

Development and characterization of smart composites reinforced with fibrillated cellulose and nickel-titanium alloy

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ABSTRACT

The current study presents the synergistic effects of fibrillated cellulose (FC) and nickel-titanium (NiTi) alloy on the performance properties of smart composites. Epoxy resin was reinforced with loadings of 1 %, 3 %, and 5 % FC and 3 % NiTi. The composites were produced using the casting method. The morphological properties have been analyzed using scanning electron microscopy (SEM). For mechanical properties, yield strength, modulus of elasticity, hardness, and impact energy were determined. The corrosion rate was determined via electrochemical corrosion testing. The recovery test was used to measure the shape-memory of the composites. The self-healing of the artificial defect in the composites was observed using a thermal camera. The yield strength, modulus of elasticity, hardness, and impact energy of composites reinforced with 5 % FC and 3 % NiTi increased by 168.2 %, 290 %, 33.3 %, and 114.3 %, respectively, compared to pure epoxy resin. There has been a 56.3 % decrease in the corrosion rate. The percentage of composites that returned from the final state to the original state after a deformation was 4 %. Self-healing analysis revealed that the scratch defect in composites was healed after 24 h. It is concluded that smart composites can be used in the aviation and automotive industries.

1. Introduction

Materials science has played an important role in the ground-breaking developments in the field of technology in recent years. Smart materials are one of the most interesting research areas of the 21st century. They have become advanced materials that solve material problems during on-site use. Smart materials are new types of materials designed to change their properties (shape, stiffness, fluidity, light transmittance, vibration damping, etc.) in a controlled manner depending on external factors such as temperature, pH, stress, humidity, electric charge, magnetic field, light, etc., or any stimulus. Smart materials can perceive the changes in their environment, show autonomous behavior, and respond to the stimulus. In recent years, self-healing and shape-memory materials have attracted the increasing interest of the scientific community [1].

Self-healing materials are those that, in the case of physical damage, may recover all or some of their original qualities [2]. The innate ability of biological systems to heal themselves serves as inspiration for the self-

healing mechanism, which is capable of self-healing [3]. Some of the self-healing mechanisms are that human skin repairs itself after being damaged; lizards repair and restore the damage after their tails are severed; planarians repair themselves even if they are divided into many parts; zebrafish repair and restore the damage even if a part of the heart is cut off; and axolotls restore the lost limb even after losing a limb. Today, many approaches have been applied to provide materials with the capability to self-heal. Two main approaches are based on mechanisms (internal and external self-healing) and working chemistry (autonomous and non-autonomous) [4]. Various techniques include capsule-based healing [5,6], fiber and/or vascular healing [7], and external stimulus-induced healing [8]. The external self-healing approach provides self-healing through the use of healing agents, whereas the internal self-healing approach provides a healing process that does not require any healing agents. Several authors were successful in producing epoxy-containing microcapsules [9,10].

Shape-memory materials (SMMs) can return to their original shape after being deformed by a specific stimulus, and they retain their

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original shape even when exposed to external stimuli [11]. These shape alterations arise from the temperature-induced austenite-to-martensite phase transformation [12]. While the martensite phase is stable at low temperatures, the austenite phase is stable at high temperatures. The creation of these thermomechanical characteristics in shape-memory alloys is due to a reversible solid-solid phase change [13]. Nickel-titanium alloy, also known as nitinol or NiTi, is a shape-memory material that is widely used in a variety of applications, including biomedical, aerospace, automotive, and micro-electromechanical systems (MEMS) [14]. It has special properties, including shape-memory behavior, superelasticity, biocompatibility and corrosion resistance [15,16]. In the literature, several investigations on NiTi wires have been carried out in recent years [17,18]. Dawood et al. [19] investigated the bonding behavior of shape-memory nitinol wires to carbon fiber-reinforced polymers. The authors reported that superelastic SMA wires embedded in fiber-reinforced composites can be used to characterize the bonding behavior. Salowitz et al. [20] tested the self-healing capabilities of a metal matrix composite reinforced with nickel-titanium (NiTi) shape-memory alloy wires. The authors reported that the specimens recovered 92 % of their original strength, indicating good adhesion between the NiTi fibers and the Sn–Bi matrix. Chen et al. [21] developed a valve prototype with a polymer matrix reinforced with NiTi wires in the radial direction. The authors reported that the results of the study showed that the maximum fundamental stress in the PET fabric matrix and the percentage of area corresponding to the high stress level were significantly reduced due to the NiTi wires. Although many studies have been carried out on NiTi to date, most of the reported studies are limited to shape-memory wires. It has rarely been reported that NiTi powders have been introduced into epoxy resin, and there is a lack of evidence in the literature.

Nanotechnology has had a rapid impact on the field of smart materials in recent years, enabling this field to develop faster and differently [22,23]. With the use of nanotechnology, it will be possible to create not only smart materials that can react but also new-generation materials and systems that can exhibit highly complex behaviors in the coming years. Cellulose is a most impressive natural biopolymer and a renewable, sustainable raw material [24,25]. Nanocellulose is a term used to describe nano-sized cellulosic materials with lengths ranging from 1 to 100 nm from trees, annual plants, agricultural leftovers, algae, bacteria, or tunicates for nanotechnology applications [26]. Fibrillated cellulose (FC), micro/nanofibrillated cellulose (MFC/NFC), or cellulose nanofibrils (CNFs) derived from renewable biomass are currently attracting more attention as they have the characteristics of biocompatibility, high mechanical strength, low thermal expansion, and environmental benefits [27–30]. FC consists of both crystalline and amorphous areas; hence, it has excellent mechanical properties when compared to glass and natural fibers [31].

