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ABSTRACT: This paper presents a review of fly-ash as a Supplementary Cementitious Material (SCM) in concrete in terms of its effects on hydration and durability. The climate change agenda has focused the cement and concrete industry on using low embodied CO_2 materials and much effort has been made on incorporating industrial by-products into cement as SCMs. With worldwide cement production (circa 4 billion tonnes) currently accounting for approximately 8% of global CO_2 emissions and 7% of industry energy use, the use of suitable SCMs to partially replace cement in concrete is extremely important. However, while coal-fired power stations are in the decline, due to the need for more sustainable energy generation, there remains stockpiles of fly-ash for potential use as an SCM. This creates opportunities for ashes not previously used in concrete to be studied both in terms of its behaviour during hydration and durability performance in harsh environments. However, these new fly-ash sources need to be studied carefully due to uncertainties about their physical and chemical constituents, reactivity, long term stability and phase relationships and minor elements distribution due to the variability in the source of coal. The work presented includes a review of fly-ash in terms of its effects during cement hydration and contribution to concretes performance in harsh environments from the literature.

KEY WORDS: Cement; Fly ash; Concrete; Cement Hydration; SCM.

1 INTRODUCTION

Concrete is one of the leading construction materials in the world today with approximately 14 billion cubic meters of concrete cast each year, according to the Global Cement and Concrete Association (GCCA) [32]. In Ireland, the consumption of concrete is approximately 15 million cubic meters per annum and according to Aecom, there was a 15% growth in the Irish construction industry in 2021 [1].

However, concrete has a problem [2]. Due to its usage, cement production now contributes to 5-10% of global CO_2 emissions. With the GCCA 2050: Concrete Future introduced, methods to producing carbon neutral construction is set in motion. This policy encourages construction industries and researchers to innovate towards producing low carbon structures. One such action that will accelerate the CO_2 reduction is increasing the clinker substitutions in concrete. Various Supplementary Cementitious Materials (SCMs) such as fly ash, calcined clays, ground granulated blast-furnace slag, and ground limestone can be used to replace the heavy CO_2 emitters [32]. The use of these SCMs, with no additional clinkering process involved, can lead to significant reduction in the CO_2 emissions produced for as less cement will be used in concrete.

One material commonly used to replace cement is Pulverised Fuel Ash (PFA), also known as fly ash (in the USA). Generally considered as a waste material, it is a by-product formed by the combustion of coal. Fly-ash particles tends to be spherical in shape (see Figure 1), ranging in micrometre particle sizes with a relatively high reactivity depending on the chemical composition of the individual particles [3,4]. Fly ash has been used as a SCM in concrete for some time in the UK, Europe, USA and Asia due to the improvements it offers in durability, workability, and higher compressive strengths compared to plain Ordinary Portland Cement (OPC) concretes. According to ASTM standard [5], the three classifications of fly ash, Class C, Class F and Class N which differ through their chemical compositions [5,6]. However, due to its pozzolanic properties, Class C and Class F fly ash are mostly used as cement substitutes with the main difference between them is the Calcium (Ca) content (Class C has a higher Ca content than Class F). Also, Class F has a higher Silica (Si), Alumina (Al) and Iron (Fe) content [7].

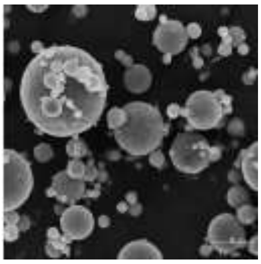


Figure 1. Fly Ash at Low Magnification. [3]

This paper will present a short review of the influence fly-ash has as a SCM during cement hydration and in concrete.

2 FLY ASH IN CEMENT

2.1 Material Properties of Fly Ash

Multiple tests and research have been conducted over the years to obtain the desired amount of fly ash that could be added to concrete to achieve a greater strength than with OPC. The nature and origin of the used coal along with the furnace design and operation contributes to the physical properties of the fly ash produced. The cooling time of the burned material contribute to the crystallization of the fly ash particles. Rapid cooling of the ash attributes to the formation of higher proportions of glassy phases. Tests such as X-ray diffraction (XRD), X-ray fluorescence (XRF) and scanning electron microscopy (SEM) have been used to compile the crystalline phases of fly ash [8]. Minerals such as magnetite, quartz, hematite, mullite and calcite are the most common minerals found in fly ash from bituminous coal.

