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# Experimental investigation of the healing properties of the microvascular channels-based self-healing glass fibers/epoxy composites containing the three-part healant

Mohammad Amin Mohammadi[a,b], Reza Eslami-Farsani[a], Hossein Ebrahimnezhad-Khaljiri[a,*]

[a] Faculty of Materials Science and Engineering, K. N. Toosi University of Technology, Tehran, Iran

[b] Centre of Research in Advanced Materials and Structures, Faculty of Mechanical Engineering, K. N. Toosi University of Technology, Tehran, Iran

Corresponding author: ebrahimnezhad-k@email.kntu.ac.ir

## Abstract

In this study, the self-healing ability of the E-glass fibers/epoxy composites based on microvascular channels under flexural and tensile loading was investigated by a three-part healant. The fabrication of the microvascular channels was conducted through embedding the solid preforms and removing them. The epoxy resin and premix anhydride hardener-$CuBr_2$ (2-Methylimidazole) as the healing agents were used to study the self-healing ability of the composites at different healing times (4, 7 and 11 days) and various volume fractions for recovering the tensile (2.5, 4 and 8 vol%) and flexural (2, 3.2 and 3.7 vol%) strength. The optimum recovered tensile strength belonged to the composite containing the 4 vol% healant. The obtained healing efficiencies of this composite after 4, 7 and 11 days were 57, 68 and 69%, respectively. In the flexural test, the composite with the 3.2 vol% healant had the maximum healing efficiencies of 3, 46 and 44%, as compared with other composites. By using the field emission scanning electron microscopy and energy dispersed spectroscopy analysis, the healing ability of the composite was confirmed by this healing system.

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**Keywords**: Smart composite, Self-healing, Microvascular channels, Flexural strength, Tensile strength, Mechanical recovery

## 1. Introduction

Polymeric composite structures are widely used in various industries because of their high strength-to-weight ratio, chemical stability in different atmospheric conditions and low density [1,2]. Despite such advantages, these materials have some disadvantages under their service conditions [3]. One of the most common issues is the presence of microcracks in these structures, detecting which could be too difficult. This is because they occur in relatively low impact energies and there is often no visible indication of damage on the materials' surfaces [4]. These microcracks can joint together and grow; so, they can cause catastrophic failures and destruct the structures. To overcome this issue, controlling the microcrack behaviors is so necessary. Using the nanomaterials like nanoalumina [5], nanozirconia [6], carbon nanotubes [7], nanoclay [8] and graphene oxide [9] in the composite structures is one of the methods for controlling microcrack behaviors by pinning [10], deflecting [11], bowing [12] and bridging mechanisms [13] which have been done by many researchers in the last decade. The idea of composite structures with the self-healing ability is another way for controlling crack behaviors which has been developed by some researchers [14–16].

The composites with the self-healing ability can eliminate the maintenance costs, as well as saving time and increasing the safety, due to the in-situ healing ability of damages [17]. This ability can be created into the composites by intrinsic and extrinsic healing systems [18,19]. Generally, for fabricating an extrinsic self-healing composite, the healing agents (healants) should be reserved inside the reservoirs of microcapsules or microvascular networks [20,21].

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Nowadays, the microvascular networks have attracted much attention, because they are one of the newest methods for reserving healants [22].

Patrick et al. [23], for example, studied the isolated and interpenetrating 3D microvascular networks via the vaporization of the sacrificial component technique. They evaluated the self-healing ability of the structures by using the double cantilever beam test. Their results showed that these structures could completely recover their initial properties. Toohey et al. [24] also made a polymer incorporated with the hollow micro-channels through the fugitive direct ink writing method to determine the healing efficiency of the materials. They assessed the effect of the catalyst on the healing efficiency and cycling, finding that increasing the catalyst led to more healing cycles. The maximum healing efficiency was found to be about 70% in this research work. Further, Norris et al. [25] embedded the glass vessels with the diameter of 500 μm on the mid-plane of carbon fibers composites and investigated the self-sensing and healing ability of the composites by applying the separated microvascular channels. These structures could recover approximately 94% of the initial properties after 24 h.

In another research, Trask and Bond [26] made the vascular networks within the carbon fibre-epoxy composite by the lost-wax process. The novelty of that work was using low-melting temperature metal wires (solder wires with the diameter of 250 μm) as the removable solid preforms between carbon plies with different angles (0, 45 and 90°). After curing, the solder wires were removed, creating orthogonal vascules in the composites.

The non-removable/removable solid preforms processes are cheaper and simpler than other methods, because other methods need expensive and advanced equipment. Generally, in the non-removable solid preforms method, the preforms have a poor interface with the matrix; so, regardless of the used preforms material for creating channels (glass, polymer or metal), they decrease the mechanical properties of the composites [27]. In the removable solid preforms

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method, there are hollow channels in the structure without any walls and interfaces between hollow channels and the composite (unlike non-removable solid preforms). Therefore, after damage creation, the healing agents can flow to the damage sites as soon as possible. On the other hand, in the non-removable preforms, because of the interface between the composite and the tube, the tubes' walls need a particular threshold of energy for destruction. Therefore, without any destruction of hollow fibers (tubes), the healant cannot be diffused to the damage area [28]. Although the removable solid preforms methods have resolved this issue, deformation (de-shaping) of channels can occur during the fabrication process of the composites.

This research work, therefore, aimed to investigate the healing time and the healant percentage of an E-glass fibers/epoxy composite containing microvascular channels to achieve the highest healing efficiency under tensile and flexural loading via anhydride healants. To fabricate one-dimensional and straight microvascular channels for storing the healing agents, the removable solid core method was used. The polyamide cores (polymeric wires) were applied, because of their rigidity for eliminating the de-shaped channels problem. By using the anhydride healants, we could ensure embedding/de-embedding the rigid polyamide cores and fabricate the hollow cores; these were the novelties of this research work.

## 2. Experimental

### 2.1. Materials

A two-part resin system consisting of epoxy bisphenol F (ML-506) and polyamine hardener (HA-11) was prepared (Mokarrar Engineering Materials Co., Tehran, Iran) for the fabrication of the composites. The ratio of resin to hardener was 100 to 15 wt%. It should be noted that the reported $T_g$ at 75% cross link density for this epoxy matrix was 158 ˚C [15]. The

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plain weave E-glass fibers (LINTEX International Co., China) with the surface density of 400 $g/m^2$ and the tensile strength of 2400 MPa were used as the reinforcement. Polymeric wires (polyamide fiber) with the diameter of 500 and 700 μm were also used as the re-tractable preforms for creating the microvascular channels into the structure. The three-part healing agents consisting of the epoxy resin (ML-526), maleic anhydride hardener (HA-59) and $CuBr_2$ (2-Methylimidazole) accelerator (Mokarrar Engineering Materials Co.) were used to heal the composite structures. The mixing ratio of the epoxy, hardener and accelerator was 100:90:2 wt%. It should be noted that the percentage of the accelerator was selected according to the previous research works [29,30].

