

Service Life and Life-Cycle Assessment of Reinforced Concrete with Fly ash and Limestone Calcined Clay Cement

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ABSTRACT

Environmental impact due to the emission of carbon dioxide during concrete production can be taken care by reducing the clinker content in the cement. The clinker content can be reduced by replacing it with fly ash and limestone calcined clay. Such systems can have a potential to exhibit enhanced durability/service life when exposed to chloride and carbon dioxide. However, estimating probabilistic service life of concretes with such alternative binder systems is difficult due to the lack of quantitative estimates of the input parameters such as chloride diffusion coefficient (D_{cl}), ageing coefficient (m), carbonation coefficient (K_{CO_2}), and chloride threshold (Cl_{th}). This paper presents the experimentally observed estimates of these parameters for the following systems: (i) 100% OPC, (ii) 70% OPC + 30% fly ash, and (iii) limestone calcined clay cement (LC3) – known as OPC, PFA, and LC3 concretes, respectively, herein. A total of three concrete mixes were designed. Also, based on these input parameters, the probabilistic service life estimates of a bridge pier and a girder made of these three concretes and exposed to chlorides and carbon dioxide are presented. For chloride ingress study, Fick's 2nd Law of diffusion and Cl_{th} have been used. For carbonation study, a recently developed model for estimating carbonation depth (using mixture proportion) have been used. Then, the life-cycle assessment (LCA) of these three concrete systems in terms of the CO₂ emissions per unit of concrete per year of estimated service life is presented - for both chloride and carbonation induced corrosion. In chloride laden environments, the service life of the bridge pier and girder systems could be enhanced by about 10 times by using fly ash or LC3 concretes – for similar strength grade concretes. Also, the average annual CO₂ emissions (during the expected service life) of PFA and LC3 concretes could be about 3 and 7 times, respectively, lower than that of OPC concretes of similar strength grade. In case of carbonation-induced corrosion, the limited experimental data indicate that the PFA and LC3 concretes could exhibit a lower service life and higher average annual CO₂ emissions (during the expected service life) than OPC concretes.

Keywords: Concrete; chloride; carbonation; corrosion; durability; service life; life-cycle assessment

1.0 INTRODUCTION

Recent research on the properties and behaviour of limestone calcined clay cement (LC3), a ternary blend with 50% partial substitution of clinker with limestone and calcined clay (Antoni *et al.* 2012; Bishnoi *et al.* 2014; Emmanuel *et al.* 2016), shows that this binder can significantly reduce the carbon footprint of cement and concrete. It was found that the mechanical characteristics of concretes with LC3 are comparable, if not better in some aspects, with ordinary Portland cement (OPC) and cement-fly ash concretes. It was also shown that concrete made with LC3 can exhibit high resistance against chloride permeability and migration, mainly due to the refined pore structure of the hydrated binder phase (Dhandapani *et al.* 2018). On the other hand, accelerated carbonation tests have indicated that the reduced clinker content in LC3 could result in a

lower CaO buffer capacity, which in turn may lower the resistance to carbonation (Leemann *et al.* 2015). For many structural projects considering various concretes, there is the need for quantitative estimate of the service life and carbon footprint so that decisions on material selection can be made with multiple perspectives in mind. This paper presents the probabilistic estimates of service life with concretes made of OPC, LC3, and OPC-fly ash systems, considering the durability limited by chloride attack or carbonation, and integrates these estimates with the CO₂ emissions that can be attributed to the concretes.

1.1 Service Life Estimation

In general, the service life of concrete structures subjected to chloride ingress or carbonation can be divided into initiation and propagation phases (see Fig. 1). In most cases, as the duration of propagation phase can be influenced significantly by

factors such as cracking and could be much smaller than that of the initiation phase, it is conservative to assume that the service life is equal to the duration of the initiation phase.

