significantly lower shrinkage was observed for the high-volume fly ash concrete than for the control concrete at the first day. At an age of six months, the 70% fly ash mixture showed the lowest shrinkage compared to the control mixture.

Gesoğlu et al[43] replaced Portland cement with Class F fly ash at dosages of 20%, 40%, and 60%. It was observed that drying shrinkage was reduced by using Class F fly ash, and this beneficial effect appeared to be more pronounced with increasing replacement levels. A similar conclusion was drawn in another investigation of fine Class F fly ash effects on shrinkage [44].

2.4.2. Slag Cement

Slag cement is also an industrial by-product. It is essentially a nonmetallic product, consisting essentially of silicates and aluminosilicates of calcium and of other bases, that is developed in a molten condition simultaneously with iron in a blast furnace, and then rapidly cooled into glassy form. The use of slag in concrete is regulated by ASTM C989[45] and the typical dosages fall between 25 and 70percent[39]. Slag has little effect on shrinkage at normal dosages, but finely ground slag is suspected to increase shrinkage due to increase in water demand[26]. Nonetheless, ACI 233 commentary[16] advised that a potential for reducing shrinkage in a concrete mixture may be realized when the higher strength afforded by slag cement in a mixture allows the use of a lower total cementitious materials (paste) content.

Brooks et al[46] found shrinkage between concrete containing only portland cement and concrete containing slag cement to be similar. In contrast, Lankard[47] reported that that concrete containing 30% slag cement showed less drying shrinkage than the 100% portland-cement mixture. This contrast appears to be supported by the report from Gesoğlu et al[43] that the shrinkage decreased with increased amounts of slag cement

There seem to be other conflicting reports that seem to suggest higher shrinkage with the use of slag. Khatri et al.[48] showed that the drying shrinkage values of all slag mixtures were higher than those for the mixes prepared from ordinary cement. Collins and Sanjayan[49] stated that alkali-activated slag concrete had higher drying shrinkage than plain concrete by 1.6 to 2.1 times. Similarly, Deshpande et al[50] studied a 30% volume replacement of Portland cement by slag cement without changing the water or aggregate content in concrete. An increased early age shrinkage was observed, although the ultimate shrinkage did not appear to be significantly affected.

2.4.3. Silica Fume

Silica fume is a by-product resulting from the reduction of high-purity quartz with coal or coke and wood chips in an electric arc furnace during the production of silicon metal or ferrosilicon alloys. ASTM C1240[51] is used to regulate the quality of silica fume. ACI Committee 211 [15] recommends a dosage between 5% and 15% but ACI Committee 209 [38] reports that shrinkage decreases with replacements less than 7.5%. Like slag, the high fineness silica fume can lead to an increase in water demand which will increase shrinkage[26]. However, ACI Committee 234 [17] cautioned that the influence of high-range water-reducing admixtures(HRWRA) seems to generate some confusion in the analysis of shrinkage data.

Khatri et al[48] reported an increase in early age shrinkage for concrete containing silica fume compared to the control mixture, but long-term shrinkage was less for the concrete using silica fume. Alsayed[52] monitored the drying shrinkage of silica fume concrete for three years subjected to controlled laboratory or hot-dry field conditions. The author similarly concluded that 10% silica fume greatly reduces the three-year drying shrinkage, the stress due to shrinkage strain, and the rate of first month drying shrinkage of concrete. In addition, adding 10% of cement weight as silica fume to a concrete mix greatly reduces the influence of curing conditions on the rate of drying shrinkage.

Furthermore, Silica fume concrete has been reported to have lower shrinkage than normal portland cement concrete with the same w/cm ratio but marginally lower binder content[53]. The lower drying shrinkage of silica fume concrete could be attributed to the lower w/cm ratio used in the study[48].

Rao[54] conducted a project on shrinkage behavior with silica fume replacement rates from 0% to 30% by weight. A constant w/cm ratio of 0.5 and cementitious material to sand ratio

of 1 to 3 by mass were used. The results indicated a significant increase in shrinkage with the use of silica fume, which further increased with the replacement dosage of silica fume. The author concluded that the addition of silica fume increases the content of C-S-H, which is an important factor causing shrinkage at an age of 28 days. Whiting et al. (2000) studied the effects of silica fume on drying shrinkage and the cracking tendency of concrete bridge decks. They recommended a silica fume dosage of less than 8% unless demanded by the situation but did not give clear justification.

The drying shrinkage of silica fume–incorporated concrete was 10% to 22% higher than that of ordinary Portland cement with the same w/cm ratio and same binder content for the short term[55].

2.4.4. Metakaolin

Metakaolin ($\text{Al}_2\text{O}_3:2\text{SiO}_2$) is a natural pozzolan produced by heating kaolin-containing clays over a temperature range of about 600 to 900 °C (1100 to 1650 °F) above which it recrystallizes, into mullite ($\text{Al}_6\text{Si}_2\text{O}_{13}$) or spinel (MgAl_2O_4) and amorphous silica[56].

Metakaolin has been marketed under the description of high reactivity metakaolin. Regardless, its use in concrete is regulated by ASTM C618[41] and its typical additions may range between 5% to 15%[26, 39]. Current literature show that higher replacements metakaolin seem to decrease shrinkage but its high level of fineness of could have an adverse effect on shrinkage.

Zhang and Malhotra[57] compared the drying shrinkage strain of a control mixture and concretes containing 10% replacement by silica fume and metakaolin. They reported a consistently better drying shrinkage strain over the entire period of drying. Similar observation was made by Brooks and Megat-Johari[58] in that the total shrinkage measured from 24 h was

reduced using metakaolin while drying shrinkage was significantly less for the metakaolin concretes than for the control concrete by about 50%.

