

Using Fly Ash to Achieve Low Embodied CO₂ Concrete

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Abstract

Twenty-first century design is increasingly being influenced by the specification of carbon-critical materials. The use of fly ash (FA) will, therefore, be an ever more important contributor to this requirement. This paper describes research carried out, aimed at developing practical guidance on how to exploit FA to achieve low embodied CO₂ in concrete susceptible to corrosion induced by chloride ingress and carbonation. Data is presented for concretes with, (i) high FA contents, (ii) FA ternary cement combinations and (iii) coarse FA materials. The initial part of the paper establishes the effect that these have on the durability properties, making comparisons with those of reference Portland cement and 'conventional' FA concretes. It is shown that care has to be taken to adjust the concrete mix constituent proportions in order to match the properties of the references mixes. The paper quantifies the embodied CO₂ of the concretes, using recently agreed UK figures for the various constituent materials. It is also shown that in some cases there is a need to optimise and balance embodied CO₂ and durability performance.

Keywords: Embodied CO₂, high fly ash contents, fly ash ternary cement combinations, coarse fly ash, durability performance, material selection.

1 Introduction

There is world-wide concern with regard to climate change and little doubt that Greenhouse gases, in particular nitrous oxides and carbon dioxide, are explicitly involved in this process. Whilst there is disagreement over whether anthropogenic or natural carbon dioxide is involved, it would be ill-advised to ignore the role of the former, when simple solutions can be adopted towards reducing this. Portland cement (PC) production makes a significant contribution to anthropogenic CO₂ output and has been increasing globally. For over 70 years, the benefits of fly ash (FA) in concrete have been demonstrated and although the origins of its use have been based on technical performance requirements, e.g. reduced heat [1], its effect has always been to reduce the embodied CO₂ of concrete mixes.

The levels of embodied CO₂ in the production of concrete constituent materials have recently been enumerated in the UK, with the values agreed given in Table 1 [2-5]. Given this, it is now possible to control the level of CO₂ in a wide-range of concrete products from RC structural elements, through to

aerated concrete blocks. However, it is important that this is carried out in a rational and objective manner, through which the technical performance requirements for the applications are maintained. It is important to recognise that where several performance requirements apply, a balance of the constituents in concrete mixes may be required and even the construction process adjusted to meet them and best utilise the benefits of FA. In other words, modification beyond straight replacement of FA may be necessary. This paper addresses these issues in more detail by examining the resistance of fly ash concrete to chloride ingress and carbonation, both directly and in terms of lowest practicable embodied CO₂. Data is presented from studies considering, (i) high FA contents, (ii) FA ternary cement combinations and (iii) coarse FA materials.

2 High FA Content Concrete

High FA content in this paper refers to concrete where the material is used as a cement component at levels beyond 30%, commonly adopted in structural concrete [6]. In some situations, concrete mixes containing more than 50% FA by mass of the cementitious material have been used [7]. Furthermore, current European standards allow a maximum FA level of 55% by mass [8, 9]. The results presented here cover mixes with FA up to 45% by mass. One of the issues associated with this is that low early strengths may result, which can have implications for the construction process in terms of formwork striking times.

2.1 Materials and Mix Proportions

The main characteristics of the cements and addition used in this study are given in Reference 6. RHPC was used to control the early strength of concrete with high FA contents to match those containing PC-only. The concretes contained crushed single-size coarse aggregate (maximum size 20mm) and a crushed sand, with a fine filler also included in the PC control mixes. The mix proportions for the concretes are given in Table 2.

The water contents of these were controlled to achieve the target slump range of 60 to 90 mm and the w/c ratio adjusted to give strengths of 35, 50 and 70 N/mm². FA was included with PC at levels of 30 and 45%, with the latter also being combined with RHPC. All of the test concretes were cured in water at 20°C to 28 days, prior to pre-conditioning / testing.

2.2 Durability Performance

Chloride diffusion and accelerated carbonation (4% CO₂, 55% RH) tests were carried out following the methods described by Dhir et al [10, 11].

The coefficient of chloride diffusion for PC, PC/30FA, PC/45FA and RHPC/45FA concretes are given in Fig. 1. The results generally agree with behaviour noted previously for FA concretes, indicating significantly lower diffusion coefficients for these than PC concretes, when compared on an equal strength basis. In addition, the 45% FA content concretes had lower coefficient values than those with 30% FA.

