

Modeling of Pozzolanic Reaction of Siliceous Fly Ash  
in Cement System based on its Material Characterization

(クリンカーとの相互作用を考慮した物理化学キャラクターに基づくフ  
ライアッシュのポゾラン反応モデル)

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Fly ash is a by-product of burning pulverized coal in thermal power plants. It is a high potential supplementary cementitious material to improve concrete performance in many aspects. For example, fly ash could not only modify the workability of concrete and reduce its water demand but also chemically react with calcium hydroxide to form cementitious compounds and further enhance concrete strength and durability. Moreover, replacing part of the cement by fly ash as cementitious material is also beneficial to circular economy and sustainable development through reducing the energy consumption and CO<sub>2</sub> emission during production of cement.

Although these potential benefits for using fly ash as a supplementary cementitious material with cement has been well known since the start of the last century, it has not yet been widely applied in concrete constructions and only around 30 mass % of produced fly ash is used right now. The primary reason is due to its intrinsic and significant heterogeneity and variability. Its properties are highly affected by the coal type, combustion temperature, production processing, and other factors and therefore the reactivity of fly ash can be profoundly different even produced in the same power plant but from different batches.

Another drawback of fly ash is its slow reaction rate compared to cement, especially for siliceous fly ash. At high fly ash replacement ratio engineering problems may be encountered with extended setting time and slow strength development, resulting in low early-age strengths and delays in the rate of construction. This shortcoming becomes particularly pronounced in winter construction. Therefore, high temperature curing is often used in fly ash concrete to accelerate its reaction. Unfortunately, current information is still far from enough to understand its temperature-dependent behavior and the corresponding effect on concrete performance as well.

Therefore, it is promising and necessary to study and model pozzolanic reaction of fly ash in cement systems. For this purpose, through experimentally studying the temperature-dependent dissolution and pozzolanic reaction mechanism of fly ash in alkaline system and cement system respectively, this thesis is aim to develop a new method to characterize fly ash temperature-dependent reactivity and finally propose a unified model which is capable of covering different siliceous fly ashes and predict their temperature-dependent pozzolanic reaction in cement systems.

In this study, three different types of siliceous fly ashes were first selected based on its solubility in alkaline condition. The material properties characterization of these fly ashes, including phase assemblage and specific surface area, was studied by SEM-EDS full element mapping test with a developed image analysis method. During these analyses, a developed phase segmentation criteria was also proposed to distinguish and then quantify the amount of reactive phases of fly ash.

Subsequently, the temperature-dependent dissolution process and pozzolanic reaction degree in cement systems of these fly ashes were investigated in NaOH solution and in blended cement pastes under sealed curing conditions at 20 and 60 °C respectively. Based on these experiment result and fly ash material characterization, the temperature-dependent reactivity of indivial amorphous phase and integral fly ash were comprehensively studied and the corresponding mechanisms were also proposed to explain the reactivity variance of fly ashes.

The interaction between fly ash and cement and calcium hydroxide consumption were also investigated and studied in this study. Experiment result showed that cement hydrations of fly ash cement paste were accelerated in room temperature but retarded at high temperature, which is the competition result of filler effect and dilute effect of fly ash and the C-S-H gel morphology effect induced by variances of calcium hydroxide content in paste.

Based on all information, a temperature-dependent two-phase reaction model both considering fly ash material variances and cement and fly ash and cement interaction was finally proposed to predict the temperature-dependent pozzolanic reaction of siliceous fly ash in cement systems and finally validated through comparisons with the experimental experimental results.