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Moisture-induced Debonding in Concrete-epoxy Interface

Concrete-epoxy interface can be found in many strengthened or retrofitted structures, especially in Hong Kong where concrete structures are the majority. In practice, epoxy is usually used in repairing concrete structures because of its excellent adhesion, chemical and heat resistance, good mechanical properties and electrical insulating properties. The bond between concrete and epoxy has been found to be strong enough such that failure does not occur at the interface under dry condition. However, prior research studies have indicated that moisture-affected debonding in a concrete-epoxy bonded system is a complex phenomenon that may often involve a distinctive dry-to-wet debonding mode shift from material decohesion (concrete delamination) to interface separation in which concrete-epoxy interface becomes the critical region of failure. Such premature failures may occur regardless of the durability of the individual constituent materials forming the bonded material systems. In this paper, a review on moisture degradation of concrete-epoxy interface is presented, followed by an explanation of such phenomenon using an atomistic approach which can fundamentally describe the interaction between the bonded system and the surrounding water molecules. It is highly recommended that the durability of concrete-epoxy interface should be carefully considered in the design stage due to the detrimental moisture effect.

Keywords: *Concrete, Epoxy, Fracture, Interface, Moisture, Molecular Dynamics*

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Introduction

Concrete-epoxy interface can be found in various structural applications including strengthening and retrofitting of existing reinforced concrete (RC) using fiber-reinforced polymer (FRP) materials or steel plates. In Hong Kong, such interface can be readily found when polymers are used for repairing RC surface. Epoxy is desirable for this application from both economical and technical considerations in a sense that the deteriorated RC should be retrofitted with impermeable material which can be well bonded with the parent concrete. Interface between concrete and epoxy is usually regarded to be stronger when compared to the strength of the concrete substrate as found in many existing data on the failure of FRP-bonded concrete beams (Teng *et al.*, 2002). This is generally true when the concrete-epoxy interface is under dry condition, as evidenced by the existing experimental data (Lau and Büyüköztürk, 2010). However, based on these prior research studies, it is found that the concrete-epoxy interface may represent the most critical region when subjected to moisture (Lyons *et al.*, 2002; Au and Büyüköztürk, 2006; Ouyang and Wan, 2008). In Hong Kong, the aging problem of existing RC structures becomes an important social issue and a safe and reliable technique is required for strengthening and retrofitting purposes. A complete understanding on the durability of concrete-epoxy interface is therefore necessary in order to ensure the public safety against the potential danger due to the presence of such interface in the repaired structures. In the following sections, a review of concrete-epoxy interface research with a focus on the severe moisture damage on such interface will be discussed, followed by a fundamental explanation of this phenomenon using an atomistic approach. The objective of this paper is to deliver an understanding on this complex moisture-induced debonding in concrete-epoxy systems with a fundamental explanation using an atomistic approach. It is hoped that, through this review paper, the practising engineers are more aware of the structural integrity of concrete-epoxy interface, especially in a humid region such as Hong Kong.

Strength-based and Fracture-based Approaches

The research methodology in the past few decades on the interfacial debonding issue can be differentiated into two major categories, namely the strength-based approach and the fracture-based approach. Strength-based models are most widely used to predict the global debonding failure of FRP bonded concrete systems, which consists of concrete, epoxy and FRP, at a structural scale. These models have been developed based on interfacial stresses, concrete tooth, and bond strength concepts (Smith and Teng, 2001) and they all implicitly assume a debonding mode involving material decohesion that the material limits are being utilised. It is currently known that high shear and normal stresses always develop at plate ends and at crack locations within a retrofit span. Approximate closed form models have been developed to determine such stresses, with a majority based on the strain compatibility condition and linear elastic behaviour of materials (Toutanji and Gomez, 1997; Malek *et al.*, 1998; Smith and Teng, 2001). By inputting these computed interfacial stresses into a failure criterion, a debonding failure load could be predicted. Early research suggested that debonding failure takes place when the maximum shear and normal stresses reach certain limits (Roberts, 1989; Triantafillou and Plevris, 1992).

