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Service life estimation of cracked and healed concrete in marine environment

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ABSTRACT: In the aggressive seawater environment, the durability of concrete is strongly influenced by the presence of chlorides and sulfates. Marine structures mostly have an important social function with a high economic impact, which makes durability a key issue. In addition, early-age cracks are a common problem, specifically for massive structural components. Repair of cracks is expensive and often impossible due to inaccessibility. Self-healing concrete is a promising solution to make marine structures more durable. In this study, capsules containing Polyurethane (PU) prepolymers were embedded in the concrete to release their contents when cracks appear. In cracked mortar, the chloride diffusion coefficients in the zone immediately around the crack significantly increased compared to uncracked mortar. The crack width dependency could be introduced into the service life model using a crack effect function. For crack widths in the range of 100 μm to 300 μm a service life decrease of around 80% was calculated. Autonomous crack healing had a beneficial influence on the resistance against chloride diffusion. However, for about one third of the cracks the healing mechanism failed, probably due to shifting of the tubes, tubes not rupturing properly, too high capillary forces in the tubes, etc. Nevertheless, on average, the service life of autonomously healed structures by means of encapsulated polyurethane increased with around 100% compared to cracked, unhealed structures. Moreover, in the most beneficial situation of proper healing, a service life increase of 150–550% was obtained, reaching values similar as for sound structures.

1 INTRODUCTION

A lot of deterioration is reported for constructions in marine environments. In this aggressive environment, the durability of concrete is strongly influenced by the presence of chlorides and sulfates, the main components of sea water. On the one hand, sulfate attack will degrade the concrete directly by forming expansive reaction products as well as strength decreasing products. On the other hand, chlorides will attack the concrete indirectly, by initiating corrosion of the reinforcement steel. In addition, concrete has a high sensitivity to crack formation because of its low tensile strength. That is why reinforcement is used in concrete to carry the tensile loads. Reinforced concrete has an increased resistance to cracking but does not eliminate it. This means that when cracks appear in concrete structures, aggressive substances like chlorides and sulfates may penetrate faster into the concrete and as a consequence faster deterioration can result. According to the standard NBN EN 1992-1-1 (2010), the allowable crack width in reinforced concrete structures depends on the environmental class and is in the range of 300 μm to 400 μm. However, according to Ismail et al. (2008) crack widths of 200 μm and wider allow unlimited chloride diffusion perpendicular to the crack faces. Djerbi et al. (2008) and Audenaert et al. (2009) found a bilinear relation between the chloride migration coefficient and the crack width. For crack widths between 0 μm and 100 μm, the migration coefficient increased with increasing crack width, and for crack widths between 100 μm and 200 μm this increase was less clear and the migration was rather constant. On the other hand, Sahmaran (2007) concluded that when the crack width increases, the diffusion coefficient increases as well, and this effect was even more distinct when the crack width was larger than 135 μm. Similarly, Kwon et al. (2009) found for on-site drilled cores that chloride diffusion coefficients kept increasing with crack widths up to 300 μm. Furthermore, apparent diffusion coefficients calculated by Mu (2012) show rising values with increasing crack width from 0 μm to 450 μm, and only when the crack width exceeded 450 μm, did the diffusion coefficient remain constant. In addition, his results indicate that concrete cracking accelerates chloride diffusion and steeply reduces the time to de-passivation of the reinforcement. Besides, the crack density had more significant influences on the de-passivation time of rebars than the crack depth, which was in turn more important than the crack width.

A possible solution to prevent maintenance and repair is self-healing of cracks in concrete (Van Tittelboom and De Belie, 2013, Van Tittelboom et al., 2011). Self-healing concrete has the ability to recover without external intervention. Nevertheless, specific data on degradation of self-healing concrete in aggressive environments are only sparsely available. These data are important to prove that an important service life extension of structures and concomitantly reduced repair and maintenance can be achieved by applying self-healing concrete.

2 MATERIALS AND METHODS

2.1 *Healing agent*

A commercially available polyurethane healing agent from BASF was used. One component of the two component agent consisted of a prepolymer of polyurethane (PU) which starts foaming in moist surroundings. The second component consisted of a mixture of 10% accelerator and distilled water. The expanding reaction of this healing agent may lead to an increase in volume of 25 to 30 times. This reaction is the driving force for the healing agent to flow out of the capsules. So, the additional volume created by the cracks may be filled up.

