Estimated Service Life of Carbonation Exposed (Cracked) Concrete with Pozzolans or Self-healing Agents

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ABSTRACT

Today, concrete with large portions of ordinary Portland cement (OPC) replaced by fly ash and self-healing concrete count as potential sustainable alternatives to traditional concrete. For the first concrete type, the increased sustainability lies in lowering the carbon footprint which is largely attributable to cement. For the second one that should be able to heal cracks autonomously upon occurrence, an extended service life is the key objective. In this paper, both concrete types were evaluated in terms of service life in carbonation exposed environments. When using the probabilistic model for carbonation-induced steel depassivation of fib Bulletin 34 with mix specific curing exponents, it was found that in uncracked High-Volume Fly Ash (HVFA) and Fly Ash + Silica Fume (FA+SF) concrete carbonation should not reach the reinforcing steel at a typical cover depth of 35 mm within the envisaged design service life of 100 years. This requires a longer optimal curing period though (HVFA: ≥ 20 days; FA+SF: ≥ 9.1 days versus OPC: ≥ 4.2 days). Evaluation of cracked concrete suggests that in presence of a 25 mm deep, 300 μm wide crack the depassivation period would take no longer than 5 years. In case of partial crack healing with the proposed encapsulated polymer, this could be extended to only 11 years regardless of the carrier concrete type (OPC, HVFA or FA+SF, all properly cured for a sufficiently long time). A 100 % crack healing efficiency implying a return to the uncracked state should therefore always be aimed for. The now considered polymer seems reasonably efficient since this was the case for 8 out of 9 samples. To account for the risk of insufficient curing in combination with possibly having an unhealed/partially healed crack anyway, OPC binder systems still have the preference as carrier concrete for incorporation of the healing agent in exposure class XC3.

Keywords: High-Volume Fly Ash (HVFA) concrete, Self-healing concrete, Cracks, Carbonation, Service life.

1.0 INTRODUCTION

Nowadays, more and more attention goes to a performance-based design of steel reinforced concrete structures. Gradually, this design strategy is replacing deemed-to-satisfy approaches involving a prescribed cement content and water-to-cement ratio per exposure class as prescribed by the concrete standards, e.g. NBN B15-001 (BIN, 2004). The most straightforward way of defining expected concrete durability performance would consist of quantifying the estimated service life of the envisaged concrete mixture for given exposure conditions (Van den Heede and De Belie, 2012). Several engineering codes with probabilistic service life prediction models for carbonation/chloride-induced corrosion are available in that perspective, e.g. fib Bulletin 34 (fib, 2006). Performance-based approaches will allow for an easier market introduction of novel concrete types, e.g. concrete with high portions of OPC replaced by supplementary cementitious materials (SCMs) or concrete with autonomous crack healing properties (Van den Heede, 2014; Van Belleghem et al., 2017). By incorporation of high volumes of fly ash at the expense of cement, the lower portlandite content is further decreased by its pozzolanic reaction. Hence,

the buffering capacity versus CO_2, and thus the carbonation resistance of the HVFA concrete is reduced (Lammertijn and De Belie, 2007). On the other hand, if cured well, concrete with SCMs could feature a low permeability and possibly result in an satisfactory service life performance anyhow. Probabilistic service life estimation cf. fib Bulletin 34 indicates that this could hold true for HVFA concrete and FA+SF concrete (Van den Heede, 2014). Yet, in our previous work the same default regression exponent for curing b_c, i.e. -0.576, was used for all concrete mixtures regardless of their binder composition. In this paper, additional service life calculations were performed after experimental or literature based determination of a mix specific b_c value for OPC, HVFA and FA+SF concrete.

Given the earlier mentioned shift in focus towards service life related performance based design, quite some research time and effort is being invested today in self-healing concrete. Concrete is known for having a low tensile strength which makes it very susceptible to cracking. These cracks count as preferential pathways for CO_2 and result in faster carbonation. Over the years, the Magnel Laboratory for Concrete Research of Ghent University has developed dedicated experience in three major

healing mechanisms, i.e. through encapsulation of polymers (Van Tittelboom, 2012), mixing in superabsorbent polymers and mircrofibres (Snoeck, 2015) or inclusion of encapsulated bacterial spores (Wang, 2013). Regarding the first mechanism, initial service life assessment estimations have already been done for both marine and carbonation exposed environments (Van Belleghem et al., 2017; Van den Heede et al., 2018), be it for only one type of binder system per exposure class. In this paper, the feasibility in terms of service life performance of various concrete compositions (OPC versus HVFA versus FA+SF) as carrier for the encapsulated polymer for self-healing purposes in carbonation exposed environments was investigated.

