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Cinnamon and Eucalyptus Extracts: A Promising Natural Approach for Durable Mosquito-Repellent Fabrics with Multifunctionality

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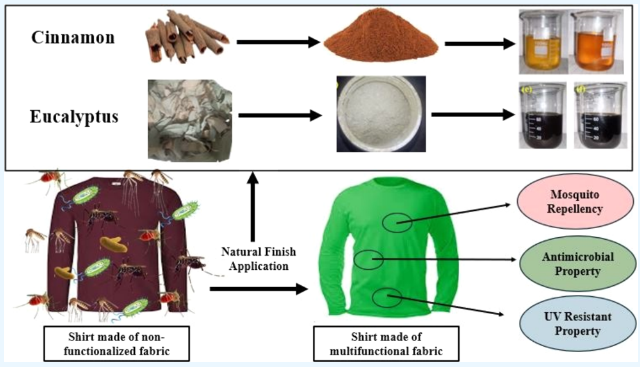
ABSTRACT: Mosquitoes are highly important carriers of diseases, such as malaria, dengue, chikungunya, and various other life-threatening illnesses. Traditionally, many chemicals, such as plant extracts, oils, and smoke, have been employed for the purpose of repelling mosquitoes. Various plants possess essential oils and chemicals that have been proven to be good insect repellents and are commonly regarded as weeds. The present study focused on the development of eco-friendly, nonhazardous mosquito-repellent fabrics using cinnamon and eucalyptus extracts. First, eucalyptus and cinnamon extracts were produced separately using ethanol and water as solvents with and without heating. Forty-eight different fabric samples were prepared by applying these extracts at three levels of process application temperature. A steam dye bath sampling machine was utilized to execute the extraction application process on fabric samples. The mosquito repellency performances of all of the samples were evaluated using the cage test method. The cage test revealed that all of the samples of eucalyptus and cinnamon extract-applied fabrics showed mosquito repellency performance at some level. However, the fabric samples treated with the heated extract of eucalyptus ethanol (EE-H) at 60 °C showed the best results in terms of mosquito repellency (85.56%) among all combinations. In addition to repellency, the impact of washing durability, UV shielding, and antibacterial performance was also evaluated. This research demonstrated a new method for creating a fabric that repels mosquitoes and has effective antibacterial properties as well as promising ultraviolet protection factor (UPF) rating. This fabric protects the wearer from the significant health risks posed by mosquitoes and harmful UV radiation while also maintaining its cleanliness. Moreover, the utilization and implementation of plant-derived coatings on textiles contribute to the advancement of sustainable methods (SDG 9 and SDG 12) in the chemical processing industry of textiles, ultimately leading to a reduction in their environmental footprint.



■ INTRODUCTION

The word mosquito dates back to 1572 in the English language. This word was embraced to substitute the name “biting flies” to avoid misperception of house flies. The bites of mosquitoes resulted in not only pain but also a variety of diseases like chikungunya and malaria. All over the world, mosquito bites are deemed as an annoyance. Malaria is caused by infection from protozoans transferred by mosquitoes. It is accountable for dengue, yellow fever, epidemic polyarthritis, hemorrhagic fever, and various types of encephalitis.[1] According to a WHO report, in 2022, malaria caused the deaths of 608 000 people all over the world.[2] In 2019, 3.10 million people in the American territory, 042 million people in Philippine, 0.320 million population in Vietnam, 1.01 million human beings in Bangladesh, and 0.131 million people in Malaysia were affected by dengue fever.[3]

Mosquito repellents are divided into two groups, i.e., chemical repellents and natural repellents. At first, people employed mosquito repellents on their skin in the form of lotions. However, its effectiveness was valid for a short period of time. Moreover, they can be dangerous as they are in direct contact with human body.[4] From past research, it was noted that synthetic microbial agents are significantly efficient in obstructing growth of microbes. On the other hand, they are not biodegradable and environment-friendly.[5] Synthetic mosquitocides like *N,N*-diethyl-*m*-toluamide (DEET) and permethrin have been extensively used in the past.[6−9] However, both these chemicals have toxicological properties, which cause pricking, burning, and scratching on the skin of

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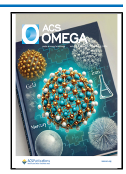

the human body.[10] On the contrary, a lot of extracts from plants generate antimicrobial properties due to their diversified natural inherited chemical composition.[11−13] Furthermore, extracts from plants were employed as a critical source of medicines throughout history.[14−16] For example, plant extracts inculcate a variety of chemical active constituents like tannins, phenols, quercetin, polyphenols, and carotenes, and can mediate and inhibit various insects like carpet beetle, mosquitoes and moths.[17] Many researchers have investigated the use of natural components like castor oil,[10] mint leaves,[10] fresh moringa leaves,[18] neem and tulsi,[10] sweet basil and eucalyptus,[19] and marigold petals[20] to prepare mosquito-repellent textiles.

Gupta and Singh[20] developed an environment-friendly mosquito-repellent finish on textile substrates by employing the herbal extract of mint leaves. The finishes were employed on the textile substrate by a pad dry curing methodology. Afterward, the mosquito-repellent behavior test was performed on these substrates. The washing durability was evaluated after the third, fifth, seventh and ninth washes. After application of finishes, the textile substrate delivered 100% mosquito-repellent behavior.[21] Gupta and Singh[20] in another study employed Marigold (Tagetes erecta) flower petals employed on the substrate of cotton. They examined the mosquito-repellent behavior of cotton fabric against *Culex quinquefasciatus* (female mosquito) and unknown home mosquitoes, which showed 66.25 and 100% mosquito repellency, respectively. Zayed et al.[22] noted 98% repellent property and a 95% death rate after 12 h of utilizing the extract of *Citrus sinensis* peel on the surface of cotton substrate.[22] Sánchez-Borzone et al.[23] determined that mint plants have ketone monoterpenes, which include pulegone, carvone, and menthone and are extremely lethal to most active insects. Mia et al.[10] in their research utilized plants like neem (*Azadirachta indica*), tulsi (*Ocimum tenuiflorum*), and mint leaf (Mentha) on cotton substrate and investigated their effectiveness against the mosquitoes. Each extract displayed suitable repellency and mint leaf displayed 90% repellency. But a considerable loss of antimosquito efficacy was reported after the ninth washing cycle. A study conducted by Kantheti et al.[19] concentrated on the synthesis of environment-friendly, not harmful mosquito-repellent substrates employing the extracts from thirteen plants, i.e., rind of orange and lemon, flowers of French marigold, wild sage and spearmint, betel, moringa, custard apple, eucalyptus, lemongrass, neem, sweet basil, and leaves of holy basil by employing the improved cage test method. Three plant sources, i.e., flowers of French marigold, leaves of sweet basil, and eucalyptus, on testing had exceptional mosquito-repellent property in comparison with the remaining tested sources.

