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Development of mosquito repellent cotton using marigold

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Abstract
From centuries man has always concerned about his protection which then leads to number of developments in every field with regards to textile. Global warming is the cause of the distribution of mosquitoes which has expanded from tropical regions to northern latitudes and that leads to a spread in sources of viral infection from mosquitoes. Hence, personal protective measures must be taken to protect human from mosquito bites**.** The major tool in mosquito control operation was the application of synthetic insecticides. But this has not been very successful because it has harmful effect on human health i.e. unpleasant smell; oily feeling to some users and potential toxicity. Using natural plant products is a simple and sustainable method of mosquito control. Repellents of plant origin do not pose hazards of toxicity to human and domestic animals and are easily biodegradable. Enhancing the health and hygiene qualities of consumer product make it necessary to find a new ways of applications on textiles with medicinal plants. Marigold (petals) methanol extract was used for mosquito repellent finish on woven cotton. Fabric was enzymatically desized, scoured and bleached. For mosquito repellent finishing on fabric two application techniques i.e. direct and resin cross-linking were used through pad-dry-cure method. Mosquito repellent finished samples were tested against mosquitoes for evaluating their efficacy and durability to washing and sun-drying.

Keywords: Application techniques, marigold, methanol extract, mosquito repellent finish, pad-dry-cure method

1. Introduction
After fabricating the mansions of fashion and comfort, textiles are now moving towards high-tech era of performance, which has brought up diversification and expansion of technologies. Mosquitoes are the major vectors for the transmission of various tropical and subtropical diseases which cause devastating effects to human. The most common dreadful diseases associated with mosquitoes are malaria, yellow fever, filariasis, schistosomiasis, japanese encephalitis (JE) and dengue hemorrhagic fever. Therefore, the studies in search of novel entities from plants to prevent proliferation of mosquito borne diseases and to protect environment from the application of chemical pesticides, the mosquito control is essential. There has been recent concern from consumers in clothing and textile materials to provide fabrics and products with greater wearing comfort while remain hygienic and odor free in use (Shahid and Mohammad, 2013) [9]. Natural products are safe for human when compared to the synthetic compounds. Therefore, it is the hour to launch extensive search to explore eco-friendly biological materials for control of insect pests. According to studies, plants are considered to be the primary source of natural bio-acting agents because of their numerous active compounds including simple phenols, phenolic acids, quinones, flavonoids, flavones, flavonols, tannins, coumarins, terpenoids, essential oils, alkaloids, lactins and polypeptides (Murugesh and Ravindra, 2015) [5].
The textile sector is facing new challenges in the modern days and every technician is giving the best to face these challenges. The world that would lead us would be astonishingly hi-tech and materialistic. To ensure our security and safety from the future hazards, we need to equally develop the technology for our protection. Protective textiles are among one such smart application of smart technology in textiles. Protective textiles refer to those textiles products which have a functionality of giving protection from something in some or the other sense.

Mosquito repellent finished textiles are also a part of protective textiles which help in protection from the species that are prone to cause damage in some or the other manner (Saraf and Alat, 2005) [8].

2. Materials and methods

The present study was undertaken to develop mosquito repellent cotton with application of marigold (petals) methanol extract using direct and resin cross-linking application techniques by pad-dry-cure method.

2.1 Selection and procurement of raw materials and auxiliaries

100 percent woven cotton commonly used by consumers for apparels was selected and procured from market of Hisar city, Haryana state. Different eco-friendly and laboratory grade auxiliaries were selected for the study on the basis of easy availability and cost effectiveness. Citric acid was used as cross-linking agent in padding bath for direct application technique. Fixapret F-eco was used as resin cross-linking agent and magnesium chloride as catalyst, as it allows the resin cross-linking reaction to be carried out within the temperature and curing range usually employed in textile industry.

2.2 Preparation of selected fabric and plant material

Americos amylase 543 for desizing, Palkoscour APCL enzyme for scouring and Peroxyme for bleaching of cotton fabric were used.

The collected fresh petals of marigold were washed and allowed to dry in shade to avoid breakdown of important compounds. Grinding of dried petals of marigold was done in grinder and mixer and the powder was sieved to remove the dirt and unkind particles. The dry powder of petals was subjected to methanol extraction using soxhlet process.

2.3 Application of mosquito repellent finish on fabric

(i) Direct technique: 10g/l marigold (petals) extract, 5 percent citric acid was mixed in 1:20 MLR and fabric immersed for 20 minutes. Padding was done in padding mangle at 2.7psi pressure and fabric was dried in preheated oven at 80 ^{0}C for 5 minutes and cured at 120 ^{0}C for 60 seconds.