Güneyisi and Mermerdaş[59] reported that when the shrinkage strain at 60 days of drying was considered, the metakaolin concrete had remarkably lower shrinkage strain in comparison to the plain concrete. It was pointed out that the higher the replacement of metakaolin, the higher the reduction in the shrinkage, irrespective of w/cm ratio. In Hassan et al[60], drying shrinkage (after 400 days) decreased from 8% to 22% when metakaolin content increased from 0% to 11%. Lu[61] confirmed that the effect of metakaolin on the drying shrinkage of cement pastes is closely related to the metakaolin content and maturity of the pastes.

2.4.5. Zeolite

Zeolites are natural volcanic or synthesized aluminosilicate minerals with typical microporous framework structure. Their utilization as pozzolans dates to ancient times when the mixture of zeolites containing tuff and lime was used as hydraulic binder[62]. But its application as a popular type of natural pozzolans in the manufacture of pozzolanic cements began from the first decades of the 20th century[63]. And as a natural pozzolan, its use in concrete is controlled by ASTM C618[41] and its dosage in concrete may range between 5% to 15% although there are reports of higher replacement. As far as drying shrinkage is concerned, it has been reported that concrete with cement replacement of 15% natural zeolite achieves a suitable drying shrinkage[64].

Najimi et al[29] reported that mixing a natural zeolite (clinoptilolite) into cement at 15 % and 30 % resulted in sharp declines of drying shrinkage of 16% and 36%, respectively, at the age of 90 days. Markiv et al[65] observed a similar result when replacing 10 % of cement with

clinoptilolite. Concrete samples containing the natural zeolite experienced 2.6-fold decreases in drying shrinkage compared to control specimens.

In contrast, Jana[66] observed that the drying shrinkage of natural zeolite mixtures at 10% and 20% Portland cement replacement levels were similar or slightly higher than that of the control mixture, whereas by replacing 30% of cement with zeolite, the drying shrinkage was about 20% more than the control mixture. Also, Kasai et al[67] examined drying shrinkage for the mortars blended with clinoptilolite or mordenite type zeolite. Both clinoptilolite and mordenite blended mortars experienced higher shrinkage than the control mortar and the mordenite blended mortar showed slightly greater shrinkage than clinoptilolite blended mortar.

2.4.6. Limestone

Limestone powder is crushed and ground from natural limestone and is mainly composed of skeletal fragment of organisms. As an ingredient in Portland cement, limestone is limited to a maximum of 5% but as a cement replacement in blended cement, it is permitted to go up to 15% [68]. In terms of shrinkage, cements containing limestone may exhibit similar or lower drying shrinkage depending on the fineness [27].

Alunno-Rossetti and Curcio [69] measured the shrinkage of two series of concrete mixtures produced using the four cements (eight mixes in all). The results showed that the rate of shrinkage and total amount of drying shrinkage at 1 year was essentially the same for comparable concrete mixes produced with Portland Cement and Portland Limestone Cement from the same plant. But Rezvani et al [28] reported that concretes containing limestone exhibited a lower or equal drying shrinkage compared to reference samples made of comparable compressive strength.

Dhir et al[70] also reported reduced shrinkage in concretes produced with cements containing up to 45% ground limestone (blended not interground). Similarly, Shrinkage data from Bucher et al[27] on three cements produced with 0%, 5% and 10% limestone, showed that the amount of unrestrained drying shrinkage decreased with increasing limestone content.

In terms of restrained shrinkage, Bentz et al[71] compared concretes made with cements without limestone to cement made with 10% limestone at three different fineness levels. Results indicated that restrained shrinkage was similar to the control for the finest limestone and somewhat lower for the concrete with the coarsest limestone.

2.5. Shrinkage Test Methodologies

Free shrinkage is the term associated with the method of test used to evaluate the shrinkage of unrestrained concrete specimens in a controlled environment. The shrinkage strain, normally the longitudinal strain, is measured at regular intervals. Restrained shrinkage on the other hand, is based on test methods used to evaluate the cracking tendency of concrete. The most common restrained shrinkage test involves a concrete ring that is cast on the outside of a restraining steel ring. The stresses due to shrinkage and the age of specimen when the first crack appears are monitored using strain gauges mounted to the steel.

Free and restrained shrinkage tests are typically performed simultaneously because free shrinkage test does not, by itself, evaluate the cracking tendency of concrete. Mokarem, Weyers, and Lane[72] reported that the potential for cracking could be minimized by limiting the unrestrained shrinkage of concrete. They stated that, length change should be limited to 0.0400 percent ($400 \mu\epsilon$) at 28 days and 0.0500 percent ($500 \mu\epsilon$) at 90 days to reduce the probability of cracking due to drying shrinkage. Thus, although the free shrinkage test does not directly evaluate cracking tendency, it has the potential to be used in a performance-based specification

for restrained concrete systems like bridge decks. A summary of the various shrinkage test methodologies is provided in Table 2.2.

According to ASTM C1581[73], monitoring the strain gages soon after casting provides information on the internal deformations caused by autogenous shrinkage and heat of hydration.

