Antibacterial finish for cotton fabric from herbal products

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An ecofriendly natural antibacterial finish has been prepared from the plant extracts for textile application. Herbal extracts from Ocimum sanctum (tulsi leaf) and rind of Punica granatum (pomegranate) have been applied to cotton fabric by the method of direct application, micro-encapsulation, resin cross-linking and their combinations. All the treatments show good antibacterial properties for the fabrics. Except the method of direct application, all other treatments show good washing durability up to 15 washes. The surface morphological studies using SEM show the surface coating, microcapsules and some fibrillation. The GC-MS studies reveal that the major components responsible for the antibacterial properties are Eugenol, Germacrene and Phytol. A small decrease in tensile strength and crease recovery angle is observed for resin treated and micro-encapsulated fabrics respectively. But in the combined processes no significant changes are observed.

Keywords: Antibacterial, Cross-linking, Eugenol, Micro-encapsulation, Pomegranate, Phytol, Tulsi

1 Introduction

Recent market survey has quite convincingly shown that the apparel consumers all over the world are demanding functionality in the product. Some of the best examples of functionality are product attributes such as wrinkle resistance, soil release, water repellency, flame retardancy, fade resistance and resistance to microbial invasion. Among these, the antimicrobial property of fabric is being considered to be an important and inevitable parameter for garments which are in direct contact with human body. Cotton textiles in contact with the human body offer an ideal environment for microbial growth. Microbial infestations possess danger to both living and non-living matters. Obnoxious smell from the inner garment (such as socks), spread of diseases, staining and degradation of textiles are some of the detrimental effects of bad microbes. The consumers are now increasingly aware of the hygienic life style and there is a necessity and expectation of a wide range of textile products finished with antimicrobial properties. Many commercial products are currently available in the market with a range of antimicrobial properties under different trade names for textile industry. Majority of such products are synthetic based and may not be environment friendly. Their compliance to the regulations imposed by international bodies, such as EPU is essential. There are many natural/herbal products which show antimicrobial properties. Chitosan, a naturally occurring biopolymer, has opened up a new avenue in this area of research. Extracts from different parts of diverse species of plants like root, flower, leaves, seeds, etc. exhibit antibacterial properties. Many of the plants contain compounds like phenolic, terpenoids, flavonoids, alkaloids, polypeptide, polycetylenes, etc. which are acting as antibacterial. Some of them act as bactericides (which kills bacteria) and some act as bacteriostatic (interfere with the multiplication, growth or activity of bacteria).

The present investigation aims at developing an ecofriendly natural antibacterial finish from plant extracts for textile application. Some selective species of plants were identified and screened for their antibacterial activities. Their extracts were applied to cotton fabrics. An extensive study was conducted to assess the antibacterial effectiveness of the herb by employing standard test method (AATCC-147, 2004) and the findings are discussed in this paper. Herbal extracts from tulsi leaf (Ocimum sanctum) and rind of pomegranate (Punica granatum) were selected for the study. As garments are subjected to washing, the wash durability of finishes is a major issue. Even though many of the herbal extracts have shown good antibacterial property after applying on cotton fabric, their wash durability is poor. Hence, in this study different techniques like micro-encapsulation and

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cross-linking were used to fix the herbal extract on the fabric. Quantitative analyses of antibacterial property of the treated and washed fabrics were estimated as per AATCC-100 test method. The physical properties of the treated fabric like crease recovery angle and breaking strength were also studied. The structural morphology of the fabrics was evaluated by using scanning electron microscope. Different components present in the herbal extracts were identified with the help of GC-MS.

2 Materials and Methods

2.1 Method of Extraction

Fresh tulsi leaf (Ocimum sanctum) and pomegranate fruits (Punica granatum) were procured from local market of Mumbai. Scoured and bleached cotton fabric was purchased from Century Mill, Mumbai. Acacia gum was purchased from Loba Chem., Mumbai. Non-formaldehyde based resin (KRISOF NIL/F) was supplied by Britacel Silicon Pvt. Ltd., Mumbai.