(ii) Resin cross-linking technique: 40g/l Fixapret F-eco and 8g/l magnesium chloride mixed along with 10g/l marigold (petals) extract in 1:20 MLR and fabric immersed for 20 minutes. Padding, drying and curing parameters were same as per direct technique.

2.4 Assessment of efficiency of finish

The finished fabric was evaluated against mosquito repellency by modified cage method (Prabha and Vasugi, 2012) [6].

a. Mosquito rearing

Mosquito reared in laboratory under controlled conditions i.e. 28±2 ^{0}C temperature with 60 to 70 percent RH. *Anopheles Stephensi* female mosquitoes were identified based on morphologic keys and was collected during the evening hours in specially designed organdie fabric constructed cage measuring 12”×12”×12”.

Resin (wet kishmish) as food was placed inside the cage and 10 percent glucose wet pad on upper facets of the cage were kept overnight. The females hatched two days after they fed. Hatched larvae was filtered and placed in a white coated tray having boiled water at normal temperature and added a pinch of fish food (tetra bits/aquaricare) in that tray. Larvae were shifted in two more trays for better growth. Whole procedure was repeated until the pupae bent and left their outer skin in water. Collected the pupae and placed the beaker in an empty cage provided with resin and glucose. Pupae were allowed to emerge to adults for 2-3 days.

b. Repellency behavioral test

Laboratory tests were performed during daylight to evaluate the efficiency of repellency activity and each test was replicated three times. Mosquitoes were deprived/starved of all nutrition and water for a minimum of four hours before exposure. For conducting experiment, two controlled samples were hung on the opposite facets of an empty cage and two finished samples on another opposite facets of that cage. Upper facet was used for observing the mosquitoes (specimens). 50 mosquitoes were exposed in each cage. Number of mosquitoes on controlled and finished samples were noted down at every 15 minutes of interval for 60 minutes and calculated from the following formula:

$$\text{Mosquito Repellency (\%)} = \frac{\text{Number of specimen on controlled sample}}{\text{Total exposed specimen in the cage}} \times 100$$

2.5 Assessment of durability of finish: Assessment of durability of finished cotton samples to washing as per the recommendation of IS: 3361-1979 and sun-drying was tested using IS: 686-1985 standard test method in the month of June (temperature 45±2 ^{0}C).

3. Results

3.1 Assessment of efficacy of finish

The mosquito repellency in terms of repelled mosquitoes from the finished samples and settled on the controlled samples. The data related to assessment of repellency against mosquitoes is presented in Table 1.

It is clear from Table 1 that with direct application technique, repellency was 96 to 98 percent whereas with resin cross-linking technique repellency was 94 to 96 percent after 60 minutes.

Table 1: Efficacy of mosquito repellent finish n=50

Time of observation (mins)	Application Techniques					
	Direct		Repellency (%)	Resin cross-linking		Repellency (%)
	C	T		C	T	
15	48	2	96	47	3	94
30	48	2	96	47	3	94
45	48	2	96	48	2	96
60	49	1	98	48	2	96

C -- Number of mosquitoes settled on controlled sample,
T -- Number of mosquitoes settled on treated sample,
n= no. of mosquitoes exposed in each cage

3.2 Assessment of durability of finish

The mosquito repellency durability to washing and sun drying of mosquito repellent finished fabric is of considerable importance to the consumers for longer life of repellency. The data related to durability of finish is presented in Table 2 and 3.

(a) Durability to washing: Finished samples were assessed for repellency of mosquitoes after washing on the basis of laboratory cage method. It is evident from Table 2 that with marigold (petals) on cotton fabric, repellency was observed by direct (70%); by resin cross linking technique (76%) after 5 wash cycles. However, mosquito repellency was durable till 10 wash cycles in direct and resin cross-linking techniques treated with marigold (petals) extract as reported by World Health Organization, 1996 that if any mosquito repellent finish given to the textile material declines below 50 percent, finish is not acceptable.

Hence it can be concluded from Table 2 that as the wash cycles increased the durability of finish decreased by both the application techniques.

Table 2: Durability of finish after washings, n=50

Application Techniques	Wash cycles	Durability to washing (%)
		Repellency (%)
Direct	0	98
	5	70
	10	52
Resin cross-linking	0	96
	5	76
	10	56

0– treated control sample (without washing), one wash cycle- 45minutes
n= no. of mosquitoes exposed in each cage

(b) Durability to sun-drying: Data revealed from Table 3 that with direct application technique, mosquito repellency of finish was durable till 2 hours of sun drying whereas till 3 hours with resin cross-linking technique.

Table 3: Durability of finish after sun-drying, n=50

		Durability to sun-drying (%)
Application Techniques	**No. of hours**	**Repellency (%)**
Direct	0	98
	1	66
	2	56
Resin cross-linking	0	96
	1	74
	2	60
	3	50

0– treated control sample (without sun-drying)
n= no. of mosquitoes exposed in each cage

In direct technique, repellency was observed 66 and 56 percent after exposure in sun for 1 and 2 hours respectively. In resin cross-linking technique, 74 percent repellency was observed after 1 hour which was decreased after 2 and 3 hours i.e. 60 and 50 percent.

