Fly Ash Route to Low Embodied CO_2 and Implications for Concrete Construction

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KEYWORDS: Embodied CO_2, water and energy in concrete; fly ash; permeation; carbonation; chloride ingress.

ABSTRACT:

This paper addresses the drive to reduce the amount of CO_2 embodied in concrete to achieve durable and sustainable construction and the implications this has for the construction industry. Shortcomings of the current methods of specifying concrete are discussed and an alternative offered that overcomes the drawbacks of mix limitation or prescription approaches. At previous WOCA conferences, the authors have demonstrated that recycled and secondary aggregates can be used to reduce demand on quarried materials. However, an adjustment to mix constituent proportions using fly ash (FA) is required to enable 'equal' performance to be maintained. In this paper, the authors progress this concept to enumerate the embodied CO_2 of typical structural concretes. The UK has now agreed the various embodied CO_2 levels for all relevant cementitious materials. Using this framework, the role of FA, of up to 55% of the total cement content, to displace Portland cement (PC) is demonstrated. These 'low carbon' concretes can enhance, or at least match, the properties of equivalent, 'traditional' all PC concretes. However, while resistance to chloride ingress is improved, with carbonation a 'trade-off' between these is likely to be required. Furthermore, it is shown that this approach extends the time to achieve early strength, which may have implications for formwork removal, prestressing and live load application. It is suggested that alternative cements that have a high early strength development in combination with FA could offset this.

INTRODUCTION

The choices now available to engineers for specifying concrete constituent materials for different applications are vast. These options include a wide range of cement types and additions, fibres, additives, new-generation admixtures and alternatives to natural aggregates. This has given unprecedented opportunities to tailor concrete for an infinite number of end uses.

While initial cost will always be a major driver, more enlightened designers and clients are evaluating the whole-life cost. This has been enabled with the increased understanding of materials, their interaction, and how they function under different conditions, and hence a growing number of issues now need to be considered as part of specification. Furthermore, clients, and indeed a majority of the public, are expecting that sustainability should be at the core of the design and construction processes.

One aspect of this is the amount of CO_2 that has been released during the production of a particular material, usually referred to as embodied CO_2. Although disputed in some quarters, Portland cement (PC) clinker is implicated in the greenhouse debate given that its manufacture results in around 8% of the total anthropogenic CO_2 produced worldwide. There are, therefore, clear obligations to use the material in a responsible manner. With the demand by designers in many markets, 'embodied CO_2 calculators' are being made available to the construction community to evaluate the effect of using different combinations of materials. Indeed, this type of approach is being used in LEED, BREEAM and CEEQUAL environmental management systems.

The levels of embodied CO_2 have now been agreed in the UK for the key concrete constituent materials and those for the main types of cement and additions are given in Table 1 below.

Table 1. UK agreed embodied CO_2 of concrete constituent materials.[1,2]

CONSTITUENT MATERIAL	Embodied CO_2 (kg CO_2 / tonne)
Portland cement	930
Fly ash	4
GGBS	52
Metakaolin	330
Silica fume	14
Limestone	32
Minor additional constituents	32

In this respect, the use of fly ash (FA) in concrete is highly desirable as it has, by far, the lowest embodied CO_2 content. In this paper, the authors look at the role FA can play in low carbon concrete design and the implications this has on construction processes, particularly on site and the need to balance these with durability.

EUROPEAN APPROACH TO SPECIFICATION OF CEMENT, COMBINATIONS AND CONCRETE DURABILITY

Europe is attempting to harmonise construction standards in all member countries but this has been a slow process and even after 25 years work there are still differences in national requirements. To a certain extent this can be expected as the climatic conditions of Europe are diverse but in the main it continues to use existing practices that account for these differences. However, there is a common agreement with regard to the environmental exposure conditions for which the engineer is expected to specify concrete with suitable mix constituents and proportions. In this paper, the authors will consider the design of concrete with FA to resist (i) carbonation and (ii) chloride ingress, both directly and in terms of lowest practicable embodied CO_2.

The use of FA in concrete is well established in the UK, having been first used in post-war dam construction, following successful projects in the US in the 1930's. This led to standards for FA emerging in 1965 (as a filler) and in 1982 (as a cement component). Today, FA is widely used throughout Europe, particularly where concrete durability is a key design requirement.