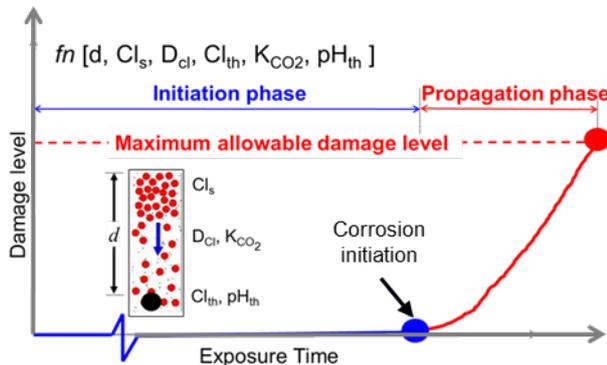


Fig. 1. Parameters influencing the corrosion initiation phase

In the case of chloride-induced corrosion, chloride ions diffuse through the cover concrete, reach the steel surface, and initiate corrosion when the concentration reaches a threshold value (defined as chloride threshold, Cl_{th}). Consequently, corrosion initiation is governed by the surface chloride concentration (Cl_s), the chloride diffusion coefficient (D_{Cl}) and the related ageing coefficient (m), the Cl_{th} , and the critical cover depth (x) in the structure being assessed.

In the case of carbonation-induced corrosion, the carbonation coefficient (K_{CO_2}) and the pH threshold (pH_{th}) are the key material parameters governing corrosion initiation, where K_{CO_2} is a function of concrete mix proportions and the microclimate or internal relative humidity (Parrott 1994; Nilsson *et al.* 1996). It should be noted that the internal relative humidity ($RH_{internal}$) at various depths can be influenced by the external relative humidity as well as the pore structure of concrete cover (Andrade *et al.* 1999). In particular, the carbonation rate can be significant when the $RH_{internal}$ is about 65 to 80% (Papadakis 1991). The pH_{th} has been estimated to be between 9 and 10 for various concretes (Czarnecki and Woyciechowski 2015).

1.2 Life Cycle Assessment (LCA)

Concrete production contributes to about 5 to 8% of the total man made global CO_2 emissions and has the danger of increasing, especially in countries like India due to the increase in per capita requirement of cement and concrete (Scrivener *et al.* 2016). Also, globally it is expected that the cement production will increase by four times by the year 2050 (Imbabi *et al.* 2012). Among the concrete ingredients, cement holds the major share in CO_2 emissions. Choosing an alternative cementing material, which would partially replace the clinker content - to reduce the negative environmental impact of concrete is important. It is, therefore, desired that, in general,

the concrete used in RC structures is made using locally available raw materials with low CO_2 footprint and high durability. LCA is a tool that could give an overall estimate of the environmental impact of cement and concrete; and is becoming more and more important in construction projects due to the increasing awareness on the global warming, resource depletion, and energy consumption. Here, the LCA is carried out based on the data obtained from typical cement plants in India, along with commonly used databases and conversion factors (Gettu *et al.*, 2016a, 2016b).

2.0 RESEARCH SIGNIFICANCE

Many new alternative binding materials (such as LC3) are being introduced into the concrete construction industry, for which the service life and life cycle assessments are becoming important for the material selection. This paper highlights the importance of combining the results of these two key assessments for the purpose of sustainability, along the lines of the works of Van den Heede and De Belie (2012) and Muller *et al.* (2014). Moreover, the present work provides experimental data on the key transport and corrosion initiation parameters – for both the chloride- and carbonation-induced corrosion, along with the CO_2 footprint from life cycle assessment. Also, the framework for combining the service life and life-cycle assessments is demonstrated for concretes containing OPC, fly ash and LC3 and exposed to chlorides and carbon dioxide.

3.0 METHODOLOGY AND RESULTS

3.1 Materials

The three binders of the concretes assessed here were made of i) 100% OPC, ii) OPC with 30% replacement with Class F fly ash, iii) LC3, and these concrete mixes are denoted as OPC, PFA, and LC3, respectively. The OPC and PFA mixes used 53 Grade OPC conforming to the IS 12269 (2013). The LC3 was a composition of 50% OPC clinker, 31% calcined clay, 15% limestone, and 4% gypsum and was obtained through an industrial scale trial production. Table 1 provides the oxide composition of OPC, fly ash, and LC3 used in the study.

Table 2 provides the mix proportions of the concretes used in this study, all with a design 28-day cube compressive strength of 35 MPa. A slump in the range of 80-120 mm was obtained by adjusting the dosage of a polycarboxylic ether (PCE) based superplasticizer, in the range of 0.2 to 1% solids by weight of the binder.