4. Discussion

4.1 Direct technique: For preparation of padding bath, 5 percent citric acid was used and mixed with 10g/l of plant extract. Thilagavathi *et al.,* (2007) [11] imparted cotton fabric using extracts of neem and Mexican daisy by direct application using pad-dry-cure method. Results are supported by the findings of Rani (2014) [7] as stated that 6 percent citric acid in padding bath with MLR 1:20 and 30 minutes of treatment time were optimized for direct application of essential oils on woven and knitted cotton fabrics by pad-dry-cure method.

4.2 Resin cross-linking technique: For application of mosquito repellent finish through resin cross-linking technique on cotton fabric, proportion of non-formaldehyde based eco-friendly 40g/l resin cross-linking agent i.e. Fixapret F-eco and 8g/l compatible catalyst i.e. magnesium chloride was mixed with 10g/l plant extract for preparation of padding bath. Results of Rani (2014) [7] supported the result as four essential oils were treated through resin cross-linking technique on woven and knitted cotton fabrics by pad-dry-cure method. Thite and Gudiyawar (2015) [12] imparted to the cotton fabric using citronella oil by resin finishing using dip-dry-cure method. Gupta (2016) [4] also studied on woven and knitted cotton fabrics with guava methanol extract using resin cross-linking application technique.

4.3 Efficacy assessment of finished fabric

Results of Table 1 show that cotton fabric samples finished with direct technique using marigold (petals) showed maximum repellency (96 to 98%) between 15 to 60 minutes observation time followed by samples finished with resin cross-linking technique (94 to 96%). It might be due to better adhesion of extract in direct technique with cross-linking agent reaction whereas with good catalyst reaction with extract in resin cross linking technique. Study carried out by Bhanupriya and Maheshwari (2013) [2] revealed that rosemary (herbal) and toluamide complex (conventional) treated samples repelled 92 and 84 percent mosquitoes treated in excito repellency test chambers. Anuradha *et al.* (2016) [7] tested the marine seagrass of *Halophila ovalis* against the *Culex quinquefesciatus* and found 95 percent protection up to 4 hours at 250 percent concentration of *Halophila ovalis* extract followed by 200, 150 and 100 percent concentration of extract.

4.4 Durability assessment of finished fabric

The mosquito repellency durability was assessed after washing

and sun drying. Mosquito repellent finish with direct and resin cross-linking techniques was durable till only 10 wash cycles. Mosquito repellency was gradually decreased after every wash cycle with both application techniques. It may be due to the reason of repeatedly washing may cause the removal of plant extract and other auxiliaries bonded with fabric structure. Maximum repellency of finished cotton was found after 2 hours exposure in sun treated with resin cross-linking followed by direct technique. No difference was observed in repellency after 2 and 3 hours exposure in sun in finished samples using resin cross-linking technique. It may be due to an interaction between the hydrocarbon, oxide of nitrogen and oxygen that cause release of core material. Sumithra and Raja (2012) [10] reported that 100 percent cotton fabric showed good efficiency of finishes even after 30 industrial washes as compared to other three blended denim fabrics. Geethadevi and Maheshwari (2015) [3] reported that the fabric treated with natural repellent using exhausted method from combination of essential oil such as thyme oil, cypress oil, grapefruit oil applied on bamboo/tencel fabric with 50/50 presented the longer efficacy of mosquito repellence up to 30 washes with no allergic reaction to wearer.

5. Summary and conclusion

The present study 'Development of mosquito repellent cotton using marigold' was carried out to see the efficiency and durability of the finish and with its application on cotton.

Enzymatic desizing, scouring and bleaching were done to prepare the fabric for mosquito repellent finish. Marigold (petals) was selected on basis of mosquito repellency properties. Extraction was done in methanol to get the extract of plant material. The fabric was treated with plant extract using two application techniques i.e. direct and resin cross-linking techniques. For direct technique, the concentration of citric acid was 5 percent and for resin cross-linking technique, 40g/l Fixapret F-eco with 8g/l catalyst proportion along with 10g/l extract, MLR 1:20 and treatment time 20 minutes. The optimum temperature for drying were 80 ^{0}C for 5 minutes and curing temperature were 120 ^{0}C for 60 seconds.

Highest efficacy was observed in samples using direct technique followed by resin cross-linking.

Durability to washing was observed till 10 wash cycles using both techniques. Durability to sun-drying was till 2 hours exposure in sun using direct technique whereas till 3 hours using resin cross-linking technique.

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