### 2.2. Specimens fabrication process

To fabricate the microvascular channels into the composite, a fixture was designed (Figs. 1a and b). The composite with 35 vol% of the reinforcement was manufactured by the hand lay-up process. Also, the microvascular channels were embedded into the composite by applying the retractable solid preforms/cores technique. First, the four plies of the glass fibers were cut with the proper dimensions. Then, these fibers were wetted by the resin-hardener mixture. After that, the glass fibers and the uncured mixture were set on the fixture plate and rigid polymeric wires were embedded into the structure. It should be noted that these rigid wires were covered by silicone wax for easier pulling out from structure after curing process. In the following, the wires were stretched in a parallel manner and fixed (as can be seen in Fig. 1c).

To reduce the trapped air voids into the composites, a press was applied on the composites during the curing process. After curing for 4 h at room temperature, the embedded solid preforms were pulled out from the structure to keep the hollow channels and reserve the healing

agents for them (shown in Fig. 1d). After 5 days as the post curing time, the laminate sheets were machined as the tensile and flexural samples. Afterwards, the healants were injected into the hollow micro-channels via the insulin syringe (seen Fig. 1e). Fig. 1f shows the fabricated smart composite containing the filled micro-channels by the healant. Then, the fabricated composites were damaged for assessing the healing process (Fig. 1g). After that, the samples were placed in the oven at 130 ˚C for 30 min to ensure healing the damaged zones (Fig. 1h).

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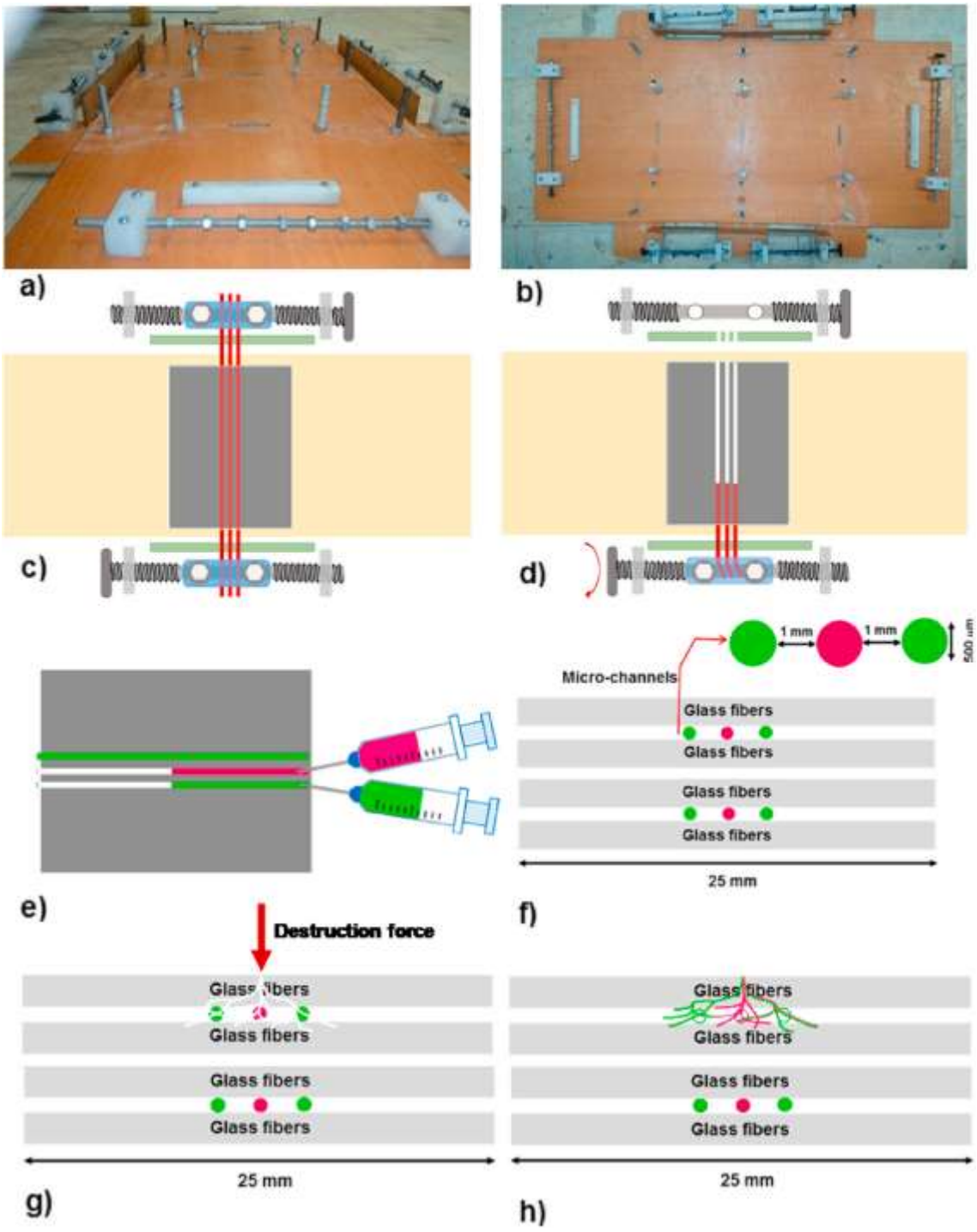


**Fig. 1.** The fabrication process of the self-healing composites: a and b) the designed fixture, c) embedding and fixing the polyamide wires, d) removing the polyamide wires, e) filling the composite by the three-part healant, f) the cross-section of the composite, g) damaging the composite, and h) healing the composite

