- Technical Paper -

MULTIPHASE MODELLING OF POZZOLANIC REACTION OF LOW-CALCIUM FLY ASH IN CEMENT SYSTEMS CONSIDERING THE INFLUENCE OF CRYSTALLINE MULLITE

Tiao WANG*1 and Ishida Tetsuya*2

ABSTRACT

The dissolution processes of three fly ashes were observed in alkaline conditions. The relationship between the reactivity and material properties were studied. The influence of mullite on the reactivity of Al-silicate glass is studied by synthetized glass-mullite intermixture and Al-silicate glass. Experimental results showed that mullite is crystallisation product of Al-silicate glass. The reactivity of fly ash depends on its mullite content. A multiphase model of pozzolanic reaction of fly ash was proposed. Numerical result shows that it could capture the reactivity variations of fly ash.

Keywords: Fly ash; Reactivity; Multiphase modelling; Mullite; Chemical composition

1. INTRODUCTION

The use of fly ash to improve concrete performance has increased worldwide in recent decades [1, 2]. However, as a by-product of coal-fired power plants, fly ash is an extremely heterogeneous cementitious material. Durdziński et al. [3] investigated European high-calcium fly ash by scanning electron microscopy–energy-dispersive X-ray spectroscopy (SEM-EDS) full-element mapping analysis. They found that high-calcium fly ash has different glass components [3]. Takahashi et al. [4, 5] observed that fly ash particles are heterogeneous multiphase mixtures of amorphous and crystalline components, especially crystalline mullite. These intrinsic heterogeneities and variabilities make the reactivity of fly ash vary across a wide range, which limits its engineering applications.

The reactivity of fly ash depends on its chemical composition. Snellings [6] studied synthesised calcium aluminosilicate glasses and found that the dissolution rates are scaled linearly with the glass Ca/(Al+Si) molar ratio. Bumrongjaroen et al. [7] found that the reactivity of fly ash-type glass is highly related to the number of non-bridging oxygens per tetrahedral network-forming ions (NBO/T), which is related to its chemical composition. Durdziński et al. [8] further confirmed this conclusion by comparing the dissolution rates of synthesised glass with various chemical compositions. They subsequently proposed a new advanced technique of SEM-EDS full-element mapping to identify and characterise various glass components of fly ash [3]. Therefore, hydraulic or basicity indices, which are the glass polymerisation degree expressed as the amount of network-modifying to network-forming elements such as $(CaO+MgO+Al_2O_3)/SiO_2$ (JIS A 6206) [9], are usually used to describe fly ash reactivity in engineering practices.

Recent studies [10, 11] further found that, besides chemical composition, the crystallinity degree of the material is another crucial parameter affecting its reactivity. Wolff-Boenisch et al. [10] found that the dissolution rates of natural Si-rich silicate glasses are significantly faster than their crystalline counterparts.

We recently studied the reactivity of three low-calcium fly ashes in alkaline conditions and cement systems [12]. The reactivities of these three fly ashes are significantly different although they have similar chemical compositions [12]. It shows that chemical composition of fly ash is not enough to explain their reactivity variations. SEM-EDS mapping result further shows that most mullite is widely distributed in Al-silicate glasses as small crystalline solids [12]. Therefore, we proposed a hypothesis that mullite is crystallisation product of Al-silicate glasses and the reactivity of fly ash is affected by the mullite content of fly ash [12]. Based on this hypothesis, the reactivity variations were successfully captured by multiphase pozzolanic reaction model [12]. Although some efforts have been done, our previous study did not directly observe the influence of mullite on the reactivity of fly ash.

Against this background, this paper reports the further study of these three fly ashes. The reactivity of these fly ashes was observed long-term in alkaline conditions by measuring the dissolved concentrations of the various elements. The pozzolanic reaction degree of these fly ashes were studied by selective dissolution tests. The calcium hydroxide contents were determined by thermogravimetric analysis test (TGA test). The chemical and mineralogical influences were studied using synthesised silicate and Al–silicate glass and a glass–mullite intermixture. Experiment result shows that mullite is crystallisation product of Al-silicate and chemical and mineralogical has significant influence on the reactivity of fly ash. Based on these experiment

*1 Assistant Prof., Dept. of Civil Engineering, The University of Tokyo, Dr.E., JCI Member
*2 Prof., Dept. of Civil Engineering, The University of Tokyo, Dr.E., JCI Member

results, a multiphase pozzolanic reaction model was proposed [12]. This model could predict the pozzolanic reaction degree of fly ash and calcium hydroxide content of fly ash cement paste.

