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COTTON AND POLYESTER TREATED WITH TRAGACANTH GUM LOADED WITH CLOVE ESSENTIAL OIL NANOEMULSION FOR MOSQUITO REPELLENT

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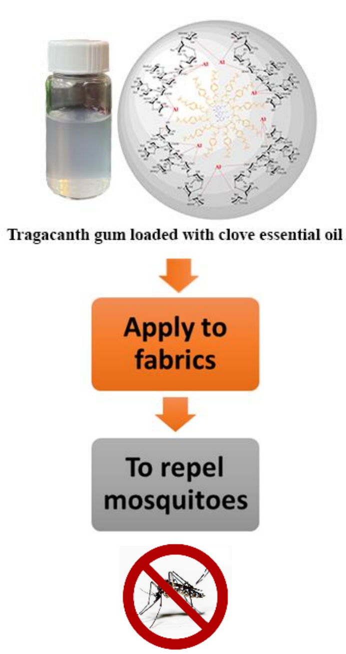
Graphical abstract



Abstract

Clove essential oil has been revealed to have a mosquito-repellent effect. Nevertheless, its application in topical preparations is minimal due to its immediate volatility. Nanoemulsion has been regarded as a promising carrier for essential oils. Here, we studied the effectiveness of cotton and polyester treated with tragacanth gum loaded with clove essential oil nanoemulsion to repel two mosquito species, namely *Aedes aegypti* (day biter) and *Anopheles latens* (night biter) for the first time. The nanoemulsion formulation was characterized using scanning electron microscopy, transmission electron microscopy, Fourier transform infrared spectroscopy, zeta potential and polydispersity index. Tragacanth gum loaded with clove essential oil nanoemulsion formulation exhibits 98% encapsulation efficiency with a -42.3 mV zeta potential and 0.3 polydispersity index. Following 5 cycles of washing, 60% of the tragacanth gum loaded with clove essential oil nanoemulsion retained on cotton meanwhile 46% of the nanoemulsion formulation retained on polyester. From an Excito chamber study, 64% of *Ae. aegypti* were successfully repelled from cotton whereas 53% of the same mosquito species were repelled from polyester. Overall, the nanoemulsion treatment was able to functionalize the fabrics up to a "very good" level of repellence against mosquitoes even after 5 cycles of washing.

Keywords: *Aedes aegypti*, clove essential oil, cotton, nanoemulsion, polyester

Abstrak

Minyak pati cengkih mempunyai sifat sebagai penghalau nyamuk. Namun begitu, penggunaannya dalam penyediaan topikal adalah minima disebabkan kadar kemeruapan minyak pati yang sangat cepat. Nanoemulsi dianggap sebagai medium pembawa yang paling baik untuk minyak pati cengkih. Di sini, pertama kalinya, kami menguji keberkesanan fabrik kapas dan poliester yang telah dirawat bersama formulasi nanoemulsi getah tragacanth yang mengandungi minyak pati cengkih untuk menghalau nyamuk spesis *Aedes aegypti* (aktif pada waktu siang) dan *Anopheles latens* (aktif pada waktu malam). Formulasi nanoemulsi ini menjalani process pencirian melalui Spektroskopi inframerah transformasi Fourier, potensi zeta dan indeks polidispersiti. Formulasi nanoemulsi getah tragacanth yang mengandungi minyak pati cengkih mempamerkan kecekapan enkapsulasi 98% dengan potensi zeta -42.3 mV dan indeks polidispersiti 0.3. Berdasarkan proses pembasuhan sebanyak 5 kitaran ke atas formulasi nanoemulsi getah tragacanth yang mengandungi minyak pati cengkih menunjukkan baki 60% formulasi nanoemulsi yang masih kekal pada fabrik kapas manakala baki 46% formulasi nanoemulsi kekal pada fabrik poliester. Berdasarkan eksperimen kadar penghalau nyamuk menggunakan ruangan Excito, fabrik kapas dan poliester yang telah dirawat dengan formulasi nanoemulsi, fabrik kapas tersebut berjaya menghalau 64% nyamuk *Ae. aegypti* manakala fabrik poliester pula menghalau sebanyak 53% nyamuk spesis yang sama. Secara keseluruhannya, rawatan formulasi nanoemulsi ke atas fabrik dapat meningkatkan keberkesanan fabrik sehingga tahap penghalauan "sangat baik" terhadap nyamuk walaupun selepas 5 kitaran pembasuhan.

Kata kunci: *Aedes aegypti,* minyak pati cengkih, kapas, nanoemulsi, poliester

1.0 INTRODUCTION

Mosquitoes are one of the life-threatening species on earth. Due to their high morbidity and mortality, dengue and malaria are the two major public health concerns [1]. According to the World Health Organization, it is estimated that about 100 to 400 million dengue infection cases are reported every year [2] and there were 241 million malaria cases in 2020 worldwide [3]. The European Centre for Disease Prevention and Control has reported that 37,950 and 68,908 dengue cases were recorded in Malaysia and Indonesia from January to September 2022, respectively [4]. As the growing number of dengue and malaria cases is proportionally correlated with mortality, the application of repellents to control mosquito vectors is on an upward trend.

Permethrin-based formulations are commonly used to repel insects, most likely mosquitoes [5]. However, long-term use of permethrin-based formulations causes adverse toxicity effects to human by inducing a decrease in the activity of the channels in the nervous system. These could affect behavioral performance and neurologic functions [6]. According to Sun *et al*. [7], a study on toxicity of permethrin to human organs has resulted in organ liver and kidney damages due to low dose long term exposure. Therefore, plant essential oils have been suggested as alternatives to permethrin. Despite their great ability in controlling mosquito, the active ingredients in essential oils are highly volatile [8].