Strength-based approach is good if the material system is free of cracks (no singularities). However, in reality, cracks always exist in various length scales and such an approach intrinsically neglects the failure process of local debonding regions, which is closely related to the fracture properties of the bi-material system. Hence, it is necessary to find another model which can capture the effect of local defects or cracks on an entire structural element. Fracture-based approach is suitable for this purpose. There is a limited number of fracture-based models which have also been proposed to describe debonding as a local consideration. Here, some basic knowledge of fracture mechanics should be given. Based on the concept of linear elastic fracture mechanics, it is known that stress intensity factor is the only parameter to describe the fracture process. At the critical stage when crack is initiated, the corresponding stress intensity factor is regarded as the fracture toughness which is a material property. Interface fracture toughness (Γ), which refers to the critical energy release rate in a bi-layer material system, can be used to quantify the resistance to crack initiation in local regions, and to predict the crack propagation based on the kink criterion (He and Hutchinson, 1989). The derivation of the interface fracture toughness through experiment can be found in Suo and Hutchinson (1990). Fracture-based mechanical tests aim at determining the fracture toughness required to produce the debonding failure in a bi-layer material system. Different fracture-based test procedures have been proposed for the determination of bond properties of composite layered materials. Detailed discussions of these procedures and their applicability for predicting the bond behaviour can be found in various publications (Taljsten, 1996; Giurgiutiu *et al.*, 2001; Qiao and Xu, 2004; Yuan *et al.*, 2004; Au and Büyüköztürk, 2006; Coronado and Lopez, 2008; Lau and Büyüköztürk, 2010). Two common types of fracture tests are described in this paper for readers' interest.

To study the bonding between concrete and epoxy, fracture tests using modified double cantilever beams (MDCB) were usually adopted (Karbhari and Engineer, 1996; Karbhari *et al.*, 1997; Wan *et al.*, 2006; Ouyang and Wan, 2008; Ouyang and Wan, 2009). With these specimens, loads can be easily applied at the end of FRP strips. Boyajian *et al.* (2005) devised a single contoured-cantilever beam specimen to facilitate mode I (opening mode as shown in Fig 1) ie fracture characterisation of FRP-concrete bonded system under various environmental conditions. This specimen is derived from the conventional double-cantilever beam (DCB) but only employs a single contoured arm such that a constant compliance gradient is attained for a convenient computation of fracture toughness. Qiao and Xu (2004), on the other hand, developed a notched three-point

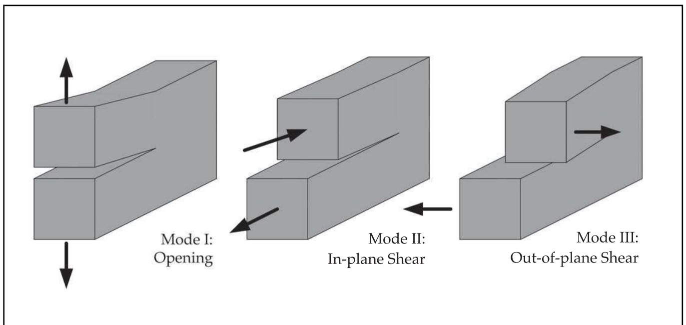

Figure 1 – There are three ways of applying a driving force to enable crack propagation: mode I crack – opening mode (tensile stress normal to the plane of the crack); mode II crack – sliding mode (shear stress acting parallel to the plane of the crack and perpendicular to the crack front); mode III crack – tearing mode (shear stress acting parallel to the plane of the crack and parallel to the crack front)

