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Development of a Smart Textile with Medicinal Properties Using Microencapsulated Camphor, Turpentine and Coconut Oils

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Abstract:
A mixture of oils containing 13%(v/v) of camphor oil, 43.5% (v/v) of turpentine oil and 43.5% (v/v) of coconut oil (CTC oil) was selected for this research due to the valuable medicinal properties of its ingredients such as high antioxidant capacity. Microencapsulated CTC oil was then used to develop a smart cotton fabric with several smart activities. CTC oil microcapsules were synthesized by the complex coacervation method using gelatin and sodium alginate as wall materials. UV visible spectrometry confirmed the encapsulation of CTC oil in microcapsules. Morphology of the CTC microcapsules appeared to be irregular in shape under optical and scanning electron microscopic images and was sized between 20–400 nm. The loading of the microcapsules was found to be 238 µL/g and the loading efficiency was 19%. The antioxidant activity of CTC oil microcapsules was 18±(0) µg PGE /mg in the Folin Ciocalteu assay. The CTC oil microcapsules displayed lower cytotoxic activity compared to the unencapsulated oil. The CTC microcapsules showed considerable repellency toward Aedes aegypti mosquitoes. The stability investigations of microcapsules indicated their stability under varying light conditions, selected surfactants and within the pH range of 5-10. The CTC microcapsules were incorporated onto cotton fabrics using the pad dry cure method and a dip coating method using succinic acid as the binder. The SEM images of cotton fabrics developed under both conditions confirmed the attachment of microcapsules on to cotton fibers. The CTC microcapsule incorporated cotton fabrics displayed significant antioxidant activity and mosquito repellent activity. The cotton fabric developed by the dip coating method using succinic acid binder displayed significant antioxidant activity whereas the fabric prepared by pad dry cure method resulted the best mosquito repellent cotton fabric. The antioxidant activity and the repellent activity of cotton fabrics still retained up to a considerable extent even after subjecting to a mild washing cycle.

Keywords: *Microencapsulation, camphor oil, turpentine oil, smart textile, antioxidant activity, mosquito repellency*

1. Introduction

In the present, people are trying to make the industrial and other processes more greener and cleaner *via* different pathways and one of them is the development of smart textiles.(Shrimali & Dedhia, 2015) Smart textile is a textile that can sense and respond in a controlled manner to environmental stimuli.(Tang & Stylios, 2001) Smart textiles are being utilized in many fields such as pharmaceutics, military forces, food industry and pest control.(Anitha, Ramachandran, Rajendran, & Mahalakshmi, 2011; Nelson, 2014; Ocepek & Forte-tav, 2002) Use of microncapsulated bioactive ingredients to develop various types of smart textiles with beneficial properties has been previously reported. (Dubey, Shami, & Rao, 2009a)(Alonso et al., 2013)

Microencapsulation of such ingredients helps to reduce oxidative degradation, mask the undesirable smells and flavors and allow controlled release of the encapsulated ingredients. (A & Jyothika, 2015; Bakry et al., 2016; Dubey et al., 2009a; Dubey, Shami, & Rao, 2009b; Jyothi et al., 2010; Xiao, Liu, Zhu, Zhou, & Niu, 2013)Microencapsulation also help to improve the shelf life of unstable bioactive ingredients including drugs (e.g. Vanillin).(Gumi, Gascon, Torras, & Garcia, 2009; Martins & Rodrigues, 2014).

During microencapsulation, a single or a mixture of active materials is coated by a coating material.(Martins, Barreiro, Coelho, & Rodrigues, 2014; Pakzad, Alemzadeh, & Kazemi, 2013; Sovilj & Katona, 2010)Controlled release activity of microcapsules is especially useful when delivering various compounds including drugs, pesticides, fragrances, etc. to specific targets at suitable rates with improved efficacy, safety and convenience.(Martins et al., 2014)The release of the encapsulated core material can occur via deterioration of capsule wall by physical or chemical means or by diffusion of core ingredients through the polymer wall.(Cheng, Yuen, Kan, & Cheuk, 2008; Keyan, Ramachandran, Shumugasundaram, Balasubramaniam, & Ragavendra, 2012; Nack, 2000; Souto, Fangueiro, & Zille, 2014)The controlled release character of the microcapsules depends on the structure, particle size, loading, degree of cross linking on capsule wall and dispersing medium.(Dong et al., 2011)

During this research, a mixture of camphor (13%), turpentine oil (43.5%) and coconut oil (43.5%) (CTC oil) was encapsulated using gelatin and sodium alginate wall materials via the complex coacervation method. This is a simple and inexpensive phase separation process that can be used to synthesize microcapsules. (Arshady, 1990; Burgess & Carless, 1985; Jun-xia, Hai-yan, & Jian, 2011; Pakzad et al., 2013)

The CTC oil mixture was selected based on the well-known medicinal characteristics of its components. Camphor oil extracted from various parts of Cinnamomum camphora trees constituents of numerous natural products such as piene, camphene, camphora, barneol, terpene and small traces of safrole.(Joshi, Padalia, Bisht, & Mathela, 2009; Zuccarini, 2009) Camphor oil is known for its decongestant, antispasmodic, anesthetic, sedative, anti-inflammatory and insect repellent properties.(Ja et al., 2006; Liu et al., 2006) Additionally, camphor oil has an aroma which helps to repel insects such as flies, mosquitoes, beetles and bugs.(Liu et al., 2006) Turpentine oil is extracted from young shoots, flowers and fruits of the turpentine tree.(Musa, Tzakou, & Couladis, 2009) The major components of the turpentine oil was α-pinene,β-pinene, sabinene and terpen-4-ol (Musa et al., 2009) and this oil is commonly used in the pharmaceutical industry, perfume industry, food additives and insecticides.(Mercier, Prost, & Prost, 2009)(Mercier et al., 2009)(Lucia et al., 2007)

2. Materials and Methods

2.1. Materials

Camphor oil was purchased from Beam chemicals Pvt Ltd, Sri Lanka. Turpentine oil and coconut oil were purchased from Glorchem Enterprise Pvt Ltd, SriLanka. Solvents (commercial grade) and other chemicals (analytical grade) were obtained from the Department of Chemistry, University of Colombo, Sri Lanka. All the solvents were double distilled prior to use.