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### 2.3. Mechanical testing

The tensile and flexural strength of the specimens was obtained from stress-strain curves, according to ASTM D3039 [31] and ASTM D790M [32], respectively, by using a Hounsfield universal testing machine (Model H25KS, UK). The tensile test specimens were damaged through three-point bending as a statistic method with the deflection of 8 mm. The flexural test specimens were damaged through the Charpy low-velocity impact as a dynamic method with the 103 J impact energy. After the initial damaging, the microcracks and delamination would occur in the structure, which could be regarded as the pre-requisite for triggering the healing mechanism. The most important parameter considered in the evaluation of the healing ability was the ultimate strength, which was determined for each samples. Afterwards, the healing efficiency of the samples (η) were assessed by using equation (1) [33].

$$\eta = \frac{\sigma_{healed} - \sigma_{damage}}{\sigma_{virgin} - \sigma_{damage}} \quad (1)$$

where $\sigma_{healed}$, $\sigma_{damaged}$ and $\sigma_{virgin}$ refer to the tensile or flexural strength of the healed, damaged (0 day healed) and virgin (without any damage) samples, respectively. The samples were named by the four-part parameter. The first parameter is the type of test, where F is the flexural test and T is the tensile one. The second one shows the undamaged (U) or damaged (D) mode of the samples. The third parameter is the vol % of the healing agent, which was 0, 2.5, 4 and 8 for the tensile samples and 0, 2, 3.2 and 3.7 for the flexural ones. The last parameter represents the healing time (0, 4, 7 and 11 days). It should be noted that the healing volume fraction of 2.5, 4 and 8 in the tensile samples was obtained by embedding 6, 10 and 10 micro-channels with the diameter of 500, 500 and 700 μm, respectively. Also, the healing volume fraction of 2, 3.2 and 3.7 in the flexural samples was obtained in a similar way.

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### 2.4. Characterization

To confirm the healing of these smart composites, the field emission scanning electron microscopy (FESEM), Mira 3 Tescan (Czech) was used. Also, by using the energy dispersed spectroscopy (EDS) analysis, the existence of the catalyst elements was characterized.

## 3. Results and discussion

### 3.1. Tensile test

Fig. 2 shows the stress-strain curves of the composites. The obtained results from Fig. 2; and equation (1) are reported in Fig. 3. According to Fig. 3, it could be seen that there were no impressive changes in the virgin and neat samples. The tensile strength for the neat sample was about 238 MPa, whereas for the composites containing 2.5, 4 and 8 vol %, this was 237, 230, and 217 MPa, respectively. By comparing these results, it could be said that by increasing the volume fraction of the healant, the tensile strength of those composites had a reducing trend, which was 0.4, 3.4 and 8.8%, respectively. According to Fig. 3, it could be seen that the composite with the 2.5 vol% healant had the tensile strength of 117 MPa, after creating the initial damage. This means that this composite kept 49.4% of the initial tensile strength and lost 50.6% of it. By increasing the volume fraction of the healant, the percentage of the remaining tensile strength had the reducing trend; so, this percentage reached to 41.6%. Comparing the healant volume fraction of 2.5 and 4 showed that the reduction of tensile strength was negligible.

Through the initial damage creation, different defects could be created in the structure, like fiber breakage, surface cracks, fiber pull out and delamination. The interlaminar shear stress could be regarded as the main reason for delamination, which had no major impact on the neat sample. However, it could influence the samples with the micro- channels, due to the air voids,

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porosity, gallery and resin concentration zones. The air voids could be reduced by using the static press during the manufacturing process. However, the created porosities were an avoidable problem causing the poor bonding between the matrix and plies, thereby increasing the delamination phenomenon. According to the literature [34], the sources of porosities are the resin concentration zones, mass conservation and an uneven local pressure distribution (Fig. 4).

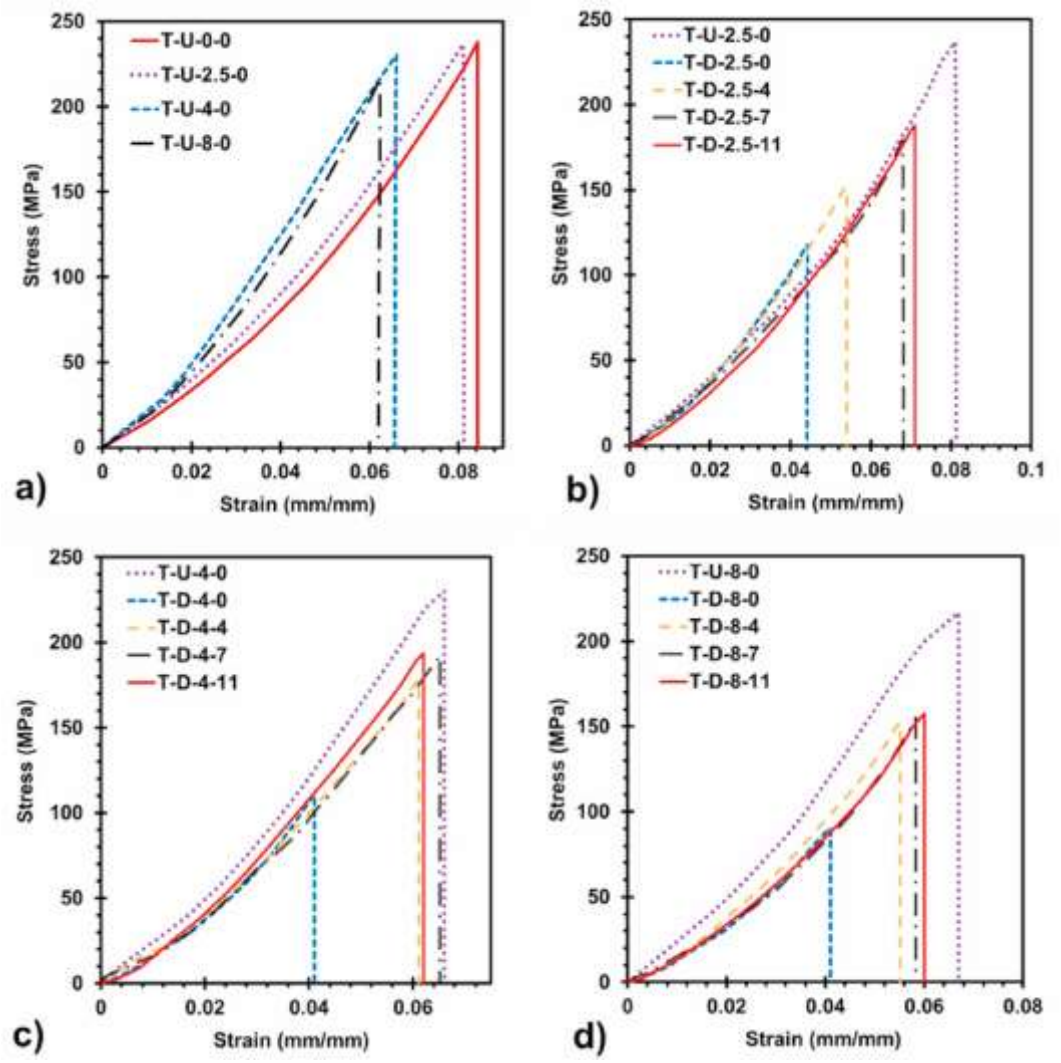


**Fig. 2.** The stress-strain curves of the composites containing various volume fractions of the healant under tensile loading: a) virgin composites, b) the composite with 2.5 vol% healant, c) the composite with 4 vol% healant, and d) the composite with 8 vol% healant.

