

# DESIGN OF POLYMERIC CAPSULES FOR AUTONOMOUS HEALING OF CRACKS IN CEMENTITIOUS MATERIALS

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## ABSTRACT

Now, most of the capsules used to contain polymeric healing agents in self-healing concrete, are made of glass. However, glass capsules cannot be mixed in concrete and are therefore placed manually into the moulds during concrete casting in laboratory tests. This represents a major drawback for an eventual industrialisation.

In this study, polymeric capsules were designed to meet three requirements: breakage upon crack appearance, compatibility with the polymeric healing agent and survival during concrete mixing. Three different polymers with a low glass transition temperature ( $T_g$ ) were selected (PLA – PS – P(MMA-n-BMA)). These polymers are brittle at 20°C, and consequently have the possibility to break upon crack appearance, but are rubbery above their glass transition temperature and, consequently, can survive mixing upon heating.

Differential Scanning Calorimetry and Dynamic Mechanical Analysis were performed to define the glass transition temperature of the selected polymers and to quantify the evolution of their mechanical properties with increasing temperature.

Concrete mixing tests were performed both at 20°C and at a temperature above the  $T_g$  of the capsules. Mixing at increased temperature was done by previously heating the capsules and the concrete components. The survival rates increased drastically when the capsules and the concrete components were heated. Even capsules with a thin wall (thickness 0.4 mm) resisted a 2 minute concrete mixing process, whereas none of them survived at 20°C.

In addition, the compatibility of the capsules with a two-component polyurethane healing agent was studied. The pre-polymer hardened after some days.

This research revealed that suitable design of polymeric capsules can help to meet the requirements for self-healing concrete even though further research is needed before a possible use in industry.

## 1. INTRODUCTION

Encapsulation of polymer-based healing agents is a very effective method to obtain autonomous healing of concrete cracks. However, up to now, capsules used to store the healing agent cannot resist the concrete mixing process, which is a major drawback on the way of a possible industrialization. Therefore, in this project, we attempt to create polymeric capsules which are able to survive the concrete mixing process without any particular protection, and to break upon crack appearance without human intervention.

## 2. MATERIALS

Polymeric hollow tubes with a length of 5 cm, inner diameters between 1 mm and 4 mm and outer diameter between 2 mm and 7 mm, were extruded from polymers which are brittle at room temperature and have a relatively low glass transition temperature ( $T_g$ ): Poly(lactic acid) (PLA), Polystyrene (PS) and Poly(methyl methacrylate/n-butyl methacrylate) (P(MMA-n-BMA)). In order to study the possibility of capsule breakage with crack appearance, mortar specimens containing capsules were prepared. First, a layer of 10 mm mortar with a W/C ratio of 0.4 was poured into 40 mm x 40 mm x 160 mm moulds. Two steel reinforcing bars with a diameter of 2 mm were placed on top of the mortar layer to avoid premature failure during crack formation. Two to four capsules were positioned in the middle of the specimen where the crack was presumed to appear. Some capsules which survived mixing were embedded, as well as 'new' capsules which were not mixed previously. Then the moulds were filled completely with mortar. The specimens were demoulded one day after casting.

## 3. METHODS

In order to prove the efficiency of adjusting the brittleness of polymeric capsules with temperature, mixing tests have been performed in a 20L planetary mixer to compare the resistance of non-heated and heated capsules. Hollow tubes with a length of 5 cm were used as capsules. After sealing one end, capsules were filled with water mixed with red dye, and finally sealed at the other end. For the concrete mixing test with heated capsules, capsules were heated in an oven during 20 to 40 minutes before the mixing test. The mixing processes are summarized in Figure 1.

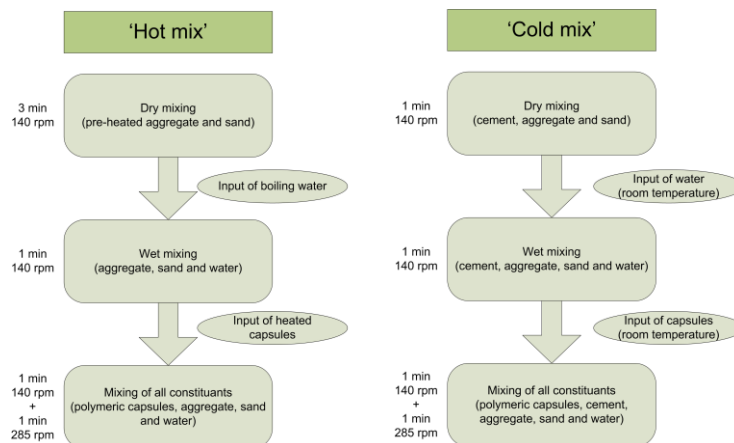


Figure 1: Mixing processes to study the resistance of the polymeric capsules

When the mortar samples, described in section 2, were 28 days old, three-point-bending tests were performed following 2 successive methods. The first method was used to see if the capsules break when a crack is created and its width increases at a speed of 0.001 mm/s until 0.4 mm (measured by linear variable differential transformer (LVDT)). Capsule breakage was expected and some pictures were taken when leaching dye of the broken capsules was detected. In the second method the mortar sample was completely broken to see whether the capsules break at higher crack width or whether they slip in the matrix.

#### 4. RESULTS

Almost all polymeric capsules survived the concrete mixing process while they were heated above their  $T_g$ . The differences between 'hot' and 'cold' mixes are detailed in Figure 2. The improvement in survival ratio after heated is assumed to be caused by the more flexible behavior of the capsules above their  $T_g$ . Even the very small PLA capsules with a wall thickness around 0.4 - 0.5 mm survived the mixing process whereas none survived the cold mixing process.