ROLE OF FA IN CEMENT COMBINATIONS

As noted above, recent developments in concrete construction mean that there are now a much wider range of constituent materials than have traditionally been available. Indeed, progress has been made in this area with the development of new European cement and concrete standards (e.g. BS EN 197-1, BS EN 206-1 and BS 8500), which provide a framework for engineers to use these materials for a given set of conditions. They also, in many cases, offer environmental benefits.

It could, however, be argued that such standards, which are prescription led, are restricting, to some extent, in the way they permit materials to be combined. Furthermore, while the literature indicates that various cement combinations have been investigated [3,4] and there are examples of their use to meet particular specification requirements in practice, these have tended to be limited. This section describes research carried out to identify what is achievable with FA in cement combinations, since this is likely to be an important issue that would need to be balanced alongside environmental considerations.

Materials and Mix Proportions

A PC and four additions, FA, limestone (LS), metakaolin (MK) and silica fume (SF) were used. All materials conformed to appropriate standards and their main physical and chemical characteristics are given in Table 2. The concretes included coarse aggregate in two fractions (4/10mm and 10/20mm) and a fine aggregate (0/4mm) conforming to BS EN 12620. A high-range water-reducing admixture was used to enable the same water/cement (w/c) ratio to be adopted for all mixes and to control workability.

The concretes had a fixed free water content of 165 l/m^3 with admixture added to achieve the target slump range of 60 to 90 mm. The w/c ratios used were 0.35, 0.50 and 0.65 to provide a range of typical concretes and strengths used in structures. PC was replaced with FA at levels of 20, 35 and 55% and then part of the FA in the 35% FA concrete replaced with each of the other additions at a level of 10%. All concretes were cured in water at 20^oC to 28 days, prior to testing or exposure.

Table 2. Properties of cement and additions

PROPERTY	CEMENT/ADDITION TYPE				
	PC	FA	MK	LS	SF
Blaine Fineness, m^2/kg	410	10.0[a)]	12,400[b)]	1,550[b)]	24,000[b)]
Loss on Ignition, %	-	6.1	-	43.6	-
Particle Density, g/cm^3	3.14	2.20	2.59	2.63	2.10
Initial Setting Time, min.	135	-	-	-	-
Main Oxides, %					
CaO	64.6	2.8	0.2	55.3	0.4
SiO_2	20.0	44.0	57.4	0.4	96.8
Al_2O_3	4.6	21.7	38.6	0.3	0.5
Fe_2O_3	3.8	8.7	0.6	0.0	0.1
K_2O	0.6	2.3	2.3	0.0	0.5
Na_2O	0.3	0.8	0.1	0.1	0.2
SO_3	3.1	1.3	0.0	0.0	0.1

[a)] 45µ sieve retention, % by mass. [b)] Data obtained from BET specific surface
- not tested

Permeation

The rate of transportation of water through concrete is an important factor influencing the durability of concrete. While chemical processes are also often important, measurement of the permeation properties provides an insight to its microstructure.

The results from absorption tests under a 200 mm head (initial surface absorption test at 10 minutes, ISA-10 (BS 1881, Part 207)) on the binary and ternary combination concretes are shown in Figure 1. These give expected behaviour in terms of the effect of w/c ratio, with the ranking of concretes generally maintained across the range considered. The binary combinations indicate similar values to the PC reference at 20% FA and gradual increases compared to this at 35 and 55%.

The introduction of the second addition gave a mixed performance with MK higher than the binary combination concrete, and SF and LS lower, with the latter similar to or less than that of PC. In the water penetration tests, carried out under a 500 kPa head (BS EN 12390-8), similar type results were obtained to those of the absorption tests. In this case, PC concrete was lower than the binary combination concretes, which again gave progressively higher values with increasing FA level. The introduction of the second addition gave either similar results to the binary combination concrete, or improved lower values.

