



## Investigation of the Effect of Microcapsule Additive on Mechanical and Physical Properties of Concrete

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### HIGHLIGHTS

- Concrete mix has been reinforced with SiO<sub>2</sub> microcapsules to seal concrete cracks.
- Great Self-healing capacity of concrete that contain SiO<sub>2</sub> microcapsules.
- Crack of concrete with SiO<sub>2</sub> microcapsules was self-healed in 28 days.

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### ABSTRACT

Cracks are a fundamental problem in concrete, so this research investigates an experimental study of the effect of adding SiO<sub>2</sub> on the self-healing behaviour of concrete. The study is done by studying the physical properties, microstructure, and mechanical properties of concrete. Microcapsules were prepared to contain (cement and nano SiO<sub>2</sub>) in a (palettization (fluidize bed coating) method) with (5,10, and 15) % of cement weight. The ratio of the prepared concrete mixture was (1: 2: 4) and the ratio of water to cement (W / C) 0.45 by the weight of cement was also added as 0.6% Superplasticizer to all concrete mixtures. SiO<sub>2</sub> MC is added with (2.5) % of the cement weight in the concrete mixture. In this study, porosity, density, and water absorption tests were performed to study the physical properties, compressive and bending strength to study mechanical properties. So, referring to the results, (SiO<sub>2</sub> MC) was used as an effective effect in the self-healing cracking mechanism, and in improving the bending strength and compressive strength. Also, 10 % wt. was recorded as the best addition because it has a positive effect on properties to be applied in construction.

## 1. Introduction

Over time, concrete structures are more susceptible to cracking, as we notice that small cracks have no effect on these structures, but the problem lies in the large cracks that cause the breakdown of concrete structures. These cracks will allow gases, chemicals, rain, etc. to enter, which will reduce the life of the structures and reduce the physical and mechanical properties. Structure's maintenance and rebuilding are more difficult and expensive, including labor and energy costs. In recent years, a new approach has been proposed to improve the physical and chemical properties of concrete and heal cracks. These materials can treat and improve the properties by producing precipitated crystals such as calcium carbonate [1]. There are several methods of self-healing materials, including a self-healing agent (fibers, nanocrystals, and mineral additives (i.e., stretching agents) and the other type is a self-healing agent (micro-capsules, tubes, vascular capsule, cement-coated granules, and bacteria)[2]. There are many techniques for creating capsules, from mixing to the end in complex and delicate ways. There are many challenges to this method, ranging from the ability of micro-capsules to stick without breaking during mixing, to their longevity within the base material [3]. In this work cement and silicon oxide were used as the nucleus of the microcapsule. Previous studies showed the positive effect when using silicon oxide nanoparticles as additives, according to Ali Haidari et al. [4], where adding a small amount of (Nano SiO<sub>2</sub>) to the concrete improved its compressive strength and decreased its ability to absorb water and the physical properties of concrete. In another study, Aggarwal et al. [5], it was noted that the samples containing (Nano SiO<sub>2</sub>) increased the compressive strength to them with the increase of (Nano SiO<sub>2</sub>), and they noted the increase in tensile strength and durability compared to the standard sample. While the polymer material was used as a coating for the microcapsules, many researchers used polymer materials, for example: epoxy [6], PMMA [7], polyurethane [8], and polystyrene. Biqin Dong et al. [9] studied how to use polystyrene as a coating and how to release healing agents. Benoit Hilloulin et al. [10] studied the ability of microcapsules made of polystyrene shell to resist concrete mixes. The use of micro-capsules is of great importance in improving many properties in addition to the ability to self-healing. Many studies have used the micro-capsule technique and have noted a positive effect in treating cracks and improving the properties. Zhengxian Yang et al. [11] used micro-capsules in the concrete mixture as the results showed the ability of micro-capsules to self-heal as well as

an increase in resistance to bending and compression. Likewise, Wei Du et al. [12] used micro-capsules to treat concrete cracks and add an improvement in their properties. The results were evident from the ability of the micro-capsules to heal in a few periods of time. The purpose of this study is preparing a concrete mixture suitable for concrete applications that can self-repair by adding different proportions of SiO<sub>2</sub> micro capsules, in order to observe the effect of these capsules on the efficiency of self-healing, by studying the physical and mechanical properties of them and also to observe the slit-closing mechanism in the micro-capsule-containing samples compared to the standard sample.