Table 2.2. Summary of Shrinkage Testing Methodology[74]

Test Method	Brief Description	Advantages	Disadvantages
Prism test in free condition	ASTM C 157	Most commonly used; easy to conduct	Unable to correlate lab test to realistic condition because shrinkage is maybe restrained in some way. Also strain capacity is hardly to accurately detect concrete cracking process.
Prism test in restrained condition	ASTM C 341	Uniaxial stress testing; possibility of using large size aggregate; easy to use portable data logger to detect cracking by an abrupt change of voltage	Not easy to set up equipment
Plate test	Cracking length and depth can be measured and so does weighted value for cracking potential comparisons	Can be used to determine the cracking potential due to drying shrinkage; easy to conduct.	Very hard to be consistent because it can provide a biaxial restraint but highly depended on the geometry and boundary conditions
Ring test in free condition	Geometry can be the same as restrained condition for comparisons	High and nearly constant restraint; applicable on both mortar and concrete test; lower effects of geometry and boundary conditions due to symmetric geometry	Hard to set up; depends on geometry and boundary conditions when investigating cracking potential
Ring test in restrained condition	ASTM C 1581, associated with fracture mechanics theory or mechanism of concrete specimens	Same as ring test in free condition; axisymmetric nature of geometry eliminates the need to consider tractions on the inner surface of steel ring; effect of creep has been taken into account when collecting strains results from strain gages	Hard to set up; level of restraint of concrete cause non-consistent of cracking potential; linear elasticity may not be accurate for such a non-linear relationship for cracking potential estimation

3. EXPERIMENTAL PROGRAM

3.1. Materials

3.1.1. Cements

To study the influence of cement properties on drying shrinkage, 5 different cements (See Table 3.1) were used. The equivalent alkali ($\text{Na}_2\text{O}_{\text{Eq.}}$) contents were 0.55%, 0.71%, 1.01% (TI/II, HTCA, HA respectively) whereas the Tricalcium Aluminate (C_3A) contents were 6%, 9%, 11% (TI/II, HA, HTCA respectively). The four cements had Blaine Fineness 393, 368, 386, 317 (m^2/kg) in order to investigate the role of cement fineness on drying shrinkage. Also, the effect of limestone on drying shrinkage was investigated at 9% cement replacement (TIL) compared to the other cements (TI/II, HTCA, HA) which had up to 5% limestone content.

Table 3.1. Chemical and Physical Properties of Cement Used

Item	TI/II*	HTCA**	HA*	CG*	TIL***
SiO ₂ (%)	20.0	-	19.0	21.07	18.6
Al ₂ O ₃ (%)	4.6	-	4.88	4.25	3.7
Fe ₂ O ₃ (%)	3.2	-	2.31	3.13	3.4
CaO (%)	64.3	-	63.15	65	61.5
MgO (%)	2.3	1.2	2.97	0.97	4.3
SO ₃ (%)	3.3	3.3	3.22	2.88	3.3
Loss on Ignition (%)	3.1	2.4	2.41	0.75	4.6
Insoluble Residue (%)	0.42	0.52	0.35	0.21	-
CO ₂ (%)	1.8	1.2	1.38	-	-
CaCO ₃ in Limestone (%)	89	80.7	95.8	-	-
C ₃ S (%)	61	59	60.96	61.62	55
C ₂ S (%)	9	14	8.28	12.44	11
C ₃ A (%)	6	11	8.99	8.83	4
C ₄ AF (%)	10	6	7	9.3	10
Na ₂ O Eq. (%)	0.55	0.71	1.01	0.52	0.45
Limestone (%)	4.6	3.1	3.27	-	9
Blaine Fineness (m^2/kg)	393	368	386	316.94	497
*Limits Specified in ASTM C150[75]					
**Limits Specified in ASTM C-114[76]					
***Limits Specified in ASTM C595[41]					

3.1.2. SCMs

In this research five different types of SCMs were used based on their commercial availability and potential drying shrinkage properties. Table 3.2 and 3.3 shows the chemical and physical composition of the SCMs. The main components of a natural zeolite are identified as clinoptilolite and opal CT, accompanied by other components consisting of Smectite/illite, Plagioclase, Quartz and K-feldspar. Typically, the level of pozzolanic activity of natural zeolite is characterized by the amounts of active phases, i.e. clinoptilolite and opal CT[29]. Therefore, a finely grounded natural zeolite containing 97% on clinoptilolite alone, was used to ensure higher pozzolanic reaction with cement.

Table 3.2. Chemical and Physical Properties of FA, MK, Z

Item	Class C/F Fly Ash (FA)*	Metakaolin (MK)*	Zeolite (Z)*
SiO ₂ (%)	50.81	51-53	66.7
Al ₂ O ₃ (%)	15.48	42-44	11.48
Fe ₂ O ₃ (%)	5.52	<2.2	0.9
Sum (SiO ₂ -Al ₂ O ₃ -Fe ₂ O ₃)	71.81	<99.2	79.08
SO ₃ (%)	0.75	<3.0	-
CaO (%)	13.77	<0.20	1.33
MgO (%)	4.40	<0.10	0.27
Na ₂ O (%)	3.91	<0.05	1.8
K ₂ O (%)	2.23	<0.40	3.42
Loss on Ignition (%)	0.15	<0.50	-
Na ₂ O Eq. (%)	1.9	<0.31	4.05
Fineness,	22.30**	15***	40***
* Limits Specified in ASTM C618[41]			
**% retained on 45-µm sieve			
***BET			

Table 3.3. Chemical and Physical Properties of Slag Used

Item	Slag*
Sulfide Sulfur (S), (%)	0.82
SO ₃ (%)	0.75
Chlorides (%)	0.32
Na ₂ O Eq. (%)	0.83
Blaine Fineness(m ² /kg)	474
*Limits Specified in ASTM C989[45]	

3.1.3. Aggregates

For this experimental program, a locally sourced 3/8" Pea Gravel was used as coarse aggregates while river sand was used as the fine aggregate. The size of coarse aggregates was limited by ASTM C1581[73] which requires the maximum nominal size of the coarse aggregate to be 13 mm [0.5 in.] or less. A combined gradation curve is presented in Figure 3.1

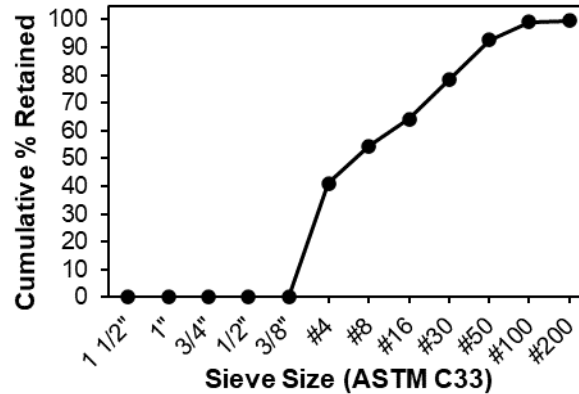


Figure 3.1. Combined Gradation Curve

3.2. Mixture Proportion

A key aspect of the research was to study the effect of SCMs at moderate to high dosages while keeping paste content constant. Recommended dosages were based on guidance from ACI Committee 211[39] and other literature but ultimately, trial mixes were used to determine the final SCM dosages with the aim of maintaining a reasonable slump across mixes. High dosage fly ash concrete tends to generate unusually high slumps even without water reducer in the mix.