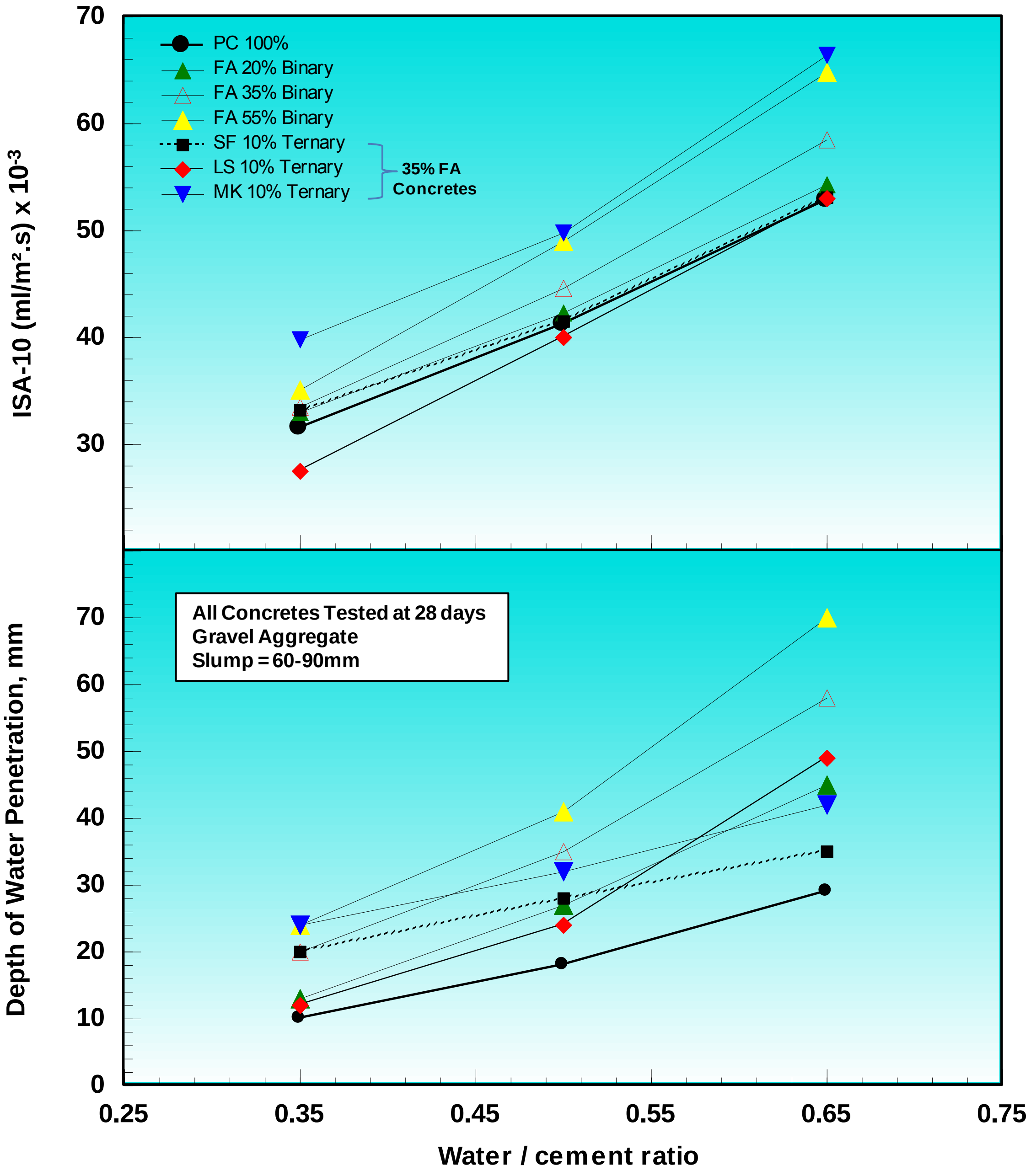

Figure 1. Comparison of absorption and water penetration of binary and ternary concretes

As noted elsewhere,[5] the results indicate that these materials cannot directly replace PC to achieve equal properties. When comparisons of the absorption tests are made at equal strength, with adjustments to w/c ratio to achieve this, the binary and ternary combination concretes give similar and in some cases better performance than PC.

Chloride ingress

The ingress of chloride into concrete is mainly an issue for structures in coastal or highway environments. In these situations, chloride passes into the material in solution and with its gradual build-up around steel reinforcement, corrosion can initiate, and with adequate levels of oxygen available, cause severe damage to structural elements.[6]

The influence of the test concretes on non-steady state migration of chlorides (using the NT Build 492 test) at 0.50 w/c ratio is shown in Figure 2. This indicates that for the various cement combinations, chloride migration reduced with curing time (28 to 180 days, 20°C in water). For the FA binary combination concretes, the migration reduced with increasing FA level compared to that of PC, reflecting its enhanced alumina content, which can bind chlorides.