2.0   MATERIALS AND METHODS

2.1   Concrete Mixtures

Given the higher susceptibility to carbonation of concrete with high volumes of fly ash, the preference usually goes to OPC binders in environments where carbonation-induced corrosion is at risk. When looking at exposure class XC3 (moderate humidity, sheltered from rain), NBN B15-001 normally prescribes a minimum cement content of 300 kg/m³ and a water-to-cement (W/C) ratio of maximum 0.55 (BIB, 2004). Therefore, a mixture T(0.55) with those requirements was manufactured as reference. One could choose to replace part of the cement with pozzolanic fly ash. However, the maximum fly ash replacement level is strongly limited by the k-value (= 0.4) that is imposed onto fly ash by NBN B15-001 (BIN, 2004). In fact, this approach implies that 50% cement replacement levels are not allowed when designing for exposure class XC3. Nonetheless, since HVFA concrete counts as a potentially 'green' alternative to traditional OPC concrete, it was decided to consider this type of mixture anyway. Although its carbonation resistance is expected to be less, the resulting service life performance in view of a typical design value of 100 years might still be acceptable. The following two mixtures with high cement replacement levels were therefore considered, i.e. mixture F50 with a total binder content of 450 kg/m³ of which 50% fly ash and with a water-to-binder ratio of 0.35, and mixture F40S10 with a total binder content of 340 kg/m³ of which 40% is fly ash and 10% silica fume, and with a W/B ratio of also 0.35. Those mix proportions are identical to the ones used in Van den Heede and De Belie (2015).

Until now, comparative service life assessment of carbonation exposed cracked and self-healing concrete remained limited to the OPC reference alone (e.g. as in Van den Heede et al. (2018)). In this study, the options of having a self-healing F50 and F40S10 were additionally considered. This was done for the encapsulated polymer-based healing agent with the most effective healing performance

according to Van den Heede et al. (2018), i.e. a commercial low viscosity polyurethane (PU) precursor with addition of 5 wt. % of accelerator and 5 wt. % of benzoyl peroxide (BPO). The additions to the precursor pressurize the healing agent inside the capsules and guarantee a better flow-out from the capsules upon crack occurrence. As such, 8/9 cylindrical concrete samples showed full crack healing during a colorimetric carbonation screening test. The cracks were induced artificially by putting thin metal plates with a predefined length (60 mm), depth (25 mm) and thickness (300 µm) in the sample molds upon casting. In case of self-healing concrete, the thin metal plates contained three holes with three capsules (length: 35 mm, inner diameter: 3 mm) going through. By pulling out the thin metal plates from the hardened samples, capsule rupture and flow-out of the healing agent occurred. As such, the healing mechanism was triggered in an artificial manner for the screening test. Full crack healing implied that the carbonation front of the sample cross-sectional surface as visualized with colour indicator phenolphtahalein would no longer extend around the crack location and beyond the crack tip.

2.2   Inverse effective carbonation resistance

The accelerated inverse effective carbonation resistance $R_{ACC,0}^{-1}$ for compositions T(0.55), F50 and F40S10 was already determined in Van den Heede and De Belie (2015) and amounts to 2684 ± 303, 11292 ± 343 and 5835 ± 165 (mm²/years)/(kg/m³), respectively. As expected, the OPC reference is the least susceptible to carbonation. With $R_{ACC,0}^{-1}$ values that are more than twice and four times the reference value, mixtures F40S10 and F50 follow in terms of increasing carbonation susceptibility.

In Van den Heede et al. (2018) it was shown that the $R_{ACC,0}^{-1}$ values for uncracked concrete are not valid for the crack region itself. Lower carbonation depths from the crack tip onwards suggests a lower carbonation rate in the crack region which can most probably be attributed to the fact that the humidity conditions inside the crack are less favourable for carbonation to occur to its fullest extent than at the surface. The carbonation rates from the uncracked surface and from the crack tip are estimated to differ with a factor 3 ± 1. For self-healing concrete with partial healing of the crack in relation to uncracked concrete, this factor seems to increase to 8 ± 2. In case of self-healing concrete with full crack healing, no factor needs to be applied to the $R_{ACC,0}^{-1}$ value. Note that simply assuming another $R_{ACC,0}^{-1}$ value for cracked and partially healed self-healing concrete does not suffice when predicting the time to carbonation-induced depassivation of embedded reinforcing steel. The modified $R_{ACC,0}^{-1}$ values are to be combined with a reduced concrete cover on top of the reinforcing steel to account for the crack depth, i.e. 25 mm in this case study.