2.2 *Capsules with healing agent*

In order to carry the healing agent and to trigger the healing action upon crack appearance, tubular glass capsules were used for proof-of-concept, similar to the capsules used by (Van Tittelboom et al., 2011). Due to their high brittleness, the tubes will easily break whenever cracks appear in the mortar matrix. Borosilicate glass tubes with an internal diameter of 3 mm, an outer diameter of 3.35 mm and a length of 35 mm were used. It should be noted that polymeric tubes that can survive concrete mixing and yet release their contents upon crack formation, are currently under development.

First, the tubes were sealed with polymethylmethacrylate at one end. Then half of the tubes were filled with the prepolymer and the other half was filled with a mixture of accelerator and water, through injection by means of a syringe. Afterwards, the open ends were sealed as well.

2.3 *Samples with(out) self-healing properties*

Cylindrical PVC moulds with a height of 50 mm and an inner diameter of 100 mm were used to produce all of the specimens. These moulds were clamped on a wooden plate in order to prevent leakage of concrete and mortar at the bottom. For the self-healing mortar samples (coded PU), nylon wires were attached to the PVC moulds and the capsules were glued on them by means of a droplet of MMA in order to prevent the capsules from floating during casting. Six capsules were equally divided along the diameter over two layers, at 12.5 mm and 37.5 mm height. First, a 20 mm mortar layer was brought into the moulds. When this layer was compacted on a vibration table, the moulds were completely filled with mortar and vibrated again. The mortar was prepared with Ordinary Portland Cement CEM I 52.5 N, a sand 0/4 over cement ratio of 3/1 and a water/cement ratio of 0.45. The reference samples without self-healing properties (uncracked – coded REF; cracked – coded CR) were prepared in the same way as described above, however, samples belonging to these series contained no glass tubes. After casting, the specimens were placed in an air-conditioned room with a temperature of 20°C and a relative humidity of at least 95%. Demoulding took place the next day whereupon the specimens were stored again under the same conditions until the age of 21 days. Then, the specimens were coated with epoxy at the sides. This coating acted as a kind of reinforcement during crack formation. The coating was allowed to harden for 3 days under laboratory conditions.

2.4 *Crack formation and healing*

Realistic cracks of 100, 200 or 300 μm were produced by means of a crack width controlled splitting test. The embedded tubes broke during crack formation and both components of the healing agent were released into the crack due to capillary forces. Upon contact of both components, PU foam was formed, resulting in crack healing. The polymerization reaction is usually complete within one day. After crack formation, the outermost 5 mm (casting surface) was cut off. Then, the specimens were coated again except for the test surface. Before immersion in the test solutions, the crack widths were measured microscopically.

2.5 *Chloride diffusion tests*

The resistance to chloride penetration of reference and autonomously healed mortar samples, was evaluated experimentally using the diffusion test as described in (NT Build 443, 1995). For every test series, at least 9 specimens were tested of which at least 3 were used for obtaining chloride profiles. At the age of 28 days, the coated specimens were placed in a 4 g/l $Ca(OH)_2$ solution for 7 days. Afterwards, the specimens were placed in a closed container with a 165 g/l NaCl solution.

After 7 weeks storage in the test solution, the specimens were split orthogonal to the crack and the chloride penetration was visualized by spraying 0.1 Mol/l $AgNO_3$ on both halves of the split specimens. Next, total chloride profiles were obtained by means of potentiometric titration (Maes, 2015). To do so, powder was collected by grinding layers of 2 mm thickness up to a depth of 20 mm. Powder was first collected in a zone of 18 × 75 mm^2 with the crack in the middle (coded C); afterwards, powder was collected from separated zones of 28.5 × 75 mm^2 both 9 mm away from the crack (coded R) (Figure 1). As such, the total ground surface had a diameter of 75 mm.