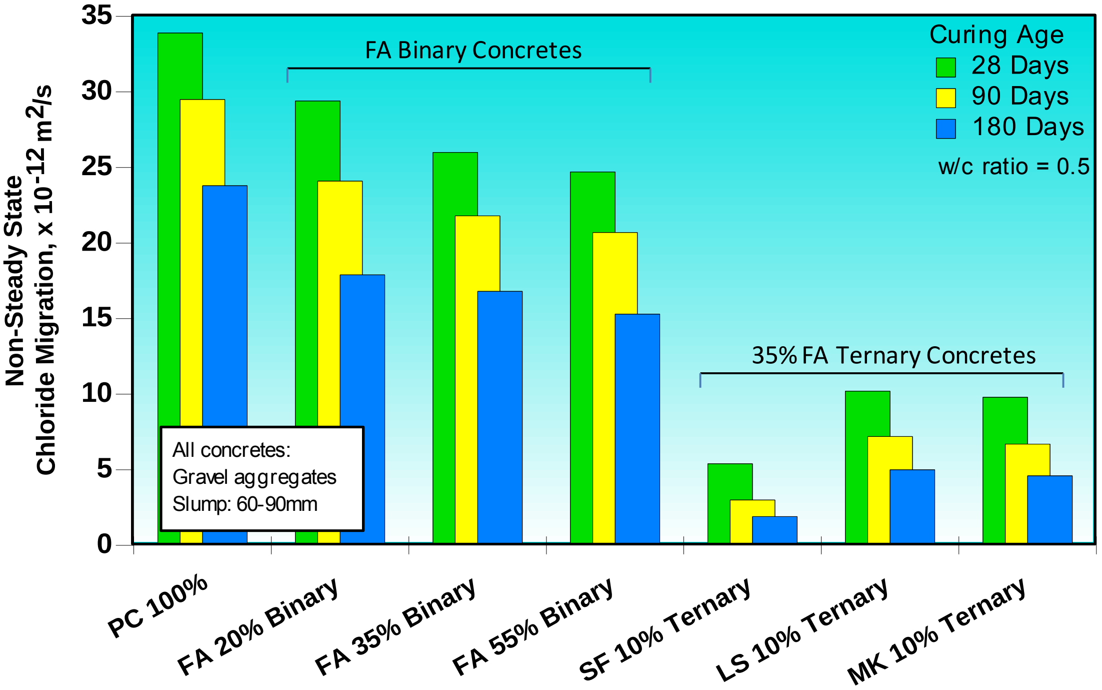

Figure 2. Comparison of non-steady state chloride migration of FA binary and ternary concretes

The inclusion of second additions, MK, SF or LS, significantly reduced the chloride migration value, with LS and MK showing similar benefits compared to the FA binary combination and SF having the greatest effect. These materials, given their fineness, are likely to contribute physically to reduce chloride migration (and to chloride binding in the case of MK).

Carbonation

Carbonation of concrete is due to the diffusion of atmospheric CO_2 into the surface of the material and interaction with the lime-rich microstructure and pore fluids, leading to a significant reduction in alkalinity. This process continues from the surface and can become a problem for embedded steel once it has penetrated the cover depth. Indeed, with the loss of passivity and in the presence of sufficient moisture and oxygen, expansive reinforcement corrosion occurs. The behaviour of concrete made with different cement types is not consistent, for example in FA and other pozzolanic cement concretes, the pore fluid is less alkaline than that of an equivalent PC. This can lead to a more rapid ingress of CO_2, however, this may be partially offset by enhanced permeation properties and as a result it is difficult to predict carbonation resistance without direct testing.

Since carbonation is a very slow process taking decades under natural exposure, in terms of laboratory testing it is necessary to use accelerated methods in order to compare the likely behaviour of different concretes.[7,8] It should be noted that there is an on-going debate as to whether this truly reflects atmospheric carbonation and, thus, accelerated data should be used carefully for durability modelling purposes.

Figure 3 show the depths of accelerated carbonation of the binary and ternary combination concretes at 0.50 w/c ratio following 20 weeks exposure (4% CO_2, 55% RH, 20°C). As indicated the carbonation depth of FA binary concretes was found to increase with FA level.

