Fourth International Conference on Sustainable Construction Materials and Technologies
http://www.claisse.info/Proceedings.htm

SCMT4
Las Vegas, USA, August 7-11, 2016

Strength Development and Shrinkage Cracking Resistance of High Alite Cement Concrete with Fly Ash

Shingo Miyazawa[1a,] Takashi Yokomuro[1b], Etsuo Sakai[2], Nobukazu Nito[3], and Hiroshi Hirao[4]

[1]Ashikaga Institute of Technology, 268-1 Ohmae, Ashigaka, 326-8558, JAPAN
[1a]Email: <smiyazaw@ashitech.ac.jp>, [1b]Email:<yokomuro@ashitech.ac.jp>

[2]Tokyo Institute of Technology, 2-12-1 O-okayama, Meguro-ku, Tokyo, 152-8552 JAPAN
Email:<esakai@ceram.titech.ac.jp>

[3]DC Co., Ltd., 8 Higashida, Kawasaki-ku, Kawasaki, 210-0005, JAPAN
Email:<nito_nobukazu@dccorp.jp>

[4]Taiheiyo Cement Corporation, 2-4-2 Osaku, Sakura, 285-8655 JAPAN
Email:<Hirao@taiheiyo-cement.co.jp>.

ABSTRACT

In order to reduce CO_2 emission in the process of cement production, it is efficient to use blended cement, such as blast-furnace slag cement and fly ash (FA) cement, widely for concrete structures instead of ordinary Portland cement. However, in Japan, FA cement has rarely been used for concrete structures, since strength development of concrete with FA cement is relatively slow at early ages. The purpose of this study is to propose FA cement which can be widely used for general concrete structures. In this study, cement samples with alite contents increased up to 72 %, by Bogue equation, were manufactured on trial at an actual cement plant and strength development of FA cement concrete using the high alite cement was experimentally investigated. Tendency of shrinkage cracking in concrete under drying condition was also investigated with uniaxial restraint tests, in which restraint stress and drying period until cracking were observed. It was shown by the test results that early-age strength development of FA cement concrete could be improved by using the high alite cement, especially at low temperatures. It was suggested that shrinkage cracking resistance of concrete with the proposed FA cement could be higher than with ordinary Portland cement.

INTRODUCTION

During the production of ordinary Portland cement (OPC), large amount of CO_2, which is thought to be a main cause of global warming, is emitted into the air, since limestone ($CaCO_3$) decarbonates through the process of clinker production. On the other hand, blended cements such as blast-furnace slag (BFS) cement and fly ash (FA) cement give off less amount of CO_2 than OPC. Therefore, blended cement has been widely used from environmental point of view in many countries.

In Japan, conventional blended cements are generally produced by blending FA or BFS with OPC, whose C_3S content ranges typically from 55 % to 60 %. Although BFS cement has often been applied to civil engineering structures, FA cement has rarely been used for concrete structures except for dam concrete. Furthermore, the consumption of FA cement accounts for no more than 0.1 % of the total cement consumption, while the consumption of OPC accounts for about 70 %. It should be also pointed out that the addition of BFS or FA at concrete mixing plants is very rare in this country because of insufficient facilities in concrete mixing plants and delivery systems. Therefore, it is expected that FA cement is extensively used for concrete structures to reduce CO_2 emission in cement industry.

In order to increase application of FA cement to general concrete structures, it is desirable to improve early-age strength development of concrete. Several measures to accelerate strength development of FA cement have been suggested in published papers. It was reported from experimental study that fine particles in FA made higher contribution toward strength development than coarse particles [Erdoğdu and Türker 1998]. It was also reported that strength development of FA cement mortar was accelerated at early ages by replacing a part of FA by limestone powder [Weerdt *et al*. 2011].

There are some researches on the effect of mineral composition of clinker on strength development of blended cement. It was reported that early-age compressive strength of FA cement concrete could be improved by using high-early strength Portland cement with C_3S content of 65 % [Morioka *et al*. 2002]. It was also reported that clinkers with C_3S content up to 73 % prepared in a laboratory could be effectively used for FA cement with a high replacement ratio [Hou *et al*. 2007]. However, these studies have not been applied to practical uses. The authors reported that clinkers with very high C_3S content up to 71 %, by Bogue equation, were manufactured at an actual cement plant [Nito *et al*. 2014], and that strength development of BFS cement could be improved by using the high C_3S clinker [Miyazawa *et al*. 2014].

