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Anti-moth efficacy of neem (*Azadirachta indica* A.Juss.) on woollen fabric

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Neem (*Azadirachta indica*) is well known medicinal plant in Indian subcontinent. It has wide spectrum of biological activity as antifeedant and acts as insecticides. An attempt has been made to study efficacy of Neem extract as anti-moth agent and natural dye. Results reveal that anti-moth efficacy of bark extract found better than neem leaves extract dyed fabric. In both case stannous chloride mordanted fabrics shows anti-moth properties similar to commercial anti-moth agents due to formation of organotin compounds. Both leaves and bark extracts also have very good dye ability for wool fabric. However, fastness to light and washing are improved after mordanting.

Keywords: Anti-moth efficacy, Azadirachtin, Natural dye, Neem, Wool

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The infestation of carpet beetle on woollen fabrics due to their keratinous nature has an economic loss. *Tinea pellionella* has been found as an important pest of woollen textiles. In a study on incubation, larva development and damage by cloth is bearing moth. Cheema[1] elucidated that both temperature and humidity have been found to influence the larval development and the number of larval instars. High humidity of 90% at 25 °C favour short duration of larval development and the shortest larval period. The behaviour of the moth larvae observed to direct sunlight, diffused light alternated to darkness and continuous darkness. He found that the amount of damage was significantly higher for continuous darkness compared to alternating light and darkness.

The conventional method of preventing and controlling clothes infestations is by the use of insecticides. The synthetic pyrethroids, permethrin and cyperethrin are the main active ingredients of moth-proofers. Woollen goods are treated with permethrin yarn-dyeing stage to provide long-term protection against moth attack. The main problems associated with the use of these conventional insecticides to control clothes moths are toxicity to non-target organisms in the environment, including humans, and the development of resistance in the pests. In the view of the above fact, there is need for a natural and sustainable alternatives for protection of woollens from different cloth bearing moth.

Neem (*Azadirachta indica* A. Juss.) is the most versatile medicinal plant having a wide spectrum of biological activity and part of human society to combat diseases, from the dawn of civilization. Joshi *et al.*[2] reviewed natural product based bioactive agents such as chitosan, natural dyes, neem extract and other herbal products for antimicrobial finishing of textile substrates. Chandrasekaran *et al.*[3] confirmed the presence of bioactive compounds in the herbal extracts using High Performance Liquid Chromatography (HPLC) test. They also confirmed the curative healthcare, antimicrobial and antibacterial efficacy of these products.

By structural and chemical composition, neem is a diversified plant and about 135 compounds have been isolated from different parts of Neem. These compounds are divided in two major classes of isoprenoids and others[4]. The isoprenoids include diterpenoids and triterpenoids containing protomeliacins, limonoids, azadirone and its derivatives, gedunin and its derivatives, vilasinin type of compounds and csecomeliacins such as nimbin, salanin and azadirachtin. Phytochemical studies founded that neem contained significant quantities of saponin and cyanide though in moderate levels which is not deleterious to human but effective to moth and beetles[5].

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Neem extracts have been use to affect more than 200 insect species by various ways including repelling adults and larvae, disrupting developmental processes, inducing adult sterility and disturbing adult behaviour. The main active ingredient isolated and identified in neem extracts is azadirachtin. It reduce the manifestation of insects at slow rate including complete or partial antifeedant response, delayed and/or disrupted moulting and inhibited reproduction by reduces fecundity and fertility and shortens the life span of adult insects[6-8].

Gerard & Ruf[9] also observed different modes of action of neem extract on survival and feeding of larvae of four keratinophagous insects. Neem extract reduced feeding damage on woollen cloth to below accepted limits of International Wool Secretariat (IWS) for insect resistance at rates of 2.6 mg azadirachtin/gm for *H. pseudospertella* and 1.3 mg/gm for the remaining species. *Azadirachtin* has potential as a control agent for keratinophagous insects and may provide an environmentally acceptable alternative to current insect-proofing treatments.

Woollen are premium apparels, natural dyeing will further value added to the fabric. Hence, in the present study, an attempt has been made to impart moth proofing properties to woollen fabrics with the extract obtained from Neem leave and bark which may also provide colour on the woollen fabrics.

