



## Research article

# High linoleic waste sunflower oil: A distinctive recycled source of self-healing agent for smart metal coatings

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## ABSTRACT

High Linoleic Waste Sunflower Oil (HLWSO) is a new self-healing agent viable to be encapsulated. Meanwhile, the unique mechanism behind the synthesis of microcapsules is deeply illustrated; the emulsification process of HLWSO by anionic surfactant and microencapsulation of HLWSO. In addition, the application of microencapsulated HLWSO in the coating matrix by the layering method is presented followed by a detailed explanation of the self-healing mechanism of a smart coating incorporating the polymerization mechanism of HLWSO developed by the diene structure. Microencapsulation of high linoleic waste sunflower oil (HLWSO) is a wall formation process in which urea-formaldehyde (UF) is attached with emulsified HLWSO to form a microcapsule. In this study, the HLWSO from recycled cooking oil is uniquely bonded with a diene structure, thus possessing the ability to dry fast and be encapsulated via the in-situ polymerization method. The microencapsulated HLWSO was characterized using Field Emission Scanning Electron Microscopy (FESEM) and Fourier Transformation Infra-Red Spectroscopy (FTIR). The optimum microcapsules synthesized from HLWSO resulted in a smooth shell structure with 2.88  $\mu\text{m}$  diameter microcapsules at 0.31  $\mu\text{m}$  shell thickness and 66% core content. It was demonstrated that increased stirring speed decreases the size, shell thickness, and core content of the microcapsules. The FTIR results indicated that HLWSO as a core, while urea-formaldehyde acted as a shell of microcapsules. The scratch on the coating matrix embedded with HLWSO was healed after five days. The corrosion rate of optimum sample was 0.0574 mm/year, with an optimum reduction of 58% from the reference sample. This study revealed that the HLWSO from recycled sources is a viable self-healing agent to be microencapsulated. The smart coating embedded with HLWSO also displayed self-healing performance, reduced corrosion rate and beneficial for the advancement of corrosion control technology.

## 1. Introduction

The optimization of recycled and waste material has become a major industrial demand for minimizing costs. Thus, high linoleic waste sunflower oil (HLWSO) sourced from waste cooking oil is very significant for a cost-saving self-healing agent as exclusively measured in this study. The unique properties of HLWSO containing high levels of linoleic acid fall under the drying category of vegetable oil, which can be used as a self-healing agent due to its fast-drying ability, suitability as cooking oil, and abundance. The

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frying process enhances the viscosity of HLWSO. The enhancement of viscosity is a factor in the performance of drying ability compared to established self-healing agents including tung and linseed oil. Tung and linseed oils are drying but cannot be used as cooking oil due to rancidity issues [1]. Most previous studies have included both drying and non-drying types of vegetable oils. Drying vegetable oils such as tung oil [2,3], linseed oil [4], and currently, sunflower oil [5,6] are chosen by researchers due to their accelerated drying ability [7–9] that very much suits the needs of coating behavior. Palm oil [10] and coconut oil [11,12] are the non-drying types of vegetable oil that have also been explored. However, normal metal coatings suffer from micro-cracks, causing the coating structure to lose its function to protect the metal component [13]. Hence, a viable modification to improve this coating structure, such as in epoxy or metal paint, should be applied by consolidating the coating system with a smart approach of microcapsules synthesized from a self-healing agent [14–17]. The self-healing mechanism was able to embark dual-functionality such self-healing and self-lubricating [18] and triple-action such self-healing, self-response and self-controlling [19]. Generally, all industries use a manual re-coating approach or replacement of damaged instruments [18,19]. Nonetheless, the manpower required for manual re-coating maintenance is a major challenge during this global uncertainty of spreading pandemic COVID-19 [20,21].

The evolution of natural self-healing agents has led to current advancements in the manipulation of cooking oils such as palm oil [10] and sunflower oil [22,23] as healing agents to meet the demand for future green resources. Encapsulating palm oil requires more chemical procedures and in the form of palm oil-alkyd base [10] to improve drying ability. Currently, engineering technology is making advancements toward enhancing the value of recycled materials as useful resources. Owing to this challenge, the investigation of HLWSO as a recyclable material and self-healing agent in microencapsulation is inclusively covered in this research. Additionally, the mechanism of diene structure in HLWSO that induces a unique polymerization process is also presented in this discovery. Microencapsulation of HLWSO by the in-situ polymerization method was used to synthesize microcapsules. The microcapsules were embedded with epoxy coating by layering method and applied to a mild steel substrate. Once the coated mild steel was scratched, the waste sunflower oil was released and polymerized, thus healing the scratch.

## 2. Experimental method

### 2.1. Materials

The microencapsulation materials included formaldehyde solution (PC Laboratory Reagent, C0330, 37 wt%); ethylene maleic anhydride (QRêc, M1037); resorcinol (R&M Chemicals); ammonium chloride (Bendosen Laboratory Chemicals, C0059); sodium hydroxide (Merck, B0805498227), 1-octanol (Bendosen Laboratory Chemicals, C0567); urea (HmbG Chemicals, C1044) and hexane (HmbG, C0980). The high linoleic waste sunflower oil (HLWSO) was obtained from recycled waste cooking oil heated at 180 °C (reused twice, used to cook fish crackers) from Tesco, Malaysia.

