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Polystyrene microcapsules containing linseed oil and SiC nanoparticles as a lubricant additive for boosting the self-healing and self-lubricating efficiency of epoxy coatings

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Abstract

Friction coefficients and specific wear rates of a multifunctional smart nanocomposite epoxy coating with embedded linseed oil-filled microcapsules and SiC nanoparticles were determined using six coatings synthesized with 15 wt.% microcapsules and 1, 2, 3, and 4 wt.% of SiC nanoparticles. In rotational pin-on-disk testing, all samples containing microcapsules and SiC nanoparticles exhibited reduced friction coefficients. The sample containing 3 wt.% SiC nanoparticles showed the highest decrease in coefficient of friction by 31.79% and a specific wear rate by 59.04% compared with the neat epoxy sample. The results are discussed concerning lubricating rollers, polishing operations of SiC nanoparticles, a solid/liquid lubricant film, and electron microscopy (FESEM/EDX) studies.

Keywords: Smart nanocomposite coating; Self-lubricating; Microcapsules; SiC nanoparticle

1. Introduction

Epoxy coatings are widely used due to their superior adhesion to the substrate and chemical resistance in wet areas such as docks, water tanks, pipelines, and cooling towers [1]. Due to epoxy's cross-linked structure, stress, impact, and wear induce micro-cracks that spread

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inside its structure, resulting in a rupture in the form of debris [2]. Adjusting the glass transition temperature (T_g) and curing temperatures can increase epoxy strength, but the gain is not substantial [3,4]. Depending on the coating's function in service, specific strategies may be used to mitigate epoxy's drawbacks, such as embedding microcapsules into the epoxy matrix and developing a self-healing smart coating [5]. These coatings have been studied extensively recently and offer new application capabilities [6–8]. Protection against corrosion is one of their functions [9].

Depending on the encapsulated core, microcapsules perform different functions in these systems. Boumezgane et al. inserted microcapsules with a PMMA shell and ionic polydimethylsiloxane oligomers core into epoxy to create a self-healing coating and improve steel corrosion resistance. The resulting coating protects the steel from corrosion [10]. Song et al. developed an epoxy-based self-healing system that repairs cracks by discoloration [11]. Schmark et al. found that an epoxy coating sample containing 15% polydimethylsiloxane microcapsules (M-PDMS-a) and 15% polydimethylsiloxane triethylenetetramine microcapsules (PDMS-a/m-TETA) had the highest corrosion resistance [12].

One of the newest ways to choose an encapsulating agent is to develop another property along with corrosion resistance. Creating wear resistance is one of these innovations. Encapsulating polymerizable organic oils in the air, such as tung and linseed oils, makes self-lubricating and self-healing coatings [13]. Zhang et al. inserted dibutyl phthalate and linseed oil microcapsules with urea and formaldehyde shells in epoxy to create self-lubricating and self-healing coatings. The synthetic coating self-heals by 96 vol.% and reduces friction by 92% [14]. Li et al. incorporated various quantities of microcapsules with polyurea-formaldehyde shells and tung oil cores into an epoxy matrix. The wear rate was reduced by 78.6% in coatings containing 10 wt.% microcapsules [15]. Sun et al. encapsulated liquid 4,4′-bis-methylene

cyclohexane diisocyanate (HMDI) in two-layered poly urea and poly urea-formaldehyde (PUF) shells. Microcapsules in an epoxy matrix prevented substrate corrosion. The friction coefficient was also reduced by 78.9% [16].

Most research on self-healing epoxy coatings with microcapsules has focused on corrosion resistance. Microcapsules are designed to have poor mechanical properties [17]. They degrade the coating's mechanical properties. The three essential coating features are desired mechanical properties, corrosion resistance, and wear resistance [18]. Depending on the service conditions, one of these three coating qualities, such as corrosion resistance, may be more critical, but upgrading all three is necessary under challenging service conditions. Each coating must have the qualities required to maintain a high level of durability in service. Hence, it is critical to improve mechanical properties, wear resistance, and corrosion resistance concurrently [6,7,19].

Adding a second agent to the self-healing coating and creating a smart composite coating can improve these three features simultaneously and synergistically. This second agent is usually made up of nanoparticles. They have been studied for their unique properties as a single and secondary additive to epoxy coatings. A literature review suggests that adding SiC nanoparticles to epoxy coatings alone can improve their thermal, mechanical, anti-wear, and anti-corrosion properties. Rodgers et al. found that the infusion of SiC nanoparticles into SC-15 epoxy resin improved thermal and mechanical properties [20]. Cong et al. studied the fabrication of nano-ceramics modified epoxy coatings. They found that the dispersion of ceramic particles in epoxy resin improved the coatings' wear and corrosion resistance [21]. Zhou et al. compared epoxy composites filled with nano-SiC particles and micro-SiC particles and found that the nanocomposites exhibited better mechanical and electrical properties [22]. Dai et al. investigated the effect of Si-based compound nanoparticles on the anticorrosive properties of epoxy coatings. They found that SiC-modified epoxy exhibited the lowest

corrosion current density and the highest coating resistance [23]. Vinodhini et al. found that a nanocomposite coating with 2 wt.% SiC nanoparticles showed excellent corrosion protection efficiency and decreased corrosion current density [24].

Therefore, it can be concluded that microcapsules and nanoparticles enhance neat epoxy coatings' corrosion resistance, mechanical properties, and wear resistance, increasing their longevity [25–27]. Researchers studied the impact of adding microcapsules and nanoparticles to epoxy coatings. Ghorbani et al. add PMMA microcapsules containing linseed oil and TiO2 nanoparticles to epoxy coating. Coatings containing 15 wt.% microcapsules and 12-16 wt.% TiO2 nanoparticles were completely corrosion-resistant. Hardness and wear rate of coatings with 15% wt.% microcapsules and 12% wt.% TiO2 nanoparticles increased 11.8% and 88.7%, respectively [28]. Sharma et al. evaluated infusing epoxy with urea-formaldehyde (UF) microcapsules containing tung oil and nano-clay. Adding these two agents to epoxy retards corrosion in epoxy-coated rebar [29]. Tao et al. added PMMA microcapsules containing lactone, epoxy monomer, phenyl acetate, and silica nanoparticles to a waterborne epoxy matrix coating. These three types of microcapsules perform self-healing and self-reporting functions. These two agents at 15 wt.% improved the coating's corrosion resistance [30].

Consequently, exploring an effective combination of microcapsules and nanoparticles is a worthwhile subject of study. Coatings made with this combination have better final properties than coatings made with microcapsules and nanoparticles individually. This work integrates self-healing linseed oil microcapsules and silicon carbide nanoparticles (SiC NPs) into an epoxy matrix to fabricate a multifunctional smart coating. Utilizing polystyrene as the microcapsule shell material presents certain advantages, such as its ability to form thin, porous morphologies with high thermal stability. Additionally, the permeability of the polystyrene shell can be tuned for the desired application by setting the cor-shell ratio. Also, SiC (a well-

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known abrasive) at low nanoparticle loadings can act as a solid lubricant due to the formation of a tribo-film on the contacting surfaces. This tribo-film is able to mitigate direct contact between high asperities of the sliding surfaces, thus reducing friction and wear [31–34].

The microcapsule and nanoparticle characteristics are investigated along with the resulting changes in the nanocomposite coating's hardness, corrosion resistance, and wear resistance. This pioneering dual-functionality coating could provide a new solution to enhance the longevity of epoxy-coated surfaces across numerous demanding applications.

The novelty of this study can be summarized as follows: (i) This is the first study that developed microcapsules with a linseed oil core and polystyrene shell. (ii) The possibility of using synthesized microcapsules in combination with SiC NPs in the epoxy coating will be investigated. (iii) Developing a new smart nanocomposite coating with various reliable applications. (iv) It evaluates synthesized coatings' mechanical, corrosive, and wear characteristics to provide a comprehensive understanding of their anticipated capabilities. (v) The systematic design of coatings with increasing SiC NPs weight allowed the most accurate interpretation of changes. The information gathered may aid in improving the quality of epoxy coatings and their application in the industry.

2. Materials and experimental procedures

2.1. Materials

Diglycidyl ether bisphenol A epoxy resin (DGEBA) and its special hardener, aliphatic epoxide D.E.R. 732 (epoxy resin-to-hardener mixing ratio of 100:38), were supplied by (Sigma Aldrich Chemical Industries) in the form of a colorless liquid with a molecular weight of 340.41 for making coatings. The combined density of epoxy resin and hardener is 1.11 g/cm^3. The microcapsule additive with polystyrene (average molecular weight about 35,000 g/mol in

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the form of beads) as a microcapsule shell and linseed oil as a microcapsule core were supplied by (Sigma Aldrich Chemical Industries). Dichloromethane (DCM) with a purity of greater than 99.5 wt.% as a solvent for the microcapsule shell and core, sodium dodecyl sulphate (SDS) with a molecular weight of 288.37 g/mol, and carboxymethyl cellulose (CMC) with a viscosity of 400–800 cps in 2% water at 25 °C, as aqueous mediums (surfactants) for microcapsule synthesis, also, were supplied by (Sigma Aldrich Chemical Industries). Beta SiC NPs with a particle size of 45-65 nm, purity above 99%, and a spherical morphology were provided by (US Research Nanomaterials, Inc.) as nanoparticle additives to coatings. DIN Ck 10 sheets (40×30×2 mm) and DIN Ck 45 rods (Ø10×50 mm) were used as steel substrates for coating. In all experiments, deionized water was utilized. All materials were used in their original form without any further processing.

2.2. The method of making samples

2.2.1. Aqueous solutions preparation

The aqueous solution for microcapsule synthesis was prepared by dissolving CMC and SDS separately in deionized water at 1 wt.% and 1.5 wt.%, respectively, using a magnetic stirrer (PIT300, Pole Ideal Tajhiz, Iran). Water was heated to 60 °C to facilitate and accelerate the dissolution of these compounds. After dissolving the substances and producing a clear solution, the final surfactant was prepared by combining the two solutions (in a one-to-one ratio) and cooling to ambient temperature.

2.2.2. Synthesis of microcapsules

Microcapsules with polystyrene shells containing linseed oil were prepared using the solvent evaporation method. 2.0 g linseed oil and 1.0 g polystyrene (core-to-shell ratio of 2-to-

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1) were dissolved in 30 ml of dichloromethane. The solution was then added dropwise by syringe (1 mm I.D. needle) to 300 ml of combined surfactant, stirring at a rate of 1200 rpm using a three-blade mechanical stirrer (AT-MD20, FALC, Italy). An external thermometer checked the emulsion's temperature. After reacting for 2.5 h at 40 °C, DCM was entirely evaporated. The microcapsules were gathered by filtering using Whatman Ashless 41 filter paper and were washed multiple times with distilled water before being dried for several days at room temperature. Before incorporation into the coating, the microcapsules were sieved to exclude the potential of using sticky microcapsules.

2.2.3. Preparation of coating

This study developed six coatings, including a control epoxy coating with no additives, a smart epoxy coating with 15 wt.% microcapsules, and four smart nanocomposite epoxy coatings with 15 wt.% microcapsules and SiC NPs at concentrations of 1, 2, 3, and 4 wt.%, respectively. A 15% microcapsule loading was selected based on the previous study [28] that provided sufficient self-healing ability without compromising the coating's mechanical integrity. Higher microcapsule content can make the coating more brittle. On the other hand, 1-4% SiC NPs were chosen to impart solid lubricant effects across a range of low nanoparticle loadings. Higher SiC contents may adversely aggregate in the coating, so a modest nanoparticle variation was investigated.

To prepare coatings, SiC NPs were added to epoxy resin at 1, 2, 3, and 4 wt.% of epoxy resin & hardener weight and stirred for 30 min at 1000 rpm using a 3-blade mechanical stirrer. The mixture was then mixed for 15 min at 100% power with an ultrasonic homogenizer mixer (UP200H, Hielscher, Germany) in an ice-water bath. Since ultrasonic waves heat the mixture, decreasing the viscosity of the epoxy resin facilitates the integration of SiC NPs. Epoxy resin

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and SiC NPs were mixed with mechanical and ultrasonic mixers three times. Then, the microcapsules (15 wt.% of epoxy resin & hardener weight) were manually mixed into the epoxy resin and SiC NPs combination using a metal spatula for about 20 min. The mixture was then placed in a vacuum chamber for 2 h to remove air bubbles. Eliminating trapped air is essential for coatings containing nanoparticles since the coating's viscosity rises substantially as the nanoparticles' weight percentage increases. So, if not done correctly, it would reduce the coating's qualities. Finally, a desired quantity of hardener (with a 100-to-38 mixing ratio by weight, epoxy-to-hardener) was added and manually mixed for 10 min.