In a study conducted by Specos et al.,[24] microcapsules inculcating citronella oil were developed by complicated coacervation and were employed to the cotton substrate to investigate the repellency efficacy of acquired fabrics. Citronella ejaculated from treated substrate was examined in some way by extracting the substance of major constituents. The property of repellency was evaluated by subjecting a human hand and arm protected with treated textile substrate to Aedes aegypti mosquitoes. Microencapsulated citronella-treated textile substrate showed a significant and prolonged defense against insects in comparison to substrates sprayed with an ethanol solution of crucial oil, ensuring a repellency impact higher than 90% for 3 weeks.

Shrimali and Manoj Dedhia[25] conducted a study in which antimicrobial finish was applied to cotton substrate by utilizing ethanolic and acetonic extricates of cinnamon bark and *Garcinia indica* by direct employment and by microencapsulation. It was noted that both herbal extricates after their impregnation on cotton substrate impart superior antimicrobial property against Gram-positive and Gram-negative bacteria such as *Staphylococcus aureus* and *Klebsiella pneumoniae*. It was noted that the outcomes of washing fastness were not good due to the less long-lasting finish. The antimicrobial performance reduced with every wash, and after the fifth washing cycle, no action was noted against the chosen microbes. Parvez et al.[26] investigated the utility of extracts (alcoholic) from peppermint leaf stems and garlic cloves as mosquito-repellent finished material on knitted fabric. A varying concentration (5, 15, 25, and 35%) of peppermint garlic extract (PGE) was developed and employed to the fabric utilizing an exhaust dyeing procedure to evaluate the mosquito (*Aedes aegypti* L.) repellent properties. The outcomes showed that PGE-treated substrates, i.e., 25% PGE and 35% PGE, had the highest mosquito mortality and repellency rates.

In this study, cinnamon bark and eucalyptus leaves in powdered form were mixed with distilled water and ethanol to prepare mosquito-repellent finishes. Bleached cotton fabric samples were treated with cinnamon and eucalyptus extracts using a steam/dye bath at temperatures of 40, 60, and 80 °C, respectively. The mosquito-repellent ability of extracts put on a textile substrate was evaluated using the cage test method. Subsequently, a Fourier transform infrared (FTIR) analysis was conducted. The durability of extracts applied on fabric was evaluated for one and five washing cycles. This was due to the fact that a limited number of studies were performed on the washing fastness of natural mosquito-repellent extracts applied on a textile substrate. Hence, this research has the potential to pave the way for the creation of a novel category of eco-friendly and long-lasting mosquito-repellent textiles. The innovative repellent fabrics discussed below are anticipated to have a substantial impact on the worldwide reduction of mosquito-borne diseases.

MATERIALS AND METHODS

Materials. The experiment utilized cinnamon bark, eucalyptus leaves, distilled water, ethanol, and citric acid. These materials were utilized in the formulation of mosquito-repellent finishing. The study utilized 100% cotton cloth with a weight of 140 g/m^2, which was obtained from a local market after undergoing scouring and bleaching processes. A specimen of eucalyptus leaves was collected from a solitary tree located near National Textile University, Faisalabad. The cinnamon bark was obtained from the local market in Faisalabad, Pakistan. Marjan Chemicals Pakistan provided laboratory-grade citric acid, ethanol, and distilled water for purchase.

Methodology. *Preparation of Cinnamon and Eucalyptus Extract.* Extraction was accomplished using both water and ethanol as a medium of extraction. In cinnamon bark and eucalyptus leaves, both the ingredients were grounded into fine powder form and then used for extraction. The extraction was carried out in two ways (unheated and heated) and by two mediums (water and ethanol). Cinnamon powder was then immersed in water at 1:10 ratio. The mixture was left for 24 h at room temperature. After 24 h, half of the cinnamon extract was separated and filtered and was named as the unheated extract of cinnamon in water (CW-UH). The other half of the

Figure 1. Cinnamon water extracts: unheated (left bottle) and heated and filtered (right bottle).

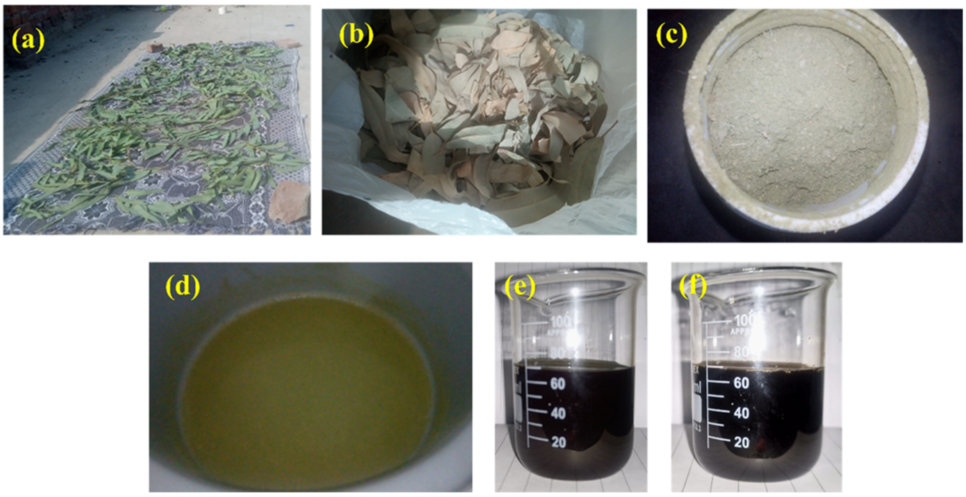

Figure 2. Steps involved in extraction of eucalyptus-based natural finish: (a) eucalyptus green leaves, (b) dried leaves, (c) powdered eucalyptus leaves, (d) powdered eucalyptus leaves in water, (e) eucalyptus and water extract unheated, and (f) eucalyptus and water extract heated.

extract was taken as unfiltered and heated for 1 h at 60 °C, and then it was filtered and named as the heated extract of cinnamon in water (CW-H), as shown in Figure 1.

Likewise, the ethanol extract of cinnamon was obtained. The fine powder of cinnamon was immersed in ethanol in a closed container. The cinnamon powder to ethanol ratio was taken as 1:10. This was set to settle for 24 h at room temperature. After 24 h, half of the extract was filtered and named as the unheated extract of cinnamon in ethanol (CE-UH). The other half of the extract was taken unfiltered, heated for 1 h at 60 °C, and then filtered, named as the heated extract of cinnamon in ethanol (CE-H). Eucalyptus leaves were taken from a single tree (Figure 2a). Leaves were then set to dry in sunlight for a straight 10 days, daily from from 8 am to 5 pm (Figure 2b). The dried leaves were then grinded into a fine powder (Figure 2c). Powdered leaves were immersed in water at 1:10 ratio. Eucalyptus powder was kept immersed in 1500 mL of distilled water at room temperature for 24 h to get the extract (Figure 2d). After 24 h, half of the mixture was separated and filtered, named as the unheated extract of eucalyptus in water (EW-UH). The other half of the mixture was taken as unfiltered, heated at 60 °C for an hour, and filtered, named as heated extract of eucalyptus in water (EW-H).

A fine powder of eucalyptus leaves was immersed in ethanol in a closed container. The powder to ethanol ratio was taken as 1:10. 2000 mL of ethanol was mixed with eucalyptus leaves powder to form a solution, which was then set to settle for 24 h at room temperature. The extract was filtered after 24 h, named as the unheated extract of eucalyptus in ethanol (EE-UH) (Figure 2e). The other half of the extract was taken as unfiltered, heated for 1 h at 60 °C, and was then filtered, named as the heated extract of eucalyptus in ethanol (EE-H) (Figure 2f).

