



Clinkering-free cementation by fly ash carbonation

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ABSTRACT

The production of ordinary portland cement (OPC) is a CO₂ intensive process. Specifically, OPC clinkering reactions not only require substantial energy in the form of heat, but they also result in the release of CO₂; i.e., from both the decarbonation of limestone and the combustion of fuel to provide heat. To create alternatives to this CO₂ intensive process, this paper demonstrates a new route for clinkering-free cementation by the carbonation of fly ash; i.e., a by-product of coal combustion. It is shown that in moist environments and at sub-boiling temperatures, Ca-rich fly ashes react readily with gas-phase CO₂ to produce robustly cemented solids. After seven days of exposure to vapor-phase CO₂ at 75 °C, such formulations achieve a compressive strength of around 35 MPa and take-up 9% CO₂ (i.e., by mass of fly ash solids). On the other hand, Ca-poor fly ashes due to their reduced alkalinity (i.e., low abundance of mobile Ca- or Mg-species), show limited potential for CO₂ uptake and strength gain—although this deficiency can be somewhat addressed by the provision of supplemental/extrinsic Ca agents. The roles of CO₂ concentration and processing temperature are discussed, and linked to the progress of reactions and the development of microstructure. The outcomes create new pathways for achieving clinkering-free cementation while enabling the beneficial utilization (“upcycling”) of emitted CO₂ and fly ash; i.e., two abundant, but underutilized industrial by-products.

1. Introduction and background

Over the last century, for reasons of its low-cost and the widespread geographical abundance of its raw materials, ordinary portland cement (OPC) concrete has been used as the primary material for the construction of buildings and other infrastructure [1–3]. However, the production of OPC is a highly energy- and CO₂-intensive process. For example, at a production level of 4.2 billion tons annually [4] (equivalent to > 30 billion tons of concrete produced [5]), OPC production accounts for approximately 3% of primary energy use and results in nearly 9% of anthropogenic CO₂ emissions, globally [2]. Such CO₂ release is attributed to factors including: (i) the combustion of fuel required for clinkering the raw materials (i.e., limestone and clay) at 1450 °C [6,7], and, (ii) the release of CO₂ during the calcination of limestone in the cement kiln [2,7]. As a result, around 0.9 tons of CO₂

are emitted per ton of OPC produced [8]. Therefore, there is great need to reduce the CO₂ footprint of cement, and secure alternative solutions for ‘cementation’ as required for building and infrastructure construction.

Furthermore, there exist unique challenges associated with the production of electricity using coal (or natural gas) as the fuel source. For example, coal power is not only associated with significant CO₂ emissions (i.e., 30% of anthropogenic CO₂ emissions worldwide [9]), but also results in the accumulation of significant quantities of solid wastes such as fly ash (600 million tons annually worldwide [10]). While considerable efforts have been made to replace OPC in the binder fraction of concrete by supplementary cementitious materials (SCMs) such as fly ash, the extent of such utilization remains limited. For example, in the U.S., only around 45% of all fly ash produced annually is beneficially utilized to partially replace in the concrete [11]. In spite of

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supportive frameworks [12], such limited use is due to factors including: (i) the presence of impurities including air-pollution control (APC) residues and unburnt carbon as a result of which some fly ashes are non-compliant (e.g., as per ASTM C618 [13]) for use in traditional OPC concrete, due to durability concerns [14,15], and, (ii) increasing cement replacement (i.e., fly ash dosage) levels to greater than 25 mass % is often associated with extended setting times and slow strength gain resulting in reduced constructability of the concrete [14,16].

Clearly, there is an immediate need to valorize or beneficially utilize (“upcycle”) vapor and solid wastes associated with coal power production. However, given the tremendous scale of waste production, there is a need to secure upcycling opportunities of some prominence; e.g., within the construction sector wherein large-scale utilization of upcycled materials can be achieved. This condition could be satisfied if the “upcycled solution” is able to serve as an alternative to OPC (and OPC-concrete) so long as it is able to fulfill the functional and performance requirements of construction. Mineral carbonation (i.e., conversion of vapor phase CO₂ into a carbonaceous mineral, e.g., CaCO₃) has been proposed as a promising route to sequester CO₂ in alkaline solids [17–19]. In such a process, CO₂ is sequestered by the chemical reaction of CO₂ streams with light-metal oxides to form thermodynamically stable carbonates; thus enabling permanent and safe storage of CO₂ [19]. While numerous studies have examined different alkaline waste streams to render cementation solutions—for example, coal combustion residues [20], municipal incinerator wastes [21], and wastes from iron and steel production [22,23]—the low production throughput, or severe operating conditions (i.e., high temperature and elevated CO₂ pressure) [7,21,24,25] have made typical approaches difficult to implement at a practical scale [17]. As such, in this study, two abundant by-products secured from coal-fired power plants (i.e., fly ash and CO₂ borne in flue gas) are utilized to demonstrate a route towards achieving cementation, by the carbonation of fly ash and without any need for clinkering (i.e., the traditional high temperature process of OPC production). It is shown that Ca-rich fly ashes react readily with CO₂ under moist conditions, at atmospheric pressure and at sub-boiling temperatures. The influences of Ca availability in the fly ash, CO₂ concentration, and processing temperature on reaction kinetics and strength gain are discussed. Taken together, the outcomes of this study create new opportunities for the simultaneous valorization of solid wastes and flue gas borne CO₂, within an integrated process.