Non-steady-state diffusion coefficients and chloride surface concentrations were obtained by fitting Equation 1.

$$C(x,t) = C_s - (C_s - C_i)\,\mathrm{erf}\left(\frac{x}{\sqrt{4D_{nssd}t}}\right) \quad (1)$$

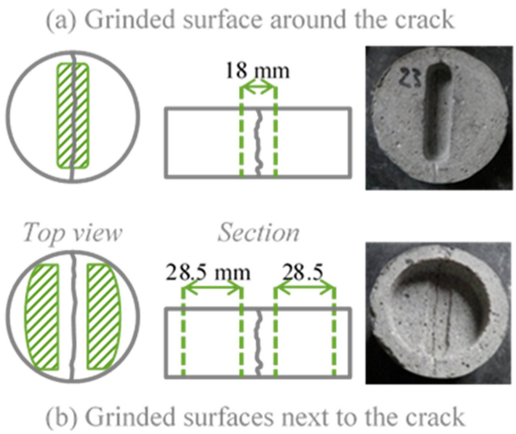

Figure 1. Grinding method to collect powder around the crack (a) and next to the crack (b).

where $C(x, t)$ is the chloride concentration at depth x and time t (mass % concrete), C_i the initial chloride concentration (mass % concrete), C_s the chloride concentration at the surface (mass % concrete), D_{nssd} the non-steady-state diffusion coefficient (m^2/s), x the distance from the surface until the middle of the considered layer (m) and t the exposure time (s).

It is reasonable to assume that the initial chloride concentration C_i equaled 0% (in reality, this value can differ from 0% and in current research an average concentration of 0.15 ± 0.05 wt.% binder was found). The first layer was excluded from the regression analysis, since the measured chloride concentration in this layer is generally considered not representative.

3 RESULTS AND DISCUSSION

3.1 *Chloride penetration profiles*

Figure 2 shows the total chloride profiles for specimens with initial crack widths of 100 µm and 300 µm, respectively. Each profile is the average of three individual profiles and refitted with the obtained diffusion coefficient.

It is clear that the highest chloride concentrations at every depth are found in the 18 mm wide zone with the crack in the middle, regardless of the crack width. Next, it can be seen that in the zones further away from the healed cracks the profiles are comparable to chloride profiles obtained for uncracked mortar specimens (REF) which implies a limited chloride penetration perpendicular to the crack wall. In between these two profiles, the profiles obtained in the zone around the healed cracks are situated. So, based on these chloride profiles it seems that autonomous crack healing has a beneficial influence on the resistance against chloride penetration compared to cracked mortar. Note that the PU polymerization reaction occurs very fast (within one day) and the healing action could be considered as completed, before immersion of the specimens in the chloride solution. Nevertheless, the chloride concentrations are higher than in uncracked mortar.

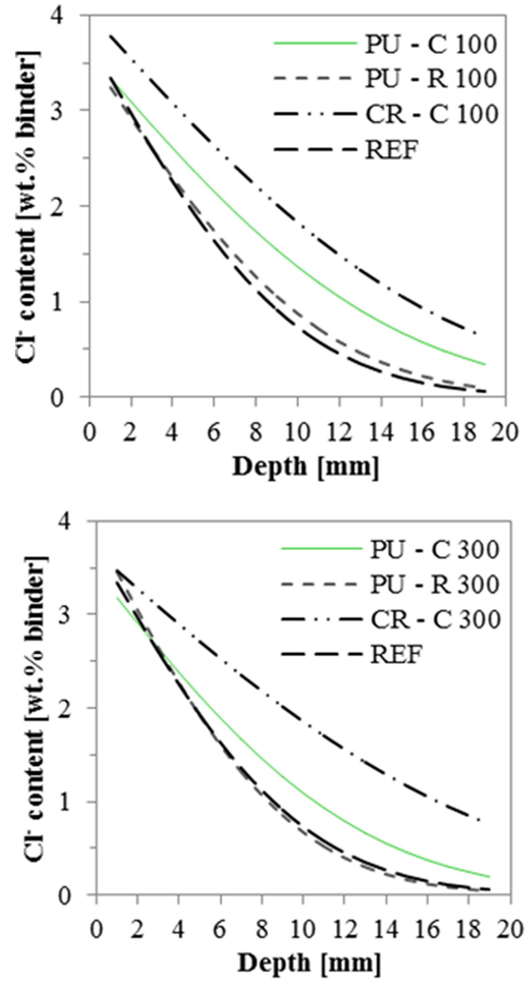

Figure 2. Average total chloride profiles around (C) and further away from (R) the cracks for healed (PU) and unhealed (CR) mortar specimens with an initial crack width of 100 µm (top) and 300 µm (bottom) and an uncracked reference (REF), obtained after 7 weeks immersion in NaCl solution.