The chemical aspects of the fly-ash however are characterized by the lower Ca content than plain OPC, which influences the type and volume of solid hydrates formed during hydration, which influence the long-term strength and durability of the concrete. Blending fly ash with OPC decreases the amount of portlandite in the hydrated mixture while increasing the amount of low Ca/Si content C-S-H. In addition, the volume of AFm phases is increased due to the high quantities of Al_2O_3 present in the ash. Addition of further gypsum ($CaSO_4$) can increase the amount of ettringite formed giving a higher total volume of hydrates. When high quantities of Al-rich fly ash are blended with OPC, strätlingite is formed as shown in Figure 2. According to Hanehara *et al*. [10], with a substitution of 60% fly ash, the volume of portlandite becomes so low, self-neutralization may occur. During this phenomenon, the alkalinity of the hardened cement and concrete is lowered due to the calcium hydroxide being excessively consumed by the pozzolanic reaction. Studies [9,10] have shown that after a hydration period of 1 year and longer, the complete depletion of the portlandite was observed, along with a significant decrease in the amount of ettringite precipitated and an increase in the AFm phases with increasing fly ash reaction.

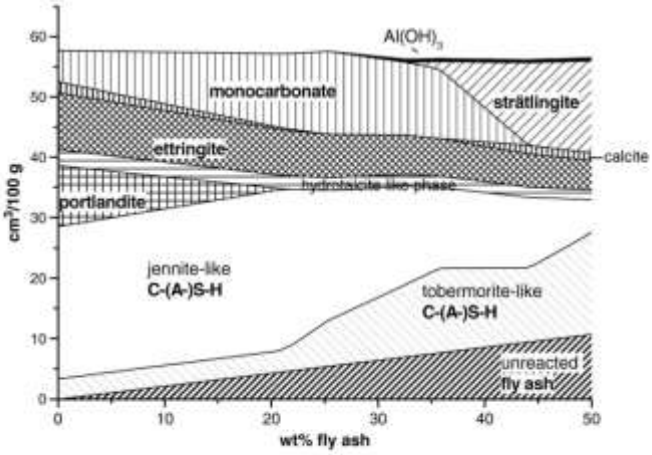
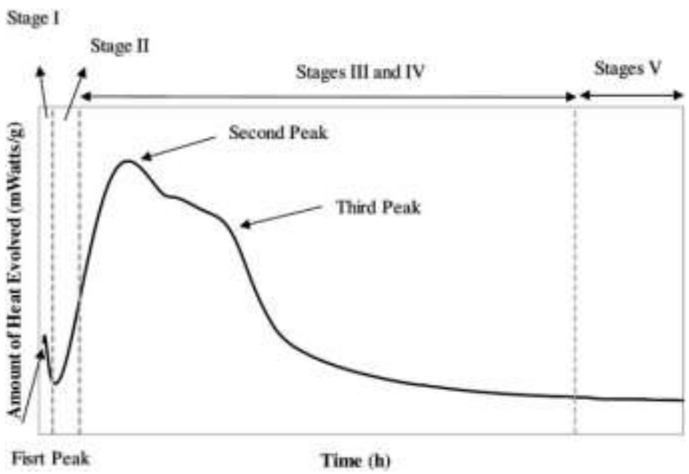


Figure 2. Phase assemblage of hydrated Portland cement upon blending with fly ash [9]

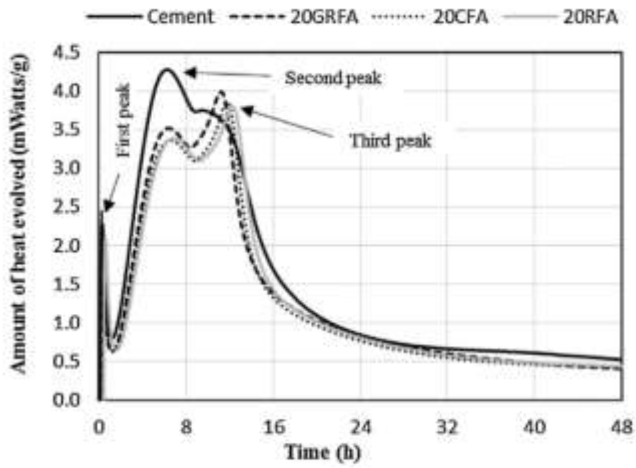
2.2 Heat of Hydration / Cement Hydration