## 2. Experimental Part

This work included two methods: The first is the combination of micro-capsules (SiO<sub>2</sub> MC) with concrete in different proportions (2.5%) of cement weight, and their effect on concrete performance is studied. Second, cracks are made in the concrete and the effect of (SiO<sub>2</sub> MC) on the mechanism and healing period is studied. In this study, ordinary Portland cement of the first type is used, which is imported from the Iraqi State Cement Company. We see that the chemical analysis in Table 1 is consistent with ASTM 105. The average volume of dry used cement powder passing through a 38 μm sieve.

Capsules are manufactured by a pelletizing process (fluidized bed coating). A sieving process is performed for the fine capsules to determine their size. MC contains different proportions of (Nano SiO<sub>2</sub> from a company called (ULTRANANOTECH.PVT. LTD) and cement) and polystyrene polymers (SIR) as shown in Table 2. Properties of (SiO<sub>2</sub>MC) are shown in Table 3. Microcapsules are shown in Figure 1. Cement Concrete was mixed at room temperature. First, the dry concrete mixture (cement, fine aggregate certified in EN 196-1, and coarse aggregate (Al-nibae 19/5)) was mixed, then we added microcapsules (2.5% by weight of cement) to the concrete mix. All concrete samples have water to cement ratio (w / c) of 0.45 and a ratio of cement, fine aggregate, and coarse aggregate materials 1: 2: 4, according to (BS 1881-111: 1983), [13], with the addition of 0.6% by weight of cement from Glinume 51 superplasticizers (BASF). Before applying the concrete, to ensure easy sample extraction, the inner mold is lubricated. 100 \* 100 \* 100mm molds were used. Plastic foil is used to cover molded samples for the first 24 hours to prevent moisture loss. After drying, they are immersed in water for various treatment periods until the time of testing. The proportions used in the formation of cement concrete are shown in Table 4.

**Table 1:** Cement Compound

Oxide composition	Abbreviation	% by Weight
Lime	CaO	63.37
Silica	SiO <sub>2</sub>	18.75
Alumina	Al <sub>2</sub> O <sub>3</sub>	5.29
Iron oxide	Fe <sub>2</sub> O <sub>3</sub>	4.28
Sulphate	SO <sub>3</sub>	2.25
Magnesia	MgO	2.36
Loss of Ignition	L.O.I.	1.55
Lime saturation factor	L.S.F.	0.90
Insoluble residue	I.R.	1.15

**Table 2:** The proportions of the micro-capsule contents

Capsule					
Mix	(wt.%)	Nano powder use	Nano powder (g)	Cement (g)	Polymer(g)
MS1	5	SiO <sub>2</sub>	3	57	2
MS2	10	SiO <sub>2</sub>	6	54	3
MS3	15	SiO <sub>2</sub>	9	51	4

**Table 3:** Typical Properties of SiO<sub>2</sub>MC

SiO <sub>2</sub> MC	Core/Shell material	Morphology	Size	Color	Thickness of the shell
Specification	Nano particle SiO <sub>2</sub> (core)PS (shell)	Aspheric	212(μm)	Gray	2 (μm)

**Table 4:** The proportions of concrete mixture in (Kg/m<sup>3</sup>) with 2.5% SiO<sub>2</sub> MC by weight of the cement

Mix	Type of MC	Ages(day)	Cement	Coarse aggregate	Fine aggregate	MC
M0	-	3,7,28,90	280	560	1120	-
M1	MS1	3,7,28,90	273	560	1120	7
M2	MS2	3,7,28,90	273	560	1120	7
M3	MS3	3,7,28,90	273	560	1120	7



Figure 1: SiO<sub>2</sub> Microcapsules

### 3. Testing

#### 3.1 Compressive Strength

The compressive strength of the cubes was obtained by using a compacting device for concrete samples made with dimensions (100 \* 100 \* 100) mm. In accordance with BS 1881-116: 1983 [14], assay was performed for each concrete sample for a period of 3,7,28, and 90 days.