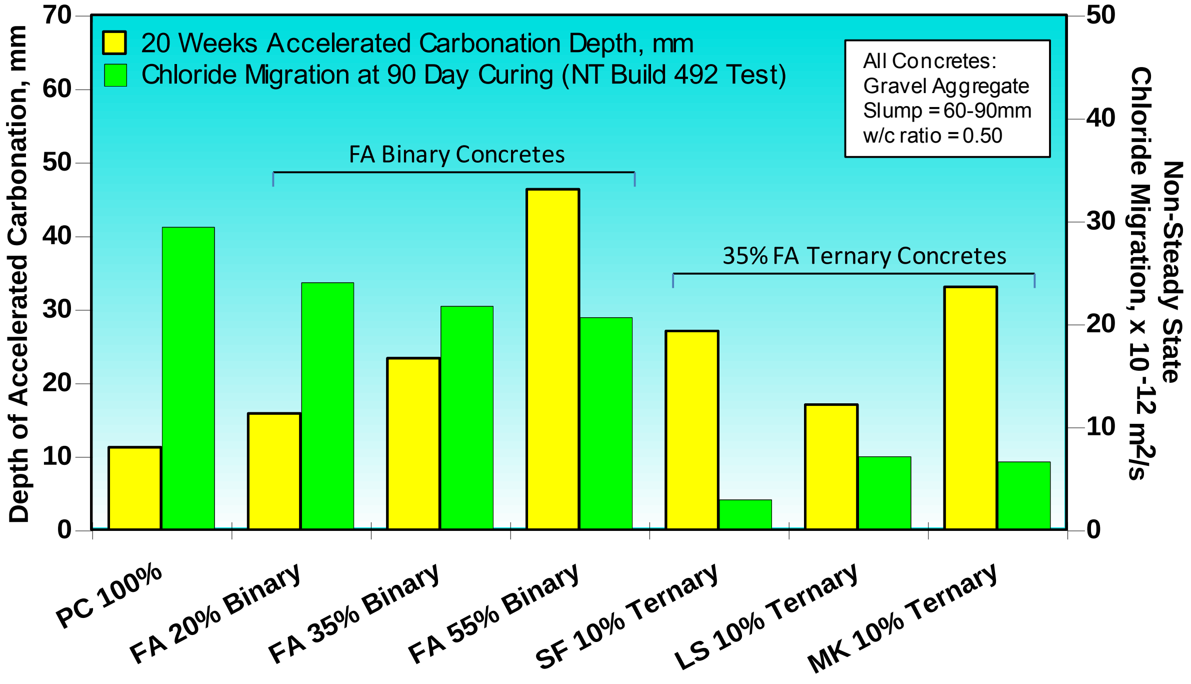

Figure 3. Comparison of 20 weeks accelerated carbonation absorption and 90 day curing non-steady state chloride migration for FA binary and ternary concretes

The inclusion of second additions gave increases in carbonation depth compared to the binary combination concrete, except for LS where there was a slight reduction. The results reflect the basis of comparison (i.e. equal w/c ratio) and the accelerated / dry conditions of the test and highlight the effects resulting from consumption of lime by pozzolanic materials.

It is apparent that the trend for carbonation is the opposite of that noted for chloride migration, also shown in the figure. This indicates that where both processes occur in a particular exposure, a 'trade-off' is likely to be necessary in material selection. It also highlights the difficulties likely to exist in balancing the different factors when trying to optimise performance, including environmental.

EXAMINING EMBODIED CO_2, WATER AND ENERGY

In order to compare potential sustainability in terms of environmental impact, the concretes were evaluated for embodied CO_2, water and energy of their constituent materials.

Tables 1 and 3 show the values used in these comparisons for the individual constituents on a tonnage basis. The data, mainly based on information supplied by the UK concrete industry, covers the CO_2 released, and energy and water required, 'cradle to gate', for these materials.

Table 3. Embodied energy and water in the production (cradle to gate) of constituent materials for concrete

CONSTITUENT MATERIAL	EMBODIED ENERGY, kWh/t [a)]	EMBODIED WATER, litres/t [a)]
Portland cement	1194	45
Fly ash	9.3	0
Limestone	12.7	48
Metakaolin	400 [b)]	0 [c)]
Silica Fume	10 [b)]	0 [c)]
Aggregate	12.7	48
Admixture	2500	650

[a)] Based on Reference 9 unless otherwise stated
[b)] Based on embodied CO_2 ratios between PC and metakaolin / silica fume
[c)] Based on private communication with materials suppliers.