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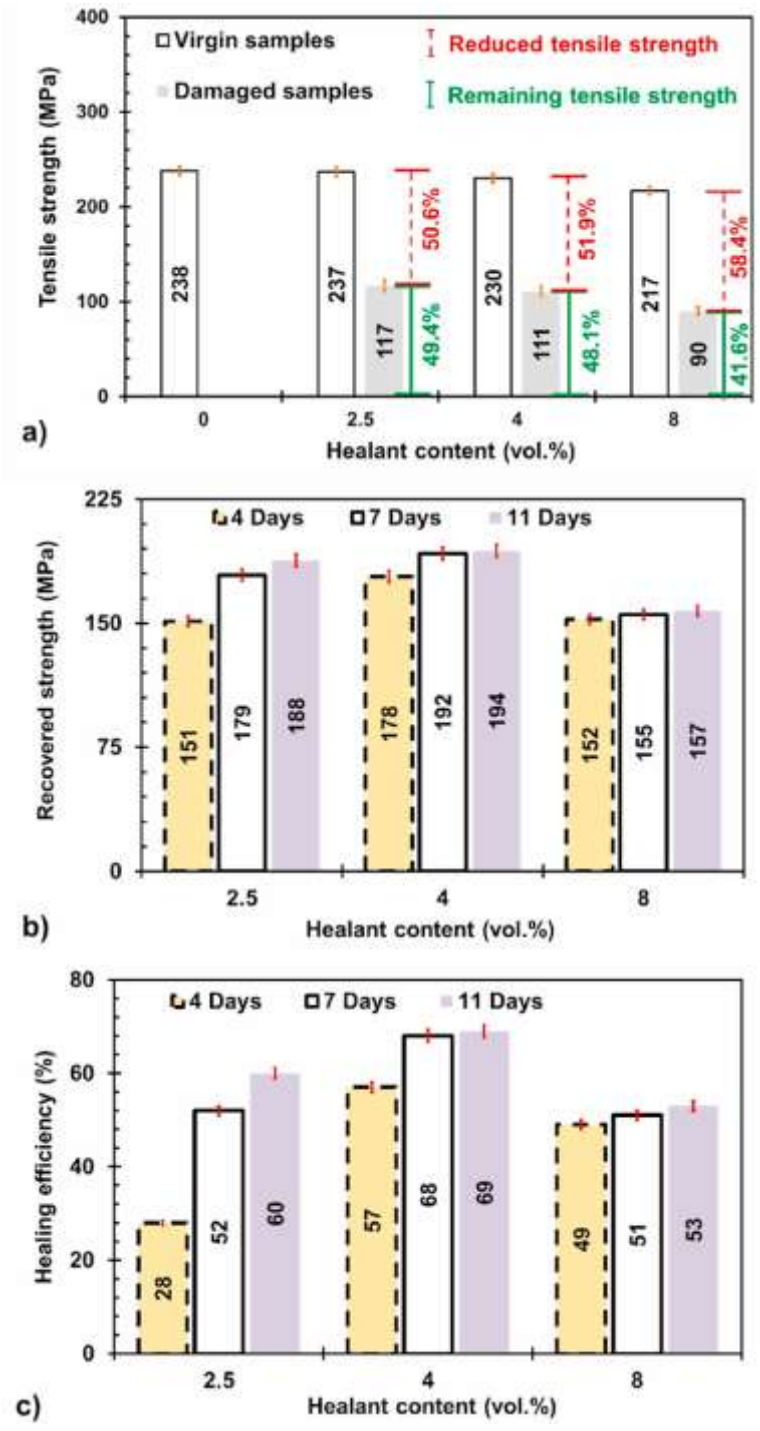


**Fig. 3.** The tensile properties of the self-healing composites, a) the initial, remaining and reduced tensile strength, b) the recovered strength, and c) the healing efficiency.

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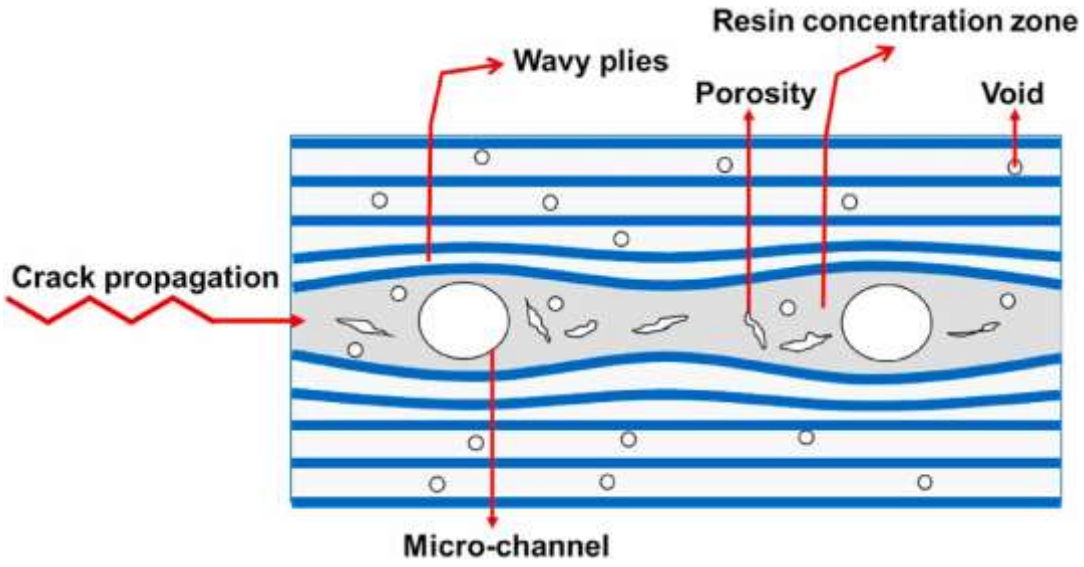