To study the efficiency of the healing mechanism itself the individual chloride profiles per specimen are shown in figure 3.

It can be seen that for none of the three tested specimens was a similar profile measured. For specimens with a 100 µm crack only one of the three tested specimens was (almost) totally healed (PU – C 3) while one showed no healing at all (PU – C 1). The other specimen (PU – C 2) showed a chloride profile similar to the average profile in Figure 2, namely in between healed and unhealed. The same trends were found for initial crack widths of 200 µm and 300 µm.

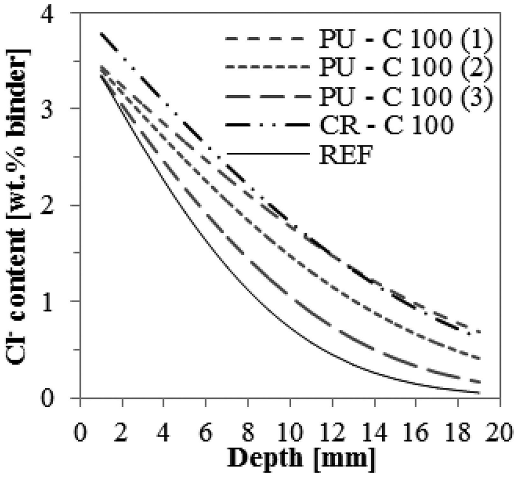

Figure 3. Individual total chloride profiles around the crack for autonomously healed mortar specimens with an initial crack width of 100 μm, after 7 weeks immersion in NaCl solution.

3.2 *Chloride diffusion coefficients*

Figure 4 gives the individual total chloride diffusion coefficients with an indication of the mean value per test series.

In general, regardless of the crack widths, the average diffusion coefficients in the zone around a PU-healed crack are lower than the diffusion coefficients in the zone around an unhealed crack, indicating the beneficial influence of PU-crack healing. In addition, based on the statistical analysis (Student-Newman-Keuls test with level of significance = 0.05), it seems that the diffusion coefficients obtained for healed mortar with initial crack widths of 100 μm are not significantly different from the diffusion coefficients obtained for healed mortar with initial crack widths of 300 μm. Furthermore, it is clear from the statistical analysis that none of the chloride diffusion coefficients measured around a 100 μm healed crack or a 300 μm healed crack are significantly different from the reference diffusion coefficients in uncracked mortar.

From Figure 4, it can also be observed that the scatter on the diffusion coefficients measured around the PU-healed cracks is clearly larger than those on the other diffusion coefficients. This was also clear from the individual chloride profiles and can be explained by the fact that only a part of the PU-healed cracks are really sealed. The latter is probably due to falling short of the healing mechanism rather than the fact that chlorides penetrate through the PU-foam which should fill up the crack. During a visual inspection of the specimens and the glass tubes after splitting, it was observed that in the autonomously healed specimens without improved resistance against chloride penetration, the healing agents were still (partially) in the tubes, maybe due to the higher capillary forces in the capsules than in the cracks. Furthermore, some

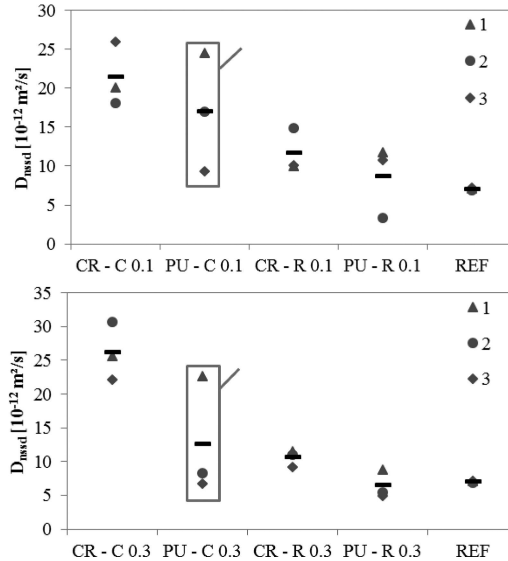

Figure 4. Total chloride diffusion coefficients in different zones around (C) or further away from (R) a 100 μm (top) or 300 μm (bottom) crack, after 7 weeks immersion in a 165 g/l NaCl solution. With indication of individual and mean values.

tubes may have shifted if not properly fixed, which could explain why the PU-prepolymer did not react with the accelerator causing a fast foaming reaction. Also in some cases, crack formation may not trigger the healing mechanism.