The purpose of this study is to propose FA cement which can be widely used not only for civil engineering concrete structures but also reinforced concrete buildings. In order to improve strength development of FA cement at early ages, some samples of clinker with very high C_3S content about 70 %, by Bogue equation, were manufactured at an actual cement plant. Replacement ratio of FA was taken to be 15 % or 20 %, and the effect of the contents free CaO and SO_3 of FA cement were investigated. Strength development of concrete with FA cement using the high C_3S clinkers was experimentally investigated, taking account of the effect of curing temperature. Tendency of cracking of concrete due to dying shrinkage was also investigated with uniaxial restraint tests.

EXPERIMENTAL PROCEDURES

Materials

The samples of high alite cement A_1 and A_2 were produced at an actual cement plant, which has been used for production of OPC for commercial use. Chemical compositions and mineral composition of materials used in the experiments are shown in Table 1 and Table 2, in which mineral compositions of the cement samples are calculated with Bogue equations. Samples of cement A_1 and A_2, which were to be used as base cement for FA cement, have larger C_3S content and larger free CaO content than OPC. Blaine fineness of the samples of cement A_1 and A_2 is higher than OPC.

In general, OPC with free CaO content of less than 1 % is used as base cement for FA cement. However, the optimum free CaO content as the base cement for FA cement has not been investigated. In this study, samples of cement A_1 and A_2 with relatively higher free CaO content were prepared. Cement A_1 with free CaO content of 1.8 % was used in Series I and Series II, and cement A_2 with free CaO content of 3.0 % was used in series III. For the FA cement samples in Series II and III, anhydrous gypsum was added at mixing concrete in amount such that SO_3 content of FA cement was 2.9 % and 3.4 %, respectively. In Series I, on the other hand, additional anhydrous gypsum was not added at mixing concrete.

Samples of FA (FA_1 and FA_2) used in this study are by-products from two different coal-fired power plants. Their properties, which are shown in Table 1 and Table 3, agree with the requirements of Class II fly ash specified by JIS A 6201 [Japanese Standards Association 2012]. Class II fly ash, which corresponds to

Table 1. Chemical Composition of Cements and Fly Ash

Cement	Series	Chemical composition of cement (%)									
		ig-loss	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	Na_2O	K_2O	f.CaO
A_1	I, II	0.90	19.8	5.39	2.87	65.6	1.64	2.43	0.31	0.35	1.8
A_2	III	1.29	18.8	5.30	2.77	64.8	2.12	3.36	0.41	0.40	3.0
N_1	I, II	0.87	21.0	5.86	3.08	63.2	2.08	2.39	0.45	0.38	0.8
N_2	III	1.09	20.7	5.33	3.11	64.0	2.01	2.24	0.44	0.34	0.6
FA_1	I, II	2.04	59.2	25.4	5.31	2.86	1.21	0.46	0.62	1.40	-
FA_2	III	1.10	58.5	27.6	4.60	3.10	1.1	0.20	1.10	0.85	-
BB	I	1.26	26.0	9.26	1.77	53.6	3.84	2.24	0.33	0.29	-

Table 2. Physical Properties and Mineral composition of Cements

Cement	Series	Density (g/cm^3)	Blaine fineness (cm^2/g)	Mineral composition (%)			
				C_3S	C_2S	C_3A	C_4AF
A_1	I, II	3.15	4690	69	5	10	9
A_2	III	3.10	4820	72	0	9	8
N_1	I, II	3.16	3290	59*	14*	9*	10*
N_2	III	3.16	3340	57	16	9	10
BB	I	3.04	4080	-	-	-	-

*: minor additional constituents excluded

Table 3. Properties of Fly Ash (FA)

Item		FA_1	FA_2	JIS A 6201
SiO_2, %		59.2	58.5	≥45.0
Moisture content %		<0.5*	0.1*	≤1.0
Loss on ignition %		2.04	1.1	≤5.0
Density, g/cm^3		2.28	2.24*	≥1.95
Fineness				
Amount retained when wet-sieved on 45μm sieve, %		12*	9*	≤40
Blaine, cm^2/g		3510	3690	≥2500
Flow, percent of control, %		110+	112*	≥95
Strength activity index, %	7 days	72*	74	—
	28 days	85*	84	≥80
	91 days	103*	102	≥90

* *obtained from quality control data at the plant*

Class F fly ash specified by ASTM C618-05 [American Society of Testing and Materials 2005], is typical FA for concrete in Japan.