**Fig. 4.** Different defects into the self-healing composite based on the micro-channel healing system.

In other words, with the constant volume fraction, embedding the polyamide wires could lead to creating the resin concentration zones for adhering those wires between the composite's layers. Therefore, based on the mass conservation rule, the resin was transferred from other areas. This transferring could create the porosities in the composites. By increasing the volume fraction of the micro-channels, the percentage of the porosities had an increasing trend, which led to easily propagating the microcracks into the composites. Based on these influential factors, it could be mentioned that by increasing the volume fraction of the micro- channel in the composite, the tensile strength and the remaining tensile strength had the reducing trend. This was because the porosities, small air voids and cracks could join together, leading to the delaminating or failure of the composite structure.

Fig. 3b and c shows the recovered tensile strength and the healing efficiency of the composites after the healing process. According to Fig. 3c, it could be realized that the composite containing the 2.5 vol% healant had the minimum healing efficiency after 4 days,

which was 28%. By enhancing the healing time, the healing efficiency had the increasing trend, so it reached to 60%. To better understand this trend, it would be better to investigate the healing process. Percolating the healant from the micro-channel to the damage area was done based on their pressure disparity. The pressure disparity could cause the healant to flow into the damage area by the capillary force [28]. By raising the healing time, the flowing of the healant to damage area was increased, improving the healing efficiency. Increasing the volume fraction of healant is another way that can improve the healing ability by raising the pressure disparity.

According to Fig. 3c, it could be observed that by increasing the healant volume fraction from 2.5 to 4%, the healing efficiency was raised to 57%after 4 days. By embedding the 8 vol% healant, the healing efficiency was, however, reduced. This reduction was due to the increase of the micro-channels size and number, which led to raising the fiber distortion angle [35]. The rise of that angel led to enhancing the fiber volume fraction above and below channels [34], which caused the increase of the local stress-strain distributions and reduced the performance of the structure. Also, increasing the micro-channels size could raise the wavy piles and resin concentration zones. Another factor that should be noted here is the interlaminar shear strength. The interlaminar shear strength is sensitive to the voids, porosities vacancies and micro-channels as the hand-made defects in the structure. So, the presence of micro-channels and the increase of their diameters could lead to the increase of the galleries, significantly diminishing the load bearing capacity of the structure, because of the lack of the reinforcement in these areas [36]. In the light of the above, it can be said that the composite containing the 4 vol% healant after 7 days had the optimum results, as compared with other samples.

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### 3.2. Flexural test

The stress-strain curves of the composites have been shown in Fig. 5. As can be seen, the flexural strength, the remaining flexural strength, the reduction percentage, the recovery of flexural strength and the healing efficiency were calculated, as illustrated in Fig. 6. As shown, by raising the healant percentage, the remaining flexural strength had the increasing trend, whereas there was a decreasing for the tensile strength. This behavior under flexural loading was due to the damaged width and the resin concentration zones. There is a dichotomy for the resin concentration zone length and width in the structures. The length and width of these zones are related to the number and diameter of the micro-channels. To further explain, Fig. 7 is brought for illustration.

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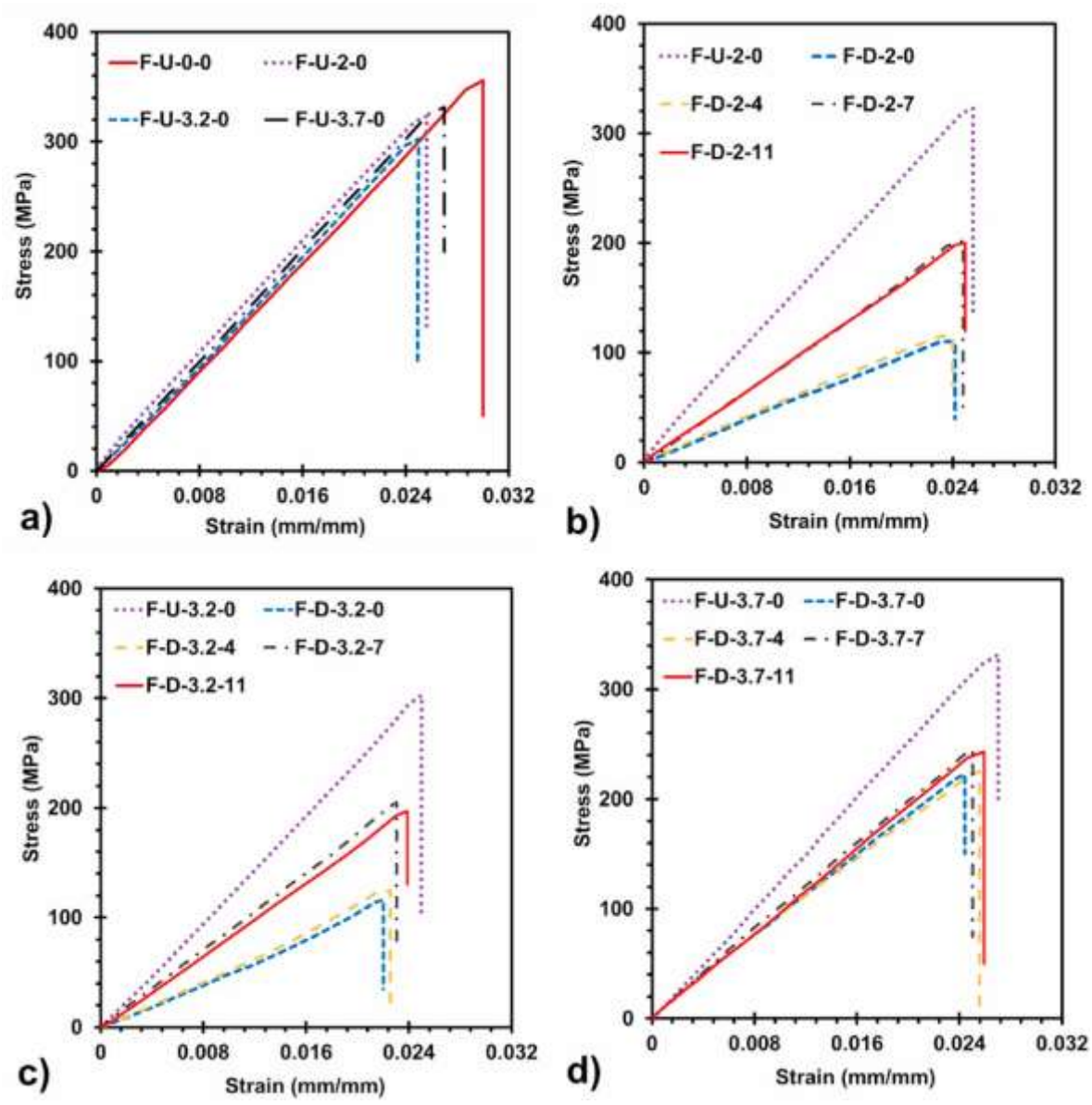