For epoxy coating containing only 15 wt.% microcapsules, SiC NPs mixing was abolished. Neat epoxy coating as a control sample was prepared by adding a hardener to the epoxy without adding microcapsules and SiC NPs. The composition of coatings with their code has been listed in Table 1. M refers to the microcapsule, and S refers to SiC NPs.

Table 1. Composition of coating samples

Sample	Microcapsule (wt.%)	SiC NPs (wt.%)
0M0S	0	0
15M0S	15	0
15M1S	15	1
15M2S	15	2
15M3S	15	3
15M4S	15	4

2.2.4. Coating the substrates

Sheets and rods were used as substrates for coatings in self-healing and self-lubricating evaluations. Before coating, the substrates' surfaces were meticulously cleaned using P80

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sandpaper to remove grease, dirt, and surface oxides, leaving only straight parallel lines. After rinsing with acetone, they were dried using a laboratory heater. The steel substrates were then placed in a Teflon mold and coated with the prepared coatings using an applicator (SZQ 3679, Moderner, China) to a wet thickness of 80 μm. After 24 h exposure at room temperature, the coatings were put in an oven heated to 80 °C for 2 h, followed by another 72 h exposure at room temperature. When fully cured, most epoxy systems shrink by around 5 vol.% [35]. As a result, the coatings' dry thickness will not be less than 75 μm.

2.3. Characterizations

2.3.1. Characterizations of additives

The primary instrument utilized to investigate the essential features needed for microcapsules was the optical microscope (OM). The shape and general morphology of the microcapsules were determined using OM (IM7200, MEIJI TECHNO, Japan), which was equipped with a Canon EOS 1100D digital camera. The microcapsules were placed on a microscope slide, and by adding a few drops of deionized water to them and putting a mirror under them, they were photographed at ×100 magnification. All observations were done at an ambient temperature.

The microcapsules' surface morphology and shell thickness were seen and measured using a field emission scanning electron microscope (FESEM) (MIRA3, TESCAN, Czech), which operated in the secondary electron mode at a voltage of 15.0 kV. Microcapsules were placed on sticky tape and burst with a mortar to determine the shell thickness. Additionally, this test was utilized to examine the scratch surface after the corrosion test and the wear surface after the pin-on-disk test. Sputter-coated gold created a conductive surface for FESEM observations. The SiC NPs' distribution was analyzed using energy dispersion spectroscopy (EDS) installed

on the FESEM. SiC NPs contain silicon; therefore, a quantitative map study of silicon in the epoxy allowed for identifying the spreading of SiC NPs in the matrix.

The microcapsules mean particle size and distribution were determined using a laser particle size analyzer (ANALYSETTE 22, FRITSCH, Germany). 0.05 g of microcapsule powder was stirred using a magnetic stirrer for 10 minutes in 150 ml of deionized water to prepare the test sample. Each sample was tested three times to ensure accuracy, and the mean and standard deviation were reported.

Weighing the raw materials and synthesized microcapsules provides essential information about the microcapsules' characteristics, such as the microencapsulation process's efficiency, the oil content of the microcapsules, and oil encapsulation's efficiency. The literature provides the formula for determining these characteristics [36–38]. The efficiency of the microencapsulation process was calculated by dividing the weight of the synthesized microcapsules by the weight of the initial raw materials used. The oil content of these microcapsules was found using the solvent extraction method, which measures the weight ratio of the oil within the microcapsules. This involved crushing a known weight of microcapsules and using ultrasonic mixing to separate and dissolve the oil in methanol, followed by filtration and drying to ascertain the weight of the residual shell. The weight of the oil was then calculated by subtracting the weight of the shell from the total microcapsule weight. The efficiency of oil encapsulation was established by comparing the weight of the oil extracted to the initial oil weight used in the formulation. Results of these measurements were taken across three independent samples for accuracy and are presented as mean values with their standard deviations.

Fourier Transform Infrared Spectroscopy (FTIR) analyses (Tensor 27, Bruker, Germany) were carried out to characterize the chemical structure of the microcapsules, polystyrene, and

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linseed oil. Microcapsules were carefully placed on an attenuated total reflectance (ATR) crystal, and spectra were collected over a range of 4000 to 400 cm^{-1} with a resolution of 4 cm^{-1}. Each spectrum represents an accumulation of 64 scans to ensure adequate signal-to-noise ratios. The characteristic peaks corresponding to polystyrene and linseed oil chemical groups were identified and analyzed.

The thermal stability of the microcapsules, polystyrene, and linseed oil were evaluated using Thermogravimetric Analysis (TGA) (TG 209F3, NETZSCH, Germany). Approximately 10 mg of microcapsules were subjected to a controlled temperature program from 25 °C to 600 °C at a heating rate of 10 °C/min under a nitrogen atmosphere. The weight loss of the capsules was continuously measured as a function of temperature to determine the thermal degradation profile of the polystyrene shell and the linseed oil core.

The distribution of SiC NPs in the epoxy matrix and their morphology were investigated using a transmission electron microscope (TECNAI G2 F20, FEI, USA). Observations were performed in TEM and selected area electron diffraction (SAED) modes. An energy dispersive spectroscopy (EDS) system installed on a TEM was used to evaluate the purity of SiC NPs. The powder of SiC NPs was dispersed in deionized water and then placed on a copper grid with an amorphous carbon substrate.

2.3.2. Characterizations of coatings

The Shore D scale hardness of the coatings was determined in accordance with ASTM D2240 (LD0559, TQC, Germany). Hardness was tested 15 times for each coating, and the mean and standard deviation were reported.

The salt immersion corrosion test determined the coating's corrosion resistance or self-healing capabilities. Cross-shaped grooves were drawn on sheet-shaped specimens using a 0.2

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mm thick surgical razor. Rust can only be seen if the scratches go deep enough to reach the steel substrate. The coatings healed for 72 h. Finally, scratched coatings were submerged in a 10% NaCl solution for 216 h and photographed after 24, 72, 120, 176, and 216 h to demonstrate the corrosive effects. The FESEM examined the healing scratch groove.

Electrochemical Impedance Spectroscopy (EIS) (WizEIS-8100premium, WIZMAC, South Korea) was used to investigate the self-healing functionality of all coatings after 216 h expousing in in a 10 wt.% NaCl aqueous solution. The process involved initially introducing a defined scratch on the epoxy-coated metal specimens to simulate damage. The scratched specimens were then left uninterrupted for 24 h at room temperature to allow the microcapsules' core material to release. Subsequently, the treated specimens were placed in a 10 wt.% NaCl aqueous solution, ensuring that a uniform area of 1 cm × 1 cm was exposed to the electrolyte. EIS measurements were executed using a three-electrode cell configuration with the prepared specimens serving as the working electrode, alongside a saturated calomel electrode (SCE) as the reference electrode and a platinum wire as the counter electrode. Tests were conducted over a frequency range from 100 kHz to 10 mHz, with an excitation signal of 10 mV rms superimposed on the open circuit potential. Post-testing, impedance data were fitted to appropriate equivalent circuit models using ZSimpWin software. Model parameters were iteratively adjusted until the chi-square value was minimized to less than 0.01, ensuring an accurate representation of the coating's electrochemical response post-healing in the saline solution.

The coating's wear behavior was examined using friction and wear test with a pin-on-disc configuration in accordance with the ASTM G99 standard (WTH02, TSN, Iran). The applied force was 7 N, the distance was 1000 m, the linear velocity was 0.15 m/s, and the device rotated at 358 rpm. All tests were conducted in a typical laboratory environment. After testing each

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sample, the instantaneous frictional force at each distance, weight reduction, and the related diagrams were acquired. Each coating's wear test was repeated three times to ensure accuracy, and the mean and standard deviation summarized the results. Prior to the commencement of testing, the tribometer was subjected to meticulous calibration procedures to ensure its ability to detect subtle fluctuations in frictional force. The manufacturer's specifications confirm that the resolution of the tribometer is sufficiently high to guarantee the accuracy of the measurements. FESEM determined the coating's surface morphology.

The instantaneous coefficient of friction (COF) values at each distance were calculated by dividing the instantaneous frictional force by the force of normal (7 N). The mean dynamic COF was calculated using instantaneous COF values at each distance, and the specific wear rate (SWR) was calculated using equation (1) [39] in terms of 10^{-13} m^3/N.m. The SWR (K_w) is force-dependent and more accurate than other wear relationships.

$$K_w = \Delta m/\rho.F_N.L \quad (1)$$

Where Δm (g) is the weight loss value of each sample, ρ (g/mm^3) is the density of each coating, F_N (N) is the force of normal (= 7 N), and L (m) is the distance traveled (= 1000 m). The density of each coating was calculated based on the density of epoxy (1.11 g/mm^3), microcapsule density (~linseed oil, 0.93 g/mm^3), and SiC NPs density (3.22 g/mm^3), and their weight percentage in the coating.

3. Results and discussion

3.1. Shape and morphology of the microcapsules

Fig. 1 shows the synthesized microcapsules. The absence of agglomeration and connections between the microcapsules is apparent. Also, air trapping in the microcapsules and free oil was not observed. Lack of free oil means optimal oil-shell surroundings. Synthesized

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microcapsules are spherical and scraggly, with a clear border between them. Their size range is predicted to be 10–30 µm based on OM visual observations but will be determined using a particle size analyzer.

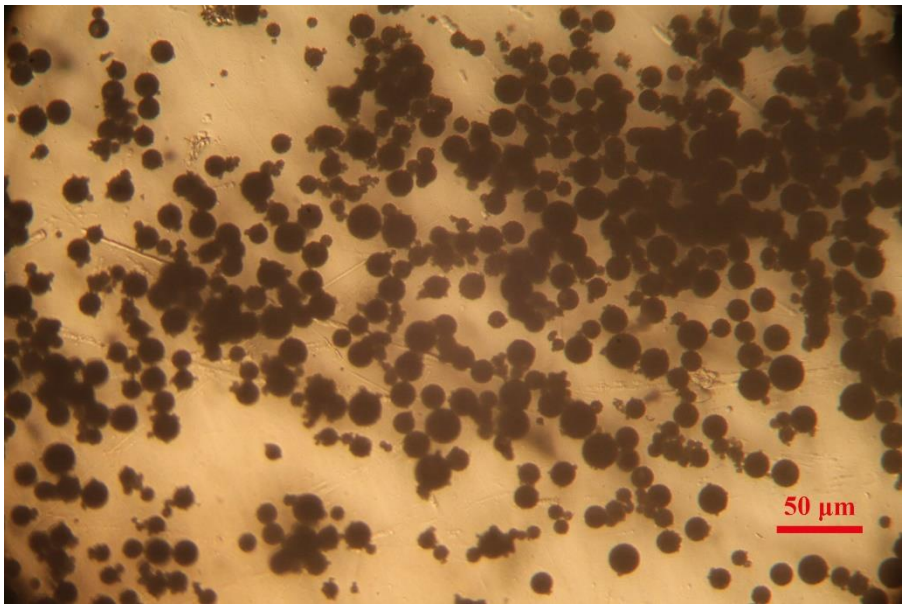

Fig. 1. OM image of the microcapsules.

3.2. Surface morphology and the microcapsule's shell thickness

FESEM micrographs in Fig. 2 show the microcapsules' appearance, shell thickness, and roughness. Microcapsules are spherical and monolithic, with negligible inter-capsular stickiness. The 1-4 µm appropriate thickness of the microcapsule shells has prevented the microcapsules' surface from exhibiting indentations. The microcapsules' outer surface is rough and porous. Microcapsules with high surface roughness, like the synthesized microcapsules, provide exceptional adhesion to the epoxy substrate, and this excellent adhesion also facilitates the release of the healing agent [40,41]. If the adhesion of microcapsules to the matrix weakens, cracks or stress propagate in the matrix without rupturing the microcapsules [37]. But if their

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adhesion to the matrix is favorable, a crack or stress may rupture the microcapsules and release their contents into the matrix.