For comparison, conventional FA cement, in which 15 % of N_2 is replaced by FA_2, was used in series III. Blast-furnace slag Portland cement type B (BB), which is widely used in Japan, was also used in series I. The base cement for BB was OPC and the replacement ratio of BFS was about 40 %.

The samples of cement N_1 and N_2, which fall into a category of OPC, were used for making control specimens. In series I and II, the cement N_1 contains minor additional constituents up to 5 %, which is the maximum requirement specified by JIS R 5210 (Japanese Standards Association, 2009), and the mineral compositions in Table 2 are for the cement clinkers, excluding minor additional constituents. In Series III, the sample of cement N_2 does not contain minor additional constituents.

Fine aggregate and coarse aggregate used to make concrete specimens were river sand and crushed hard sandstone respectively, and the result of sieve analysis of them are shown in Table 4. Air-entraining and water-reducing agent (ad), whose main ingredients are lignin-sulfonic acid compound and polycarboxylic acid, was used for all the mixtures. In addition, in order to obtain the target air content of concrete, two types of air-entraining agents were used. Alkylcarboxylic acid type agent was used for concretes with FA and alkylether type agent was used for concrete without FA.

Mix proportion of concrete

The mix proportions of concrete prepared for the experiments are shown in Table 5. Water to cement ratio of concretes was fixed at 0.55. Slump was 19 ± 2.5 cm and air content was 5.0 ± 1.5 %. The content of air-entraining and water-reducing agent (ad) was taken to be 1.0 % for all the mix proportions.

Table 4. Results of Sieve Analysis of Aggregates

Sieve size (mm)		25	20	15	10	5	2.5	1.2	0.6	0.3	0.15
Cumulative percentage passing (%)	Coarse aggregate	100	96	74	23	0	0				
	Fine aggregate					100	89	63	39	19	6

Table 5. Mix Proportions of Concrete

Series	Cement	W/(C+FA)	s/a (%)	Unit content (kg/m^3)					Ad x(C+FA) %
				W	C	FA	S	G	
I	A_1-$FA_1$20(2.0)	0.55	46.0	180	262	65	802	945	1.0
	N_1			180	327	-	812	957	1.0
	BB			175	318	-	817	963	1.0
II	A_1-$FA_1$20(2.9)	0.55	46.0	180	262	65	802	945	1.0
	N_1			180	327	-	812	957	1.0
III	A_2-$FA_2$15(3.4)	0.55	46.0	180	278	49	802	946	1.0
	N_2-$FA_2$15(3.4)			175	271	48	814	960	1.0
	N_2			180	327	0	812	957	1.0

ad : air-entraining and water-reducing agent

Test methods. The specimens for measurements of compressive strength were cylinders with 100 mm in diameter and 200 mm in length. Strength development of concrete under standard curing was measured up to 91 days and early-age strength development of concrete under sealed curing was measured up to 7 days. In case of standard curing, concrete was casted at 20 ± 2 °C and the specimens were cured under water at 20 °C until measurements of compressive strength at the ages of 3, 7, 28 and 91 days. In case of sealed cuing, the top surface of specimens was sealed immediately after casting in order to prevent evaporation and the specimens were stored in rooms at 20 ± 1 °C and 10 ± 1 °C until measurements of compressive strength at the ages of 1, 2, 3, 5 and 7 days. Three specimens were prepared for each mixture and for each curing condition.

In order to study resistance to shrinkage cracking, restraint stress of concrete caused by drying shrinkage was measured by uniaxial restraint tests as shown in Figure 1. A deformed reinforcing bar with nominal diameter of 25 mm was embedded at the center of concrete beam specimens of 100 mm in width and depth and 1100 mm in length as a retraining body. Both the libs and lugs of the reinforcing bars were removed within the central portion with 300 mm in length and this portion was sealed with Teflon sheet with 0.2 mm in thickness in order to eliminate bond between reinforcing bar and concrete, as shown in Figure 1 (a). After casting, concrete specimens were cured in sealed condition until the age of 3 days and they were exposed to drying condition at 20 ± 1 °C and 60 ± 5 % R.H. as shown in Figure 1 (b). Restraint stress of concrete caused by drying shrinkage was obtained from the strain of the embedded reinforcing bar which was measured with electric wire strain gauges attached on two opposite surfaces of the central portion of the reinforcing bar. During the drying period, the time at cracking in the specimens was detected from a sudden decrease in strain of the reinforcing bar. Three specimens were prepared for each mixture.