2. EXPERIMENTAL MATERIAL PROPERTIES

2.1 Fly ash

Three commercial fly ashes were selected form among 11 types of commercially available fly ashes in Japan according to their solubilities in NaOH solution. All three fly ashes satisfied the JIS II category specified by Japan Industrial Standard JISA 6201 [13]. The material properties were reported in previous studies [14, 15]. The chemical and mineralogical compositions of these fly ashes are summarised in Table 1 [14]. The densities and specific surface areas determined by laser diffraction SSA_{LD} and image analysis SSA_{img} are also listed in Table 1[15].

Table 1 Material properties of fly ash [14]

Chemical composition (wt. %)				Minermical compostion (wt. %)			
Item	FA1	FA2	FA3	Item	FA1	FA2	FA3
Al_2O_3	20.23	21.35	19.78	Mullite	10.92	6.19	14.18
SiO_2	64.1	60.57	63.78	Quartz	7.21	4.89	10.28
CaO	2.19	2.01	4.65	Lime	0.04	0.08	0.23
Fe_2O_3	4.17	5.2	4.12	Hematite	0.33	0.34	0.62
Na_2O	0.58	0.68	0.54	Magnetite	0.36	0.52	0.59
MgO	0.7	0.75	0.69	Glass	81.15	87.99	74.09
K_2O	1.24	1.4	1.07	Physicla properties			
TiO_2	1.4	1.18	1.24	Density (cm^3/g)	2.283	2.344	2.339
Carbon	3.09	3.29	2.52	SSA_{LD} (cm^2/g)	5756	5669	5450
Sum	97.7	96.43	98.39	SSA_{Img} (cm^2/g)	7802	5585	5380

These fly ashes are further studied by SEM-EDS mapping analysis (e.g. Fig. 1 (a)) [14]. The major glass components are silicate and Al-silicate. It is also found that mullite usually exists as a small solid and is widely distributed in fly ash particles (Fig. 1 (b)) [12]. Thus, a transitional phase between pure mullite and glass – the glass-mullite intermixture phase – was proposed between the two phases [14]. A segmentation criterion based on chemical composition was proposed to distinguish various crystalline and glass components (Fig. 2 (a)) [14]. The SEM-EDS mapping results are shown in Fig. 2 (b) and summarized in Table 2 [14]. These three fly ashes have similar silicate and Al-silicate glass contents.

Table 2 SEM-EDS mapping result (wt. %) [14]

	Total glass	Silicate	Al-silicate	Glass-mullite	Mullite	Qiartz	Crytalline iron	Lime
FA1	81.76	10.83	48.40	33.44	0.01	7.18	0.14	0.02
FA2	85.78	10.36	44.85	36.72	0.04	7.10	0.85	0.08
FA3	75.14	13.76	46.37	29.14	0.05	10.31	0.25	0.13

2.2 Synthesised materials

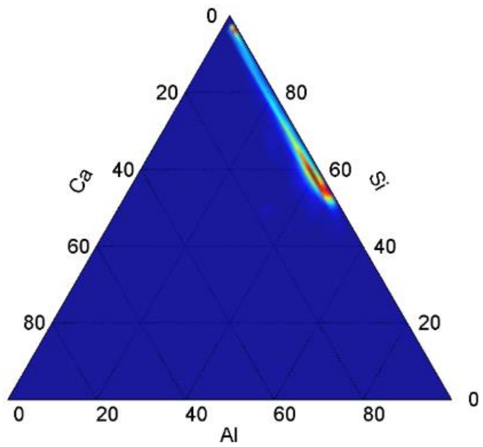
The model glass–mullite intermixture and its counterpart Al–silicate glass was synthesised from designed mixtures of reagent-grade Al_2O_3, SiO_2, $CaCO_3$, Na_2CO_3 and MgO. The samples were first heated to 950 °C at a slow rate of 5 °C/min to ensure that the $CaCO_3$ completely decarbonated. The samples were then heated to 1600 °C at a rate of 10 °C/min and kept at that temperature for 4 h to ensure the powders were completely and homogeneously melted. The model glass was quenched in deionized water but the model glass–mullite intermixture was cooled in air.