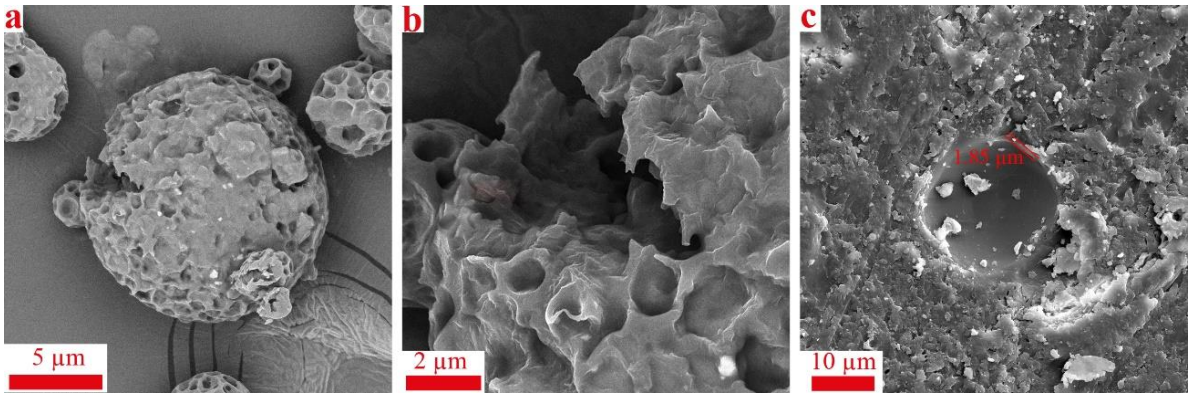

Fig. 2. FESEM micrographs of the microcapsules: (a) appearance and shape, (b) surface roughness and shell thickness, (c) shell thickness.

3.3. Particle size and size distribution of the microcapsules

Microcapsule sizes affect the amount of repair content delivered to the cracked surface. Fig. 3 shows the microcapsules' particle size distribution and mean particle size. The microcapsule size distribution curve showed a 95% Gaussian fit. The particle size distribution device needs unagglomerated particles to measure microcapsule size correctly. If particles agglomerate, the device treats them as one, and thus, the particle size curve will not be Gaussian. The diameter range and mean microcapsule diameter were 6-26 µm and 15.3 µm, respectively. Microcapsule diameter matches OM's visual observations.

Turbulent fluid flow around mechanical stirrer blades causes variable microcapsule sizes and particle diameter instability. Smaller microvortices exert less shear stress on developing microcapsules near the blades, increasing their diameter. Bigger microvortices in zones farthest from the blades impose more shear stress on developing microcapsules, reducing their size [42].

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Larger synthesized microcapsules often contain more repair agents, which improves healing, and they break faster and more efficiently than smaller ones under external force [43]. When a microcapsule with a diameter of 24 µm is broken apart, for instance, the contents are equivalent to 216 microcapsules with a diameter of 4 µm. The less healing agent is released when an external force ruptures fewer of these 216 microcapsules. On the other hand, the disadvantage of the large microcapsule is that it is easily broken, which weakens the coating because it leaves a large hole and degrades its properties. Therefore, obtaining a variety of sizes is preferable.

It is preferred that the diameter of the microcapsules not exceed one-third of the coating thickness [37,44]. Since the coating thickness is 75 µm, it is desirable to place microcapsules with a diameter of less than 25 µm; consequently, the diameter range of 6-26 µm is quite suitable.

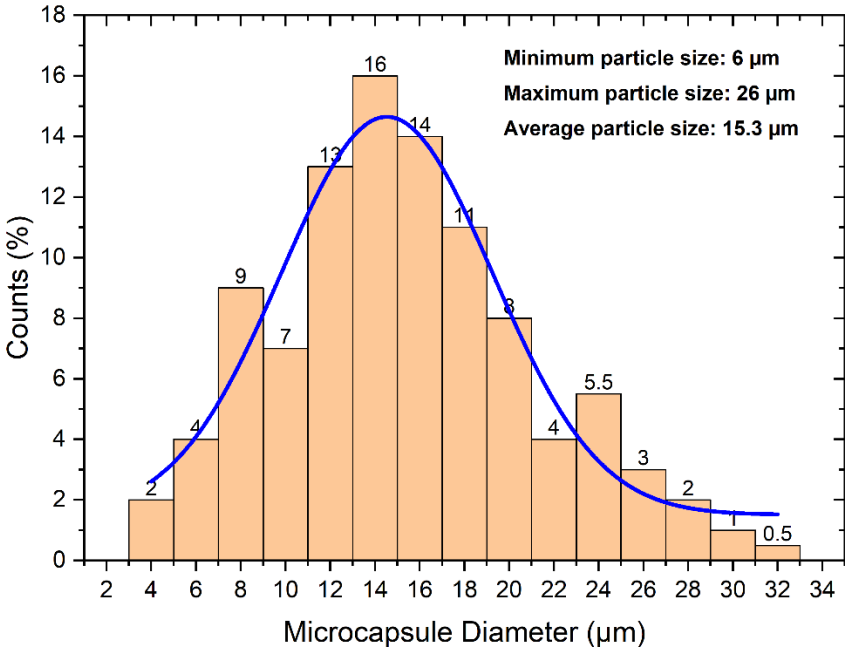

Fig. 3. The microcapsules' particle size distribution curve.

3.4. The characteristics of the microcapsules

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The microcapsules with the desired characteristics guarantee the anticipated features of self-healing coatings. The microencapsulation process's efficiency, the oil content of the microcapsules, and oil encapsulation's efficiency were 72.64 ± 2.58%, 72.79 ± 2.85%, and 73.81 ± 1.43%, respectively. These values are higher than 70%, suggesting that the synthesized microcapsules have acceptable qualities for self-healing coatings. Indeed, values greater than 70% indicate that raw materials have been mainly converted into microcapsules and that more microcapsules are synthesized; furthermore, a large volume of the microcapsule content is filled with oil, and the shell surrounds the primary oil during microencapsulation [45]. The increased linseed oil in polystyrene microcapsules is crucial for this self-healing coating. This helps coatings become more self-heal and self-lubricated and delivers more repair agents.

3.5. The chemical structure of the microcapsules

The FTIR spectral analysis provides robust evidence for the successful encapsulation of linseed oil within a polystyrene shell. The distinct peaks observed in the FTIR spectra of the microcapsules, linseed oil, and polystyrene polymer (Fig. 4) confirm the presence of both core and shell materials. Polystyrene shows peaks at 3063 and 3030 cm^{-1}, which are characteristic of the aromatic C-H stretching vibration absorption. Additionally, the aliphatic C-H stretching vibrations at 2921 and 2846 cm^{-1} originate from the methylene groups in the polystyrene structure. Further validation of the polystyrene shell is evidenced by the aromatic C=C stretching vibrations at 1601, 1498, and 1457 cm^{-1}, along with the C-H bending peaks at 756 and 698 cm^{-1}.

The linseed oil core is confirmed by observing several critical functional group vibrations. The peak at 3029 cm^{-1} is attributed to the =C-H cis-double bond stretching, indicating the unsaturation of the fatty acid chains. The methylene and methyl C-H bending signals at 2942

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and 2869 cm^{-1} demonstrate the presence of aliphatic groups in the fatty acid structure. Additionally, the intense carbonyl C=O stretch at 1750 cm^{-1} and C-O stretch at 1175 cm^{-1} correspond to the ester linkages of the triglyceride molecules. Finally, the peak at 733 cm^{-1} results from the long methylene hydrocarbon chains comprising the fatty acids.

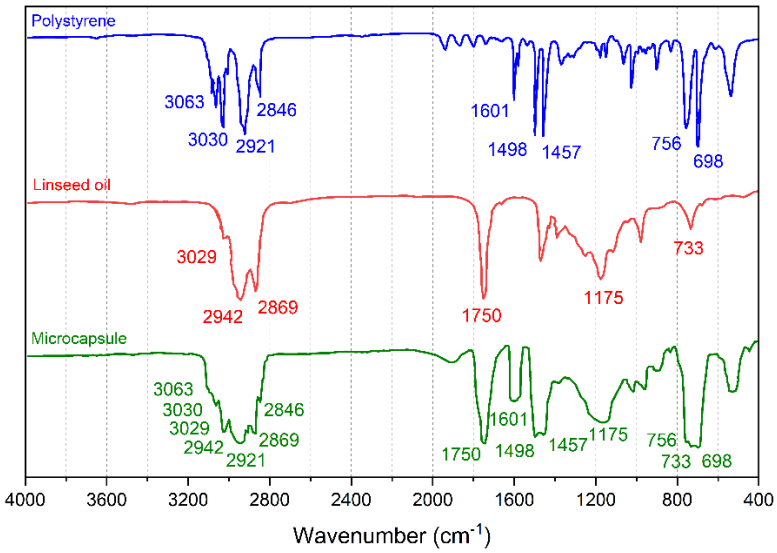

Fig. 4. FTIR spectra of the polystyrene, linseed oil, and microcapsules.

The FTIR spectrum of the microcapsules contains distinct peaks at 3063, 3030, 3029, 2942, 2921, 2869, 2846, 1750, 1601, 1498, 1457, 1175, 756, 733, and 698 cm^{-1} (Fig. 4). The combination of characteristic linseed oil and polystyrene peaks provides clear evidence for the successful encapsulation of linseed oil within a polystyrene shell. The unique spectral fingerprints of the core and shell materials are retained within the microcapsule spectrum, while peaks corresponding to potential contaminants or byproducts are conspicuously absent. This suggests a clean encapsulation process yielding the expected chemical composition. The 2800-3200 cm^{-1} region displays overlapping C-H stretching signals from the aliphatic groups in linseed oil and polystyrene. The persistence of these methylene and methyl vibrations indicates

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the core oil is thoroughly encapsulated within the polymer shell matrix. Notably, the ester carbonyl stretch at 1750 cm^{-1} retains strong intensity, implying the unsaturated fatty acid structures of the linseed oil remain primarily intact despite encapsulation. This demonstrates the structural integrity of the core oil is preserved. Furthermore, intense aromatic peaks at approximately 1600-1450 cm^{-1} and the out-of-plane C-H bending vibrations at 756 and 698 cm^{-1} confirm the presence of the polystyrene shell. The clarity of these signals corresponds to the aromatic nature of the polystyrene polymer, verifying shell formation. Finally, the absence of carbonyl stretches in the 1750-1735 cm^{-1} region suggests negligible oxidation of the linseed oil during encapsulation. This indicates the polystyrene shell provides an effective barrier, protecting the core oil from atmospheric exposure.

It is worth mentioning that additional stability testing was performed by storing the microcapsules for extended time periods and periodically checking for signs of oil oxidation. After storage for up to 6 months, FTIR analysis continued to show no detectable carbonyl peaks in the 1750-1735 cm^{-1} region. Furthermore, the oil release efficiency after storage remained unchanged compared to freshly prepared capsules.

These FTIR results align with the structure of the designed microcapsules and provide chemical evidence that the core-shell morphology was successfully achieved. The results indicate that the microencapsulation by solvent evaporation method did not notably interfere with the chemical structure of the linseed oil, which is vital for maintaining its quality and functionalities. The polystyrene shell was proven to be a suitable encapsulant material, as reflected by the distinct spectral signatures in the FTIR spectrum.

3.6. The thermal stability of the microcapsules

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TGA provides critical insights into materials' thermal stability and decomposition behavior, particularly when assessing the performance characteristics of novel microcapsule formulations. In the context of the polystyrene microcapsules containing linseed oil, the TGA test was conducted to evaluate their thermal decomposition behavior, a defining aspect of their applicability as a lubricant additive in epoxy coatings. The TGA curves in Fig. 5 reveal three distinct weight loss profiles for linseed oil, polystyrene, and the microcapsules.

The linseed oil displays an initial weight loss beginning at approximately 300 °C, with a gradual decline continuing until ~430 °C when 100% degradation occurs. This protracted decomposition is attributed to the stepwise scission of fatty acid chains as temperature increases. In contrast, the polystyrene polymer exhibits enhanced thermal stability, with the onset of degradation at 320 °C followed by rapid breakdown approaching 0 wt.% above 450 °C. This abrupt weight loss is characteristic of thermoplastic polymers and highlights the better thermal stability of polystyrene versus linseed oil. Notably, the initial polystyrene decomposition temperature of 320 °C is substantially higher than conventional shell materials like poly (melamine-formaldehyde) (260 °C), polyurethane-formaldehyde (240 °C), and polyurea (200 °C). This enables the potential application of the polystyrene microcapsules at elevated working temperatures.