### 2.2. Preparation of microcapsules

Microcapsules were prepared by in-situ polymerization technique modifying the stabilization time and reducing the mass of chemicals [22,23] (Fig. 1). First, a 250 mL beaker was filled with 100 mL of deionized (DI) water and 2 mL of ethylene maleic anhydride (EMA) at 300 rpm stirring speed and 15 min of mixing time. EMA performed as an emulsifier and an ionic surfactant. The concentration of EMA used was 5 wt%. Then, HLWSO with a mass of 10 g was added with a 15 min stabilization time at a constant stirring speed to allow emulsification. After the HLWSO was emulsified and stabilized, the next step was done by adding 0.25 g resorcinol, 0.25 g ammonium chloride, and 2.5 g urea into the mixture. Then, 5 wt% of sodium hydroxide was added to adjust the pH of the solution to 3.5 (fixed for all experimentations). Subsequently, three drops of 1-octanol were added, followed by the addition of

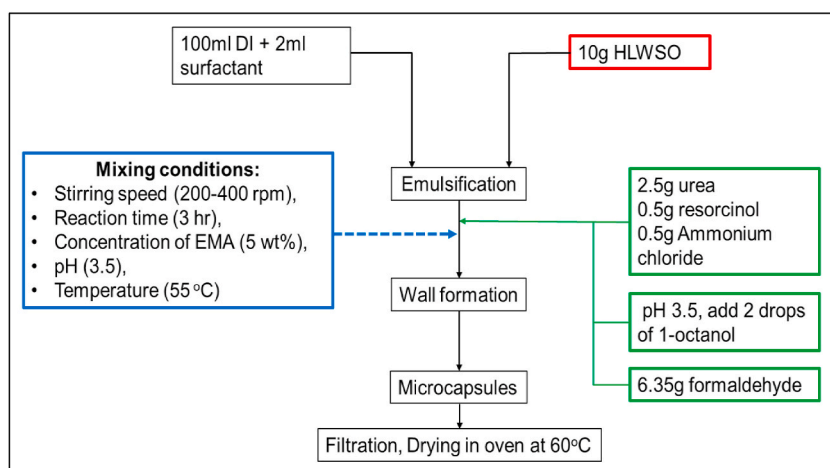


Fig. 1. The in-situ polymerization technique on microencapsulation of HLWSO.

6.25 g of formaldehyde. The stirring speed was then adjusted to 200 rpm as a studied parameter (Sample 1-S1). The mixture then was heated until 55 °C for 3 h. Once the mixture cooled, it was filtered using hexane and deionized water. The filtered microcapsules were transferred to a vacuum oven at 60 °C for 24 h. The final form of dried microcapsules existed as flowing powder form. The same procedure was applied to the studied parameters at f 300 (Sample 2-S2) and 400 rpm (Sample 3-S3) stirring speed. The parameter of stirring speed was adopted from Baharom et al. (2020) [22,23].

### 2.3. Characterization of microcapsules

Microcapsule imaging was conducted using Field Emission Scanning Electron Microscopy (FESEM) (JFM-7600 F, JEOL, Japan). The functional group of microcapsules was studied via PerkinElmer 400 N Fourier Transmission Infra-Red Spectroscopy (FTIR) characterization in the range of 400–4000  $\text{cm}^{-1}$  with a resolution of 4  $\text{cm}^{-1}$  incorporated with 32 scans/spectrum [26]. Analyses of FTIR on each microcapsule were performed in triplicate and then recorded. The core contents of the microcapsules were measured by extracting the oil through the Soxhlet extraction procedure [25–27]. The solvent used in this procedure was hexane, which is suitable for isolating suspended oil. Microcapsules were first weighed and then crushed by pestle and mortar until finely structured. The finely crushed microcapsules were then transferred to a thimble. The initial weight of the thimble was measured. The extraction process operated for 8 h; then, the thimble was transferred to a drying oven at 60 °C for 8 h. The final weight of the thimble was then measured.

The core content of microcapsules consisted of HLWSO as determined using Eqn (1): where  $W_c$  is the weight of microcapsules,  $W_{ti}$  is the initial weight of the thimble, and  $W_{tf}$  is the final weight of the thimble.

$$\text{Core content (CC) \%} = \frac{W_c - [W_{ti} - (W_{tf})]}{W_c} \times 100\% \quad (1)$$

Where,  $W_c$  was weight of microcapsules,  $W_{ti}$  was initial weight of thimble and  $W_{tf}$  was final weight of thimble.

### 2.4. Preparation of self-healing coating and evaluation of self-healing performance

Commercial grade S275JR mild steel (purchased from Oh Tung Sang Hardware Sdn. Bhd. Malaysia) with custom dimensions of 30 × 20 × 3 mm were chosen as coating substrates. The substrates were polished using sandpaper and then rinsed with acetone. 0.008 g of microcapsules containing HLWSO was measured and embedded with 0.8 g commercial grade epoxy (resin combined with hardener) with a ratio of 1:100. The epoxy coating and microcapsules were then applied to the metal substrate by the layering method (Table 1). The coating systems were dried and cured for 7 days, followed by artificial scratching with a sharp knife to reach the coating substrate to investigate the healing performance and anti-corrosion effects after immersion in sodium chloride, NaCl (3.5 wt%) for 672 h. The self-healing performance was evaluated under Scanning Electron Microscope (SEM) (Hitachi S-3400 N).

### 2.5. Anti-corrosion evaluation

The mass of coated mild steel samples before the immersion test was measured and noted as initial mass. Then, after 672 h (28 days) of immersion in 3.5 wt% NaCl, the mass loss calculation was derived from Eqn (2) [30].

$$\text{Mass loss (w)} = m_i - m_f \quad (2)$$

Where,  $m_i$  is an initial mass of sample and  $m_f$  is a final mass of sample.

Meanwhile, the corrosion rate of coated mild steel samples was measured to study the anti-corrosion ability of samples, which was derived from Eqn (3) [30].

$$\text{Corrosion rate (mm / year)} = \frac{w}{\rho \times A \times t} \quad (3)$$

Where,  $w$  is mass loss (g),  $\rho$  is density of S275JR mild steel ( $\text{g}/\text{mm}^3$ ),  $A$  is surface area of sample ( $\text{mm}^2$ ) and  $t$  is total immersion time (year).