The chemical compositions of these glasses were measured by X-ray fluorescence (XRF) and are listed in Table 3. The particle size distribution (PSD) of these glasses was measured by laser diffraction (LD) and deduced by the Mie optical model. The median diameters are listed in Table 3.

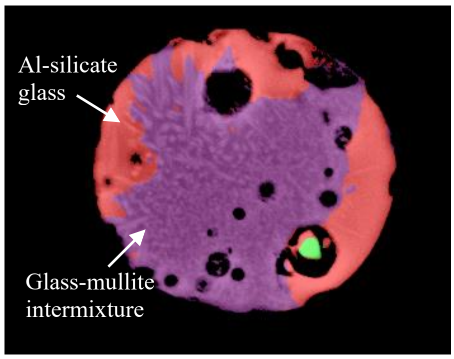
Table 3 Synthesised material properties

	Al_2O_3 (%) (Al at%)	SiO_2 (%) (Si at%)	CaO (%) (Ca at%)	Density (g/cm^3)	D_{50} (μm)	Molar mass (g/mol)
Al–silicate	32.69 (39.44)	54.94 (56.24)	3.94 (4.32)	2.656	46.97	72.53
Glass–mullite	31.11 (37.63)	56.73 (58.22)	3.78 (4.16)	2.662	71.707	71.92

The XRD scans of these synthesised materials are shown in Fig. 3. An apparent crystalline peak was not found for the synthesised glass, but a crystalline mullite peak appeared in the glass–mullite intermixture. Crystalline mullite is the crystallisation product of Al–silicate glass. The mullite content of the glass–mullite intermixture was 13.11%, which was determined by the internal standard method with Rietveld analyses. It can thus be seen that the combustion conditions highly affected the mineralogical composition of the fly ash, which experimentally proves our previous discussions [12, 14, 17].

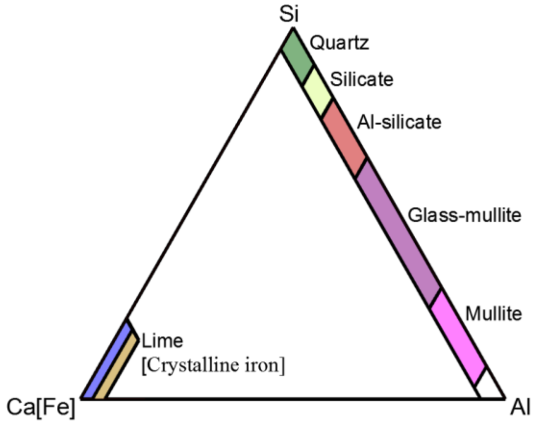


(a) Chemical distribution of FA1 [12]

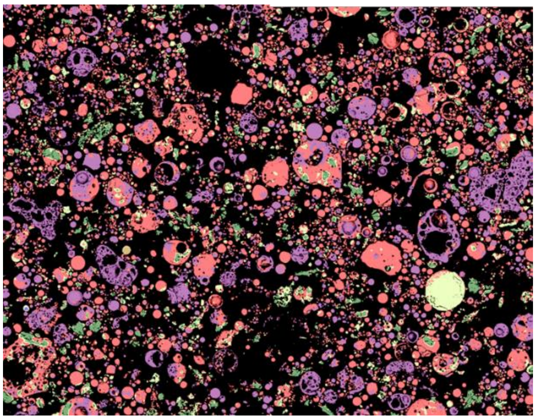


(b) Al-silicate glass and glass-mullite intermixture [12]

Fig. 1 Chemical distribution of FA1 and glass-mullite intermixture [12,14,17]