It is impossible to hold the absolute volume relationship and total weight of the cementitious material constant across mixes with different pozzolanic materials because of their different specific gravities. In this experiment, the absolute volume relationship of the cementitious materials was prioritized over total weight since the paste content (volume) was held constant with equal water content for all mixes. This resulted in the water/binder ratio

remaining between 0.43 and 0.48 as paste content (without air) was kept at $26 \pm 0.5\%$ across mixes.

Table 3.4 shows a matrix of the different cementitious material combinations and the final SCM dosages used. The experimental program for this study allowed for 4 plain cement mixes, 6 binary cementitious blends and 5 ternary cementitious blends. The concrete batch proportions for the 15 mixes is presented in Table 3.5. The air content ranged between 5 – 8% with 3-6 inch slump.

Table 3.4. Cementitious Combinations and SCM Dosages

CEMENT \ SCM	No SCMs	FA (35%)	SF (10%)	S (50%)	MK (10%)	Z (15%)
TI/II -CONTROL	X	X	X	X	X	X
HTCA (high C3A)	X					
HA (high Alkali)	X					
CG (Coarse Grind)	X					
TIL (9% Limestone)	X	X	X	X	X	X

Table 3.5. Mix Proportions

Mix Code		Cement (lbs)	SCM (lbs)	Coarse Agg. (lbs)	Fine Agg. (lbs)	W/R (oz)	AE-90 (oz)	Water (gal)
Plain	TI/II	587	-	1584	1467	36	2	30
	HTCA	587	-	1584	1467	55	2	30
	HA	587	-	1584	1467	55	2	30
	CG	587	-	1584	1467	32	2	30
Binary	TI/II-SF	508	56	1584	1467	56	3	30
	TI/II-FA	354	191	1584	1467	-	2	30
	TI/II-MK	517	57	1584	1467	56	3	30
	TI/II-S	285	285	1584	1467	33	2	30
	TI/II-Z	453	80	1584	1467	55	3	30
	TIL	585	-	1584	1467	36	2	30
Ternary	TIL-SF	506	56	1584	1467	56	3	30
	TIL-FA	354	190	1584	1467	-	2	30
	TIL-S	284	284	1584	1467	33	2	30
	TIL-MK	516	57	1584	1467	57	3	30
	TIL-Z	452	80	1584	1467	64	3	30

3.3. Specimen Preparation and Test Methods

Concrete materials were batched and mixed in accordance with ASTM 192[77] to prepare;

- 4x8 cylinder concrete specimen for semi-adiabatic calorimetry tests[78]
- 3 x 3 x 11 ¼ in prisms for free drying shrinkage testing, ASTM C157[79] whereas concrete
- Ring specimens for restrained drying shrinkage testing, ASTM C1581[73].

The curing and testing regimen of the concrete specimens was in accordance with the ASTM test methods except for the free shrinkage specimens. The free shrinkage prisms were kept in the mold in a moist room for 24±1hrs and later stored in lime saturated water for 2 days after demolding. Initial comparator reading was taken after curing and further readings were taken at 4, 11, 25 and 53days of drying. Both the free and restrained drying shrinkage specimens were stored in the same drying environment with constant air temperature of $73.5 \pm 3.5^{\circ}\text{F}$ and relative humidity of $50 \pm 4\%$.

4. RESULTS AND DISCUSSIONS

4.1. Influence of Paste Quality on Free Water

In this study, the strain measurements for the first 24hrs after casting was juxtaposed against the hydration curve from the semi-adiabatic calorimetry results (See Figure 4.1). Results showed some potential relationships between the strain curve and the hydration curve, as the temperature due to heat of hydration appeared with observed correlate to strains.