A comparison between the depths of accelerated carbonation for these concretes following 10 and 20 weeks exposure is made in Fig. 2. As indicated, these were generally greater for FA concretes than those of PC at equal strength, with differences tending to increase at higher FA level, for 35 and 50 N/mm² but similar values noted at 70 N/mm².

2.3 Embodied CO₂

The concretes were evaluated for embodied CO₂ of their constituent materials. Table 1 shows the values used in the comparisons, which are mainly based on information given by the UK concrete industry.

Fig. 3 compares the calculated embodied CO₂ of the concretes. The inclusion of FA lead to reductions in this compared to PC and, as might be expected, and the effect increased with FA level. With RHPC used instead of PC, similar values / slight reductions in embodied CO₂ were noted (reflecting minor reductions in RHPC/FA contents in these concretes).

3 FA Ternary Cement Combinations

In this research, various additions, including FA, limestone (LS), metakaolin (MK) and silica fume (SF) were used with PC. The FA content of the concretes ranged from 20 to 55%, covering that given in current standards [8, 9], and the mixes were considered at equal w/c ratio. Interpolation was used to obtain the mix proportions for a given strength and to enable embodied CO₂ to also be compared on this basis.

3.1 Materials and Mix Proportions

The characteristics of the PC and four additions used have been given previously [6]. The concretes contained natural gravel in two fractions (4/10mm and 10/20mm) and a medium grade sand.

The concretes had a free water content of 165 l/m³ with a high-range water-reducing admixture used to give a slump in the range of 60 to 90 mm. The w/c ratios were 0.35, 0.50 and 0.65, selected to cover the practical range. FA was used at levels of 20, 35 and 55% and then 10% of the FA at 35% was replaced with each of the other additions. Curing of concrete was carried out in water at 20°C for the required period, prior to pre-conditioning / testing.

3.2 Durability Performance

Chloride migration tests were carried out following the NT Build 492 method [13]. The effect of the test concretes on non-steady state migration of chloride at 0.50 w/c ratio is shown in Fig. 4. The data indicate that this reduced with increased curing time (28 and 90 days) in all cases.

The FA binary concretes gave lower migration compared to PC and this increased with FA level. The effect of FA on chloride ingress is similar to that shown in Fig. 1, despite differences in the basis of comparison (strength and w/c ratio). The inclusion of second additions, MK, SF or LS, further reduced chloride migration.

The accelerated carbonation depth [11] was found to increase with FA in concrete compared to the PC reference and the effect became greater at higher FA levels. The inclusion of second additions gave increases in carbonation depth compared to the binary concrete, except for LS where there was a slight reduction. The results reflect the effect of the materials on the concrete chemistry and microstructure and suggest the former has the greater influence on the process.

The behaviour for carbonation was different to that noted for chloride migration, which indicates that where more than one processes can occur, a balance would be necessary in material selection to optimise performance.

3.3 Embodied CO₂

In evaluating embodied CO₂, the concretes described above and those of a related study [14] were included and the following approaches to mix proportioning considered:

- a) equal w/c ratio of 0.5 with a fixed water content of 165 l/m³
- b) equal 28 day cube strength with a fixed water content of 165 l/m³
- c) equal 28 day strength with a variable water content and fixed water-reducing admixture content (0.6% by weight of cement plus additions)

These were examined to identify the role that factors associated with concrete mixes and basic properties have on embodied CO₂ and the results are given in Fig.5. The use of FA in concrete, particularly at high levels, gave reductions in embodied CO₂ compared to PC. The rate was influenced by how the concretes were proportioned, with greatest reductions obtained for comparisons at equal w/c. At equal strength, the reduction was less significant with fixed water content (variable water-reducing admixture), than when the water content was allowed to vary (ie fixed water-reducer content).

The inclusion of a second addition gave minor differences, which again depended on the concrete proportions. This however, had a significant reduction in chloride migration (Fig. 4). Therefore, the advantage of using ternary mixes can be seen when the comparison is based on equal chloride resistance.

4 Coarse Fly Ash

The European Standard for FA, BS EN 450-1 [15], permits the use of material up to a fineness of 40% retained on a 45µm sieve. This represents a wider property range than has been traditionally used in the UK for FA as a cement component. Work examining material from single and multiple sources indicates that reductions in strength may occur with increasing fly ash coarseness [16].