2.3 Exponent of regression for curing

In the first days after casting, several measures can be taken to prevent premature desiccation of the concrete surface. This can be done via curing with water, moist air, covering the surface with sheets, etc. Neglecting to do so will have a negative effect on the hydration process, hamper the development of the microstructure and result in inferior strength and durability properties (e.g. the resistance to CO_2 ingress and carbonation). This is especially critical when having binder systems that hydrate slowly (e.g. in case of composite cements such as CEM II, III and V or in case of partial replacement of OPC by pozzolanic fly ash or latent-hydraulic blast-furnace slag. In the prediction model of fib Bulletin 34 for carbonation-induced steel depassivation (see subsequent section), the act of curing and its duration are accounted for via the execution transfer parameter k_c which includes an exponent of regression for curing b_c. Surprisingly, fib Bulletin 34 gives only one value for b_c, i.e. -0.576 ± 0.024, which is to be applied to all binder systems. The authors considered it more appropriate to differentiate and determine binder system specific b_c values. This can be done through the Bayesian regression procedure mentioned in DARTS R2.16 (European Commission, 2004) and (Gehlen, 2000). Therefore, concrete samples need to be exposed to increased CO_2 levels for a fixed time period, while varying the curing period prior to exposure. The mean carbonation depths that are eventually obtained per subset of samples with curing period i, are to be filled out in Eq. (1).

$$k_{c,i} = \left(\frac{x_{c,i}(t)}{x_{c,7}(t)} \right)^2 \tag{1}$$

with $k_{c,i}$, the curing factor for curing period i (–), $x_{c,i}(t)$, the carbonation depth at the end of exposure for curing period i (mm), $x_{c,7}(t)$, the carbonation depth at the end of exposure for a reference curing period of 7 days. Then, the as such obtained values for $k_{c,i}$ need to be plotted as a function of the corresponding curing periods. A subsequent fitting of Eq. (2) to the data points, yields an estimated value for b_c.

$$k_c = a_c \cdot t_c^{b_c} \tag{2}$$

with a_c set equal to $7^{(-b_c)}$ cf. (Gehlen, 2000). Thus, b_c is the only output of the fitting operation. Using this method, the b_c value for composition T(0.55) was determined experimentally. Therefore, concrete cubes with a 100 mm side were stored in a carbonation chamber at a CO_2 concentration of 1%, 95 % relative humidity (RH) and 20°C for 203 days. The considered curing periods prior to exposure were 2, 7, 14 and 28 days. The corresponding carbonation depths as determined with phenolphthalein colour indicator amounted 6.0 ± 1.4 mm, 3.6 ± 1.6 mm, 2.5 ± 2.0 mm and 0.8 ± 0.5 mm,

respectively. The resulting b_c value equaled -0.807 ± 0.050 (R²: 0.978). The b_c values for the other two compositions were not determined experimentally. In a previous study by Van den Heede and De Belie (2015), they were estimated from curing age (1, 3, 7, 14 and 28 days) related carbonation depth data reported by (Burden, 2006), after 90 days of exposure for concrete with similar cement replacement levels by fly ash (50% and 40%) and a W/B ratio of 0.34 which is very close to 0.35. The as such estimated b_c values for compositions F50 and F40S10 amounted to -0.897 ± 0.010 (R²: 0.998) and -1.131 ± 0.006 (R²: 0.999). Note that the latter value should still be interpreted with caution since the 40% fly ash composition studied by (Burden, 2006), did not contain any silica fume in addition. Evidently, further experimental verification of the literature based b_c values is imperative and at the moment ongoing.