**Fig. 5.** The stress-strain curves of the composites containing various volume fractions of the healant under flexural loading, a) virgin composites, b) the composite with the 2 vol% healant, composite with the 3.2 vol% healant, and d) the composite with the 3.7 vol% healant.

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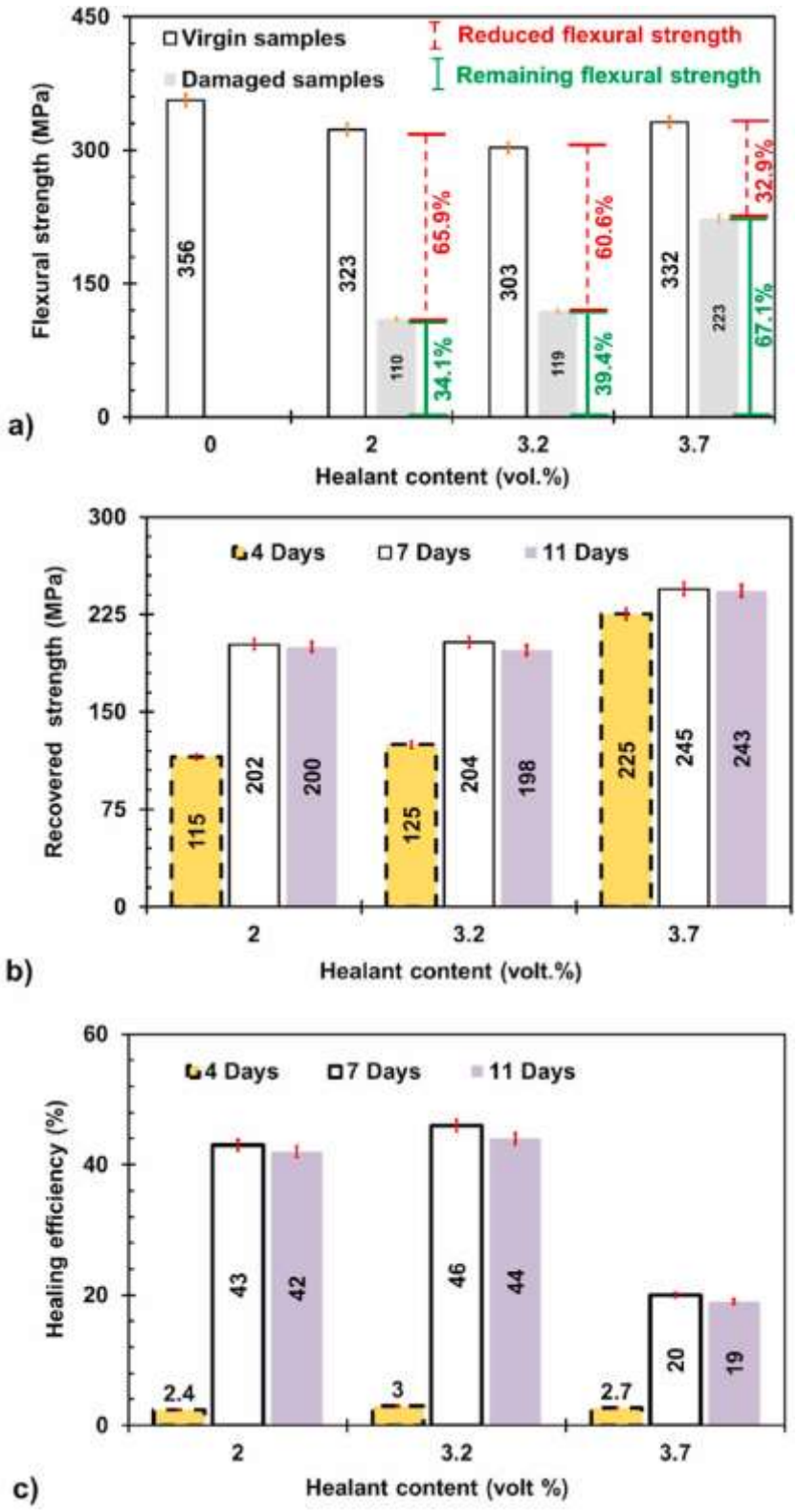


**Fig. 6.** The flexural properties of the self-healing composites, a) the initial, remaining and reduced flexural strength, b) the recovered strength, and c) the healing efficiency.

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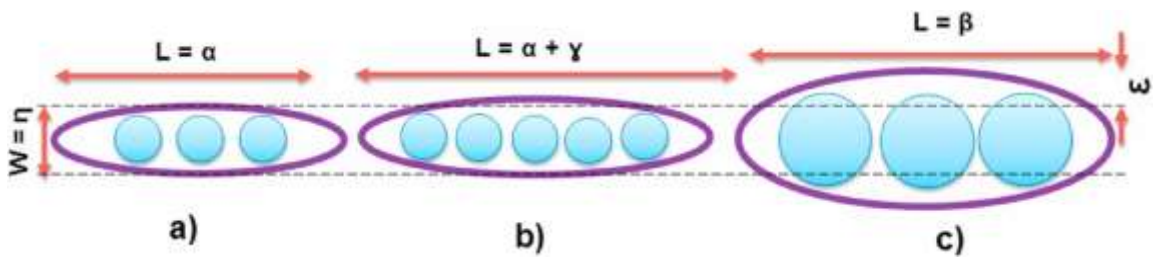


**Fig. 7.** The resin concentration zone in the composites with various volume fractions, a) 2 vol%, b) 3.2 vol%, and c) 3.7 vol%.