4 SERVICE LIFE ESTIMATION

To evaluate the influence of autonomous crack healing in concrete/mortar in a quantitative way, the service life prolongation is estimated with regard of the service life for cracked structures. To do so, a full probabilistic design approach is used.

4.1 *Apparent chloride diffusion coefficient*

The apparent chloride diffusion coefficient is calculated starting from the diffusion coefficient obtained by means of curve fitting based on Equation 1. These coefficients are tabulated in Table 1. Remark that the unit of the calculated diffusion coefficients is mm²/year (=31.54 × 10^{-12} m²/s).

Concerning the chloride diffusion coefficient for autonomously healed mortar in the zone around the crack (PU-C), a mean value can be calculated regardless of the crack width. This mean chloride diffusion coefficient amounts to 447 ± 219 mm²/year. Furthermore, the influence of autonomous crack healing will also be taken into account by means of the minimum and maximum calculated values per initial crack width.

On the other hand, the chloride diffusion coefficient for unhealed samples (CR-C) changes (to a limited

Table 1. Chloride diffusion coefficients for cracked (CR-C) and autonomously healed (PU-C) OPC mortar, after 7 weeks immersion in a 165 g/l solution.

D_{nssd}	100 μm		200 μm		300 μm		
[mm²/yr]	CR-C	PU-C	CR-C	PU-C	CR-C	PU-C	REF
Min.	572	327	662	189	762	213	215
Max.	758	776	757	599	903	716	226
Mean	654	546	704	400	824	396	222
St dev	95	224	48	205	72	278	5

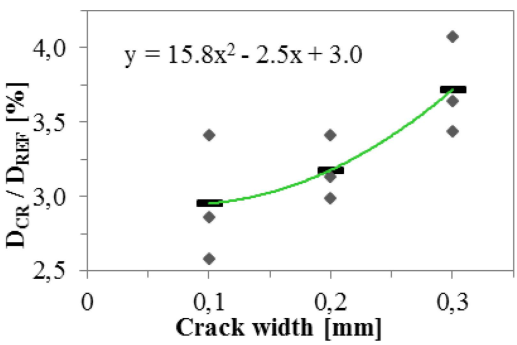

Figure 5. Relation between chloride diffusion ratio (coefficient for cracked mortar/coefficient for uncracked mortar) and crack width.

extent) in function of the crack width 'w'. So, a crack effect function is suggested based on a regression analysis as shown in Figure 5.

This crack effect function is implemented in the equation to calculate the apparent chloride diffusion coefficient by means of the parameter k_{cr}. So in this particular case, the apparent chloride diffusion coefficient can be calculated from equations (2) to (5).

$$D_{app,C} = k_e \cdot D_{nssd,REF} \cdot k_t \cdot W(t) \cdot k_{CR} \quad (2)$$

$$k_e = \left(b_e \left(\frac{1}{T_{ref}} - \frac{1}{T_{real}} \right) \right) \quad (3)$$

$$W(t) = \left(\frac{t_0}{t} \right)^a \quad (4)$$

$$k_{CR} = 15.8\, w^2 - 2.5\, w + 3.0 \quad (0.1 \text{ mm} \leq w \leq 0.3 \text{ mm}) \quad (5)$$

with the apparent chloride diffusion coefficient D_{app} (mm²/years) calculated starting from the chloride diffusion coefficient for uncracked mortar $D_{nssd,REF}$ (mm²/years) taking into account a transfer parameter k_t (–), an ageing function W(t) with the ageing exponent a (–), the reference point of time t_0 (years) and the time t (years), an environmental factor k_e (–) including a regression variable b_e (K) as well as the standard test temperature T_{ref} (K), the temperature of the structural element or the ambient temperature T_{real} (K), and at last a crack effect function k_{CR} (–) with the crack width w (mm).

Table 2. Default input parameters for service life prediction.