2.4 Service Life Prediction Model

In fib Bulletin 34 (fib, 2006), a limit state function is defined that allows for an estimation of the time to carbonation-induced depassivation of embedded rebars. Eq. (3) shows the overall form of this limit state function. Since this study focuses on exposure class XC3, it does not include a weather function. Subfunction k_e (Eq. (4)) accounts for the difference in RH between the expected outdoor environment and the carbonation chamber used for the accelerated compliance test, while subfunction k_c (Eq. (5)) handles curing.

$$g(d, x_c(t)) = d - \sqrt{2 \cdot k_e \cdot k_c \cdot \left(k_t \cdot R_{ACC,0}^{-1} + \varepsilon_t \right) \cdot C_S} \cdot \sqrt{t} \tag{3}$$

$$k_e = \left(\left(1 - \left(\frac{RH_{real}}{100} \right)^{f_e} \right) \middle/ \left(1 - \left(\frac{RH_{ref}}{100} \right)^{f_e} \right) \right)^{g_e} \tag{4}$$

$$k_c = \left(\frac{t_c}{7} \right)^{b_c} \tag{5}$$

with d, concrete cover (mm), $x_c(t)$, carbonation depth (mm) at time t (years), k_e, environmental function (–), k_c, execution transfer parameter (–), k_t, regression parameter (–), $R_{ACC,0}^{-1}$, accelerated inverse effective carbonation resistance ((mm²/years)/(kg/m³)), ε_t, error term ((mm²/years)/(kg/m³)), C_S, expected CO_2 concentration (kg/m³), RH_{real}, RH of the environment (%), RH_{ref}, reference RH during the accelerated carbonation test (%), f_e, regression parameter (–), g_e, regression parameter (–), t_c, curing period (d), b_c, exponent of regression for curing (–). Table 1 gives an overview of the mean values, standard deviations, distribution types, etc., that are assigned to each of the above mentioned model input variables when considering the uncracked or fully healed condition of the concrete. The cracked and partially healed condition for each concrete composition were simulated by assuming a concrete

cover of only 10 mm (= 35 mm design cover minus the crack depth of 25 mm), and $R_{ACC,0}^{-1}$ values to which earlier mentioned factors 3 ± 1 or 8 ± 2 were applied.

Table 1. Characterization of the input variables for the service life prediction model.

Variable	Distribution	Mean	Stdv.
$d_{uncr. / healed}$	Lognormal	35	8
k_t	Normal	1.25	0.35
$R_{ACC,0}^{-1}{}_{uncr. / healed}$ (T(0.55))	Normal	2684	303
$R_{ACC,0}^{-1}{}_{uncr. / healed}$ (F50)	Normal	11292	343
$R_{ACC,0}^{-1}{}_{uncr. / healed}$ (F40S10)	Normal	5835	165
ε_t	Normal	315.5	48
C_S	Normal	0.00082	0.0001
RH_{real} (between 40-100%)	Beta	79	9
RH_{ref}	Constant	60	–
f_e	Constant	5.0	–
g_e	Constant	2.5	–
t_c	Constant	3 or 28	–
b_c (default)	Normal	-0.576	0.024
b_c (T(0.55))	Normal	-0.807	0.050
b_c (F50)	Normal	-0.897	0.010
b_c (F40S10)	Normal	-1.131	0.006

Reliability indices (β) and probabilities of failure (P_f) as a function of time were calculated using the First Order Reliability Method (FORM) available in the probabilistic Comrel software. In accordance with fib Bulletin 34 (fib, 2006), these parameters need to meet the requirements for the depassivation limit state (β ≥ 1.3 and P_f ≤ 0.10) to qualify for use in exposure class XC3.

3.0 RESULTS AND DISCUSSION

3.1 Service Life Uncracked/Fully Healed Concrete

The time to carbonation-induced depassivation of reinforcing steel embedded in uncracked concrete or self-healing concrete with a 100% crack healing efficiency varies considerably with the type of binder system, the exponent of regression for curing b_c, and the curing period t_c (Fig. 1). For OPC binder systems with a rather rapid hydration process, a short curing period of a few days should normally be sufficient. However, Fig. 1a indicates that only 3 days of optimal curing implies a time to steel depassivation of less than a typical design service life of 100 years. Under assumption of the experimentally determined b_c value of -0.807 this criterion is not met (77 years < 100 years). Using the default b_c value of fib Bulletin 34 (-0.576) in combination with a 3 d curing period yields a significantly higher time to carbonation-induced steel depassivation (i.e. 94 years). Clearly, the value of b_c plays quite a role in the service life assessment. Therefore, it is highly recommended to use mix specific instead of default b_c values. When adopting an optimal curing period of no less than 28 days for OPC reference T(0.55), a design service life of at least 100 years is easily obtained. It should be noted that as soon as the applied curing period exceeds

the reference time period of 7 days, use of the mix specific b_c value as model input results in a longer depassivation period than using the default value (468 years versus 340 years). This is in contrast with the earlier calculation outcome for the 3-day curing period. This is simply a consequence of the mathematical form of the subfunction for curing in the prediction model of fib Bulletin 34 (fib, 2006). The choice of having a reference curing period of 7 days and the inclusion of this value in the denominator of the subfunction implies this.