Application on Fabric Samples. Bleached cotton fabric samples were cut in the required length and weight. The weight of each sample was taken as 4 g. The cinnamon extract solution was prepared by taking 240 mL extract in a beaker and 6% of citric acid was added to it as a cross-linker, dissolved by continuous stirring. The cotton fabric was soaked in the beaker containing extracts. A steam bath was used to treat the cotton fabric with cinnamon and eucalyptus extracts (Figure 3) for 30 min at different temperatures as given in Table 1. Lastly, the cotton fabric samples were removed from the beakers and air-dried for 24 h. The same treatment procedure was adopted for treatment of the cotton fabric with all extracts. The treated and

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Figure 3. Steam/dye bath.

Table 1. Design of the Experiment

sample no.	extract type	sample code[a]	application temperature 40 °C	60 °C	80 °C
1	cinnamon + water	CW-H	√		
2	cinnamon + water			√	
3	cinnamon + water				√
4	eucalyptus + water	EW-H	√		
5	eucalyptus + water			√	
6	eucalyptus + water				√
7	cinnamon + ethanol	CE-H	√		
8	cinnamon + ethanol			√	
9	cinnamon + ethanol				√
10	eucalyptus + ethanol	EE-H	√		
11	eucalyptus + ethanol			√	
12	eucalyptus + ethanol				√
13	cinnamon + water	CW-UH	√		
14	cinnamon + water			√	
15	cinnamon + water				√
16	eucalyptus + water	EW-UH	√		
17	eucalyptus + water			√	
18	eucalyptus + water				√
19	cinnamon + ethanol	CE-UH	√		
20	cinnamon + ethanol			√	
21	cinnamon + ethanol				√
22	eucalyptus + ethanol	EE-UH	√		
23	eucalyptus + ethanol			√	
24	eucalyptus + ethanol				√

[a]Heated extract (H); unheated extract finish (UH)

untreated fabric samples are shown in Figure 4 for visual elaboration.

Washing Durability Test. The wash durability of the fabrics was evaluated by subjecting them to washing in a Laundr-o-meter (SDATLAS, M228AA) using the AATCC test method 61-1A. The fabrics were washed once and five times and then assessed for their effectiveness in repelling mosquitoes. After undergoing a single Launder-o-meter washing, each sample was washed and air-dried. This process was repeated for a total of five washings. A mosquito repellency test was performed on unwashed samples as well as those that had undergone one wash and five washes. The results of the unwashed and washed samples were compared.

Mosquito Repellency Test. Evaluation of mosquito-repellent properties was carried out using a modified WHO cage test technique, and 30 mosquitoes were taken in the cage. The mosquitoes used in this study were starved for 10 h. Mosquitoes were caught while biting humans between 7 and 10 pm using a net. The cage used in the tests is shown in Figure 5. The treated fabric samples were compared to the untreated fabrics to check the mosquito repellent's effectiveness. The mosquitoes were allowed to settle for 5 min. The number of mosquitoes resting on the treated and untreated fabric samples was counted after 5,10, 15, 20, 25, and 30 min intervals. The untreated fabric sample was first tested. For each sample, the percentages of mosquito repellency were determined using the formula given below.

$$\text{repellency rate formula} = \frac{\text{total no. of mosquitoes} - \text{no. of mosquitoes landed}}{\text{total no. of mosquitoes under test}} \times 100$$

Statistical Analysis. Statistical analysis was also performed to evaluate the relationship between key factors and mosquito repellency. The analysis of variance (ANNOVA) technique was applied using Minitab software. The statistical significance of input variables such as extract type and temperature was concluded against the mosquito repellency of all of the samples.[27,28]

FTIR Analysis. Fourier-transform infrared (FTIR) spectroscopy is employed to identify chemical bonds and functional groups by using the distinctive absorption of infrared light in the vibrational modes of the bonds. In order to confirm the presence of functional groups on the surface of the cotton fabric, both before and after treatment with various cinnamon and eucalyptus extracts, an FTIR study was conducted using Nexus-870. Additionally, the FTIR spectra were examined.

Ultraviolet Protection Factor (UPF) Test. UPF is a crucial factor in textiles, as it provides protection to the wearer against harmful ultraviolet (UV) radiations. The UPF test was conducted in accordance with the techniques outlined in the AATCC 183 standard method. The fabric samples, which were conditioned at a temperature of 21 ± 1 °C and a relative humidity of 65 ± 2%, were tested for UPF. A UV-2000 Transmittance Analyzer was used to perform UPF testing. The data included UVA, UVB, and average UPF measurements.

Antibacterial Activity Test. The antibacterial activity test was performed on unheated extracts-treated fabric samples that were processed at the optimum temperature parameter (60 °C) against *S. aureus* bacteria qualitatively using the disc diffusion test method (AATCC 147).

■ RESULTS AND DISCUSSION

All of the samples were prepared and tested for mosquito repellency to evaluate the efficacy of the applied extracts. The results of samples with different extract combinations and post-treatment processes are shown in Tables 2–6, respectively. The mosquito repellency test was performed by adopting the cage test method. The count of mosquitoes that contacted the sample during the exposure time was expressed in percentage as a performance indicator for all of the samples. The variation of mosquito count after a 5 min interval was used to average out the performance, which is shown as a graph in Figures 7 and 10 for heated and unheated extract samples.

Mosquito Repellency of Heated Extracts (Unwashed). In the case of heated extract application, four different combinations of cinnamon and eucalyptus with water and

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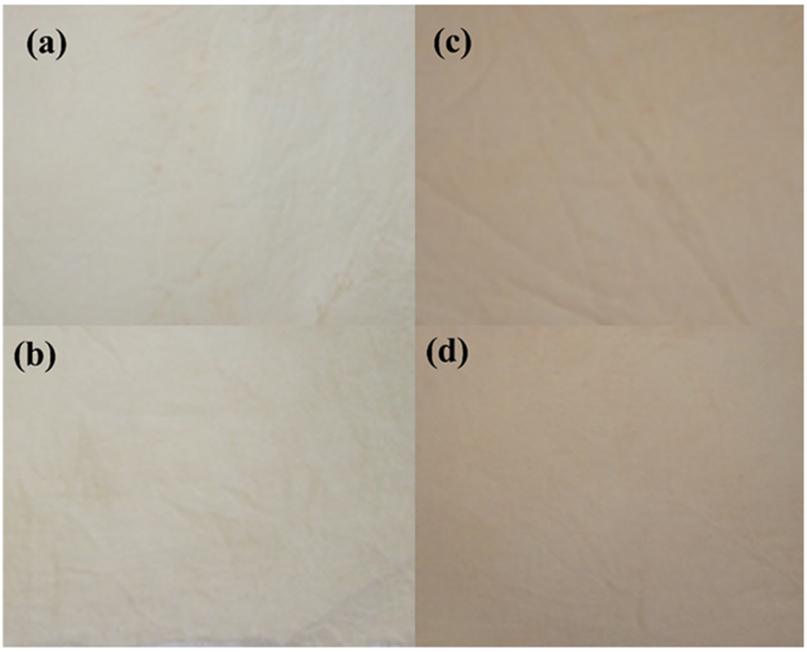

Figure 4. Treated samples with cinnamon and eucalyptus water extracts: (a) EW-UH, (b) EW-UH, (c) CW-UH, and (d) CW-H.