Epoxy is one of the most versatile polymers widely used in many industries, especially in composite production [32]. Its uses are limited to high-performance adhesives, though, as it cures to produce a very complex cross-linked network structure that is weak and brittle by nature in addition to having low thermal stability and viscoelastic properties. [33]. As a result, in order to increase their applications, epoxy resins need to be reinforced with additives [34]. The most promising option is to include some dispersed micro- and nanoparticles in the second phase, such as cellulose nanomaterials, metal particles during polymerization, or additives [35]. In this context, many studies have been conducted in recent years to enhance the mechanical properties of CNF-reinforced epoxy composites. Over the last 10 years, research has shown that NFC/CNF can be used alone or mixed with epoxy and other polymers to produce composites with improved performance properties. Barari et al. [36] reported that CNF-reinforced composites showed improved mechanical and tribological properties compared to pure epoxy samples. The authors also found that composites reinforced with chemically modified CNFs performed better than untreated composites. Li et al. [37] reported that high aspect ratio cellulose nanofibrils

improved the mechanical properties, manifested by a high modulus of elasticity, high stiffness, and high yield strength. Nair et al. [38] found that the modulus, strength, and strain of composites obtained by adding 18–23 wt% CNF to epoxy increased significantly. Roszowska-Jarosz et al. [39] reported a satisfactory improvement in the mechanical properties of epoxy composites reinforced with nanocellulose between 0.5 and 1.5 wt%. Kurita et al. [40] reported that when the CNF volume fraction was higher than 0.37 % by volume, it was strengthened by mechanical interaction, and the tensile and flexural moduli of epoxy resin increased with the addition of CNF. Despite many studies about NFC, there is not any study in the literature on the shape-memory, self-healing, and electrochemical corrosion properties of FC-containing composites.

The aim and rationale of the study are to prevent problems by preserving the strength properties of composites in case of damage by providing shape-memory and self-healing properties. The importance and novelty of this study are to evaluate the synergistic effects of incorporating a lignocellulosic bionanomaterial FC filler and a biocompatibility metallic smart material NiTi alloy into the epoxy at different loading levels to extend the service life, reduce the cost, and expand the application areas of composites. To the best of our knowledge, this is the first study on how the synergistic effect of lignocellulosic biomaterial and powder metallic material influences the performance properties such as mechanical, electrochemical corrosion, shape-memory, and self-healing properties of the composites.

2. Experimental

2.1. Materials

AC-510 transparent epoxy resin was used as the polymer matrix. Both liquid resin and hardener were purchased from Armor Chemical (Istanbul, Türkiye). Fibrillated cellulose (FC) and nickel-titanium alloy (NiTi) were used as reinforcements for the epoxy matrix composition. FC was produced at the Advanced Structures & Composites Center at the University of Maine (Orono, ME, USA). FC obtained from wood pulp by the 2,2,6,6,6-tetramethyl-piperidin-1-yl-oxyl (TEMPO) method was used. The FC used as reinforcement in the study is shown in Fig. 1.

Nickel (CAS Number 7440-2-0) and titanium (CAS Number 7440-32-6) powders were purchased from Alfa Aesar (Ward Hill, Massachusetts, USA). NiTi alloys (Ni:Ti = 50:50) with an average size of 30–50 nm and >99 % purity were produced.

2.2. Preparation of nickel-titanium alloy

Nickel (Ni) and titanium (Ti) powders were weighed in quantities determined according to the composition of the targeted NiTi alloy. Weighed Ni and Ti metal powders were placed in a suitable container

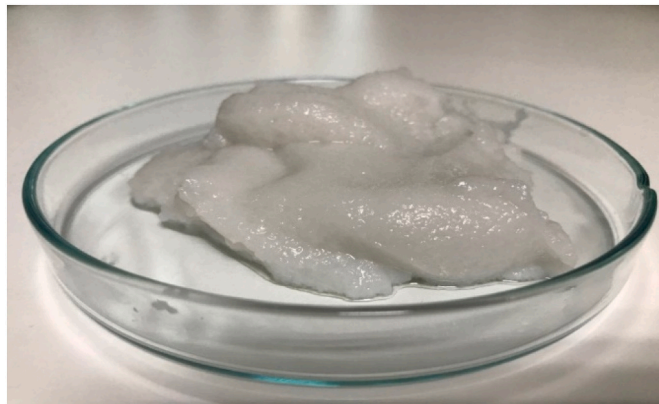


Fig. 1. Fibrillated cellulose.

with a metal powder/ball ratio of 1:10. 6 mm-diameter zirconia (ZrO₂) balls were used as grinders. Ni and Ti powders were mechanically ground in a ball mill at 400 rpm rotation speed for 15 h, and a NiTi alloy was produced with the mechanical alloying method.

2.3. Manufacturing of smart composites

In order to prove that the synergistic effect of FC and NiTi enhances the performance properties of smart composites, only pure epoxy and pure NiTi have been produced for comparison.

The manufacturing parameters of the composites are presented in Table 1.

The liquid epoxy resin and hardener components were mixed for 300 s at 2000 rpm in an ultrasonic homogenizer at a mixing ratio of 2 Part A (epoxy resin) + 1 Part B (hardener). NiTi was added to the prepared epoxy and hardener liquid mixture at 3 wt% and mixed in an ultrasonic homogenizer at 2000 rpm for 300 s. Then, FC was added to the mixture at 1 %, 3 %, and 5 % and mixed in an ultrasonic homogenizer at 2000 rpm for 300 s. The reinforced-liquid mixture was poured onto a one-piece open-mold polytetrafluoroethylene (PTFE) panel. The samples were left to cure at room temperature for 96 h. At the end of four days, the composites produced were cut to the sample sizes specified in the relevant standard for each analysis. Three specimens were produced for each group, and the tests were performed in three repetitions.

2.4. Characterization

2.4.1. Scanning electron microscopy (SEM)

The microstructure and phase dispersion of the samples were studied using a scanning electron microscope (SEM) and the Phenom Pure G6 (Thermo Fisher Scientific). SEM surface morphology examinations were performed using a BSE detector at an accelerating voltage of 5 kV and a working distance of approximately 7.00 mm.

2.4.2. Tensile test

The tensile test is used to measure the force required to break the material and how much the sample stretches or elongates to reach this breaking point. The tensile test was performed according to the ASTM D-3039 standard [41]. The dog-bone-shape composites were prepared with dimensions of 250 mm in length, 25 mm in width, and 2.5 mm in thickness, and the tensile test was performed with a working speed of 10 mm/min using a DVT Devotrans device (Istanbul, Türkiye). Three test specimens were used for each sample, and average results were obtained.