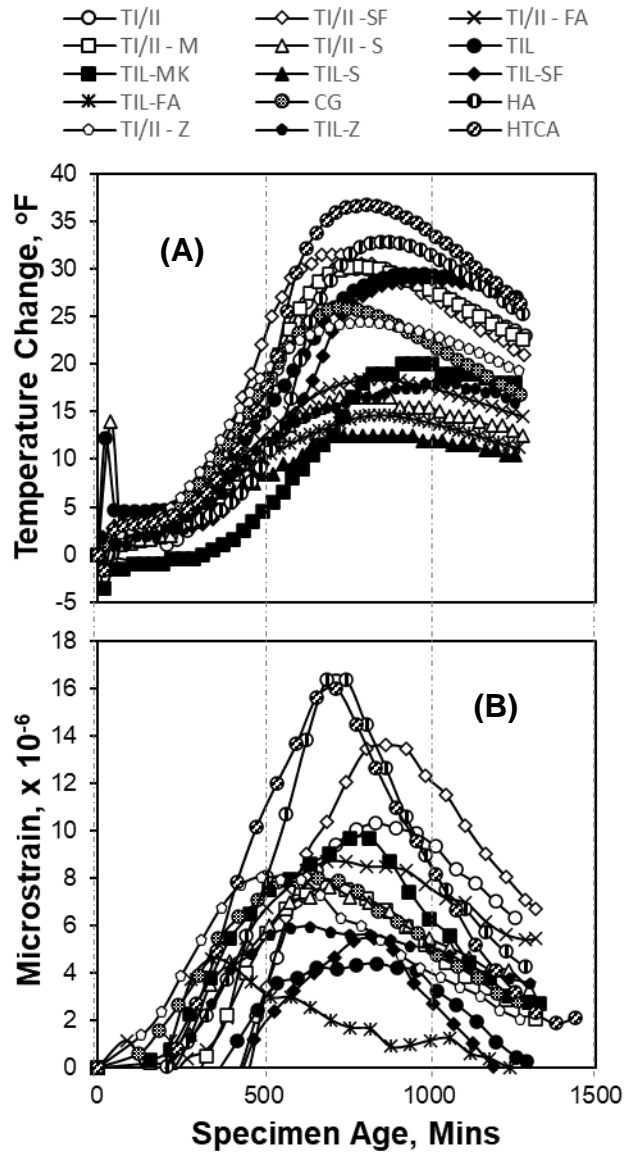


Figure 4.1. Hydration Curve (A) and Internal Deformations (B)

It was observed that cementitious materials with high peak temperature during hydration process tend to produce high peak strains and contribute to high early age shrinkage. This was particularly evident in cement only mixes (TI/II, HTCA, HA, CG) as shown in Figure 4.2 but was less apparent in mixes with binary and ternary cementitious blends.

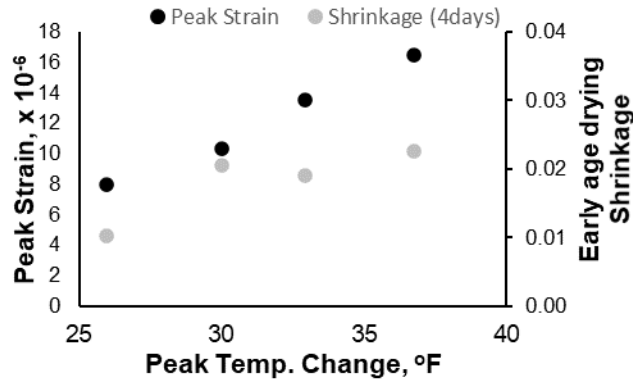


Figure 4.2. Relationship Between Early Drying Shrinkage and Initial Deformities Due to Heat of Hydration and Autogenous Shrinkage

4.2. Effect of Paste Quality on Drying Shrinkage

As it has been established that free water plays a role within the concrete microstructure at the early ages in dictating the extent of drying shrinkage; it is important to consider the action of heat of hydration and self-desiccation on free water at early ages. Observed results seem to indicate that the quality of the paste material in concrete appears to impact how the self-desiccation process and heat of hydration affects free water and later drying shrinkage. For instance the choice of binary or ternary blend can provide some microstructure benefits[14].

Cementitious blends with fly ash have been reported to provide reduced heat of hydration[80-84], reportedly because of a slower reaction rate. This means that free water is available during the initial stages of drying to offset early-age drying shrinkage. Zeolite handles free water differently within the concrete microstructure through an internal curing process[29]. During mixing and initial hydration of a cementitious blend with zeolite, when the heat of

hydration and self-desiccation process are its peak; the zeolite particles protect free water from rapid dissipation by absorbing it and releasing it later back into the concrete microstructure to counteract drying shrinkage. In Figure 4.3 we see that among the binary and ternary cementitious blends, mixtures that contained Fly ash (TI/II-FA and TIL-FA) and Zeolite (TI/II-Z and TIL-Z) showed lower shrinkage during early ages.

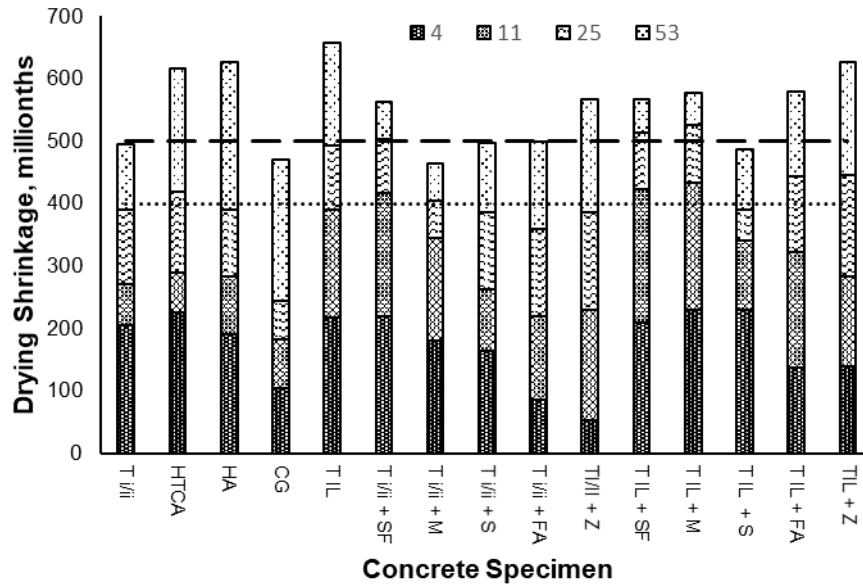


Figure 4.3. Free Shrinkage Over 53 Day Drying Period

In contrast, silica fume and metakaolin are known to be highly reactive pozzolans with high water demand due to their fine particle size. Their aggressive use of free water aggravating self-desiccation can adversely exacerbate their early age drying shrinkage behavior.

Cementitious blends with metakaolin (TI/II-M, TIL-M) and silica fume (TI/II-SF, TIL-SF) showed higher shrinkage on day 4 and 11 of drying compared to the control mix (TI/II).