2.2. Preparation of CTC Oil Microcapsules via Complex Coacervation

A 8.0 mL of CTC oil mixture was mixed with 100 mL of 3% (w/v) gelatin solution followed by drop wise addition of 100 mL of 1% (w/v) sodium alginate solution while stirring. Thereafter, the pH of the mixture was set to 3.2 - 4.0 by adding 25% (V/V) glacial acetic acid drop wise. Then the mixture was stirred continuously for one hour at 600 rpm. The mixture was cooled to 5-10 ˚C and 3 - 5 mL of saturated $CaCl_2$ was added to it. The prepared microcapsules were filtered using suction filtration and washed with DCM to remove the remaining un encapsulated oil. The washed capsules were dried at 40˚C for one hour using an oven. The microcapsules were kept in the refrigerator until further use.

2.3. Verification of Encapsulation

Two portions of 0.05 g of washed microcapsules were used with DCM as mentioned in previously published protocols with slight modifications.(Rasanganie & Perera, 2018)(Wijesirigunawardana & Perera, 2018) The UV absorbances were measured at 245 nm. (The wavelength which gives the maximum absorbance for the CTC oil). A solution of gelatin and sodium alginate in DCM solvent was used as the blank. The prepared CTC oil microcapsules were observed under the optical microscope(A12.0907-B) (under ×40 magnification) and the scanning electron microscope (EVO 18Research-ZEISS) (under × 3.50 K magnification) to investigate their morphology.

2.4. Determination of the Loading and Loading Efficiency of CTC Oil Microcapsules

A concentration series of CTC oil (600 ppm, 800 ppm, 1000 ppm, 1200 ppm, 1400 ppm, 1600 ppm) was prepared using DCM. A standard curve was developed using the absorbance values of the concentration series at 245 nm. Then the previously published protocols were used with slight modifications to determine the loading ofmicrocapsules.(Wijesirigunawardana & Perera, 2018)(Rasanganie & Perera, 2018) Loading efficiency was calculated using the following equation.

$$\text{Loading efficiency} = \frac{\text{Total volume of encapsulated oil in microcapsules} \times 100\%}{\text{Total volume of oil used for microencapsulation}}$$

2.5. Investigation of Biological Activities of CTC Oil Microcapsules

2.5.1. Folin Ciocalteu Antioxidant Assay

Previously published protocols were used with slight modifications.(Rasanganie & Perera, 2018; Wijesirigunawardana & Perera, 2018)

2.5.2. Cytotoxic Activity – Brine Shrimp Lethality Assay

The percentage mortality was calculated using the following equation and the cytotoxic activity was investigated according to previously published protocols with slight modifications.(Rasanganie & Perera, 2018; Wijesirigunawardana & Perera, 2018)

$$\% \text{ mortality} = \frac{\text{Number of brine shrimps dead} \times 100\%}{\text{Total number of brine shrimps used}}$$

2.6. Investigation of the Stability of Synthesized Microcapsules under Different Conditions

2.6.1. Thermal Stability

Portions of 0.10 g of microcapsules were separately kept under 10 ˚C, 30 ˚C, 40 ˚C, and 45˚ C, 50 ˚C and 60 ˚C for 3 hours. After 3 hours, 2 mL of methanol was added to each sample and the supernatant was collected. The absorbance of these solution obtained from the uncrushed samples was recorded at 245 nm. Then, 2 mL of methanol was freshly added to each sample and the microcapsules were well crushed using a glass rod followed by sonication for 10 minutes. The absorbance of the solutions obtained from the crushed samples was also measured at 245 nm. Pure unencapsulated oil samples (an equivalent amount to that encapsulated in the microcapsules) were also subjected to the same temperature and were dissolved in 2 mL of methanol. The absorbance of these samples was also measured at 245 nm. The experiment was carried out in triplicate.

2.6.2. Light Stability

Three portions of 0.10 g of microcapsules were kept in the dark, under sunlight and under a lamp for 6 hours respectively. A volume of 2 mL of methanol was added to each sample and the absorbance of the uncrushed sample was measured at 245 nm. Then each sample was mechanically crushed 2 mL of methanol was added followed by sonication. The Folin Ciocalteu assay was carried out for all the samples and the UV absorbance at 245 nm was also recorded. The same steps were repeated with the pure oil (an equivalent to that encapsulated in 0.10 g microcapsules). The experiment was carried out in triplicate.

2.6.3. Stability against Selected Surfactants and Oxidizing Agents

A portion of 0.10 g of microcapsules was incubated with 1 mL of 5% H_2O_2, 5% Triton-X100, 5% Tween-80 and 5% SDS solutions separately for 3 hours at room temperature. Each mixture was centrifuged and the UV absorbance values of supernatant of uncrushed microcapsules were recorded at 245 nm. Then these microcapsules were crushed well and transferred into 2 mL of methanol followed by sonication for 10 minutes. The antioxidant capacity was observed using Folin Ciocalteu assay for each sample. Pure oil which is similar to the loading of 0.10 g of microcapsules was also added to the above surfactant solutions and antioxidant capacity of these solutions were measured after 3 hours incubation period. All the experiments were carried out in triplicate.

2.7. Mosquito Repellent Activity of CTC Oil Microcapsules

A set of Aedes aegypti mosquitoes (n =30) was placed inside a static air repellency apparatus(Navanjalee, Panagoda, & Perera, 2018).The two sides of the static air repellency apparatus was covered with a net and the samples were mounted on to these two sides. Control sample was placed on one side and the test sample was placed on the other side. After mounting the sample, it was kept to stabilize for 2 minutes and then the number of mosquitoe in each side was recorded. The mosquito repellency assay was carried out with intact microcapsules (0.30 g), crushed microcapsules (0.30 g), an eqiuvalent amount of core oil (to that encapsulated inside the used microcapsules; 57 µL) and a higher amount of core oil sample (2 mL). The number of mosquitoes in the repellent side (side where the sample was placed) and non repellent side (where control sample was placed) was recorded at 1 hour intervals for 6 hours. All the samples were tested in triplicates. The percentage mosquito repellencey was calculated using the following equation.

$$\text{Percentage spatial repellency (\%)} = \frac{(C - T)}{\text{Total}} \times 100\%$$

C – Number of mosquitoes in control side
T- Number of mosquitoes in test side
Total – Total number of mosquitoes

2.8. Incorporation of CTC Oil Microcapsules onto a Cotton Fabric

2.8.1 Pad Dry Cure Method

A portion of 0.10 g of microcapsules was mixed with 1.0 mL of distilled water to make a 10 % microcapsule sample. Then the fabric was immersed in this solution for 2-3 minutes and the treated fabric was squeezed using a roller. Thereafter, the fabric was dried at 70 ˚C for 3 minutes using the oven. After the preparation of microcapsules incorporated cotton fabric, one piece of it was washed under 500 rpm to investigate the tolerance of in the developed fabric towards the washing process. The SEM images of both unwashed and the washed fabric pieces were obtained to verify the adhesion of microcapsules.