In this study, minor adjustments to the concrete mixes through the w/c ratio were considered to take account of the effect of FA fineness and achieve equal strength.

4.1 Materials and Mix Proportions

A PC and four FAs with different fineness (45µm sieve retention = 3.5, 13.5, 27.0 and 35.0%) and similar LOI values (between 3.5 – 5.5%) were used, see Reference 16. The aggregates were similar to those used in the 'high FA content concrete' research (Section 2). The concrete mix constituent proportions, adjusted as indicated above, are given in Table 3. A FA level of 30% was used and curing of the test concretes was as described for the other test series above.

4.2 Durability Performance

The coefficient of chloride diffusion and the depths of accelerated carbonation following 10 and 20 weeks exposure [10, 11] for PC/FA concretes of strength 35 N/mm², with different FA fineness are compared in Fig. 6. The results indicate similar general trends to those noted above between PC and the various FA concretes considered. With small adjustments to the concrete mix constituent proportions to achieve the same strength, the variations in FA fineness between 3.5 to 35.0% gave only minor effects on the durability in terms of chloride diffusion and accelerated carbonation.

4.3 Embodied CO₂

Fig. 7 compares the calculated embodied CO₂ of the concretes. It was assumed for FA that this did not change with the variation in fineness.

Therefore, the four FA concretes essentially gave similar embodied CO₂ at the same strength (with only minor differences in the concrete mix proportions). As with the data shown in Figs. 3 and 5, the inclusion of FA lead to reductions in embodied CO₂ compared to PC and this effect increased with strength.

5 Balancing Durability Performance and Sustainability

With increasing consideration being given to sustainability, optimising materials to meet several requirements is becoming increasingly important. Given the performance effects noted above, different concretes would be appropriate when optimising for chloride exposures, compared to those for carbonation.

In order to demonstrate how the approach to material selection could be carried out, taking account of embodied CO₂, two exposure classes covering corrosion due to chlorides from seawater (XS3) and carbonation (XC3/4) were considered. The minimum requirements for concrete exposed to XS3 (for nominal cover depth 50mm+Δ_c) and XC3/4 conditions (for nominal cover depth 30mm+Δ_c) are given in Table 4 (50 years' service). Options for meeting these based on the 'FA ternary cement combinations' research are considered in the following sections.

5.1 Chloride Exposure Conditions

Table 5 gives compressive strength, chloride resistance and embodied CO₂ for concretes with FA levels up to 55%, and could be used in structures exposed to a marine environment. The minimum requirements for the specification and the 'limiting factors' which are controlling are indicated for each concrete.

The binary concretes indicate that the 55% FA concrete gives the 'best' performance in terms of chloride resistance and embodied CO₂. When ternary combination concretes are used, chloride resistance can be further improved, while embodied CO₂ is similar to the FA binary concretes at the same level. In this case the 45%FA/10%SF concrete had the 'best' performance.

5.2 Carbonation Exposure Conditions

Table 6 makes a similar comparison for carbonation to that for chloride ingress above. This indicates that cement combinations with lower embodied CO₂ had greatest carbonation depths. Thus in the case of carbonation environments, a balance would have to be made, such as reducing the w/c ratio (which could increase cost) and/or increasing the depth of cover (which could affect structural performance).

A weighting factor method [17] is considered as a pragmatic route to balancing durability and embodied CO₂. A weighted objective function, $h(\mathbf{F}(\mathbf{x}))$, can be established as follows:

$$h(\mathbf{F}(\mathbf{x})) = w_{Carb} f_{Carb}(\mathbf{x}) + w_{CO_2} f_{CO_2}(\mathbf{x})$$

where: w_{Carb} is the weighting factor for carbonation performance, $0 \leq w_{Carb} \leq 1$

w_{CO_2} is the weighting factor for embodied CO₂, $0 \leq w_{CO_2} \leq 1$, and $w_{Carb} + w_{CO_2} = 1$

$f_{Carb}(\mathbf{x})$ is the normalised carbonation value
 $f_{CO_2}(\mathbf{x})$ is the normalised embodied CO₂, and
 \mathbf{x} is a set of the cement combinations in consideration.