Figure 5. Cage used to test mosquito repellency.

Table 2. Mosquito Repellency Percentage (%) of Samples (Unwashed) Treated with Heated Extracts

sr. no.	sample code	treated at temperature (°C)	mosquitoes landed average ± SD	repellency (%)
1	CW-H	40	14.66 ± 2.05	51.13
		60	12.33 ± 2.35	58.9
		80	11.16 ± 1.84	60.2
2	EW-H	40	7.33 ± 3.02	75.56
		60	6.33 ± 3.15	78.9
		80	7.33 ± 2.88	78
3	CE-H	40	10.33 ± 2.62	65.56
		60	18.33 ± 2.88	72.23
		80	13.66 ± 2.17	54.46
4	EE-H	40	6.33 ± 3.15	78.9
		60	4.33 ± 1.42	85.56
		80	8.33 ± 2.88	72.23

ethanol were applied on the fabric samples at three different levels of temperature (40, 60, and 80 °C) as shown in Table 2. It is obvious that the effects of extract type on the mosquito repellency of the treated fabric are statistically significant (p-values < 0.05), and the effect of temperature is statistically insignificant, as illustrated in Table S1. The study (Figure 6 and Table 2) revealed that at 40 °C the fabric samples treated using eucalyptus ethanol (EE-H) extract displayed the highest mosquito repellency, which is 78.9%, followed by the samples treated with eucalyptus water (EW-H) extract (75.56%), cinnamon ethanol (CE-H) extract (65.56%), and cinnamon water (CW-H) extract (51.13%). However, the sample treated with eucalyptus ethanol (EE-H) extract showed the highest mosquito repellency (85.56%) at 60 °C application temperature, followed by samples treated with eucalyptus water (EW-H) extract (78.9%), cinnamon ethanol (CE-H) extract (72.23%), and cinnamon water (CW-H) extract (51.13%). Furthermore, the samples treated at 80 °C with ethanol—water (EW-H) extract and eucalyptus ethanol (EE-H) extract depicted a mosquito repellency performance of 78 and 72.23%, respectively, followed by samples treated with cinnamon ethanol (CE-H) extract (54.46%) and cinnamon water (CW-H) extract (62.2%) (Figure 7).

Overall, all of the samples of heated extract exhibited the mosquito-repellent property. However, the sample treated with eucalyptus ethanol (EE-H) extract at 60 °C showed the highest mosquito repellency performance among all of the 12 samples. The results also displayed that the efficacy of ethanol as a solvent for the heated extraction scenario is comparatively better than that of water in both plant extracts, which resulted in better performance in all resultant samples. Furthermore, eucalyptus extract samples showed higher performance results

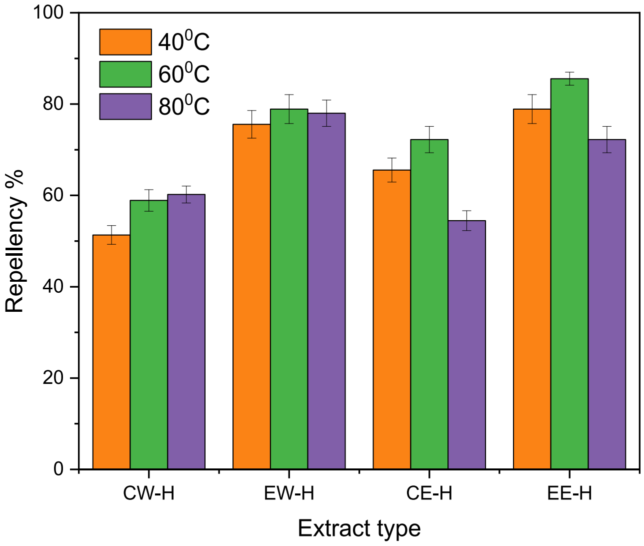

Figure 6. Mosquito repellency % of samples treated with heated extracts of cinnamon and eucalyptus at various temperatures (unwashed samples).

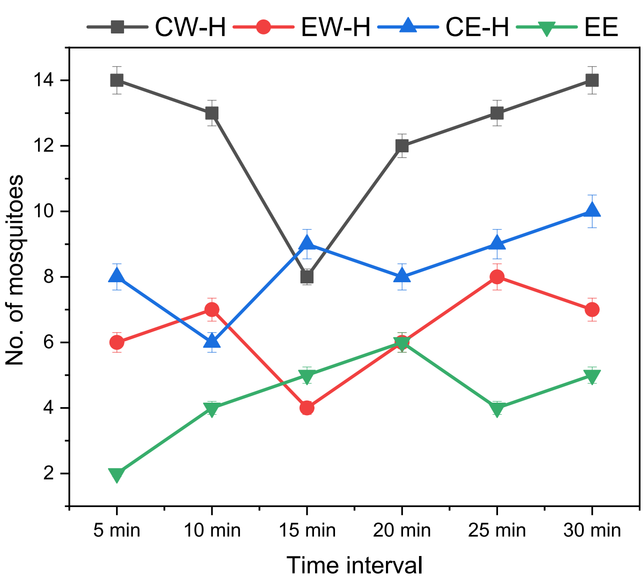

Figure 7. Mosquito repellency % at various time intervals of unwashed samples treated with heated extracts.

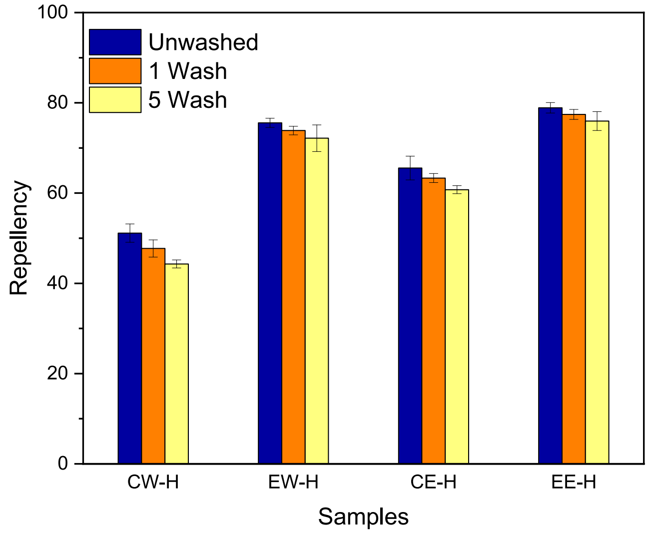

Figure 8. Mosquito repellency of samples treated (60 °C) with unheated extract after washing.

compared to cinnamon extract-applied samples at all temperature levels. In the case of temperature of application process, the results depicted that from 40 to 60 °C the mosquito repellency performance increased and declined at 80 °C, which explains the optimization of process temperature up to 60 °C. This phenomenon illustrated that at 60 °C maximum extract was attracted, settled down, and retained its position on the fabric sample surface.