**Table 1**  
The layering conditions of microcapsules and epoxy coating in coating matrix.

Samples	Stirring speed of microencapsulation (rpm)	Layer 1 (epoxy)	Layer 2 (microcapsules)	Layer 3 (epoxy)
1	Reference coating without microcapsules	0.8 g	–	–
2	SC1 (200 rpm)	0.4 g	0.4 g	0.4 g
3	SC2 (300 rpm)			
4	SC3 (400 rpm)			

### 3. Result and discussion

#### 3.1. Microcapsule's characteristics

The character of microcapsules is important to perform as smart material in self-healing mechanisms. The size, shell thickness, shell structure, shape, and core content affect the suitability of embedding microcapsules in the coating matrix. As shown in Fig. 2 and described in Table 2, the microcapsules synthesized under different in-situ polymerization parameters displayed various surface morphology and conditions. FESEM results at different stirring speed showed in Fig. 2a for 200 rpm, Fig. 2b for 300 rpm and Fig. 2c for 400 rpm. The results concluded that the mean diameter of microcapsules was in the range of 2.36–3.71  $\mu\text{m}$ . The results on the diameter range of the microcapsules were compared with Boura et al. (2012) produced microcapsules, which ranged in size from 750 nm to 6  $\mu\text{m}$  [31]. Whereas the shell thickness of microcapsules was in the range of 0.04–0.43  $\mu\text{m}$ . All samples displayed the spherical shape of microcapsules with smooth shell structure [11]. The balancing effect of the ideal stirring speed that works together with the ideal concentration of surfactant was the factor to generate a smooth shell structure [32]. The samples were found agglomerated UF as a shell is classified as an adhesive material that is supposed to attach to the micelle of HLWSO during wall formation. The attachment

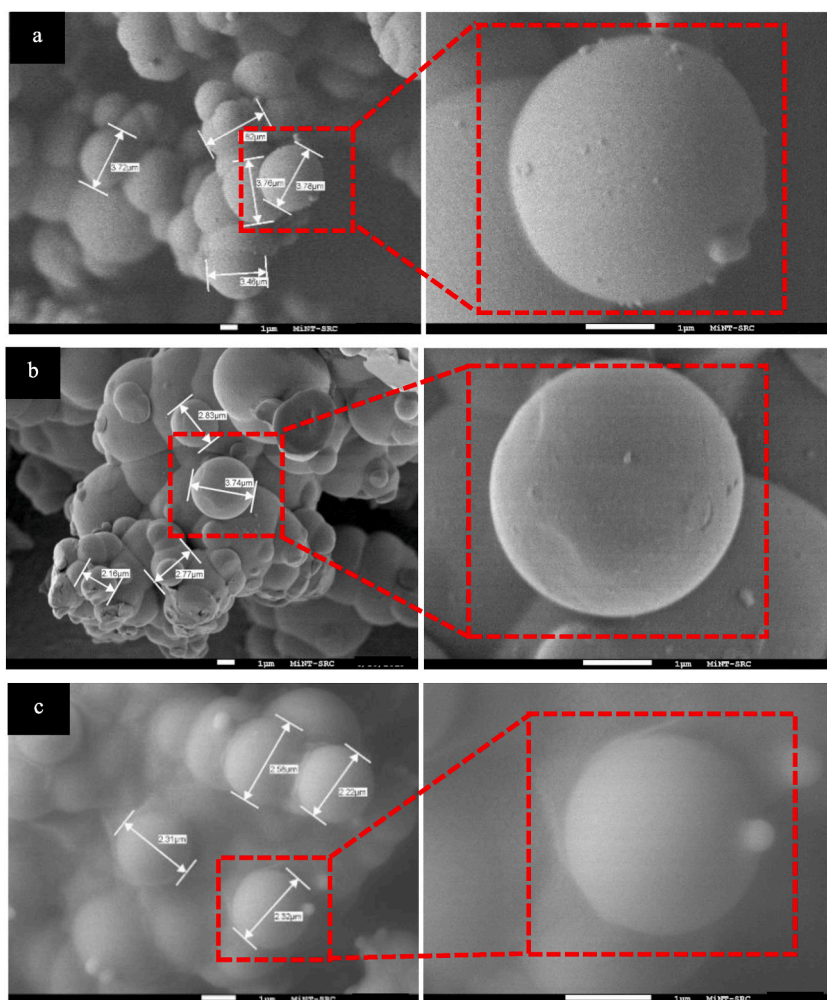


Fig. 2. The FESEM surface morphology of microcapsules at different stirring speed; (a) 200 rpm, (b) 300 rpm and (c) 400 rpm.

**Table 2**

The surface morphology and conditions of microcapsules.

No	Sample	Mean diameter ( $\mu\text{m}$ )	Shell thickness ( $\mu\text{m}$ )	Core content (%)	Shape	Shell structure	Microcapsule's condition
1	S1	3.71	0.43	69	Spherical	Smooth	Agglomerated
2	S2	2.88	0.31	66	Spherical	Smooth	Agglomerated
3	S3	2.36	0.04	64	Spherical	Smooth	Agglomerated



process was possibly interrupted by the adhesive structure of UF. The stirring force has induced the collision of microcapsules triggering the agglomeration phenomenon [19,30,31]. Sondari et al. (2010) stated that the agglomeration of microcapsules might be caused by a reaction with circulated air during the vacuum filtration procedure [34].

### 3.2. Emulsification process of HLWSO by anionic surfactant

Ethylene maleic anhydride (EMA) was observed as a suitable emulsifier and surfactant incorporating with new self-healing agent of HLWSO (Fig. 3). The process began with EMA being attracted to surround the HLWSO droplets by immersing its hydrophobic tail. The EMA acted as an emulsifier, stretching the HLWSO droplet. The HLWSO droplet was divided into two smaller, separated droplets. Then, EMA was employed as a surfactant with a hydrophilic head, reducing the surface tension in the water phase to shape the HLWSO droplet into a spherical micelle. Finally, the micelle carrying negative charges attracted urea-formaldehyde (UF) to bind the micelle to generate microcapsules.