bending beam that employs a fictitious crack model based approach to characterise mode I fracture of FRP-concrete bonded regions. It was found from this model that the interface fracture toughness is not strongly dependent on the loading rates but increases with increasing epoxy curing time. Wan *et al.* (2004) attempted to assess the mixed-mode fracture energy by means of crack opening displacement (COD) measurement using computer vision on MDCB specimens. However, the interface fracture toughness estimated by these tests was often with an order of magnitude higher than the fracture energy of the concrete substrate (Kaplan, 1961; Moavenzadeh and Kuguel, 1969; Brown, 1972; Jenq and Shah, 1985). This phenomenon has been found to be not realistic when the material system is in wet condition (Au and Büyüköztürk, 2006; Lau and Büyüköztürk, 2010). In fact, the mechanical test involving primarily bending of the film may not be representative in quantifying the interface fracture toughness of the bonded system because a significant portion of the applied work is converted to the plastic strain energy in the thin layered material that is not related to the crack initiation or propagation. Such plasticity in the constituent material eventually leads to the over-estimation of the interface fracture toughness. The four-point bending tests on sandwiched beam specimens have been shown to be robust in quantifying the interface fracture toughness in bi-layer materials by incorporating the relationship between the interface fracture toughness and the phase angle (Dundurs, 1969). Experimental studies using this type of specimen for the determination of fracture toughness at the concrete-epoxy interface can be found in recent publications (Lau and Büyüköztürk, 2010; Qiao *et al.*, 2011).

Severe Degradation of Concrete-epoxy Interface under Moisture

The research on concrete-epoxy interface is greatly initiated by the durability issue of the FRP-bonded concrete system. Many studies have shown that the structural performance of FRP retrofitted systems can be impeded due to the introduction of various environmental effects, and the debonding in the FRP-concrete bonded region may play a major role in such premature failures (Karbhari *et al.*, 2003; Abanilla *et al.*, 2006a; Abanilla *et al.*, 2006b; Boyajian *et al.*, 2007; Imani *et al.*, 2010; Cromwell *et al.*, 2011). Experimental works by Karbhari and Engineer (1996) and Grace (2004) investigated the influence of several environmental conditions such as salt water, moisture, heat, and alkalinity on the structural performance of FRP-strengthened concrete beams. In particular, moisture effect was identified as an important environmental deterioration mechanism promoting premature system failures. Debonding was observed as one of the main failure modes (Grace, 2004). Findings in these investigations regarding the detrimental effect of moisture were in agreement with other independent studies conducted on FRP-plated concrete that were subjected to accelerated wet/dry cycles. Reductions in ultimate strength and stiffness due to such cycling were generally observed. Chajes *et al.* (1995) reported decrements of 20% – 30% in ultimate strength for FRP retrofitted specimens which were subjected to 100 wet/dry cycles. In one study (Myers *et al.*, 2001), it was experimentally determined that FRP-bonded concrete specimens can lose up to 85% of its flexural stiffness after 20 wet/dry cycles. Ultimate failure load also depreciates at various degrees, depending on the type of epoxy being used. It was reported that the range of the observed strength reductions was up to 33% on FRP-plated concrete beams that were subjected to 300 wet/dry cycles in salt water (Toutanji and Gomez, 1997). Failure was reported as a debonding mode that generally took place near the FRP-concrete bonded region.

Based on the above reported findings, it shows that the bonded region between FRP and concrete is critical under the effect of moisture. For a further investigation on this particular region, a fracture approach has been adopted. The severe degradation of concrete-epoxy interface under moisture has been demonstrated in various kinds of fracture samples including the peel specimens and the sandwiched bending beam specimens. It was reported that the interface fracture toughness dropped significantly after exposure to the 100°C wet environment (Lyons *et al.*, 2002). Tests on MDCB after wet/dry cycles also showed a marked reduction in fracture toughness of FRP-concrete bonded region (Qiao and Xu, 2004). Moisture affects not only the adhesive bond after bonded systems are in service, but also during the application of FRP on concrete surface. Tests on Carbon FRP bonded to concrete with initially damp surface using a modified cantilever beam indicated reduction in bond strength for both dry and moisture-conditioned specimens (Wan *et al.*, 2006). A shift in failure mode has also been observed for both glass and carbon FRP bonded concrete systems before and after moisture conditioning. Control specimens usually exhibit a conventional concrete fracture type of failure, while moisture-conditioned specimens fail mostly at the concrete-epoxy interface as shown in Fig 2 (Au and Büyüköztürk, 2006). This indicates that an interface weakening mechanism is activated by the presence of water molecules. Since the failure under the effect of moisture generally occurs by either concrete delamination or concrete-epoxy interface separation, some studies have been devoted to determining the extent of degradation of bi-material systems. Results from fracture test on bi-material specimens have shown that the interface between concrete and epoxy become weakened due to moisture exposure (Frigione *et al.*, 2006).