Parameter		Mean	St. Dev.	Lower bound	Upper bound
C_{crit} [wt%]	Beta	1.90	0.15	0.2	3
C_0 [wt%]	Normal	0.10	0.025	–	–
$C_{s,\Delta x}$ [wt%]	Normal	3.0	0.8	–	–
d [mm]	Lognorm	60	8	–	–
Δx [mm]	Constant	0	–	–	–
b_e [K]	Normal	4800	700	–	–
T_{ref} [K]	Constant	293		–	–
T_{real} [K]	Normal	293	5	–	–
$D_{nssd,REF}$ [mm²/year]	Normal		Table 1 (REF)		
k_t [–]	Constant	1	–	–	–
t_0 [year]	Constant		0.211 (28 days curing + 49 days immersion)		
a [–]	Beta	0.40	0.12	0	1
w [mm]	Constant	[0.1–0.3]	–	–	–

4.2 *Effect of crack width on service life*

The results of the service life prediction can vary a lot depending on the assumed values for the concrete cover, the critical chloride content, etc. In this paper the influence of these parameters on the specific service life is not discussed since the aim is to evaluate the effect of cracks in cementitious materials and the autonomous healing.

The default parameters as described in fib Bulletin 34, Duracrete or the CUR guideline VC 81 are taken into account, see Table 2, however some of them are adapted in order to have a reference service life of about 100 years. This is obtained by changing the concrete cover and the ageing factor in accordance with acceptable values found in literature (Van den Heede et al., 2014), 60 ± 8 mm and 0.40 ± 0.12, respectively. In addition, the crack effect function parameter k_{cr} is introduced in the model taking the crack width w into account.

The results considering the influence of crack widths between 100 and 300 μm (CR – xx) are shown in Figure 6. The curves indicating the probability of failure P_f in function of time for cracked cementitious materials are plotted against the same curve for sound material (REF). The end of service life corresponds to a probability of failure of 0.10. It is clear that the presence of cracks induces a significant service life reduction. The service life for structures decreases from 103 years for the uncracked situation to 23 years in case of cracks with a width of 100 μm, and 20 and 16 years for 200 and 300 μm wide cracks. The obtained values are in reasonable agreement with the results obtained by Kwon et al. (2009) for in-situ cracked concrete of port wharves which had been operated for 8 and 11 years.

It is furthermore described in literature that microcracks caused by service loads should be taken into

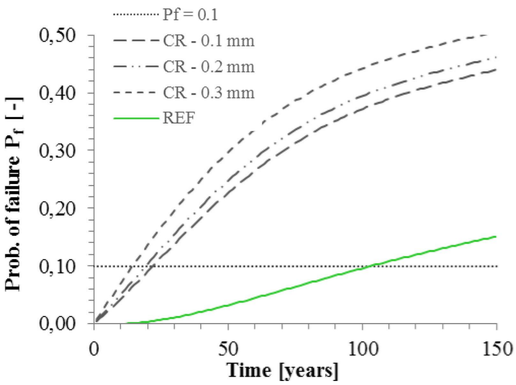

Figure 6. Service life prediction for cracked OPC mortar.

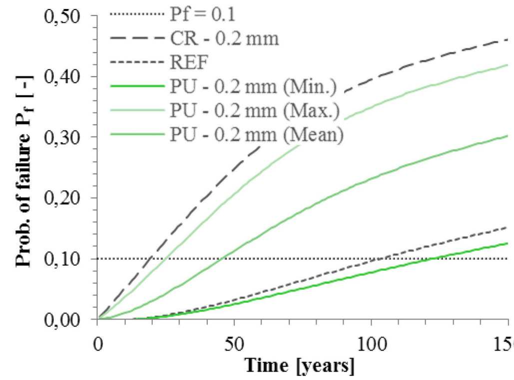

Figure 7. Service life of healed OPC mortar with initial crack width 200 μm.

account during service life prediction. The chloride migration coefficient of concrete under loading is different and can be significantly higher than that without loading. Concrete in aggressive environments with service loading of over 50% of the ultimate compressive stress can have a 2–4 times higher chloride migration coefficient compared to concrete without loading (Wang et al., 2016). However, when a permanent compressive load of 30% of the ultimate load is applied, the average diffusion coefficient decreased to about 70% of the diffusion coefficient for an non-loaded concrete at the same age (Egüez Alava et al., 2015).

