



Hydration of coal–biomass fly ash cement

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ABSTRACT

This paper presents possibilities of use of fly ashes from co-combustion bituminous coal and biomass in cement production process. Both fly ashes coming from co-combustion bituminous coal and biomass and the ones from bituminous coal combustion were analysed. The following properties of cement were tested: heat of hydration, Ca(OH)₂ content, unreacted C₃S content and microstructure. The results showed that fly ashes from co-combustion coal and biomass retard cement hydration. Cement samples containing coal-biomass fly ashes demonstrate adverse features like lower heat of hydration, higher Ca(OH)₂ content and lower rate of C₃S hydration in comparison to the ones containing fly ashes from bituminous coal. The incorporation of coal-biomass fly ashes in cement results in an increase of porosity of cement paste, leading to a microstructure of lower density.

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1. Introduction

Fly ash is a waste that is released from coal combustion process in electric power stations [1,2]. However, the coal combustion process inevitably comes with CO₂ emission to atmosphere. The Kyoto Protocol to the United Nations Framework Convention on Climate Change (UNFCCC) – adopted in Kyoto on December 11 of 1997 – stabilized the concentration of greenhouse gasses in the atmosphere, which are responsible for climatic changes in the world. The Kyoto Protocol establishes specific reduction of greenhouse gas emissions for the period 2008–2012 for Parties included in Annex I to the Convention, which includes industrialized countries and countries undergoing the process of transition to a market economy. That is for each country a percentage of greenhouse gases emission has been fixed in relation to 1990, which will have to be reached in the period from 2008 to 2012, with a global reduction equivalent to 5%. Poland has adopted already the national renewable energy strategy of 7.5% in 2010 and is harmonising its environmental and energy policy with those of European Union.

According to disposal of Minister of Economy from May 30 of 2003 [3], many electric power stations in Poland have implemented process of co-combustion coal and biomass. In 2004, biomass provided approximately 11–14% of the world's primary energy consumption (European Statistical Office), whereas the biomass utilization for energy power production in Poland was only 4.5%.

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The results of analysis show that quantitative chemical composition of coal and biomass is very different [4,5]. The biomass contains less oxygen and carbon as coal, similarly the content of sulphur, nitrogen and ash in biomass is also less than those in coal. This results in higher content of volatile matter and higher activity of biomass. The adverse feature of biomass is its moisture content, which leads to decrease in its calorific value. High content of calcium, alkalis and phosphorus as well as high concentration of Cl⁻ ions are usually regarded as characteristic features of biomass. The variability of properties of biomass results in variable properties of fly ashes resulting from co-combustion of coal and biomass.

Fly ash has been used as additive to ordinary Portland cement (OPC) or as admixture for concrete [1,2]. The differences in chemical composition between coal and biomass are reflected in higher content of calcium, magnesium, and phosphorus in coal–biomass fly ashes [6]. These properties of coal–biomass fly ashes have an impact on quality of blended cements. For example, high content of calcium or magnesium, due to their delayed hydration, can cause inner stresses that in turn lead to destructive changes in a concrete structure. The P³⁺ ions contained in fly ashes can react with Ca²⁺ ions in the liquid phase of cement paste forming a dense coating of Ca₃(PO₄)₂ on unhydrated cement grains, which retards cement hydration, especially C₃S hydration and consequently decreases compressive strength of cement, especially for early hardening period.

Role of fly ashes from bituminous coal combustion in OPC hydration was investigated [7–14]. With these fly ashes, induction period can be lengthened or remain unchanged and main peak intensity are increased or decreased [11–16]. Retarding of C₃S hydration is attributed to dissolution of fly ashes and incorporating

into solution alumina and organic ions [11]. At 28 day, C_3S hydration degree is greater by 3–9% in comparison to OPC [14]. In fly ash cement, besides C–S–H from the C_3S hydration, C–S–H from pozzolanic reaction is also found. At 8 day, CaO/SiO₂ molar ratio of C–S–H around C_3S grains decreases from 1.71 to 1.55 [17], but after 4 years – reaches 1.01 [18]. The content of Ca(OH)₂ decreases slightly or distinctly in spite of progress of OPC hydration [9,10,13,19–21].

According to authors [22–24], coal–biomass fly ashes from co-combustion are able to meet the requirements of the European standard (EN 450-1). These fly ashes perform as well or much better than pure coal fly ashes in related to strength built up by pozzolanic reaction in mortar, although they consume the similar amount of Ca(OH)₂ in pozzolanic reaction [23]. The coal–biomass fly ashes are more efficient in depressing alkali–silica-reaction expansion even in the presence of high alkali ion concentrations [24]. The results show that co-combustion fly ashes adsorb alkali ions from the pore solution and form non-expansive products instead of the expansive alkali silica gel. Performance tests of concrete confirm the suitability of fly ashes from high percentages of co-combustion [22]. The initial strength of this concrete is comparable to that of concrete made with coal fly ashes and differences are very small. The 28-day compressive strengths were practically equal. This means that there is lack of negative influence of specific components (such as phosphate) on the hydration of fly ash cement. It all depends on the nature of the co-combustion fuel and especially on its inorganic matter and ash content.