2.4.3. Ultrasonic speed measurement

Due to the structure of the specimen and the difference in the measurement point, the modulus of elasticity can be found by different methods. In this study, ultrasonic velocity testing was used to determine the modulus of elasticity of the composites with an ultrasonic flaw detector (General Electric, USM Go).

Firstly, the surfaces of the composites were ground to sufficient flatness. Longitudinal and transversal ultrasonic wave velocities were measured using both pulse-echo and transmission methods. A pulse-receiver-type ultrasonic instrument and a 4 MHz normal beam

longitudinal wave transducer were used for the ultrasonic velocity measurements. As a coupling agent, oil was used for longitudinal waves and honey for transverse waves. A uniform and constant force is applied to the probe against the composites to maintain a constant couplant layer at the interface. Unknown velocities were determined by comparative measurements on a reference block whose ultrasonic velocity is accurately known. Ultrasonic velocities were measured from their back-wall echoes by changing the velocity on the instrument until the thickness of the composites was obtained with 0.01 mm precision.

The modulus of elasticity (E) was calculated by the following equation:

$$E = \rho V_T^2 \frac{3V_L^2 - 4V_T^2}{V_L^2 - V_T^2} \quad (1)$$

Among them, V_T and V_L are the ultrasonic longitudinal and transverse velocities, and ρ is the density symbol.

2.4.4. Impact test

The impact test is used to determine the impact resistance of the specimen against sudden loading and/or the amount of energy required by dropping a weight from a certain height.

In this study, the drop method was used to determine the impact energy. In the drop method, a hammer with a certain mass is dropped from different heights based on tests and samples. The impact test was performed according to the ASTM D1709-22 standard [42]. The composites for the impact test were determined using a weight drop impact testing Devotrans machine (Istanbul, Türkiye). A hammer was dropped on a 230 mm × 230 mm composite material from a height of 1.5 m (150 cm), with weights ranging from 180 to 400 g in 45-g increments. The results are given by averaging the hardness values obtained.

The impact energy of the composites was calculated using the following formula:

$$E_p = m \cdot g \cdot h \quad (2)$$

Among them, E_p is potential energy (joule), m is mass (kg), g is gravity (m/s²), and h is the height symbol. The acceleration of gravity is equal to the number 9.8, which corresponds to approximately 10.

2.4.5. Hardness test

The hardness of a material is the resistance shown against the immersion of another harder object. The Shore A scale is frequently used for measuring softer materials, whereas the Shore D scale is used for measuring rigid materials. Hardness test specimens were made with Shore D according to the ASTM D2240-15 standard [43]. The area and depth of the mark left on the surface of the material by the notcher, called a durometer, to which a certain amount of force was applied, were determined. The composites were placed on a flat, hard surface ~3 mm thick. The durometer was held firmly, and the notcher was placed on the test specimen in contact with the test substrate. The upper and lower surfaces of the specimens produced for this purpose and Shore D hardness were measured at three points. The results are given by averaging the hardness values obtained.

2.4.6. Electrochemical corrosion analysis

An aqueous solution containing 5 % NaCl was prepared. The pH value of the solution used was measured at 6.3. In electrochemical corrosion analysis, a polarization cell was set up consisting of an electrolyte solution, a reference electrode, a counter electrode(s), and a composite connected to a sample holder. The sample is also called the working electrode. The electrodes were connected to an electronic instrument, a 1-liter corrosion cell, using a potentiostat (Interface 1000, Gamry). A standard calomel electrode (SCE) was used as a reference electrode, and a high-density graphite electrode was used as a counter electrode. The composite sample was connected to the cell as a working electrode. The tests were carried out at room temperature. Data

Table 1
Manufacturing parameters of the composites.

Groups	FC content (%)	NiTi content (%)
Pure Epoxy	–	–
Pure NiTi	–	3
A	1	3
B	3	3
C	5	3

acquisition was carried out through computer software (Framework, Version 6.04, DC105, Instruments, USA), while data analysis was carried out by Echem Analyst Software, Version 6.04 (Gamry Instruments, USA). Tafel curves were obtained by scanning in the determined potential range using the reference electrode and cathode. Corrosion current density (I_{corr}) and corrosion potential (E_{corr}) values were determined from these curves, and the corrosion rate was calculated based on these data.

2.4.7. Shape-memory analysis

The shape-memory ability of composites was determined by the recovery test, which measures the extent to which the material can return to its initial shape after deformation. Initially, the first size of the composites was measured. The composites were placed between the press jaws, and a force of approximately 150 MPa was applied. Then the composites were placed in an oven at a temperature of 45 °C and kept in the oven. After waiting for 5 min, the last size of the composites was measured.

The recovery ability was calculated by the following formula:

$$\% \text{Recovery} = 100 \times \frac{\text{Last Size} - \text{First Size}}{\text{First Size}} \quad (3)$$

2.4.8. Self-healing analysis

Composites of the dimensions represented in Fig. 2 were prepared for the self-healing analysis. A scratch with a width of 1 mm and a length of 10–15 mm was created on the composites. The self-healing of the artificial defect (scratch or notch) in the composites was observed for 24 h using a thermal camera (IR imaging) device (Bosch GTC 400C). The thermal imager has a microbolometer-type infrared (IR) sensor. The thermal imager is capable of measuring temperatures between –10 and 400 °C. The resolution of the thermal imager is 0.1 °C.

3. Results and discussion

3.1. Scanning electron microscopy (SEM)

The SEM micrographs of the rupture surfaces as a result of mechanical analyses of the FC/NiTi-reinforced smart composites are shown in Figs. 3–5.