Assuming equal consideration for carbonation and embodied CO₂, i.e. $w_{Carb} = w_{CO_2} = 0.5$, the minimum h value is obtained with the ternary mix, 45%FA/10%LS, when the figures in Table 6 are considered. This suggests that the ternary cement combinations with LS as a second addition could be an option to balance carbonation resistance and embodied CO₂. It should be noted that this result depends on the weighting factor and if this is changed, depending on specific requirements, a different result would be obtained.

6 Concluding Remarks

As the demands on low carbon and carbon-critical design intensify, so there is an increasing need to look to a range of material options. Given its wide availability and environmental credentials, FA has significant potential in this role. However, with ever greater demands, balancing of these may be necessary to optimise performance.

The research described has highlighted the importance of how FA is used. In general, best performance with the material was obtained when concrete was designed at equal 28 day strength. For example, when this was followed, there was little effect of different FA properties on durability. Similarly, in the case of chloride, increasing FA content, or the use of ternary cements gave enhanced performance, however, the opposite was found for carbonation.

In terms of embodied CO₂, this tended to mirror the material effects notes for chloride ingress, while again, the opposite tended to occur with carbonation. The implication of this is, with increased chloride resistance, good environmental performance may be expected however, with carbonation a balance between this and the environmental requirements would need to be considered in optimising concrete. This may also apply if chloride/carbonation and embodied CO₂ were to be considered.

The work, therefore, provides an indication of how both durability and sustainability can be considered collectively. However, it was noted that some of the concretes extended the time to achieve early strength, particularly with high FA contents, which may have implications for formwork removal and this factor may also need to be considered in the material selection process.

7 Acknowledgements

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Table 1. UK agreed embodied CO₂ in the production of concrete constituent materials [2, 3, 4].

Constituent Material	Embodied CO ₂ , kg CO ₂ /t
Portland cement (PC)	930
Rapid hardening PC	930+0.435*25=941 ^{b)}
Fly ash	4
Silica fume	14
Limestone	32
Metakaolin	330
Aggregate	4
Admixture	220
Water	0.21 ^{c)}

- a) Based on [3] unless otherwise stated
b) Estimated from national average CO₂ emitted per unit of electricity (0.435 kg/kWh) [4]. Assumed typical mill power consumption for RHPC to fineness 490 m²/kg (55 kWh/t) and for PC to 320 m²/kg (30 kWh/t) [5].
c) Based on [4].

Table 2. Mix proportions (for high FA content concrete series)

Mix Code	Design Strength, N/mm ²	Mix Proportions, kg/m ³			
		PC	FA	Water	Aggregate ^{a)}
PC Control	35	305	-	190	1900
	50	385	-	190	1830
	70	510	-	190	1725
PC/30FA	35	240	105	170	1885
	50	290	125	170	1820
	70	360	160	170	1700
PC/45FA	35	200	165	165	1870
	50	270	220	165	1755
	70	350	285	165	1545
RHPC/45FA	35	195	160	170	1875
	50	245	200	170	1795
	70	335	275	170	1640

- a) Total aggregates included crushed coarse (4/20 mm) and fine (0/4mm) aggregates and filler sand (0.15 – 1 mm, only used in PC mixes).

Table 3. Mix proportions (for study of effects of coarse fly ashes)

Mix Code	Design Strength, N/mm ²	Mix Proportions, kg/m ³				w/c ratio	WR Dosage, % of cement
		PC	FA	Water	Aggregate		
PC Control	25	225	-	175	1985	0.77	-
	35	270	-	175	1945	0.65	-
	50	370	-	175	1850	0.47	-
	60	450	-	175	1765	0.38	-
PC /30FA1 (45µm ret. = 3.5%)	25	190	85	165	1950	0.60	0.55
	35	230	95	165	1910	0.51	0.55
	50	295	125	165	1820	0.39	0.66
	60	350	150	165	1745	0.34	0.88
PC /30FA2 (45µm ret. = 13.5%)	25	190	85	160	1970	0.58	0.99
	35	230	95	160	1925	0.49	0.99
	50	295	125	160	1830	0.38	1.10
	60	350	150	160	1755	0.32	1.32
PC /30FA3 (45µm ret. = 27.0%)	25	190	85	160	1970	0.58	0.99
	35	230	95	160	1925	0.49	0.99
	50	295	125	160	1830	0.38	1.10
	60	350	150	160	1755	0.32	1.32
PC /30FA4 (45µm ret. = 35.0%)	25	190	85	155	1980	0.56	1.43
	35	230	95	155	1935	0.47	1.43
	50	295	125	155	1840	0.36	1.65
	60	350	150	155	1765	0.31	1.65

Table 4. Concrete requirements for XS3 (chloride) and XC3/4 (carbonation) exposure classes as specified in BS 8500 for 50 years of design life for different cement combinations.