As motivated by this interesting shift in failure mode, a careful material characterisation of concrete-epoxy interface with the consideration of moisture effect was conducted using the four-point bending test setup (Lau and Büyüköztürk, 2010). In that experiment, sandwiched beam specimens were used for quantifying the mode I interface fracture

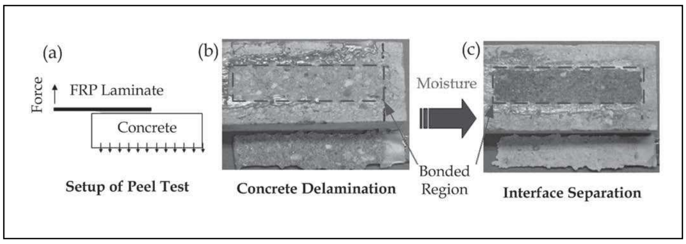

Figure 2 – (a) Schematic Diagram of the Experimental Setup of Peel Test, (b) Failure Mode of Dry FRP-bonded Concrete Specimen, (c) Failure Mode of Wet FRP-bonded Concrete Sample (Au and Büyüköztürk, 2006)

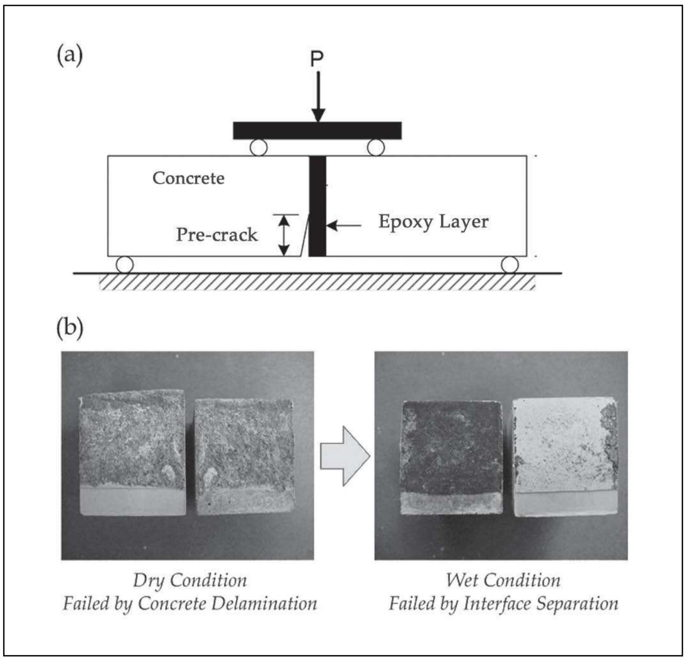

Figure 3 – (a) Schematic Diagram of the Four-point Bending Test, (b) Failure Surface after the Bending Test (Lau, 2010)

toughness. Typical failure modes of the sandwiched bending beam specimens with and without moisture conditioning are shown in Fig 3(a). Fig 3(b) shows the failure surfaces of both the dry sandwiched bending specimen and the wet specimen with a four-week moisture duration (Lau, 2010). The good agreement in the observed failure mode from different experiments further validates the detrimental effect of moisture on concrete-epoxy interface.