The surface of the pure epoxy composite has a smooth and glassy exterior surface. This reveals its typical brittle structure. The crack pattern is indicative of the brittleness of the pure epoxy composite, which leads to a very low resistance to crack propagation during impact fracture, reducing the mechanical strength [44,45]. Figs. 3 to 5 show the SEM micrographs of the fractured surfaces of the FC/NiTi-reinforced smart composites. Fig. 3 shows that the fibers are located where the

polymer breaks. Fig. 4 shows that some voids are opened and channels are available. Fig. 5 shows that the particles are aggregated in voids and spherical cavities. These micrographs show that the crack pattern for all the smart composites is quite different from that of the pure epoxy composite. The images of FC/NiTi-reinforced composites exhibited rough fractured surfaces with white spots, mainly due to the presence of FC. The roughness and irregular fracture surface of smart composites indicate that they are less brittle, with the presence of sharp edges and deep line markings between crack surfaces resulting from rapid crack propagation [46,47]. Proper dispersion and strong adhesion between the FC/NiTi and epoxy matrix resulted in improved yield strength, modulus of elasticity, hardness, and impact energy in the composites.

3.2. Mechanical analysis

As a result of the increasing mechanical effect, materials first change their shape, then lose their strength and break. Under low stresses, the shape change is elastic, and the shape change that occurs with the force applied to the material within the elastic limit disappears when the force is removed. When the stress value exceeds the elastic limit, plastic deformation occurs. The plastic deformation is permanent, and the deformation does not recover even if the force is removed. During the tensile test within the scope of the study, there was no slippage at the place where the specimen was fixed to the jaw. No fracture or crack formation was observed in the same area.

The mechanical properties of the pure epoxy, pure NiTi, and FC/NiTi-reinforced composites are presented in Table 2.

3.2.1. Yield strength

Yield is the end of elastic deformation and the beginning of plastic deformation during deformation. The yield strength defines the stress value at which plastic deformation begins. The yield strength was determined by determining the point where there is a deviation from linearity in the tensile/compression curve. The yield strength of all groups reinforced with combines FC and NiTi is higher than that of pure epoxy and pure NiTi. There is an increase of 81.8 % in yield strength values in group A, 131.8 % in group B, and 168.2 % in group C compared to pure epoxy. There is an increase of 2.6 % in yield strength values in group A, 30.8 % in group B, and 51.3 % in group C compared to pure NiTi. Although the NiTi ratio was constant, increasing the FC ratio from 1 % to 5 % also increased the modulus of elasticity. Composites reinforced with 5 % FC and 3 % NiTi showed the best synergistic effect, increasing by 168.2 % compared to pure epoxy and 51.3 % compared to only NiTi. Due to the synergistic effect, FC/NiTi-reinforced composites are harder and more durable than pure epoxy and pure NiTi composites, as evidenced by their higher yield strength. The small size and high surface area of the FC have provided more interface area, which led to good interfacial bonding between the FC, NiTi and epoxy matrix. This bonding prevented quick crack propagation and promoted better stress transfer between these three components. The synergistic effect of FC and NiTi, mainly due to the establishment of specific interactions and stress transfer between them, improved the yield strength. This is verified by data from SEM micrographs. The results obtained are supported by the results of studies conducted by other scientists [48–50].

3.2.2. Modulus of elasticity

The modulus of elasticity was observed to have a similar trend as that of the yield strength, where it also increased as the FC loading increased from 1 % to 5 %. There is a 150 % increase in the modulus of elasticity values in group A, 205 % in group B, and 290 % in group C compared to pure epoxy. There is an 11.1 % increase in the modulus of elasticity values in group A, 35.5 % in group B, and 73.3 % in group C compared to pure NiTi. The images from the SEM indicated strong adhesion between FC, NiTi and the epoxy polymer matrix, which was attributed to this result. The fibril structure of FC, providing a higher surface area, interacted with NiTi, increasing the composites' modulus of elasticity.

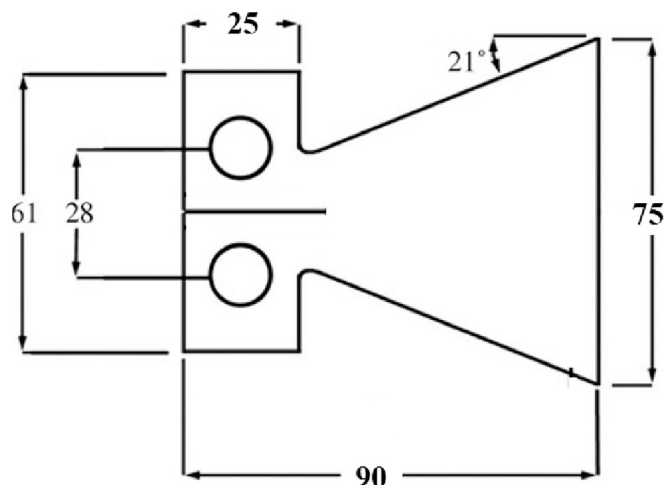


Fig. 2. Composites dimensions for self-healing analysis.