Mosquito Repellency of Heated Extracts (after Washing). An analysis was conducted to assess the effect of laundry on fabric samples. This was done by subjecting all of the samples in the heated extract category to five wash cycles and subsequently measuring the effectiveness of mosquito repellency. The study (Figure 8 and Table 3) revealed that at 40 °C temperature the fabric samples treated by using eucalyptus ethanol (EE-H) extract displayed the highest mosquito repellency, which is 75.96%, followed by the samples treated with eucalyptus water (EW-H) extract (72.16%), cinnamon ethanol (CE-H) extract (60.76%) and cinnamon water (CW-H) extract (44.3%), respectively. However, the sample treated with eucalyptus ethanol (EE-H) extract showed the highest mosquito repellency of 83.33% at 60 °C application temperature, followed by samples treated with eucalyptus water (EW-H) extract (75.96%), cinnamon ethanol (CE-H) extract (68.33%), and cinnamon water (CW-H) extract (53.33%). Furthermore, the samples treated at 80 °C with ethanol–water (EW-H) extract and eucalyptus ethanol (EE-H) extract depicted a similar mosquito repellency performance (68.33%), followed by samples treated with cinnamon ethanol (CE-H) extract (48.1%) and cinnamon water (CW-H) extract (38.6%).

Like the unwashed samples, all of the washed samples of the heated extract had mosquito-repellent properties. However, the protection efficacy of the washed samples was marginally reduced due to the influence of detergent and rigorous washing cycles. Similarly, the sample treated with eucalyptus ethanol (EE-H) extract at 60 °C exhibited the most effective mosquito repellency performance after washing compared to the other 12 samples. Moreover, the observed decrease in mosquito repellency during wash cycles serves as evidence for the endurance and effectiveness of the applied natural finish.

Mosquito Repellency of Unheated Extracts (Unwashed). In the case of unheated extract application, four different combinations of cinnamon and eucalyptus with water and ethanol were applied on fabric samples at three different levels of temperature (40, 60, 80 °C) as shown in Table 4. Analysis of variance (ANNOVA) results showed that the effects of extract type on the mosquito repellency of the treated fabric and the effect of temperature both are statistically significant (p-values < 0.05), as illustrated in Table S2. The study (Figure 9 and Table 4) revealed that at 40 °C, the fabric samples treated using eucalyptus ethanol (EE-UH) extract displayed the highest mosquito repellency, which is 75.56%, followed by the samples treated with eucalyptus water (EW-UH) extract (72.23%), cinnamon ethanol (CE-UH) extract (62.23%), and cinnamon water (CW-UH) extract (47.8%). However, the sample treated with eucalyptus ethanol (EE-UH) extract showed the highest mosquito repellency of 82.23% at

Table 3. Mosquito Repellency % of Samples Treated with Heated Extract after Washing

sr. no.	sample code	treated at temperature (°C)	samples tested ⟨mosquitoes landed⟩ (average)		total mosquitoes	repellency (%)	
			one wash	five wash		one wash	five wash
1	CW-H	40	15.68	16.71	30	47.73	44.3
		60	13.19	14		56	53.33
		80	13.29	18.42		42.36	38.6
2	EW-H	40	7.84	8.35	30	73.86	72.16
		60	6.77	7.21		77.43	75.96
		80	6.77	9.5		70	68.33
3	CE-H	40	11	11.77	30	63.33	60.76
		60	9	9.5		70	68.33
		80	14.61	15.57		51.3	48.1
4	EE-H	40	6.77	7.21	30	77.43	75.96
		60	4.63	5		84.56	83.33
		80	9	9.5		70	68.33

Table 4. Mosquito Repellency Percentage (%) of Samples Treated with Unheated Extract at Various Time Intervals (Unwashed Samples)

sr. no.	sample code	treated at temperature (°C)	mosquitoes landed average ± SD	repellency (%)
1	CW-UH	40	15.66 ± 1.91	47.8
		60	13.33 ± 2.22	55.56
		80	10.16 ± 1.71	42.2
2	EW-UH	40	7.33 ± 2.88	72.23
		60	7.33 ± 3.03	75.56
		80	9.33 ± 2.75	68.9
3	CE-UH	40	11.33 ± 2.49	62.33
		60	9.33 ± 2.73	68.8
		80	14.66 ± 2.04	51.13
4	EE-UH	40	7.33 ± 2.02	75.56
		60	5.33 ± 2.28	82.23
		80	9.33 ± 2.75	68.9

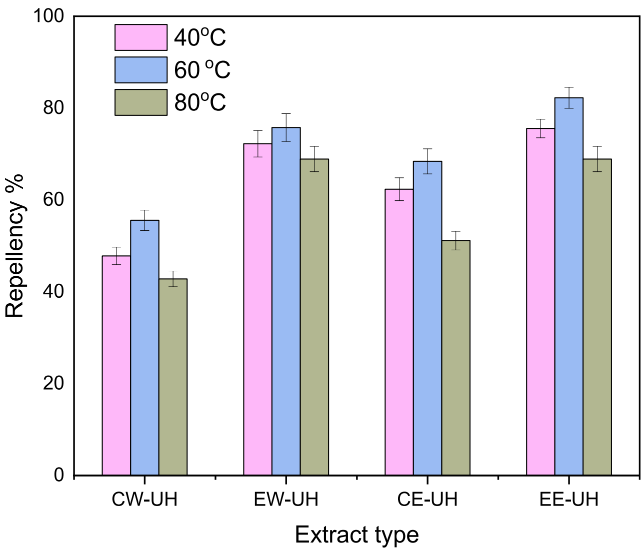

Figure 9. Mosquito repellency % of samples treated with unheated extracts of cinnamon and eucalyptus at various temperatures (unwashed samples).

60 °C application temperature, followed by samples treated with eucalyptus water (EW-UH) extract (75.56%), cinnamon ethanol (CE-UH) extract (68.8%), and cinnamon water (CW-UH) extract (55.56%). Furthermore, the samples treated at 80 °C with ethanol–water (EW-UH) extract and eucalyptus ethanol (EE-UH) extract depicted similar mosquito repellency performance, 68.9%, followed by samples treated with cinnamon ethanol (CE-UH) extract (51.13%) and cinnamon water (CW-UH) extract (42.2%).

Overall, all of the samples of heated extract exhibited mosquito-repellent property. However, the sample treated with eucalyptus ethanol (EE-UH) extract at 60 °C showed the highest mosquito repellency performance among all 12 samples. The results also illustrated that the efficacy of ethanol as a solvent for heated extraction scenario is comparatively better than that of water in both plant extracts, which resulted in the better performance in all resultant samples. Furthermore, eucalyptus extract samples showed higher performance results compared to cinnamon extract-applied samples at all temperature levels. In the case of application process temperature, the results depicted that from 40 to 60 °C the mosquito repellency performance increased and it declined at 80 °C, which explains the optimization of process temperature up to 60 °C. This phenomenon illustrated that at 60 °C, maximum extract was attracted, deposited, and retained its position on the fabric sample surface (Figure 10).