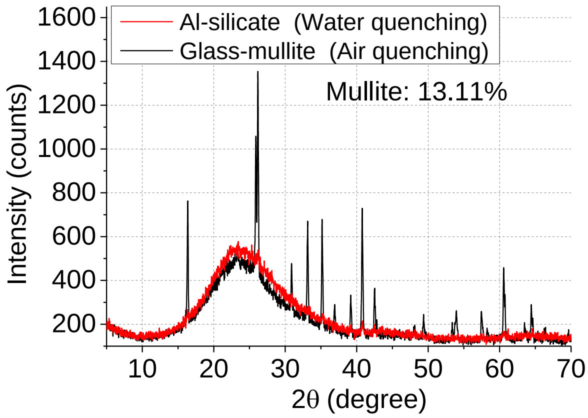


(a) SEM-EDS segmentation criterion [17]



(b) SEM-EDS mapping result of FA1 [17]

Fig. 2 Mineralogical segmentation criterion and SEM-EDS mapping result [12,14,17]



(b) Synthesised Al–silicate glass and glass–mullite intermixture

Fig. 3 XRD scans of synthesised materials

2.3 Cement

The ordinary Portland cement (OPC) was used in this study. The phase compositions ware analysed by XRD (XRD 6100, Shimadzu). Rietveld analyses were carried out using commercial software Siroquant (version 3.0). The cement material properties are listed in Table 4.

Table 4 Cement material properties [17]

	C_2S (wt. %)	C_3S (wt. %)	C_3A (wt. %)	C_4AF (wt. %)	Fineness (cm^2/g)	Density (g/cm^3)
OPC	15.9	65.5	8.07	8.77	3350	3.15

3. EXPERIMENT PROCEDURES

3.1 Alkaline dissolution test

The reactivity of the selected fly ashes and synthesised glass were investigated in NaOH solutions at 20 °C. The theoretical pH of this alkaline solution is 13.2 at 20 °C, which corresponds to the pore solution of common Portland cement paste suggested by Durdziński et al. [8]. Around 0.5 g of the tested materials was added to 1 L of a prepared NaOH solution (i.e., water–solid ratio is 2000) in a 1500 mL air-tight polypropylene bottle. A high water–solid ratio was used in this test is to ensure far-from-equilibrium conditions and to limit the precipitation of hydrates during long term measurements. All bottles were sealed with tape and stored in a curing room at 20 ± 2 °C. On the designated sampling days, 10 mL aliquots were sampled from the NaOH solutions and stored in small, air-tight polypropylene bottles. Ten millilitres of the NaOH (0.14 mol/L) solution was added back to the original bottles to keep the volume in the reactor constant. The dilution effect was taken into account during data analysis. The silicon, aluminium and calcium aqueous concentrations of the samples were measured with an ICPS 7000 system (Shimadzu). The experimental errors were expected to be less than 5%.

3.2 Pozzolanic reaction degree in cement systems

Three series of fly ash cement pastes with a water–to–binder ratio of 0.4 and a fly ash replacement ratio by mass of 0.3 were prepared for sealed curing at 20 °C to further investigate the reactivity of fly ash. The pastes were cured at 20 °C in petri dishes until testing after 7, 14, 28, 56, 91 and 182 days. The pozzolanic reaction degree of the fly ash was determined by the selective dissolution method with ethylenediaminetetraacetic acid (EDTA) and the procedures proposed by Haha et al. [16].

3.3 TGA test

The initial mass of raw materials, calcium hydroxide content and bound water content of cement pastes were determined by TGA tests. The tests were conducted using a Thermo plus EVO2 (Rigaku) TGA analyser. Cement samples were first degassed for 1 hour at 30 °C under a N_2 protective atmosphere to evacuate the remaining isopropanol and physically absorbed water. Afterward, heating from 30 °C to 1000 °C at a rate of 15 °C/min was carried out under a protective N_2 atmosphere to minimise sample carbonation.

4. EXPERIMENT RESULT AND DISCUSSIONS

4.1 Dissolution reactivity of fly ash

The dissolution reactivities are estimated by the composited dissolution degree proposed previously (i.e. Eq. (1)) [12]

$$D_{com} = \frac{m_{SiO_2} + m_{Al_2O_3} + m_{CaO}}{m_f\left(c'_{SiO_2} + c'_{Al_2O_3} + c'_{CaO}\right)} \quad (1)$$

where m_f is the mass of fly ash powder added to the NaOH solution; m_{SiO_2} is the dissolved SiO_2 content estimated by the concentration of Si ions in alkali solution; and C'_{SiO_2} is the mass fraction ratio of the SiO_2 content of the specified fly ash given in Table 1. Similar definitions were used for the compositions of Al_2O_3 and CaO.