### 3.3. Microencapsulation of HLWSO

The investigation on modulating recycling waste vegetable oil as a self-healing agent on microencapsulation of HLWSO involved several steps (Fig. 4). HLWSO acted as a self-healing agent via resorcinol, which carried negative charges pulled by a positive charge from emulsified HLWSO. The preferable pH on the cross-linked HLWSO and resorcinol was in the range of pH 3–4; in this research, pH

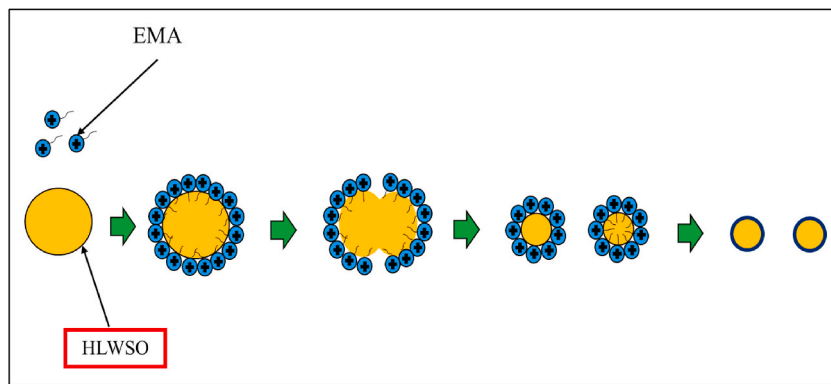


Fig. 3. Emulsification of high linoleic waste sunflower oil, HLWSO by ethylene maleic anhydride, EMA.

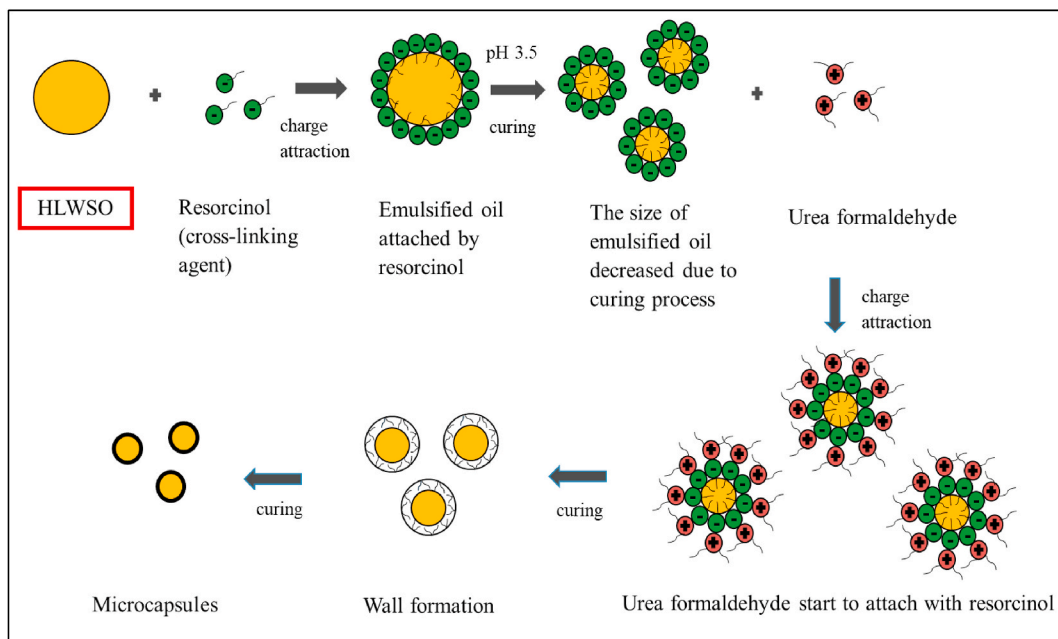


Fig. 4. Microencapsulation process flow with HLWSO as core and UF as a shell.

3.5 was chosen as a fixed parameter. The UF mixture was added after that, carrying positive charges attracted to the negative charges from resorcinol binding the emulsified HLWSO. This existing region generated a wall formation concept of microencapsulation. The curing process was performed twice in this wall formation region until microcapsules were fabricated.

### 3.4. The effect of stirring speed on microcapsules

Increased stirring speed decreased the core content, diameter and shell thickness of the microcapsules (Fig. 5) [14,24,33]. The higher stirring speed induced more collisions with the emulsified HLWSO, thus generating smaller droplets and lesser core content (Fig. 5a). The diameter of microcapsules was simultaneously depending on the size of emulsified oil that representing core content. Due to that, the higher the stirring speed was resulted smaller diameter of microcapsules (Fig. 5b). The increment of stirring speed was relatively induced wide range of shell thickness that were reduced from 1.43 to 0.04  $\mu\text{m}$ . The higher the stirring speed was promoted the higher restriction of UF to attach on the wall of emulsified HLWSO to form shell (Fig. 5c). The increment of stirring speed was also reduced the core content, but able to maintain in the range of 64–69%. (Fig. 5c). The core content percentage found indicated that the HLWSO as a self-healing agent was viable to be encapsulated and most importantly, the amount of core content was found to be adequate to be unleashed during the crack of microcapsules. The unleashed core content was needed to cover micro-cracks in the coating matrix [15,34].

### 3.5. The presence of core and shell of a microcapsule

The core and shell of microcapsules were identified by disclosing their functional group under FTIR. Fig. 6 indicates the pattern of microcapsule from samples at 200 rpm, 300 rpm, and 400 rpm. The FTIR pattern demonstrated the main peaks of HLWSO presence at