Eugenol, a naturally occurring aromatic phenol essential oil derived from *Syzygium aromaticum* (cloves), has received much attention for its ability to repel and eliminate a variety of pest species [9,10]. Eugenol is the major active ingredient of clove essential oil. Synthetic repellents and plant-based repellents are comparable. Nevertheless, due to their volatility, essential oil repellents often have a limited lifespan in terms of efficacy. For instance, Eden *et al.* [11] conducted a study on an efficacy of citronella essential oil's larvicidal and repellent actions against mosquitoes. They found both effects endure for only two hours.

An effective release system is required to improve the efficacy of such active ingredients. In this context, nanoemulsions have received great attention from scientists mainly due to simple procedure of preparation, use small amount of surfactant and relatively cheaper as compared to other conventional preparation methods [12-14]. Wei *et al*. [15] have prepared nanoemulsion via low-energy emulsification method and used it to carry fenpropathrin. Meanwhile, Kawish *et al*. [16] have developed and evaluated docetaxel-nanoemulsion for improvement of therapeutic acitivity.

Recent studies have shown that biopolymer based nanoemulsions are able to enhance the effectiveness of various drug deliveries [17-20]. For example, a chitosan-based nanoemulsion was able to release triclabendazole over 6 hours of observation [21]. Tragacanth gum has several uses in healthcare provision, most notably for the administration of drugs. Tragacanth gum is frequently used as an emulsifier in the food, pharmaceutical, and related sectors because of its outstanding stability over a wide pH and temperature range and unusually extended shelf life [22,23].

Hence, we investigate the potential of tragacanth gum encapsulated with clove essential oil for the development of a nanoemulsion formulation suitable for use as mosquito repellent textile fabrics. This research entailed two innovative strategies, namely: (1) the encapsulation of clove essential oil into nanoemulsion via ultrasonic emulsification method and (2) the formation of protective-layer micelles for stabilisation of clove essential oil in nanoemulsion.

2.0 MATERIALS AND METHODS

2.1 Materials

Tragacanth gum and Triton X-100 surfactant were supplied by Sigma-Aldrich. Clove essential oil was obtained from Tradeserve Resources. Aluminium chloride and ethanol were purchased from Hmbg Reagents & Chemicals.

2.2 Preparation of Nanoemulsion

The nanoemulsion was prepared according to the procedure outlined by Ghayempour *et al*. [24], but with some modifications. The mixture of clove essential oil (0.1 mL) and Triton X-100 (1 mL) was stirred for 30 min at 900 rpm using a magnetic stirrer. Then, 15 mL of 0.25% w/v tragacanth gum and 0.1 mL of 0.25% w/v aluminium chloride were added to the mixture and stirred for another 30 min. The mixture was sonicated using an ultrasonic homogenizer set at 150 W and 24 kHz for 15 min.

2.3 Characterisation Studies

Fourier transform infrared spectra of raw materials and nanoemulsions were recorded using attenuated total reflectance (ATR) method on a Spectrum 3 PerkinElmer spectrometer. Fourier transform infrared analysis was conducted for 32 cumulative scans within a 4000 to 40 cm^{-1} wavenumber at 4 cm^{-1} resolution. The surface morphology of aggregates was observed using a Jeol JSM-IT200 scanning electron microscope. All fabrics sample were coated with platinum using a sputter coater before scanning electron microscope analysis began.

The internal morphology of the oil droplets in the nanoemulsion was observed using a LEO Libra-120 transmission electron microscope. The nanoemulsion formulation was dropped on a copper grid and left to air-dry before imaging. The stability of the tragacanth gum loaded with clove essential oil nanoemulsion was evaluated in terms of zeta potential, polydispersity index and particle size, which were based on dynamic scattering light technique using Malvern Zetasizer nano.

2.4 Encapsulation Efficiency

The encapsulation efficiency of nanoemulsion was determined using an ultrafiltration method. A cellulose ultrafiltration tube (10 kDA) containing nanoemulsion formulation was centrifuged at 200 rpm for 45 min. Since free clove essential oil can only infiltrate from the tube layer, the difference between the total amount of essential oil added to the nanoemulsion and the amount of free clove essential oil (unencapsulated) detected in the filtrate was used to calculate the quantity of essential oil that was encapsulated. The concentration of clove essential oil was determined using an Agilent Cary 60 Ultraviolet-Visible Spectrophotometer set at 282 nm. The encapsulation efficiency was calculated as Mahdi *et al*. [18]:

$$\text{Encapsulation efficiency (\%)} = \frac{\text{Amount of encapsulated clove essential oil}}{\text{Total amount clove essential oil}} \times 100$$

2.5 Applying Tragacanth Gum Loaded with Clove Essential Oil Nanoemulsion to Fabrics

The set of tragacanth gum loaded with clove essential oil nanoemulsion (2 mL) and clove essential oil (2 mL) were sprayed directly onto 6 x 6 cm of polyester and cotton fabrics using a trigger spray. The fabrics were dried at room temperature (25 ± 1 °C).

2.6 Wash Durability

Wash durability tests were performed at room temperature (25 ± 1 °C) using 5 cycles of washing. Specifically, treated fabrics were immersed in *Savon de Marseille* detergent solution (600 mL, 2.5 g/L) and stirred for 10 min at 200 rpm. The cotton and polyester fabrics were then air dried after being rinsed twice with deionised water.