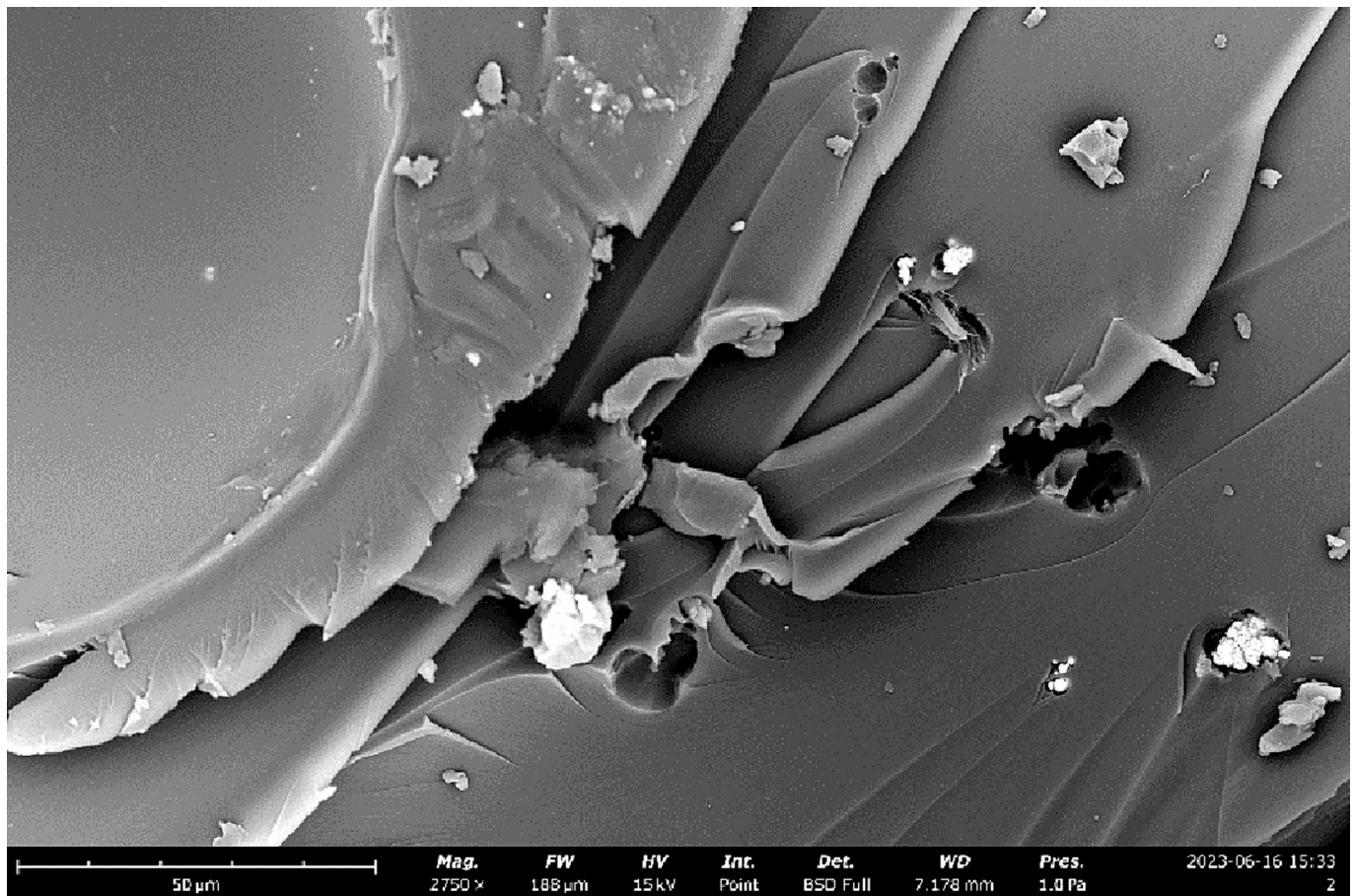


Fig. 3. SEM micrographs of the fractured surfaces of the 1 % FC and 3 % NiTi-reinforced smart composites.

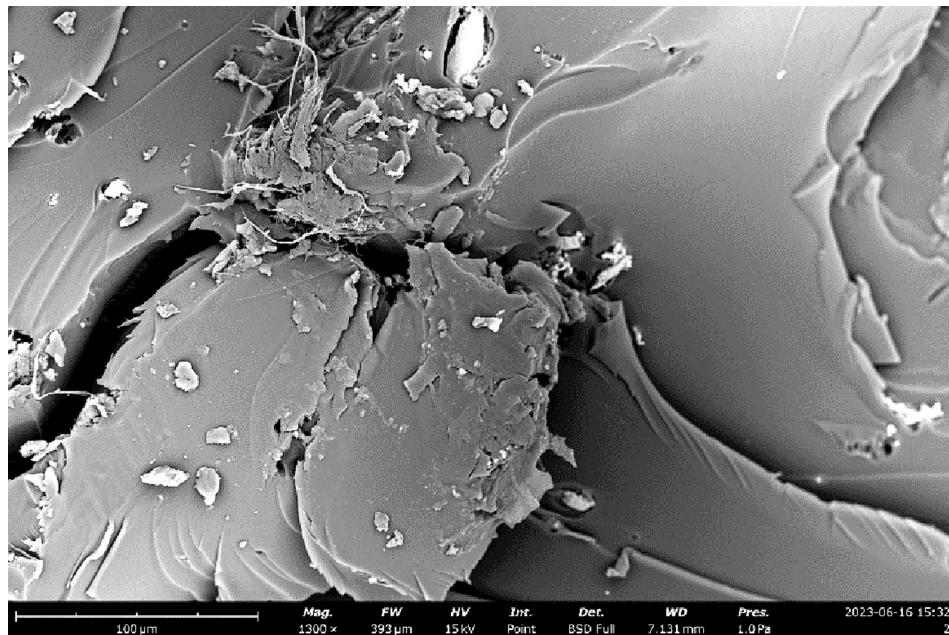


Fig. 4. SEM micrographs of the fractured surfaces of the 3 % FC and 3 % NiTi-reinforced smart composites.

This increase was attributed to the nanoscale of the fibrils, which provided more surface area, as well as the good interaction and bonding between the FC, NiTi and epoxy matrix, which resulted in better stress transfer; therefore, the elastic deformation of the composites increased.

By adding NFC to the epoxy matrix, voids in composites can be filled, and ultimately, the void in the polymer fibril matrix can be closed [48]. As the amount of empty space between the particles decreases, the flexibility of the polymer chain reduces, and the modulus of elasticity