Materials and methods

Materials

The leaves and bark of Neem tree were used for dye source and anti-moth agent. The leaves and bark of neem tree are collected from ICAR-Central Sheep and Wool Research Institute campus and dried on wooden platform in the lab. Pure wool fabric was mild scoured with a 0.5 gm per litre non ionic detergent at 50 ^{o}C for 30 min, washed with warm water, rinsed with cold water and dried at ambient condition. $Al_2(SO_4)_3$, $SnCl_2$, and $FeSO_4$ were used as mordants. All other chemicals were of LR grade.

Colourant extraction

A quantity of 100gm of pulverized neem leaves and bark were soaked overnight in five litres of water. The colourant was extracted from leaves/bark solution at 100^{o}C for 60 min.

Dyeing

The woollen fabric was dyed with 5% colorant extracted from leaves/ bark (owm) at 100 ^{o}C with 1:40 material to liquor ratio at *p*H 5-6 in presence of 0.5 gm/L acetic acid solution for 1 hr in a water bath. This will followed to meta-mordanting with 3% mordant (owm) as per procedure described else ware[10].

Colorimetric and fastness properties

The colorimetric values L, a, b, and K/S of dyed samples and detergent washed samples were evaluated using a JAYPAK4802 Colour matching system (Jay Instruments Ltd, Mumbai, India) at D65 illuminate/ 10^{o} observer fitted with spectrophotometer. The light and washing fastness of the dyed samples was evaluated as per standard procedure ISO 105-A02[11].

Anti-moth properties

The dyed woollen fabrics along with un-dyed (control) sample were kept in petri dishes by adding ten alive adult carpet beetle moths as per method described by Kato *et al.*[12]. Petri dishes were kept in incubator for 15 days at the temperature range of 30-35°C and 50-60% of relative humidity. After that, the anti-moth properties were characterized based on weight loss in the fabric due to moth attack, visual examination of fabric for damage and number of moths alive after the test. A synthetic anti-moth chemical Eulon treated wool fabric was also kept in similar condition and evaluated to compare the results.

Result and discussion

Dyeing behaviour

The alkaline extract (0.1% Na_2Co_3) for neem leaves and bark observed to 2.76% and 11.64%, respectively. Since, both neem leaves and bark has good quantity of flavonoids which may yield yellow dyes giving brilliant and fast yellow colours on wool[13]. Fabric samples were dyed with colourant alone and with mordants, viz. Aluminium sulphate, tin chloride and ferrous sulphate by meta-mordanting to obtain different shade. The different shades produced are given in Table 1.

Colorimetric coordinates of the dyed fabric and colour strength (K/s)

The colour coordinate values as measured by the spectrophotometer fitted with computer colour matching system, of the dyed fabrics with colour extracts in different medium are given in Table 2.

Table 1—Shades produced by Neem leaves and bark extract

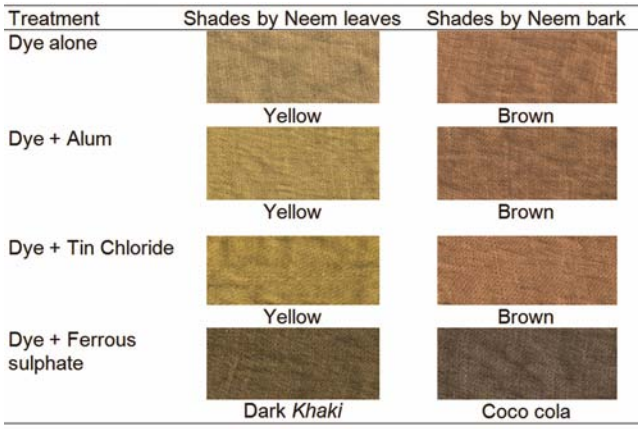

Table 2—L*, a*, b* and colour strength value of dyed woollen fabrics

Particulars	L*	a*	b*	K/S
Substrate	87.91	46.29	27.18	0.742
		Neem leaves		
Dye alone	77.88	42.76	31.86	1.111
Dye + Al	77.68	43.56	34.23	2.005
Dye + Sn	78.44	44.56	38.72	3.540
Dye + Fe	57.04	0.08	10.72	2.018
		Neem bark		
Dye alone	72.77	46.52	27.15	2.055
Dye + Al	71.10	45.02	26.23	2.303
Dye + Sn	73.01	46.90	28.14	3.49
Dye + Fe	63.17	34.97	17.59	1.059