As a guide, Mokarem et al[72] recommended that length change should be limited to 400millionth at 28 days and 500 millionth at 90 days to reduce the probability of cracking due to drying shrinkage. It is worth mentioning that only 3 out of the 15 mixes (CG, TI/II-M and TIL-S) were within the proposed limit of 400 and 500 millionth at 56days.

Interestingly, no apparent correlation was observed between the measured shrinkage (unrestrained) at 56 days and the age at which restrained shrinkage cracking was recorded for all the specimens. However, there appears to be some relationship between the level of shrinkage at the early ages (7 and 14 days) and cracking potential of concrete specimens.

Specimens (HTCA,HA, TI/II-SF, TI/II-M, TI/II-S, TIL-SF, TIL-M, TIL-S) with high early age drying shrinkage seems to have cracked earlier in comparison to the control mix TI/II whereas specimens (CG, TI/II-FA, TI/II-Z, TIL-FA, TIL-Z) with equal or lower early age drying shrinkage seem to have performed better than the control (See Figure 4.4). Mixture TIL showed an interesting deviation from this relationship discussed in section 3.3 below.

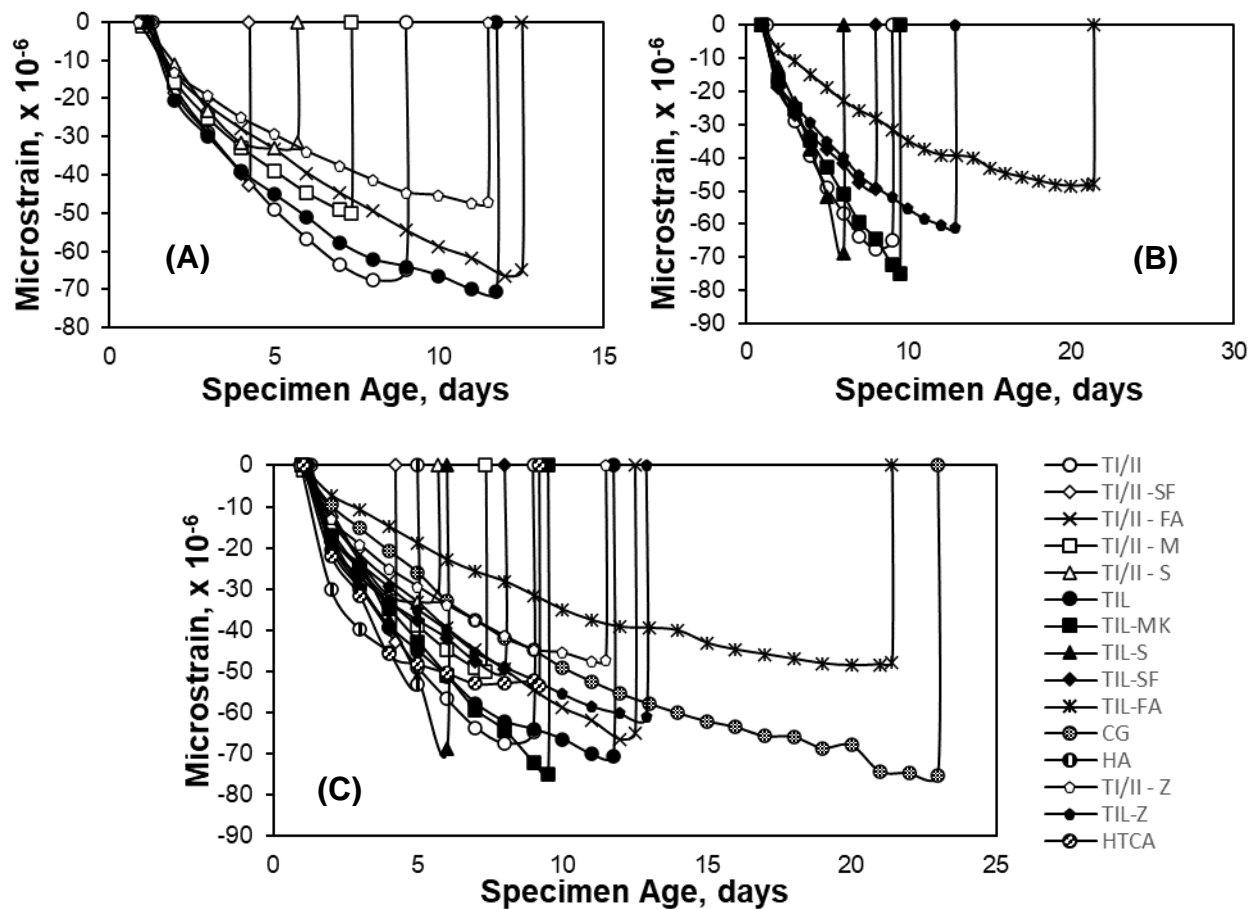


Figure 4.4. Restrained Shrinkage: Binary Mixes and Control(A), Ternary Mixes and Control (B) and All Mixes (C)

4.3. Influence of Limestone in Drying Shrinkage Durability

One of the objectives of this study was to understand how the presence of limestone affects the paste quality in terms of drying shrinkage cracking resistance. As reported by Bucher et al[27], cements containing limestone may exhibit similar or lower drying shrinkage depending on the fineness. TIL had a Blaine fineness of $497\text{m}^2/\text{kg}$ compared to the control cement (TI/II) which was $393\text{m}^2/\text{kg}$. Therefore, the high level of drying shrinkage exhibited by TIL mix in comparison to TI/II was expected (See Figure 4) but it is somewhat surprising that TIL mix had better drying shrinkage cracking resistance than the TI/II mix (See Figure 5).