Heat of hydration in cement refers to the exothermic reaction when cement is mixed with water. This production of heat during the curing of concrete has correlation to the composition and fineness of the cement powder, the water to cement (w/c) ratio and cement type. The cement hydration process can be distinguished by five stages as shown in Figure 3a [11].

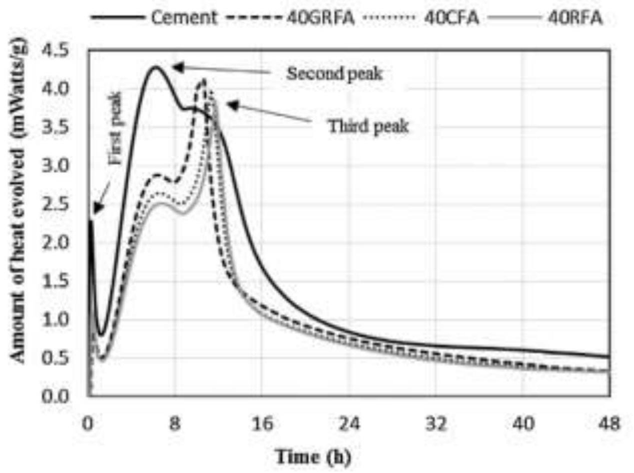
With the addition of fly ash as a replacement to cement, the time taken for the heat of hydration to reach its maximum peak is less compared to plain OPC due to the fineness of the fly ash particles. This is a result of the lower specific surface area and reduced solubility of the aluminosilicate in the fly ash. Figures 3b and 3c illustrates the heat evolution of different types of fly ashes, Classified fly ash (CFA), Run-of-station fly ash (RFA) and Grounded run-of-station fly ash (GRFA), with a



(a)



(b)



(c)

Figure 3. Heat Evolution Curve (a) cement hydration heat evolution curve (b) heat evolution curve for 20% fly ash blended cement paste (c) heat evolution curve for 40% fly ash blended cement paste [13].

substitution level of 20% and 40% in the blended cement paste. Studies have shown [12] that the fineness of the fly ash has a correlation with the peak intensities for the heat of hydration of blended cement.

It has been observed in the literature [14,15] that in blended cements, the intensity of the second peak decreases due to the dilution effect, which occurs when the cement content is reduced as the portion of fly ash increases (Figure 3(b) & (c)). This low pozzolanic activity could also be attributed to the lower solubility of the fly ash in the pore solution. Finally, the third peak was recorded to be more evident for the fly ash blended cement compared to plain OPC. This enhancement of the reaction was found to be due to the fly ash providing more nucleation sites for the calcium aluminates to precipitate, known as the filler effect, thereby increasing the reaction of tricalcium aluminate (C_3A) with the gypsum present in the cement and the conversion of ettringite to monosulfate [14,15].

3 FLY ASH IN CONCRETE

When fly ash is added to concrete as a cement replacement, it has been reported in the literature that its performance compared to plain mixes is improved. This section looks at how the addition of fly-ash improves the performance of concrete in different harsh environments.

3.1 Compressive Strength

The compressive strength of OPC increases rapidly during the early stages. Figure 4 displays the compressive strength of concrete with varying amount of fly ash at 3, 7, 28 and 90 days. As can be seen, the compressive strength of those concretes with fly ash is more gradually increasing over time compared to the plain OPC mixes. At 3 days, all fly ash concretes were found to have a lower compressive strength than the plain cement mixture. As the percentage of fly ash increased, the initial hydration rate of cement is reduced and therefore a mixture with 25% fly ash had the lowest compressive strength compared to others. This can be attributed to the dilution effect of the fly ash, which lowers the relative proportion of cement and dilutes the active component in the blended cement pastes due to the lower pozzolanic activity of the ash early on.