According to Fig. 7, by increasing the healant vol% from 2 to 3.2, the length of the resin concentration zone was raised from α to $\alpha+\gamma$, but the width of it remained at a constant value of η and the thickness had no significant change. According to the obtained results, as shown in Fig. 6a, by increasing the healant from 2 to 3.2 vol%, despite the increase of the resin concentration zone length, the remaining flexural strength was raised from 110 to 119 MPa. By considering this point, it could be mentioned that the number of the micro-channels factor had no significant effect on the reduction of the flexural strength. However, by increasing the healant vol% from 3.2 to 3.7, although the number of micro-channels was decreased, the diameter of them was increased, which led to the increase of the width of the resin concentration zone up to $\eta+2\epsilon$ (seen Fig. 7). This increase enhanced the thickness of the composite, consequently improving the remaining flexural strength. In the light of above, it could be mentioned that the micro-channels diameter factor had the most effect on the flexural strength than the number of micro-channel factors through the initial damage creation.

Based on the literature [34], the micro-channels diameter increases the fiber distortion angle and height, which could enhance the fiber distortion and fiber volume fraction through the thickness direction. Enhancement of the volume fraction can raise the absorbing energy during the initial damaging process, consequently increasing the remaining flexural strength.

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For this reason, in the composite with the micro-channel diameter of 700 μm, the remaining flexural strength was higher than that of the composite with the 500 μm micro-channel diameter. Although improving the absorption energy increased the remaining mechanical strength, it could reduce the healing ability. This is because, for healing the damage area, the minimum destruction force is needed. In other words, the mechanism for the activation of the healing process is the destruction of the micro-channels [37]. Therefore, by reducing the destruction micro-channels, the percolation of the healant into the damage zones is reduced. For this reason, the healing efficiencies of the composites in the flexural loading were lower than those in the tensile loading.

A similar behavior could be observed by comparing the composite containing 3.2 and 3.7 vol% of the healant in Fig. 6b and c. According to these figures, it could be seen that the composite with the 3.7 vol% healant had 225, 245 and 243 MPa flexural strength, after 4, 7 and 11 days of the healing process, respectively. On the other hand, the recovered flexural strength after those days for the composite with the 3.2 vol% healant was 125, 204 and 198 MPa. In contrast, upon 4, 7 and 11 days, the composite containing the 3.2 vol% healant, with 3, 46 and 44% healing efficiency, had higher healing efficiencies in comparison to the one containing the 3.7 vol%, with the healing efficiency of 2.7, 20 and 19%. Therefore, it could be said that in the composite structures, in addition to the healing ability of the structure, the remaining initial properties could play an important role in recovering the mechanical properties.

### 3.3. Fracture analysis

To confirm the created damage and percolate the healant into the damaged zone in the flexural and tensile samples, these samples were filled by ultraviolet (UV) dyes. Fig. 8a–c

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shows the composites before and after damage under flexural (for the tensile sample) and impact (for the flexural sample) loading. As can be seen, after the creation of damages, the healant was diffused into the damaged areas, filling them. This means that these structures had the self-healing ability. Fig. 8d and e represent the composite containing the 2.5 and 4 vol% healant. According to these figures, it was observed that the healant could approach the edge of cracks in the samples containing the 2.5 vol% of it, whereas in the sample containing the 4 vol% of that, the crack completely filled the damaged zone. This means that by raising the healant volume fraction, the healing ability of the composite could be increased. For the characterization of the healed damage zones, the microscopic investigation would be so necessary.

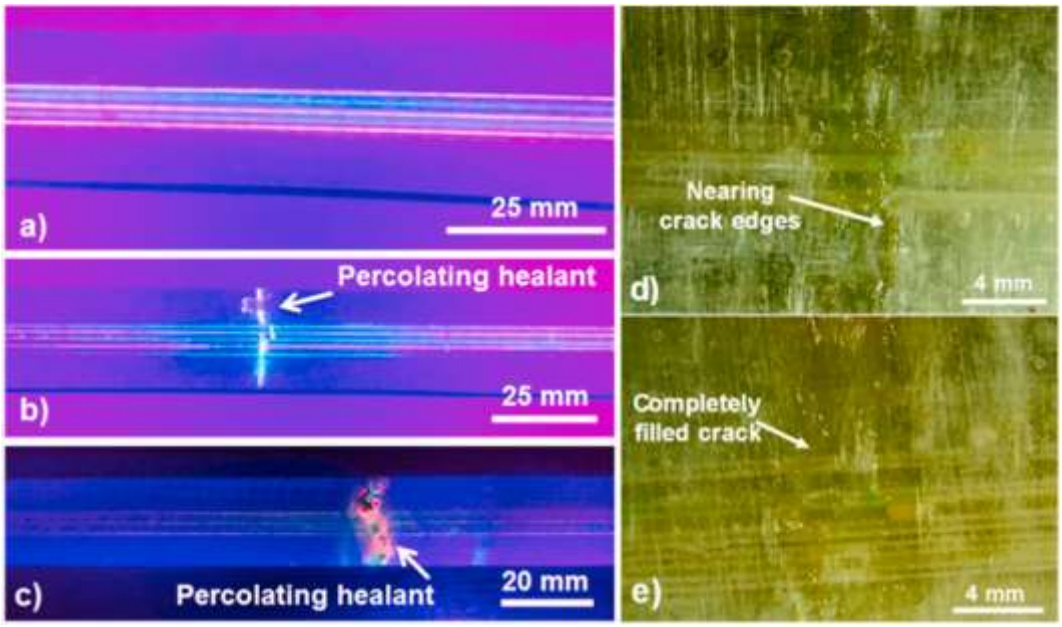


**Fig. 8.** Macrograph of the self-healing composite, a) before the damage containing the UV dyes, b) the damaged tensile sample containing the UV dyes, c) the damaged flexural sample containing the UV dyes, and d and e) the percolating healant in the composite containing the 2 and 4 vol% healant.