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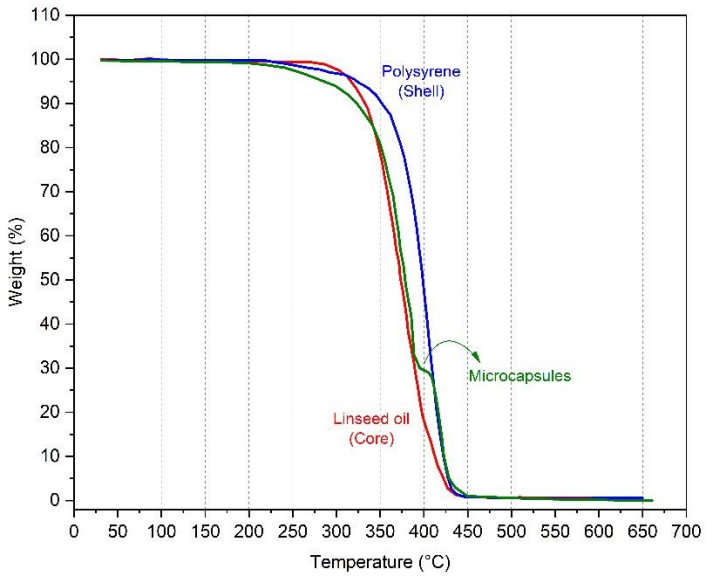

Fig. 5. TGA curves of the polystyrene, linseed oil, and microcapsules.

The thermal decomposition profile of the microcapsules exhibits a complex, multi-stage behavior. Initial minor weight loss below 300 °C can be attributed to residual solvent or moisture evaporation. A significant degradation event occurs near 300 °C as temperature increases, consistent with the onset of linseed oil core breakdown. The mass stabilizes around 35% remaining, denoting evaporation of the lower thermal stability oil core. At higher temperatures above 380 °C, the microcapsule shell degrades completely, seen as a reduction in molecular weight and associated mass loss approaching 0 wt.%. This is expected as the microcapsules are comprised entirely of organic components. The rapid decline in mass aligns with the ultimate breakdown of the polystyrene shell and complete volatilization of the linseed oil core. The distinct mass loss events correlate well with the individual TGA profiles of the linseed oil core and polystyrene shell materials. The microcapsule thermal analysis confirms the core-shell structure, with sequential degradation processes linked to the less thermally stable oil core and subsequent shell polymer breakdown at higher temperatures.

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While the distinct mass loss events in the microcapsule TGA profile confirm the core-shell structure, further examination of the individual degradation temperatures provides additional insights. The onset of linseed oil evaporation after 300 °C warrants further investigation into potential premature core release at operational temperatures, impacting epoxy coatings' self-healing and lubrication functionality. Although the higher thermal stability polystyrene shell protects the core up to 300 °C, this threshold invites the necessity of evaluating microcapsule performance at temperatures approaching this limit. Quantitatively, the rapid mass loss event centered at 350 °C corresponds to the thermal decomposition of the polystyrene shell polymer. This defines an upper bound on the thermal robustness of the microcapsule shell matrix. However, the lower core oil degradation temperature, while sufficiently high for many applications, could restrict the maximum operating temperature range. The demonstrated microcapsule stability up to 300 °C suggests suitability for environments requiring resistance up to this limit. Further work should aim to elucidate the kinetics of core release above 250 °C to establish definitive operational thermal thresholds. Nonetheless, the TGA results confirm the polystyrene shell effectively enhances the thermal protection of the encapsulated linseed oil cores.

In addition to elucidating the thermal decomposition profiles, the distinct core and shell mass loss events in the microcapsule TGA curve enable quantification of the relative core and shell content. Specifically, integration of the area under each degradation region allows the determination of the core-shell weight ratio and the oil content of the microcapsules. The initial degradation from 300-380 °C, accounting for 70% of the total mass loss, is attributed to volatilization and breakdown of the linseed oil core. The subsequent event from 380-450 °C, responsible for the remaining 30% weight loss, corresponds to the thermal decomposition of the polystyrene polymer shell. This analysis indicates a 70:30 core-shell ratio, suggesting

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efficient encapsulation of a substantial oil mass fraction. Integrating the distinct mass loss events in the microcapsule TGA profile revealed a 70% and 30% difference between the initial core degradation at 300-380 °C and subsequent shell breakdown at 380- 450 °C. Using these weight percentages, the core-shell ratio can be calculated by the equation (2):

$$\text{Core-shell ratio} = W_{core} / (W_{core} + W_{shell}) \quad (2)$$

Where W_{core} and W_{shell} are the weight losses of the core and shell regions, respectively. Inputting the experimental values gives:

$$\text{Core-shell ratio} = 70\% / (70\% + 30\%) = 0.7$$

This result confirms the core constitutes 70% of the total microcapsule weight, while the shell is 30%. The quantified 0.7 core-shell ratio aligns closely with the 2:1 core-shell ratio utilized during microcapsule synthesis. Furthermore, the 70% encapsulated linseed oil verifies the oil content determined by extraction experiments in Section 3.4.

3.7. Characteristics and Purity of SiC NPs

TEM was used to characterize SiC NPs and identify a contaminant and its composition. TEM micrographs of SiC NPs are displayed in Fig. 6. A selective area electron diffraction (SAED) pattern is seen, confirming the crystalline structure of SiC NPs. These micrographs illustrate the spherical and cylindrical morphology of SiC NPs with particle size distributions of 10–60 nm. Smart nanocomposite coatings need high-quality nanoparticles. TEM micrographs of SiC NPs and elemental EDS maps for silicon and carbon show no evidence of elemental contaminants. < 0.5 wt.% copper traces have also been found (EDX chart in Fig. 6). The TEM's copper grid with an amorphous carbon substrate is a source of contamination. They did not affect test results; large-scale nanoparticle purity is 100%.

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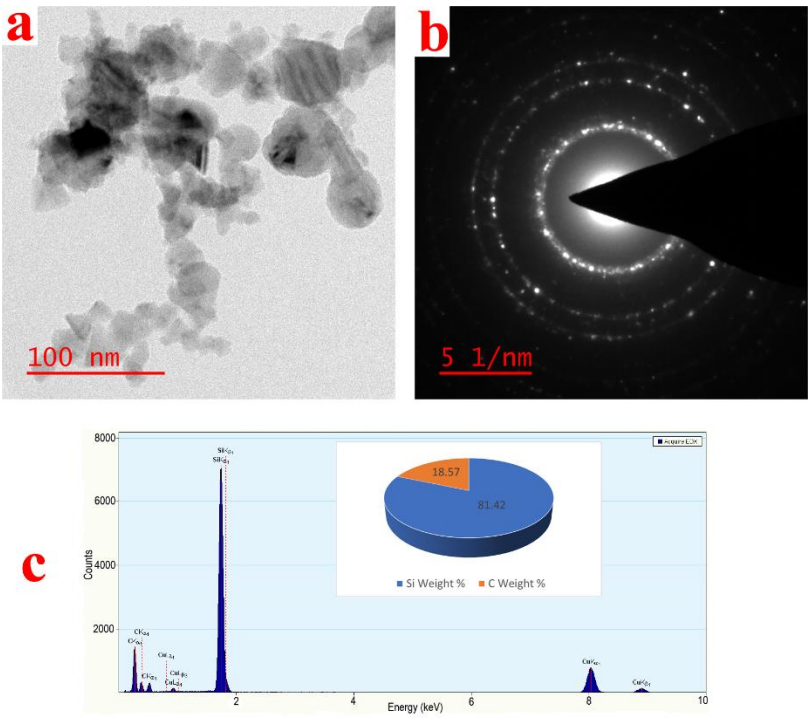

Fig. 6. (a) TEM micrograph of SiC NPs (b) corresponding SAED pattern (3) EDX of SiC NPs.

3.8. SiC NPs distribution in smart nanocomposite coatings

Additives must be dispersed uniformly throughout the coating matrix to optimize coating effectiveness. Microcapsules and nanoparticles need good dispersion to avoid aggregation. Agglomerates have a larger diameter than additives, which can cause problems. Poor additive distribution lowers coatings' mechanical, corrosion, and wear properties [46]. The SiC NPs additive distribution uniformity was determined using the FESEM-EDX mapping. Fig. 7 shows SiC NPs EDX mapping in 15M1S, 15M2S, 15M3S, and 15M4S samples. Homogeneous samples with uniform SiC NPs distribution were obtained, as demonstrated. While the SiC NPs are well-dispersed throughout the coating matrix, leading to a generally homogeneous distribution as demonstrated by FESEM-EDX mapping, there may be localized instances of agglomeration, particularly in the samples with higher SiC NPs content.

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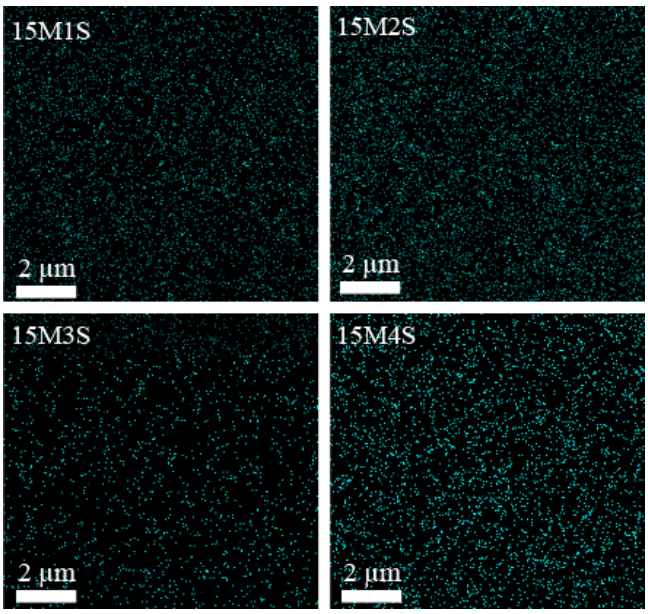

Fig. 7. EDX Si mapping of smart nanocomposite coating samples.

The TEM micrograph of the 15M3S sample shows the actual distribution of SiC NPs in the epoxy matrix (Fig. 8). The micrograph is from a microcapsule-free zone due to their large size at this magnification. Clearly, increasing SiC NPs content increases agglomeration potential. However, the coating's optimal preparation and the compatibility of the SiC NPs with the epoxy matrix allowed for excellent distribution of the 3 wt.% SiC NPs. Also, due to the excessive compressive stress that emerges during the curing process, the dispersion of SiC NPs should get better compared to the as-received state [47], and accumulation should decrease. Hence, as shown in Fig. 8, coatings are isotropic due to homogeneous dispersion.

Also, Fig. 8 exhibits no noticeable voids or porosity within the 15M3S coating sample. Upon careful examination of the image, there are no visible regions characterized by empty spaces or voids that could compromise the structural integrity of the coating. The absence of voids or porosity is an important characteristic, as it indicates a high level of quality and uniformity in the coating. This observation suggests that the manufacturing process and

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nanoparticle dispersion methods have been effectively managed, resulting in a high-quality and dense nanocomposite coating. Such a defect-free structure ensures the coating's mechanical strength and overall application performance.

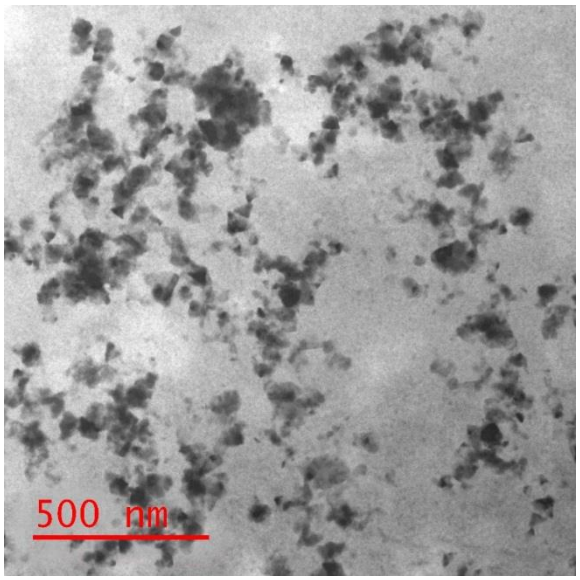

Fig. 8. TEM micrograph of the 15M3S coating sample filled with 3 wt.% of SiC NPs.

3.9. The hardness of the coatings

Fig. 9 shows the hardness test results for the mechanical properties of the coating. Adding 15% microcapsules decreased the hardness, whereas adding SiC NPs, on the other hand, could increase it with a slow slope. Indeed, microcapsules with a weak polystyrene shell and the inability to withstand shear stress reduce a coating's hardness [48–51], and SiC NPs, which have high strength and hardness, increase it. SiC NPs are exceptionally hard and exhibit high abrasion resistance. When incorporated into a coating, they act as a reinforcement phase, effectively increasing the hardness of the coating. Thus, the additives' kind, shape, strength, and weight percentage dictate each coating's ultimate hardness.

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The 15M4S sample's maximum hardness of 79.7 ± 0.3 Shore D (16.18% more than the 0M0S coating sample with 68.6 ± 0.3 Shore D) is attributable to the coating's higher weight percentage of SiC NPs compared to other samples and their resistance to the indenter's force in the hardness machine. The hardness impact of 4 wt.% SiC NPs is more robust in this coating than the weakening effect of 15 wt.% microcapsules. On the other side, the low hardness of the 15M0S sample, 64 ± 0.2 Shore D (-6.71 % from the 0M0S sample), is due to the weakening impact of the coating's microcapsules, which provide little resistance to the hardness machine's indenter force.