2.2 GC – MS Spectral Analysis

The component identification of herbal extract was done by using Shimadzu GC–2010 gas chromatograph coupled with QP2010 mass spectrometer. Samples were injected into a fused capillary silica column (RTX–5 ms) of 30m × 0.25 mm × 0.25 µm. Helium was used as carrier gas. Injector and detector temperatures were kept at 250°C. Column temperature was programmed at 60°C for 3 min and then raised up to 160°C (@ 5°C/min) and kept as such for 2 min. Further temperature was raised up to 300°C (@10°C/min). Mass spectra were acquired over a 40–500 amu range in EI Mode. The eluted compounds were identified by comparing the mass spectral data with the standard data available in the library of National Institute of Standards and Technology.

2.3 Antibacterial Activity Assessment

Antibacterial activities of the treated fabrics were evaluated by both qualitative (AATCC-147) and quantitative (AATCC-100) methods. For parallel streak method (AATCC-147-1988), specimen of test material, including the corresponding untreated control, were placed in contact with nutrient agar which has been previously streaked with inoculums of a test bacterium. After incubation, a clear area of interrupted growth underneath and along the sides of the test material indicates antibacterial activity of the specimen. For quantitative analysis, AATCC-100-2004 test was used. Control swatches were inoculated with the test organism. After incubation the bacteria are eluted from the swatches by shaking a known amount of neutralizing solution. The number of bacteria present in this liquid was determined and the percentage reduction by the treated material was calculated. In both cases the bacteria used was Staphylococcus aureus (ATCC 6538) (Gram positive) and Klebsiella pneumoniae (ATCC 4352) (Gram negative).

2.4 Fabric Treatment with Herbal Products

2.4.1 Direct Application method

Methanol extracts of the herbs were directly applied on 100% cotton fabric by pad-dry-cure method. 1% of the herbal extract was applied on cotton fabric along with 6% citric acid as cross-linking agent by pad-dry-cure method. Padding was carried out in a pneumatic padding mangle at a pressure of 3 psi to get a pick up of 100% on weight of fabric. Drying and curing were carried out in spooner at 80°C for 5 min and 150°C for 3 min respectively.

2.4.2 Micro-encapsulation Method

Micro-encapsulation is one of the novel methods of getting functional finishes on textiles. It is a process by which very tiny droplets of liquid or particles of solid are covered with a continuous film of polymeric material. Micro-encapsulation is more advantageous to conventional processes in terms of economy, energy saving, ecofriendliness and controlled release of substance. The agents reside in colloidal suspension within the amorphous zone of the binder so that a reservoir of agent is present in solid solution within the polymer matrix. Such treated fabrics were reported to be durable to a few number of wash cycles. The prolonged bioactivity of the fabric is due to slow diffusion of the antimicrobial agent out of the polymer reservoir. In the present work, micro-encapsulation was done using herbal extract as core material and gum acacia as wall materials. Ten grams of acacia powder was allowed to swell for 15 min in 100 mL of hot water. To this mixture, 50 mL of hot
water was added and stirred for 15 min maintaining the temperature between 40°C and 50°C. One and a half gram of core material (herbal extract) was slowly added under stirring condition. Stirring was continued for another 15 min and then 10 mL of 20% sodium sulphate and 6 g of citric acid were added. The stirring was stopped and the mixture was freeze dried in a freezer to develop microcapsules. Cotton fabric was immersed in the microcapsule solution and padded through pneumatic padding mangle at a pressure of 3 psi. The treated fabric was dried at 80°C for 5 min.

2.4.3 Cross-linking Method

One gram of herbal extract was mixed with 100 mL (120 gpL concentration) non-formaldehyde based resin (KRISOF NIL/F) and 2.0 g MgCl₂ was added as a catalyst. Cotton fabric was dipped in the resin solution and padded through a pneumatic padding mangle. The treated fabric was dried at 80°C for 5 min and cured at 150°C for 3 min.

2.4.4 Micro-encapsulation and Cross-linking Method

Micro-encapsulation is a physico-chemical mode of application of finishes to textiles, whereas cross-linking is a chemical method of applying finishes to textiles. To enhance the functional property of a textile it is possible to combine both physico-chemical and chemical techniques i.e. combination of micro-encapsulation and cross-linking. In the present work first micro-encapsulation was done using herbal extract of core material and acacia gum as wall material. Before applying on the fabric 18 g non-formaldehyde resin and 3 g MgCl₂ were added. Cotton fabric was dipped in the solution and padded through a pneumatic padding mangle. The treated fabric was dried at 80°C for 5 min and cured at 150°C for 3 min.