The L* value of the dyed fabric has not observed significant change with mordenting using alum or tin chloride. Whereas, it has reduced after mordanting with ferrous sulphate. The lower L* value by 25-30% for different mordanted fabrics indicates the darker shades compared to un-mordanted fabric. This attributed to dye-metal coordinate complex which reduces the lightness of dyed textiles with modification in the chrome of the shade. The a* and b* values of the dyed samples have also change marginally after mordanting with alum and tin chloride. Although the fabrics appeared same yellowish green and brown tone with change in hue. After mordanting with ferrous sulphate Dark khaki and Coco-cola shades were produced for neem leaves and bark, respectively. The change in tone, viz. a* and b* values showed shade change from reddish to green and yellowish to blue for both.

The K/S values of woollen fabrics are found higher for mordanted samples as compared to dye extract alone. The highest values of 3.54 and 3.49 are observed for fabrics mordanted with Stannous Chloride. This may be attributed to higher aggregation of dye-metal complex during mordanting with tin chloride as compared to other mordants[14].

Table 3—Light and washing fastness of woollen fabrics dyed with neem leaves and bark

Fabric dyed with	Neem leaves		Neem bark	
	Light	Washing	Light	Washing
Dye extract alone	3/4	4	4	4
Dye extract + Al	3	5	4	5
Dye extract + Sn	2/3	5	¾	5
Dye extract + Fe	3/4	4	¾	4

Fastness properties

Colour fastness is the resistance to change in any of its colour characteristics of dyed fabrics. Most of the natural dyes have moderate to good, wash and light fastness properties on woollens. The light and washing fastness of woollen fabrics dyed with colour extracts obtained from neem leaves and bark are given in Table 3.

From the results, it is observed that light fastness of fabric dyed Neem bark extract is falling between 3/4 to 4, which is better than compare to neem leaves dyed fabric observed fair to good (2/3 to 3/4) light fastness properties. The washing fastness of dyed fabrics without mordant is very good. The fastness to washing improves from very good to excellent for meta-mordanted dyed fabrics with alum and tin chloride. Whereas, fabrics dyed using ferrous mordant are not observed any significant change in washing fastness property. The metal complex bonding formation between dye and mordant is attributed to better fastness properties.

Anti-moth efficacy of neem leaves and bark extract

Among the various limonoid components of the neem, Azadirachtin ($C_{35}H_{44}O_{16}$) is the main active ingredient of Neem. It is a triterpenoid limonoid obtained from various parts of the neem tree and has many anti infective and anti microbial properties[15]. It also exhibits antifeedant, insect repellent and insect sterilization properties[4]. Insects treated with Azadirachtin during the larval and pupal stages, comprising 60-70 % of their lives, generally die within 3-14 days. It interferes to key insect molting hormone and prevents larvae and pupae from completing the molting process. In HPLC quantification of azadirachtin and nimbin

Table 4—Anti-moth properties of neem bark and leaves extract

Particulars	Weight loss (%)	Moths alive	Visual examination
Neem bark extract	0.41	20%	Not damaged
Neem bark extract + Aluminium Sulphate	0.29	Nil	Not damaged
Neem bark + Tin Chloride	0.27	Nil	Not damaged
Neem bark + Ferrous Sulphate	0.56	10%	Least Attacked
Neem leaves extract	1.40	40%	Attacked
Neem leaves extract + Aluminium Sulphate	0.57	10%	Least Attacked
Neem leaves + Tin Chloride	0.54	Nil	Least Attacked
Neem leaves + Ferrous Sulphate	1.01	20%	Attacked
Commercial Anti-moth (Eulon)	0.17	Nil	Not damaged
Control	16.11	70%	Attacked

in different neem parts in different solvent was reported by, Ghimeray *et al.*[16]. They observed high concentration of Azadirachtin and Nimbin in neem bark compared to neem leaf, i.e., 5 times and 2 times, respectively. Though, highest concentration of Azadirachtin has been found in Neem seed which was double to the quantity in neem bark in Hexane.

