



Development of mosquito-repellent and antibacterial nylon

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In the present study, a substantive derivative of N, N-diethyl-3-methylbenzamide (DEET), i.e. 4-amino-N, N-diethyl-3-methylbenzamide (DEET-NH₂), has been applied on nylon fabric by the exhaust process. The change in the appearance of the fabric after finishing and the efficacy of functional properties, like mosquito repellency and antibacterial activity, has been evaluated using standard methods. The finished fabric shows mosquito repellency (100%) and antibacterial activity (>90%), which are found durable until at least 10 washes.

Keywords: Antibacterial, Fibre technology, Mosquito-repellent, Nylon, Protective textile

Notwithstanding hundreds of years of control endeavours, mosquito-borne diseases are prospering around the world. Mosquito-borne diseases are liable for generous worldwide dismalness and mortality. Mosquitoes are attracted towards humans with the smell of lactic acid, gnawing one human to another human, thus acting as a vector for transmission of deadly diseases. A billion individuals experienced the ill effects of dengue caused with the assistance of *Aedes aegypti*. Not many vaccines are accessible to counter the mosquito-borne diseases¹. The best solution is to utilise the safeguards that can protect us from mosquito nibbles. Market contains lotions and cream for the counteraction of mosquito chomps. Direct utilisation of such creams can cause unpleasant feelings on human skin², and these salves also lose viability following a few hours of use.

DEET(N,N-Diethyl-3-methylbenzamide)is a well-known mosquito repellent and is most widely utilised in lotions and creams available in the market. The study suggested that lotions can be composed of 5-100% DEET³. DEET was first structured by the United States armed forces for the security of their

soldiers in mosquito-prone zones. DEET was utilised as a mosquito repellent compound in the pharmaceutical sector^{4,5}. For ensuring the slow release of DEET, the DEET- β -cyclodextrin complex was covalently attached to cellulosic fabric⁶. In order to achieve the prolonged mosquito repellent effect, microcapsules of DEET were made and applied to bed nets, which exhibited mosquito repellency for as long as a half year⁷. Microcapsules of DEET were prepared using chitosan and butyl acrylate polymer to accomplish antibacterial and mosquito repellent properties⁸. A reactive dye was synthesised with the help of DEET-NH₂ for imparting mosquito repellency to nylon fabric⁹. DEET-NH₂ and Bronner's acid were used for the synthesis of mosquito-repellent reactive dye¹⁰. DEET-NH₂ was reacted to cotton fabric using DMDHEU (Dimethylol dihydroxy ethylene urea) as a crosslinker¹¹. Copper-benzene-1,3,5-tricarboxylic acid was consolidated into the matrix of natural fibres, like cotton, wool, linen, and silk, followed by the incorporation of DEET in the previously modified fabric¹². DEET-NH₂ was also utilised for the *in-situ* synthesis of mosquito repellent antibacterial azoic colorants¹³. Recently, multifunctional disperse dye was prepared by reacting DEET-NH₂ with 4-hydroxy coumarin¹⁴. An outstanding mosquito repellency (100 %) was achieved on cotton fabric using DEET and a binder¹⁵.

A major problem associated with available mosquito repellents, like DEET, is that they lack substantivity towards the textile substrates and provide a poor attachment with the textiles in the absence of a cross-linker or binder. Mosquito repellent dyes and microcapsules were prepared from DEET to achieve durable mosquito repellency. However, a facile method of application of DEET for functional modification of textiles is still desired. The adequacy of mosquito-repellent textiles can also be upgraded by the chemical bonding of the mosquito repellent chemicals to the textile structure. Thus, mosquito repellents can be chemically modified to increase the substantivity towards textiles. This type of chemically modified mosquito-repellent can be directly attached to textiles without using any binder or cross-linker. Most of the works reported the attachment of DEET-NH₂ with textiles through a cross-linker or by preparing a dye molecule from

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DEET-NH₂. The utilisation of cross-linkers affects the handle properties of textile fabrics. The application of DEET-NH₂ to textiles without the use of a cross-linker has not been explored. Hence, motivation is generated to study the cross-linker-free finishing of textiles by using DEET-NH₂. Nylon fibre is generally used in home textiles, military items and apparel. As it is used in many technical applications, it is an ideal substrate for functional modification. To be best of our knowledge, no work has been accounted for the direct binding of DEET-NH₂ with nylon. Hence, we have explored a direct application of DEET-NH₂ to nylon using an exhaust process of finishing, resulting in a wash-durable mosquito repellent-cum-antibacterial nylon. The change in the appearance of the fabric after finishing was studied. The efficacy and durability of both functional properties were also analysed.