In Figure 4.4(A) both TI/II and TIL had similar tensile strains until day 5, after which a significant disparity was noticed. Also, since TIL eventually cracked at a higher strain than TI/II, it can be argued that the presence of limestone may have improved the tensile capacity of TIL, hence its prolonged age of cracking despite the high shrinkage. Similar improvement in drying shrinkage cracking resistance was observed for ternary cementitious blends that had limestone (See Figure 4.5).

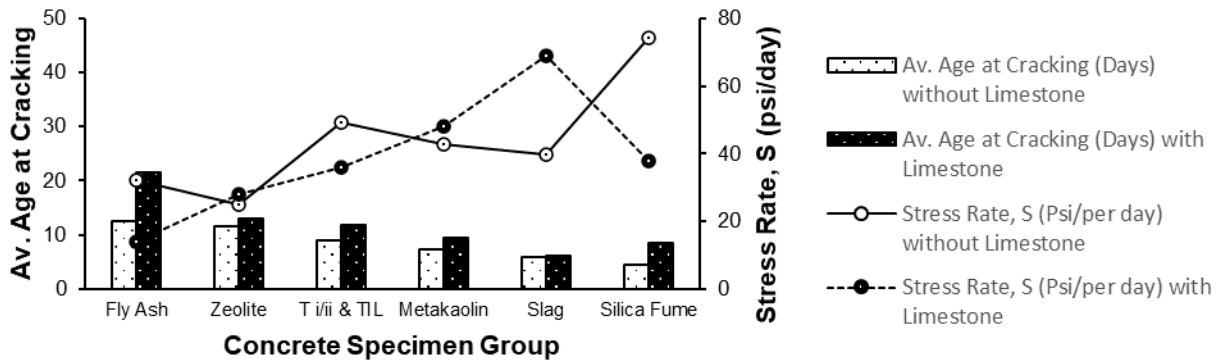


Figure 4.5. Effect of Limestone on Drying Shrinkage Cracking

Further statistical validation through a randomized block design (Table 4.1) was used to ascertain, if indeed, the presence of limestone in the cementitious blends impacted the drying

shrinkage cracking durability as opposed to the uniqueness of the SCM. The average age at cracking was calculated from three ring test samples for each mix and analyzed for variance using Minitab at a 95% level of confidence (See ANOVA output in Table 4.2).

Table 4.1. Randomized Block Design for Effect of Limestone Against SCMs

BLOCK (Effect of SCM)	TREATMENTS (Effect of Limestone)	
	Without Limestone	With Limestone
Silica fume Mix	TI/II-SF	TIL-SF
Metakaolin Mix	TI/II-MK	TIL-MK
Slag Mix	TI/II-S	TIL-S
Fly ash Mix	TI/II-FA	TIL-FA
Zeolite Mix	TI/II-Z	TIL-Z
No SCM Mix	TI/II	TIL

Table 4.2. ANOVA Table for Effect of Limestone Against SCMs

Source	DF	Adj SS	Adj MS	F-Value	P-Value
Block	5	171.58	34.316	7.40	0.023
Treatment	1	31.01	31.014	6.69	0.049
Error	5	23.17	4.635		
Total	11	225.77			

A P-value of 0.023 for the block and 0.049 for the treatments are lower than α value of 0.05 and therefore, statistically significant. This means that whereas limestone has some effect, its overall effect on the cementitious blend is limited by other additives and/or SCMs in the blend. In figure 4.5 , notice that the average age at cracking improved substantially with the Type IL blend with fly ash, compared to TI/II-FA, but the slag and zeolite blends barely improved with their respective Type IL blends. It is possible that the variations in the amount of limestone due to differing SCM percentages within the cementitious blends may have had some effect, but this was not investigated.

4.4. Role of Cement Chemical and Physical Property on Drying Shrinkage

It was noted that cementitious fineness and chemical composition (mainly C_3A and Na_2O_{Eq}) may have had some influence on drying shrinkage at different stages of the various concrete specimens. However, the extent of influence was much clearer in the plain cement mixes (TI/II, HTCA, HA, CG) as opposed to the binary and ternary cementitious blends.

4.4.1. Early Stages

At the early stages of the concrete hydration, the C_3A and Na_2O_{Eq} content seem to have had some effect on the heat of hydration which in turn may have affected the availability of free water (discussed earlier in section 3.3). In Figure 4.6, notice that cements with higher contents of C_3A and/or Na_2O_{Eq} produced higher peak temperatures with lower Blaine fineness than the control cement. It is worth mentioning that this does not negate the influence of cement fineness on heat of hydration as widely known, but rather explains the nature of its influence.

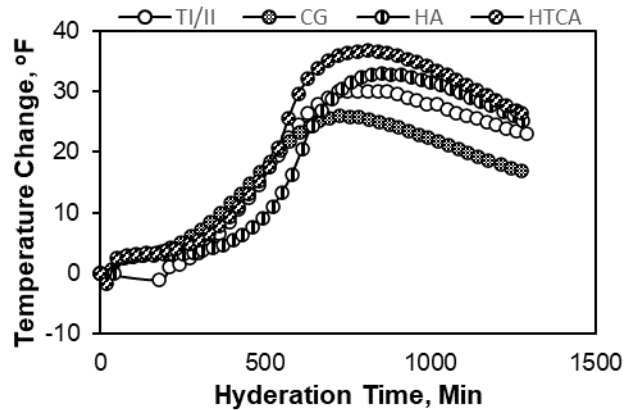


Figure 4.6. Semi-Adiabatic Calorimetry for Plain Mixes

An increase in alkali content has been known to accelerate hydration of cement and lead to greater heat evolution[23, 85-87]. This is because the increased alkali cations in the liquid phase of fresh cementitious mixture accelerates the hydration of C_3A by depressing the Ca^{2+} cations released from gypsum thereby decreasing its effectiveness[88]. The C_3A compound itself

is considered the most reactive among the cement compounds and liberates a large amount of heat during the first few hours of hydration[26, 89].