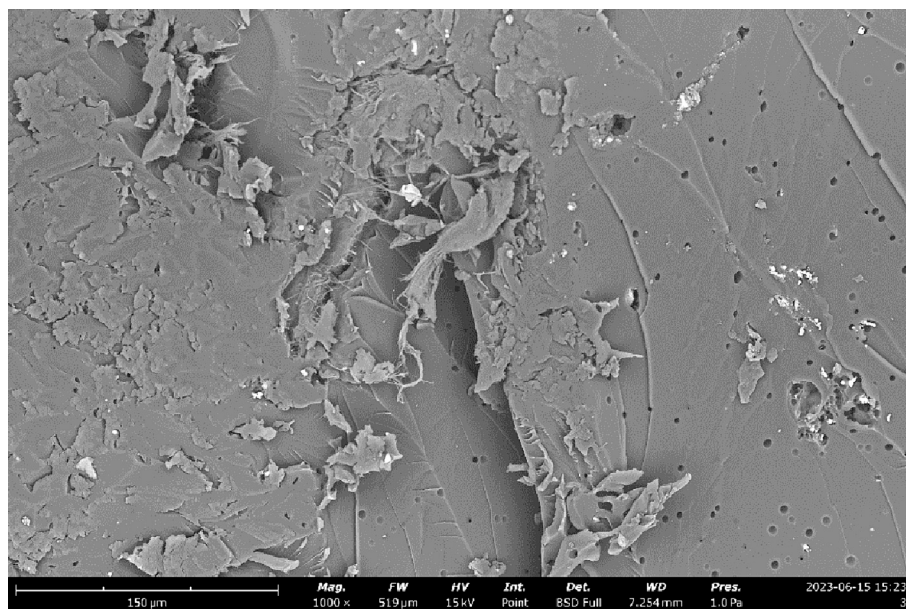


Fig. 5. SEM micrographs of the fractured surfaces of the 5 % FC and 3 % NiTi-reinforced smart composites.

Table 2

Mechanical properties of pure epoxy, pure NiTi, and FC/NiTi-reinforced smart composites.

Groups	Yield Strength (MPa)	Modulus of Elasticity (GPa)	Hardness (Shore D)	Impact Energy (kJ/m ²)
Pure Epoxy	22 (2.08)	2.0 (0.36)	60 (0.58)	28 (1.53)
Pure NiTi	39 (1.44)	4.5 (0.15)	65 (0.57)	40 (1.55)
A	40 (1.52)	5.0 (0.18)	66 (0.58)	41 (2.52)
B	51 (2.52)	6.1 (0.28)	74 (0.58)	54 (1.52)
C	59 (1.53)	7.8 (0.40)	80 (0.58)	60 (3.51)

Standard deviations are given in parentheses.

increases [51]. The results obtained are supported by Wongjaiyen et al. [32]. The authors investigated the effect of cellulose nanofibers on the mechanical properties of epoxy nanocomposites. The authors reported that the modulus of elasticity increased with increasing CNF content up to 3 wt%.

3.2.3. Hardness

The Shore D hardness scale provides values between 0 and 100. Higher values indicate harder materials that have higher durability against indentation. The hardness of all groups reinforced with FC or NiTi is higher than that of pure epoxy. This is verified by data from SEM micrographs. There is a 10 % increase in hardness resistance values in group A, 23.3 % in group B, and 33.3 % in group C compared to pure epoxy. There is a 1.5 % increase in hardness resistance values in group A, 13.8 % in group B, and 23.1 % in group C compared to pure NiTi. Increasing the FC content from 1 % to 5 % increased the resistance of smart composites to hardness. 5 % FC and 3 % NiTi-reinforced composites indicated the highest synergistic effect, increasing by 33.3 % compared to pure epoxy and 23.1 % compared to only NiTi. The hardness of reinforced composites was significantly increased due to the network structure between the fibrous cellulose and the NiTi and its synergistic effect. The NiTi has higher hardness and stronger work-hardening properties, while the FC has high strength, excellent

stiffness, a high aspect ratio, a high surface area, and strong interactions with polymeric compounds, providing a synergistic effect that results in improved hardness. The results obtained are supported by the results of studies conducted by other scientists. Shankar and Goriparthi [52] developed epoxy composites containing nickel-titanium alloys and tested their mechanical properties. The authors reported that up to 4 % NiTi reinforcement improves hardness properties.

3.2.4. Impact energy

Impact strength indicates the ability of the polymer to absorb energy during crack propagation and rapid impact loading in the matrix. The impact energy of all FC/NiTi-reinforced groups is higher than the impact energy of pure epoxy and pure NiTi. The maximum impact strength (60 kJ/m²) was observed in group C, and there was a 114.3 % increase in impact energy in the group at the 5 % FC and 3 % NiTi loading. There is a 92.9 % increase in impact energy values in group B and 46.4 % in group A, compared to pure epoxy. There is a 2.5 % increase in impact energy values in group A, 35 % in group B, and 50 % in group C compared to pure NiTi. It is apparent that the addition of FC/NiTi leads to better impact energy compared with that of the pure epoxy composites, indicating the positive impacts of the reinforcement due to the energy absorption between the fibrillated cellulose and the NiTi and its synergistic effect. The impact strength of smart composites increased with the addition of FC. Increasing the NFC load leads to an increasing change in impact strength as a result of the small size of the NFC and its higher surface area [53]. NiTi has energy absorption under impact or some other high strain rate. Since the composite was subjected to an impact, rapid crack propagation started. When the crack propagation reached the FC and NiTi in the composite, the FC and NiTi absorbed the impact energy and stopped the crack propagation due to strong interfacial interactions between the FC, NiTi, and the epoxy matrix. The absorption of impact energy by both FC and NiTi resulted in a synergistic effect with higher impact energy compared to pure epoxy. The seen decreased impact energy of pure epoxy justifies its brittleness [54]. The results obtained are supported by the results of studies conducted by other scientists. Roszowska-Jarosz et al. [39] investigated the mechanical properties of biocomposites produced using nanocellulose as a reinforcement material and epoxy resin as a matrix material. The authors reported that the impact resistance of the nanocomposites was doubled compared to the reinforced epoxy resin, and the highest breaking stress value was obtained at a 1 % nanocellulose loading level, which was also

15 % higher than the unreinforced epoxy resin. It has been reported that even with very low nanocellulose addition, the mechanical properties of nanocomposites are improved, while other studies have reported that much more nanocellulose addition is required. Xu et al. [55] investigated the impact energy of glass fiber-reinforced epoxy composites coated with shape-memory alloy wires by the vacuum-assisted resin infusion method. Authors reported that the impact energy of shape-memory alloy composites increases with temperature. Raghavan et al. [56] investigated the mechanical properties, such as tensile and impact properties, of polymer composites reinforced with superelastic shape-memory alloys. Authors reported that the tensile and impact resistance properties of the composites were improved by the incorporation of shape-memory alloy fibers.