RESULTS AND DISCUSSIONS

Environmental performance of cement

In the manufacture process of the samples of high alite cement A_1 and A_2 in the actual cement plant, the

(a) Apparatus for restraint stress test (b) Specimens stored in drying condition

Figure 1. Test for Restraint Stress of Concrete due to Drying shrinkage

kinds of raw materials for clinker including wastes and industrial by-products such as coal ash and construction generated soil were the same as those for OPC which had commercially been produced. From environmental point of view, the following points can be pointed out regarding to production of the clinker samples for A_1 and A_2.

(1) It can be said that the amount of limestone required for producing A_1 and A_2 clinkers are about the same as that for N_1 and N_2 clinkers, which can be clarified from the equivalent CaO contents in these clinkers (see Table 1). Therefore, CO_2 emission due to decarbonation of limestone in the production of A_1 and A_2 clinkers is equivalent to OPC clinker.

(2) Since the temperatures in the kiln during the calcination of A_1 and A_2 are about the same as N_1 and N_2, it can be said that CO_2 emissions caused by calcination energy of A_1 and A_2 are equivalent to that of OPC.
(3) The consumption of electric power in the processes of law material treatment and grinding is about the same among A_1, A_2, N_1 and N_2. The reason why specific surface areas of A_1 and A_2 are higher than N_1 and N_2 is that these clinkers contain larger amount of C_3S with high grindability.

Therefore, it can be said that CO_2 emission due to the production of FA cement using the high alite cement is about the same as that of conventional FA cement at the same replacement ratio of FA. This means that CO_2 emission of FA cement using the high alite cement is less than that of OPC without FA.

Compressive strength under water at 20 ºC. In a published paper [Miyazawa et al. 2015], properties of FA cement using high alite cement with alite content of 69.3 %, belite content of 2.7 % and free CaO content of 3.2 % were studied, and the relation between replacement ratio of FA and compressive strength of concrete was given as shown in Figure 2. It can be seen from the results that compressive strength of concrete using high alite cement decreased with increase in replacement ratio of FA at 28 days or earlier. At 91 days, however, concrete with 10 % FA gained higher strength than concrete without FA. Although compressive strength of high alite cement concrete without FA did not increase beyond the age of 28 days, compressive strength of concrete with FA increased even after 28 days, which may be due to pozzolanic reactivity of FA. When compared to OPC concrete without FA, compressive strength of high alite cement concrete up to 7 days was equivalent at FA replacement up to 20 %, but strengths at 28 days and 91 days are lower at FA replacement of more than 10 %. It is desirable that the replacement ratio of FA is more than around 15 % in order to control alkali aggregate reaction. Therefore, the influence of chemical composition of clinker and replacement ratio of FA on strength development should be investigated.

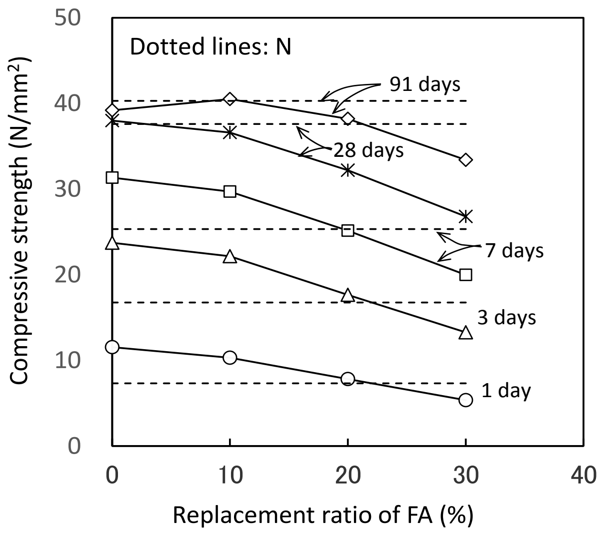


Figure 2. Compressive Strength of Concrete under Water at 20 oC

Table 6 shows compressive strength of concrete cured under water at 20 ºC. It can be seen from this table that early-age strength of high alite cement concrete containing FA at 15 % or 20 % replacement ratios is more than those of OPC (N_1 and N_2) and ordinary blended cement (BB and N_2-$FA_2$15). Furthermore, in series III, compressive strength of A_2-$FA_2$15(3.4) is equivalent to N_2 even at 28 days and 91 days. The contents of SO_3 and free CaO are different among series I, II and III, but the difference in strength development of high alite FA concretes is not so large. Belite content of A_1 is larger than that of A_2, but contribution to later-age strength development by belite is not clear. So, more studies should be conducted in the future.