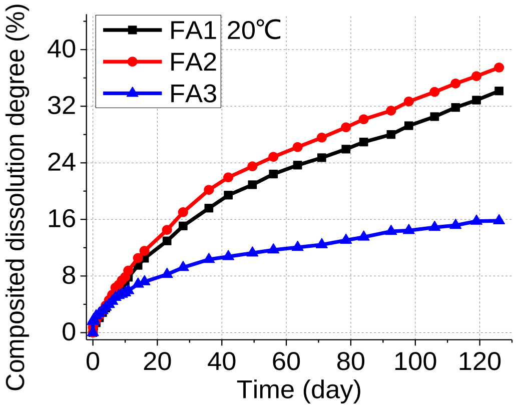


Fig. 4 Composited dissolution degree of fly ashes [12].

Our previous study had reported the composited dissolution degree of the three different fly ashes [12]. Fig. 4 presents it for comparison. FA1 and FA2 have similar reactivities, but FA3 is less reactive than FA1 and FA2. However, the SSAs and combined mineralogical compositions are insufficient to explain the variations in the dissolution processes shown in Fig. 4. FA3 had similar mineralogical composition and SSA as FA2, but it showed much lower reactivity than FA2. That is probably because of the variations of crystalline mullite content in these fly ashes, which will be discussed in following sections.

4.2 Pozzolanic reaction degree of fly ash

The pozzolanic reaction degrees of these fly ash were studied by selective dissolution tests. Fig. 5 presents our previous result of pozzolanic reaction degrees of these fly ashes in OPC system [12]. The pozzolanic reaction degrees in OPC systems agree well with the dissolution processes in alkaline conditions. FA1 and FA2 have similar pozzolanic reactivities, but FA3 is less reactive than FA1 and FA2. It shows that dissolution process is the controlling step of pozzolanic reaction of fly ash in cement systems [12]. The alkaline dissolution test can be used to study the reactivity of fly ash.

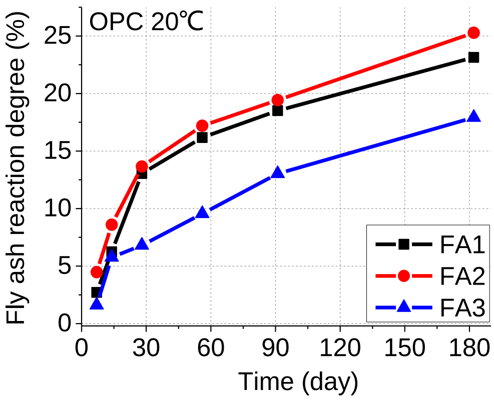


Fig. 5 Pozzolanic reaction degree of fly ashes [12]

4.3 Calcium hydroxide content

Fig. 6 shows the calcium hydroxide content of fly ash blended cement paste. The experimental result agrees well with the pozzolanic reaction processes cement paste. FA1 is slightly higher than FA2 and FA3 is the highest one amongst these three fly ashes. It further shows that fly ash have different reactivities although they have similar silicate and Al-silicate glass contents.

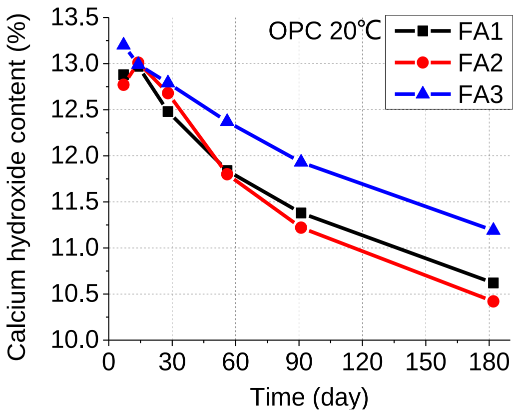


Fig. 6 Calcium hydroxide contents of fly ash cement paste [17]

4.4 Dissolution reactivity of synthesized materials

An alkaline dissolution test was conducted to study the influence of crystallisation on the reactivity of the Al–silicate glass. The absolute SSAs of these samples were determined by the PSD results. Linear regression analysis was performed to estimate the normalised dissolution rates with the absolute SSAs. The adjusted R-squared values are both higher than 0.97. If we assume that the crystalline mullite solids of the glass–mullite intermixture have the same SSA as the glass content, the relative dissolution rates of the glass component of glass-mullite intermixture can be estimated.