2.5 Wash Durability of Finish

The antibacterial activity of the finished samples was evaluated after being subjected to several wash cycles. The finished fabrics were washed using standard detergent (3% owf) at 40°C in an automatic washing machine using the method ISO: 6330–1984 E.

2.6 Characterization of Treated Fabric

The surface morphology of treated fabrics was established by using scanning electron microscope (JEOL, SSM-5400). Tensile strength, one of the important physical parameters, of the treated and untreated (control) fabrics was evaluated by using Instron tensile tester as per the ASTM D 5035–2006 method. Crease recovery angles of treated and control fabrics were measured on a crease recovery tester using the method AATCC 66-2003. The absorbency of treated and untreated fabrics was evaluated by water drop method as per AATCC 79-2000 standards.

3 Results and Discussion

The yields of methanol extracts of dry tulsi leaf and pomegranate rinds are found to be 24% and 45%; moisture contents are 7.9 % and 11.0 %; and pH of the extracts are 4.5 and 3.8 respectively. GC-MS spectral data of components in methanolic extraction of tulsi leaf and pomegranate rind are given in Table 1.

In gas chromatography, we could separate out many components and the separated components were identified by using NIST library search method. Table 1 indicates that the majority of components found in Tulsi extract are Eugenol methyl ether (30%), linoleic acid methyl ester (11.1%), ascorbic acid 2,6 dihexadecanoate (7.9%), caryophyllene (5.5%), phytol (3.9%) and germacrene (2.2%)<12,13>. Among these compounds, caryophyllene, phytol and germacrene belong to terpenes category and are reported to be antimicrobial compounds<14-16>.

Table 1 also indicates that the main ingredients found in the rind of pomegranate are 2 Furan carboxaldehyde; 5–5 hydroxy methyl (40.5%); 1,2,3 benzene triol (10.6%); 2-furaldehyde (4.7%); and 9 octadeanoic acid (4.3%).

The antibacterial properties of materials can be studied by qualitative (AATCC-147) as well as quantitative (AATCC-100) test methods. However, it has been found by the authors that the qualitative test is good for testing the main agent or the treated fabrics, provided the antibacterial agents used are capable of leaching out. On the other hand, quantitative test is the proper indicator of degree of antibacterial activity when the antibacterial agents are fixed on to the textile material or are unable to leach out. The different tests carried out in this study are based on such consideration.

Antibacterial activities of tulsi leaf extract treated fabrics and pomegranate rind extract treated fabrics using qualitative and quantitative methods are given in Table 2. Parallel streak method results clearly show that all treated fabrics are having very good antibacterial properties to both Gram positive and Gram negative micro-organisms. The treated fabrics
do not allow the growth of bacteria under the test specimens. All the treated fabrics show a zone of inhibition ranging from 9.9 mm to 12.5 mm for Gram positive and from 4.9 mm to 6.6 mm for Gram negative bacteria. The untreated fabric (control) shows bacterial growth under the test specimen. It is also indicated by quantitative results that irrespective of method and methodology adopted for application, the % reduction value to both the microorganism (Sa and Kp) are found to be >90% for tulsi and >85% for pomegranate extract treated fabrics (Table 2). The untreated fabric shows zero % reduction to both the micro-organisms. The zone of inhibition values indicate that both the herbal extracts not only prevent the growth of bacteria under the fabric, but also leaches out and kills the bacteria (bactericides).
is in agreement with the test results of % reduction value as shown in Table 2. Wash durability of the herbal treated fabrics for a few wash cycles are shown in Table 3.

Table 3 shows that tulsi extract and pomegranate extract when directly applied on cotton fabrics are having very good antibacterial properties to both Sa and Kp micro-organisms, but show poor wash durability. Before washing, the pomegranate treated fabric shows that the % reduction values for Sa and Kp are 99.9 and 90.8 respectively. After one wash, the % reduction values drop drastically and are 33.0% and 30.0% for Sa and Kp respectively. The same trend is observed in the case of tulsi extract treated fabrics. This indicates that direct treatment is only a superficial coating on the fabric surface without any firm bonding and gets removed by washing.