2.7 Retention of Clove Essential Oil on Fabrics

The solvent extraction technique was used to study the removal and retention of deposited clove essential oil from the fabrics. The 1 x 1 cm of treated fabric was placed in a tube filled with ethanol. Then, the tube was vortexed for 5 min before sonicating for another 30 min, to extract the essential oil. The ultraviolet-visible spectrophotometer was used to measure the quantity of clove essential oil that was retained on the fabric. The percentage of clove essential oil retained on the fabrics was calculated using the following equation as Hebeish *et al*. [25]:

$$\text{Retention (\%)} = \frac{\text{Amount of clove essential oil after washing}}{\text{Amount of clove essential oil introduced to fabric}} \times 100$$

2.8 Mosquito Repellency Test

The Excito chamber method was employed to test the mosquito repellency as outlined by Xin and Wang [26]. Both *Aedes aegypti* and *Anopheles latens* mosquitoes were reared at the Faculty of Health Sciences, Universiti Kebangsaan Malaysia. The animal research ethics (Ref. No. 2021-0008-35) approval was obtained from the Research Ethics Committee of Universiti Pendidikan Sultan Idris.

In this Excito chamber method, tragacanth gum loaded with clove essential oil nanoemulsion was applied to fabrics before and after washing. For repellency test, 50 adult female mosquitoes were famished for 4 h. Then, the response of mosquitoes was monitored closely in relation of the quantity of mosquitoes stayed within the treated fabric chamber and the quantity of mosquitoes that fled to another space. The number of mosquitoes escaped from treated fabrics in the chamber was recorded at 5 min intervals within 30 min of total observation period. The mosquito repellency was calculated as Xin and Wang [26]:

$$\text{Mosquito repellency (\%)} = \frac{\text{Number of mosquito escaped}}{\text{Number of mosquito exposed}} \times 100$$

3.0 RESULTS AND DISCUSSION

3.1 Fourier Transform Infrared Spectroscopy Analysis

The Fourier transform infrared spectrum of tragacanth gum is shown in Figure 1(a), and consists of absorption bands indicative of stretching vibrations at wavenumbers of 3432 cm^{-1} (-OH), 2923 cm^{-1} (aliphatic $–CH_2$), 1800 cm^{-1} (C=O stretching of -COOH), 1630 cm^{-1} (carboxylate peaks) and 1027 cm^{-1} (stretching vibrations of C-O-C in the glycosidic group) [23,27]. Figure 1(b) depicts the spectrum of clove essential oil. Absorption bands at 3521 cm^{-1} (phenolic O-H), 2936 cm^{-1} (C-H in aromatic ring), 1638 cm^{-1} (C-H of benzene), 1376 cm^{-1} (C-H deformation of eugenol methyl), 1265 cm^{-1} (C-O of phenolic hydroxyl) and 1032 cm^{-1} (C-O-C of aromatic ether) represent eugenol [9,10].

The spectrum of the nanoemulsion is presented in Figure 1(c), which clearly shows that it possesses both functional group characteristics of tragacanth gum (Fig. 1a) and clove essential oil (Figure 1b). From Figure 1c, the appearance of peaks at 3370 cm^{-1}, 2918 cm^{-1}, 1640 cm^{-1} and 1513 cm^{-1} confirms the formation of tragacanth gum loaded with clove essential oil nanoemulsion. The aforementioned peaks were shifted from 3432 cm^{-1}, 2923 cm^{-1}, 1638 cm^{-1} and 1510 cm^{-1}, respectively. A new peak observed at 1111 cm^{-1} can be related to the presence of Al-O-C in the nanoemulsion. In this study, aluminium chloride was used as a linking agent for carboxylic groups of tragacanth gum.

In this nanoemulsion formulation, up to 90% of the colloidal system consists of aquous phase (tragacanth gum solution) and 10% of dispersed phase (surfactant with oil). Tragacanth gum was used as the main wall material to encapsulate the clove essential oil by ultrasonic emulsification.

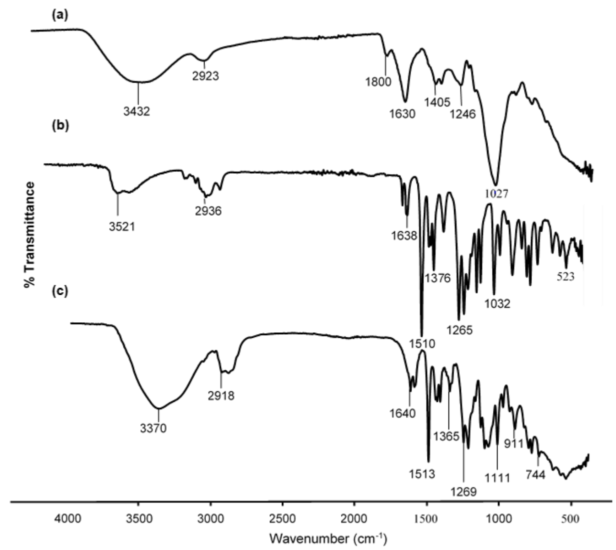


Figure 1 Fourier transform infrared spectra: (a) tragancanth gum, (b) clove essential oil, and (c) tragacanth gum loaded with clove essential oil nanoemulsion

Figure 2 summarises a schematic of tragacanth gum loaded with clove essential oil nanoemulsion formation. Triton X-100 surfactant possesses both hydrophobic and hydrophilic terminals. The hydrophobic terminals of Triton X-100 dissolve in oil, and the hydrophilic terminals dissolve in the aqueous phase (tragacanth gum solution), forming a dispersion of small clove essential oil droplets.

After incorporation of tragacanth gum solution into the mixture of Triton X-100 and clove essential oil create formation of wall, since carboxylic groups has a ability of bonding with other group. The hydrophilic nature of tragacanth gum (OH and COOH groups) interacts with the hydrophilic terminal of the surfactant, producing a stable nanoemulsion. Clove essential oil was trapped in the hydrophobic center, and hydrophilic tragacanth gum was placed around the nanoemulsion. Addition of aluminium chloride is to increase the homogeneity dispersion of particles in nanoemulsion system and provide a better stability to the system.