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Fig. 9 shows the FESEM analysis of the damaged zone of the composite. After the initial damage creation, the generated cracks led to the connection of the micro-channels. Then, the micro-channels content slowly flowed to the damaged site. So, after the passage of time, the healants remained in form of a rough surface on the fracture surface of the composite, which could be more recognizable than the smooth surface of it. To confirm this about the rough surface of the healed samples, the EDS analysis was conducted, as can be seen in Fig. 9b. The Cu and Br elements were indicative of the healed structure in the damage zone. Another interesting phenomenon in Fig. 9a is how the micro-channel was blocked by the healants. As can be seen, the hardener and premix catalyst reached to resin, which blocked the micro-channel. Fig. 9c, shows the fracture surface samples after the tensile test. It was clear that the healing agents were diffused into the damage site, creating the rough polymeric shell. To confirm this, the EDS analysis was done, as marked by an arrow in this figure. The Cu and Br elements were representative of the catalyst and the hardener premix in the damage area (Fig. 9d). The closing crack area can be seen in Fig. 9e. According to this figure, the healants filled the damaged zone. By using the EDS analysis, as shown in Fig. 9f, the existence of the Cu and Br elements in it was proved, which was the sign of the healed area. In the light of the above, it could be said that the crack as the provocative agent activated the healing system by breaking the micro-channels. Then, the pressure disparity between the micro-channels and the composites acted as the agent for flowing the healants into the damage zones. After that, the healants formed the polymeric shells in the damaged areas. These polymeric shells were near the edges of the cracks, recovering the initial tensile and flexural strength of the composites.

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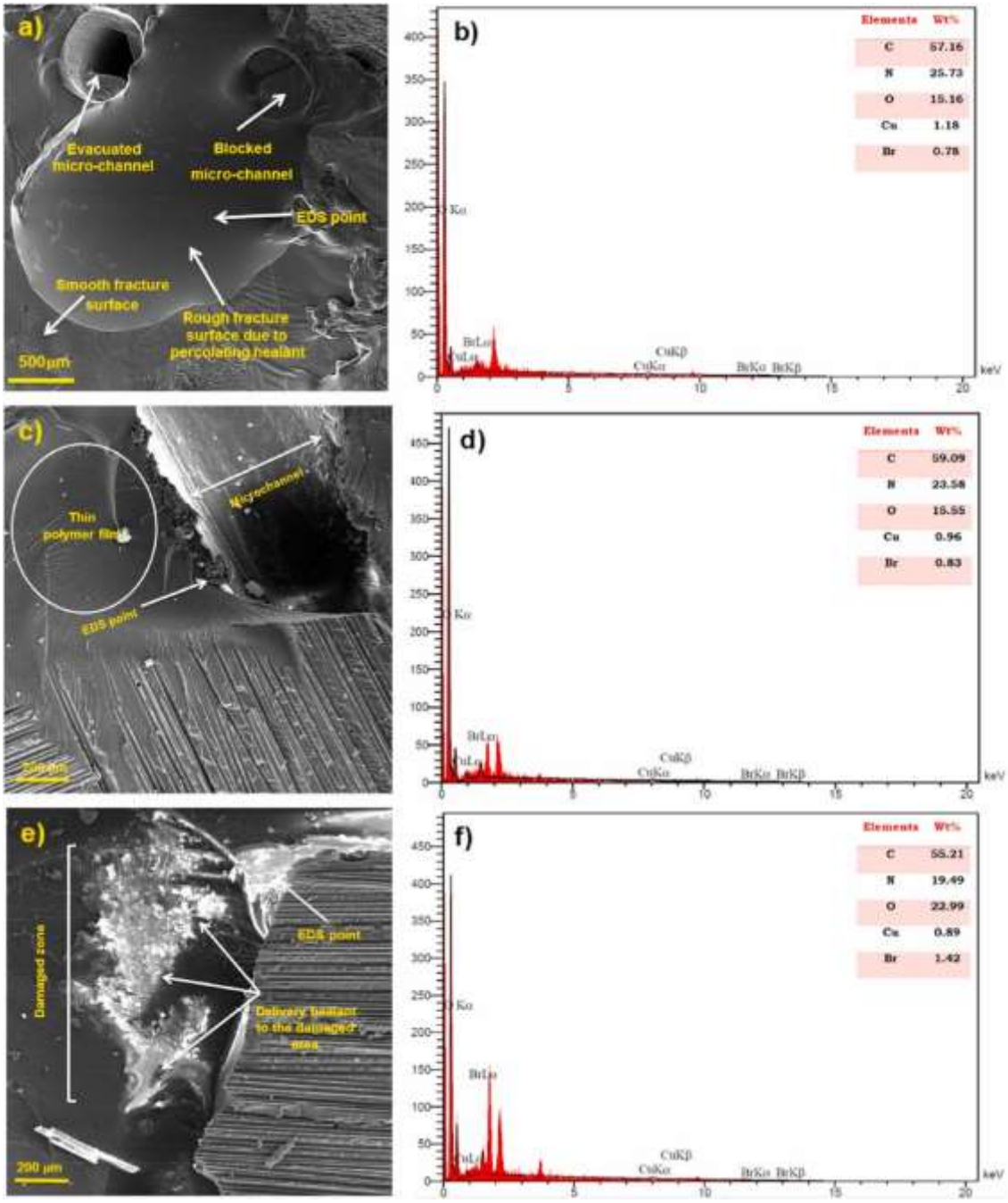


**Fig. 9.** The FESEM and EDS analysis showing the healing process, a and b) the healant flowing, c and d) the formation of the polymeric shell, and e and f) the crack filled by the healing system.

## 4. Conclusions

In this research work, the self-healing ability of the glass fibers-epoxy smart composites based on the microvascular channel under the tensile and flexural loading was investigated. In this healing system, the epoxy resin and the premix anhydride hardener-$CuBr_2$(2-Methylimidazole), were used as the three-part healant. The percentage of the healant for the tensile test was 2.5, 4 and 8 vol%, whereas for the flexural test, this was 2, 3.2 and 3.7 vol%. Also, the healing time for both tests was 4, 7 and 11 days. The results obtained from the tensile test showed that the composite containing the 4 vol% healant had the maximum recovered tensile strength, which was 178, 192 and 194 MPa after 4, 7 and 11 days, respectively. Also, the maximum healing efficiencies in the tensile test belonged to those composites, which were 57, 68 and 69%. The maximum recovered flexural strength was related to the composites containing the 3.7 vol% healant, which was 225, 245 and 243 MPa, after the healing times of 4, 7 and 11 days. On the other hand, the composite with the 3.2 vol% healant had the maximum healing efficiencies, which were 3, 46 and 44%. Based on the obtained healing efficiency, the composite could not recover their initial tensile and flexural properties, after the healing time of 4 days. However, after 7 days, the composite had the proper recovered strength and healing efficiency. Also, it was found that these properties had negligible changes after 11 days.

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