Repellency of Unheated Extracts (after Washing). The impact of laundry on the fabric sample was analyzed by passing through all of the samples of the unheated extract category under five wash cycles, and then measuring the efficacy of mosquito repellency for analysis. After five washes, the study (Figure 11 and Table 5) revealed that at 40 °C temperature the fabric samples treated using eucalyptus ethanol (EE-UH) extract displayed the highest mosquito repellency, which is 70%, followed by the samples treated with eucalyptus water (EW-UH) extract (66.66%), cinnamon ethanol (CE-UH) extract (55.46%), and cinnamon water (CW-UH) extract (38.3%). However, the sample treated with eucalyptus ethanol (EE-UH) extract showed the highest mosquito repellency, 78.33%, at 60 °C application temperature, followed by samples treated with eucalyptus water (EW-UH) extract (71.36%), cinnamon ethanol (CE-UH) extract (62.83%), and cinnamon water (CW-UH) extract (47.6%). Furthermore, the samples treated at 80 °C with ethanol–water (EW-UH) extract and eucalyptus ethanol (EE-UH) extract depicted a similar mosquito repellency performance, 63.33%, followed by

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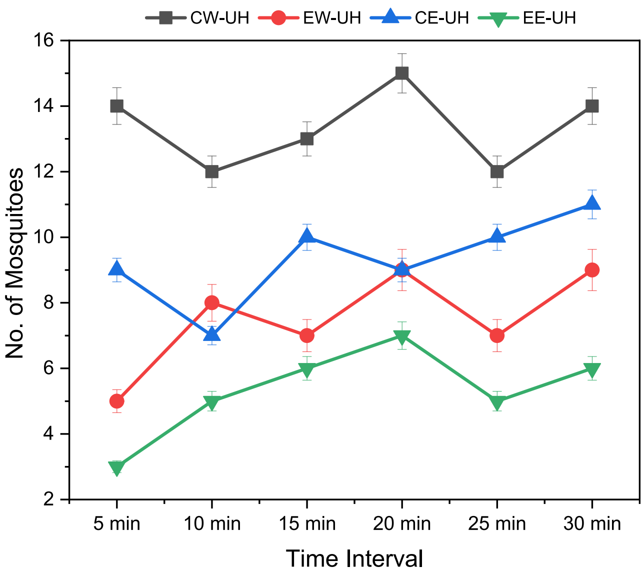

Figure 10. Mosquito repellency % at various time intervals of washed samples treated with unheated extracts.

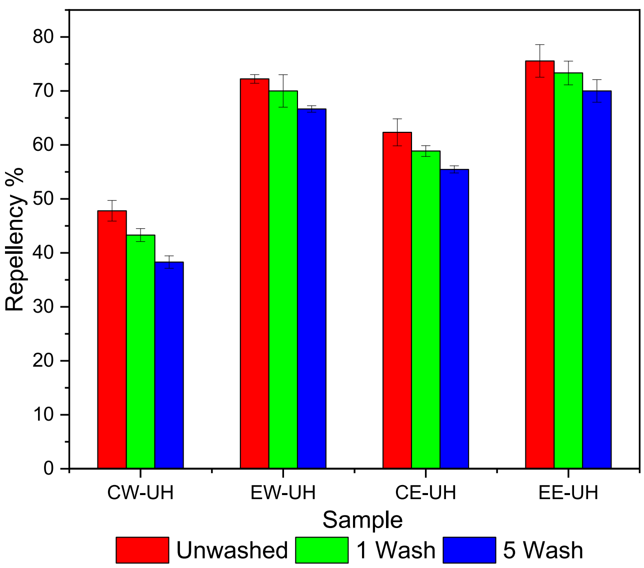

Figure 11. Mosquito repellency % of samples treated with unheated extracts (unwashed, one washed, five washed samples) (60 °C).

samples treated with cinnamon ethanol (CE-UH) extract (42.33%) and cinnamon water (CW-UH) extract (32.53%).

Similar to unwashed samples, all of the washed samples of the heated extract exhibited mosquito-repellent property, but the protection performance of all of the washed samples was gradually decreased after five washes (Figure 11) due to the impact of detergent and severe wash cycles. The sample treated with eucalyptus ethanol (EE-UH) extract at 60 °C showed the highest mosquito repellency performance after washing among all 12 samples. After the rigorous wash cycles, the slight decrease in mosquito repellency is also evidence for the durability and efficacy of the applied finish.

■ DISCUSSION

The primary objective of this research was to implement the use of plant (eucalyptus and cinnamon) base natural finish for mosquito protection application in fabrics so that the ultimate clothing made from those fabrics can protect the wearer from the hazardous impact of mosquitoes. This study involved the construction of 48 distinct combinations of samples and the measurement of their effectiveness in repelling mosquitoes. The reaction mechanism of cinnamon and eucalyptus extracts with bleached cotton fabric samples is shown in Figure 12. The results of these studies showed that eucalyptus and cinnamon extract-based finishes are effective against mosquitoes and all of the samples showed some level of protection. Eucalyptus extract-based samples showed better repellency performance in all categories compared to cinnamon. This property of eucalyptus is associated with the intrinsic molecular behavior of this plant that shows strong adulticidal properties.[29] In terms of solvents, ethanol-based extract samples depicted promising results compared to those of water. Ethanol as a solvent illustrates better extraction properties compared to other solvents such as hydrocarbons and supercritical CO_2.[30] Consequently, all of the samples that used ethanol as a solvent exhibited encouraging outcomes in this study. Nevertheless, the impact of heating during the extraction mechanism was negligible in terms of their resultant samples' repellency performance for both eucalyptus and cinnamon categories. Although the extract produced through heating showed slightly increased repellency performance, that was not significant. The results also illustrated that application process temperature has a significant impact on the samples' performance. All of the fabric samples processed at 60 °C showed maximum mosquito

Table 5. Mosquito Repellency % of Washed Samples Treated with Unheated Extracts

			samples tested (mosquitoes landed) (average)			repellency (%)	
sr. no.	sample code	treated at temperature (°C)	one wash	five wash	total mosquitoes	one wash	five wash
1	CW-UH	40	17.00	18.50	30	43.30	38.30
		60	14.52	15.72	30	51.60	47.60
		80	15.76	18.57	30	37.46	32.53
2	EW-UH	40	9.00	10.00	30	70.00	66.66
		60	8.00	8.60	30	73.30	71.36
		80	9.16	9.90	30	66.13	63.30
3	CE-UH	40	12.34	13.36	30	58.86	55.46
		60	10.16	11.15	30	66.13	62.83
		80	16.00	17.3	30	46.66	42.33
4	EE-UH	40	8.00	9.00	30	73.33	70.00
		60	5.80	6.50	30	80.66	78.33
		80	10.16	11.00	30	66.13	63.33