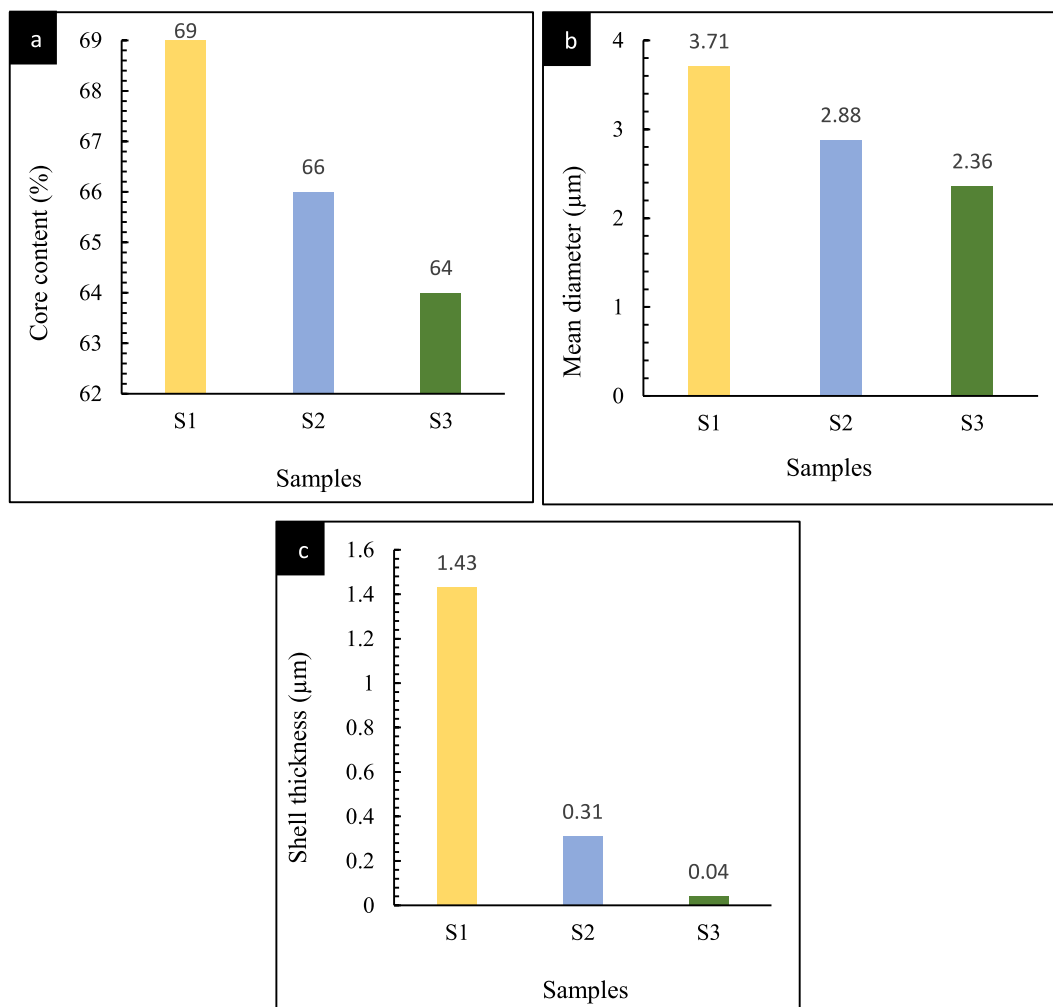


Fig. 5. The effect of stirring speed on microcapsules; (a) core content, (b) mean diameter and (c) shell thickness.

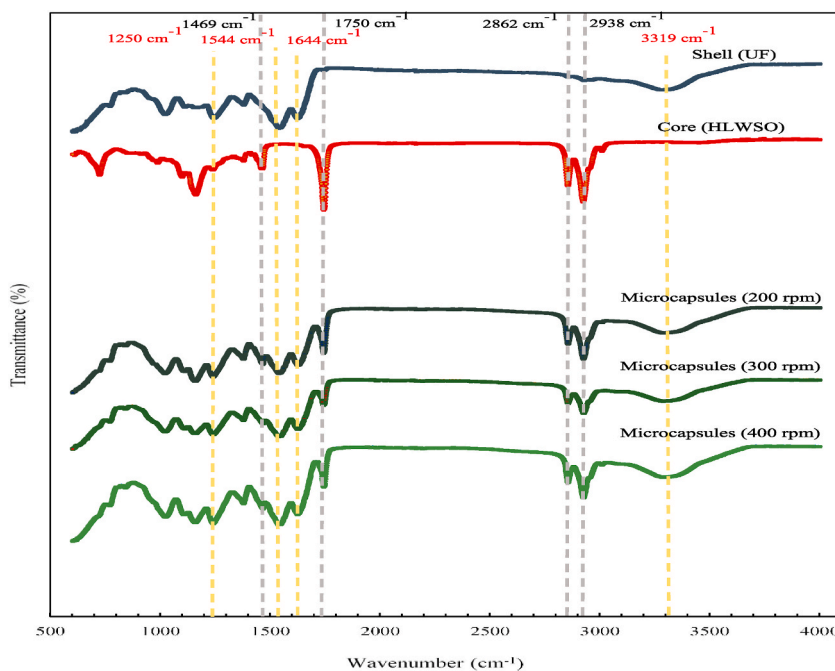


Fig. 6. The FTIR results on microcapsules containing HLWSO at different stirring speed (200, 300 and 400 rpm).

2938  $\text{cm}^{-1}$  and 2862  $\text{cm}^{-1}$  (C–H stretching vibrations), 1750  $\text{cm}^{-1}$  (C=O stretching absorption), and 1469  $\text{cm}^{-1}$  (C–C stretching vibrations) that also appeared on every microcapsule. This result conveys HLWSO as a core of microcapsules. Apart from that, the UF spectrum resulting from FTIR graph mapping revealed N–H, C=O and C–N stretching vibration at 1250  $\text{cm}^{-1}$ , 1544  $\text{cm}^{-1}$ , and 1644  $\text{cm}^{-1}$ , whereas, at 3319  $\text{cm}^{-1}$  peaks, N–H stretching absorptions were observed. The same stretching absorption peak also existed on the graph of microcapsules. This phenomenon proved that UF appeared as a shell of microcapsules on each variant parameter [2].