In papers [25,26], another concept for the assessment of the quality of co-combustion fly ashes for use as additive to OPC is developed based on the relation between the chemical and phase composition of coal–biomass fly ashes and properties of cement. The results show that incorporation of these fly ashes in cement results in a lengthened induction period and delayed cement hydration [25]. The consequence of that is lower cement hydration degree, especially in initial days of hydration. The decreases in compression strength are 17% and 20% – respectively – for 20% and 40% cement replacement range by fly ashes [25,26]. This difference decreases to 30% after 28 days and to 24% after 180 days of hydration. The lower early compressive strength of cement with coal–biomass fly ash is attributed to increase in total pore volume and forming microstructure of higher permeability. The drop of early compressive strength of this cement is also connected with retarding influence of P³⁺ ions on the cement hydration reaction.

This paper presents possibilities of use of fly ashes from co-combustion of coal and biomass as additive to OPC. The basic researches were carried out to investigate hydration of cement with fly ashes from co-combustion of bituminous coal and biomass. The fly ashes came from the 3rd hopper in electrostatic precipitator (ESP) system. The cement hydration was examined by determination of heat of hydration, Ca(OH)₂ content, unreacted C_3S content and analysis of cement paste microstructure. The experimental part was completing by assessing the mechanical performance (compressive strength) of coal–biomass fly ash cement.

Table 1
Chemical analysis of Portland cement clinker.

Chemical analysis (wt%)	Portland cement clinker
SiO ₂	21.6
Al ₂ O ₃	4.7
Fe ₂ O ₃	2.5
CaO	66.8
MgO	1.7
SO ₃	1.5
Na ₂ O	0.14
K ₂ O	1.06
Na ₂ O _e	0.84
CaO _{free}	1.1

Table 2
The physical properties of fly ashes.

Physical properties	FA1	FA2
Density (g/cm ³)	2.35	2.26
Blaine value (m ² /kg)	480	280
Residue on 45-μm sieve (%)	30.0	52.9
Mean particle size (μm)	33.2	48.0

2. Experimental

2.1. Materials

Portland cement Type I according to PN-EN 197-1 [27] was used in these investigations. Portland cement had been obtained by grinding the Portland cement clinker together with gypsum (5 wt%) to fineness of 350 m²/kg. The chemical analysis of Portland cement clinker is shown in Table 1. The mineral composition of Portland cement clinker as calculated by conventional Bogue's equations are C_3S 72.7%, C_2S 7.2%, C_3A 8.2% and C_4AF 7.6%.

Fly ashes came from the 3rd hopper in electrostatic precipitator (ESP) system. The first fly ash (FA1) was a waste in bituminous coal combustion, the second one (FA2) came from co-combustion of bituminous coal and biomass. The proportion of biomass in the fuel was 10% by total mass of fuel. The particles of fly ashes from co-combustion of bituminous coal and biomass were less spherical and glassy than those of fly ashes from bituminous coal combustion. The physical properties of fly ashes are presented in Table 2. The chemical analysis of fly ashes is shown in Table 3.

The Blaine specific surface area of FA2 is significantly lower than that of FA1 (Table 2). It is attributed to amount of fly ash particles that passed through a 45-μm sieve. For FA2, the amount of these fly ashes particles is greater by 76.3% as compared to that found in FA1. The mean particle size of FA1 and FA2 is 33.2 and 48.1 μm, respectively. The difference of density value of fly ashes does not exceed 4% (Table 2).

The fly ashes represent typical composition of class F fly ashes according to ASTM C618 [28] (Table 3). The total content of SiO₂, Al₂O₃ and Fe₂O₃ is more than 70%. For FA2, the content of silica, aluminium as well as potassium and sodium is lower, but the content of iron, calcium and magnesium is significantly higher. Because of the free calcium oxide content is lower than 1.0%, FA2 can not be responsible to destructive changes in a concrete structure. The content of magnesium oxide is greater than 4.0% by mass, but the results of autoclave expansion – determined according to ASTM C151 [29] on 40% FA2 plus 60% control cement by mass – show that FA2 is conforming to the requirements for soundness. Expansion of cement containing FA2 is lower than 1.0%. The soluble phosphate content in FA2, calculated as phosphorus pentoxide, is twice higher than that of FA1, but does not exceed 100 mg/kg (Table 3).

The results of X-ray diffraction (Fig. 1) show two crystalline phases in FA1 – quartz (β-SiO₂) and mullite, whereas in FA2 – also periclase and free lime. The FA2 gives higher intensity of quartz peak (26.6°2θ) and consequently smaller intensity of background (14–40°2θ). The decrease in quartz content and increase in SiO₂ content in glass is attributed to change of FA2 glass structure, confirmed by results of IR, MAS-NMR and DTA/DSC investigations [30].

The pozzolanic activity of fly ashes – determined according to ASTM C379 [31] – is 27.2% and 13.7%, respectively, for FA1 and FA2. The lower pozzolanic activity of FA2 is attributed to its coarse grinding as well as to different glass structure, exhibiting higher degree of polymerisation of [SiO₄]⁴⁻ [30,32].

The content of fly ashes in blended cement was 20% and 40% by mass of cement. For cement replacement by fly ash from bituminous coal combustion, mixes are marked as M20FA1 and M40FA1. For cement replacement by coal–biomass fly ash, mixes are marked as M20FA2 and M40FA2.

Table 3
The chemical analysis of fly ashes.

Chemical analysis (wt%)	FA1	FA2
LOI	1.1	0.9
SiO ₂	52.0	48.7
Al ₂ O ₃	28.5	21.2
Fe ₂ O ₃	7.5	10.1
CaO	3.4	8.7
MgO	1.9	5.4
SO ₃	0.9	1.1
Na ₂ O	1.79	1.48
K ₂ O	2.91	2.42
Na ₂ O _e	3.70	3.07
P ₂ O ₅	0.001	0.002
CaO _{free}	–	0.4

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