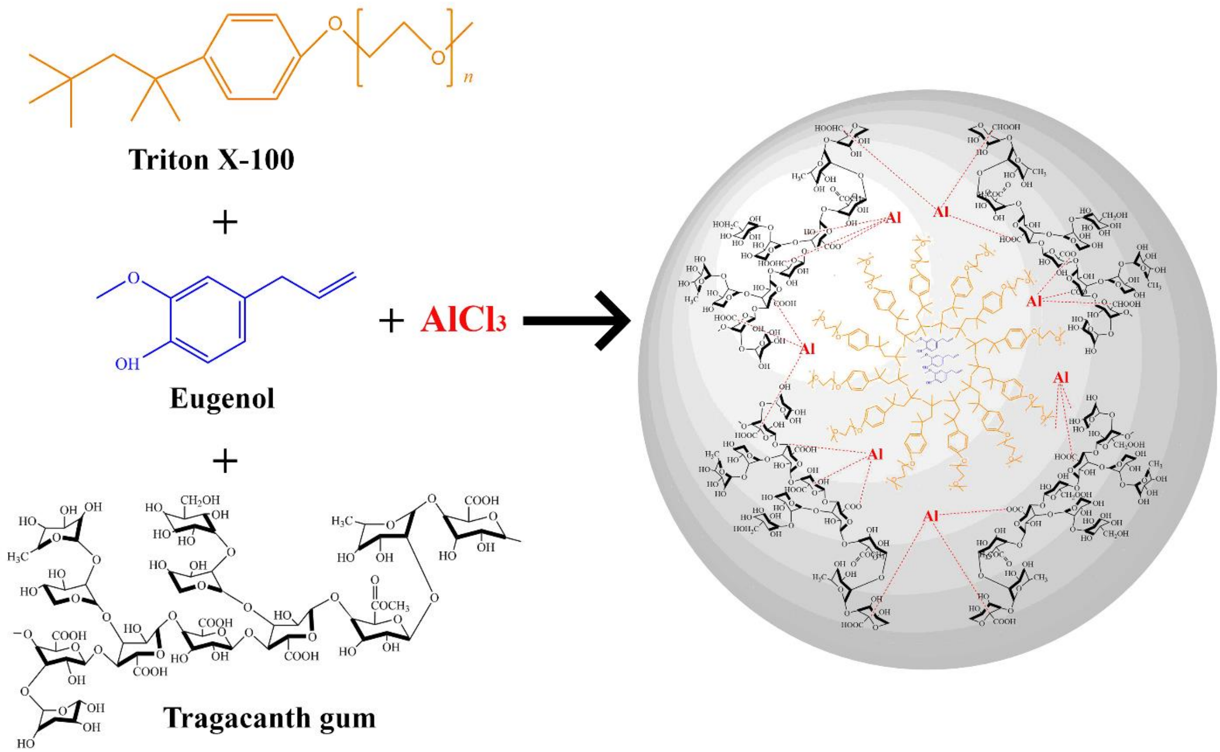


Figure 2 Proposed mechanism for the formation of tragacanth gum loaded with clove essential oil nanoemulsion

3.2 Transmission Electron Microscopy Analysis

Figure 3 depicts the internal morphology of tragacanth gum loaded with clove essential oil nanoemulsion at 100,000× magnification. It was observed that the particles in the nanoemulsion were fairly uniformly distributed, indicating that the nanoemulsion has a relatively high homogeneity (Figure 3(a)).

From Figure 3(b), the nanoemulsion was non-aggregated and exhibited discrete droplets of spherical shape with size ranged from 31.7 to 51.6 nm. According to Jafari and McClements [28], the system remained dispersed with no separation due to its small droplet sizes which prevent flocculation, resulting in improved stability of the nanoemulsion

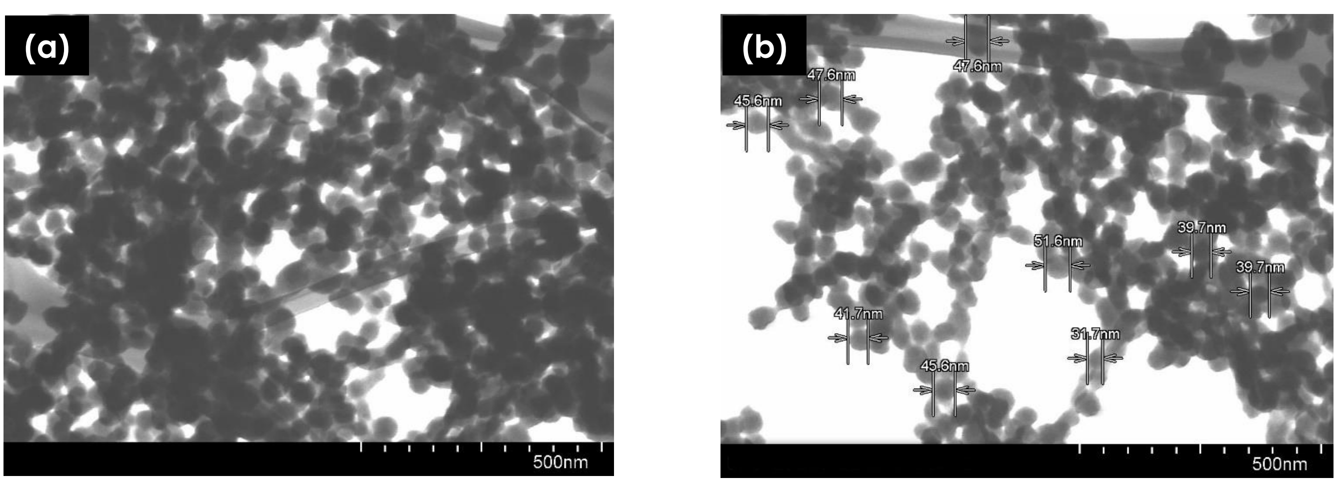


Figure 3 Transmission electron microscopy image of tragacanth gum loaded with clove essential oil nanoemulsion at 100,000× magnification: (a) the shape and particle distribution of nanoemulsion particles, and (b) the size of nanoemulsion particles

3.3 Physicochemical Stability

The particle size of tragacanth gum loaded with clove essential oil nanoemulsion was measured as 47.5 nm and the nanoemulsion remained stable until 30 days of observation with no significant change in physical appearance. Meanwhile its polydispersity index was determined as 0.3. According to the International Organization for Standardization polydispersity index values in the range of 0.05-0.70 are known to be monodisperse distribution, while values > 0.70 are polydisperse distribution of particles [29]. The low value of polydispersity index (0.3) reflects the homogeneity distribution of droplets in nanoemulsion.