Experimental

Nylon 6 fabric (EPI96, PPI84, and GSM60.3) was purchased from the market. The fabric was washed with non-ionic detergent (Lissapol N) before its use for further treatments. Acetic acid was purchased from Sigma Chemicals. All reagents/chemicals were used as received without any purification.

The preparation of DEET-NH₂ was done according to the previously reported method¹⁴. DEET-NH₂ stock solution was prepared in dilute acetic acid at pH 4.5-5. Nylon fabric was treated with DEET-NH₂ (1%, 3%, and 5% owf) in rota dyer with a material-to-liquor ratio of 1:10 at 90°C for 1h. After the treatment, fabrics were dried in the air. The finished nylon samples were evaluated for colour coordinates (L*, a*, b*) and colour strength (K/S) under light D65 and 10° observer using a spectrophotometer. The fabric was washed using the technique as per standard AATCC61-A¹⁶. The antibacterial activity of nylon samples was evaluated according to the AATCC-100 test method¹⁶.

The mosquito repellency was estimated according to the 'arm in cage method' against Anopheles female mosquitoes¹⁴. A cage filled with mosquitoes was used to evaluate mosquito repellent testing of fabrics. Untreated and treated fabrics wrapped on the user's arm were kept in the cage, and repellency was calculated using the following formula:

$$\text{Mosquito repellency} = (U - T)/U$$

where *U* indicates the number of mosquitoes landing on untreated nylon; and *T*, the number of mosquitoes landing on treated nylon.

Results and Discussion

Colour Coordinates, CIE Whiteness index, and Colour Strength of Finished Nylon

Figure 1 shows the change in the CIE whiteness index (CIE WI) of nylon after the DEET-NH₂ treatment. The decrease in the whiteness of nylon after DEET-NH₂ treatment can be clearly seen. The increase in K/S values of nylon samples after DEET-NH₂ treatment can also be concluded (Table 1). A decrease in L* and an increase in a* and b* values are observed with DEET-NH₂ treatments. Figure 2 demonstrates the probable mechanism of ionic interaction. Aromatic amines tend to generate a positive charge (like basic dye) in the water at acidic pH. Nylon fibre is amphoteric and has both negative and positive charges, depending on the pH. Both charges are in equal quantity at the isoelectric point (4.5 pH). Thus, NH₃⁺ from DEET-NH₂ can make ionic bonding with the COO⁻ group of nylon (Fig. 2).

Table 1 — Colour coordinates, colour strength (K/S) of nylon samples

Treatment of DEET-NH ₂ (% owf)	Colour coordinates ^a			K/S ^a
	L*	a*	b*	
0	89.43	1.13	-4.03	0.075
1	87.65	2.34	-3.56	0.139
3	86.45	3.65	-2.24	0.206
5	85.34	4.32	-1.23	0.299

^aAverage value of three determinations.

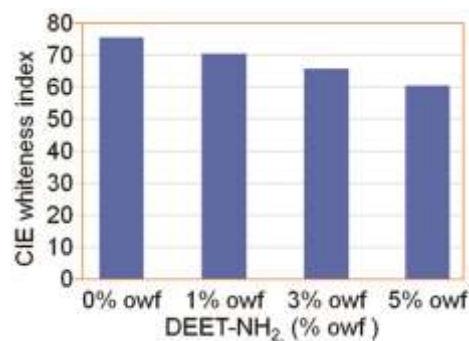


Fig. 1 — CIE whiteness index of DEET-NH₂ treated nylon fabric.

