# Comparative Study on Different Polymer Tubes as Carriers of Healing Agent for Self-Healing Concrete

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Keywords: capsules, concrete, crack, polymer, self-healing

Abstract ID No : 248

## Abstract

Nowadays, concrete is the most widely used construction material in the world. Despite many technical and economic advantages, crack initiation and propagation in concrete elements under different types of loading which consequently require periodical maintenances, are problematic in reality. Autonomous healing is a recently upcoming strategy for overcoming this drawback of concrete structures. However, the bottleneck for valorisation of self-healing concrete is developing suitable capsules. This paper presents initial results of a research work which investigates suitability of polymeric tubes as carriers of healing agents for self-healing concrete. Several glassy polymers were evaluated in terms of their compatibility with different healing agents, survival after the concrete mixing process and breakage upon crack formation. It was observed that polymethyl methacrylate (PMMA) had the highest compatibility with different healing agents. Although different polymeric tubes showed acceptable performance with regard to their survival rate after concrete mixing, further modifications are required to ensure that they can be broken upon crack formation.

## 1. Introduction

Concrete which simply consists of Portland cement, water and aggregates is the main construction material in the world. Although it has high compressive strength, it is brittle under tensile loads. This results in crack initiation and propagation in concrete elements which consequently require periodical maintenances. It is estimated that approximately 50% of the annual construction budget is used as the maintenance costs for the existing structures such as concrete bridges, tunnels and retaining walls in the European Union [1, 2]. Self-healing concrete is an innovative approach for overcoming these barriers. The healing process can be performed by autonomous healing system which is based on using capsules containing healing agents. These capsules can be open to release the healing agent upon crack formation. However, the bottleneck for valorisation of self-healing concrete is developing suitable capsules which (1) can survive during concrete mixing; (2) are compatible with the healing agent and resist the high pH environment of concrete; (3) break upon crack formation while they show enough bonding with surrounding matrix and (4) do not negatively affect the properties of fresh and hardened concrete. Up to now, most research has been done by using glass tubes for proof-of-concept [3] which is not appropriate for practical applications. This paper evaluates the feasibility of using different types of polymeric tubes as carriers of healing agent for self-healing concrete.

## 2. Materials and experimental methods

Three glassy polymers with low glass transition temperature ($T_g$) including polylactic acid (PLA) with and without impact modifiers, polystyrene (PS) and polymethyl methacrylate (PMMA) were identified for use as carriers of healing agents. Moreover, the most common healing agents including one component polyurethane (PU), pre-polymer of PU and an accelerator (two-component PU), methyl methacrylate (MMA) based agent and dicyclopentadiene (DCPD) were selected for use in the compatibility test between healing agents and three identified polymers. To investigate the compatibility between healing agents and the identified encapsulation materials, gel permeation chromatography (GPC) for measuring molecular weight and Differential Scanning Calorimetry (DSC) for determining $T_g$ were used, respectively. Furthermore, different polymeric tubes were extruded with an average wall thickness of 0.5 mm and the outer and inner diameter of 5 and 4 mm, respectively. These hollow tubes were cut to capsules with a length of 50 mm and their ends were sealed with MMA glue, one before and another one after filling the tubes with green dye for visualizing capsule breakage during the survival test of tubes after concrete mixing, as well as breakage upon crack formation in mortar samples. Moreover, DCPD was added to the dye inside the capsules which were used for the breakage test due to its specific strong smell which could be distinguished upon breakage of the tubes in mortar samples. Normal-strength concrete (ratio of fine and coarse aggregate to cement: 1.9 and 3.5, respectively) with water to cement ratio of 0.47 containing crushed aggregates with the maximum size of 20 mm was used for the survival test. In this test, ten prepared tubes were added to ten litre of the concrete mixture which was already made and mixed with the tubes for two minutes. Then, the number of survived and unbroken tubes were calculated after retrieval from the fresh concrete mixture. In addition, standard mortar in accordance with the standard NBN EN 196-1 (water to cement ratio of 0.5 and aggregate to cement ratio of 3) was used for making prismatic samples for the breakage upon crack formation. Each sample contained one polymeric tube filled with DCPD and green dye. Moreover, two steel reinforcing bars were used for each mortar sample to avoid premature failure during crack formation. A linear variable differential transformer (LVDT, Solarton AX/0.5/S) was used for measuring the crack width during the three-point-bending test.