Uncracked and fully healed HVFA concrete F50 was characterized by a much higher inverse effective carbonation resistance than OPC reference T(0.55) (Table 1: 11292 versus 2684 (mm²/years)/(kg/m³)). Evidently, this affected the service life prediction outcome quite drastically (Fig. 1b). With only 3 days of optimal curing the time to depassivation equaled only 18-24 years. Curing up to 28 days has a strong beneficial effect as it prolongs the depassivation period to 87-135 years. As such, a design service life of at least 100 years is still achieved under consideration of the HVFA mix specific regression exponent for curing b_c (-0.897) as estimated from literature data by (Burden, 2006). This demonstrates once more that it is highly important to determine b_c for each type of binder.

It seems worthwhile to incorporate 10% silica fume in the binder system. It lowers the susceptibility to carbonation of composition F40S10 significantly (Table 1: $R_{ACC,0}^{-1}$: 5835 (mm²/years)/(kg/m³)) as opposed to HVFA concrete F50. In terms of service life this means that the estimated depassivation period would amount to 28-45 years and 165-357 years for a 3-day and 28-day optimal curing period, respectively (Fig. 1c). Despite a higher carbonation resistance for mix F40S10, curing remains a key issue. A depassivation period exceeding 100 years cannot be achieved with only 3 days of curing, given a mix specific b_c value of -1.131. For a 28-day curing period it can be achieved.

From Fig. 1 it is clear that for none of the considered uncracked/healed concrete compositions it seems necessary to have a total of 28 days of optimal curing to have a depassivation period of precisely 100 years. Therefore, an additional series of probabilistic service life prediction calculations was conducted in Comrel to pinpoint for each concrete composition the curing period that corresponds with this event. These calculations were done while assuming the mix specific curing exponents b_c per concrete composition. The as such obtained results have been included in Table 2. The advised curing periods in view of a 100-year design service life for compositions T(0.55), F50 and F40S10 would amount to 4.2, 20.0 and 9.1 days, respectively.

Table 2. Mix specific optimal curing period for a design service life of 100 years.

Composition	T(0.55)	F50	F40S10
$t_{c\_100\ years}$	4.2	20.0	9.1
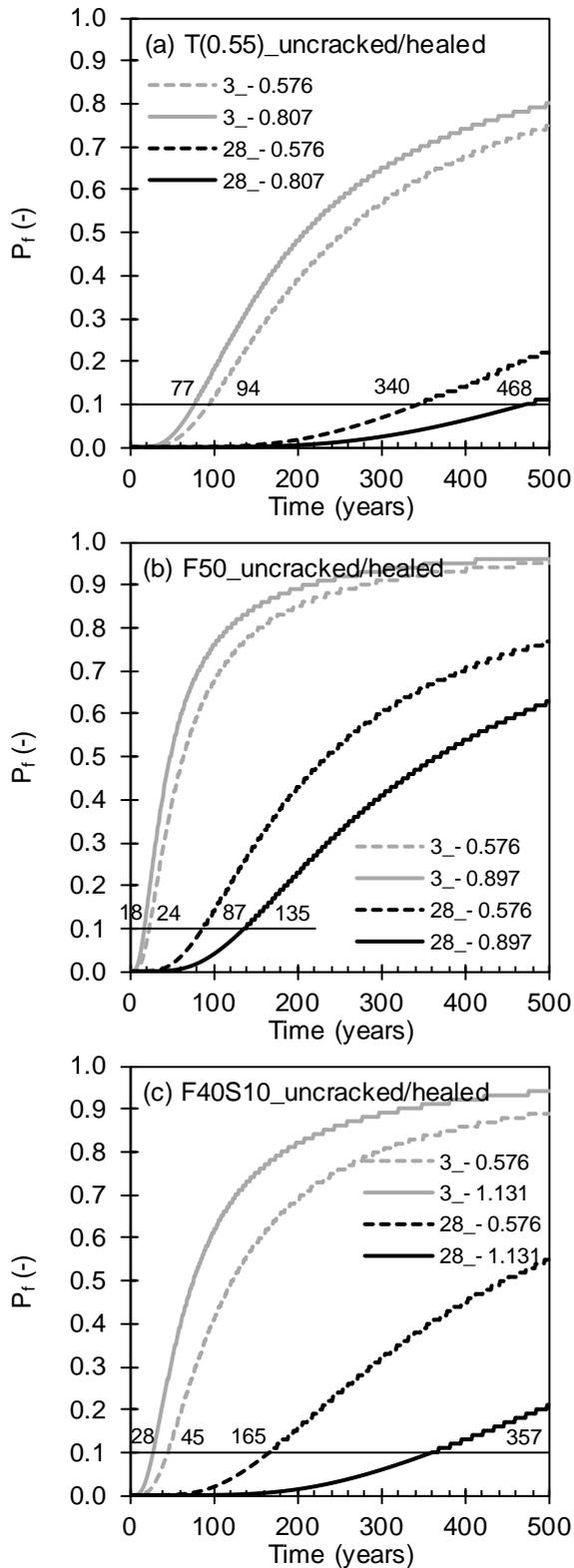
Fig. 1. Probability of failure P_f as a function of time for uncracked/fully healed T(0.55) (a), F50 (b) and F40S10 (c) concrete while assuming 2 curing periods (3 and 28 days) as well as default and mix-specific b_c values.