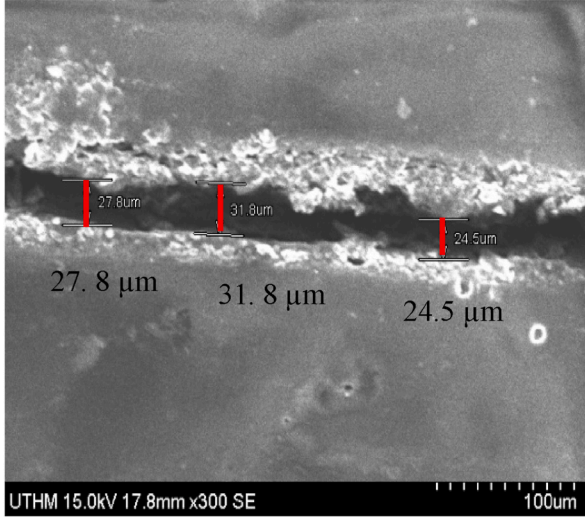
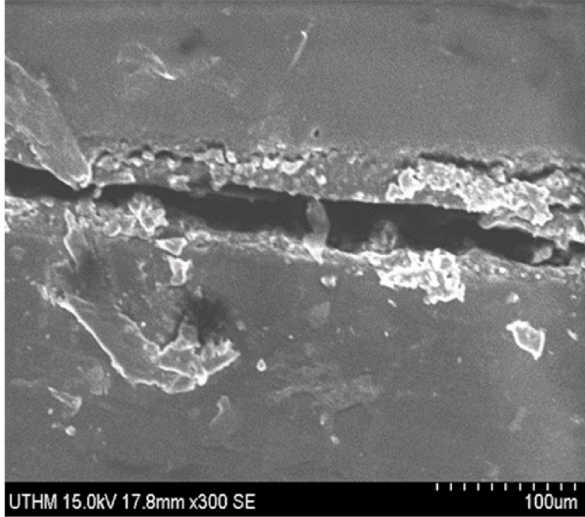
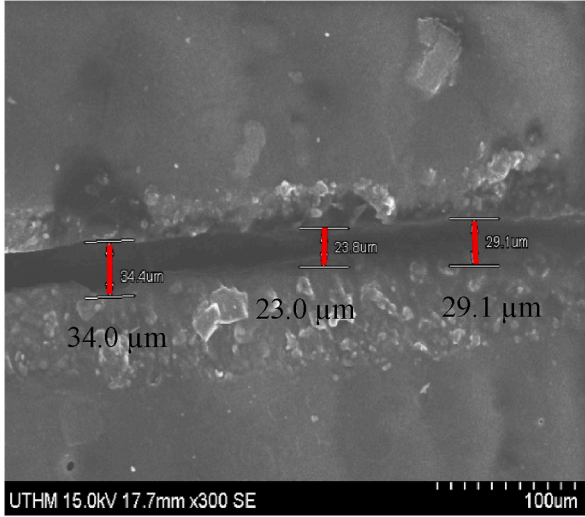
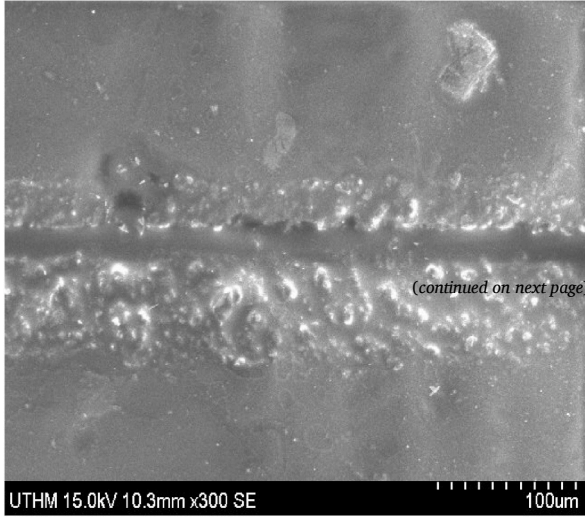
### 3.6. Self-healing performance

The self-healing performance of the studied coating sample by the layering method is shown by SEM analysis. Table 3 illustrates the self-healing performance of coating samples. The coating system of the reference sample displayed microstructure with unhealed and uneven fractured surface with signs of crack deviation on the 5th day. The samples illustrated the same lengths of fractured zones from day 1 until day 5, showing that no healing action occurred on the reference sample due to the absence of microcapsules embedded in Ref. [9]. Meanwhile, SC1, SC2 and SC3 were healing. A self-healing action occurred in the sample as the reactive healing mechanism from the microencapsulated HLWSO was unleashed by capillary action and started to fill the scratch zone [13]. The embedment of microcapsules in the coating matrix using the sandwich method generated a self-healing mechanism in the coating system. The coating matrix based on multilayering systems and induced with a green inhibitor produced a better coating system, which would be beneficial to the industry [37]. The embedment of microcapsules containing HLWSO as a drying oil successfully generated protection of the coating substrate via self-healing performance on scratches, in addition to general anti-corrosive properties [38].

### 3.7. Self-healing mechanism

The mechanism behind the self-healing of the scratched zone is illustrated in Fig. 7. The microcapsules were ruptured from the existence of the scratch, unleashing the HLWSO that filled the scratch and then polymerized, thus fully healing the scratch. The microcapsules were layered in an epoxy coating and located in the second layer of the coating system for all SC1, SC2 and SC3. The location of microcapsules in the middle of layer 1 and layer 2 were called ‘trap state’. The diameter range of microcapsules were 2.36 (S3), 2.88 (S2) and 3.71  $\mu\text{m}$  (S3) and the scratch width created was between 20 and 30  $\mu\text{m}$ . Since the core content of 64–69% and the number of microcapsules present in the coating matrix were uncountable, the unleashed HLWSO was highly able to fully cover the scratched zone [2,9]. This phenomenon highlights the benefits of embedding microcapsules into coating systems through stacking. The coating system’s ‘‘trap state’’ of the microcapsules has decreased the condition of scattered microcapsules under standard embedment (stirring method). In addition, the ‘‘trap state’’ of microcapsules was able to disregard the impact of microcapsule size because of the narrow range of sizes (S1, S2 and S3). Due to the ‘‘trap state’’ in the second layer (layer 2), the scratch will be able to penetrate and break microcapsules of various sizes (S1, S2, and S3), releasing the HLWSO that filled the scratch and performing self-healing for all SC1, SC2 and SC3.

