

Carbonation of composite cements with high mineral admixture content used for radioactive waste encapsulation



H. Kinoshita^{a,*}, C. Circhirillo^b, I. SanMartin^c, C.A. Utton^a, P.H.R. Borges^d, C.J. Lynsdale^e, N.B. Milestone^a

^a Department of Materials Science and Engineering, The University of Sheffield, Sheffield, UK

^b Materials Science and Engineering, National Institute Applied Science, Lyon, France

^c Department of Engineering, Public University of Navarra, Pamplona, Spain

^d Department of Civil Engineering, Federal Centre of Technology Education of Minas Gerais – CEFET/MG, Belo Horizonte, Brazil

^e Department of Civil Engineering, The University of Sheffield, Sheffield, UK

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ABSTRACT

Carbonation of ordinary Portland cement occurs naturally. This process is, however, not sufficient for the application of CO₂ sequestration due to the very slow kinetics of the diffusion-controlled process. The present study shows that the carbonation can be enhanced in the hardened cement systems blended with blast furnace slag or pulverised fuel ash under the condition tested. Both blended systems, with a very high replacement level of Portland cement with slag or fly ash, indicated nearly two times of CO₂ intake compared to the pure Portland cement system in the particle form, and the system with blast furnace slag gained significantly more CO₂ than other samples in the powdered form. The carbonation of the slag containing particles appeared to result in a significant release of H₂O, which may have caused the coarse pore structure due to the shrinkage of C–S–H. This seems to be a very attractive system to capture CO₂ as the sufficient level of porosity and free water could be maintained in the system. The lower Ca²⁺ concentration in the system appeared to favour the formation of vaterite as the carbonation product.

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1. Introduction

Climate change due to carbon dioxide (CO₂) emission is a serious concern in modern society (Metz et al., 2005). To minimise the impact, it is necessary to stabilise the CO₂ concentrations in the atmosphere. In recent years, cementitious waste materials have been considered to be potential materials for CO₂ sequestration via mineral carbonation (Lackner et al., 1995) owing to their high concentration of calcium oxide (CaO) (Fernández Bertos et al., 2004; Iizuka et al., 2004; Stolaroff et al., 2005). The CO₂ sequestration by the carbonation of such materials would have an advantage in accessibility, as the cementitious waste materials are available without mining efforts. It is especially beneficial for the cement industry, which contributes approximately 6% of man-made CO₂ emission in the world (Metz et al., 2005), as they could recover the CO₂ they produced using their own waste materials.

The cement industry has been making continuous efforts to reduce their CO₂ emission. For example in the UK, they have reduced the CO₂ emission over 29% between 1990 and 2006 by reducing the fossil fuel consumption through the use of waste-derived alternatives for kiln fuels and virgin raw materials (British Cement Association, 2007). Although this effort will continue and they will

make further progress, there will be a limitation in the reduction of CO₂ emission as approximately half of the emission is originated from the decarbonation of limestone (CaCO₃), one of the key components of raw materials (World Business Council for Sustainable Development, 2012). In order to further reduce the net CO₂ emission in the current process, the CO₂ generated from the decarbonation of limestone has to be recovered, and the recovery of CO₂ by cementitious materials can offer a good option.

The carbonation of cementitious materials occurs naturally. However, the overall natural carbonation of cementitious materials is usually limited due to the very slow kinetics of the diffusion-controlled process (Fernández Bertos et al., 2004). To increase the extent of carbonation, different reaction conditions have been investigated e.g., a high pressure (Reardon et al., 1989), a vacuum environment (Venhuis and Reardon, 2001) and use of supercritical CO₂ (Short et al., 2001). On the other hand, it has been reported that cements and concretes can carbonate more readily when mineral admixtures such as blast furnace slag (BFS) or pulverised fuel ash (PFA) are incorporated at a high replacement level (Ngala and Page, 1997; Osborne, 1999; Gruyaert et al., 2013). Materials of this type can be beneficial in terms of CO₂ reduction because such materials may be able to contribute to the CO₂ reduction in two ways: reducing the use of ordinary Portland cement (OPC) and sequestering CO₂ during their life time as products and/or after becoming wastes.

* Corresponding author. Tel.: +44 114 222 5930.

E-mail address: h.kinoshita@sheffield.ac.uk (H. Kinoshita).

The mechanisms of the enhanced carbonation in these blended cement systems have not been well understood due to the complex nature of the systems. Hydrated cements and concretes can contain various anhydrous and hydrated phases originated from OPC in addition to the water, admixtures and aggregates (Sharp et al., 2003) which may or may not react with the OPC. Borges et al. (2010) recently investigated the carbonation behaviour of blended cement systems used for radioactive waste encapsulation, and provided the quantified data on carbonation of portlandite ($\text{Ca}(\text{OH})_2$) and calcium silicate hydrate (C–S–H) gel in the system. One of the advantages of studying such systems is that they usually contain a significant amount of BFS or PFA with no other additives, resulting in relatively simpler systems. For example, the systems studied by Borges et al. have a very high replacement level of OPC with BFS or PFA, but contain no other aggregates such as sand, which would make the effects of admixtures more prominent.