### 3.2 Service Life Cracked/Partially Healed Concrete

The service life assessment done so far related to the ideal condition of concrete being uncracked or in case of a self-healing concrete with a crack healing efficiency of 100%. These conditions are seldom encountered in practice. The low tensile strength of traditional concrete implies that concrete structures are almost never free of cracks. Moreover, although capsule breakage in PU-based self-healing concrete may be adequately triggered upon crack occurrence, the flow-out of the healing agent as well as the subsequent crack filling and sealing is not always full proof. As indicated earlier, the intended healing of a 25 mm deep, 300 μm wide artificially induced crack with an encapsulated low viscosity PU-precursor + accelerator + BPO was for 8 out of 9 samples fully successful. Still, one sample showed only partial healing.

Therefore, it was necessary to also study the effect of partial healing on the service life and make the comparison with the fully cracked condition. Such calculations were performed for the three binder systems under investigation, i.e. T(0.55), F50 and F40S10, while assuming mix specific b_c values and an optimal curing period of 28 days (Fig. 2). At the location of a 25 mm, 300 μm wide crack, the time to carbonation-induced steel depassivation for T(0.55), F50 and F40S10, with a cover depth of 35 mm, drops drastically to 24, 7, and 18 years, respectively (Fig. 2a). For each of the considered concrete mixes this is far below the envisaged design service life of 100 years. This means that multiple interventions will be needed in the course of time for repair actions. Partial healing of the crack with encapsulated PU more than doubles these time spans to 52, 15 and 40 years, respectively (Fig. 2b). Thus, incorporation of the proposed autonomous healing mechanism has a pronounced beneficial effect on the service life. However, repair actions will remain necessary within a timespan of 100 years. This means that one should always pursue to optimize potentially interesting techniques for self-healing to a 100% crack healing efficiency.

It should be noted that the now observed variation in service life performance between the different types of concrete in cracked and partially healed condition solely relates to the different curing behaviour of each concrete type. If one redoes the service life calculations while assuming per concrete type the required curing period for a 100-year service life in uncracked/fully healed state, then the time to steel depassivation for cracked and partially healed concrete would equal 5 and 11 years for all compositions.