However, as shown in Figure 4, the strength of the fly ash mixture increases over time. Notably, the mixture with 10% and 15% fly ash content were found to acquire a higher strength than the plain cement mixture and the fly ash mixture of 20% and 25% after 90 days of curing with the mixture of 15% attaining the maximum compressive strength at 79.5MPa. From the provided data, it could be seen that all fly ash mixtures had a higher compressive strength compared to plain OPC after 90 days of curing. This development of compressive strength at a later stage is due to the increasing pozzolanic effect over time, set in motion by the calcium hydroxide (CH) formed during the cement hydration. However, with a 20% fly ash content, the compressive strength has notably decreased due to the portlandite in the PFA cement being dissolved at the 20% substitution level. When fly-ash is present, large amounts of C-(A)-S-H gel are formed when the amorphous SiO_2 and Al_2O_3 released during the reaction react with the CH, creating a denser microstructure, improving the compressive strength of fly ash concrete significantly [16,17].

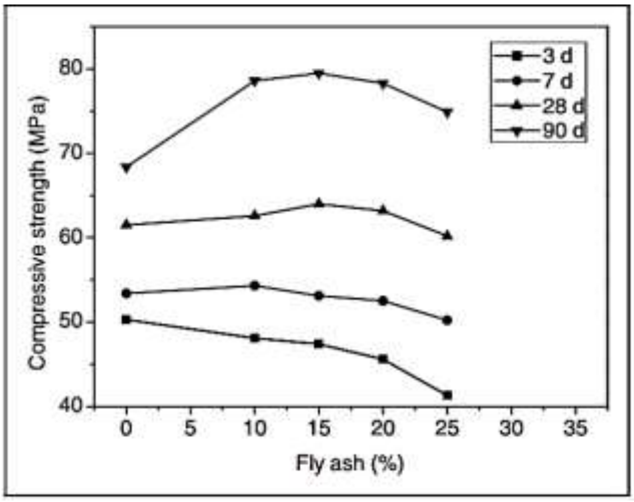


Figure 4. Compressive Strength of Concrete with Varying Fly Ash Content [16].

3.2 Flexural Strength

The flexural strength of concrete has been shown to have a similar strength development. An initial lower flexural strength compared to OPC slowly increases over time, as shown in Figure 5. However, unlike the compressive strength, the sample with 20% substitution of fly ash is shown to have a flexural strength higher than that of OPC. This is also shown in Table 1 [18]. This increase in the flexural strength is due to the fineness of the fly ash and the morphology of the particles as the fine particles fill the pores during the hydration process, producing a denser microstructure.

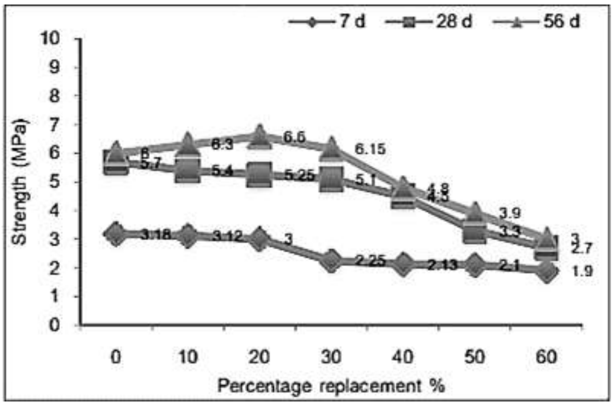


Figure 5. Flexural Strength of Concrete [18].

Table 1. Chemical Properties of Fly Ash and OPC

Beam designation	Fly ash (%)	Flexural strength (MPa)		
		7 d	28 d	56 d
B1	0	3.18	5.70	6.00
B2	10	3.12	5.40	6.30
B3	20	3.00	5.25	6.60
B4	30	2.25	5.10	6.15
B5	40	2.13	4.50	4.80
B6	50	2.10	3.30	3.90
B7	60	1.9	2.70	3.00

3.3 Effect of fly ash on fresh concrete

3.3.1 Workability

The degree of fineness of fly ash can contribute to the workable properties of fresh concrete. The increased fineness of the fly ash particles increases the lubricant effect in compared to cement particles which improves the workability of the blended cement paste. Figure 6 displays the flow of fly ash blended pastes assessed using the mortar flow test according to ASTM C1437 [31] with cement replacement levels of 20% and 40%. It can be seen that the workability of the blended fly ash cement mixture has an increased flow compared to OPC. This is due to the ball bearing effect of the blended cement mixture which is improved because of the spherical shape of the fly ash particles. The glassy and smooth surface of the fly ash reduces their water absorption capacity compared to plain cement particles help to improve the flow of the mixture [14].

