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SUSTAINABLE ANTIMICROBIAL FINISHING OF FABRICS USING NATURAL BIOACTIVE AGENTS - A REVIEW

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ABSTRACT

Antimicrobial textiles have gained much attention and popularity in the market and in day- to-day life during the last two decades. The textile industries continue to introduce different methods in their production to enhance the quality of their products and to satisfy their customers. Increased competition in the industry has lead to the development of different synthetic antimicrobial agents. Though these synthetic agents are used widely, they pose a threat to both the users and the environment. Taking into consideration these threatening issues, several environmental bodies have implemented rules in the use of synthetic agents. On the contrary, the natural antimicrobial agents which have less adverse effect on humans and are eco friendly, are gaining much attention. Several natural agents such as basil (active agent is eugenol), neem (active limnoids like azadirachtin, nimbinin), turmeric (curcumin), clove oil, chitosan, sercin, onion, aloevera and pomegranate have antimicrobial properties. This review paper highlights the different natural antimicrobial sources, their effect on public and the possibilities of using these agents in textiles to impart antimicrobial properties and to develop different products.

KEYWORDS: *Antimicrobial agents, Eco friendly agents, health impact*

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INTRODUCTION

During the past few decades, human health has been seriously bothered and threatened by various health issues. Among the many infectious diseases prevalent in the society, indoor microbial contamination and infection are more common. Textiles are susceptible to cross contamination as they are conducive to the growth of pathogenic microorganisms. The availability of large surface area and moisture content are the main factors that promote the growth of microbes. Growth of microbes leads to unpleasant odour, pigmentation and deterioration in quality, which not only affect the products but also the wearer. To enhance the quality of textile products innovative methods are in use such as fragrance textiles, insect repellent textiles, textiles immobilized with active agents for various purposes and textiles coated with phase changing materials (PCMS). Antimicrobial finishing on textile mainly focuses on three objectives 1) To control and avoid infection 2) To reduce odour and pigmentation 3) To control bio deterioration. Two different modes of action are reported for antimicrobials such as bound antimicrobials and leaching antimicrobials. Several synthetic agents like triclosan, quaternary ammonium compounds, nano silver materials are available for antimicrobial finishing[1] of textiles. Synthesis of nano particles is expensive whereas the other synthetic agents cause many environmental issues[2]. Considering these problems, regulations in the use of these chemicals have been implemented by various environmental bodies such as EPU[3]. The active agent used in antimicrobial finishing should be effective only against undesirable organisms. It should also be nontoxic, safe and biodegradable and should have long durability. The natural agents with their effective antimicrobial activity and less adverse consequences are being used at present to overcome the problems caused by synthetic agents. India has a rich biodiversity with more than 450 plants available for dye extraction and antimicrobial finishing[4]. The use of different natural antimicrobial agents like aloe vera, eucalyptus, turmeric, neem, and basil has been already reported in textile production. This shows that most of the natural antimicrobial agents are mainly derived from different medicinal plants [5]. Though much research has been done on the technical and functional aspects of production or synthetic antimicrobial finishing, only a few research papers are available on natural antimicrobial finishing. This paper offers a comprehensive, critical review of the use of natural products and their applications in antimicrobial finishing finishing on textiles. Numerous antimicrobial agents reported have been taken from natural sources. The following (Table 1) lists the active agents taken from plants and their modes of action against microbes.Table 2 represents their spectrum of activity. Disc diffusion method (AATCC 90) was used for the study.Zone of inhibition was observed beneath fabric which was treated with the antimicrobial extracts.

Table 1
Active agents from plants

Class	Subclass	Example	Mechanism
Phenolics	Simple phenols	Catechol	Substrate deprivation
	Flavonoids	Chrysin	Bind to adhesins complex with cell wall
	Flavones	Abyssinone	Interact with enzymes
	Flavonols Tannins	Ellagitanninn	Bind to protein Bind to adhesin Substrate deprivation Membrane disruption
	Coumarins	Warfarin	Interaction with eukaryotic DNA
Terpenoids		Capsaicin	Membrane disruption
Alkaloids		Berberine, Piperine	Interactive in to cell wall and/or DNA
Lectines and Polypeptides		Mannose	Block viral fusion or adsorption

Table 2
Antibacterial spectrum of herbal products from different sources

Antimicrobial Agents	Antimicrobial Spectrum	Reference
Terpenoids[6,7]	*S.aureus,P.aeruginosa, V.cholera*	*Bach et al.,2011* Mathabe*et al*., 2008
Lectins and polypeptides[8,9]	*S.aureus,B.subtilis, E.coli,P.aeruginosa ,C.albicans*	Kheeree*et al*.,2010 Petnual*et al*.,2010
Flavonoids[10,11]	*K.pneumoniae, S.centerica, P.aeruginosa ,S.aureus,E.coli, Actinobacterbaumannii*	Orhan*et al*.,2010 Özçelik*et al*.,2008
Quinones[12,13]	*S.aerreus, B.subtils, P.aeruginosa*	Ignacimuthu*et al*.,2009 Singh *et al*.,2006
Tannins[14,15]	*B.cerus, Listeria monocytogenes, S.centerica*	Engels *et al*.,2009 Scalbert*et al*.,1991
Coumarins[5,16,17]	*Listeria monocytogens, S.aerus, E.coli,V.parahaemolyticus*	Cowan *et al*., 2009 Venugopala*et al*.,2013 Saleem*et al*., 2010

MECHANISM OF ANTIMICROBIAL AGENTS AGAINST INVADING MICROBES

Mode of action of each antimicrobial agent will differ on textile. This depends on factors like chemical and structure nature and affinity level to certain target sites within the cells. The Mode of action is classified as follows by different researchers[18,19].