In Phytochemical analysis of different neem part by Asobara *et al.*[17]. It was found to higher concentration of alkaloid, flavonid, phynate and saponin in neem leaves. Whereas, level of cyanogenic glucosides (HCN) was almost twice in neem bark. Results for anti-moth efficacy of wool fabric dyed with neem leave and bark extracts with and without mordant are given in Table 4.

Anti-moth efficacy of neem leaves

Anti-moth efficacy in terms of weight loss due to moth attack is found poor (1.4%) for fabric dyed with neem leaves extract without mordant, as compared to commercial anti-moth product Eulon (0.17%). The low anti-moth efficacy is attributed to moderate tannin content in neem leaves and lower content of azadirachtin, nimbin and cyanogenic glucosides (HCN) as compared to neem bark. This result to less effectiveness of anti-feedent properties with neem leaves extract dyed fabrics and incubated carpet beetle may digest these organic compounds and survived as observed to 40% alive moths after 15 days of incubation for fabric dye alone[18]. The anti-moth efficacy is found to improve after mordanting which gives comparatively lower fabric weight loss and 0-20% alive moth on the completion of incubation period. Meta-mordanted neem dyed fabric with Alum and Ferrous is observed to fabric weight loss in range of 0.6-1.1%. However, it is exceptionally very good for fabric mordanted with tin chloride, i.e., least fabric weight loss (0.27-0.54%) with none of the moth alive. This is due to formation of organotin compound with reaction of azadirachtin and cyanogenic glucosides with Sn^{2+} ion. Formation of organotin compound is confirmed by Fourier Transform Infra Red (FTIR) Spectra (Fig. 1). In FTIR spectra of Neem leaves and tin chloride mordanted fabric have strong peaks appeared at 468 and 453 cm^{-1} assigned to Sn–C and Sn <–N, respectively. But these peaks are not present in the FTIR spectra of grey fabric and fabric dyed with Neem leaves extract alone. Organotin compound disrupt oxidative phosphorylation by inhibition of the mitochondrial ATP synthase enzyme[19]. Organotin compound is block the bioenergetics system and results to demise of moth.

Anti-moth efficacy of neem bark

Neem bark extract with and without mordant showed very good anti-moth properties. It has to found less than 0.3% loss in fabric weight with death of all the moth placed for incubation with fabric dyed with bark extract and mordanted with alum and tin chloride. Whereas, fabric dye with extract alone or simultaneous mordanted with ferrous sulphate also showed very good anti-moth properties and left only 10-20% of moth alive after incubation period with 0.41% and 0.56 % fabric weight loss, respectively (Fig. 2). The above results shows that alkaloid, flavonoid and saponin of neem bark extract on their dye fabric is digestive to wool moth similar to neem leave extract dyed fabrics. But the high concentration of azadirachtin, i.e., five times and almost double the quantity of cyanogenic glucosides (HCN) in neem bark may be the reason of higher anti-moth efficacy of neem