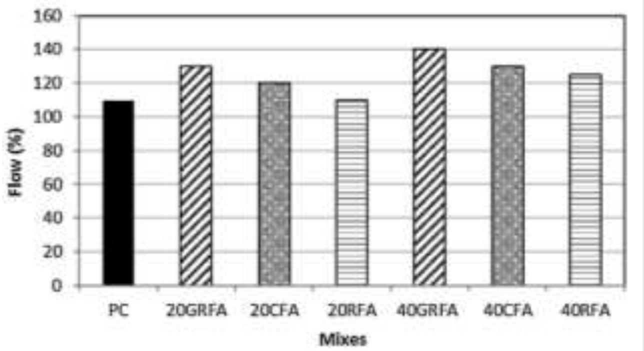


Figure 6. Flow rate of fly ash cement with varying fineness and cement replacement levels [14].

Slump tests by Mahajan *et al* [19] noted that the round spherical shape of the fly ash improves the workability. Concrete produced with fly ash has been shown to have improved workability compared to OPC concrete due to the improved cohesiveness of the fly ash mixture [20]. Figure 7 shows with increasing fly ash content, the concrete slump also increases, thereby reducing the water demand of the blended concrete paste for workability purposes [21].

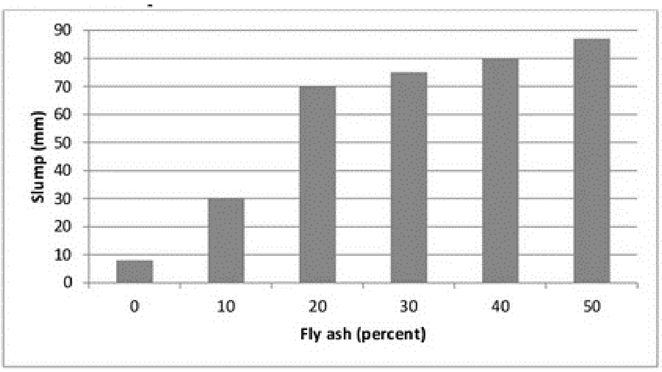


Figure 7. Slump for various fly ash replacement levels in concrete mix [21].

3.3.2 Bleeding

Fly ash in concrete has less bleeding and segregation compared to OPC due to the reduced water demand during hydration. High fly ash content in concrete with low water content can reduce the effect of bleeding on concrete to a point of practically eliminating it. Gebler *et al* [29] noted reduced bleeding was a result of the lower water requirement of fly ash mortar. Therefore, during the production of concrete containing PFA, the water usage in the paste can be reduced according to the rate of replacement of cement to fly ash [20,22,29].

3.3.3 Setting time

The use of fly ash in the concrete mixture can retard the setting time of freshly poured concrete, depending on (1) cement type and content, (2) w/c ratio, (3) the type and amount of chemical admixtures, (4) temperature and (5) the quantity and composition of the fly ash. The setting time of PFA concrete is controlled by the chemical reactions occurring between the cement compounds. Silicates and aluminates present in reacted PFA cement mixture tends to dominate during the early stages of hydration which contributes to the setting and early strength development. This is due to the silicates and aluminates being the predominant component responsible for the formation of the principal product of hydration, C-A-S-H.

The climate of the region affects the setting time of the concrete as hot climate regions experience a reduced retardation which allows more time to place and complete the pouring of the concrete. However, during cold weather, the use of fly ash can lead to significant delays, especially with high levels of replacement. Due to this delay, practical consideration could lead to a limited use of fly ash in the concrete mixture to reduce the difficulties with regards to finishing operations. The use of admixtures can partially or wholly decrease the setting time. Ref [22] found that Class C fly ash generally delays the setting time compared to Class F and concluded that the increased hydraulic reactivity of the fly ash, as well as the calcium content, was the main reason why [20, 23, 24].