By observing the sandwiched beam specimens carefully, it was noticed that dry specimens exhibited failure in concrete itself, rendering a cohesive type of failure. The pre-crack, which was placed at the concrete-epoxy interface, kinked into the concrete upon reaching the peak load and the specimen failed by concrete delamination. Failure surface was felt powdery to a touch and small grains of sand were clearly seen at the failure surface. Wet sandwiched bending beam specimens, on the other hand, exhibited a distinctive concrete-epoxy interface separation. For the specimens under moisture conditioning of two weeks and four weeks, a relatively small amount of loose concrete particles adhered to the epoxy layer. However, for the specimen under moisture conditioning over four weeks, a clear separation between concrete and epoxy was observed. It was noted that the crack did not kink into concrete substrate at all during the entire testing process, but remained propagating along the moist concrete-epoxy interface. It was observed that the shift of fracture failure mode from concrete delamination to interface separation is accompanied by a substantial decrease of the mode I interface fracture toughness. Such trend has also been reported in FRP-bonded concrete system (Lyons *et al.*, 2002; Au and Büyüköztürk, 2006; Ouyang and Wan, 2008). It is noticed that the change of the interface fracture toughness has a strong correlation with the shift of failure mode. The variations of mode I fracture toughness at concrete-epoxy interface under different moisture durations are shown in Fig 4 (Lau, 2010). It is noticed that there is a 50% reduction in the interface fracture toughness and an asymptotic behaviour can be achieved with increasing moisture ingress. The deterioration can be even worse as reported in another independent study that the prolonged exposure to moisture condition can result in significant degradation of the concrete-epoxy bond strength and the degradation can be as much as 70% for specimens moisture-conditioned for eight weeks (Tuakta and Büyüköztürk, 2011). Besides the studies on static moisture ingress, the cyclic moisture effect has also been investigated recently (Tuakta and Büyüköztürk, 2011). Moisture reversal and cyclic moisture conditioning tests showed that the adhesive bond cannot regain its original bond strength after successive wet/dry cycles. Also, the residual bond strength decreases when the number of wet/dry cycles and the intermediate conditioning duration increase.

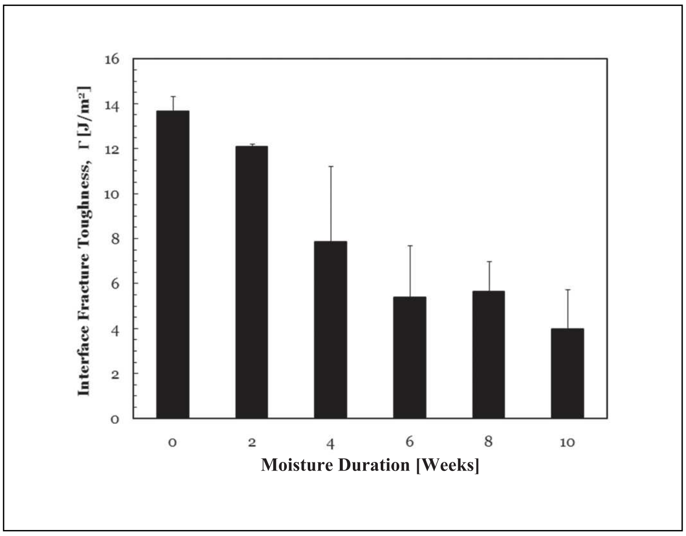

Figure 4 – Interface Fracture Toughness Decreases with the Duration of Moisture Conditioning (Lau, 2010)

Explanation of the Weakened Concrete-epoxy Interface

In general, interfacial crack can propagate either in bulk materials (material decohesion) or along the interfaces (interface separation). Reported empirical and analytical formulae available to predict interfacial fracture behaviour include: (1) crack kinking by maximum hoop stress (Yuuki and Xu, 1992) which means that the crack will propagate in a direction with the maximum hoop stress; (2) crack kinking by energy release rate (Hutchinson and Suo, 1991; Nishioka *et al.*, 2003) which is usually referred as kink criterion. The kink criterion states that when:

$$\frac{\Gamma_i}{\Gamma_s} > \frac{G_i}{G^t_{max}} \tag{1}$$

is satisfied, the parent crack that lies at the interface of the two adjoining materials will tend to kink into the substrate in consideration. Here, Γ_i is the interface fracture toughness. Γ_s is the substrate fracture toughness in mode I. G_i is the interface fracture energy release rate (of the parent crack), and G^t_{max} is the maximum fracture energy release rate for the kink crack at any putative kink angle. One should note that the ratio is less than unity because G_i is always less than G^t_{max}, due to the definition of G_i.