3.2.5. Comparison of mechanical properties

A comparison was made with the results of other similar studies to show the strengths of the study. Comparisons of FC/NiTi-reinforced composites with those of other published works are presented in Table 3.

Due to the unique characteristics of FC and NiTi, which acted synergistically to enhance their mechanical properties, the yield strength, modulus of elasticity, hardness resistance, and impact energy were all increased. As synergistic effects of fibrillated cellulose and NiTi resulted, good stress transfer resulted in increased yield strength, strong work hardening resulted in increased hardness resistance, and energy absorption resulted in increased impact energy. This shows that the mechanical properties of the smart composites produced have been greatly improved, and the values obtained are higher than the values reported to have been obtained in previous studies of similar type.

3.3. Electrochemical corrosion analysis

The corrosion rate is the thickness reduction value in μm that occurs on the corroded material surface in 1 year. Table 4 shows the corrosion rate values of the pure epoxy and FC/NiTi-reinforced composites.

Table 4 indicates that the corrosion rate of all FC/NiTi-reinforced groups is lower than the impact energy of pure epoxy and pure NiTi. The corrosion rate of the smart composites ranged between 0.07 and 0.09 $\mu\text{m}/\text{year}$. Increasing the FC level from 1 % to 5 % with a constant NiTi content of 3 % resulted in a 43.8 % and 56.3 % reduction in corrosion values, respectively. It was determined that FC did not have a negative effect on corrosion. The decrease in corrosion rate is attributable to the presence of NiTi, which has higher corrosion resistance and dominates when evaluated in terms of the synergistic effect of fibrillated cellulose and nickel titanium alloy. The corrosion resistance of NiTi due to its high titanium content is very good [61]. Also, since metals are conductive and the co-atomic intermetallic compound NiTi has exceptional corrosion resistance, the corrosion rate was reduced. The results obtained are supported by the results of studies conducted by other scientists. Zhang et al. [62] investigated the effect on the corrosion behavior of NiTi alloy produced by electron beam melting using different production parameters. The authors reported that a superior corrosion resistance equivalent to that of the wrought alloy was observed when the alloys were evenly distributed. It has been determined by different scientists that high corrosion potential and low corrosion current density are obtained with NiTi [63–66].

Table 3

Comparison of FC/NiTi-reinforced composites with those of published other works.

Published works	Yield strength (MPa)	Modulus of elasticity (GPa)	Hardness (Shore D)	Impact energy (kJ/m^2)	References
Current study	59	7.80	80	60	–
CNF scaffold- reinforced epoxy composites	48	0.60	70	–	[57]
CNF-reinforced epoxy nanocomposites	25	0.90	–	4.29	[58]
CNF-reinforced epoxy-based nanocomposites	42	0.96	–	4.49	[59]
CNC-reinforced epoxy nanocomposites	51	1.90	–	–	[60]

Table 4

Corrosion rate values of pure epoxy, pure NiTi, and FC/NiTi-reinforced smart composites.

Groups	Corrosion rate ($\mu\text{m}/\text{year}$)
Pure Epoxy	0.16
Pure NiTi	0.10
A	0.09
B	0.07
C	0.07

3.4. Shape-memory analysis

Table 5 shows the percentage recovery values of the pure epoxy and FC/NiTi-reinforced composites. Shape-memory values are the percentage of the composite recovering in length or reaching the initial length.

FC/NiTi-reinforced composites were able to return to their original shape to a certain extent after being deformed. The percentages of the composites returning from the final state to the original state after a deformation were obtained as 0 % in pure epoxy, 3 % in pure NiTi, 4 % in group A, 3 % in group B, and 4 % in group C. The shape-memory property shows that FC/NiTi-reinforced groups are smart composites. In terms of synergistic effect, it has been determined that FC has no significant negative impact on shape memory. The ability to shape-memory is due to the presence of NiTi, which has the capacity to memorize its original shape after being deformed into a temporary shape under the application of external stimulus. This shape-memory capability is based on a phase transition. These shape changes occurred between martensite and austenite. The martensite phase is stable at lower temperatures, while the austenite phase is stable at higher temperatures. The formation of these thermomechanical properties resulted from reversible solid-to-solid phase transformations [12]. Heating the deformed composites to a temperature higher than the austenite temperature allowed the inelastic strain to be recovered by transferring the existing martensite to the austenite, providing shape-memory. Basically, as a result of deforming the material in the martensite phase, which is a low-temperature phase, and raising it back to the austenite phase temperature, it was able to remember its shape by returning to the original positions of its atoms.

3.5. Self-healing analysis

Fig. 6 shows that the image on the left shows damage to the composites, while the image on the right shows the self-heal of the damage to the composites after 24 h of the notch or scratch defect.

Table 5

Recovery ability of pure epoxy, pure NiTi, and FC/NiTi-reinforced smart composites.