Antioxidant activity and mosquito repellent activity of both unwashed and washed cotton fabric pieces obtained from the pad dry cure method was investigated according to previously stated protocols.

2.8.2. Use of Succinic Acid Binder

A portion of 0.3 g of microcapsules was mixed with 3.0 ml of saturated succinic acid and stirred overnight. Then the microcapsules were filtered and resuspended in 3 mL of distilled water to make 10 % microcapsule solution. A piece of fabric was dipped in this solution and then the cloth was air dried and ironed. The fabric was washed under 500 rpm to

investigate the stability of microcapsules towards washing process. Then the SEM images of both unwashed and washed fabric samples were observed to verify the presence of microcapsules in the fabric.

Antioxidant activity and mosquito repellent activity of both unwashed and washed cotton fabric pieces obtained from the dip coating method was investigated according to the previously stated protocols.

3. Results and Discussion

3.1. Synthesis and Characterization of CTC Oil Microcapsules

The CTC microcapsules used in this study was successfully synthesized using the complex coacervation method (Figure 1).

Figure 1: Gelatin-Sodium Alginate Based CTC Microcapsules Obtained From the Complex Coacervation Method

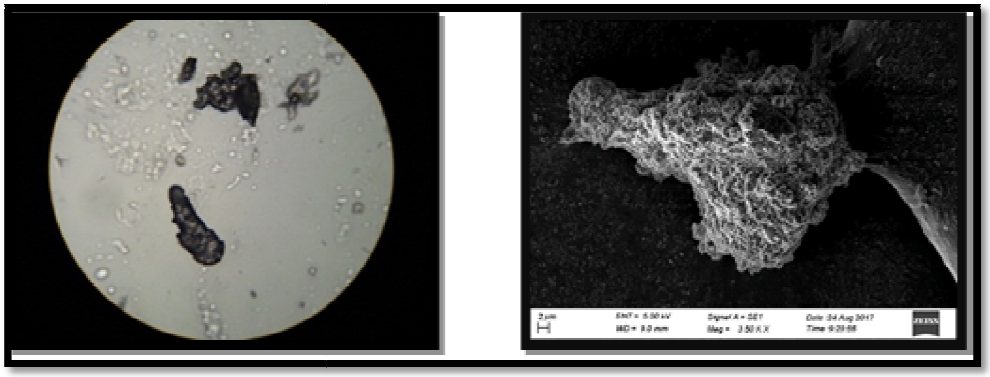

Figure 2: The Optical Microscopic Image of CTC Microcapsules (×10 Magnifications) The Scanning Electron Microscopic Image of CTC Microcapsules (×3.50 K Magnifications)

According to the optical and SEM images (Figure 2b), the CTC microcapsules were irregular in shape with rough walls. The size of the microcapsules varied between 20-400 µm.

The UV visible spectra were used to verify the encapsulation of CTC oil in the synthesized microcapsules. The maximum absorbance of the crushed and uncrushed samples was measured at 245 nm, which was the distinctive absorbance peak given by the CTC oil mixture (Supporting information Figure S1).

Test Sample	UV Absorbance Value
Uncrushed Sample	0.070 ±0.1
Crushed Sample	0.510±0.1

Table 1: UV Absorbance Values for Crushed and Uncrushed Samples at 245nm

As shown in Table 1, the absorbance for the mechanically crushed microcapsule sample was significantly higher than the absorbance obtained for the uncrushed microcapsule sample. Therefore, it can be concluded that the CTC oil

mixture was successfully encapsulated with gelatin and sodium alginate wall materials and this encapsulated oil was released upon mechanical crushing of microcapsules.

3.2. Loading of CTC Oil Microcapsules

Loading of the CTC oil microcapsules was determined using a standard curve of CTC oil (Supporting information Figure S2)and it was found to be 238 µL/g. The loading efficiency of the CTC microcapsules was found to be 19 %. (Supporting information Calculation S1 and S2)

3.3. Antioxidant Activity of CTC Oil Samples

According to the results shown in Table 2, unencapsulated CTC oil displayed a slightly higher AOC than the crushed microcapsule sample. However, it can be concluded that a significant amount of AOC of unencapsulated CTC oil was retained in the microcapsules after the encapsulation process.

Test sample	AOC (µg PGE/mg)
Unencapsulated CTC oil*	18±0
Crushed CTC microcapsules	11±0

Table 2: Antioxidant Capacity (AOC) of CTC Oil Samples
** Similar Equivalent to That Encapsulated in the Crushed Microcapsule Sample*

3.4. Cytotoxic Activity of CTC Oil Samples

The cytotoxic activity of CTC oil samples was monitored using the brine shrimp lethality assay as it corelate to the possible cytotoxic activity of test compounds towards humans.(Meyer et al., 2014; Mosmann, 1983)

According to the results shown in Figure 3(Supporting information Table S1), the percentage mortality was highest for the unencapsulated CTC oil and the crushed microcapsule sample. However, the percentage mortality of the intact microcapsule sample remained similar to the negative control did not exceed 50% even after 8 hours. These results clearly indicate that the cytotoxic activity of CTC oil can be successfully masked by microencapsulation process and the encapsulated oil can be released out upon the breakage of capsule wall. The percentage mortality of the intactmicrocapsules rose slightly faster than that of the negative control after the 5thhour and this could be due to the release of small amount of CTC oil from the microcapsules to the exterior solution via diffusion through microcapsule wall.