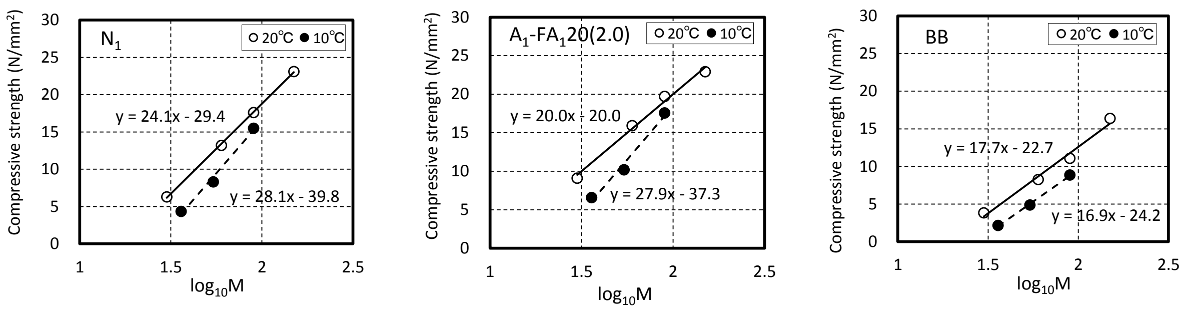
Table 6. Compressive Strength of Concrete Cured in Water at 20 ℃

Series	Cement	Copmpressive strength N/mm^2)			
		3 days	7 days	28 days	91 days
I	A_1-$FA_1$20(2.0)	18.8	25.7	33.2	38.5
	N_1	18.3	26.4	35.1	38.9
	BB	12.6	21.1	35.8	48.2
II	A_1-$FA_1$20(2.9)	19.1	23.4	28.8	36.4
	N_1	15.8	23.7	34.5	40.8
III	A_2-$FA_2$15(3.4)	19.7	26.1	30.6	36.9
	N_2-$FA_2$15(3.4)	13.5	19.1	27.4	38.4
	N_2	16.2	23.9	30.9	36.9`

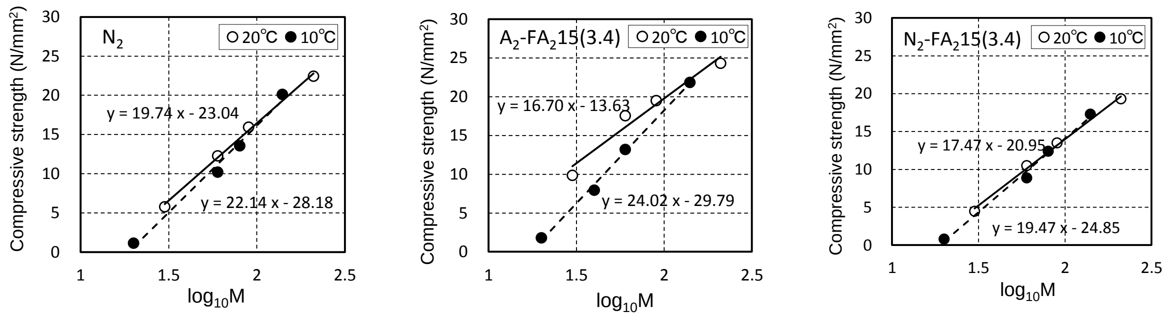
Compressive strength of sealed concrete at early ages cured at different temperatures. Test results for compressive strength of concrete at early ages up to 7 days, in which the specimens were cured in sealed condition at 10 ºC and 20 ºC, are shown in Figurer 3. In this figure, the observed compressive strength is plotted against maturity calculated by equation (1), since compressive strength of concrete at various temperatures has often been estimated on the basis of maturity concept.

$$M(t) = \sum_{z=1}^{t}(\theta_z + 10) \quad (1)$$

Where: $M(t)$ = maturity at age t (ºC･day), θ_z = the average temperature at age z (ºC) and t, z = age of concrete (day).



