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Using CO2-Embedded Coal and MSWI Fly Ashes as Sustainable Alternative SCMs: A Feasibility Study

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Using CO_2-Embedded Coal and MSWI Fly Ashes as Sustainable Alternative SCMs: A Feasibility Study

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Abstract . This study explores using carbonated coal fly ash (CCFA) and municipal solid waste incineration (MSWI) fly ash (CMFA) as supplementary cementitious materials. Carbonation treatment typically increase surface area and enable remarkable CO_2 uptake, with CMFA showing improved reactivity compared to uncarbonated ashes. Compressive strength tests revealed that CMFA outperformed uncarbonated MFA, while CCFA-incorporated mortar showed lower strength than uncarbonated counterpart. All blends consisting of Portland-limestone cement and carbonated ashes appear to meet the requirements of ASTM C1157. It suggests carbonated coal and MSWI fly ashes are viable alternative SCMs for sustainable concrete production.

Keywords: Fly ash, Carbonation, Supplementary cementitious materials (SCMs).

1 Introduction

Global warming poses severe environmental risks, necessitating a 50%–60% reduction in emissions by 2050 [1]. Simultaneously, municipal solid waste management remains a significant challenge, with the U.S. generating 292.4 million tons annually [2]. While incineration reduces waste volume and recovers energy, it produces hazardous residues requiring costly treatment before landfilling. MSWI ashes, rich in cementitious compounds, show potential as supplementary cementitious materials (SCMs) [3]. However, challenges such as heavy metal leaching and chloride content limit their application. Carbonation is a process that permanently lock CO_2 into stable carbonates emerges as a promising solution, immobilizing heavy metals and enhancing SCM properties [4]. Nikulshina et al. [5] demonstrated that introducing water significantly enhances carbonation rates by dissolving CO_2 and forming $Ca(OH)_2$, which reacts with calcium compounds in MSWI fly ash. Similarly, coal fly ash with high calcium content exhibits strong carbonation potential. This study uniquely compares the carbonation performance of MSWI fly ash (MFA) and coal fly ash (CFA) while assessing their effectiveness as SCMs. By analyzing the performance of both carbonated and non-carbonated ashes, this study provides critical insights into carbonation process and its influence on SCM potential.

2 Materials and methods

The raw powder materials of CFA, MFA, and PLC have particle sizes (D50) of 14 μm, 40 μm, and 17.4 μm, respectively (**Fig. 1 (a)**). The pH of MFA and CFA was measured in a 1:10 water-ash mixture, yielding values of 11.9 and 10.3, respectively. As shown in **Table 1**, CFA primarily contained SiO_2 (36.7%) and CaO (29.1%), while MFA was rich in CaO (41%) but had notable impurities, including Cl (6.6%) and SO_3 (5.5%). Both carbonated CCFA and CMFA were used to replace 20% of cement in 2-inch cubic mortar samples, with uncarbonated CFA and MFA for comparison. Mortars were mixed with sand at a 1:2.75 ratio, followed by a fixed water-to-binder ratio of 0.485, and tested for compressive strength. Thermogravimetric analysis and surface area measurements of ashes were conducted.

Table. 1. Oxide composition of CFA, MFA and PLC

Oxide	CaO	SiO_2	Al_2O_3	Fe_2O_3	MgO	Cl	SO_3	Na_2O	P_2O_5	K_2O	TiO_2	ZnO	LOI
MFA	40.88	9.42	5.16	1.37	2.26	6.55	5.53	0.65	1.25	1.46	1.15	1.29	22.87
CFA	29.07	36.74	17.8	6.11	3.41	-	1.12	1.12	0.81	0.76	1.19	-	1.5
PLC	64.8	18.06	3.68	3.33	1.71	0.06	2.16	-	0.06	0.69	0.16	0.05	5.20

3 Carbonation-treatment of ashes

Dry ash was mixed with deionized water at a water-to-solid ratio of 10 mL/g, stirred at 500 rpm, and subjected to pressured CO_2 at 0.2 MPa for 24 hours until pH stabilization for maximum carbonation. The samples were then filtered, dried, and ground. Carbonation increased specific surface area of CFA (from 3.11 m^2/g to 9.63 m^2/g) and MFA (from 15.05 m^2/g to 32.82 m^2/g), which can be attributed to nanoscale carbonate deposits forming on the grain surfaces. Despite MFA having a higher CO_2 uptake, the relatively smaller net change in surface area compared to CFA is due to the formation of carbonates filling some pores.