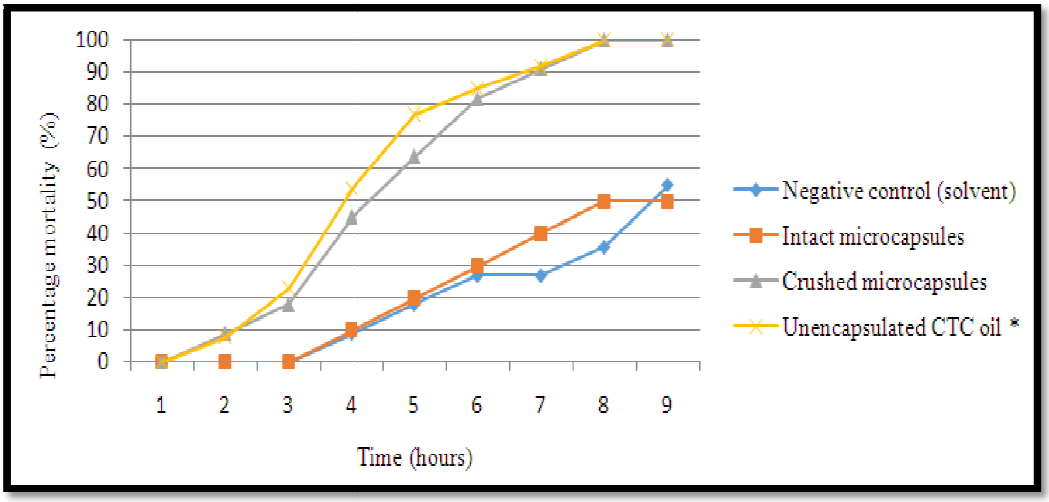

Figure 3: Percentage Mortality of Brine Shrimps against Time for Different CTC Oil Samples

unencapsulated CTC oil* - Similar equivalent to that encapsulated in the crushed microcapsule sample

3.5. Stability of CTC Oil Microcapsules under Different Conditions

3.5.1. Thermal Stability

The thermal stability of different CTC oil samples was investigated within the temperaturerange of 10˚C to 60˚C.The results obtained for the thermal stability studies are shown inFigure 4(Supporting information Table S2).

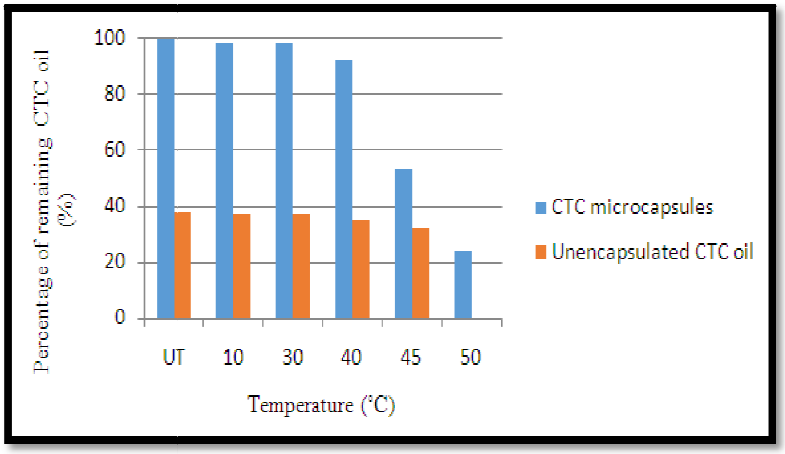

Figure 4: Percentage of Remaining CTC Oil after Thermal Treatment of Different CTC Oil Samples
UT = Untreated Sample

In the 10°C-40°C temperature range, there was no significant release of CTC oil from the microcapsules prior to crushing and 92%-98% core oil was released after crushing. This indicates the thermal stability of the capsule wall within this temperature range and the microencapsulation has also successfully reduced any evaporative loss of CTC oil as displayed by the unencapsulated oil sample. Even at 50°C, at least 24% retention of CTC oil was observed for the microcapsule sample where as the unencapsulated oil was totally lost at this temperature due to evaporation. These results indicate that microencapsulation of CTC oil can improve the thermal stability of CTC oil mixture to a significant degree.

3.5.2. Light Stability

Stability of CTC oil samples were investigated under different light conditions. According to the results in Figure5(a) (Supporting information Table S3), there was 87% and 84% of oil remaining in the CTC microcapsules subjected to artificial and sun light respectively. However, these values were only 78% and 75% for the unencapsulated oil samples. More importantly, the encapsulate CTC oil retained its AOC at a much higher level compared to that of unencapsulated CTC oil under all tested light conditions in Figure 5(b) (Supporting information Table S4). These results indicate the extra stability of encapsulated CTC oil under different light conditions.

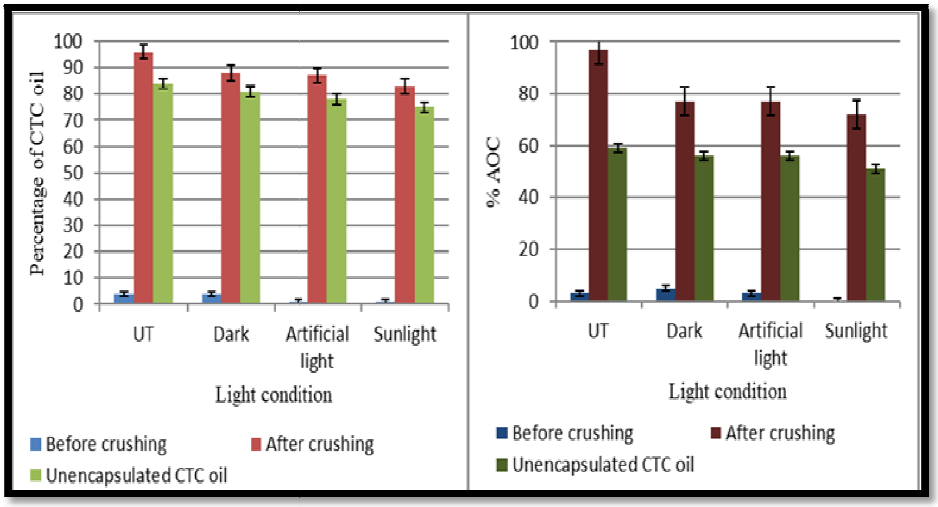

Figure 5: Percentage of Remaining CTC Oil and Percentage AOC (%AOC) of CTC Oil Samples Treated under Different Light Conditions
UT = Untreated Sample

3.5.3. Stability against Selected Surfactants and Oxidizing Agents

The AOCs of the CTC oil samples subjected to different surfactants and H_2O_2 oxidizing agent are shown in Figure 6. (Supporting information Table S5)