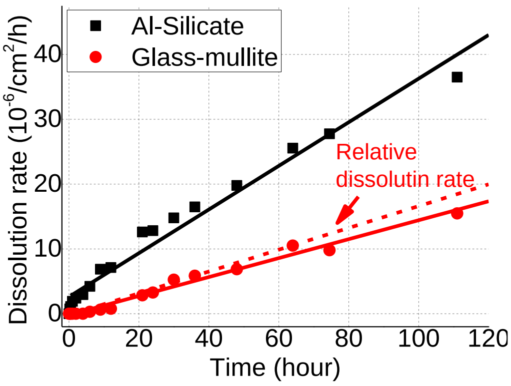


Fig. 7 Normalised dissolution rates of Al-silicate and glass-mullite intermixture

Fig. 7 presents the normalised dissolution rates of Al-silicate glass and glass-mullite. The normalised dissolution rate of the Al–silicate glass is almost two times that of the glass–mullite intermixture. It shows that the reactivity of fly ash decreases as mullite content increases. Based on this result, the reactivity varieties of these fly ashes can be more reasonably explained. FA1 has a higher specific surface area but more mullite contents (Table 5) than FA2; therefore, FA1 and FA2 have similar dissolution processes. FA3 has the lowest specific surface area and highest mullite content (Table 5), and the dissolution process is therefore the lowest amongst these three fly ashes. In conclusion, the reactivity of low-calcium fly ash depends upon its glass phase assemblage, its surface areas and the mullite-content.

Table 5 Summary of fly ash material properties

	Silicate (wt. %)	Al-silicate (wt. %)	Glass-mullite (wt. %)	Mullite (wt. %)	SSA_{Img} (cm^2/g)
FA1	10.83	48.40	33.44	10.92	7802
FA2	10.36	44.85	36.72	6.19	5585
FA3	13.76	46.37	29.14	14.18	5380

5. MULTIPHASE POZZOLANIC REACTION MODEL

DuCOM is a multiphase modelling platform for concrete materials, which is developed in recent decades by our research group. It is a multi-chemo-physic platform that is capable of simulating cement hydration, latent hydration of ground-granulated blast-furnace slag and pozzolanic reaction of fly ash, micro-pore structure formation and mass transport in concrete ranging from nanometre to micrometre scales.

Based on DuCOM system, a multiphase pozzolanic reaction model (Eqs. (2) and (6)) with silicate and Al-silicate glass was proposed to simulate the pozzolanic reaction of fly ash in cement systems [12].

$$H^{20℃}_{silicate} = f_r \times \frac{0.025}{R_p} \times \left(1-\frac{Q^c_{silicate}}{Q_{FA,\infty}}\right)^{2.0} \quad (2)$$

$$H^{20℃}_{Al-silicate} = f_r \times 0.025 \times \left(1-\frac{Q^c_{Al-silicate}}{Q_{FA,\infty}}\right)^{2.0} \quad (3)$$

$$f_r = R_{FA} \times \left(1.2SSA^N_{FA} - 0.2\right) \quad (4)$$

$$H^T_{FA} = P_{silicate} H^T_{silicate} + P_{Al-silicate} H^T_{Al-silicate} \quad (5)$$

where $H^{20℃}_{silicate}$ and $H^{20℃}_{Al-silicate}$ are the heat generation rates of the pozzolanic reactions of silicate and Al-silicate glass at 20 °C (cal/g), respectively; H^T_{FA} is the total heat generation rate of the pozzolanic reaction of fly ash at temperature T (cal/g); f_r is the pozzolanic reaction factor of fly ash considering the influence of normalised specific surface area of fly ash SSA^N_{FA}; $Q^c_{silicate}$ and $Q^c_{Al-silicate}$ are the accumulated release-heat of silicate and Al-silicate glass (cal/g), respectively; $Q_{FA,\infty}$ is the total heat of the fly ash (still assumed to be 50 cal/g in this modified model); $P_{silicate}$ and $P_{Al-silicate}$ are the mass fractions of glass silicate and Al-silicate, respectively

Recent researches reported that the Al-silicate glass is more reactive than silicate glass because additional Al element breaks the tetrahedral network of silicate glass. Therefore, a reaction reduction factor between silicate and Al-silicate glass R_p was introduced, which is assumed to be 4.0 according to the alkali dissolution test [12].