Finally, from both Fig. 1 and Fig. 2 it is quite clear that the most preferable carrier for the encapsulated healing agent in carbonation exposed environments is obviously OPC reference T(0.55). True, with a

sufficiently long curing period a time to steel depassivation of at least 100 years could be achieved for all three binder systems under investigation when uncracked or perfectly healed. However, in case of a (partially healed) crack causing a serious reduction in concrete cover (from 35 to 10 mm), the increased carbonation rate of F50 and F40S10 from the crack tip onwards seriously shortens the depassivation period. This risk should be minimized by choosing for a carbonation resistant OPC carrier concrete in exposure class XC3.
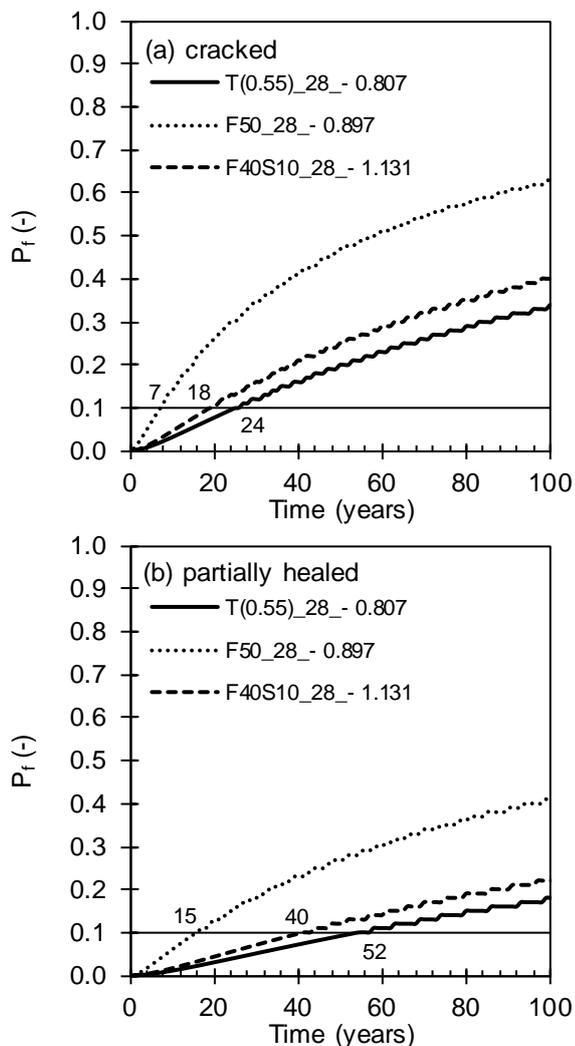
Fig. 2. Probability of failure P_f as a function of time for cracked (a) and partially healed (b) T(0.55), F50 and F40S10 concrete.

4.0   CONCLUSIONS

Although crack-free concrete with high replacement of the cement by fly ash is known for being more susceptible to carbonation than OPC concrete, probabilistic service life estimation cf. fib Bulletin 34 (fib, 2006) indicates that, if properly cured for a sufficiently long time period, carbonation-induced depassivation of steel embedded at a depth of 35 mm would not occur within an envisaged design service life of 100 years. For HVFA concrete with a

50% fly ash content that is characterized by a mix specific regression exponent for curing b_c of -0.897, the minimum optimal curing period should be at least 20 days. For FA+SF concrete containing 40% fly ash and 10% silica fume, with a mix specific b_c value of -1.131, 9.1 days of optimal curing would be sufficient. It should be noted though that these curing periods highly exceed the estimated 4.2 days that would suffice for the OPC reference in exposure class XC3. This reference was characterized by a mix specific b_c value of -0.807 that was determined experimentally. It should be emphasized that it is important to use mix specific b_c values in service life calculations. Using the default value of fib Bulletin 34 (= -0.576) for all types of binder systems tends to affect the outcome of the service life calculations significantly.

The presence of a 25 mm deep, 300 µm wide crack shortens the carbonation-induced depassivation period greatly for the studied OPC reference, HVFA and FA+SF concrete. While considering the optimal curing periods for a 100-year depassivation period in crack-free condition, the carbonation front at the location of a crack with this geometry is expected to reach the reinforcing steel after 5 years already. Giving the concrete self-healing properties by inclusion of encapsulated PU precursor + 5 wt.% accelerator + 5 wt.% BPO that is released upon crack occurrence could overcome this problem. However, a 100% crack healing efficiency that ensures return to the initial uncracked state should always be aimed for. In case of only partial crack healing, the depassivation period would only be extended from 5 to 11 years. Given the risk of insufficient curing and at the same time having an unhealed/partially healed crack, it remains important to use a carrier concrete for the capsules with healing agent with a high carbonation resistance, i.e. OPC concrete. In that perspective, self-healing concrete with high volumes of fly ash seems less suitable for exposure class XC3.

Acknowledgement

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