In this study, the nanoemulsion was further characterised using the zeta potential in order to analyse the surface charge density between particles which is crucial for prediction of coagulation stability. The zeta potential for nanoemulsion was measured as -42.3 mV. As discussed by Jafari and McClements [28], readings exceeding +30 mV or below -30 mV indicate a stable nanoemulsion. Therefore, the nanoemulsion prepared in this study can be considered as stable.

3.4 Encapsulation Efficiency

The encapsulation efficiency of the nanoemulsion for clove essential oil was determined as 98%. This can be attributed to formation of trivalent aluminium ion complex bonds, which has been discussed earlier. This finding is consistent with a previous study of encapsulation of peppermint oil in biopolymer based nanosystem [30]. It is important to note that the encapsulation efficiency of clove essential oil in tragacanth gum nanoemulsion is greater than encapsulation of essential oil in arabic gum based nanoemulsion (92%) [14].

3.5 Wash Durability

Figures 4 and 5 demonstrate the surface morphologies of untreated fabric, treated fabric and treated fabric after 5 washing cycles at 1000× magnification. The untreated cotton and polyester in Figures 4(a) and 5(a) display a smooth surface texture with visible threadlike structure of fibrils. In contrast, treated cotton and polyester Figures 4(b) and 5(b) show a rough and shiny surface morphologies which indicate the deposition of tragacanth gum loaded with clove essential oil nanoemulsion formulation on fabrics.

The surface texture of treated cotton and polyester differs significantly due to the amount of nanoemulsion sprayed on cotton being greater than that for the polyester. It is known that cotton possesses hydrophilic properties, which results in a greater affinity for the nanoemulsion formulation than the hydrophobic polyester. This finding is in accordance with scanning electron microscopy analysis on polyester and cotton fabrics treated with formulations by Bonet-Aracil *et al*. [31].

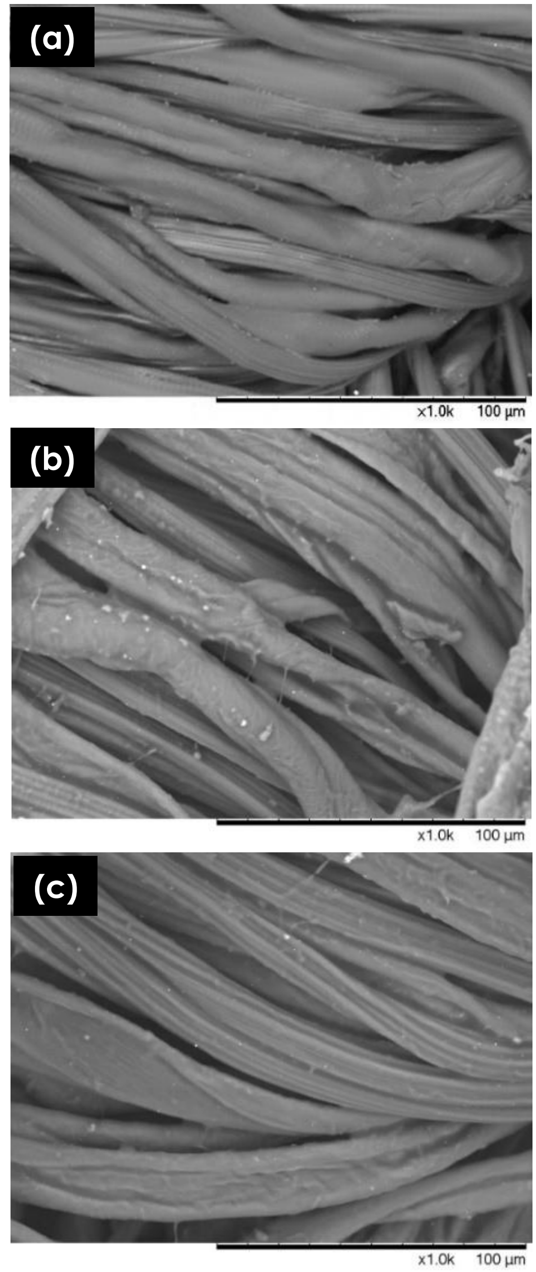


Figure 4 Scanning electron microscope images of (a) untreated cotton, (b) nanoemulsion treated cotton, and (c) nanoemulsion treated cotton after 5 washing cycles at 1000× magnification

The effects of washing on surface morphology of treated fabrics were also studied. From Figures 4(c) and 5(c) after 5 washing cycles the distinct layer of nanoemulsion formulation has been reduced, indicating that the tragacanth gum loaded with clove essential oil nanoemulsion formulation gradually decreased disproportionately with an increasing number of washing cycles. This result suggests that nanoemulsion formulation has good adhesion on fabrics which indicate high washability.