- Damage or inhibition of the cell wall synthesis
- Inhibition of cell membrane function, which disturbs the intra and extra cellular flow of substrates
- Inhibition of protein synthesis
- Inhibition of nucleic acid synthesis(DNA / RNA)
- Inhibition of metabolic process eg: Folic acid pathway

BASIL- OCIMUMTENUIFLOURUM

Ocimumtenuiflourum is cultivated for religious and traditional medicinal purposes. Some of the phytochemical constituents of basil are oleaolic acid, rosmarinic acid, eugenol and the reported essential oils are eugenol(70 %)(Figure - 1), β-caryophyllene (8 %), β-elemene (11%) and germacrene (2 %). Natural antimicrobial agents are attaining much attention than synthetic antimicrobial agents because of their efficacy and safety. The effect of methanolic extract of basil leaves on cotton fabric was studied and 73 % of bacterial reduction was reported against the test pathogens[1]. To improve the antibacterial activity the combined effect of basil with clove, neem and karanga have been reported by Sarkar*et al*.,2002[20]. A group of researchers tried three different methods of antimicrobial finishing to find out the efficiency of basil leave extract coated on cotton fabric by treating with basil leave extract by direct method, microencapsulation and with cross linking agent [21], and reported their durability. The Cotton fabric treated with cross linking agent (resin) is better than that treated with direct and encapsulated method. The tensile strength and stiffness of the material got improved with reduction in the crease recovery. Reports are available for combined antibacterial effect of basil and pomegranate on cotton fabric. The researchers had used three methods for application - direct method, micro encapsulation and microencapsulation with resin as cross linking agent against *S.aureus* and *K.pneumoniae* using AATCC 147 and AATCC 100 methods. Clear zone of inhibition was observed in AATCC 147 method and 90% of reduction was recorded in AATCC 100 method against both pathogens. Wash durability of coated fabric was also reported. After 15 wash cycles antibacterial activity gradually decreased in direct method and micro encapsulated method but fabric coated with cross linking agent (citric acid) showed good antimicrobial activity even after 15 washing cycles [22]. Comparative study of natural antimicrobial agents with synthetic was done by Rajput et al.,2017[23], reported the effect of basil as an

antibacterial agent on diapers and compared the effect with synthetic antibacterial agents such as silver nanoparticles, zinc nanoparticles and titanium dioxide powder and concluded that the natural agents have similar activity compared with synthetic agents and that 12.5 mm zone of inhibition was present against *P.aeruginosa*, 11mm against *S.aureus* followed by *E.coli* with 5mm. The effect of basil in combination with malabar nuts against *S.aureus* and *E.coli* was also done by the same researcher in which he observed 30 mm against *S.aureus* and 12 mm against *E.coli.*

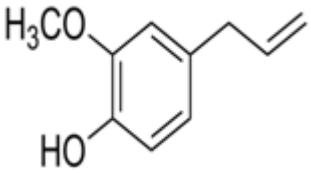

Figure 1
Eugenol, the main ingredient of basil

NEEM –AZADIRACHTAINDICA

Neem has been widely used in ayurveda, unani and in homeopathic medicines. More than 140 bioactive compounds are reported from neem for its various applications. Comparative study of dip-dry method and exhaust method was reported by Mahesh et al[24] using neem extract on cotton fabric. Observation reports suggest that exhaust method is better than dip-dry. Results were recorded as follows: the extract is more effective against Gram negative than Gram positive bacteria. After 5 wash cycles, the antimicrobial activity gradually reduced to 33.23 % and 24.31% respectively for Gram negative and Gram positive bacteria. Neem extract applied on medical textile to improve antibacterial activity by two methods, direct and microencapsulation by Ganesan[25] reported that the wash durability was poor for fabric treated by direct method but microencapsulated fabric showed higher reduction in bacterial growth even after 15 wash cycles against *S.aureus* and *E.coli*. Improvement of physical parameters by imparting antibacterial agent was reported by shahidi[26] using neem extract by treating the modified cotton with different concentrations of neem extract in the presence of silicon micro emulsion and non ionic softener. The efficacy of neem seed and bark extracts against *Bacillus subtilis* on cotton and cotton/polyester blend textiles has been reported byJoshi[27]. The effect of neem extract on diaper compared with different synthetic antibacterial agents was reported by [23] who observed that the activity of natural agents was similar to that of synthetic agents. The antibacterial properties of neem oil in combination with other herbal oils such as clove, basil and karanga have been used to impart antimicrobial finishing on cotton textiles [20]. The Methanolic extract of neem was used by Vyas *et al*., 2008[28] who followed different methods for coating and concluded that neem was better than aloe vera on cotton fabric as durability was observed even after 15 washes. Active limonoids are given in the (Figure 2)