FA Content, % by Mass	Concrete Requirements		
	Minimum Strength Class	Maximum w/c Ratio	Minimum Cement Content, kg/m ³
<i>Limiting Factors for XS3 Chloride Exposure (nominal cover depth 50mm+Δ_c)</i>			
6-20	C50	0.40	380
21-35	C35	0.50	340
36-55	C30	0.50	340
<i>Limiting Factors for XC3/4 Carbonation Exposure (nominal cover depth 30mm+Δ_c)</i>			
6-35	C35	0.60	280
36-55	C37	0.55	300

Table 5. Comparison of embodied CO₂ of concretes subjected to chloride-induced corrosion (XS3).

Cement Combination	Minimum Concrete Requirements for XS3 Exposure			Concrete Properties		
	Minimum Strength, N/mm ²	Maximum w/c Ratio	Min Cement Content, kg/m ³	28 day Cube Strength, N/mm ²	Chloride ^{a)} Migration, ×10 ⁻¹² m ² /s	Embodied CO ₂ , kg/m ³ Concrete ^{b)}
100%PC	50	0.40 ^{c)}	380	66	28.0	391
PC/20FA	50	0.40 ^{c)}	380	61	23.7	315
PC/35FA	35 ^{c)}	0.50	340	35	27.1	212
PC/55FA	30 ^{c)}	0.50	340	30	20.0	181
PC/25FA/10SF	35	0.50	340 ^{c)}	48	5.4	212
PC/25FA/10LS	35	0.50	340 ^{c)}	40	10.9	212
PC/25FA/10MK	35	0.50	340 ^{c)}	38	9.8	223
PC/45FA/10SF	30	0.50	340 ^{c)}	35	4.9	149
PC/45FA/10LS	30 ^{c)}	0.50	340	30	8.3	170
PC/45FA/10MK	30 ^{c)}	0.50	340	30	7.5	166

a) NT Build 492 test

b) Calculations based on data in Table 1.

c) Limiting factor(s) controlling the specification.

Table 6. Comparison of embodied CO₂ of concretes subjected to carbonation-induced corrosion (XC3/4).

Cement Combination	Minimum Concrete Requirements for XC3/4 Exposure			Concrete Properties		
	Minimum Strength, N/mm ²	Maximum w/c Ratio	Min Cement Content, kg/m ³	28 day Cube Strength, N/mm ²	8 Week Accelerated Carbonation ^{a)} Depth, mm	Embodied CO ₂ , kg/m ³ Concrete ^{b)}
100%PC	35	0.60	280 ^{c)}	41	11.5	268
PC/20FA	35	0.60	280 ^{c)}	36	18.5	216
PC/35FA	35 ^{c)}	0.60	280	35	18.5	209
PC/55FA	37 ^{c)}	0.55	300	37	22.5	198
PC/25FA/10SF	35 ^{c)}	0.60	280 ^{c)}	35	31.0	178
PC/25FA/10LS	35 ^{c)}	0.60	280	35	17.0	194
PC/25FA/10MK	35 ^{c)}	0.60	280	35	23.5	211
PC/45FA/10SF	37 ^{c)}	0.55	300	37	33.5	214
PC/45FA/10LS	37 ^{c)}	0.55	300	37	19.0	182
PC/45FA/10MK	37 ^{c)}	0.55	300	37	28.0	191

a) 4% CO₂, 55% RH, 20°C

b) Calculations based on figures in Table 1.

c) Limiting factor(s) controlling the specification.

Fig. 1 Comparison of coefficient of chloride diffusion for PC, conventional FA and high FA content concretes.