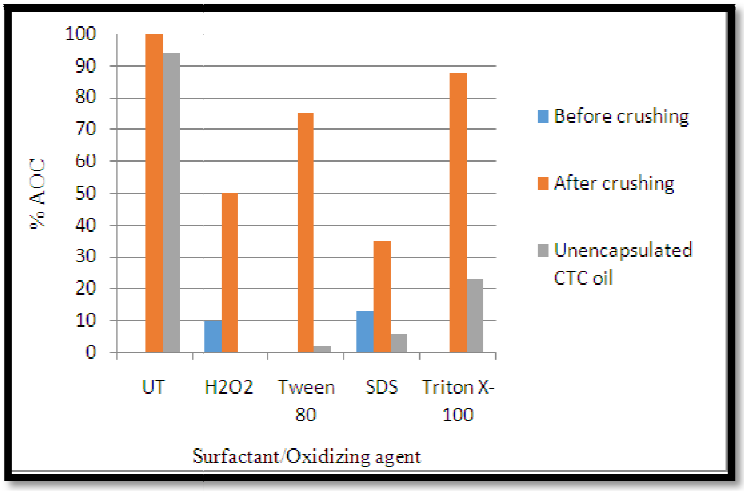

Figure 6: Percentage of AOC of CTC Oil Samples in the Presence of Different Surfactants and Oxidizing Agents
UT = Untreated Sample

The AOC of encapsulated CTC oil was significantly higher compared to that of the unencapsulated oil in the presence of all the tested surfactants and oxidizing agent (H_2O_2). The AOC of unencapsulated CTC oil was below 10% for all the surfactants and H_2O_2 oxidizing agent except for Triton X-100. However, CTC microcapsules displayed at least 50% or more AOC even after subjecting to the selected surfactants or H_2O_2, thus indicating the pH stability resulted due to microencapsulation process.

3.6. Mosquito Repellent Activity of CTC Oil Samples

The intense penetrating aroma of turpentine and camphor oils in the CTC oil mixture are known to repel small insects such as moths, bugs and mosquitoes.(Lucia et al., 2007)(Mercier et al., 2009)Mosquito repellent activity of microencapsulated and unencapsulated CTC oil samples was investigated using a static air repellency apparatus and the results are shown in Figure 7. (Supporting information Table S6)

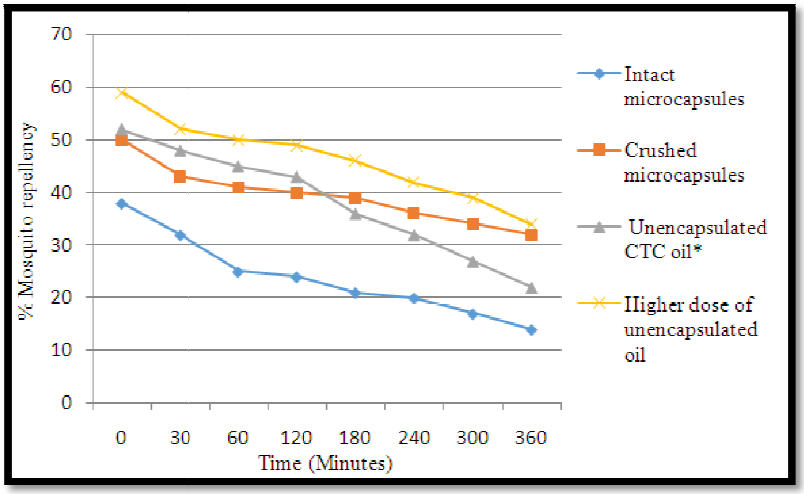

Figure 7: Change in Percentage Mosquito Repellency of Different CTC Oil Samples with Time
*Unencapsulated CTC Oil * - Equivalent Amount To That Encapsulated*

The mosquito repellent activity of all CTC oil samples diminishes with time as the intensity of its aroma is depleted. Throughout the test period, the intact microcapsules displayed a lower mosquito repellency compared to that of the crushed microcapsules. This indicates the ability of microcapsules to release its active ingredients under controlled conditions. Eventhough the mosquito repellent activity was similar for the crushed CTC microcapsulesandan equivalent amount of unencapsulated CTC oil, after 2 hours, the repellent ability of unencapsulated CTC oil was comparatively decreased than that of the crushed microcapsule sample. This result suggests the suitability of microencapsulation of CTC for prolonged repellent activity than the direct usage of this oil mixture. Furthermore, microencapsulation of the oil enables easier usage of oil on a surface such as a cloth compared to its direct application on the cloth that could be undesirable to the user.

3.7. Characterization of CTC Micro Capsule Incorporated Cotton Fabric

CTC oil microcapsules were incorporated into cotton fabrics using the pad dry cure method (no binder) and dip coating method (using succinic acid binder). The SEM images of the cotton fabrics prepared by both methods indicated successful adherence of CTC microcapsules onto cotton fibers (Figure 8 (a-d)).