For the investigation of carbonation behaviour of the cement systems with a high replacement level of OPC with BFS or PFA, the age of the testing materials is important factor. The systems with a high BFS or PFA content have hydration behaviours different from that of neat OPC system due to the latent-hydraulic and pozzolanic nature of BFS and PFA, respectively (Robeyst et al., 2008; Baert et al., 2008). This usually result in a slower development of microstructure with different product phases due to the pozzolanic reaction (Baert et al., 2008) in which silica content in the system react with the portlandite ($\text{Ca}(\text{OH})_2$) to form calcium silicate hydrate known as C–S–H gel. The overall hydration kinetics and the C–S–H product can differ depending on the amount and types of admixture (Robeyst et al., 2008; Wang and Scrivener, 1995). The importance of the age of testing materials in the study of carbonation has been recently pointed out by Gruyaert et al. (2013). According to their data on the cement samples containing BFS, the rate of carbonation ($\text{mm}/\text{day}^{1/2}$) was significantly different in the younger samples of 1–3 month old and became stable in the older samples after 6–18 months of curing.

In the present study, the carbonation behaviour of hardened cements commonly used for radioactive waste encapsulation in the UK (Milestone, 2006; Borges et al., 2010) i.e. blends of OPC with BFS or PFA was investigated. As already mentioned, these materials have a very high replacement level of OPC with BFS or PFA, but contain no other aggregates such as sand, which would make the effects of admixtures more prominent. Building on to the previous studies (Borges et al., 2010, 2012), the present study aims to obtain further insight into the carbonation of these materials, but after curing period of 15 months which is much longer than the 3 months of curing period previously studied by Borges et al. The study was conducted also with emphasis on the role of water in the system, which has not been studied previously.

2. Materials and methods

Table 1 summarises the experimental conditions used in the present work. Based on the formulation used by Borges et al. (2010), a neat OPC grout and a blend of BFS and OPC (BFS:OPC = 9:1) were prepared with a water to solids (w/s) ratio of 0.33. Similarly, a

Table 2
Composition of raw materials.

	CaO	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	SO ₃	MgO	K ₂ O	Na ₂ O	LOI ^a
OPC	64.58	20.96	5.24	2.61	2.46	2.09	0.59	0.28	0.73
BFS	42.10	34.50	13.74	0.97	^b	7.29	0.49	0.22	1.05
PFA	1.62	49.53	26.45	8.70	0.88	1.56	4.58	<0.01	4.10

^a Loss of ignition.

^b Sample contains reduced sulphur.

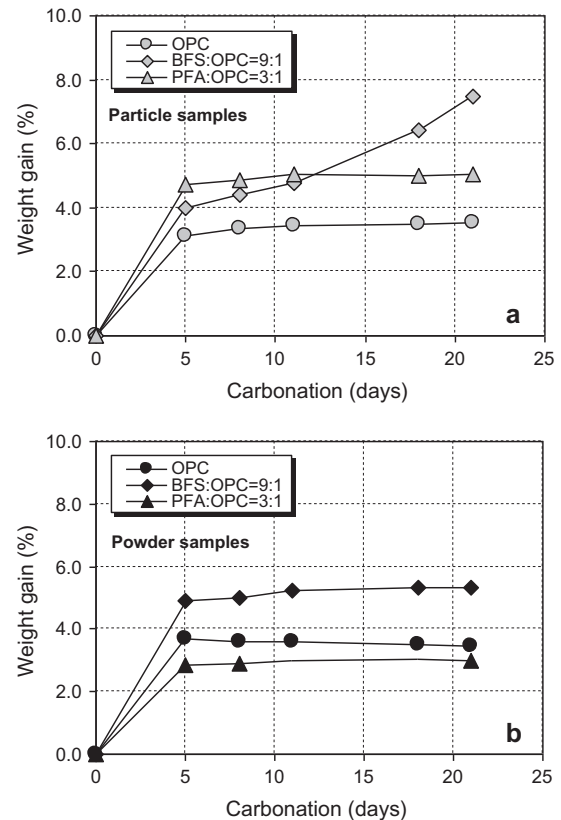


Fig. 1. Weight gain of samples during carbonation period: (a) particle samples and (b) powder samples.

w/s ratio of 0.42 was used for the blend of PFA and OPC (PFA:OPC = 3:1). The grouts were moulded in prisms of $25 \times 25 \times 285$ mm and cured at 40°C with 100% relative humidity for 15 months to assure the sufficient hydration of cement samples. The cured samples were crushed and sieved into either particles of 3.36–5.00 mm or powder of $<63 \mu\text{m}$ prior to the carbonation. A set of cylindrical samples with 25 mm in diameter for these formulations were also used to estimate the apparent porosity of the samples by water absorption of the specimens under vacuum. The specimens were oven-dried at 50°C over night, and the porosity was obtained as the volume percentage of absorbed water in the bulk volume of specimens estimated based on the dimensions. Three samples were used for each formulation to determine the average values. The chemical compositions of the raw materials, OPC, BFS and PFA used in the present study are shown in Table 2.

The carbonation was performed in a chamber filled with 15% CO_2 at $25\text{--}30^\circ\text{C}$ with 50–60% relative humidity under atmospheric pressure for 21 days. The carbonation condition is based on the previous studies (Borges et al., 2010, 2012) with an increased CO_2 concentration from 5% to promote more carbonation. The weight of samples was periodically measured during the

Table 1
Experimental details.

Formulation	w/s ratio	Curing condition	Size	Carbonation condition
OPC	0.33	40°C	Particles:	$25\text{--}30^\circ\text{C}$
BFS:OPC = 9:1	0.33	100% R.H.	3.36–5.00 mm	50–60% R.H.
PFA:OPC = 3:1	0.42	15 months	Powder:	15% CO_2 conc.
			$<63 \mu\text{m}$	21 days

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