2. Materials and methods

2.1. Materials

Class C (Ca-rich) and Class F (Ca-poor) fly ashes compliant with ASTM C618 [13] were used. An ASTM C150 [26] compliant Type I/II ordinary portland cement (OPC) was used as a cementation reference. The bulk oxide compositions of the fly ashes and OPC as determined by X-ray fluorescence (XRF) are shown in Table 1. The crystalline compositions of the Ca-rich and Ca-poor fly ashes as determined using X-ray diffraction (XRD) are shown in Table 2. It should be noted that these two fly ashes were used since they represent typical Ca-rich and Ca-poor variants in the U.S., and since Ca content is expected to strongly influence the extent of CO₂ uptake and strength development of carbonated fly ash formulations.

2.2. Experimental methods

2.2.1. Particle size distribution and specific surface area

The particle size distribution (PSD) of OPC was measured using static light scattering (SLS) using a Beckman Coulter LS13-320 particle sizing apparatus fitted with a 750 nm light source. The solid was dispersed into primary particles via ultrasonication in isopropanol (IPA), which was also used as the carrier fluid. The complex refractive index of OPC was taken as $1.70 + 0.10i$ [28]. The uncertainty in the PSD was

Table 1

The simple oxide composition of the fly ashes and OPC as determined using X-ray fluorescence (XRF) [27].

Simple Oxide	Mass (%)		
	Ca-rich Fly Ash	Ca-poor Fly Ash	Type I/II OPC
SiO ₂	35.44	53.97	20.57
Al ₂ O ₃	17.40	20.45	5.19
Fe ₂ O ₃	7.15	5.62	3.44
SO ₃	2.34	0.52	2.63
CaO	26.45	12.71	65.99
Na ₂ O	1.90	0.57	0.17
MgO	5.73	2.84	1.37
K ₂ O	0.53	1.11	0.31
P ₂ O ₅	0.95	0.30	0.08
TiO ₂	1.19	1.29	0.26
Density (kg/m ³)	2760	2470	3150
Specific surface area (SSA, m ² /kg) ^a	4292.6	616.8	442.6

^a The surface area of the Ca-rich (Class C) fly ash is significantly overestimated by N₂ adsorption due to the presence of unburnt carbon [27]. However, based on kinetic analysis of reaction rates in OPC + fly ash + water systems, it can be inferred that the reactive surface areas of both the Ca-rich and Ca-poor fly ashes are similar to each other, and that of OPC. Further discussion regarding the surface areas of these materials can be found elsewhere [27].

Table 2

The mineralogical composition of the fly ashes and OPC as determined using quantitative X-ray diffraction (XRD) and Rietveld refinement [27].

Composition	Mass %		
	Ca-rich Fly Ash	Ca-poor Fly Ash	Type I/II OPC
Lime (CaO)	1.16	–	0.5
Periclase (MgO)	3.81	0.30	–
Quartz (SiO ₂)	10.06	16.64	–
Calcite (CaCO ₃)	0	0	4.60
Mullite (Al ₆ Si ₂ O ₁₃)	0.86	5.08	–
Anhydrite (CaSO ₄)	2.80	0.97	1.2
Gypsum (CaSO ₄ ·2H ₂ O)	–	–	1.1
Magnetite (Fe ₃ O ₄)	1.66	1.76	–
Merwinite (Ca ₃ Mg(SiO ₄) ₂)	6.98	–	–
Gehlenite (Ca ₂ Al ₂ SiO ₇)	4.45	–	–
Ca ₂ SiO ₄ (C ₂ S)	4.93	–	18.00
Ca ₄ Al ₂ Fe ₂ O ₁₀ (C ₄ AF)	–	–	11.40
Ca ₃ Al ₂ O ₆ (C ₃ A)	8.03	–	6.30
Ca ₃ SiO ₅ (C ₃ S)	–	–	56.50
Amorphous	55.26	75.25	–

around 6% based on six replicate measurements. From the PSD, the specific surface area (SSA, units of m²/kg) of OPC was calculated by factoring in its density of 3150 kg/m³, whereas the SSAs of the fly ashes were determined by N₂-BET measurements, as previously reported (see Table 1) [27].

2.2.2. Carbonation processing

Cementitious formulations are processed in the form of slurries, i.e., mixtures of solids (discrete particles) in water (continuous phase) [14]. To maintain consistency with established methods of processing cementing materials, slurries of fly ash in deionized (DI) water (i.e., fly ash pastes) were formulated using a planetary mixer at a water-to-solids mass ratio of 0.20 (w/s = 0.20). The fly ash pastes offered sufficient fluidity such that they could be poured—following ASTM C192 [29]. The pastes were then cast into molds to prepare cubic specimens with a dimension of 50 mm on each side. Following 2 h of curing in the molds at temperature, T = 45 ± 0.2 °C and relative humidity, RH = 50 ± 1%, the specimens were demolded after which on account of evaporation they featured a reduced water content, i.e., w/s = 0.15, but were able to hold form; that is, they were “shape stabilized”. At this

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