Table 1. Oxide composition of the binders used

Oxide composition	OPC (%)	Fly ash (%)	LC3 (%)
Al ₂ O ₃	4.17	29.95	10.6
CaO	64.59	1.28	39.7
SiO ₂	19.01	59.32	30.3
Fe ₂ O ₃	3.89	4.32	3.5
K ₂ O	0.59	1.44	0.2
MgO	0.88	0.61	1.7
Na ₂ O	0.16	0.16	0.2
SO ₃	1.70	0.16	0.3
LOI	1.40	-	9.4

Table 2. Mix proportions of concretes per m³

Ingredients	Concrete mix code		
	OPC	PFA	LC3
Binder (kg/m ³)	310	360	360
Water (kg/m ³)	155	162	162
Water-binder ratio	0.50	0.45	0.45
Fine aggregate (kg/m ³)	695	721	687
Coarse aggregate (kg/m ³)	10 mm	496	463
	20 mm	744	694

3.2 Determination of the durability parameters

Diffusion coefficient (D_{Cl})

The bulk diffusion test (as per ASTM C1556) was carried out on 100 × 200 mm cylindrical specimens cured in a mist room for one year. The extended curing was used in order to achieve complete hydration, especially in the blended systems. After the 1-year moist curing, the curved surface of the specimen was coated with epoxy. Then, the specimen was sliced into two halves (each with 100 mm length). Two slices for each concrete mix were immersed in 2.8 M NaCl solution for a period of 56 days. Then, powder samples were collected from various depths (at a regular interval of 2 mm up to a depth of 25 mm from the surface) using a profile grinder. The chloride concentration in the collected powder samples were tested according to the SHRP 330 (1992) method to estimate the acid soluble chlorides, from which D_{Cl}-values were determined for each concrete. The D_{Cl} of LC3 is 93% and 63% lower than OPC and PFA mixes – indicating excellent resistance of LC3 against chlorides. The major reason for this could be the pore refinement in the PFA and LC3 mixes, which results in better ionic resistance (Dhandapani *et al.* 2017, 2018).

Ageing coefficient (m)

The ageing coefficient, *m*, related to the chloride diffusivity of a concrete mix can vary depending upon the binder composition, water-binder ratio and exposure condition. This is attributed to the development of the physical structure in the binder phase and the evolution of the resistance due to the chloride binding ability of alumina. Here, the value of *m* is determined using an indirect method based on

the electrical conductivity measurement on concrete, as shown in Eq. (1), which was proposed by Andrade *et al.* (2011).

$$\sigma(t) = \sigma_{ref} \left(\frac{t_{ref}}{t} \right)^m \quad (1)$$

where, σ_{ref} is the electrical conductivity at t_{ref} (i.e., the reference age of 28 days). The electrical conductivity of the concrete mixes was determined at 28, 90 and 365 days, as per the ASTM C 1760 Standard, and the ageing coefficients were calculated through the regression analysis. This approach assumes that ageing can be accounted as a function of the physical structure development and does not consider the decay in diffusivity due to chloride binding. This leads to a conservative estimate of the ageing coefficient, more specifically in the case of alumino-silicate based blended systems, where an extensive amount of binding in the cementitious matrix can occur.

The electrical conductivity (σ) results shown in Fig. 2 indicate that the OPC mix exhibits higher electrical conductivity than PFA and LC3 mixes. Using Eq. (2), the values of *m* estimated for OPC, PFA, and LC3 mixes were 0.26, 0.70, and 0.53, respectively. The values obtained for PFA and LC3 seem reasonable as they are close to the range of values reported in the literature for concretes with fly ash and metakaolin (Boddy *et al.*, 2001; Nokken *et al.* 2006; Stanish and Thomas, 2008; Park *et al.*, 2016).

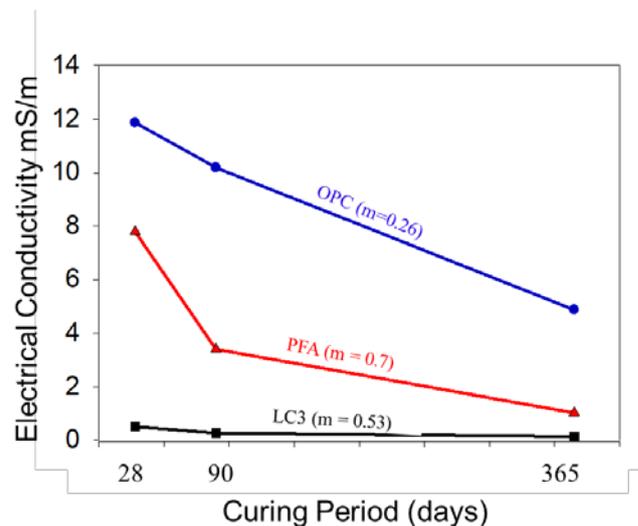


Fig. 2. Change in electrical conductivity as a function of curing period

Chloride threshold (Cl_{th})