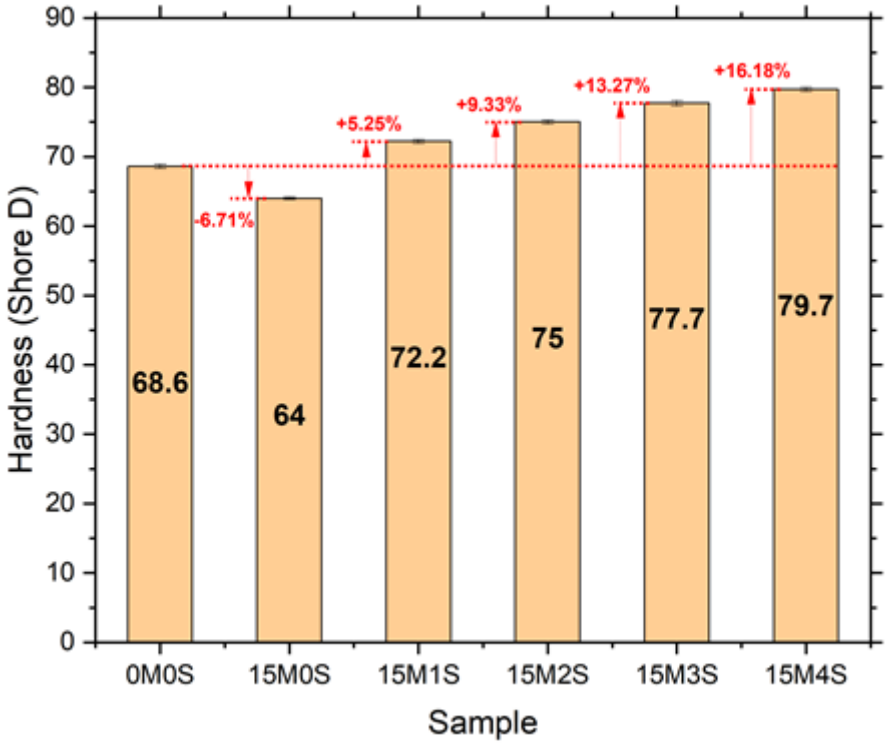

Fig. 9. Columnar chart illustrating the variation in hardness of coating samples and the percentage change compared to the 0M0S sample.

3.10. The self-healing capability of the coatings

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Fig. 10 shows how corrosive exposure affects the appearance of the coatings, especially in scratched areas. As the immersion time in the corrosive medium increased, some samples were corroded. In the 0M0S sample, corrosion begins from the start and worsens over time, resulting in severe corrosion at the end of the test. After 72 h, mild corrosion developed in the 15M0S sample and progressed throughout the test. As SiC NPs' weight percentage in the coating formulation increased, corrosion decreased. The 15M3S and 15M4S coatings showed no corrosion until the end of the test (216 h). After the test, none of the samples showed signs of coating lift or corrosive solution penetration into the substrate-coating interface. In other words, all samples rated the coatings' adherence to the substrate as excellent.

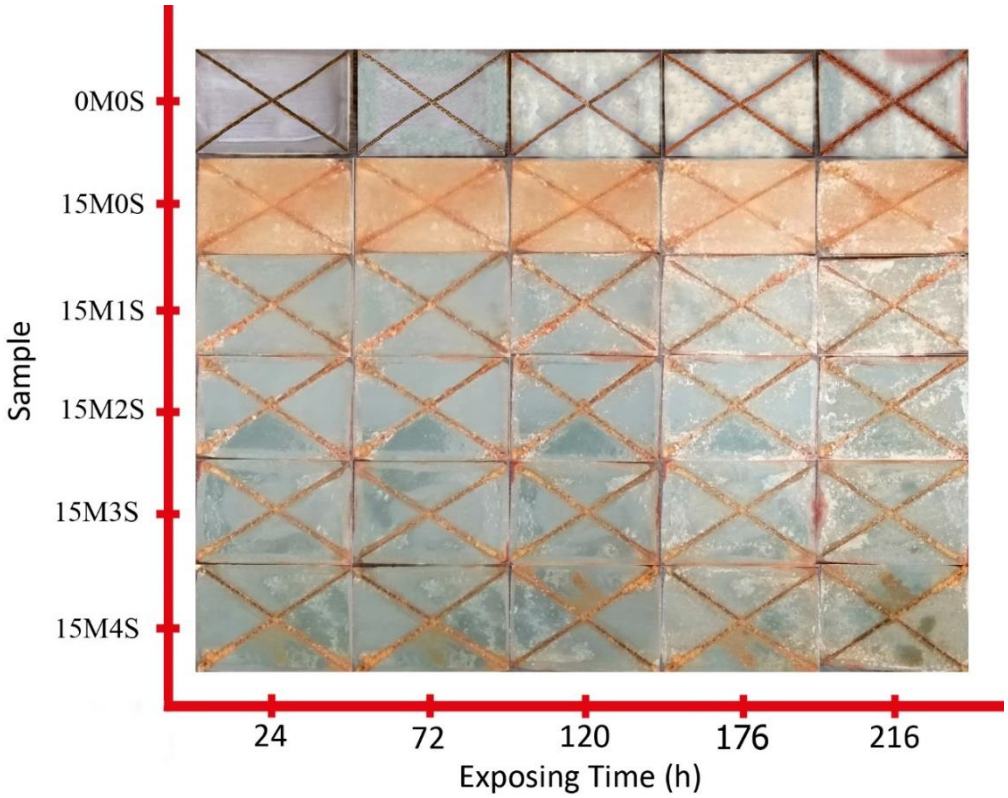

Fig. 10. Photographs of the scratched area in steel plates coated with various coating samples.

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Fig. 11 shows FESEM micrographs of each sample groove after the test. The groove in the 0M0S sample lacks a protective film, so its surface corrodes severely at the end of the test. In this sample, the groove's depth and width are the same as at the start of the test, and the substrate's exposed surface is unprotected. In other samples, however, the healing agent film is visible on the scratch, and the groove's width and depth have changed due to a newly generated protective layer. The 15M0S sample completely repaired the scratches' central section, while pores appeared in only a few areas. In other samples, SiC NPs improved the healing condition of grooves and healed their depth and width more efficiently. The 15M1S sample's groove is nearly repaired, but its width is flawed. The 15M2S sample's healing agent filled the groove's depth and width. The 15M3S sample's groove was almost completely filled with a new protective layer. The 15M4S sample's groove depth was repaired and aligned with the epoxy surface.

The proposed self-healing mechanism is such that after artificial scratching (or mechanical damage during service) on the coating, the microcapsules are broken, and linseed oil is released at the groove and reacts with air oxygen. Oil polymerizes, seals the groove, and heals the crack by forming a new layer.

SiC NPs play a crucial role in enhancing the corrosion resistance of coatings through several mechanisms. The corrosion protection conferred by SiC NPs stems from their inherent chemical stability and low diffusivity for corrosive agents. When distributed within the epoxy matrix, SiC NPs act as a physical barrier and create a tortuous path for the ingress of moisture, chemicals, and salts. This path incrementally increases the diffusion path length and creates physical hindrances, thereby significantly reducing the rate at which these agents can reach and react with the underlying, reducing the likelihood of corrosion. Moreover, the hydrophobic nature of SiC NPs aids in reducing water uptake of the epoxy matrix, providing further

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resistance to moisture-induced degradation and corrosion processes. Additionally, the SiC NPs can attract corrosion inhibitors used within the matrix due to their surface chemistry, keeping these inhibitors in place to counteract the corrosive agents more effectively. Also, SiC NPs are chemically inert and resistant to a wide range of corrosive environments, including acids and alkalis. When incorporated into epoxy coatings, SiC NPs impart their chemical stability to the coating, making it less susceptible to chemical attack. Therefore, when their weight percentage in the coating increases, it provides better protection in coatings containing SiC NPs.

Another mechanism is that adding SiC NPs can help reduce the porosity of epoxy coatings. The reduction in porosity can be attributed to the SiC NPs' ability to occupy the voids that are inherently present within the epoxy matrix. When the SiC NPs are distributed throughout the epoxy resin, they fill the interstitial spaces that could otherwise become potential sites for pores during the curing process. Fewer pores and defects mean fewer pathways for corrosive agents to penetrate the coating. This reduction in porosity enhances the overall integrity and durability of the coating. Moreover, the high surface area to volume ratio of SiC NPs facilitates a better interaction with the epoxy matrix, which promotes a more uniform cross-linking density during the curing phase. This enhanced cross-linking further densifies the material, effectively decreasing the likelihood of porosity. The nanoparticles also act as nucleation sites for the epoxy resin, providing structural integrity that can prevent the formation of voids. As the distribution of SiC NPs increases throughout the matrix, the path for any entrapped air or volatile compounds to coalesce into larger pores during the curing process becomes disrupted, leading to a decrease in the overall porosity of the cured product.

The increase in the coating's hardness due to the nanoparticles mentioned above can also make the surface more resistant to mechanical abrasion, which can expose the substrate to corrosive elements. Combining with other additives, i.e., microcapsules, can create synergistic

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effects. This combination can offer enhanced protection by providing both a physical barrier and chemical resistance so that the groove heals entirely with enough SiC NPs (> 3 wt.%). In Fig. 11, FESEM micrographs of 15M3S and 15M4S samples demonstrate complete groove healing due to sufficient SiC NPs.

Based on the above observations, it can be concluded that linseed oil microcapsules can provide self-healing in coatings. In addition, although microcapsules play a substantial role in preserving steel substrates, their presence alone at 15 wt.% is insufficient to prevent corrosion. With more than 3% SiC NPs, the substrate was completely protected. SiC NPs enhanced the self-healing property of smart coatings containing microcapsules [36]. Another finding that can be inferred is that the microcapsules remained intact in the coating throughout the preparation and application process. This is evident from the successful occurrence of self-healing in samples containing these microcapsules.

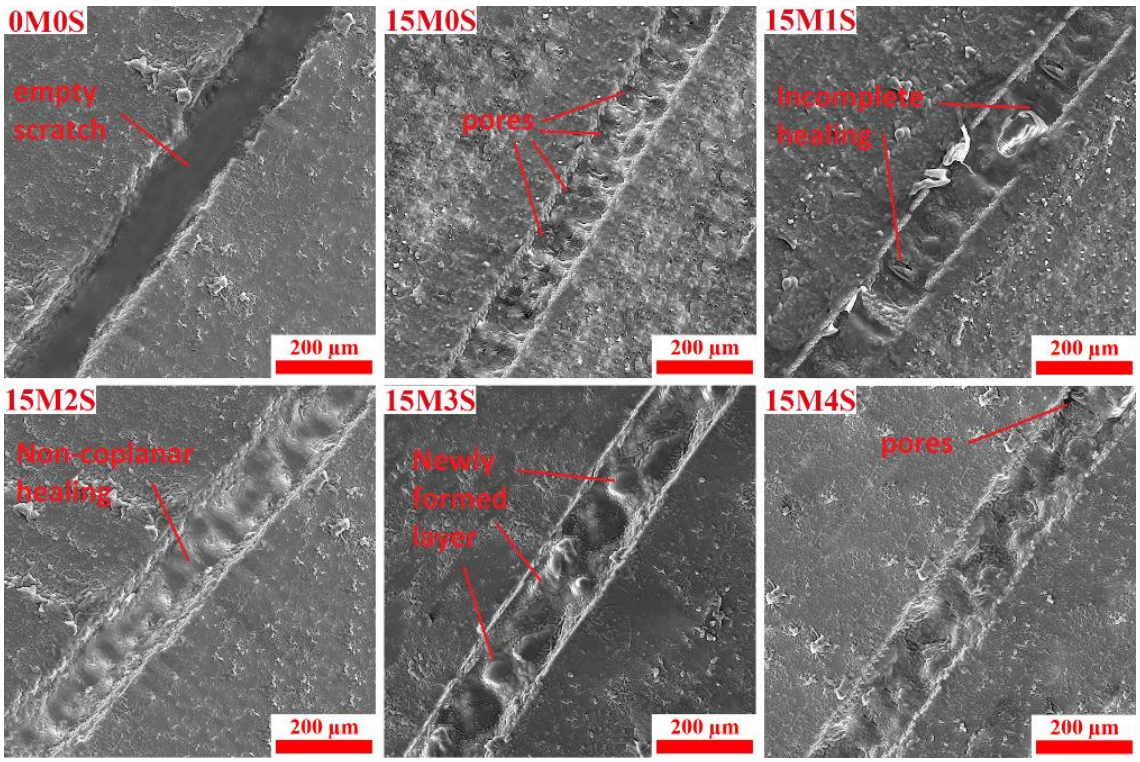

Fig. 11. FESEM micrographs of the scratched area in neat and self-healed coating samples.