From the previous study, it has been discovered that these criteria cannot fully explain the interfacial fracture behaviour involving complex material interaction and chemical reaction during moisture attack (Au and Büyüköztürk, 2006). The crack kinking criterion could well predict crack propagation observed in the dry condition. However, these criteria cannot provide the physical understanding on the decrease in strength and shift in failure mode observed in wet specimens, in which interaction at the material level between water, epoxy and concrete may have occurred in the bulk material and at the interface. The two possible explanations may be offered for this phenomenon. Firstly, plasticisation by water toughens a layer of epoxy-penetrated concrete in the vicinity of the interface as shown in Fig 5(a). As a result, the initial crack at the interface between concrete and epoxy cannot propagate into concrete due to this toughened top concrete layer, and stays at the interface instead. Secondly, the adhesive bond at the interface is weakened due to the reaction between epoxy and water as described in Fig 5(b). The initial crack, therefore, stays at the interface because it requires less energy for crack propagation. There is a need to study the interfacial debonding mechanism under the presence of water molecules using a more fundamental approach. Atomistic modelling approach employing molecular dynamics (MD) simulation has been used as a basis for a fundamental study (Buehler, 2008).

One of the challenges in MD simulation is to construct an atomistic model which can accurately describe the system. A known chemical formula for the concerned material is necessary in order to create a realistic atomistic model. Concrete is a heterogeneous material which is primarily composed of hydrated cement and aggregate. However, among all the constituent materials in concrete, silica, which has a well defined chemical formula, constitutes a large proportion as aggregate with more than 40% by weight of the solid ingredients. By investigating the epoxy-silica interface in an atomistic scale, it is expected that the structural behaviour of concrete-epoxy interface can be interpreted. Such approach has been implemented recently and it has shown the

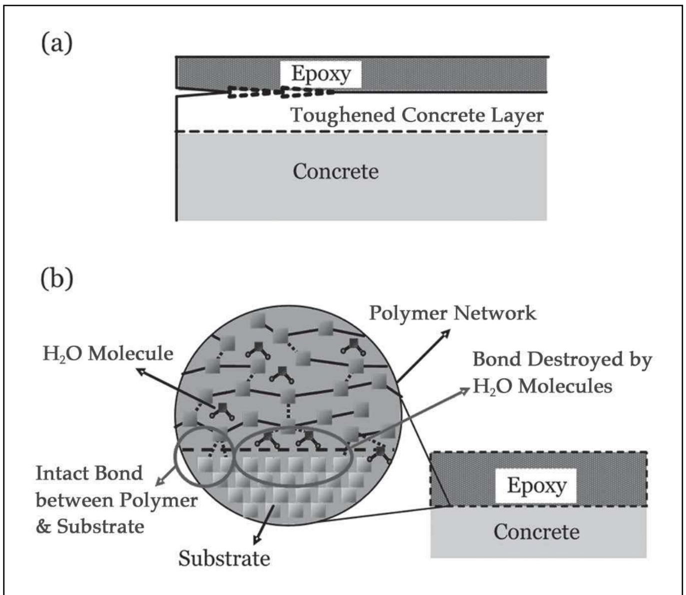

Figure 5 – (a) Plasticisation of Epoxy-penetrated Concrete Layer, and (b) Weakening of Bond between Epoxy and Concrete