4.3 *Effect of autonomous crack healing*

In order to report the effect of autonomous crack healing, different points of view are possible. Firstly, the averaged effect is studied based on the mean chloride diffusion coefficient for autonomously healed mortar, regardless of the initial crack width. This coefficient is used as an input parameter in the model without k_{CR}. Based on the averaged effect of autonomous healing, the service life is higher than for cracked mortar (Figure 7). Nevertheless, it is still clearly lower than the time to reinforcement depassivation in sound mortar. More specifically, the service life of healed mortar amounts to 32 years. This is an increase of 39%, 60% or 100% when compared to the service life for cracked mortar with crack widths of 100 μm, 200 μm and 300 μm, respectively.

So the averaged effect of autonomous crack healing by means of encapsulated PU is rather positive. To take the non-constant efficiency of autonomous crack healing into account, service lives were predicted for every initial crack width individually based on the maximum, minimum and mean measured chloride diffusion coefficient for healed mortar. Since the input parameter in the estimation of the service life for cracked mortar is an average value, it is possible that an estimation of healed mortar based on a single value results in a slightly lower service life. This is the case when the maximum measured value for the chloride diffusion coefficient in autonomously healed

Table 3. Increase of the service life for autonomously healed mortar predicted by means of the maximum, mean and minimum measured chloride diffusion coefficient and compared to cracked mortar.

Crack width [mm]	Service life increase compared to cracked mortar [%]			
	PU – Max.	PU – Mean	PU – Min.	REF
0.1	–26	22	154	348
0.2	25	120	510	415
0.3	19	181	550	544
Mean	6	108	405	436
St. Error	16	46	126	57

mortar with initial crack width 100 μm is considered, which leads to a service life of 17 years. On the other hand, the prediction based on the mean and the minimum observed value for autonomously healed mortar with initial crack width 100 μm results in a service life increase to 28 and 58 years respectively. In Figure 7 the probability of failure in function of the time for healed mortar with initial crack widths of 200 μm is visualised. The service life predicted based on the maximum measured chloride diffusion coefficient for autonomously healed mortar is equal to the service life for cracked mortar, namely 25 years. On the opposite, the calculation with the minimum measured coefficient leads to a service life of 122 years, equal to the one for sound mortar. In addition, the mean diffusion coefficient leads to 44 years.

For autonomously healed mortar with initial crack widths of 300 μm, similar observations can be made. The predicted service lives amount to 19 years, 45 years and 104 years based on the maximum, the mean and the minimum measured chloride diffusion coefficients. Starting from the aforementioned results, relative differences are calculated compared to the cracked mortar. These increases and decreases are tabulated in Table 3.

It can be observed that almost all samples benefit from a service life increase through self-healing.

Except for the samples with initial crack widths of 100 μm, this relative increase seems independent of the initial crack width. In general, it can be concluded that in the worst case scenario, an average increase of 6% is obtained, assigned to autonomous healing. In the most beneficial situation an increase of 405% is obtained. On average, autonomous crack healing induces a service life increase of 108% compared to the case without healing. In practice, the effect of self-healing will have to be weighed against other options, such as the use of stainless steel in XS3 environments.

5 CONCLUSIONS

Chloride penetration increases when a mortar is cracked and there is a limited effect of the crack width (for cracks of 100 μm to 300 μm).

The chloride diffusion coefficients in the zone immediately around the crack (9 mm at each side) significantly increase compared to uncracked mortar. For crack widths in the range of 100 μm to 300 μm, the relation between the chloride diffusion coefficient ratio and the crack width can be expressed through a crack effect function (equation 5). Service life of mortar/concrete structures containing cracks decreases with 81% compared to sound structures. These conclusions indicate the need for a proper crack repair mechanism. One of the solutions is self-healing concrete/mortar by means of encapsulated polyurethane.

Autonomous crack healing of mortar by means of encapsulated polyurethane has a beneficial influence on the resistance against chloride diffusion. In the most beneficial situation, a service life increase of 154% up to 550% can be obtained by autonomous crack healing compared to cracked unhealed structures, leading to a service life similar to the one for sound structures. On average, the service life of autonomously healed structures by means of encapsulated polyurethane increases 108 ± 46% compared to the service life of a cracked, unhealed structure.

If the healing mechanism works properly and the crack is sealed well, (almost) no chlorides will penetrate along the crack. So, autonomous crack healing by means of encapsulated polyurethane is able to improve the durability and increase the service life of mortar/concrete structures in chloride-containing environments.

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