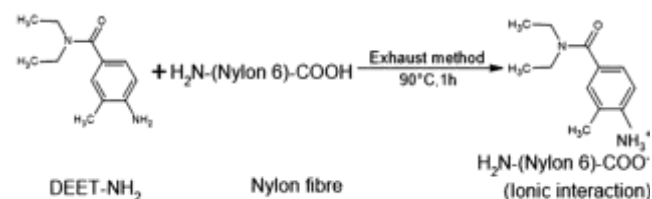


Fig. 2 — Possible mechanism of interaction between DEET-NH₂ and nylon.

Mosquito Repellency of Finished Nylon

A mosquito repellency of 61 % is obtained for the fabric treated with 1 % (owf) DEET-NH₂. The nylon sample finished with 3 % and 5 % (owf) DEET-NH₂ shows 100 % mosquito repellency. Moderate mosquito repellency is imparted to nylon with the application of 1 % (owf) DEET-NH₂, while excellent repellency (100 %) is obtained at 3 % and 5 % (owf) concentrations. Hence, 3 % amount of DEET-NH₂ is sufficient to provide excellent mosquito repellency.

The insignificant loss in mosquito repellency of finished nylon is observed after washing treatment, and the retention of 98% mosquito repellency at 3% (owf) DEET-NH₂ is observed after 10 washes. Minor loss in mosquito repellency after the washing treatments can be due to the removal of a small quantity of DEET-NH₂ from the fibre. The results indicate an increment in mosquito repellency with the increase in DEET-NH₂ concentration. The nylon fabric finished with 5% owf DEET-NH₂ retains 100% mosquito repellency even after 10 washes. DEET-NH₂ is the chemically altered structure of the DEET. On contact mode of action, the presence of moiety [(CH₃-CH₂)₂-N-C=O] makes finished nylon unfavourable for mosquito attack. Charges on oxygen and nitrogen play a significant role in mosquito repellency¹⁷. The dipole moment of the modified structure is also a vital factor in the resultant mosquito repellency. The phenyl ring can also upgrade the repellency. In the generalising term, positive potential charges can interact with negative charges of the receptors of mosquito¹⁷. A balanced polarity of modified structure (mainly nitrogen and oxygen) is responsible for mosquito repellency.

Antibacterial Activity of Finished Nylon

Finished fabrics show antibacterial activity of 99.65 %, 98.43 %, and 99.75 % against *S. aureus* at 1%, 3 % and 5 % (owf) DEET-NH₂ concentrations respectively. Similarly, finished fabrics show antibacterial activity of 97.54 %, 98.54 % and 96.76 % against *E. coli* at 1%, 3 %, 5 % (owf) DEET-NH₂ concentrations respectively. Hence, 1 % DEET-NH₂ is sufficient to impart excellent antibacterial activity (<97%) to nylon.

Even after 10 washing treatments of the sample finished with 5% (owf) DEET-NH₂, the sample retains antibacterial activities of 95.63 % against *S. aureus* and 92.45% against *E. coli*. This indicates the wash-fast attachment of DEET-NH₂ with nylon

fabric. The loss in antibacterial activity after washing might be due to the removal of a small quantity of DEET-NH₂ from the fabric surface. DEET-NH₂ must be acting as a poison for the bacterial cells (bactericidal), leading to reduced bacterial colonies. This might be due to the positive charge of DEET-NH₂ interacting with a negative charge on the bacteria surface. DEET also possesses antibacterial activity¹⁸. A consolidated impact of the DEET moiety structure and the amino group of DEET-NH₂ can impart efficient antibacterial activity to finished nylon.

A novel method of mosquito-repellent-cum-antibacterial finishing of nylon using DEET-NH₂ is established. Good interaction between nylon and DEET-NH₂ is confirmed. The finished nylon shows an excellent mosquito repellent action along with antibacterial properties. Both the functional properties are durable until at least ten washes. Nylon is a vital textile fibre, and a wash-durable, mosquito repellent and antibacterial nylon is developed. Such an approach does not need binding agents and can be easily used to make mosquito repellent-cum-antibacterial apparel, tent fabrics, and army uniforms.

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