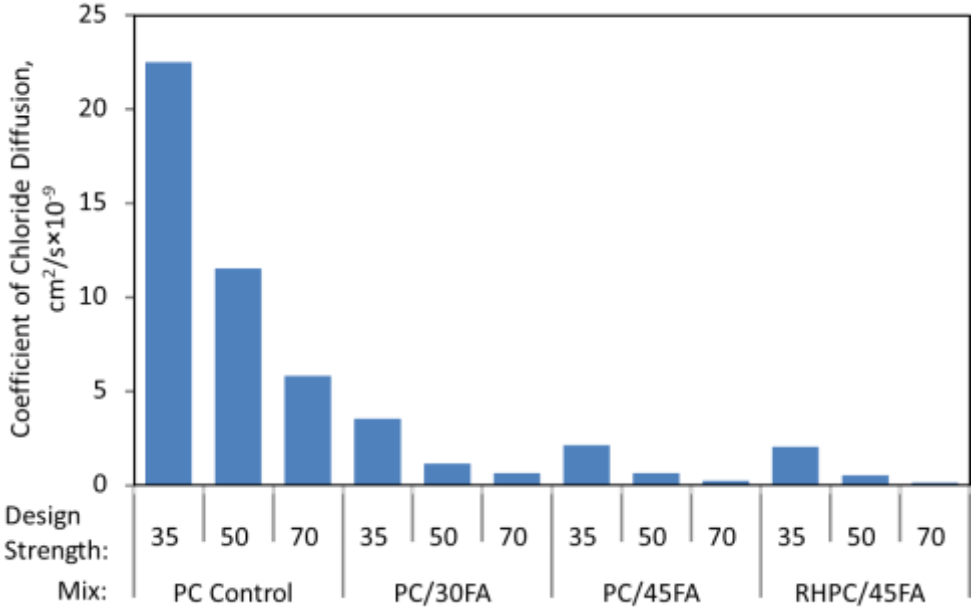


Fig. 2 Comparison of depth of accelerated carbonation for PC, conventional FA and high FA content concretes.

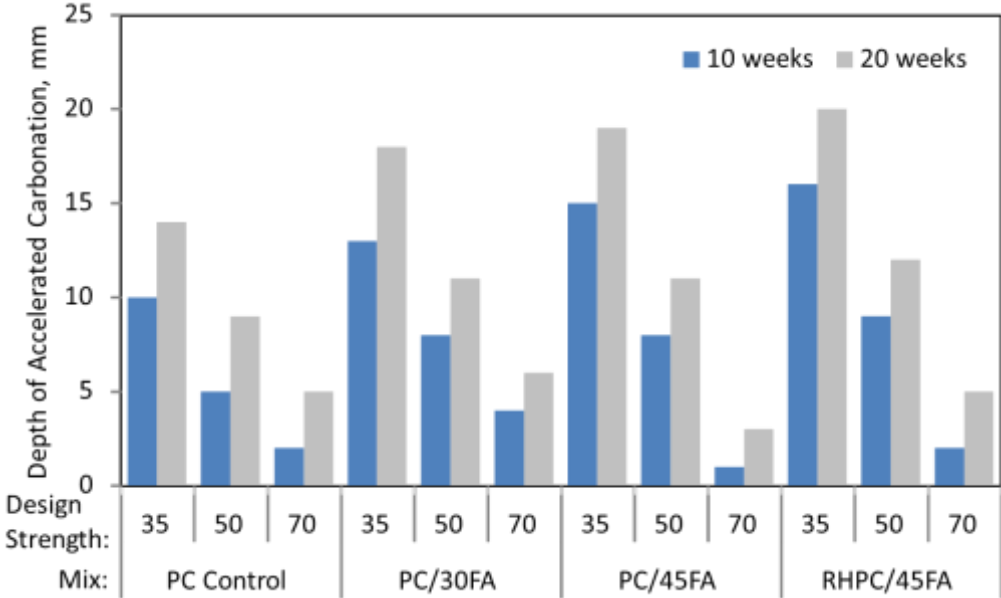


Fig. 3 Comparison of embodied CO₂ of constituent materials for PC, conventional FA and high FA content concretes.