Moreover, previous experimental results indicated that the reactivity of fly ash decreases as mullite content increases. Therefore, a relative reactivity of fly ash R_{FA} was also introduced to consider the influence of mullite (i.e. Eq. (6)) [12].

$$R_{FA} = \frac{c_{Amorphous}}{15 \times c_{mullite}} \quad (6)$$

where $c_{Amorphous}$ and $c_{mullite}$ are the amorphous content and mullite content of fly ash. It should be noted that this equation is still empirical. More studies are needed in the future to investigate the relationship between mullite content and reactivity of fly ash.

Coupled with multiphase pozzolanic reaction model, a tentative calcium hydroxide consumption model was also proposed (i.e. Eq. (7)) [17].

$$FA_{CH} = 0.035 CH^{Pot}_{CE} - P_{FA}/100 \quad (7)$$

$$CH^{Pot}_{CE} = P_{C_2S} \times PCH^{20°C}_{C_2S} + P_{C_3S} \times PCH^{20°C}_{C_3S} \quad (8)$$

where P_{C_2S} and P_{C_3S} are the mass percentage of belite and alite in cement; and $PCH^{20°C}_{C_2S}$ and $PCH^{20°C}_{C_3S}$ are the production rates of calcium hydroxide of belite and alite at 20 °C, which are assumed to be 1.74 and 1.91 in this study; P_{FA} is the fly ash replacement ratio.

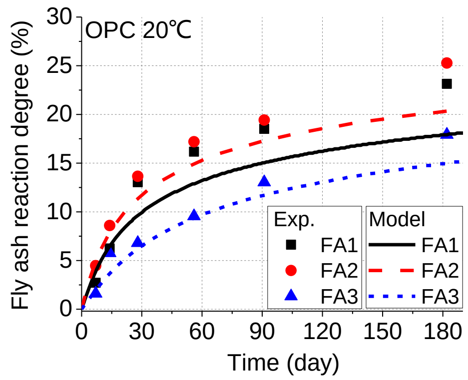


Fig. 8 Comparison of fly ash reaction degrees and multiphase reaction model [12]

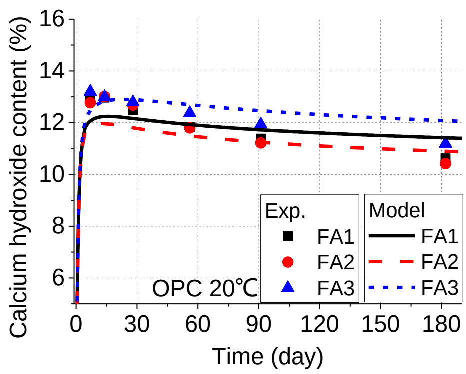


Fig. 9 Comparison of calcium hydroxide content and proposed model [17]

Fig. 8 shows numerical results of pozzolanic reaction degrees. Fig. 9 presents the numerical results of calcium hydroxide content of fly ash cement pastes. The predictions were all improved and in good agreement with the experimental results. It shows that the multiphase reaction model could simulate the pozzolanic reaction of fly ash in cement systems considering variations of fly ash material properties.

6. CONCLUSIONS

The reactivity of three low-calcium fly ashes were experimentally investigated in alkaline conditions and cement systems. The relationship between reactivity and material properties is discussed in this paper. Experimental result shows that the fly ashes have different reactivities even they have similar silicate and Al-silicate glass contents. Mullite is crystallisation product of Al-silicate glass. The reactivity of fly ash depends on its mullite content. Based on experimental observations, a multiphase pozzolanic reaction model was proposed based on fly ash material properties characterizations. Numerical results show that this proposed model could predict the pozzolanic reaction degrees and calcium hydroxide content of fly ash cement pastes considering variations of fly ash material properties.