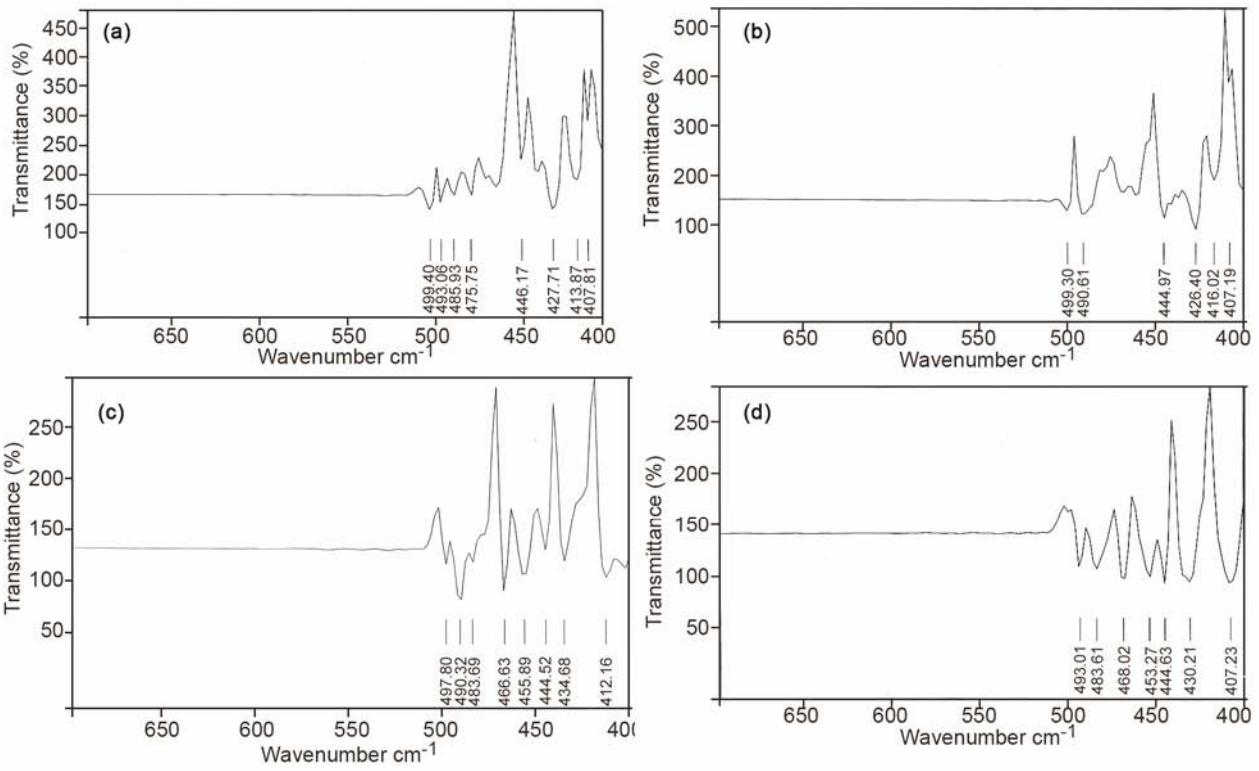

Fig. 1—FTIR spectra of grey and dyed woollen fabrics; (a) Grey wool fabric; (b) Fabric dyed Neem leaves alone; (c) Dyed fabric (Leaves extract + $Sncl_2$); (d) Dyed fabric (Bark extract + $Sncl_2$)

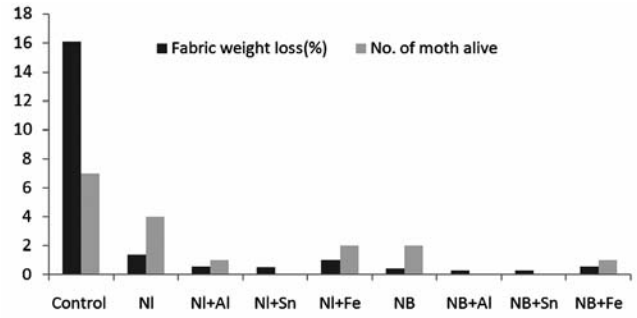

Fig. 2—Anti-moth properties of Neem bark and leaf dyed woollen fabric Neem bark extract-NB; Neem leaves extract-NL; Aluminium Sulphate-Al; Tin Chloride-Sn; Ferrous Sulphate- Fe

bark extract. Since, cyanogenic glucosides yield glucose, hydrogen cyanide and aldehyde or ketone upon hydrolysis[5]. The high concentration of azadirachtin make the fabric infeedent and under starving conditions when bark dyed wool keratin induced in the metabolic system, the high dose of hydrogen cyanide has lethal effect causes to death of moth.

Conclusion

The natural colourant obtained from neem leaves and barks are able to produce yellow and brown range of colours on woollen fabrics. Mordanting enhances the colour strength as well as light and washing fastness properties. Neem leaves extract shows lower anti-moth efficacy mainly due to lower content of azadirachtin, nimbin and cyanogenic glucosides (HCN). Anti-moth efficacy of neem bark has found better than neem leaves extract dyed fabric. It may be due to higher concentration of azadirachtin and cyanogenic glucosides (HCN). Stannous mordanted fabrics show higher anti-moth efficacy mainly due to formation of organotin compound comparable with Eulon, a commercial anti-moth agent.

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