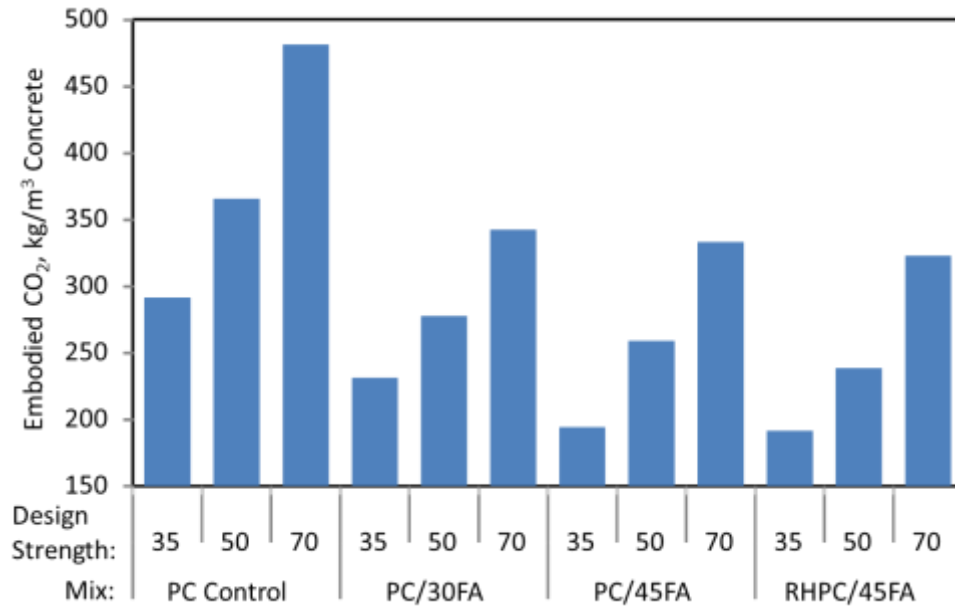


Fig. 4 Comparison of non-steady state chloride migration and accelerated carbonation test results of FA binary and ternary concretes

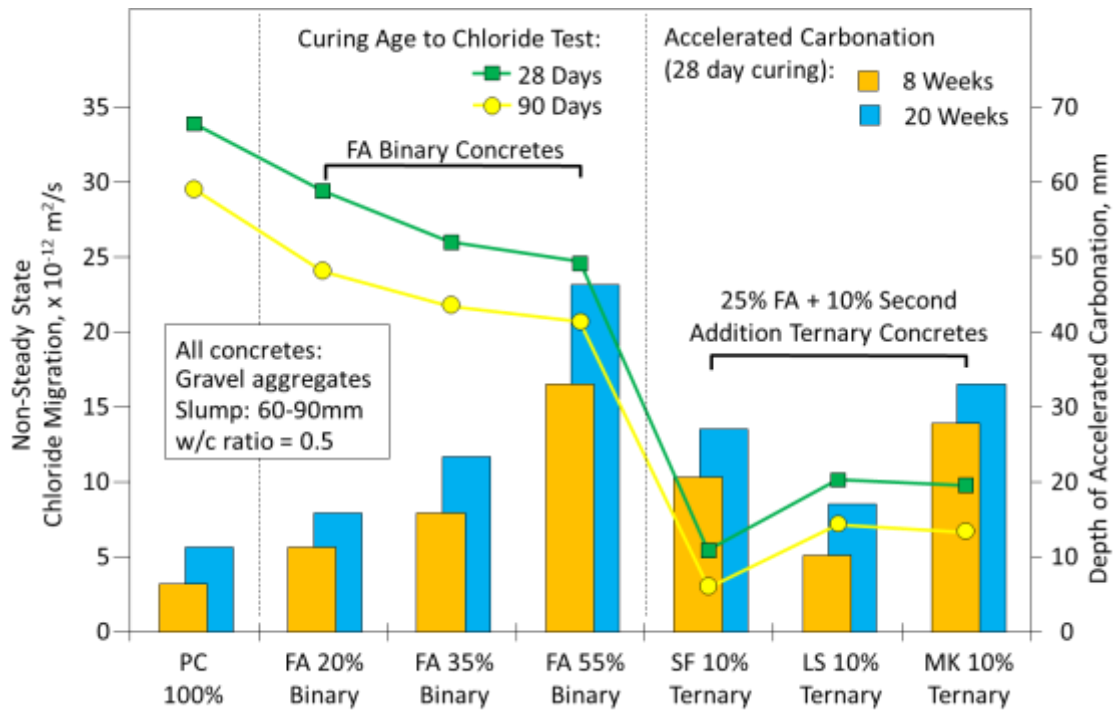


Fig. 5 Embodied CO₂ of constituent materials of PC, FA binary and ternary concretes.