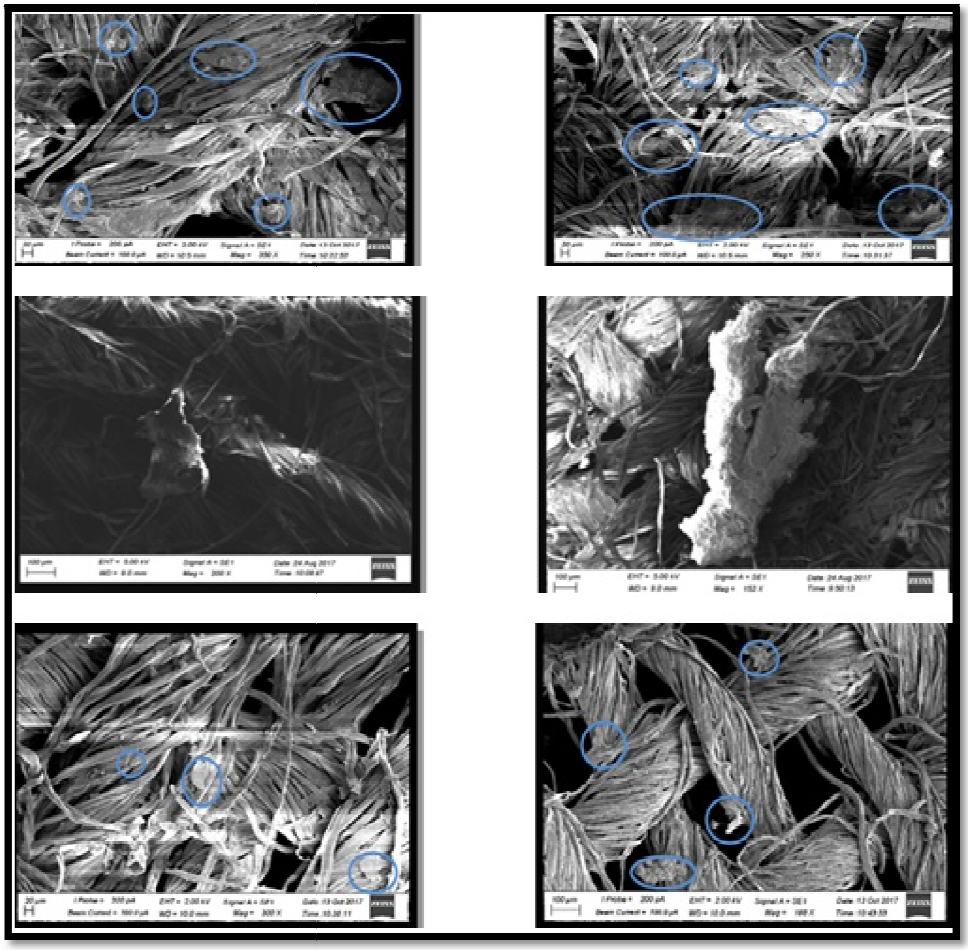

Figure 8: SEM Images of CTC Oil Microcapsules Incorporated Cotton Fabrics, Pad Dry Cure Method- before Washing (× 350 K Magnification), Pad Dry Cure Method- Before Washing (× 200 K Magnification), Pad Dry Cure Method-after Washing (×300 K Magnification), Dip Coating Method-before Washing (×250 K Magnification), Dip Coating Method-Before Washing (×152 K Magnification), Dip Coating Method-After Washing (×198K Magnification)

The SEM images also confirmed the retention of microcapsules on cotton fibers after subjecting to a mild washing cycle carried out simulating a washing machine spinning at a rotational speed of 500 rpm (Figure 8 (e-f). However, after the washing process, the number of microcapsules present in the cotton fabric was comparatively less than the number observed on the cotton fabric before washing. However, after the washing process, there were more microcapsules remaining on the cotton fabric developed using the succinic acid binder compared to that prepared by the pad dry cure method. Therefore, it can be concluded that the usage of a binder to attach the microcapsules can be more efficient for the incorporation of CTC microcapsules onto cotton fabric.

3.8. Antioxidant Activity of CTC Microcapsules Incorporated Cotton Fabrics

Due to the initial bioactivites observed in the CTC oil microcapsules, it was expected that the cotton fabrics developed using these micrcapsules should also displaythesebeneficial bioactivitiessuch as antioxidant activity and mosquito repellent activity. The AOC of CTC microcapsule incorporated cotton fabrics are shown in Figure 9.(Supporting information Table S7)

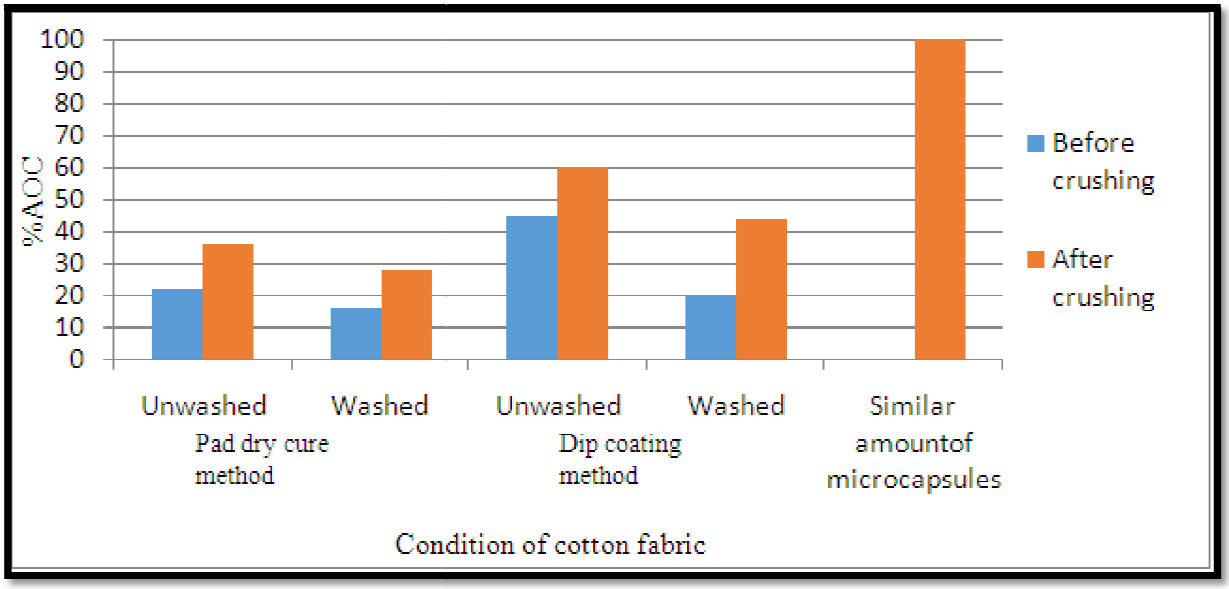

Figure 9: Percentage Each of Cotton Fabrics Pieces Developed under Different Conditions.

Both cotton fabrics developed by incrporating CTC microcapsules by the pad dry cure method as well as using succinic acid binder in the dip coating method displayed significant antioxidant activity. The highest AOC was given by the unwashed cotton fabric samples indicating the loss of some of the incorporated microcapsules during washing. However, the cotton fabrics developed using succinic acid binder displayed higher AOC compared to that of the cotton fabrics developed by the pad dry cure method. Therefore, it can be concluded that adhesion of microcapsules onto cotton fabric with a binder was more efficeient than the padding of microcapsules onto cotton fabrics.