(a) Series I



(b) Series III

Figure 3. Compressive Strength of Sealed Concrete

When compared to the case of 20 °C, development of compressive strength of concrete was delayed at 10 °C for any type of cement. However, compressive strength of concretes with A_1-$FA_1$20(2.0) and A_2-$FA_2$15(3.4) increased earlier than those with N_1 and N_2 even if they were cured at 10 °C. In Figure 3, since linear relations can be seen between $\log_{10}(M)$ and compressive strength for each test condition, fitted lines were determined by regression analysis and are shown with the equations.

At construction sites, strength development of concrete is one of the useful indices to determine the form removal time which often affects the execution speed of construction projects. For example, it is specified by Architectural Institute of Japan that forms may be removed after confirming that concrete strength reaches the specified strength of 5 or 10 N/mm^2, depending on the service life of structures [Architectural Institute of Japan 2009]. The age at which compressive strength of concrete reaches 10 N/mm^2 was obtained from the fitted line for each type of cement in Figure 3, and it is shown in Table 7. It can be seen from this table that the age for conventional BFS cement (BB) and conventional FA cement N_2-$FA_2$15(3.4), at which compressive strength of 10 N/mm^2 is attained, is later than N_2 and N_2 at each temperature. Especially at 10 °C, the age for BB is later than N_1 by more than 2 days. For concretes with A_1-$FA_1$20(2.0) and A_2-$FA_2$15(3.4), however, it is earlier than N_1 and N_2 by 0.3 - 0.7 day at both temperatures. Therefore it can be said that some delay in form removal caused by using conventional blended cement can be avoided by using the high alite cement with FA.

Shrinkage cracking resistance of concrete subjected to drying

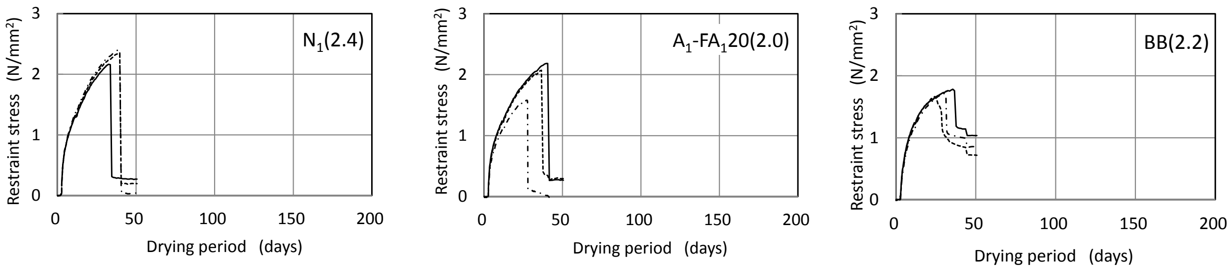
Restraint stress tests of concrete under dried condition were carried out in order to estimate resistance to shrinkage cracking of concrete with each type of cement. In this study, restraint stress of concrete subjected

Table 7. Age of Concrete to Obtain Compressive Strength of 10 N/mm^2

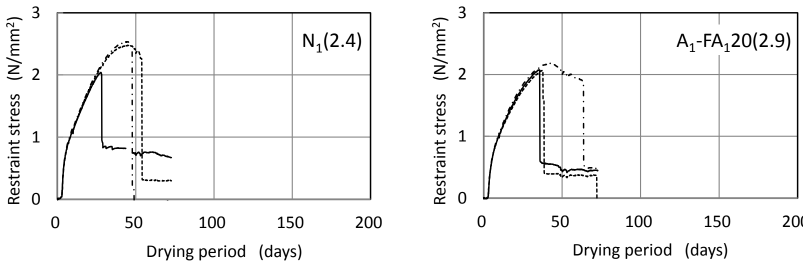
Series	Temperature	Age of concrete (days)				
		N_1 or N_2	BB	N_2-$FA_2$15(3.4)	A_1-$FA_1$20(2.0)	A_2-$FA_2$15(3.4)
I	20 °C	1.4	2.3	-	1.1	-
	10 °C	3.0	5.3	-	2.5	-
III	20 °C	1.6	-	2.0	-	0.9
	10 °C	2.7	-	3.1	-	2.3

to drying at 20 °C, 60 % R.H. after sealed curing for 3 days after casting. The test results were shown in Figure 4, in which tensile stresses are expressed as plus values on the vertical axis. It can be seen from this figure, that the restraint stresses increased with progressive drying and through cracks were observed in the specimens with sudden decreases in restraint stress in the most cases.