#### 3.2 Flexural Strength Test

The bending test is performed using prismatic mold prepared with specified dimensions of (75 \* 75 \* 300) mm, in accordance with BS 12390-5: 2009. The crack is made by means of a three-point flexural strength test. Slots of 3 mm deep below each prism in the center of the base were made using a circular saw to act as strength focus points. The applied speed was 0.04 mm / min. The results were recorded for ready-made concrete samples at the age of 28 days [15]. To determine the flexural strength, we used the following equation:

$$F_{ct} = F * I / (d_1 * (d_2)^2) \quad (1)$$

Where:  $F_{ct}$ : is the flexural strength, in MPa (N/mm<sup>2</sup>);

$F$ : is the maximum load, in N.

$I$ : is the distance between the supporting rollers, in mm;

$d_1$  and  $d_2$ : are the lateral dimensions of the specimen, in mm.

After 28 days, the restoration of the bending strength is evaluated again (2 R) after healing, and an equalizer is applied:

$$\eta\% = f_{ct1} / f_{ct2} \quad (2)$$

Where:  $\eta\%$ : is the efficiency of healing.

$f_{ct1}$ : is maximum stress for the original sample (1st R);

$f_{ct2}$ : is maximum stress for the sample after healing (2nd R)

Samples are removed from the water weekly to measure the width of the slit and imaging is taken over time to estimate the rate of strength recovery.

#### 3.3 Dry Density

Density can be calculated according to ASTM (C642-2013) specifications, where it can be calculated after drying the sample and weighing it suspended in water and weighing it after being immersed in water for 24 hours.

Dry density can be calculated from the equation [16]:

$$\text{Dry density (g/cm}^3\text{)} = [w_1 / (w_2 - w_3)] * \rho_w \quad (3)$$

Where,  $W_1$ : Mass of oven-dried sample in air, g.

$W_2$ : Mass of surface-dry sample in air after immersion and boiling, g.

$W_3$ : Apparent mass of the sample in water after immersion and boiling, g.

$\rho_w$ : Density of water, which is equal to (1 g/cm<sup>3</sup>).

#### 3.4 Porosity

This test was performed according to the procedure specified in ASTM Specifications (C642-1997). The method test was similar to that of dry density test; and the porosity can be calculated by using the following equation:

$$\text{Porosity} = [(w_2 - w_1) / (w_2 - w_3)] * 100 \% \quad (4)$$

#### 3.5 Water Absorption

Water absorption can be calculated in a similar way to calculating density (Archimedes base), except that a sample is not weighed while suspended in water. This scan is performed according to ASTM Specification (C642-1997).

water absorption in mortar samples can be calculated using the equation:

$$\text{Absorption of Water} = [(w_2 - w_1)/w_1] * 100 \% \tag{5}$$

The tests of dry density, total water absorption, and porosity were carried out in the metallurgy lab of Materials Eng. Dep./University of Technology.

## 4. Results and Discussion

### 4.1 Compressive Strength

In Figure 2 the results for the compressive strength test at ages of 3, 7, 28, and 90 days are shown for all cubic samples. At the age of 3 days, the results of the MS and 0 M samples showed a slight increase in strength, and all samples (reference and MS) also showed an excess of the required strength at 28 days. The improvement in strength of all samples can be attributed to the effect of the superplasticizer which, as a parameter, represents the lowest ratio of water to cement used in preparing the concrete mixture. It contains more C2S (Dicalcium Silicate) and less C3S (Tricalcium Silicate), which is highly correlated with potency at distant ages. Moreover, the SiO<sub>2</sub> MC 10% sample showed the highest strength at the age of 3, 7, 28, and 90 days, the decrease in strength occurs at all ages when the amount of SiO<sub>2</sub> MC exceeds 10% (percentage of added Nano powder to capsule) which agrees with previous research [17-19].