The Cl_{th} for steel embedded in OPC, PFA, and LC3 systems was determined using the “lollipop” type specimens (consisting of steel rebar embedded in mortar). The specimen was prepared using an 8 mm diameter and 70 mm long TMT/QST steel reinforcing bar with a cover depth of 10 mm (see Fig. 3). Here, ‘TMT/QST’ stands for Thermo-Mechanically Treated/Quenched and Self-Tempered steel reinforcing bar, which is widely used in today’s construction. The

mortar was prepared with a water : cement : sand ratio of 0.5 : 1 : 2.75.

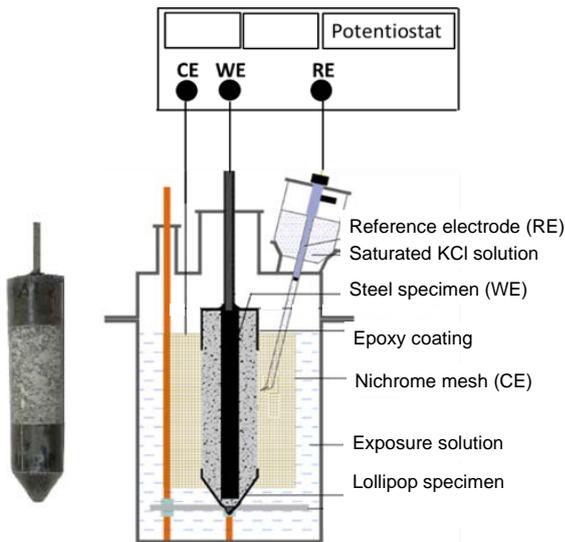


Fig. 3. Lollipop test specimen and setup for determining chloride threshold

Ten specimens each of OPC, PFA and LC3 were exposed to alternate wet-dry cycles (i.e., two days of wetting followed by five days of drying). At the end of the wet regime, linear polarization resistance (LPR) tests were conducted by sweeping the potential from -15 mV to +15 mV versus the open circuit potential (OCP) at a scan rate of 0.1667 mV/s. The polarization resistance (R_p) was monitored until it fell below $10,000 \Omega \cdot \text{cm}^2$, at which corrosion is assumed to have initiated. The linear polarization measurements were taken at the end of every wet period. The OPC, PFA, and LC3 specimens were subjected to maximum of 5, 7, and 8 wet-dry cycles, respectively. However, it should be noted that the Cl_{th} is a function of both the wet-dry cycles and the transport properties of the mortar cover. After this, the specimens were split into two and powder was collected from the mortar at the interface. The acid-soluble chloride content of the mortar was determined using the SHRP S330 (1992) procedure and defined as the Cl_{th} . Table 3 provides the Cl_{th} values obtained for the systems with OPC, PFA, and LC3. The Cl_{th} was highest for the OPC mortar (0.44% by weight of cement) due to the higher alkalinity and more buffering capacity of $\text{Ca}(\text{OH})_2$ at the interface.

Carbonation depth

Carbonation studies were carried out on concrete prisms of $100 \times 100 \times 500 \text{ mm}$. The specimens were cast and moist cured for 28 days, after which they were exposed under unsheltered conditions on the terrace of a building in Chennai, India (see Fig. 4a).

Table 3. Input parameters used for estimation of chloride induced corrosion

Properties of concrete	Concrete mix code		
	OPC	PFA	LC3
D_{Cl} at 365 days ($\times 10^{-12} \text{ m}^2/\text{s}$)	~N (18.7, 6.3)	~N (3.5, 0.7)	~N (1.3, 0.2)
Ageing coefficient, m	~N (0.26, 0.07)	~N (0.70, 0.03)	~N (0.53, 0.07)
Cl_{th} (% bwoc)	~N (0.06, 0.02)	~N (0.05, 0.02)	~N (0.03, 0.01)

This exposure condition can be classified as XC4 (cyclic wet and dry), according to EN 206:1:2000. The carbonation depth in these specimens were measured after splitting the specimen across the cross-section, and spraying phenolphthalein indicator on the freshly fractured surface (see Fig. 4b) after 1, 2, and 3 years of exposure. This colorimetric technique differentiates the carbonated and non-carbonated zone with colourless and pink colour zones, respectively. The average carbonation depths observed under unsheltered, natural exposure are shown in Fig. 5.