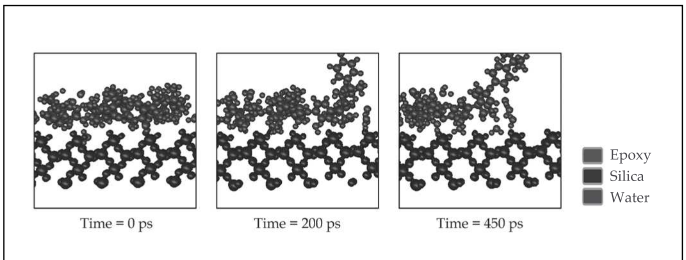

Figure 6 – Three colours are used in this figure to differentiate three different materials. Three snapshots were captured at different time for a better understanding on the interaction among epoxy, silica and water. Based on the simulation result, it is observed that water molecules really seep into the gap between epoxy and silica and eventually reduce the adhesion between epoxy and silica (Büyüköztürk et al., 2011).

ability to quantify the decrease in adhesion energy in the interface by examining the changes in the physical forces of attraction and repulsion between molecules in the two materials (Büyüköztürk *et al.*, 2011; Lau *et al.*, 2012). It was reported that the adhesion energy in a wet scenario decreased by approximately 15% compared to a dry scenario. This reduction at the molecular scale may translate into greater adhesion energy reductions in large-scale structures due to the interaction of local effects that significantly reduces their integrity. A close examination of the MD simulation process under wet condition shows that some water molecules can seep into the gap between silica and epoxy as shown in Fig 6. The presence of water molecules hinders the attraction between epoxy and silica. As a result, the adhesion between epoxy and silica is weakened.

Recommendations

Based on the results from both experiments and MD simulation, the effect of moisture on structural epoxy is detrimental with a decrease in interface fracture toughness of more than 50%, which means that the strength to resist crack initiation becomes half of its design capacity. Such decrease is significant to the safety of repaired structure and must not be overlooked. Some recommendations to remedy the moisture-induced deterioration are given as follows:

1) When structural epoxy is used for repairing the concrete cover, waterproofing spray should be applied on the repaired region once epoxy is cured in order to minimise the rate of moisture ingress to the interfacial region. In this application, remedy using waterproofing spray is suggested since the repaired concrete cover is, in general, not designed as a load carrying structural element.

2) When concrete-epoxy interface is found in the strengthening and retrofitting scenarios in which there is another structural material, such as FRP or steel, taking up the designed load, the application of waterproofing spray is expected to be not sufficient and a careful design through an appropriate computational model is highly recommended. It is known that the material properties of constituent materials, as well as the interfacial properties vary with the moisture content. It is suggested that finite element (FE) programme should be used to simulate the moisture diffusion process in the critical interfacial region. Diffusion coefficient and solubility should be quantified through moisture sorption test which are used in a FE programme to simulate moisture diffusion in the structural system. The diffusion coefficient is defined as the rate at which a diffusion process takes place, while the solubility is defined as the saturated moisture content (% by mass) (Crank, 1975). Meanwhile, many commercial FE programmes allow the fracture analysis at the interface through the implementation of cohesive zone model (CZM) (Dugdale, 1960; Barenblatt, 1962) or extended finite element method (XFEM) (Zi and Belytschko, 2003). Hence, a combined moisture diffusion and fracture analysis can be conducted in order to predict a more reliable design load for the structural element. For a more detailed procedure for conducting FE simulation on moisture diffusion and fracture analysis, it is highly recommended to refer to the documentation of the commercial FE programme (ABAQUS, 2009).

Conclusion

A review on the moisture-induced debonding in concrete-epoxy systems has been given, with the emphases on different approaches to investigate the debonding problem, as well as the important experimental finding on the severe moisture degradation of the concrete-epoxy interface. Concrete-epoxy system can readily be found in various structural applications including strengthening and repairing existing structures. In Hong Kong, aging problem of existing RC structures becomes an important social issue and a safe and reliable maintenance technique is required to ensure the public safety. The detrimental reduction in mode I fracture toughness at the concrete-epoxy interface accompanied with the dry-to-wet shift of failure is a serious warning to practising engineers that such deterioration must not be overlooked and should be considered carefully in the design stage. Recommendations for remedy have been given and it is hoped that, through this review paper, the practising engineers are more aware of the structural integrity of concrete-epoxy interface, especially in a humid region such as Hong Kong.

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