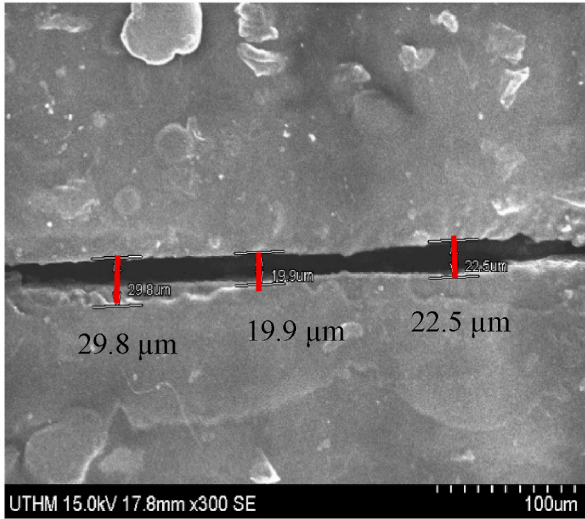
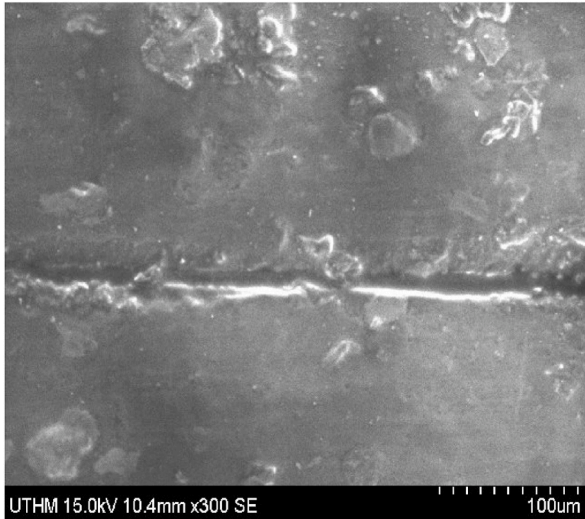
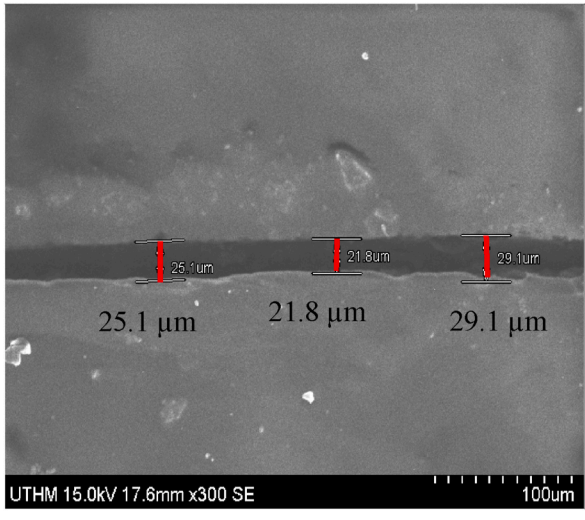
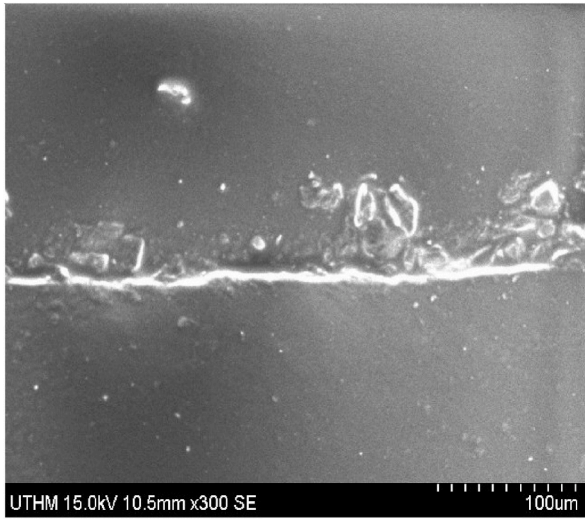
**Table 3**  
Self-healing performance of coating samples.

Sample	Day 1	Day 5
Reference sample		
SC1		

8



Table 3 (continued)

Sample	Day 1	Day 5
SC2		
SC3		



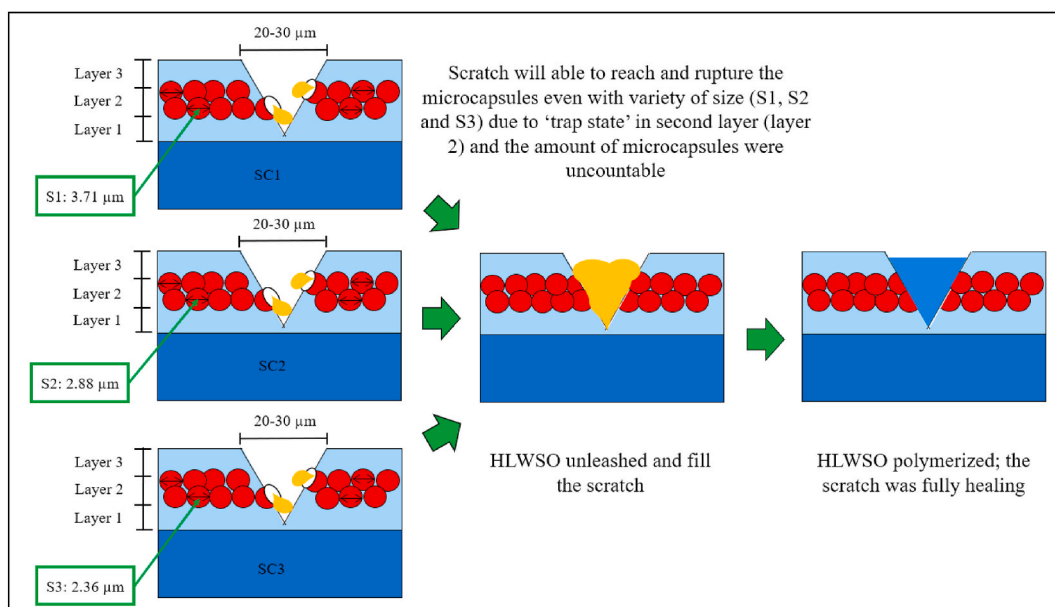


Fig. 7. Self-healing mechanism from different size of ruptured microcapsules (S1, S2 and S3) unleashing HLWSO to fully healed the scratched.