In evaluating embodied CO_2, energy and water, the concretes described above and those of a related study[5] were considered, with the following approaches to mix proportioning covered:

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 admixture content (0.6% by weight of cement)

Figures 4 and 5 compare the calculated embodied CO_2 and energy of the concretes. The inclusion of FA lead to reductions in embodied CO_2 and energy compared to PC and this effect increased with FA level. When comparing binary and ternary combination concretes, the inclusion of a second addition lead to minor differences which depended on how the concretes were proportioned.

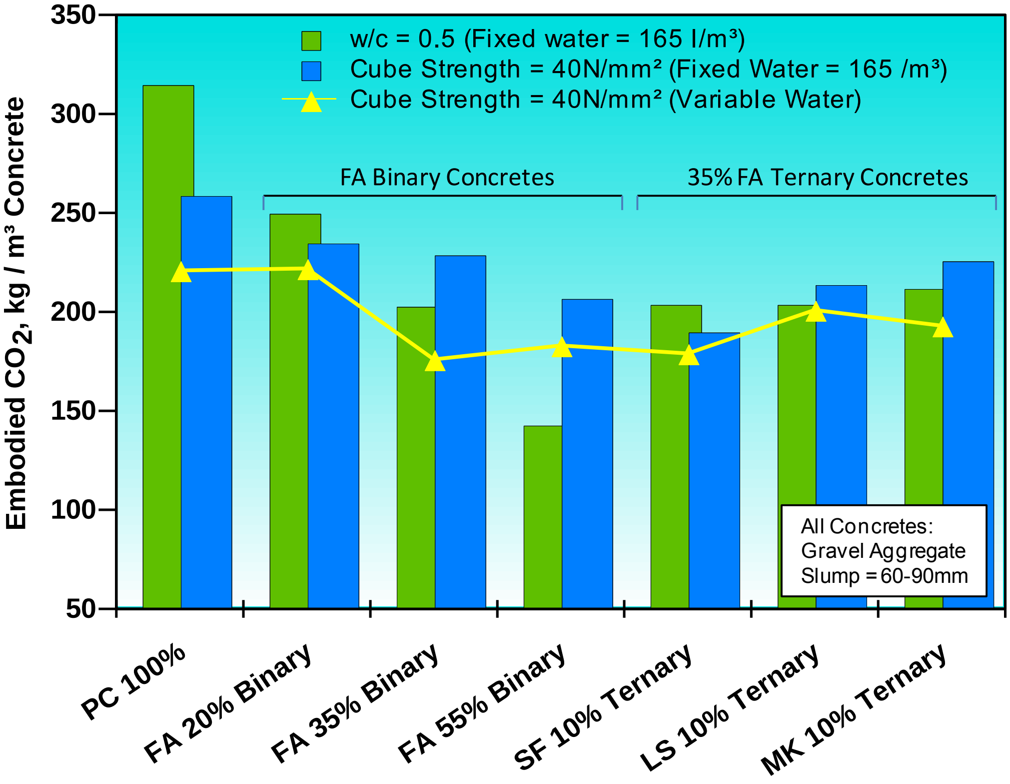

Figure 4. Embodied CO_2 of constituent materials of PC, FA binary and ternary concretes.

Figure 6 shows the effect of FA on the total water used in the mix. When comparing at equal w/c ratio or cube strength (fixed water), there was little effect of increasing FA level or including a second addition on this.

If water demand is taken into account (variable water), there is a general increase in total water with FA level and the inclusion of a second addition. This is likely to reflect the increased fines content / specific surface area of concrete in these cases.