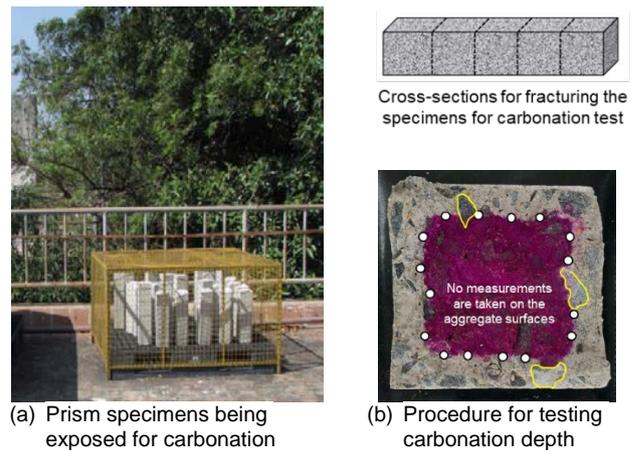


Fig. 4. Testing for carbonation

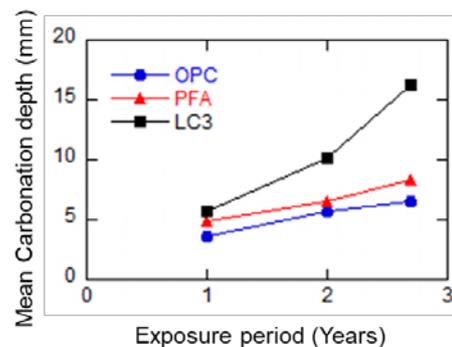


Fig. 5. Average carbonation depths observed in OPC, PFA, and LC3 concretes

4.0 SERVICE LIFE ESTIMATION

4.1 Initiation of Chloride-Induced Corrosion

Using the Fick's second law of diffusion and the input values from experiments (see Table 3), the chloride concentration at the steel-binder interface, as $C(x, t)$ (for depth, x , and time, t) is determined as follows:

$$C(x, t) = Cl_s \left(1 - \operatorname{erf} \left[\frac{x}{\sqrt{4D_{Cl}t}} \right] \right) \quad (2)$$

where, D_{Cl} is the apparent chloride diffusion coefficient of concrete, erf is the mathematical error function (Amey *et al.* 1998). The surface chloride build-up rate was assumed to be 0.04% by weight of concrete (%bwoc) for a period of 15 years (i.e., $k = 0.04$ %bwoc), which results in a maximum surface chloride concentration ($Cl_{s,max}$) of 0.6 %bwoc (after 15 years of exposure). The ageing coefficient was not considered for $t > 25$ years.

A Matlab code was developed to perform the Monte-Carlo simulations using the statistical distributions given in Table 3 and then calculating the chloride levels at the level of embedded steel at the end of every year, $C(x, t)$. The determined $C(x, t)$ in every simulation was compared with the randomly generated Cl_{th} . If the $C(x, t)$ realized at the level of steel reinforcement is more than the random realization of Cl_{th} value, then the corrosion is defined to be initiated. In this way, 1000 simulations each were done for systems with OPC, PFA, and LC3 and the cumulative probability of $C(x, t)$ exceeding the Cl_{th} was determined for each year; and defined as the cumulative probability of corrosion initiation. Then, the cumulative distribution functions (CDFs) for the time to corrosion initiation were developed.

4.2 Initiation of Carbonation-Induced Corrosion

Carbonation in concrete leads to a reduction in the pH, which in turn can lead to corrosion initiation. The rate of advancing of the carbonation front is usually modelled to estimate service life (Papadakis *et al.* 1991; Marques *et al.*, 2010). Tutti (1982) had proposed a model to estimate the carbonation depths in OPC systems using K_{CO_2} and the square root of exposure time, the suitability of which has been questioned for the concretes with SCMs (Visser *et al.* 2012). Later, researchers have modelled carbonation rate as a function of thermodynamic parameters (Shi *et al.* 2016), and durability/strength parameters (Russell *et al.* 2001, Salvoldi *et al.* 2015).