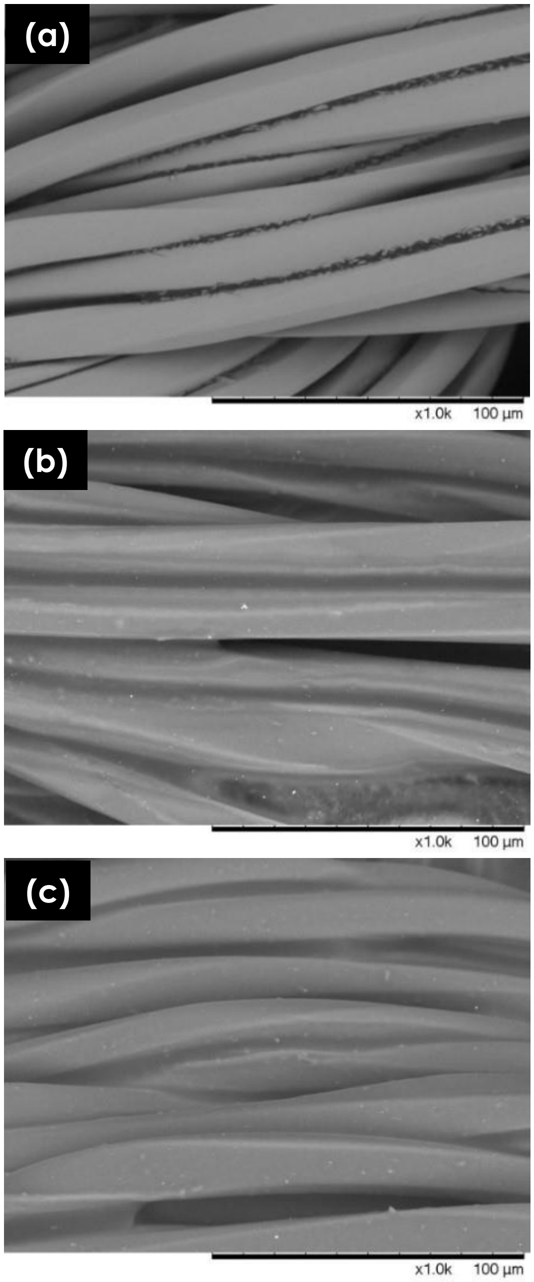


Figure 5 Scanning electron microscope images of (a) untreated polyester, (b) nanoemulsion treated polyester, and (c) nanoemulsion treated polyester after 5 washing cycles at 1000× magnification

3.6 Retention Study

Table 1 presents the percentage of clove essential oil retained on fabrics before and after 5 washing cycles. When comparing to unencapsulated clove essential oil, tragacanth gum used as a polymeric wall for clove essential oil results in a slow release of essential oil and the ability to retain on fabrics after a series of washing cycles. Based on Table 1, unencapsulated clove essential oil was successfully sprayed on cotton and polyester by 85% and 90%, respectively. On the other hand, clove essential oil encapsulated in tragacanth gum nanoemulsion was successfully sprayed on cotton and polyester by 96% and 93%, respectively.

After 5 washing cycles, unencapsulated clove essential oil recorded a low percentage of retention (12% on cotton and 14% on polyester) while encapsulated clove essential oil was able to retain 60% on cotton and 46% on polyester. Abrasion action from the washing cycle led to partial breakage of the Al-O-C bonds and prolonged release of clove essential oil from the nanoemulsion. This finding supports the hypothesis that tragacanth gum loaded with clove essential oils has high encapsulation efficiency and was able to promote the slow release of clove essential oil to the environment.

This study also proved that nanoemulsion formulations have good adhesion on fabrics, are able to prevent the complete removal of essential oils from the surface and are suitable as nanocarriers for essential oils [17-20].

Table 1 Effect of washing cycle on retention of essential oil from fabrics treated with clove essential oil and tragacanth gum loaded with clove essential oil nanoemulsion

Sample	Washing cycle	Retention (%)	
		Cotton	Polyester
Clove essential oil	Before wash	85	90
	1	60	62
	2	50	56
	3	43	45
	4	24	30
	5	12	14
Tragacanth gum loaded with clove essential oil nanoemulsion	Before wash	96	93
	1	90	86
	2	82	78
	3	75	71
	4	62	50
	5	60	46

3.7 Repellency Study

Fabrics treated with clove essential oil and tragacanth gum loaded with clove essential oil nanoemulsion formulations were subjected to repellency studies against *Aedes aegypti* and *Anopheles latens* The number of escaped mosquitoes in Excito chamber were recorded and percentage of repellency for both mosquito species are given in Table 2.

According to Bonet-Araci *et al*. [31], percentage of mosquito repellency can be classified into: (i) tolerable (40-49%), (ii) good (50-69%) and (iii) very good (higher than 69%). Based on Table 2, all fabrics treated with clove essential oil and tragacanth gum loaded with clove essential oil nanoemulsion before undergo sequence of washing cycle exhibited a 'very good' repellence results (94–99%) against *Ae. aegypti* and *An. latens*.

As for clove essential oil treated fabrics, a low repellency (20-29%) was obtained right after third washing cycle as compared to tragacanth gum loaded with clove essential oil nanoemulsion formulation which was able to maintain 'very good' repellency (46-64%) against two different mosquitoes until 5 washing cycles. These findings could be due to high encapsulation efficiency of tragacanth gum loaded with clove essential oil nanoemulsion formulation and high durability against washing cyles as discussed earlier in preceding sections.

Table 2 Effect of washing cycle on repellency against mosquitoes for fabrics treated with clove essential oil and tragacanth gum loaded with clove essential oil nanoemulsion

Sample	Wash cycle	Repellency (%)			
		Aedes aegypti		*Anopheles latens*	
		CT	PE	CT	PE
Clove essential oil	Before wash	98	99	97	95
	1	73	78	70	76
	2	65	69	60	64
	3	40	49	43	47
	4	38	40	36	38
	5	20	27	29	25
Tragacanth gum loaded with clove essential oil nanoemulsion	Before wash	96	92	94	94
	1	91	87	89	83
	2	88	75	81	70
	3	76	70	73	67
	4	73	68	66	63
	5	64	53	51	46

CT: Cotton; PE: Polyester

Slow degradation of tragacanth gum polymeric walls has caused a slow release of clove essential oil to the environment. The amount of clove essential oil in nanoemulsion was successfully retained by more than half amount of oil that was introduced to the nanoemulsion formulation. Due to this high level of retention (46-90%) for fabrics treated with tragacanth gum loaded with clove essential oil nanoemulsion formulation, they offered significant protection against tested mosquitoes. It is important to note that the hydrophilic characteristic of cotton fabric results in better adhesion for both retention and repellency treatments than polyester fabric.