Table 3 also indicates that fabrics treated with microcapsules of tulsi extract and pomegranate extracts are found to have good resistance to bacterial attack. At the same time, such treated fabrics are found to have a wash durability of up to 15 washes. In the case of tulsi extract treated fabric, in each wash cycle the antibacterial activity decreases gradually and at the end of 15th wash cycle the % reduction value reaches to 45.5 and 42.6 for Gram positive and Gram negative bacteria respectively. Almost same trend is also observed for pomegranate extract treated fabric. These results are similar to the earlier reports on micro-encapsulation of neem and Mexican daisy[10,11]. The antibacterial effect of micro-encapsulated fabric is not significantly affected due to repeated laundering when compared with that of direct treated fabrics (Table 3). This is perhaps due to the sustained release of antibacterial compounds over repeated laundering from the herbal products because of micro-encapsulation technique. While comparing between tulsi extract treated fabric and pomegranate extract treated fabrics, it is found that the rate of bacterial reduction is similar up to one wash and after that the pomegranate extract treated fabric shows reduction at a faster rate than tulsi extract treated fabric.

The fabrics treated with herbal extract along with a resin (cross-linking agent) show good resistance to bacterial attack. Such treated fabrics are also found to have a wash durability up to 15 washes. It is also found that even though citric acid is used in the direct method, the wash durability is found to be ineffective. Basically citric acid was used to study the fixation efficacy of herbal extracts on cotton fabric. Citric acid is not an effective cross-linking agent as resin.

In the case of tulsi extract treated fabric, in each wash cycle the antibacterial activity decreases gradually and at the end of 15th wash cycle the % reduction value reaches to 55 and 51.5 for Gram positive and Gram negative bacteria respectively. Almost the same trend is observed in the case of pomegranate treated fabric.

Table 3 also reveals that the herbal extracts encapsulated with acacia gum and cross-linked with a resin give good antibacterial property and wash durability up to 15 wash cycle. In the case of tulsi treated fabric, in each wash cycle the antibacterial activity decreases gradually and at the end of 15th wash cycle the % reduction value reaches to 48.9 and 50.5 for Gram positive and Gram negative bacteria respectively. Almost same trend is observed in the case of pomegranate extract treated fabrics.

Figure 1 shows an overall picture of wash durability and antibacterial activities of both the

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<th>No. of washes</th>
<th>% Bacterial reduction of fabrics</th>
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<td>Direct treated</td>
</tr>
<tr>
<td>0</td>
<td>Sa 99.9 Kp 95.3</td>
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<tr>
<td>1</td>
<td>30.0 22.3 Sa 92.6 Kp 33.0</td>
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<tr>
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<td>0 0 0 0 0 0</td>
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herbal extracts treated fabrics towards Gram positive and Gram negative bacteria with all the four different techniques.

Physical properties like crease recovery angle and tensile strength of the treated fabrics are given in Tables 4 and 5 respectively. It is observed that for the method of direct application there are no significant changes in both the physical properties. In micro-encapsulated pomegranate extract treated fabric, the crease recovery angle decreases by about 32% while in case of tulsi the decrease is about 20%. Cross-linking by resin alone increases the crease recovery angle value for tulsi as well as pomegranate. However, the combined use of micro-encapsulation and cross-linking gives optimum crease recovery angle values. The changes in tensile strength for direct and micro-encapsulation treatments for tulsi and pomegranate extract treated fabrics are very minor. Pomegranate extract cross-linked with resin decreases the tensile strength by about 15% and that of tulsi is about 13% in warp direction. But in the combined method the loss in crease recovery angle and tensile strength could be minimized.