### 4.2 Dry Density

Figure 3 shows the density test results for the tested samples containing capsules (2.5%) by weight of cement and standard sample, where we notice an increase in density of up to 1%, because of the presence of SiO<sub>2</sub> MCs in the concrete mixture that works on improving the rate of cement hydration and making denser microstructure when comparing to control samples at the same ages and then it decreases above this percentage, but the density of M2 is still higher by (2.34, 2.4, 2.50083, and 2.58)g/cm<sup>3</sup> at the same ages, respectively compared to control samples [20, 21].

### 4.3 Water Absorption

Through the results of the absorption test, Figure 4 shows that the absorption results for control sample exceeded those for MS samples, from previous results, where the addition of these MCS to concrete leads to a large decrease in the water absorption and this might be due to the result of more C-S-H gel formation and reducing the pore size to the partial or total filling of pores through the physical and chemical effects of these MCS, where the addition reduced water absorption in 3 days to (0.1, 0.07, 0.065, and 0.08)%, respectively for mix (M0, M1, M2, and M3) [18, 20, 22].

### 4.4 Porosity

The results of the porosity tests for all samples more than the control samples at all ages are shown in Figure 5. The microcapsules will act as a filler material to enhance the density of the concrete, which greatly reduces the porosity of the concrete and the CSH gel cannot grow and it limits the growth of a dense Ca(OH)<sub>2</sub> crystal causing the porosity to be reduced. The porosity could be considered as an adverse property to density, so the porosity value reduction was due to the density increase. Porosity improvement ratios in 28 day was (48.096, 58.47, and 44.636) %, respectively with (M1, M2, and M3) [18, 20].

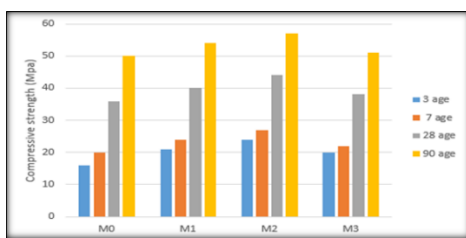


Figure 2: Compressive strength results for all samples at ages 3, 7, 28, and 90 days

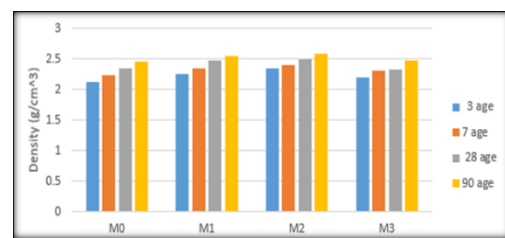


Figure 3: Dry density of all samples at all ages

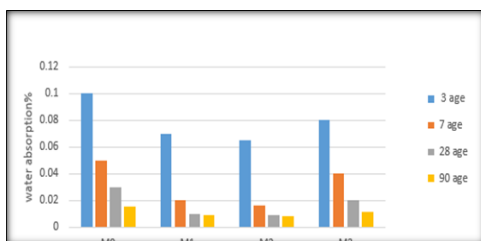


Figure 4: Water absorption percentage for all

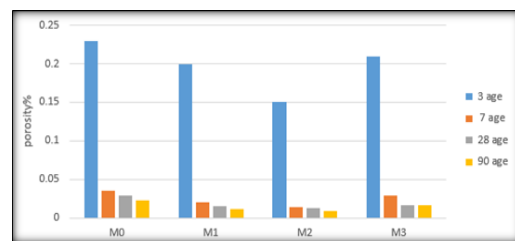


Figure 5: Porosity percentage of all samples

samples

### 4.5 Flexural Strength

From the compressive strength results, a ratio of 1% was adopted to predict the results of the flexural test. Prism of 1% is compared with the reference sample. The highest result was 17.1 Mpa at the age of 28 days, and the result of the reference sample did not exceed 15 Mpa at the same age. This corresponds to [23]. The same prism samples are used and applied to assess the bending strength recovery after healing for a period of 28 days. Whereas the recovered strength appeared to be the control sample and the sample containing the microcapsules, respectively (74.7%, and 94.2%), as shown in Figure 6 and this is evidence of the effectiveness of MC as a healing agent when compared to the control sample consistent with previous studies [24].