4.0 CONCLUSION

The feasibility of tragacanth gum loaded with clove essential oil nanoemulsion as a mosquito repellent formulation for fabrics was tested for the first time. Generally, the nanoemulsion formulation exhibits a great encapsulation efficiency, dispersion stability, retention on cotton and polyester fabrics, and has 'very good' repellency against two different mosquito species. The retention and repellency of *Aedes aegypti* from the treated cotton has been increased by 48% and 44%, respectively, than unencapsulated clove essential oil. As for future perspective, it is worth to study the feasibility of biosynthesised nanoemulsion for mosquito control at both larval and adult stages. This innovation contributes remarkably to the advancement of mosquito-repellent textile industries.

Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

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References

[1] Greisman, L., Koenig, B., Barry, M. 2019. *Control of Mosquito-borne Illnesses: A Challenge to Public Health Ethics*. The Oxford Handbook of Public Health Ethics. 458–471. Doi: https://doi.org/10.1093/oxfordhb/9780190245191.013.39.

[2] World Health Organization. 2022. Dengue and Severe Dengue. https://www.who.int/news-room/fact-sheets/detail/dengue-and-severe-dengue.

[3] World Health Organization. 2022. World Malaria Report 2021. https://www.who.int/teams/global-malaria-programme/reports/world-malaria-report-2021.

[4] European Centre for Disease Prevention and Control. 2022. Dengue Worldwide Overview. https://www.ecdc.europa.eu/en/dengue-monthly

[5] Zhu, Q., Yang, Y., Lao, Z., Zhong, Y., Zhang, K., Zhao, S. 2020. Acute and Chronic Toxicity of Deltamethrin, Permethrin, and Dihaloacetylated Heterocyclic Pyrethroids in Mice. *Pest Management Science*. 76: 4210-21. Doi: 10.1002/ps.5978.

[6] Yusof, M. Z., Cherrie, J. W., Samsuddin, N., Semple, S. 2022. Mosquito Control Workers in Malaysia: Is Lifetime Occupational Pesticide Exposure Associated with Poorer Neurobehavioral Performance. *Annals of Work Exposures and Health*. 66(8): 1044-1055. Doi: https://doi.org/10.1093/annweh/wxac038

[7] Sun, Y.-J., Liang, Y.-J., Yang, L., Long, D.-X., Wang, H.-P., Wu, Y.-J. 2022. Long-term Low-dose Exposure of Permethrin Induces Liver and Kidney Damage in Rats. *Pharmacology Toxicology*. 23(1): 46. Doi: https://doi.org/10.1186/s40360-022-00586-2.

[8] Czarnobai, D. J. B., Hummel, H. E., Gross, J. 2022. Repellent Activity of Clove Essential Oil Volatiles and Development of Nanofiber-based Dispensers against Pear Psyllids (Hemiptera: Psyllidae). *Insects*. 13(8): 743. Doi: https://doi.org/10.3390/insects13080743.

[9] Nagaraju, P. G., Sengupta, P., Chicgovinda, P. P., Rao, P. J. 2021. Nanoencapsulation of Clove Oil and Study of Physicochemical Properties, Cytotoxic, Hemolytic, and Antioxidant Activities. *Journal of Food Process Engineering*. 44(4): e13645. Doi: https://doi.org/10.1111/jfpe.13645.

[10] de Oliveira C. T., Tavares, M. I. 2021. Antioxidant and Cytotoxic Activities of Clove Oil Nanoparticles and Evaluation of Its Size and Retention Efficiency. *Materials Sciences and Applicationsz*. 13(1): 39-53. Doi: https://doi.org/10.4236/msa.2022.131003.

[11] Eden, W. T., Alighiri, D., Supardi, K. I., Cahyono, E. 2020. The Mosquito Repellent Activity of the Active Component of Air Freshener Gel from Java Citronella Oil (Cymbopogon winterianus). *Parasitology Research*. 9053741. Doi: https://doi.org/10.1155/2020/9053741.

[12] Bu, G., Zhao, C., Wang, M., Yu, Z., Yang, H., Zhu, T. 2023. The Development and Properties of Nanoemulsions Stabilized with Glycated Soybean Protein for Carrying β-carotene. *Journal of Food Engineering*. 345: 111411. Doi: https://doi.org/10.1016/j.jfoodeng.2023.111411.

[13] Tayeb, H. H., Felimban, R., Almaghrabi, S., Hasaballah, N. 2021. Nanoemulsions: Formulation, characterization, Biological Fate, and Potential Role against COVID-19 and Other Viral Outbreaks. *Colloid and Interface Science Communications*. 45: 100533. Doi: https://doi.org/10.1016/j.colcom.2021.100533.

[14] Sharifi, S., Rezazad-Bari, M., Alizadeh, M., Almasi, H., Amiri, S. 2021. Use of Whey Protein Isolate and Gum Arabic for the Co-encapsulation of Probiotic Lactobacillus plantarum and Phytosterols by Complex Coacervation: Enhanced Viability of Probiotic in Iranian White Cheese. *Food Hydrocolloids*. 113: 106496. Doi: https://doi.org/10.1016/j.foodhyd.2020.106496.