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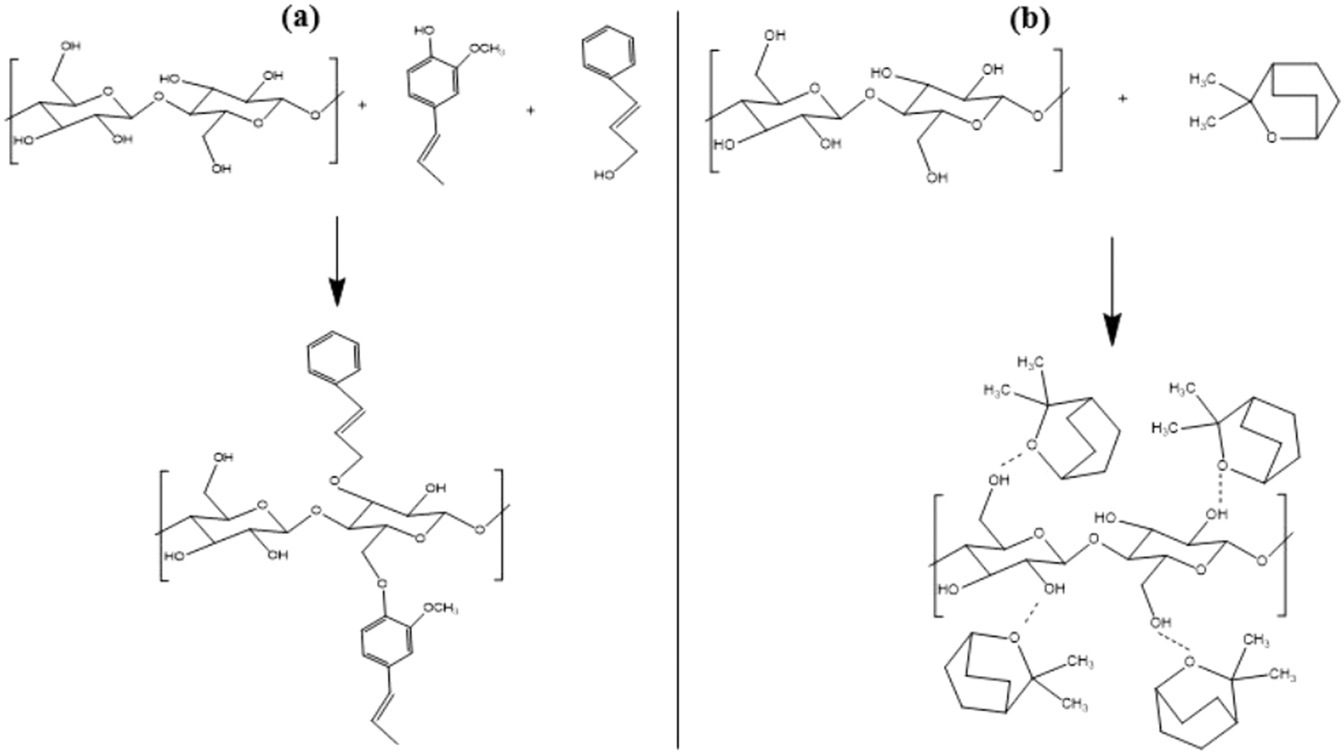

Figure 12. (a) Reaction mechanism of cinnamon with bleached cotton fabric; (b) reaction mechanism of eucalyptus with bleached cotton fabric.

repellency, beyond which a decline in performance was observed. This phenomenon is attributed to the swelling and reactivity behavior of cotton with temperature. Cotton fabric structure swells with the increase of temperature up to 90 °C and liquid penetration followed by chemical reactivity increases with the temperature rise.[31] However, it might be possible that degradation of cinnamon and eucalyptus molecules due to their high temperature sensitivity caused lesser-finish molecules to be available to deposit on the fabric samples at 80 °C temperature.[32] Furthermore, the boiling point of ethanol (78 °C) is another issue in all ethanol solvent-based extract-treated samples.[33] It evaporates at or beyond its boiling point, which affected the efficacy of the applied finish and repellency performance at 80 °C temperature. The impact of laundry washing was also analyzed to predict the durability of the developed plant-based natural finish. It is pertinent to mention that a slight gradual decrease in mosquito repellency was observed from one to five wash cycles. It occurred due to the impact of the detergent and the rigorous process of wash cycles. However, the slight gradual decrease in performance was evident in the durability of the applied finish. Furthermore, the fabric samples processed with the heated extract of eucalyptus ethanol (EE-H) at 60 °C showed the best results in terms of mosquito repellency (85.56%). In the statistical analysis of variance (ANNOVA), the extract type remained a significant factor in both the heated and unheated extract categories. However, the impact of temperature was not statistically significant for the heated extract samples, while it remained a significant factor for the unheated extract samples. Thus, this mixture can effectively be utilized to provide a biodegradable, skin-friendly, nontoxic, and environmentally friendly finish for mosquito repellency. The utilization of these finishes will enhance the reliance on plant-based finishes and ultimately encourage sustainable practices in the textile chemical processing sector.

FTIR Analysis. Fourier transmission infrared spectroscopy analysis (Figure 13) was performed on unheated extracts' fabric samples, which were processed at the optimum temperature parameter (60 °C). An unprocessed cotton fabric sample was also evaluated as a comparison to validate the finish application. The analysis showed that new peaks were formed between 3000 and 3500 cm^{-1} (O−H stretching vibration), 2700−3000 cm^{-1} (C−H stretching vibration), and 1500−1750 cm^{-1} (aromatic C═C group) wavenumbers in all extract-applied samples compared to the unprocessed cotton fabric sample. These peaks indicated the presence of O−H, C−H,

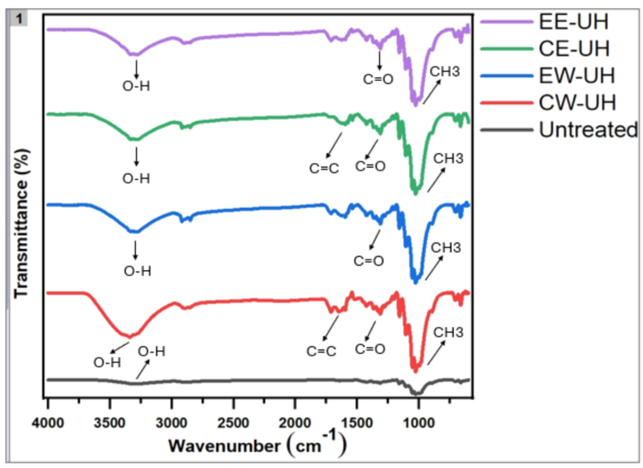

Figure 13. FTIR spectra of the samples (eucalyptus ethanol, cinnamon ethanol, eucalyptus water, and cinnamon water unheated extract applied at 60 °C compared to the untreated sample).