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3.11. EIS results of the coatings

EIS was utilized to quantitatively evaluate the self-healing capability of a series of epoxy coatings after qualitative assessment in a 10% NaCl solution did not yield discernible differences (Fig. 10). The interpreted EIS results for all the coating samples after espousing in 10% NaCl solution for 216 h, presented in Fig. 12 (Nyquist fitting diagrams) and Fig. 13 (Nyquist and Bode plots), were attained through fitting procedures using ZSimpWin 3.20 software and are subsequently quantified by their chi-square values. In addition to qualitative evaluations, the chi-square values provide a solid quantitative measure for fitting results.

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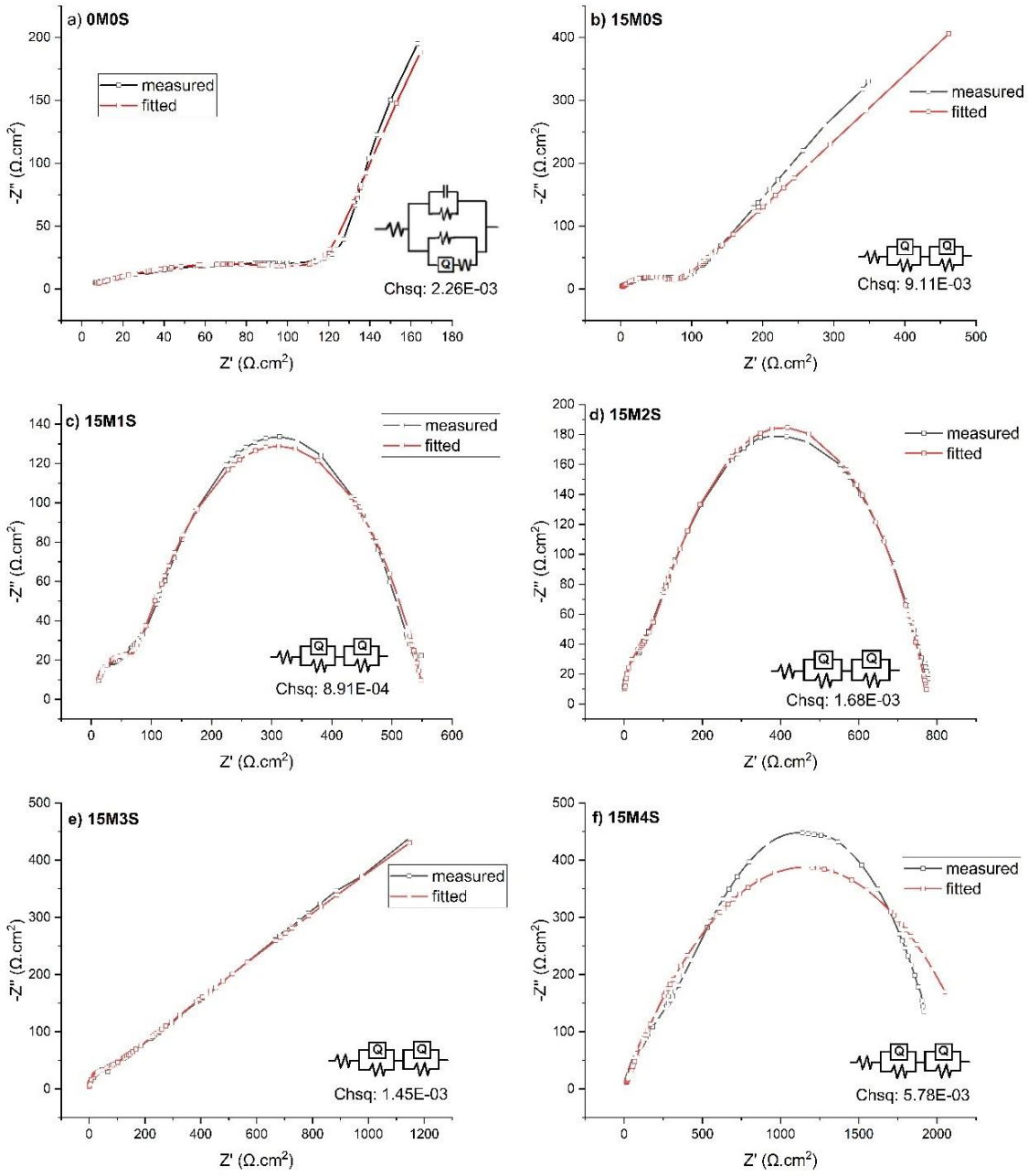

Fig. 12. Nyquist plots of the scratched coating after 216 h immersion in 10% NaCl solution, fitted with equivalent circuit models

The 0M0S sample was modeled using the equivalent circuit $R_s((C_{dl}R_p)(C_{crevice}(R_{crevice}W)))$. Consistently used in prior literature for organic epoxy coatings, this circuit attributes R_s to solution resistance, C_{dl} to double-layer capacitance, R_p to polarization resistance, $C_{crevice}$ to the

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crevice capacitance, Rcrevice to crevice resistance, and W to Warburg impedance, which represents mass transfer limitations within the double layer. For the microcapsules containing samples, an alternative equivalent circuit $R_s(Q_cR_{po})(Q_{dl}R_{ct})$ was employed. This adjustment accounts for the electrolyte penetration within the coating reaching saturation, prompting the formation of micro batteries at the interface between the coating and base metal. The terms Q_c and Q_{dl} denote the coating and electric double-layer capacitances, respectively, while R_{po} and R_{ct} stand for the microporous and charge transfer resistances. The software's fit refinement utilizing constant phase elements (Q) instead of ideal capacitors (C) addresses the inhomogeneous nature of the interface. Mathematically $ZCPE = (1/Q\omega n)e^{-ni\pi/2}$, and if n = 1, then Q = C.

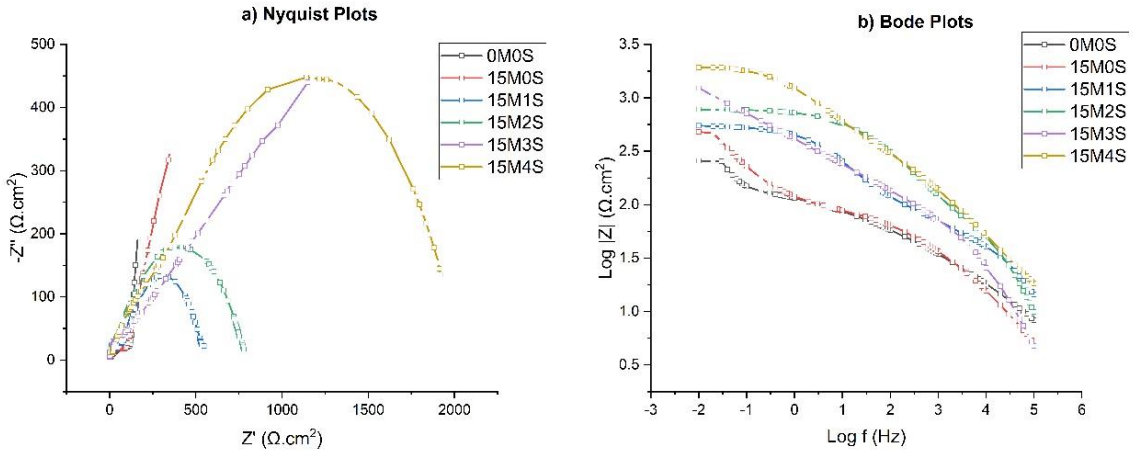

Fig. 13. Nyquist and Bode plots of the coating samples after 216 h immersion in 10% NaCl solution

The data derived from the software fitting are tabulated in Table 2. Impedance modulus (|Z|) at a low frequency of 0.01 Hz was considered for determining the coating resistance. The results indicate a direct correlation between the rise in SiC NPs content and enhanced coating

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resistance. The observed trend underscores the efficacy of SiC nanoparticle incorporation in augmenting the protective attributes of epoxy coatings.

Table 2. Electrical circuit model parameters of the scratched coating samples after espousing in 10% NaCl solution for 216 h.

Sample	R_s ($\Omega \cdot cm^2$)	C_{dl} ($F \cdot cm^{-2}$)	R_p ($\Omega \cdot cm^2$)	$C_{crevice}$ ($F \cdot cm^{-2}$)	$R_{crevice}$ ($\Omega \cdot cm^2$)	W ($\Omega \cdot cm^2/s^{½}$)	Log|Z| = $R_{coating}$
0M0S	8.43E-06	9.48E-02	23.2	2.08E-03	1.34	9.79E-09	2.41014
Sample	**R_s ($\Omega \cdot cm^2$)**	**Q_c ($F \cdot cm^{-2}$)**	**R_{po} ($\Omega \cdot cm^2$)**	**Q_{dl} ($F \cdot cm^{-2}$)**	**R_{ct} ($\Omega \cdot cm^2$)**	-	**Log|Z| = $R_{coating}$**
15M0S	11.0	3.23E-03	80.34	7.38E-03	573	-	2.68151
15M1S	17.5	6.84E-04	90.76	2.36E-04	596	-	2.73957
15M2S	23.3	1.24E-04	117.5	1.86E-04	1265	-	2.89107
15M3S	34.2	6.18E-05	135.2	8.64E-05	2730	-	3.09033
15M4S	45.2	4.66E-05	147.4	1.50E-05	2634	-	3.28404

The Nyquist and Bode plots in Fig. 13 demonstrate that as the SiC NPs' content increases, there is a corresponding increase in the total impedance, enhancing their corrosion protection performance. This is evidenced by the rise in total impedance (diameter of Nyquist plots) and low-frequency impedance modulus (|Z| at 0.01 Hz) with increasing SiC NPS content, where the neat epoxy coating 0M0S exhibits a Log|Z|0.01Hz value of 2.41. The 15M4S coating with the highest SiC NPs content reaches a significantly higher value of 3.28. In Nyquist plots, this improvement manifests as an enlarged semicircular diameter, indicative of a charge transfer controlled process. The larger the semicircle, the more excellent the charge transfer resistance, implying more effective retardation of electrochemical reactions at the interface between the coating and the corrosive electrolyte environment. This suggests that SiC NPs shield against

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corrosive agents by forming a more compact and less permeable network within the epoxy matrix.

In the context of the Bode phase plots, a single time constant observed suggests that the coatings predominantly exhibit capacitive behavior, which aligns with expectations for barrier-type coatings. This singular time constant infers that the dominant electrochemical process is related to capacitance, typically associated with the physical barrier properties of the coating. As the SiC NP content increases, so does the low-frequency impedance modulus, further demonstrating an increment in the barrier properties of the coatings. The SiC NPs likely occupy and, therefore, reduce the free volume within the epoxy matrix, resulting in a denser, more structurally sound coating. This physical densification minimizes pathways for electrolyte ingress, thereby enhancing the coating's ability to serve as an effective barrier against corrosion. Moreover, the SiC NPs might contribute to a more tortuous path for ion migration through the coating, effectively increasing the path length that corrosive agents must traverse to reach the metal surface. This increased pathway complexity could contribute to the observed increase in low-frequency impedance and the apparent improvements in overall coating performance. This interpretation is consistent with the phenomenological understanding that the scattering of electrons and defect generation at the particle-matrix interface can enhance the composite coatings' barrier properties. The incorporation of SiC NPs reinforces this interface, providing sites of heterogeneous nucleation that may slow down electrochemical reactions by reducing the active sites available for such processes on the metal surface.

3.11. The self-lubricating capability of the coatings

Microcapsules and SiC NPs affected nanocomposite coating wear. Based on the data obtained from the wear test, including instantaneous frictional force at each distance and

sample weight reduction, instantaneous COF values at each distance, and mean COF values and SWR for each sample were calculated (Figs. 14 and 15). It is worthy that the noise observed in the evolution of the COF can be attributed to various factors inherent in the tribological testing process. As the COF is a measurement of resistance at the interface of two surfaces in motion, any minute variations in surface roughness, presence of wear particles, or changes in lubrication can contribute to signal fluctuations. The instabilities observed beyond the 2% SiC NPs addition may indicate a transition in the wear mechanism, attributed to the presence of a higher concentration of SiC NPs. At these concentrations, the interaction between the particles and the counter face can lead to sporadic fluctuations in the COF. As the SiC particle concentration increases, there is likely to be a higher tendency for agglomeration, which may also cause temporary increases in the COF when these agglomerates pass between the contact surfaces.