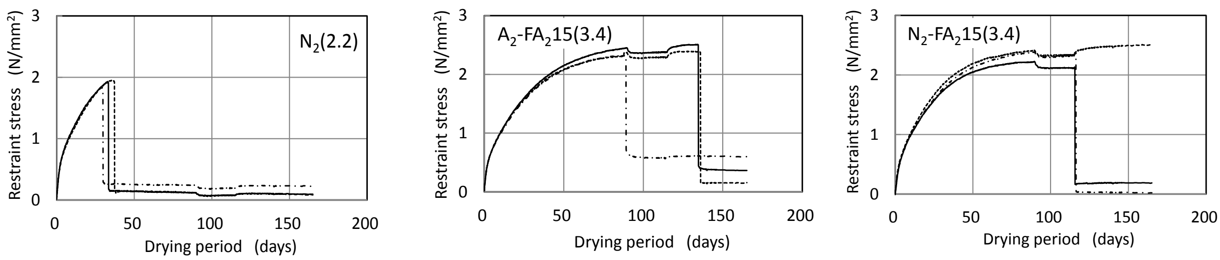
The period of drying until cracking in each specimen is shown in Table 8. It can be seen from these experimental results that the period of drying until cracking in A_1-$FA_1$20(2.0) concrete was slightly shorter than in N_1 concrete and was slightly longer than in BB concrete. It can be seen that the period of drying until cracking increased as SO_3 content was increased. In cases of A_2-$FA_2$15(3.4) and N_2-$FA_2$15(3.4), which have relatively large SO_3 content of 3.4 %, the period of drying until cracking was much longer than that of N_2. Therefore, it can be said that the resistance to shrinkage cracking of FA cement using high alite cement can be improved by increasing SO_3 content up to 3.4 %. It should be noted that the restraint stress tests in this study were conducted in a single environmental condition and in a single degree of restraint. Therefore, the influences of the conditions on test results should be investigated in the future.



(a) Series I



(b) Series II



(c) Series III

Figure 4. Restraint Stress of Concrete

Table 8. Period of Drying until Cracking in concrete specimens

Series	Cement	Period of drying until cracking (days)				Rerated to N_1/N_2
		No.1	No.2	No.3	Average	
I	N_1	34	40	40	38.0	-
	A_1-$FA_1$20(2.0)	28	37	41	35.3	0.93
	BB	29	31	37	32.3	0.85
II	N_1	28	48	54	43.5	-
	A_1-$FA_1$20(2.9)	36	39	64	46.1	1.1
III	N_2	30	34	37	33.7	-
	A_2-$FA_2$15(3.4)	89	136	138	121.0	3.6
	N_2-$FA_2$15(3.4)	116	119	>165	>117.5	>3.5

CONCLUSION

The purpose of this study is to propose general-purpose FA cement, which can be widely used for concrete structures including reinforced concrete buildings. In the experiments, some clinker samples with high alite content were experimentally produced at an actual cement plant. Strength development and restraint stress due to drying shrinkage of concrete with FA cement using the high alite clinker were experimentally investigated in comparison with those of concrete with OPC or conventional blended cement.

- Cement clinker with alite content up to 72 % by Bogue equation can be successfully produced at an actual cement plant.
- Compressive strength development of concrete with FA cement is significantly accelerated at early ages, when the high alite cement is used instead of OPC as the base material for FA cement.
- Experimental results suggest that the age of concrete at which the form removal is allowed can be set earlier for FA cement using the high alite clinker than for conventional FA cement or BFS cement.
- It is suggested from the restraint stress tests that the resistance to shrinkage cracking of concrete with FA cement using the high alite clinker can be improved by increasing SO_3 content up to 3.4 %.

ACKNOWLEDGEMENT

The study presented in this paper was carried out as a part of the activity of "Technical Committee on General-Purpose Fly Ash Cement". The authors express sincere gratitude to the parties concerned.

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