### 4.6 Evaluation of Seal of Crack by Microscopy

The cracks are observed with a microscope at 7 days of age and 28 days after healing with water. The healing values of the reference and sample SiO<sub>2</sub>MC after 28 days are shown in Figure 7, and the best decrease in the width of the cracks was in the sample of SiO<sub>2</sub>MC. Likewise, Figure 8 shows the shape of the cracks that were detected by the microscope optical magnification that equals 40 X.

### 4.7 SEM and XRD of Healed Samples

An examination of SEM and XRD was performed in addition to seeing self-healing during microscopy, to see the changes in the microstructure.

In Figure 9-a, the results of the control sample containing CH and the gel CSH produced from the residual anhydrous cement are shown. It acts as a curing agent that closes very few cracks.

In Figure 9-c the sample containing the added capsules, the healing agent's calcium hydroxide, calcium silicate hydrate, and silica appear at the interface of the crack as a cure in the concrete paste that produces the CS [25].

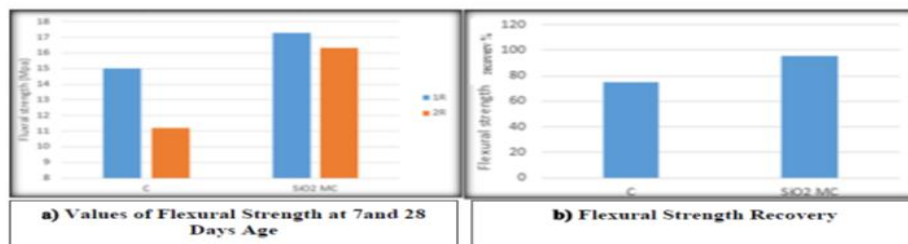


Figure 6: Flexural Strength

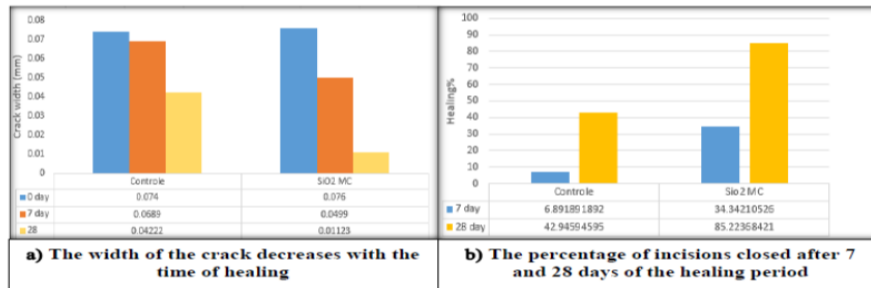


Figure 7: Healing values of the reference and MC sample



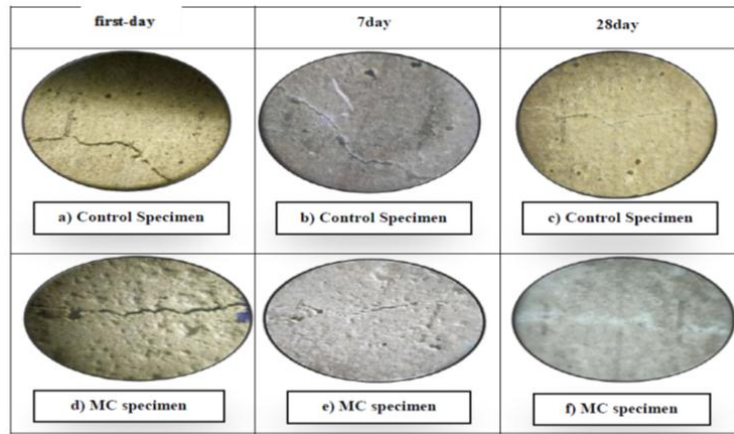


Figure 8: First-day Cracks before healing and at 7 and 28 days after healing

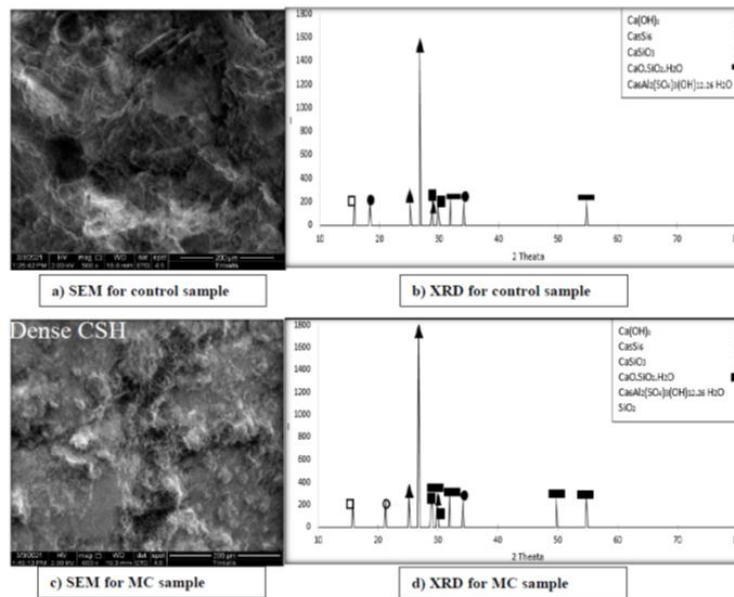


Figure 9: The XRD patterns and SEM images for the crack area of the healed samples

The XRD results also show the main components in concrete that gathered from faces of crack, as shown in Figure 9-b represented by  $(Ca(OH)_2, Ca_5Si_6, CaSiO_3, CaO.SiO_2.H_2O, \text{ and } Ca_6Al_2(SO_4)_3(OH)_{12}.26H_2O)$ , and they were identical with JCPDS No. (04-0733, 45-1480, 43-1460, 34.0002, and 41-1451), respectively, and that agrees with previous studies [26]. The peak of  $SiO_2$  is shown in Figure 9-d. CSH( $CaO.SiO_2.H_2O$ ) can be observed as healing agents that are abundant in Figure 