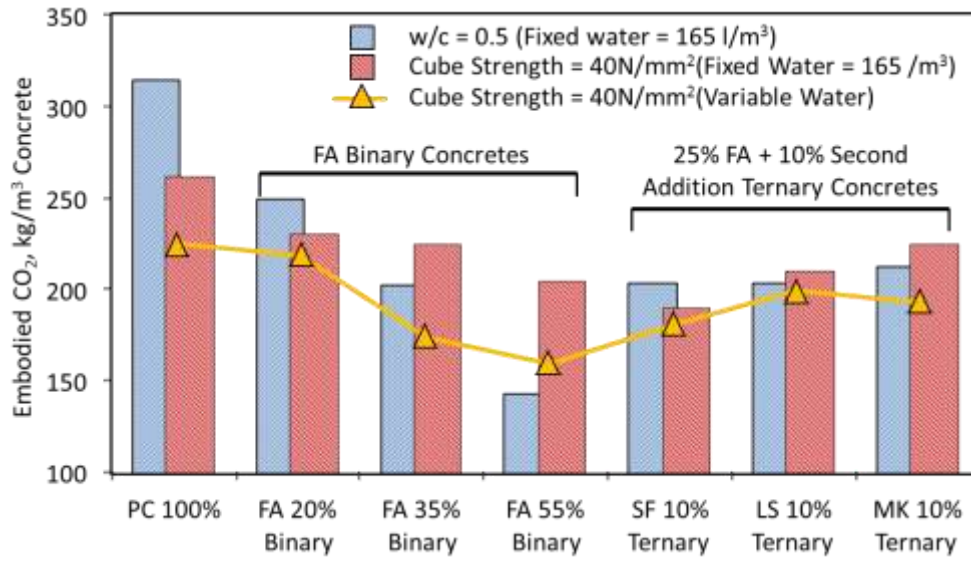


Fig. 6 Comparison of coefficient of chloride diffusion and depth of accelerated carbonation of PC/FA concretes with different FA fineness.

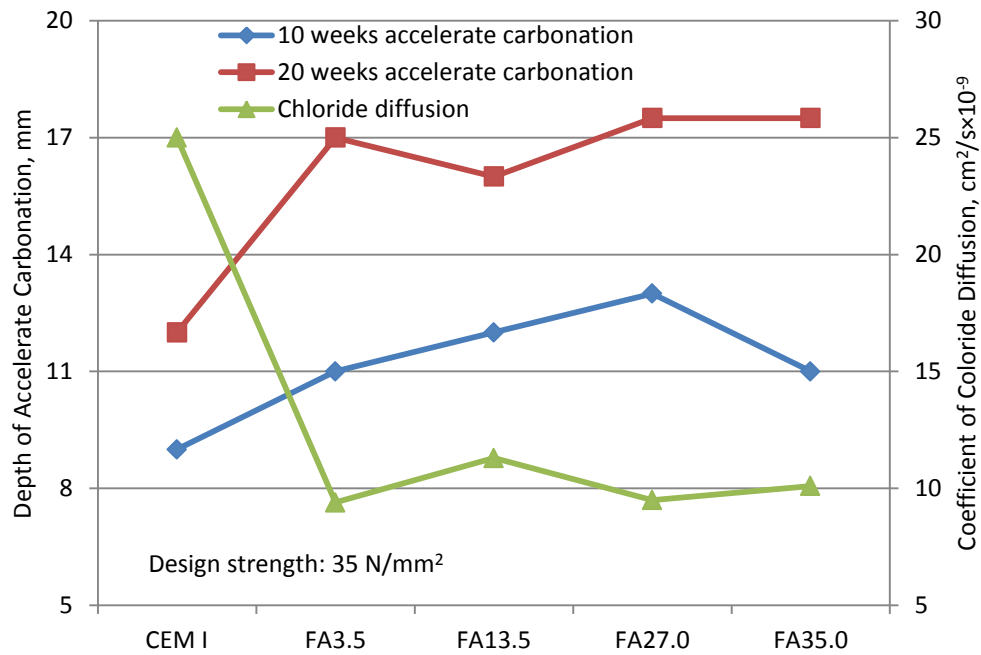


Fig. 7 Embodied CO₂ of constituent materials of PC/FA concretes with different FA fineness.