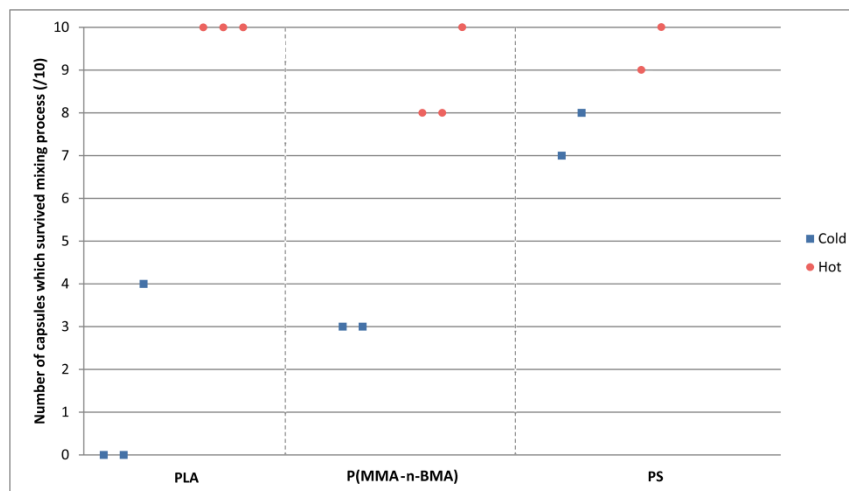


Figure 2: Survival ratios of the polymeric capsules during mixing

Some P(MMA/n-BMA) capsules broke when cracks of 0.4 mm were created. For two specimens with incorporated deformed capsules, which previously survived the hot mixing process, some capsules broke and released the water they initially contained as illustrated in Figure 3a. For 2 other specimens with capsules which previously resisted the concrete mixing process but which were not deformed, capsule breakage was observed. The crack width at which the capsules broke was deduced from the graphs representing the load in function of the crack width measured by the LVDT. When a sudden drop in the curve was observed (as illustrated in Figure 3b) and the sound of capsule breakage was heard, the capsules were supposed to be broken. Three capsules broke when the crack width was around 0.15 mm, which corresponds to a relatively small crack. Therefore, if the healing agent could be encapsulated by these capsules, healing of cracks with a width of around 0.15 mm could be possible.

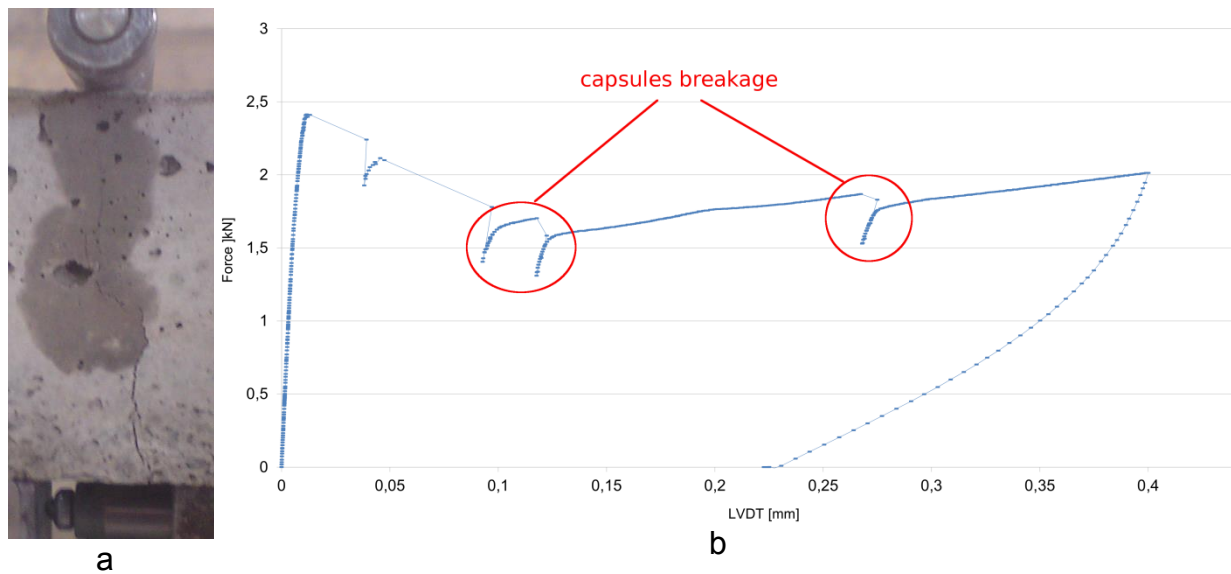


Figure 3: Breakage of P(MMA-n-BMA) capsules with visible water release (a) and sudden drop in the load curve (b).

However, none of the PS and PLA capsules broke when cracks with a width of 0.4 mm were created. No drops were observed in the load curves and no water marks were observed. During the second test (complete breakage of sample), the PS capsules broke without visible. PLA capsules also broke during the second test, but most of them broke just before specimen failure at crack widths larger than 1 mm. A plastic deformation of the capsules was observed. Moreover, some small capsules were pulled out when the two parts of the specimens were disconnected. This indicates that there is no sufficient bond between the PLA capsules and the cementitious matrix.

## 5. CONCLUSIONS

The use of polymeric capsules to obtain self-healing properties in concrete seems promising but still needs further investigation. In this study, brittle thermoplastics which can resist the concrete mixing process through heating, and break with crack appearance at room temperature have been extruded. Even if the three polymers studied are known for their brittleness, only one type of capsules broke with the creation of crack with a width smaller than 0.4 mm. Therefore the brittleness of the material is a very sensitive parameter to design capsules for self-healing concrete.