Therefore, it is likely that high cement fineness intensifies the effect of C_3A and Na_2O_{Eq} . In Figure 4.6, the CG mix had a lower peak temperature despite having a higher C_3A than the TI/II mix with similar Na_2O_{Eq} content.

4.4.2. Later Stages

Generally, at the later stages of the concrete specimen, the effect of cement chemical composition on drying shrinkage reduces as cement hydration slows. However, the mixture with high Na_2O_{Eq} content may result in reduced tensile strength which can adversely affect the drying shrinkage cracking resistance. Smaoui et al[90] reported a 5 -16% reduction in the splitting tensile strength over a 180-day testing period in a high-alkali concrete mixture (1.25% Na_2O_{Eq}) when compared to a control mixture (0.6% Na_2O_{Eq}). They also noted that the main reduction was observed at 3days, while the difference remained about the same afterwards. This may explain why the HA mix cracked earlier than the control mix despite comparatively similar tensile stresses (Figure 4.7B).

Nevertheless, cement fineness plays the most critical role during the later stages of drying. Coarser cement particles tend to offer some form of internal restraint against the shrinkage tensile stresses, thereby helping to improve shrinkage and potential shrinkage cracking[18, 50, 91, 92]. It can be seen in Figure 4.7(A) that CG showed considerably lower shrinkage compared to the finer cements (TI/II, HTCA, HA) and produced lower tensile stresses which led to better cracking resistance in Figure 4.7(B).

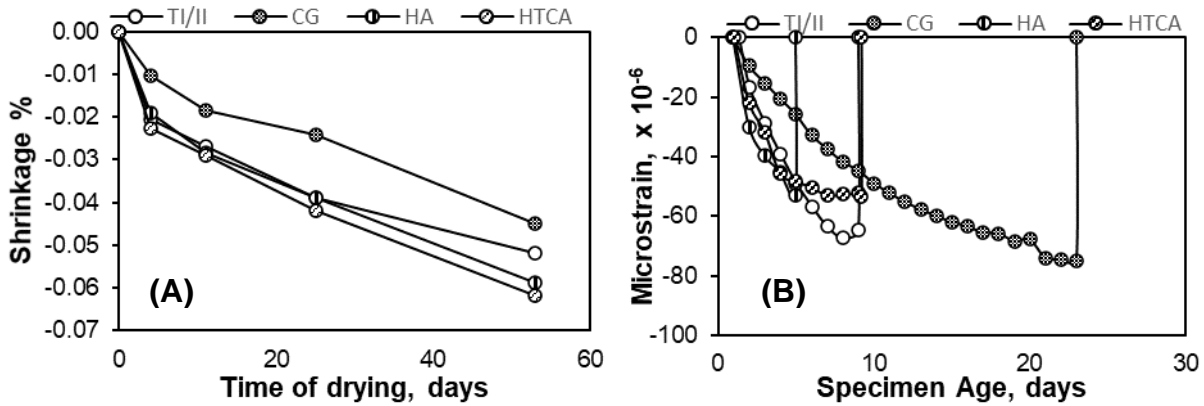


Figure 4.7. Plain Mixes: Free Shrinkage (A) and Retained Shrinkage (B)

4.5. Future Research and Significance

This study has showed how the constituents of cement paste might have influenced the drying shrinkage and related crack resistance of various concrete mixes. It is possible that decisions on the quality of cementitious paste materials alone could have some significant effect on the drying shrinkage and cracking potential of concrete.

The discovery of microcracks by researchers[93-95] in cement paste as a result of self-restraint within the cement paste may have created an avenue to study the relationship between the shrinkage in cement paste and bulk concrete. Basically, as neat cement paste element dries, a moisture gradient may develop due to uneven drying within the element – thus, the portion of the element with less moisture will shrink more than the other portion with more moist, hence a self-restraining effect is created. This self-restraint within a neat cement paste element causes tensile stresses, oriented parallel to the drying surface to occur [96, 97]. And when tensile these tensile stresses become greater than the tensile capacity of the cement paste element microcracking develops mainly perpendicular to the drying surface.

Early results from Sharma et al[92] showed a potential correlation between the drying shrinkage induced microcracks on a cement paste element and the stress rate due to shrinkage of

an externally restrained concrete element, however, further validation may be required with a much broader range of cementitious materials and sample size. It is expected that further studies into cement paste microcracking may offer some insights on the relative likelihood of early-age cracking of concrete mixtures and possibly assist in the selection of binder materials which are more resilient to drying shrinkage cracking.

5. CONCLUSION

In this research, the influence of paste quality on drying shrinkage and drying shrinkage cracking was studied in terms cementitious paste chemical composition, fineness, SCM and mineral admixture addition in binary and ternary blends. The following observations were made from the study.

- The early deformations caused by heat of hydration and autogenous shrinkage was recorded and modeled against the calorimetry profile of concrete specimens. A significant correlation was found between the peak strain from early deformations and peak temperature change.
- The constituents of the paste material influenced the action of heat of hydration and self-desiccation on free water at early ages which in turn affects shrinkage at later stages of drying. Cementitious blends with Fly ash and Zeolite were found to have mechanisms that control these two processes, hence showed better shrinkage resistance.
- Cements with high C_3A and Na_2O_{Eq} produced a large amount of heat during the early stages of hydration but their net effect on drying shrinkage was limited by the fineness of the cement. Nonetheless, high Na_2O_{Eq} content can affect the tensile capacity of the concrete and lead to premature drying shrinkage cracking.
- Cementitious blends with high limestone dosage showed high drying shrinkage but was counteracted by improved tensile capacity. Results from statistical randomized block design showed that whereas limestone has some effect on the drying shrinkage cracking resistance of concrete, its overall effect on the cementitious blend is limited by the nature and the property of the additives and/or SCM in the blend.

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