ACKNOWLEDGEMENT

The authors acknowledge the Materials Characterization and Preparation Center of Southern University of Science and Technology for its technical support on SEM-EDS mapping and laser diffraction tests. The authors also acknowledge the he State Key Laboratory of Green Building Materials of China Building Materials Academy for their technical support on synthesising glass. The authors are grateful for the financial support received from Japan Society for the Promotion of Science (Project code: 17H01284 and 19K15059).

REFERENCES

[1] M.D.A. Thomas, “Optimizing the use of fly ash in concrete”, Portland Cem. Association Skokie, IL, 2007.

[2] P.K. Mehta, P.J. Monteiro, “Concrete: Microstructure, Properties, and Materials (Third Edition)”, McGraw-Hill Education, New York, 2005.

[3] P.T. Durdziński, C.F. Dunant, M.B. Haha, K.L. Scrivener, “A new quantification method based on SEM-EDS to assess fly ash composition and study the reaction of its individual components in hydrating cement paste”, Cem. Concr. Res. 73, 2015, pp. 111-122.

[4] H. Takahashi, K. Yamada, “Characterization of fly ash dominating ASR suppressing effects by using SEM-EDS and EBSD (in Japanese)”, Concr. Res. Technol. 23(1), 2012, pp. 1-11.

[5] H. Takahashi, K. Yamada, “Critical characters of fly ash suppressing ASR”, Proc. of 14th Int. Conf. Alkali-Aggregate Reaction Concr. 2012.

[6] R. Snellings, “Solution-controlled dissolution of supplementary cementitious material glasses at pH 13: the effect of solution composition on glass dissolution rates”, J. Am. Ceram. Soc. 96(8), 2013, pp. 2467-2475.

[7] W. Bumrongjaroen, S. Swatekititham, R. Livingston, J. Schweitzer, “Synthetic glass models for investigating fly ash reactivity”, ACI special publication, 242, 2007, pp. 227-242.

[8] P.T. Durdziński, R. Snellings, C.F. Dunant, M.B. Haha, K.L. Scrivener, “Fly ash as an assemblage of model Ca–Mg–Na-Aluminosilicate glasses”, Cem. Concr. Res. 78, 2015, pp. 263-272.

[9] JIS, Standard “A 6206-2013: Ground granulated blast-furnace slag for concrete”, (in Japanese), 2015.

[10] D. Wolff-Boenisch, S.R. “Gislason, E.H. Oelkers, The effect of crystallinity on dissolution rates and CO_2 consumption capacity of silicates”, Geochim. Cosmochim. Ac. 70(4), 2006, pp. 858-870.

[11] M. Vallet-Regí, A.J. Salinas, J. Ramírez-Castellanos, J.M. González-Calbet, “Nanostructure of bioactive sol−gel glasses and organic−inorganic hybrids”, Chem. Mater. 17(7), 2005, pp. 1874-1879.

[12] T. Wang, T. Ishida, “Multiphase pozzolanic reaction model of low-calcium fly ash in cement systems”, Cem. Concr. Res. Vol. 122, Aug. 2019, pp. 274-287.

[13] JIS, Standard “A6201-2015: Fly ash for use in concrete (in Japanese)”, 2015.

[14] T. Wang, T. Ishida, R. Gu, “A study of the influence of crystal component on the reactivity of low-calcium fly ash in alkaline conditions based on SEM-EDS”, Constr. Build. Mater., 2020.

[15] T. Wang, T. Ishida, R. Gu, “A comparison of the specific surface area of fly ash measured by image analysis with conventional methods”, Constr. Build. Mater. 190(30), 2018, pp. 1163-1172.

[16] M.B. Haha, K. De Weerdt, B. Lothenbach, “Quantification of the degree of reaction of fly ash”, Cem. Concr. Res. 40(11) 17, 2010, pp. 1620-1629.

[17] T. Wang, “Modeling of pozzolanic reaction of siliceous fly ash in cement system based on its material characterization”, Ph.D. Thesis, The Univeristy of Tokyo, 2017.