3.4 Durability

3.4.1 Sulfate resistance

Sulfate attack, a common cause of deterioration, can occur when concrete is located in environments with high ionic strengths, as found in some soils, groundwater and seawater. Sulfate attack tends to soften the concrete and may lead to expansion, cracking and loss of strength due to the re-formation of gypsum, ettringite and thaumasite at high sulfate concentrations through the reactions with calcium hydroxide and calcium aluminate hydrate (Figure 8) [25,26].

Following a number of studies conducted on Class C and Class F fly ashes, it was demonstrated that sufficient use of low-calcium Class F fly ash contributed to the increased resistance of concrete to sulfate attack and high-calcium Class C fly ashes were found not as effective. Indeed, Class C may have a tendency to increase the rate and extent of the sulfate attack.

3.4.2 Carbonation

The process where CO_2 from the air penetrates into the concrete which reacts with calcium hydroxide, calcium silicate

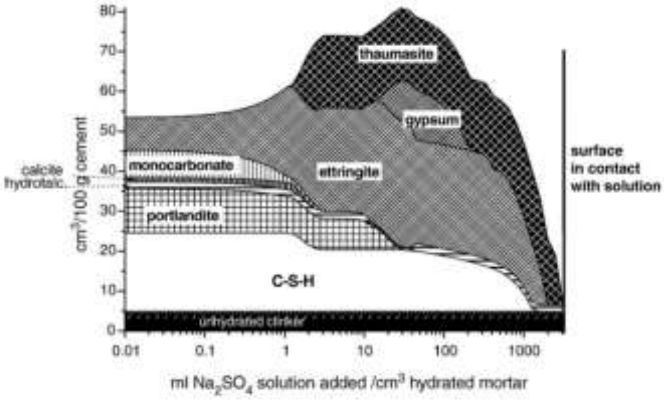


Figure 8. Calculated phase assemblage of a mortar sample immersed a Na_2SO_4 solution [30].

and aluminates, to form carbonates is known as carbonation of concrete. Carbonation reduces the alkalinity of the mixture, which leads to the corrosion of the embedded steels [22]. As the pH falls below ~10/9, hydration of the CO_2 occurs resulting in the formation of carbonic acid (H_2CO_3) which speciates into the HCO_3^- and CO_3^{2-} ions formed by the rapid dissolution of the CO_2 into the alkaline pore solution, leading to the corrosion of concrete [27]. Fly ash helps to decrease the permeability of the concrete thereby increasing its ability to resist cracking. It has been documented through various studies by multiple researchers [27,33,34,35] that fly-ash blended concrete exhibited a reduced likelihood of carbonation than of OPC mixes.

3.4.3 Chloride Resistance

Fly ash reduces the penetration of chlorides and cations due to the changes in the pore structure and the chloride binding capacity of the cement paste [28]. The chloride ions present in the concrete attacks the protective oxide film formed around the reinforcement steel in the initial high alkaline chemical environment. Fly ash reduces the chloride-ion ingress through the binding and immobilization of the chloride ions which is achieved due to the much higher quantities of active alumina present in the fly ash compared to OPC. [28]

4 RESEARCH

Research is ongoing by the authors to determine the optimum cement replacement level of conditioned pulverised fuel ash in concrete using the current stockpile deposited at the ESB Moneypoint power station in Co. Clare. This work will include a comprehensive thermodynamic cement hydration study using state of the art models and experimental techniques, assess the improved durability performance in concretes containing PFA by undertaking a suite of non-destructive tests in three exposure environments and confirm the environmental benefits of it as a SCM.

5 CONCLUSIONS

The use of fly ash as SCMs with OPC enhances the properties of plain concrete along with reducing the CO_2 emissions of cementitious materials through the avoidance of the clinkering process while utilizing the by-product formed by industrial manufacturing process. The optimum level of fly ash for the blended mixture varies on (1) the chemical and physical properties of the fly ash, (2) the proportion of fly ash along with the other materials such as aggregates present in the concrete mixture, (3) the conditions during placing and (4) the exposure conditions subjected to the fly ash concrete. The use of fly ash as a cement replacement substitute increases the overall strength, workability and durability of concrete.

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