In this paper, a preliminary model to estimate the carbonation depth as a function of mixture proportion is proposed as follows.

$$x_d = K_{CO_2} (t)^{\theta_0} \quad (3)$$

where

$$K_{CO_2} = \theta_1 \left(\frac{BC}{UW} \right) \left(\frac{w}{b} \right)^{\theta_2} \left(1 - \frac{RL}{100} \right)^{\theta_3} \quad (4)$$

- x_d = Carbonation depth (mm)
- BC = Binder content (kg/m³)
- UW = Unit weight of concrete (kg/m³)
- w/b = water to binder ratio
- t = Exposure period (years)
- RL = SCM replacement level (%)
- θ_i = Maximum likelihood estimates (i = 0, 1, 2, and 3)

Table 4 provides the maximum likelihood estimates (θ_i) of the model parameters determined using an optimization code in Matlab. Using Eq. (3) and the coefficients given in Table 4, the K_{CO_2} for the OPC, PFA, and LC3 concretes were determined to be 4.2, 4.7, and 8.8, respectively. The carbonation depth at the end of every year was estimated using Monte-Carlo simulations – using a Matlab code. Using an approach similar to that used for the chloride attack, the cumulative distribution functions (CDFs) for the time to corrosion initiation due to carbonation were developed.

Table 4. Model coefficients for three types of binder

Type of binder	θ_0	θ_1	θ_2	θ_3
OPC	0.6	129	2.0	1.0
PFA	0.7	11.8	1.9	-6.6
LC3	0.9	10.7	1.44	-3.5

5.0 LIFE CYCLE ASSESSMENT

LCA technique is used to assess the environmental aspects and potential impacts associated with a product's life cycle and the manufacturing processes involved (ISO 14040:2006). Here, SimaPro v8.0.5.13 (Pré Consultants 2016) was used to calculate the environmental impact of concrete production. It was used to compute the total equivalent CO₂ emissions due to the production of cement and concrete of different mix proportions. The four major steps followed in LCA are i) system boundary definition, ii) inventory analysis, iii) impact assessment and iv) comparative analysis, see Fig. 6.

In the present study, the boundaries corresponding to a 'ground-to-gate' system were considered. The primary source of data was a cement plant near Chennai, India and secondary sources of data were database from ecoinvent, Environmental Policy Act (EPA), and Intergovernmental Panel on Climate Change (IPCC). For the environmental impact assessment, the CO₂ conversion factors given in the IPCC 2013 Global Warming Potential (GWP) for

6.2 Carbonation Induced Corrosion

The reinforced concrete bridge pier is considered to be located at about 5 km from the sea shore and exposed to seasonal wet (rain) and dry (summer) conditions with an atmospheric carbon dioxide concentration of about 400 ppm. The service life was estimated using the Eq. (3) and the input variables, as per the mixture proportions given in Table 2.

Fig. 7 (c) shows the CDF of corrosion initiation time. The corrosion initiation time for the system with OPC concrete is seen to be much longer than that for the PFA and LC3 mixes because the rate of progress of carbonation front is primarily a function of $\text{Ca}(\text{OH})_2$ buffer capacity. Despite the better transport properties exhibited by the concretes with blended binders, the deterioration due to the reaction of CO_2 with calcium silicate leads to the coarsening of pores (Chindaprist *et al.* 2009) and a faster rate of carbonation. As shown in the last three bars in Fig. 7 (d), the CO_2 footprint per year of service life for the systems with OPC, PFA, and LC3 concretes are 2.4, 5.9, and 9.8, respectively. A comparison with this and the results from the case study on chloride-induced corrosion indicate that a binder system that leads to a longer service life and lower CO_2 footprint for a structure exposed to chlorides need not always behave the same way when exposed to carbon dioxide. This emphasizes the need for material selection based on the quantitative evaluation of both service life and CO_2 footprint.

7.0 CONCLUSIONS

Based on the experimental data and service life estimation presented in this paper, following major conclusions are drawn.