[15] Wei, N., Hou, C., Liu, Z., Liang, Q., Lv, Z., Meng, X., Feng, J. 2023. Preparation of Fenpropathrin Nanoemulsions for Eco-friendly Management of Helicoverpa Armigera: Improved Insecticidal Activity and Biocompatibility. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*. 656: 130442. Doi: https://doi.org/10.1016/j.colsurfa.2022.130442.

[16] Kawish, S. M., Hasan, N., Beg, S., Qadir, A., Jain, G. K. Aqil, M. Ahmad, F. J. 2022. Docetaxel-loaded Borage Seed Oil Nanoemulsion with Improved Antitumor Activity for Solid Tumor Treatment: Formulation Development, In Vitro, In Silico and In Vivo Evaluation. *Journal of Drug Delivery Science and Technology*. 75: 103693. Doi: https://doi.org/10.1016/j.jddst.2022.103693.

[17] Rehman, A., Qunyi, T., Sharif, H. R., Korma, S. A., Karim, A., Manzoor, M. F., Mehmood, A., Iqbal, M. W., Raza, H., Ali, A., Mehmood, T. 2021. Biopolymer based Nanoemulsion Delivery System: An Effective Approach to Boost the Antioxidant Potential of Essential Oil in Food Products. *Carbohydrate Polymers*. 2: 100082. Doi: https://doi.org/10.1016/j.carpta.2021.100082.

[18] Mahdi, A. A., Al-Maqtari, Q. A., Mohammed, J. K., Al-Ansi, W., Aqeel, S. M., Cui, H., Lin, L. 2022. Nanoencapsulation of Mandarin Essential Oil: Fabrication, Characterization, and Storage Stability. *Foods*. 11: 54. Doi: https://doi.org/10.3390/foods11010054.

[19] Mahran, H. A. 2021. Using Nanoemulsions of the Essential Oils of a Selection of Medicinal Plants from Jazan, Saudi Arabia, as a Green Larvicidal against Culex Pipiens. *PLOS ONE*. 17(5): e0267150. Doi: https://doi.org/10.1371/journal.pone.0267150.

[20] Maurice, M. N., Huseein, E. A., Monib, M. E.-S., Alsharif, F. M., Namazi, N. I., Ahmad, A. A. 2021. Evaluation of the Scolicidal Activities of Eugenol Essential Oil and its Nanoemulsion against Protoscoleces of Hydatid Cysts. *PLOS ONE*. 16(11): e0259290. Doi: https://doi.org/10.1371/journal.pone.0259290.

[21] Real, D., Hoffmann, S., Leonardi, D., Salomon, C., Goycoolea, F. M. 2018. Chitosan-based Nanodelivery Systems Applied to the Development of Novel Triclabendazole Formulations. *PLOS ONE*. 13(12): e0207625. Doi: https://doi.org/10.1371/journal.pone.0207625.

[22] Yazdi, M. E. T., Nazarnezhad, S., Mousavi, S. H., Amiri, M. S., Darroudi, M., Baino, F., Kargozar, S. 2021. Gum Tragacanth (GT): A Versatile Biocompatible Material Beyond Borders. *Molecules*. 26(6): 1510. Doi: https://doi.org/10.3390/molecules26061510.

[23] Sharma, B., Thakur, S., Mamba, G., Prateek, Gupta, R. K. Gupta, V. K., Thakur, V. K. 2021. Titania Modified Gum Tragacanth based Hydrogel Nanocomposite for Water Remediation. *Journal of Environmental Chemical Engineering*. 9(1): 104608. Doi: https://doi.org/10.1016/j.jece.2020.104608.

[24] Ghayempour, S., Montazer, M., Rad, M. M. 2015. Tragacanth Gum as a Natural Polymeric Wall for Producing Antimicrobial Nanocapsules Loaded with Plant Extract. *International Journal of Biological Macromolecules*. 81: 514-520. Doi: https://doi.org/10.1016/j.ijbiomac.2015.08.041

[25] Hebeish, A., Hamdy, I. A., El-Sawy, S. M., Abdel-Mohdy, F. A. 2010. Preparation of Durable Insect Repellent Cotton Fabric through Treatment with a Finishing Formulation Containing Cypermethrin. *The Journal of the Textile Institute*. 101(7): 627-634. Doi: https://doi.org/10.1080/00405000902732859.

[26] Xin, J. H., Wang, X. W. 2018. Insect-repellent Textiles. *Journal of Engineered Fibers and Fabrics*. 335-348. Doi: https://doi.org/10.1016/b978-0-08-101273-4.00027-5.

[27] Khan, S. A., Khan, S. B., Khan, L. U., Farooq, A., Akhtar, K., Asiri, A. M. 2018. Fourier Transform Infrared Spectroscopy: Fundamentals and Application in Functional Groups and Nanomaterials Characterization. In: Sharma, S. (eds). Handbook of Materials Characterization. Springer, Cham. Doi: https://doi.org/10.1007/978-3-319-92955-2_9.

[28] Jafari, S. M., McClements, D. J. 2018. *Nanoemulsions: Formulation, Applications, and Characterization*. Elsevier. Doi: https://doi.org/10.1016/C2016-0-01985-X.

[29] International Organization for Standardization. 2017. Particle Size Analysis-dynamic Light Scattering (DLS) (ISO Standard No. 22412: 2017). https://www.iso.org/standard/65410.html

[30] Ghayempour, S., Montazer, M. 2019. A Novel Controlled Release System based on Tragacanth Nanofibers Loaded Peppermint Oil. *Carbohydrate Polymer*. 05: 589-595. Doi: https://doi.org/10.1016/j.carbpol.2018.10.078.

[31] Bonet-Araci, M., Bou-Belda, E., Gisbert-Payá, J., Ibañez, F. 2019. In Situ Test: Cotton Sheets against Mosquito Bites in India. *Cellulose*. 26(7): 4655-4663. Doi: https://doi.org/10.1007/s10570-019-02395-z.