## 3. Results and discussion

Table 1 summarizes compatibility between different healing agents and different polymers based on the results which were obtained by GPC and DSC analysis after immersing polymers in different healing agents. It can be seen that MMA-based healing agent had the worst performance in terms of compatibility with the encapsulation materials. In terms of the identified polymers for making capsules, PMMA showed the most acceptable performance except for combination with the

MMA glue, in which it was partially dissolved after two weeks and then, completely dissolved within one month. Moreover, PS showed the worst compatibility with the healing agents as it was compatible only with two-component PU. However, it was partially dissolved after two weeks of immersion in DCPD and one-component PU and then, completely dissolved within 4 months. Therefore, it was decided that PS would not be used during the tube extrusion process and the capsules were made of PLA, PLA+10% impact modifier and PMMA. The impact modifier (BioMaxStrong) was added to the PLA for increasing its impact strength and getting higher probability of survival during the concrete mixing process.

**Table 1: Compatibility results between different polymers and different healing agents**

| | PLA | PMMA | PS |
|---|---|---|---|
| Two-component PU | - | + | + |
| One-component PU | + | + | - |
| DCPD | + | + | - |
| MMA-based glue | - | - | - |

As it was mentioned earlier, ten capsules were added to 10 litre of the prepared concrete mixture and mixed for further two minutes for measuring survival probability of the capsules during the mixing process. Overall, 90 capsules of these three polymers were evaluated in the survival test. Table 2 presents the results of the survival test which was done 3 times for each polymer type. It can be seen that the capsules which were made of PLA+10% impact modifier had the best performance in terms of the survival after mixing since none of them were broken during the mixing process. Moreover, the PLA and PMMA capsules showed similar performance so that eight tubes could survive after mixing which means an average survival probability of 80%. In general, these three polymers showed acceptable performance during the concrete mixing process and can be considered for practical applications and further investigation.

**Table 2: Results of the survival of the polymeric capsules during concrete mixing**

| Polymer | PMMA | | | PLA | | | PLA+10% impact modifier | | |
|---|---|---|---|---|---|---|---|---|---|
| Repetition | 1st | 2nd | 3rd | 1st | 2nd | 3rd | 1st | 2nd | 3rd |
| Survived capsules | 7 | 10 | 7 | 9 | 9 | 6 | 10 | 10 | 10 |
| Average % | 80% | | | 80% | | | 100% | | |

In terms of the breakage of capsules upon crack formation, a three-point-bending test was done to obtain a crack width of 0.3 mm and continued to 0.9 mm. Figure 1 shows different samples containing one capsule from PLA+10% impact modifier (left), PLA (middle) and PMMA (right). Special care was taken to consider any sign of dye, smell of DCPD and noise of possible breakage of the capsules during loading. As it can be seen, none of the capsules could be broken after loading for obtaining a crack width up to 0.9 mm. This is consistent with results that were reported by Hilloulin et al. [4].

**Figure 1: Mortar samples containing one capsule from PLA+10% impact modifier (left), PLA (middle) and PMMA (right) after three-point-bending test (crack width:0.9 mm)**

## 4. Conclusions and further research

Based on the results of this study, it can be concluded that PS is not a suitable encapsulation material due to its incompatibility with the main self-healing agents which were investigated in this work. Moreover, the PMMA tube showed promising results in terms of the compatibility with different healing agents. In addition, using impact modifier could increase the survival probability of the polymeric capsules during the concrete mixing process. In general, three investigated polymers had acceptable performance in terms of the survival ratio. However, it seems further modifications should be applied so that the extruded capsules can be broken upon crack formation during the loading. Different strategies such as using plasticizers and adding nano-particles for obtaining lower elongation at rupture are under investigation by the authors.

## References


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