3.9. Mosquito Repellent Activity of CTC Microcapsules Incorporated Cotton Fabric

Due to the high demand of consumer friendly and safer mosquito repellent methods, development of a fabric with mosquito repellent acitivty with a healthy oil mixture such as the CTC oil can be of great benefit to people.(Anuar & Yusof, 2016)

As the CTC microcapsules displayed considerable mosquito repellent activity, the cotton fabrics developed by incorporating these microcapsules were also tested for their ability to repel *Aedes aegypti* mosquitoes. The results obtained from this experiment are shown in Figure 10.(Supporting information Table S8) During this experiment, the fabric samples were hand squeezed at each time point to simulate the conditions of normal wearing of a dress by a person.

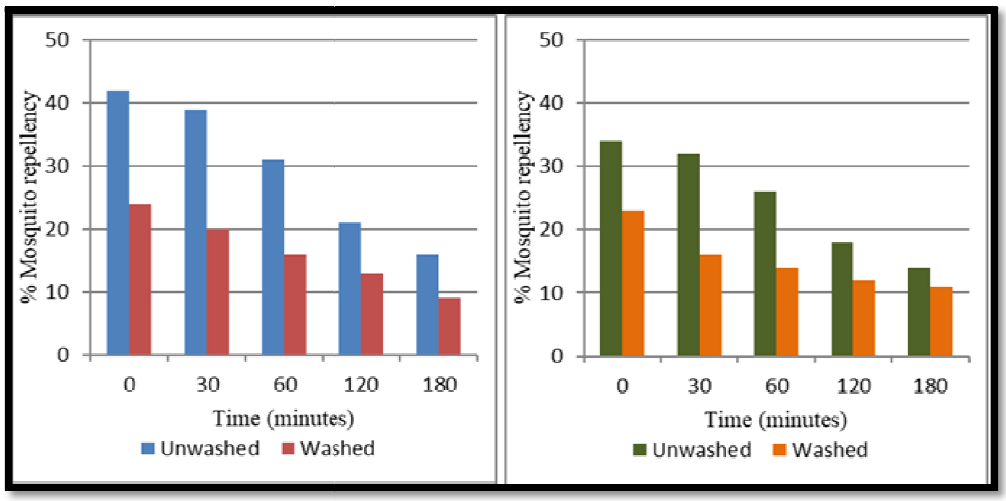

Figure 10: Percentage Mosquito Repellency of Cotton Fabric Samples Developed by Pad Dry Cure Method and Dip Coating Method

The best mosquito repellency of 42% was observed for the fabric prepared by the pad dry cure method using a 50% microcapsule solution. In contrast to the antioxidant activity, the cotton fabrics developed by the pad dry cure method displayed higher mosquito repellent activity compared to that displayed by the cotton fabrics developed using succinic acid binder. The presence of succinic acid binder on the microcapsule wall might have blocked the pores on the capsule wall, thus reducing the release of mosquito repellent actives to the exterior. However, the percentage loss of

microcapsules during the washing process could be decreased with the use of succinic acid binder. To improve the mosquito repellent activity of the CTC microcapsule containing cotton fabric, a higher concentration of microcapsules can be introduced on to the cotton fabric or microcasules with a higher loading of CTC oil can be used to develop the modified fabric. Additionally, different binders can be tested out to improve the binding of microcapsules to the cotton fibers.

4. Conclusion

CTC oil microcapsules can be successfully synthesized by the complex coacervation method using gelatin and sodium alginate as wall materials. The synthesized CTC oil microcapsules were irregular in shape and were sized between 20-400 m. CTC oil mixture had a milder smell than pure turpentine oil with a stronger aroma and microencapsulation further reduced this aroma. CTC oil microcapsules displayed significant antioxidant activity, mosquito repellent activity, thermal and light stability and stability against selected surfactants and hydrogen peroxide. Furthermore, the encapsulated CTC oil had lower cytotoxicity compared to the unencapsulated oil. The synthesized CTC microcapsules were successfully incorporated onto cotton fabrics using the pad dry cure method as well as by a dip coating method using succinic acid as the binder and the CTC microcapsule incorporated cotton fabrics displayed significant antioxidant activity and mosquito repellent activity.

5. References

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Appendix

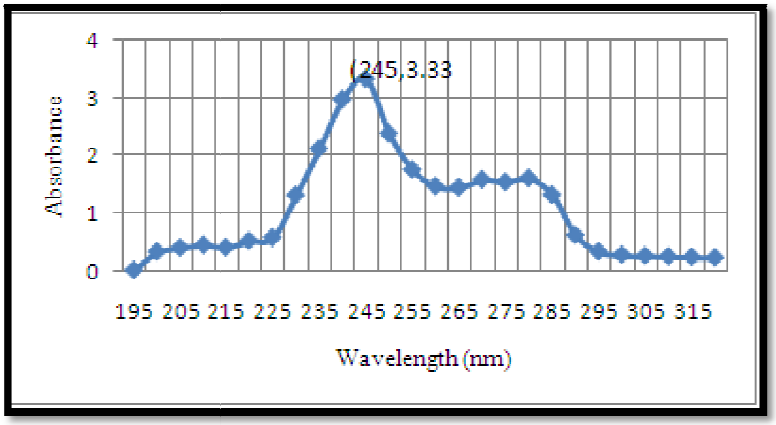

Figure 11: UV Absorbance Spectrum of CTC Oil Mixture

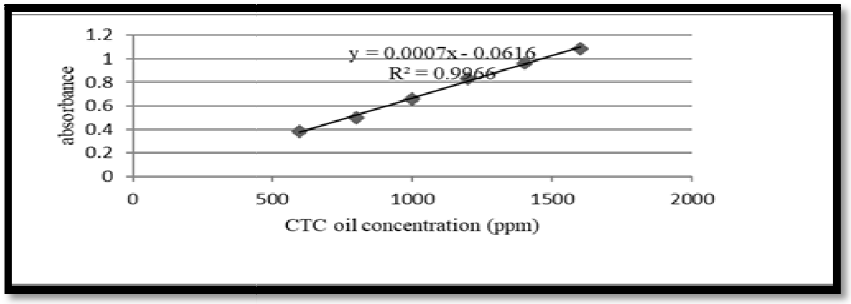

Figure 12: Standard Curve for CTC Oil

Calculation S1
Y=0.0007x-0.0616
0.510=0.0007x – 0.0616
x= 816.57 ppm
Dilution factor = 8
The initial concentration = 816.57 ppm × 8
= 6532.56 ppm
Hence, the loading of CTC oil in 1 g of microcapsules = 6532.56 µg/mL × 2 mL
0.0553 g × 0.992 g/mL
= 238 µL/g
Calculation S2
Total dry weight = 6.48 g
Amount of CTC oil used = 8mL
Total loading of CTC oil = 238 µL/g × 6.48 g
= 1542.24 µL
Loading efficiency = 1542.24 µL × 100%
8000 µL
= 19%