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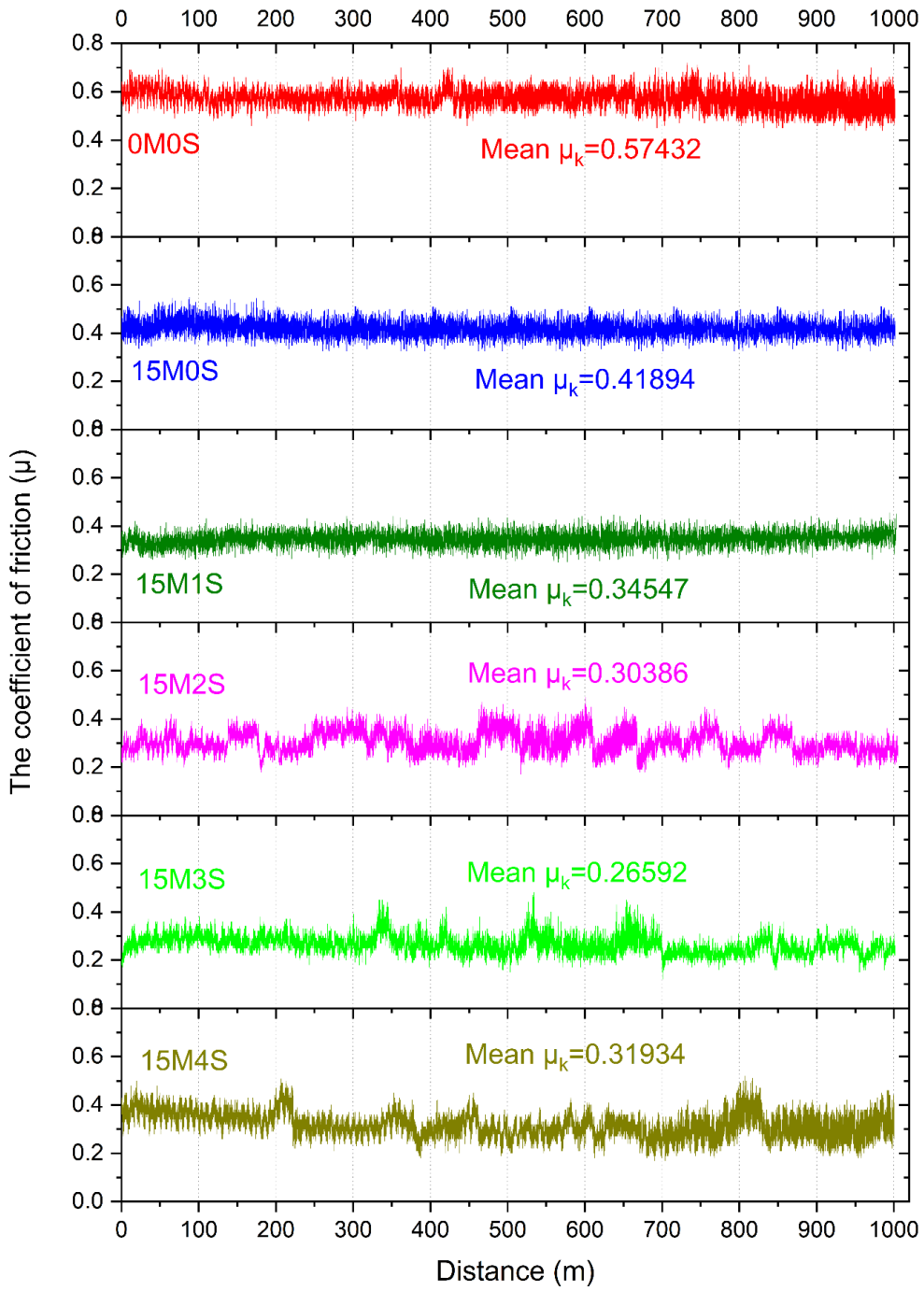

Fig. 14. Scatter chart illustrating the coefficient of friction of coating samples during wear test.

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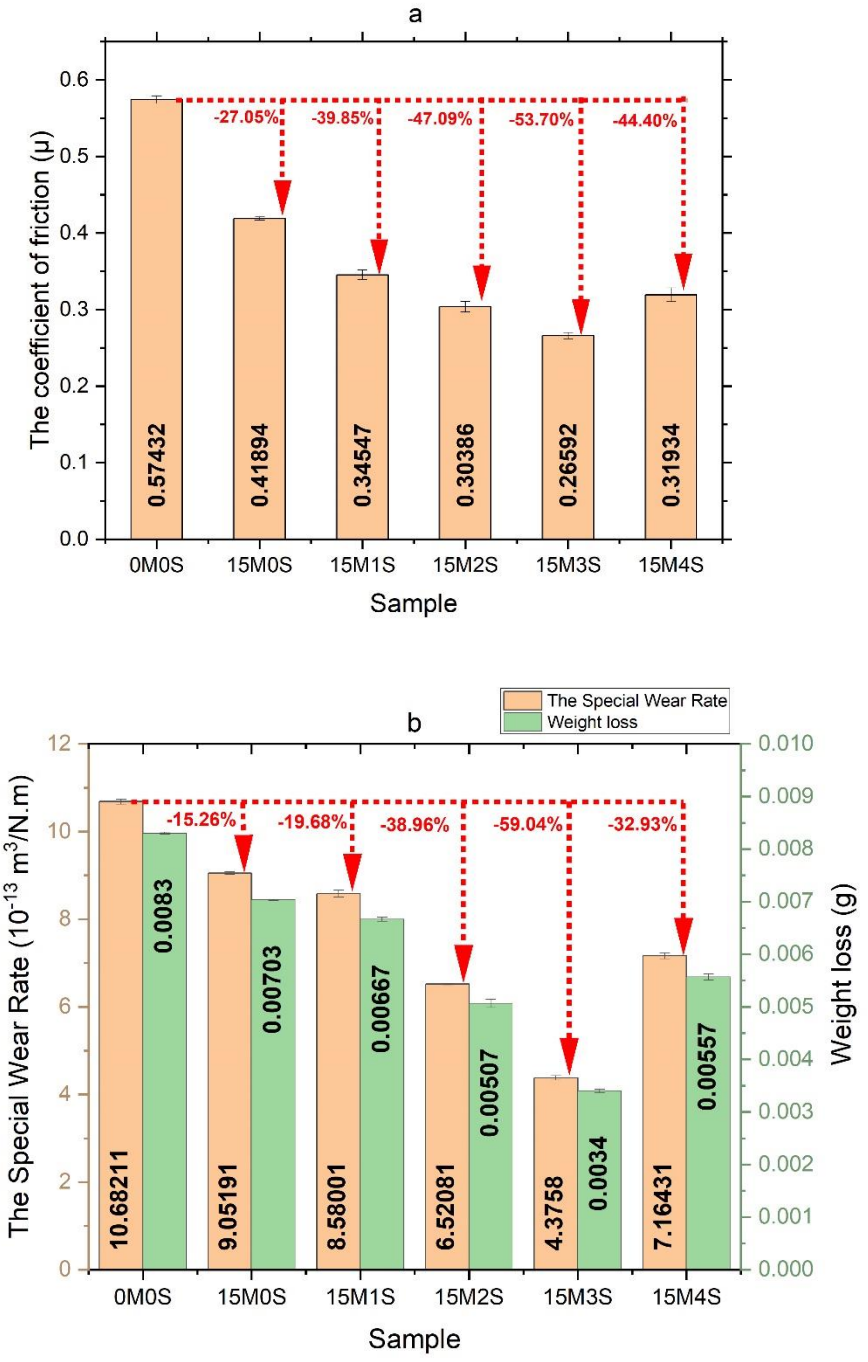

Fig. 15. Columnar chart illustrating the variation in a) the coefficient of friction, b) the specific wear rate and weight loss of coating samples, and the percentage change compared to the 0M0S coating sample.

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COF diagrams and FESEM observations were used to investigate the mechanisms affecting the wear resistance of coatings. The 0M0S sample with a COF of 0.5744 and an SWR of 10.682×10^{-13} $m^3/N.m$ has the lowest wear resistance. The 0M0S sample's FESEM micrograph shows parallel grooves on its surface, a hallmark of the abrasive wear mechanism. There are also wave-shaped wear lines and localized crumbling, which can be attributed to fatigue wear and the fatigue layering mechanism in this sample [49,52–57].

Adding 15% microcapsules to a 0M0S sample and making a 15M0S smart coating changed its wear resistance utterly. COF and SWR decreased by 27.05% and 15.26% to 0.4189 and 9.052×10^{-13} $m^3/N.m$, respectively. This improvement in wear resistance is attributed to the presence of microcapsules. The examination of FESEM micrographs reveals the formation of lubricating films on the worn surfaces, which is indicative of linseed oil's polishing effect, as evidenced by the smoother appearance of said surfaces. This polishing influence can be attributed to the linseed oil, which, upon release from the embedded microcapsules, generates a lubricating film. This film effectively reduces the direct interaction between sliding surfaces, ordinarily facilitating smoother movement and diminishing the severity of wear—a phenomenon corroborating the polishing effect discernible in the FESEM micrographs.

The following mechanism was proposed to improve wear resistance in this sample: The 15M0S sample's epoxy matrix degrades during wear testing, breaking the microcapsule shell and releasing linseed oil into the wear interface. By creating a liquid lubricant layer, COF and SWR are reduced. The cavity formed when microcapsules are emptied also reduces COF. These voids collect interface wear particles such as an epoxy matrix and a microcapsule shell (repairing mechanism). These two actions alter fatigue wear and improve 15M0S's wear properties.

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Adding SiC NPs to the 15M0S smart coating improved the wear resistance. The 15M3S sample showed the lowest COF and SWR values, 0.3918 and 4.376×10^{-13} $m^3/N.m$, representing respective decreases of 31.79% and 59.04% from the neat coating (0M0S). The improvement in wear resistance in smart nanocomposite coating samples is ascribed to the synergistic impact of microcapsules and SiC NPs. Adding SiC NPs up to 3 wt.% improves the mechanical properties, as evidenced by increased hardness. However, the wear properties deteriorate at the 15M4S sample. This demonstrates that an intermediate SiC NPs concentration maximizes wear resistance enhancement.

FESEM micrographs of these samples in Fig. 16 show that as the SiC NPs content increased, the wear surface smoothed out until the 15M3S sample, at which size and depth of the scratch wear lines decreased. The 15M1S and 15M2S samples had fewer furrows and matrix debris compared to the 15M0S sample, which is consistent with their lower SWR. The 15M3S had the smoothest wear level and lowest SWR. The 15M4S sample clearly shows an accumulation of SiC NPs extracted from smart nanocomposite coatings. Cavities seen in FESEM micrographs of smart nanocomposite coatings indicate microcapsule rupture and the creation of a lubricating layer at the interface, a process identical to that observed with the 15M0S sample.

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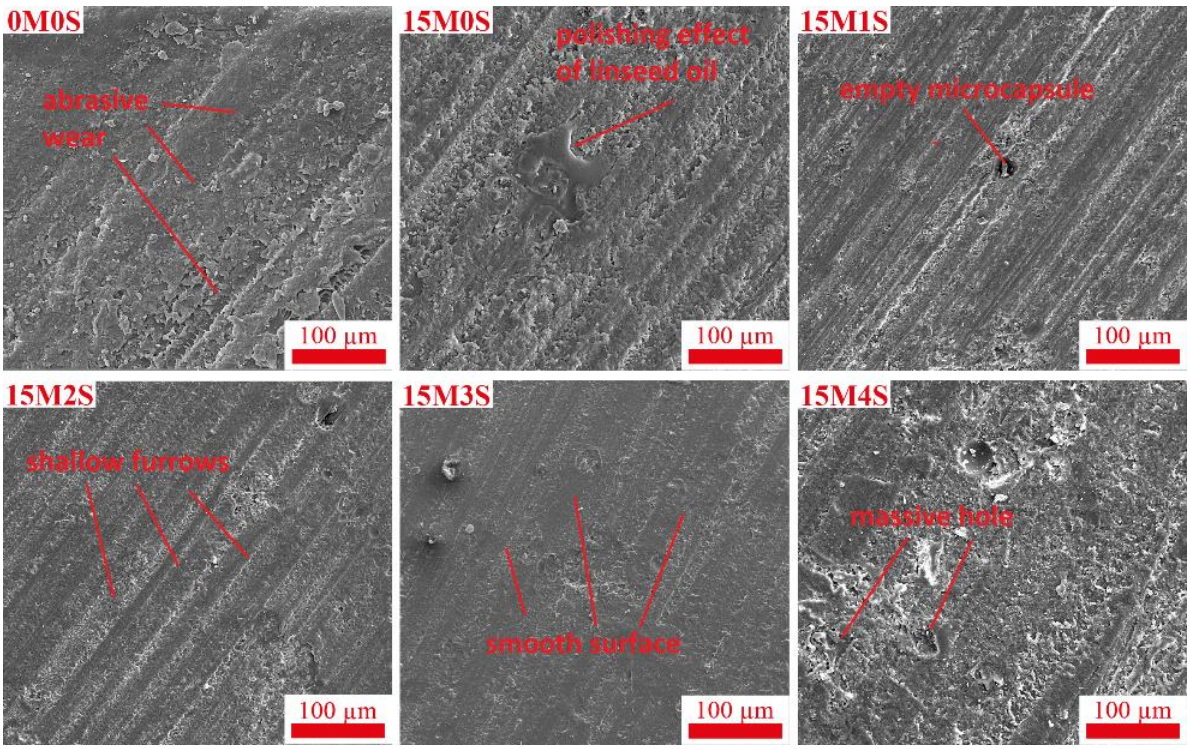

Fig. 16. FESEM micrographs of the wear surface in coating samples after the wear test.