The surface morphological features of cotton fibre (control) are depicted in Fig. 2. Figure 3 shows the SEM photographs of cotton fabric treated with extract

| Table 4 — Crease recovery angle of treated fabrics |
| Fabric | Tulsi | Pomegranate |
|        | WP   | WT   | Total | WP   | WT   | Total |
| Untreated (Control) | 80   | 80   | 160   | 80   | 82   | 162   |
| Direct treated       | 78   | 81   | 159   | 75   | 80   | 155   |
| Micro-encapsulated   | 67.0 | 61.0 | 128.0 | 50   | 55   | 110   |
| Cross-linked          | 100  | 109  | 209   | 102  | 110  | 212   |
| Micro-encapsulated & cross-linked | 87  | 85  | 173   | 55   | 65   | 120   |

WP-Warp and WT-Weft
of tulsi and pomegranate using direct method. In both the photographs one can clearly see the fibrillar structures being ruptured and as a result small micro-fibrils extruding from the main fibre. The width and length of such micro-fibrils measured are found to be 0.5 and 5.8 µm respectively. The probable reason for this type of pluckering effect on the fibre could be due to the acidic reaction of the extract on the fibre. The surface of the fibre seems to be covered with granular structure. The sizes of such granules vary from 0.2 µm to 0.6 µm. This could be a deposit of herbal extract on the surface of fibre. Such granular deposit was found less in pomegranate extract treated fabric than in tulsi extract treated fabric.

Figure 4 shows the SEM photographs of the fabric washed once after treatment with tulsi extract and pomegranate extract. It is clear from the photographs that the granular deposit on the surface reduces considerably; however, the protruding fibrils still exist on the fibre.

Figure 5 shows the SEM photographs of cotton fabric treated with micro-encapsulation method. It is clear from the photograph that microcapsules produced are of small spherical shape with a fairly

<table>
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<tr>
<th>Fabric</th>
<th>Tulsi</th>
<th>Pomegranate</th>
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<tr>
<td></td>
<td>WP</td>
<td>WT</td>
</tr>
<tr>
<td>Untreated (Control)</td>
<td>343.1</td>
<td>11.0</td>
</tr>
<tr>
<td>Direct treated</td>
<td>340.1</td>
<td>11.0</td>
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<tr>
<td>Micro-encapsulated</td>
<td>344.8</td>
<td>12.1</td>
</tr>
<tr>
<td>Cross-linked</td>
<td>297.6</td>
<td>12.3</td>
</tr>
<tr>
<td>Micro-encapsulated &amp; cross-linked</td>
<td>311.9</td>
<td>12.0</td>
</tr>
<tr>
<td></td>
<td>N</td>
<td>%E</td>
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N – Newton, E – Elongation.
uniform size distribution. It is also clear that the microcapsules are firmly fixed on the fibre assembly of the fabric. The sizes of the microcapsules are in the range of 0.13-0.5 µm. The average size of microcapsules is found to be 0.44 µm.

Water absorbency of fabrics treated with various techniques is shown in Table 6. The absorbency values indicate that the water absorbing nature of fabric has not altered much in any of the application processes, except for the direct treatment. Fabrics directly treated with herbal extracts show water repellency. This could be due to the hydrophobic nature of extract, which forms a thin coating of film on the surface of fabric. Micro-encapsulation has also made the fabric water repellent to some extent.

4 Conclusions

Both tulsi leaf and pomegranate rind extract treated fabrics show considerable zone of inhibition to Gram positive and Gram negative bacteria. There is no bacterial growth found in case of treated fabrics. This indicates that these herbal extracts act as bactericides as well as bacteriostate. Extracts can be applied directly on to the fabric by a simple process like pad-dry-cure method. The only limitation is that such direct treated fabric does not have wash durability. If the fabric is for single use application such as disposable items like surgical cloth, bandage gauze and sanitary napkin, which do not need to be washed, then this technique can be adopted. If the requirement needs several washes, such as garments like bed linen sheet in hospital and socks, then any one of the other techniques (micro-encapsulation, resin treatment or both) can be adopted. The surface morphological data reveal that micro-encapsulated fabric becomes stiff and decreases the crease recovery property. Resin treated fabric (cross-linked) imparts good crease recovery but decreases the tensile strength. The fabric treated with the combined processes, i.e. micro-encapsulation+cross-linking shows good antibacterial property and good wash durability without compromising the physical properties.

Since tulsi leaf and pomegranate fruit are abundantly available in many of the countries, the scope of implementation and commercialization of herbal extract to impart antibacterial finish in textile is high. After removal of edible seeds from pomegranate fruit the rind is considered to be a waste material. Hence, synthesis of antibacterial agent from such waste product is a novel idea. Finally, since the raw material is 100% from natural resources, it is ecofriendly having economic, social and environmental benefits.

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References