1. PFA and LC3 systems exhibited lower chloride diffusion coefficient (D_{cl}) and lower chloride threshold (Cl_{th}) than that in OPC systems with similar strength grade. However, it is seen that because of the combined effect of both lower D_{cl} and lower Cl_{th} , the PFA and LC3 systems could exhibit longer service life than OPC systems exposed to chloride environments.
2. Based on the initial 3 years of unsheltered, natural carbonation study, it was found that the concretes with fly ash and LC3 could experience faster carbonation than that experienced by OPC systems of similar strength. The suitability of LC3 when carbonation is the principal mode of deterioration needs to be investigated further.
3. In case of chloride-induced corrosion, the CO_2 footprint per year of service life for LC3 and PFA concretes is estimated to be significantly lower than that for OPC systems. The use of blended

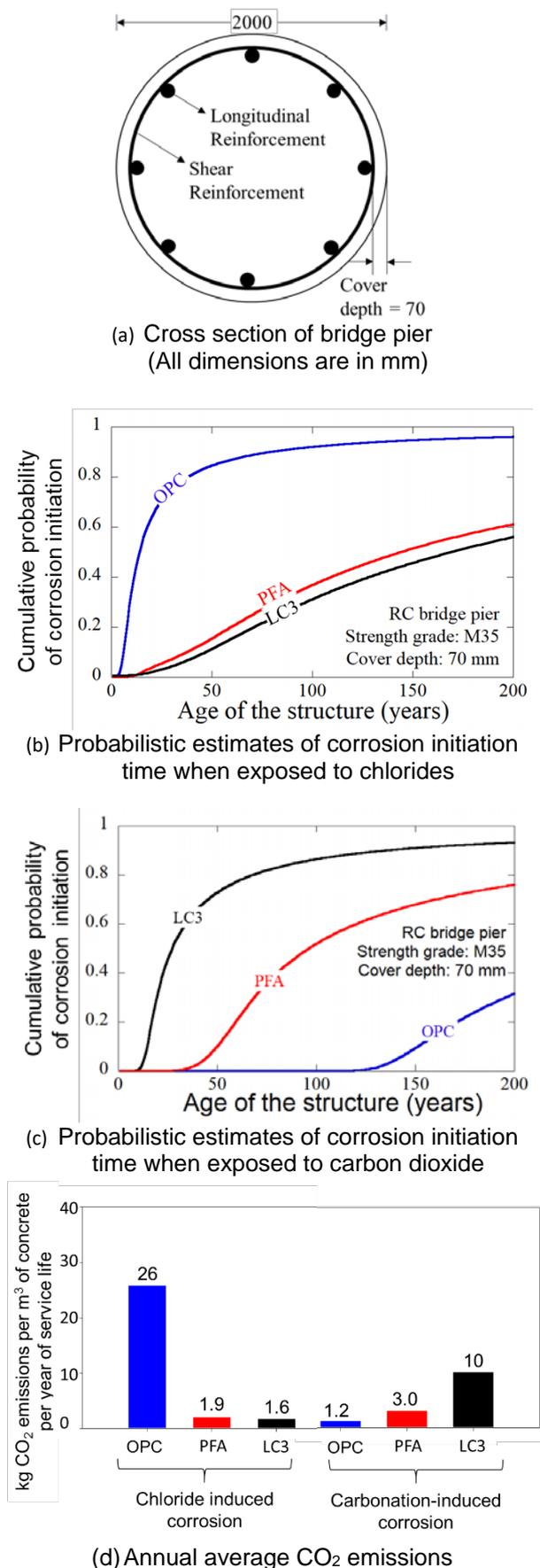


Fig. 7. Results from the case study on service life estimation and life cycle assessment.

systems (PFA and LC3) can have a major positive impact on sustainability due to the higher service life against chloride ingress and lower emissions associated with concrete structure.

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Notations

C_{th}	Chloride threshold, [%bwob]
m	Ageing coefficient
C_s	Surface chloride concentration [%bwoc]
D_{Cl}	Chloride diffusion coefficient, [m^2/s]
D_{ref}	Diffusion coefficient at t_{ref} , [m^2/s]
t	Age of the structure [years]
t_{ref}	Reference time [years]
$\sigma(t)$	Electrical conductivity at time t [mS/m]
$\sigma_{(ref)}$	Electrical conductivity at t_{ref} , [mS/m].
x	Cover depth (m)
K_{CO_2}	Carbonation coefficient (mm/ \sqrt{yr})
x_d	Carbonation depth (mm)

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