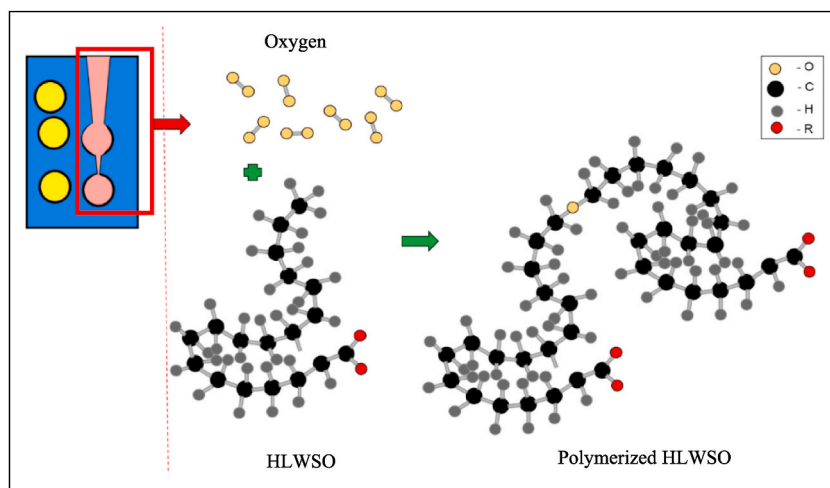


Fig. 8. Polymerization mechanism of HLWSO.

### 3.8. Polymerization mechanism of HLWSO

The polymerization mechanism of HLWSO was important in this research to identify the drying ability of the oil as a new self-healing agent. The distinctive properties of HLWSO introduced in this study were emphasized by the diene structure of linoleic acids. The diene structure derived from polyunsaturated drying oil like linoleic acids was active to create a reaction with oxygen. Monounsaturated fatty acids like oleic acids are less stable to generate reactions with oxygen to dry [39]. HLWSO as a self-healing agent has a high concentration of polyunsaturated fatty acids (linoleic) at 58%. The highest concentration of linoleic acid generates a high drying ability. Fig. 8 demonstrates that oxygen in the air reacted with carbon-oxygen bonds of unsaturated fatty acids to form hydroperoxides. The hydroperoxides then react with neighboring unsaturated fatty acids to form a polymer network as a thin layer through the process known as polymerization [40].

### 3.9. Anti-corrosion evaluation

Anti-corrosion evaluation persuades the anti-corrosion ability of smart coating containing HLWSO. Table 4 shows the mass loss and corrosion rate of samples after immersion in 3.5 wt%. NaCl for 28 days. The mass loss and corrosion rate of the reference sample were

**Table 4**  
Mass loss and corrosion rate.

No	Sample of coating system	Mass loss (g)	Corrosion rate (mm/yr)
1	Reference sample	0.04800	0.1356
2	SC1	0.02450	0.0682
3	SC2	0.02062	0.0574
4	SC3	0.03387	0.0943

0.04800 g and 0.1356 mm/yr. The reference sample had an optimum corrosion rate since there was no barrier to cover the scratch area [9,39]. The mass loss of SC1, SC2, and SC3 were 0.02450, 0.02062 and 0.03387 g and corrosion rate 0.0682, 0.0574 and 0.0943 mm/yr. The corrosion rate of the studied samples showed an improvement from the reference sample with reductions of 26–58%. Meanwhile, the SC2 showed an optimum result of anti-corrosion ability with the lowest mass loss and corrosion rate. This proved that samples embedded with microcapsules containing HLWSO induced anti-corrosion ability. The corrosion rate increased from the continuous immersion of the coating system in corrosive media (NaCl). Since the reference sample has no corrosion barrier, the intact corrosive media was higher than the studied samples [42]. In this study, SC2 had the best anti-corrosion performance, the microcapsules' 2.88  $\mu\text{m}$  diameter and 0.31  $\mu\text{m}$  shell thickness (percentage of shell: diameter was 10.8%) displayed the appropriate embedding characteristics to protect the microcapsules' ability to perform anti-corrosion. Additionally, the "trap state" that permits microcapsules to be disseminated uniformly also aids the ideal character microcapsules in providing an optimal anti-corrosion ability.

#### 4. Conclusion

The microencapsulation of HLWSO (Sample 2-S2) produced an optimum microcapsule diameter range of 2.88  $\mu\text{m}$ , 0.31  $\mu\text{m}$  shell thickness, and 66% core content. Further, increased stirring speed decreased the size of microcapsules, shell thickness, and core content. The FTIR analysis on microcapsules demonstrated the main peaks at 2938  $\text{cm}^{-1}$  (presence of HLWSO as a core) and 3319  $\text{cm}^{-1}$  (presence of urea-formaldehyde as a shell). The scratch on the coating matrix embedded with HLWSO was healed after five days with a reduced corrosion rate. The mass loss of SC2 (coating embedded with 300 rpm microcapsules) was 0.02062 g with corrosion rate of 0.0574 mm/year with an optimum reduction of 58% from the reference sample. The smart coating embedded with microcapsules containing HLWSO healed the scratch and offered anti-corrosion ability. To date, HLWSO as a new alternative self-healing agent showed polymerization ability due to high concentration of linoleic fatty acids that were constructed with diene structure. Microencapsulation of HLWSO with an ionic surfactant of ethylene maleic anhydride (EMA) has performed optimum formation of microcapsules. HLWSO as new self-healing agent also has attracted interest in optimizing recycled material. Future investigations should consider this smart coating technique with natural and environmental sources, which minimizes cost. Additionally, HLWSO is also a safe material that can improve sustainability by protecting metal while preventing pollution.

#### Credit author statement

Z. Baharom<sup>a</sup>, H.Z. Abdullah<sup>b</sup>, M.I. Idris<sup>c</sup> and Z.M.M. Ismail<sup>d,\*</sup>

a Wrote the paper, Original draft preparation, Software, Reviewing and Editing, Analysed and interpreted the data and literatures

b Reviewing and Editing

c Reviewing and Editing

d Supervision, Conceptualization, Reviewing and Editing.

#### 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.

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