TGA revealed that CCFA and CMFA contained 29.3% and 60.9% $CaCO_3$, respectively (**Fig. 1 (b)**). CFA and MFA achieved CO_2 uptakes of approximately 3.5 mmol/g and 8 mmol/g, respectively, with both reaching an 80% carbonation degree, where CFA's lower uptake is linked to its reduced CaO content. Weight losses beyond 800°C were attributed to silicates decomposition; and their reductions reflected the transformation of unstable compounds into stable calcium carbonates during carbonation.

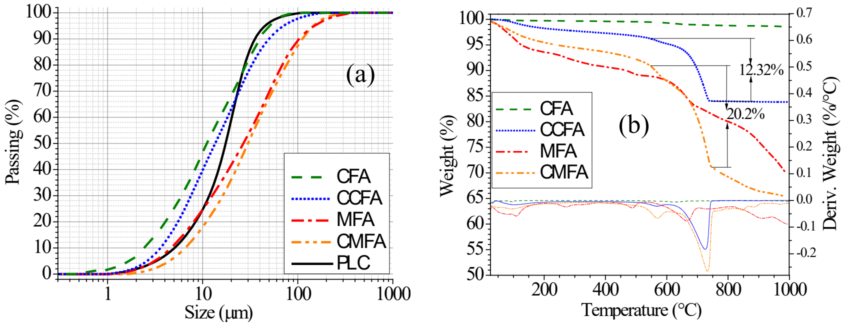

Fig. 1. (a) Cumulative particle size distributions and **(b)** TGA results of raw and carbonated CFA and MFA

4 Performance of CO_2-embeded ashes as alternative SCMs

Fig. 2a indicates the paste incorporated with 20CFA (20 wt% replacement of cement by CFA) reaching an early peak while the one with 20MFA showed a delayed initial peak due to its lower reactivity. 20CCFA demonstrated a delayed hydration start, likely due to stable carbonates hindering the initial reactions. The heat flow curve for 20CFA exhibited a single peak, which is indicative of a rapid and continuous reaction, while 20MFA showed two peaks, indicating a more complex hydration mechanism involving both silicate and aluminate phases.

As shown in **Fig. 2b**, the 7-day strength for the mortar with 20CFA was slightly lower than the control, but the 28-day strength became higher due to secondary pozzolanic reactions. However, the mortar with 20CCFA showed lower strength at both 7 and 28 days due to stable carbonate formation hindering hydration. In comparison with CFA, 20MFA exhibited lower reactivity than the control group, while after carbonation, 20CMFA demonstrated higher strength, benefiting from nano carbonates acting as nucleation sites and enhancing microstructure and hydration.

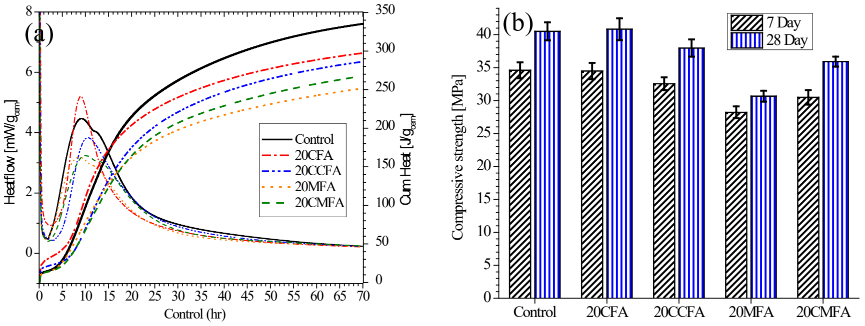

Fig. 2. (a) Calorimetric heat curves of pastes and **(b)** compressive strength of mortars with 20% replacement of cement

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