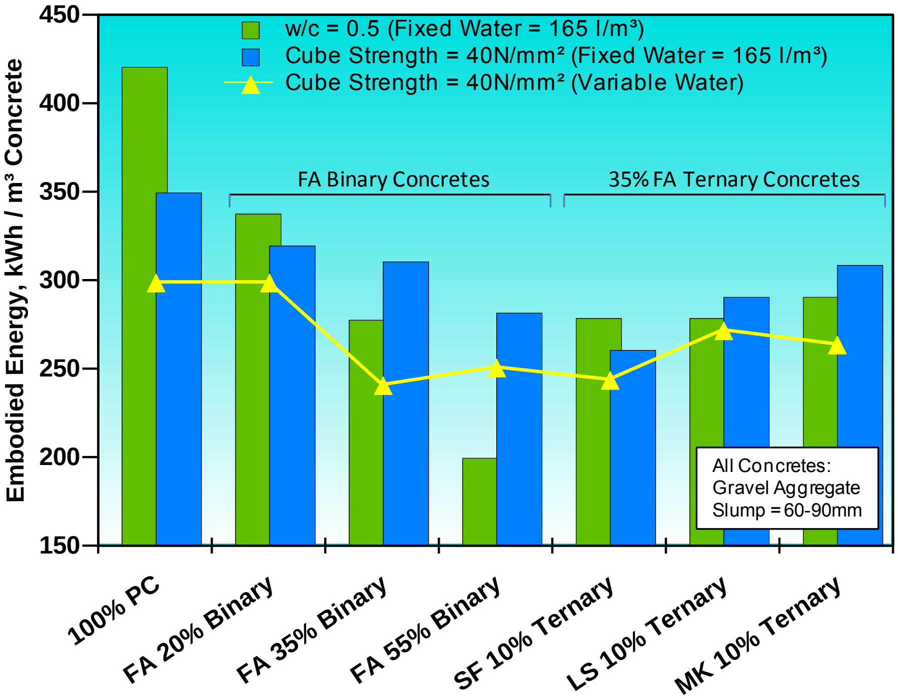

Figure 5. Embodied energy of constituent materials of PC, FA binary and ternary concretes.

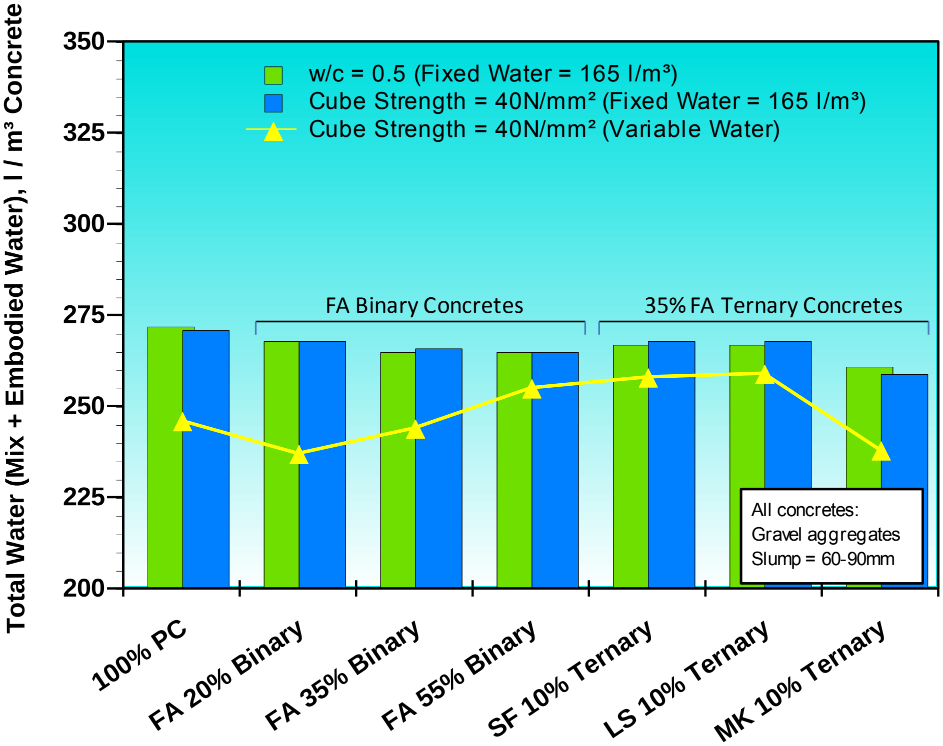

Figure 6. Total water contents (mix water plus embodied water of constituent materials) of PC, FA binary and ternary concretes.

BALANCING DURABILITY PERFORMANCE AND SUSTAINBILITY

Previous work at the University of Dundee developed a methodology for optimising cement combination concretes for use in chloride exposure conditions.[10] A similar approach was used to examine the impact of the FA-based cement combinations on the current specification standard in the UK (BS 8500) for chloride and carbonation-induced corrosion.

BS 8500 defines exposure conditions for corrosion-induced by chlorides in two categories, each with three classes:

- Corrosion induced by chlorides other than from sea water (XD classes)
- Corrosion induced by chlorides from sea water (XS classes)

In this case, XS3 has more onerous requirements when using PC and FA-based concretes than XD3. The minimum requirements for concrete exposed in XS3 conditions are shown in Table 4.