When wear occurs, a mixed texture comprising epoxy resin fragments, wear debris, oxidation products, adhesive or abrasive wear products, chemical compounds, corrosion products, and so on, in the form of lubricating rollers, are expected to form at the interface. The term "lubricating rollers" refers to a self-organizing structure of debris and material fragments that are generated during the wear process. These particles can form into spherical or cylindrical shapes under the influence of the tribological forces present at the interface between the sliding surfaces. These shapes act similarly to microscopic bearings or 'rollers,' facilitating a form of lubrication that can potentially reduce the overall friction coefficient and wear rate by creating a rolling effect rather than sliding friction. Combining the rolling effect of this texture and polishing operations of SiC NPs improves the surface quality of smart nanocomposite coating. Fig. 17 (a) demonstrates the existence of such rollers. Cavities and

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cracks formed during wear can also be filled with these rollers to improve the wear resistance of these samples [58]. Fig. 17 (b) shows how rollers repair these regions in this mechanism.

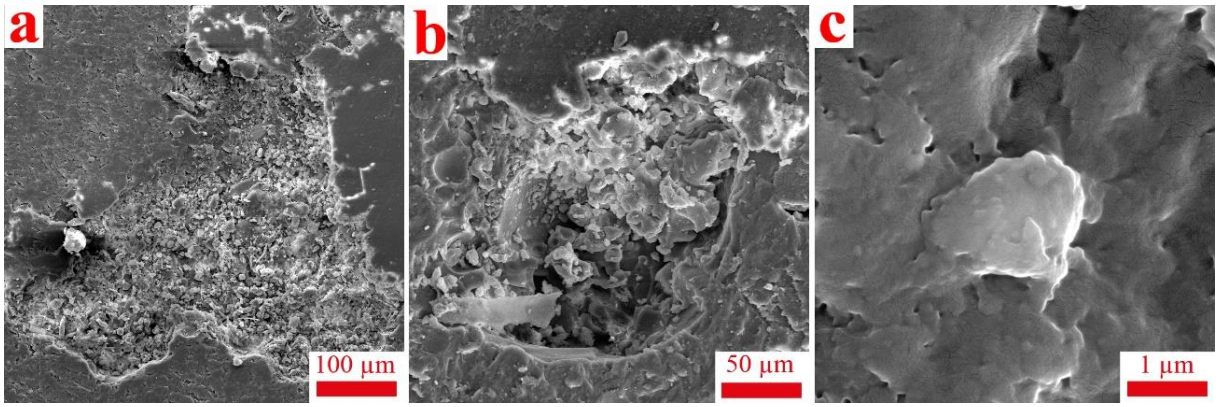

Fig. 17. FESEM micrograph of (a) rolls in the 15M2S sample, (b) SiC NPs filling cavities and cracks in the 15M3S sample, and (c) solid/liquid lubricant film in the 15M3S sample.

Creating a solid/liquid lubricant film is another mechanism that reduces COF in the 15M3S sample to a greater extent than other smart nanocomposite coatings. SiC NPs' high specific surface area and oil-friendly properties prevent oil leakage and create cohesive oily tissue at the interface. As the FESEM micrograph in Fig. 17 (c) shows, a solid/liquid lubricant film is formed with the addition of debris, and it provides a tribo-film between the two wear sides, lowering the COF in this sample. The tribo-film consists of a complex mixture of mechanically mixed layers from the original material, contaminants, and any debris generated during wear. These films form at microscopic scales under the heat and pressure of tribological loading and can consist of chemical reaction products such as oxides, as well as polymers that exhibit plastic deformation or reflow at elevated temperatures to create the 'liquid' phase observed. The micrograph obtained at high magnification enables observation of distinctive structural features characteristic of the tribo-film, which may not be discernible at lower

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magnifications or from alternative perspectives distant to the wear interface. The 15M3S sample shows this behavior more than other smart nanocomposite coatings. The EDS analysis of the tribo-film (Fig. 18) shows Si and C from SiC NPs. This much carbon can be derived from linseed oil, epoxy, and polystyrene shells.

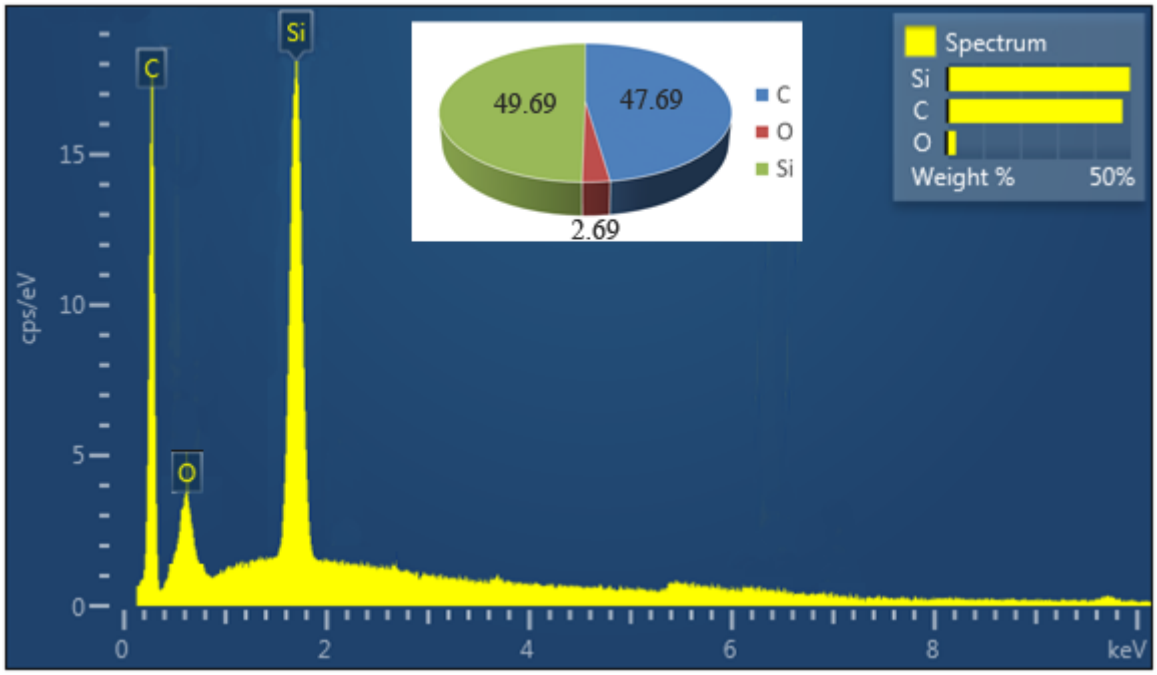

Fig. 18. EDS analysis of the film formed on the wear surface of the 15M3S smart nanocomposite epoxy coating sample.

Interconnected SiC NPs at the wear surface and large SiC NPs agglomerates in the coating bulk and wear interface increased COF and SWR in the 15M4S sample. As the amount of SiC NPs in the coating formulation increases, agglomeration probability increases during coating preparation. It is worthy that homogeneity in EDX mapping only represents the optimal preparation and proper and acceptable distribution of SiC NPS in the samples and does not preclude the presence of microscale agglomerates that may not be resolved at the scale of the mapping. When the surrounding epoxy matrix erodes, the agglomerates fall into the interface,

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leaving a massive hole in the coating. In Fig. 19 (a), a hole in the coating's wear surface has been produced, reducing its wear qualities. Fig. 19 (b) and (c) micrographs show large agglomerates larger than the solid/liquid lubricant spacer layer due to increasing COF.

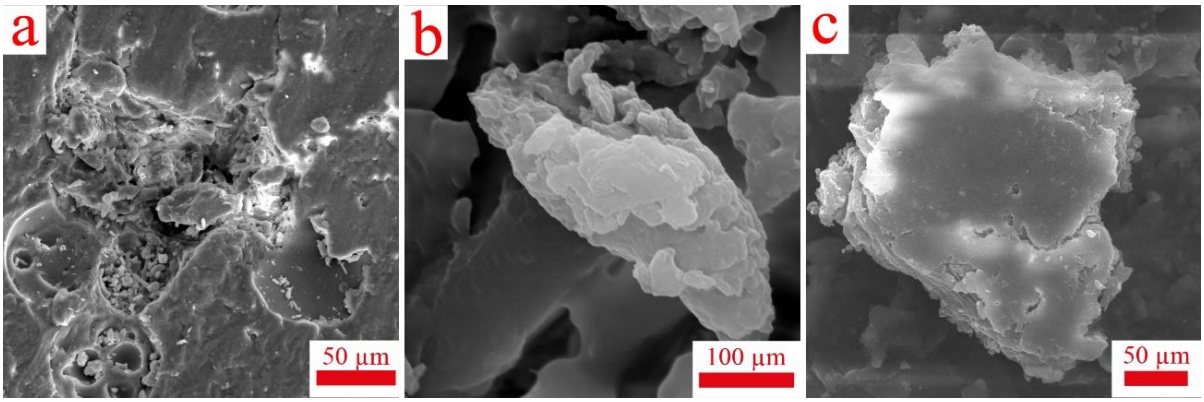

Fig. 19. FESEM micrograph of a) large hole, (b) and (c) large agglomerates in the wear surface of the 15M4S smart nanocomposite epoxy coating sample.

4. Conclusions

This study highlights the feasibility of the industrial application of self-healing and self-lubricating coatings by adding microcapsules and SiC nanoparticles into the epoxy matrix. Linseed-oil-filled polystyrene microcapsules were prepared using the solvent evaporation method. For excellent adhesion to the epoxy, the microcapsules with a spherical shape and rough surface morphology were obtained with a particle size distribution of 6-26 µm and an average particle size of 15.3 µm, which is ideal for use in the coating. Other characteristics of the synthesized microcapsules, including the microencapsulation process's efficiency, the oil content of the microcapsules, and oil encapsulation's efficiency, were also calculated. The results were 72.64 ± 2.58%, 72.79 ± 2.85%, and 73.81 ± 1.43%, respectively. Values above 70% guarantee the optimal performance of microcapsules.

TEM evaluated the SiC nanoparticles' purity and characteristics. The results showed crystalline structure, spherical and cylindrical morphology, and 10–60 nm particle size distributions. TEM determined SiC nanoparticle distribution in smart nanocomposite coatings. EDX Si mapping of SiC nanoparticles showed homogeneity in 15M1S, 15M2S, 15M3S, and 15M4S samples.

The coating's resistance to external damage is crucial, so 4 wt.% SiC nanoparticles were added to increase its hardness by 16.18% compared to neat epoxy. Self-healing capability measures corrosion resistance and longevity, another coating quality indicator. The best self-healing capability was obtained in samples with 3 wt.% and 4 wt.% SiC nanoparticles. The reduction in the specific wear rate of the synthesized coating is shown to be as low as possible in samples containing 3 wt.% SiC nanoparticles with a mean coefficient of friction of 0.3918 ± 0.00005 and a specific wear rate of $4.376 \pm 0.0006 \times 10^{-13}$ $m^3/N.m$, which were 31.79% and 59.04% improvements compared with the neat epoxy sample.

Self-healing and self-lubricating coatings ensure resistance to conventional epoxy coating applications and eliminate worry about harmful corrosion. However, periodic coating inspection is essential if the coating environment is very harsh. Most pipeline and storage tank centers use coating service companies.

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