9-d pooled on the crack face in contrast to the not appearance of peaks in the control sample. This is evidence of ( $SiO_2$ ) resulting from the broken capsules spreading on the side of the crack interacting with CH ( $Ca(OH)_2$ ) to produce a condensed layer (CSH). While virtually no peaks CH ( $Ca(OH)_2$ ) can be observed in the sample containing capsules compared to their appearance in the control sample. All other ingredients appeared without noticeable differences [27].

### 5. Conclusion

The effect of  $SiO_2$  MC addition on the concrete mixture was considered in terms of its ability to self-repair concrete cracks. The physical and mechanical properties can be deduced from the research results. Several tests were performed, including compressive strength tests where the improvement ratios for (M1, M2, and M3) at 28 days were (11.11, and 22.22, 5) % respectively, the density (5.477, 6.687, and 0.59)%, respectively, the water absorption (66.6, 70, and 33)%, respectively, the porosity of the concrete castings (48.096, 58.47, and 44.636)%, respectively, and the flexural strength of the best selected ratio, which showed improvement of (74.7%, and 94.2%). Great healing ability through microscopy, and the ability to seal concrete cracks were observed at the age of 7 and 28 days, as the percentage of improvement of crack closure was (34.3, and 85.2) %, respectively, which is better than the control sample. It achieved (6.8 and 42.9) %, respectively.  $SiO_2$  MC acts as a network that limits the content, size, microcrystals, and orientation of methane crystals.

### Author contribution

All authors contributed equally to this work.

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## Data availability statement

The data that support the findings of this study are available on request from the corresponding author.

## Conflicts of interest

The authors declare that there is no conflict of interest.

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