BS 8500 defines the exposure conditions for concretes subject to a risk of carbonation-induced corrosion in three classes (XC Classes). Of these, XC3/4 is the most likely to induce corrosion due to the continued presence of moisture and oxygen in conjunction with the carbonation process and the minimum requirements for concrete in this class are also shown in Table 4. The requirements specified in Table 4 are not, of course, coincident, which means that at the minimum point for one criteria, the other two will be exceeded.

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
Limiting Factors for XS3 Chloride Exposure (nominal cover depth 50mm)			
6-20	C50	0.40	380
21-35	C35	0.50	340
36-55	C30	0.50	340
Limiting Factors for XC3/4 Carbonation Exposure (nominal cover depth 30mm)			
6-55	C30	0.65	260

Table 5 gives comparative data for a series of concretes with varying levels of FA up to 55%, all of which conform to the requirements of BS 8500 and, therefore, could be used for infrastructure exposed to a chloride environment. The minimum requirements for specification and the ‘limiting factor(s)’ are indicated for each concrete.

Table 5. Comparison of embodied CO$_2$, water and energy 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, $\times 10^{-12}$m^2/s	Embodied CO$_2$, kg/m^3 Concrete[b]	Embodied Energy, kWh/m^3 Concrete[c]
100%PC	50	0.40 [d]	380	66	28.0	391	516
80%PC/20%FA	50	0.40 [d]	380	61	23.7	315	418
65%PC/35%FA	35 [d]	0.50	340	35	27.1	212	286
45% PC/55%FA	30 [d]	0.50	340	30	20.0	181	246

[a] NT Build 492 test

[b] Calculations based on figures in Table 1.

[c] Calculations based on figures in Table 3.

[d] Limiting factor(s) controlling the specification.

In this case the 55% FA concrete has the ‘best’ performance in terms of chloride resistance and environmental impact. This highlights the effectiveness of high FA concrete in chloride environments (despite its lower strength) and that no ‘trade-off’ with environmental performance is required.

A similar exercise, but in this case for carbonation, is shown in Table 6.

Table 6. Comparison of embodied CO$_2$, water and energy 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[a] Carbonation Depth, mm	Embodied CO$_2$, kg/m^3 Concrete[b]	Embodied Energy, kWh/m^3 Concrete[c]
100%PC	35	0.60	280[d]	42	11.5	268	354
80%PC/20%FA	35[d]	0.60	280[d]	35	17.5	216	287
65%PC/35%FA	35[d]	0.60	280	35	16.5	212	286
45% PC/55%FA	37[d]	0.55	300	37	19.5	200	272

[a] 4% CO$_2$, 55% RH, 20°C

[b] Calculations based on figures in Table 1.

[c] Calculations based on figures in Table 3.

[d] Limiting factor(s) controlling the specification.

This indicates that the cement combination with the lowest embodied CO_2 and energy had the greatest carbonation depth. Thus, a 'trade-off' 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).

CONCLUSIONS

The demands of low carbon legislation and client specification have made it clear that all PC concrete will become more difficult to justify. This makes the use of FA attractive, particularly as slag cannot provide the volumes needed worldwide. However, just displacing PC is not a solution to this issue, as a much broader range of performance requirements are necessary, not least durability and early strength.

The influence of FA in cement combinations on concrete properties is considered. In particular, the use of ternary cements offers benefits in chloride resistance over more familiar PC/FA concretes.

The environmental cost is examined (in terms of embodied CO_2, energy and water) with regard to the different concretes. This indicates that environmental benefits were achievable in the FA-based concretes, but this was influenced by how they were proportioned.

It is shown that for some properties such as resistance to chloride-induced corrosion, it is possible to achieve very good performance in terms of chloride ingress and environmental impact. However, for carbonation it is necessary to balance between these to achieve an optimum. 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, which may have implications for formwork removal and this factor may also need to be considered in the material selection process. Further environmental benefits are achievable through the use of higher FA levels but this would increase the reduction in early strength development, which would have to be offset either by activation or combining it with a high early strength cement.[11]

ACKNOWLEDGEMENTS

The data reported in this study is based on information provided from several research projects. The authors would like to acknowledge the contribution made to these by CTU staff in collaboration with industrial partners, the Mineral Products Association: Cement and Concrete and the United Kingdom Quality Ash Association.

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