Groups	Shape-memory (% Recovery)
Pure Epoxy	0
Pure NiTi	3
A	4
B	3
C	4

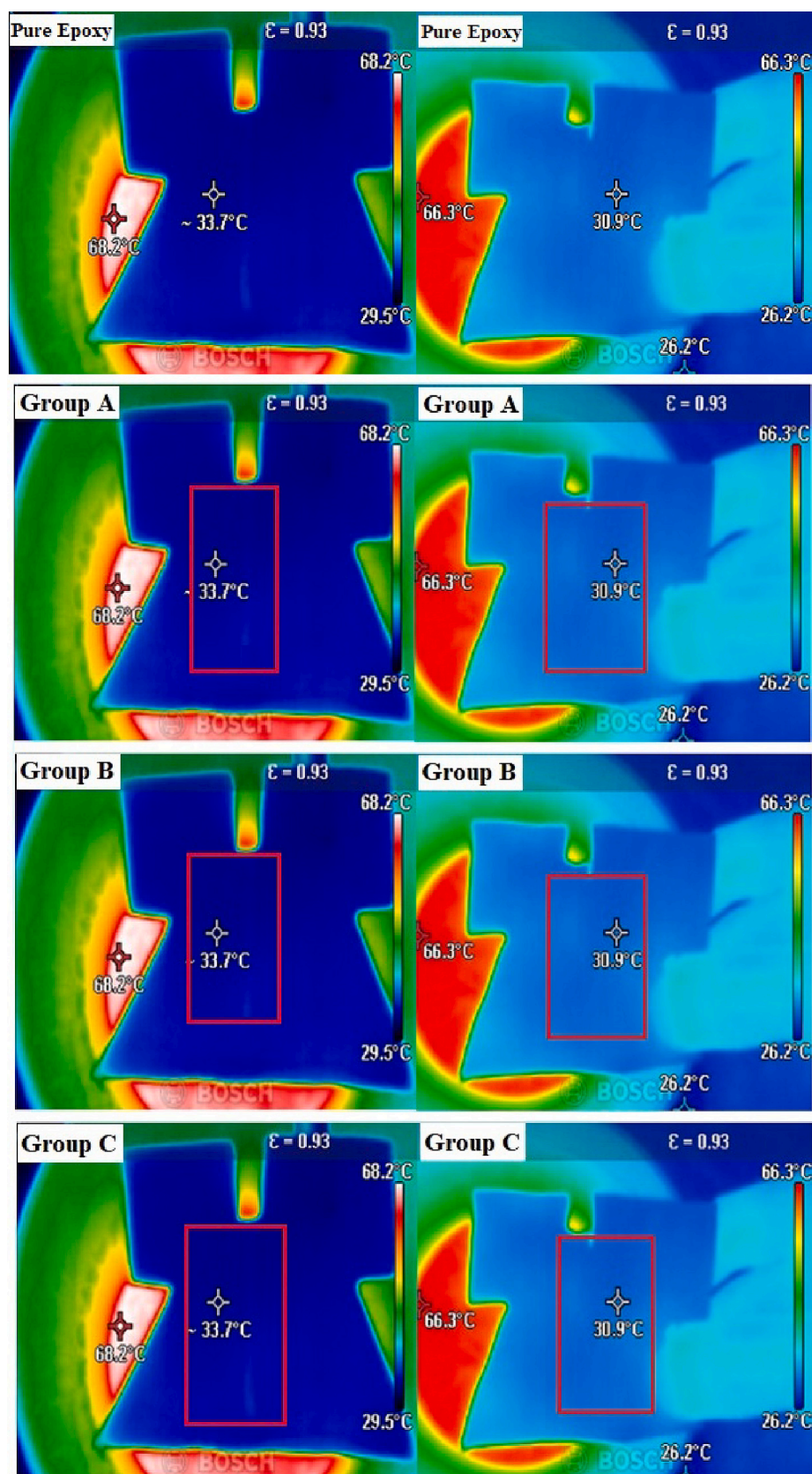


Fig. 6. Damage to the composite material (left) and the self-healing of the damaged composite material after 24 h (right).

Fig. 6 showed that pure epoxy and FC/NiTi-reinforced groups have self-healing abilities for superficial cracks. It has been reported that non-reinforced epoxies have an inherent healing capacity [67]. The self-healing mechanism of unreinforced epoxies has been fully attributed to physical phenomena. The effect is attributed to a strongly crosslinked network that hinders the molecular motion of polymer chains in completely cured, unreinforced epoxies [68]. Self-healing mechanism within the scope of the study; as a result of the damage caused by crack formation in the first stage, phase transformation occurred when the damage came to NiTi powder. In the second stage, during the phase transformation from austenite to martensite, it absorbed the energy, and the crack stopped. In this way, the cracked area was closed, and the surface integrity of the smart composite material was maintained. In general, the closure of the scratch on the composite material can be attributed to the presence of NiTi and the austenite-martensite phase transformation. Although unreinforced epoxy resins have a natural healing capacity, they are inherently brittle and have low-strength properties. NiTi provided the composites with self-healing ability, and fibrillated cellulose increased their mechanical properties. FC/NiTi-reinforced composites are both self-healing and resistant to mechanical properties due to their synergistic effect, which makes them different. This is verified by data from Table 2 and SEM micrographs.

4. Conclusion

FC/NiTi-reinforced smart composites were successfully manufactured using the casting method. Mechanical characterizations such as yield strength, modulus of elasticity, hardness, and impact energy, as well as electrochemical corrosion, shape-memory, and self-healing analyses, were presented in this work. The obtained results explicitly manifest that the synergistic effects of FC filler and NiTi alloy remarkably increased the mechanical properties, decreased corrosion rate, had shape-memory, and had self-healing of all smart composites compared to pure epoxy and pure NiTi composites. These effects can be attributed to the better reinforcing effects of the hard FC filler, the superelastic and shape-memory properties of NiTi, and their interaction to form strong interfacial interactions in the epoxy matrix. Smart composites can be utilized for advanced material applications due to their high performance properties. Smart composites are lightweight and impact-resistant, which may enable them to be used in interior and structural parts in the automotive industry. Lighter interior components can help reduce vehicle weight and CO₂ emissions. The lightweight, shape-memory, and self-healing properties of smart composites make them suitable for use in the aviation industry. In this way, the performance of aircraft can be increased and maintenance costs can be reduced.

The limitations of this study are that there are not enough studies on the topic in the literature, there is no standard or clear method for shape-memory and self-healing analysis, and it is difficult to observe and measure. It is of great importance to accurately analyze shape-memory and self-healing capabilities and develop test methods to investigate shape-memory and self-healing properties in more detail in future studies. In addition, in future studies on this subject, X-ray diffraction (XRD) and energy dispersive X-ray (EDX) analyses of smart composites can be performed. In addition, transmission electron microscopy (TEM) can be used to examine the micro- and nanostructure of the materials in more detail.

CRediT authorship contribution statement

Mert Yildirim: Writing – original draft, Methodology, Investigation, Formal analysis, Conceptualization. **Ilven Mutlu:** Writing – review & editing, Supervision. **Zeki Candan:** Writing – review & editing, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The data that has been used is confidential.

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