C−C, and C−O bonds in the processed samples, which illustrated the presence of functional groups of cotton, cinnamon molecule, and eucalyptus molecules in the applied finish. In the case of cinnamon, it contains a eugenol-based aromatic compound, which is responsible for its insect repellent property. Eugenol spectra show O−H stretching vibration, C−H stretching vibration, and C═C aromatic group peaks.[34] Therefore, the FTIR results of all cinnamon-based samples depicted the eugenol spectra and justify the mosquito repellency performance, which is directly associated with these functional groups. In contrast, eucalyptus contains a complex mixture of a variety of functional groups such as aromatic phenols, oxides, ethers, alcohols, esters, aldehydes, and ketones.[35] However, an aromatic terpene group is responsible for the mint-like pleasant fragrance of eucalyptus, which repels mosquitoes and makes it adulticidal. The hydroxyl (O−H) is the main functional group of aromatic terpene, which is responsible for the reactivity and spicy aroma smell of eucalyptus plant leaves.[36,37] Therefore, all eucalyptus extract-based samples showed a strong peak between 3000 and 3500 cm^{-1} (O−H stretching vibration) and validated the presence of the desired fragrance, which ultimately illustrates the achieved mosquito repellency performance.

Ultraviolet Protection Factor (UPF). A UPF test was conducted on fabric samples that were treated with unheated extracts and processed at an optimal temperature of 60 °C. An untreated cotton fabric sample was also evaluated as a reference to validate the induced functional application. All the processed fabrics show a UPF rating of 15 and higher, indicating effective protection against UV radiation (Table 6).

Table 6. UPF Results of Cinnamon and Eucalyptus Extract-Applied Fabric Samples

sr. no.	sample code	UVA blocking %	UVB blocking %	UPF rating
1	untreated	71.61	77.36	4
2	CW-UH	90.96	94.74	16
3	EW-UH	93.88	94.98	19
4	CE-UH	90.91	93.89	15
5	EE-UH	95.77	97.04	28

The fabric processed with eucalyptus ethanolic extract (UPF 28) exhibited superior outcomes compared with the fabrics processed with the water extracts of both agents. The discrepancy may be attributed to the variation in constituents extracted by the two systems, namely, water and ethanol.

Cinnamaldehyde, a key component of the cinnamon extract, is known to absorb UV radiation. By incorporating it into the fabric, cinnamaldehyde molecules can capture UV rays before they reach the wearer's skin. Both cinnamon and eucalyptus contain phenolic compounds, a class of natural molecules known for absorbing ultraviolet (UV) radiation. When applied to fabric, these compounds can act as a shield, absorbing UV radiation before it penetrates the fabric and reaches the skin.

Antibacterial Activity. The antibacterial activity of untreated and unwashed processed fabric samples was checked against the bacteria *S. aureus*. The results obtained reveal that the fabric samples processed with both extracts exhibited antimicrobial activity against *Staphylococcus* bacteria when applied to cotton fabric (Figure 14). However, there was no discernible difference in the activity displayed between the two extraction media. Meanwhile, the untreated sample exhibited no activity at all as no zone of inhibition can be seen around the fabric sample (Figure 14a). This antibacterial activity of processed samples may be attributed to the presence of compounds such as eugenol, cinnamon aldehyde, and other phenolic compounds in cinnamon extract and an aromatic terpene group in eucalyptus (justified by the functional groups identified in FTIR spectra), which impart antimicrobial activity to the fabric samples processed with cinnamon and eucalyptus extracts. These compounds are known to disrupt bacterial cell membranes, thus inhibiting the enzyme function, and ultimately causes bacterial death.[38,39] The antibacterial activity of cinnamon extract against a wide range of bacteria including MRSA is also reported. It implies that both extracts imparted good antibacterial properties along with mosquito repellency, which maintains the hygiene of the fabric.

■ CONCLUSIONS

This study involved the production and application of a natural finish derived from plant extracts on fabric samples in order to enhance their mosquito repellency properties. The natural extract was produced using cinnamon and eucalyptus as the

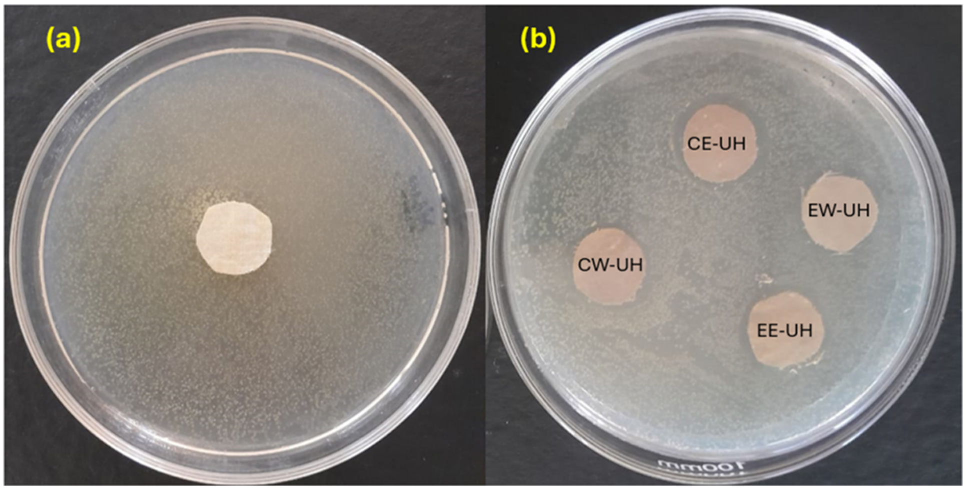

Figure 14. Antibacterial activity (a) untreated sample and (b) eucalyptus ethanol, cinnamon ethanol, eucalyptus water, and cinnamon water unheated extract applied at 60°c.

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herbal finishing agents. After application of various combinations, all of the samples were evaluated for their effectiveness in repelling mosquitoes, their antibacterial properties, and their ability to provide UV protection. The results demonstrated that eucalyptus and cinnamon extract-based finishes are efficacious in repelling mosquitoes, as all of the samples exhibited varying degrees of protection. Eucalyptus ethanol combinations depicted highest performance in all levels of application temperature (40, 60, and 80 °C). Among all combinations, the fabric samples treated with a heated extract of eucalyptus ethanol (EE-H) at a temperature of 60 °C exhibited the highest level of mosquito repellency, with a rate of 85.56%. An incremental decline in the effectiveness of mosquito repellency after 5 wash cycles was observed, which indicates the durability of the applied finish. The superior wash durability performance demonstrates that the fabric may be utilized for an extended duration without significant deterioration, and its repellency performance remains largely unaffected even after intensive wash cycles. The mosquito-repellent fabric that is being manufactured will provide extended protection to the wearer without the need for chemically based skin treatments. Furthermore, cinnamon and eucalyptus extracts showed promising results as natural UV protectants for textiles. The intrinsic ability of the fabrics to absorb UV radiation and potentially reflect light resulted in higher UPF ratings for the processed fabrics compared with the untreated fabric samples. Both extracts exhibited effective antibacterial capabilities and mosquito repellency, hence ensuring the fabric's cleanliness and hygiene. In conclusion, the inclusion of natural plant-based finishes in the textile chemical processing sector will encourage sustainable practices and mitigate the adverse environmental effects of manufacturing.

■ ASSOCIATED CONTENT

Data Availability Statement

The authors confirm that the data supporting the findings of this study are available within the article and its Supporting Information file.

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.4c04910.

Tables S1 and S2 present the results of statistical analysis (ANOVA) to validate the significance and non-significance of key variables, such as extract type and temperature, for both heated and unheated sample categories (PDF)

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Notes

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