Time (hours)	Percentage Mortality of Brine Shrimp Nauplii (%)			
	Negative control (solvent)	Intact microcapsules	Crushed microcapsules	Unencapsulated CTC oil *
0	0±0	0±0	0±0	0±0
1	0±0	0±0	9±0	8±0
2	0±0	0±0	18±0	23±0
3	9±0	10±0	45±0	54±0
4	18±1	20±0	64±0	77±0
5	27±0	30±0	82±0	85±0
6	27±0	40±0	91±0	92±0
7	36±0	50±0	100±0	100±0
8	55±0	50±0	100±0	100±0

Table 3: Percentage Mortality of Brine Shrimp Nauplii in Different CTC Oil Samples

T (˚C)	Thermally Treated CTC Oil Microcapsules						Unencapsulated CTC oil	
	Before crushing		After crushing		Total conc. of CTC oil (µL/mL)	Total % of CTC oil	Conc. of CTC oil (µL/mL)	% CTC oil
	Conc. of CTC oil (µL/mL)	% CTC oil	Conc. of CTC oil (µL/mL	% CTC oil				
UT	0±0	0±0	2.19±0.00	100±0	2.19±0.00	100±0	0.84±0.01	38±1
10	0±0	0±0	2.15±0.00	98±0	2.15±0.00	98±0	0.81±0.01	37±1
30	0±0	0±0	2.14±0.00	98±0	2.14±0.00	98±0	0.80±0.00	37±0
40	0±0	0±0	2.02±0.01	92±1	2.02±0.01	92±1	0.76±0.00	35±0
45	0±0	0±0	1.15±0.01	53±1	1.15±0.01	53±1	0.71±0.00	32±0
50	0±0	0±0	0.52±0.01	24±1	0.52±0.01	24±1	0±0	0±0
60	0±0	0±0	0±0	0±0	0±0	0±0	0±0	0±0

Table 4: Concentration and Percentage of CTC Oil in Thermally Treated CTC Oil Samples
UT = Untreated

Light Conditions	Microencapsulated CTC Oil						Unencapsulated CTC Oil	
	Before crushing		After crushing		Total conc. of CTC oil (µL/mL)	Total % of CTC oil		
	Conc. of CTC oil (µL/mL)	% CTC oil	Conc. of CTC oil (µL/mL)	%CTC oil			Conc. of CTC oil (µL/mL)	% CTC oil
UT	0.13±0.00	4±0	2.97±0.02	96±1	3.10±0.02	100±1	2.61±0.02	84±1
Dark	0.11±0.01	4±0	2.73±0.03	88±1	2.84±0.03	92±1	2.50±0.02	81±1
Artificial light	0.02±0.00	1±0	2.69±0.04	87±1	2.71±0.04	88±1	2.43±0.01	78±1
Sunlight	0.02±0.00	1±0	2.58±0.04	83±1	2.60±0.04	84±1	2.33±0.01	75±1

Table 5: Concentration and Percentage CTC Oil Samples under Different Light Conditions.
UT = Untreated Sample

Light Conditions	Microcapsules in Different Light Conditions						Unencapsulated CTC Oil	
	Before crushing		After crushing		Total AOC (µg PGE/mg)	Total % AOC		
	AOC (µg PGE/mg)	% AOC	AOC (µg PGE/mg)	% AOC			AOC (µg PGE/mg)	%AOC
UT	1±0	3±0	38±0	97±0	39±0	100±0	23±0	59±0
Dark	2±0	5±0	30±0	77±0	32±0	82±0	22±0	56±0
Artificial light	1±0	3±0	30±0	77±0	31±0	80±0	22±0	56±0
Sunlight	0±0	0±0	28±0	72±0	28±0	72±0	20±0	51±0

Table 6: The AOC of CTC Oil Samples Treated under Different Light Conditions

Surfactant	CTC Oil Microcapsules in Different Surfactants						Unencapsulated CTC oil	
	Before crushing		After crushing		Total AOC (µg PGE/mg)	Total % AOC		
	AOC (µg PGE/mg)	% AOC	AOC (µg PGE/mg)	% AOC			AOC (µg PGE/mg)	%AOC
UT	0±0	0±0	48±0	100±0	48±0	100±0	45±0	94±0
H_2O_2	5±0	10±0	24±0	50±0	29±0	60±0	0±0	0±0
Tween 80	0±0	0±0	36±0	75±0	36±0	75±0	1±0	2±0
SDS	6±0	13±0	17±0	35±0	23±0	48±0	3±0	6±0
Triton X-100	0±0	0±0	42±0	88±0	42±0	88±0	11±0	23±0

Table 7: The AOC of CTC Oil Samples in the Presence of Different Surfactants and Oxidizing Agents
UT = Untreated Sample

Time (minutes)	% Repellency			
	Intact microcapsules	Crushed microcapsules *	Equivalent amount of unencapsulated oil	Higher dose of unencapsulated oil
0	38±1	50±1	52±1	59±2
30	32±2	43±1	48±0	52±1
60	25±0	41±1	45±1	50±2
120	24±2	40±1	43±2	49±1
180	21±1	39±1	36±1	46±1
240	20±0	36±2	32±1	42±1
300	17±1	34±1	27±1	39±1
360	14±1	32±2	22±2	34±2

Table 8: Percentage Mosquito Repellency for Different CTC Oil Samples within a 6 Hour Time Period
** The Microcapsule Sample Was Crushed Prior to the Each Trial*

Sample	%AOC				
	Pad dry cure method		Dip coating method		Similar equivalent of microcapsules
	Unwashed	Washed	Unwashed	Washed	
Before crushing	22±0	16±0	45±0	20±0	0±0
After crushing	36±0	28±0	60±0	44±0	100±0
Total	58±0	44±0	105±0	64±0	100±0

Table 9: The Percentage AOC of the Microcapsule Incorporated Cotton Fabrics Prepared Using 30% Microcapsule Solutions