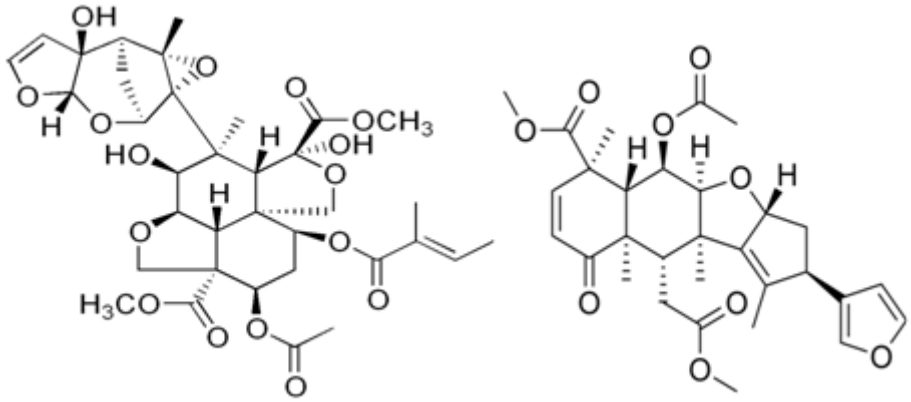

Figure 2
Azadirachtin Nimbinin
Active limonoids present in neem giving antimicrobial property

POMEGRANATE–PUNICAGRANATUM

Pomegranate is a rich source of vitamin A, D, B and calcium. Its total fat content is 1.2 g and is widely used in ayurvedic medicines. Antimicrobial property of pomegranate and its effect on fabric was reported by Gupta[29]. It was observed that the stiffness and tensile strength of the fabric improved along with durability which could also be improved by either micro encapsulation or using cross linking agent. In 2010, study conducted by Dahham*et al*., 2010[30] found that among the antibacterial activity of extracts taken from different parts of pomegranate (peel, seed, juice, whole fruit), the peel extract exhibited maximum activity against *S.aureus* and against fungus *A.niger*. The antibacterial effect of pomegranate on cotton fabric was checked by dip method followed by exhaust method and it was reported that the exhaust method was effective against both Gram positive and negative but more effective against Gram negative bacteria [24]. Three methods (direct, micro encapsulation and cross linking) were used by Sathianarayanan[22] and wash durability was found to be more effective on the fabric coated with micro encapsulation with cross linking agent.

TURMERIC-CURCUMA LONGA

Curcumin, one of the main ingredients of turmeric (Figure 3), imparts yellow colour and antibacterial property. The phytochemical compounds present in turmeric include diarylheptanoids which occur from numerous curcuminoids. Wool fabric treated with curcumin showed good antibacterial activity against *S.aureus* and *E.coli* (45 % and 30 %) even after 30 wash cycles as reported by Han*et al*., 2005[31]. A Report of antibacterial activity [24] on cotton fabric treated with turmeric against *E.coli* and *B.cereus* suggested that instead of direct method, the exhaust method was more powerful. After 5 wash cycle activity the effect reported was 33.23 % and 24.31 % against Gram positive and Gram negative bacteria. Mordant polyamide was used with turmeric and reported to be more effective against *E.coli* than *S.aureus*. Fabric finished with mordant is more efficient in washing than the normal finishing[32]

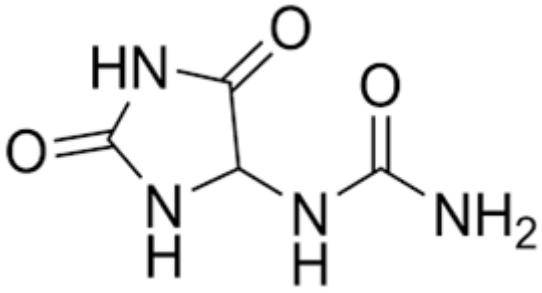

Figure 3
Curcumin present in turmeric

ALOEVERA (ALOE BARBADENSIS MILL)

It grows on tropical regions and is used for medicinal and decorative purposes. Aloe vera consists of more than 200 active ingredients. Because of its wound healing property it is used in the production of wound dressing materials. Its bioactive molecules are given in (Figure 4). Antibacterial activity of *Aloe vera* in the presence of glyoxal as cross linking agent on cotton fabric was done by following AATCC 147 & 100 methods. The experiment was performed using different concentrations of *A.vera*5,10,15,20,25 gpl respectively and good antimicrobial activity was observed on treated than untreated fabric Murugesh*et al*., 2015[33]. Effect of antibacterial activity in combination with neem and aloe vera has been reported by Vyas*et al*., 2008 [28]on cotton who found the combination more durable on fabric. In A comparative study done by Rajput *et al*.2017[23], the researchers selected many natural antibacterial agents including *Aloe vera* and compared them with synthetic antibacterial agents. The selected antibacterial agents were applied on diaper. The results revealed that natural coating had the same activity when compared with synthetic agents.

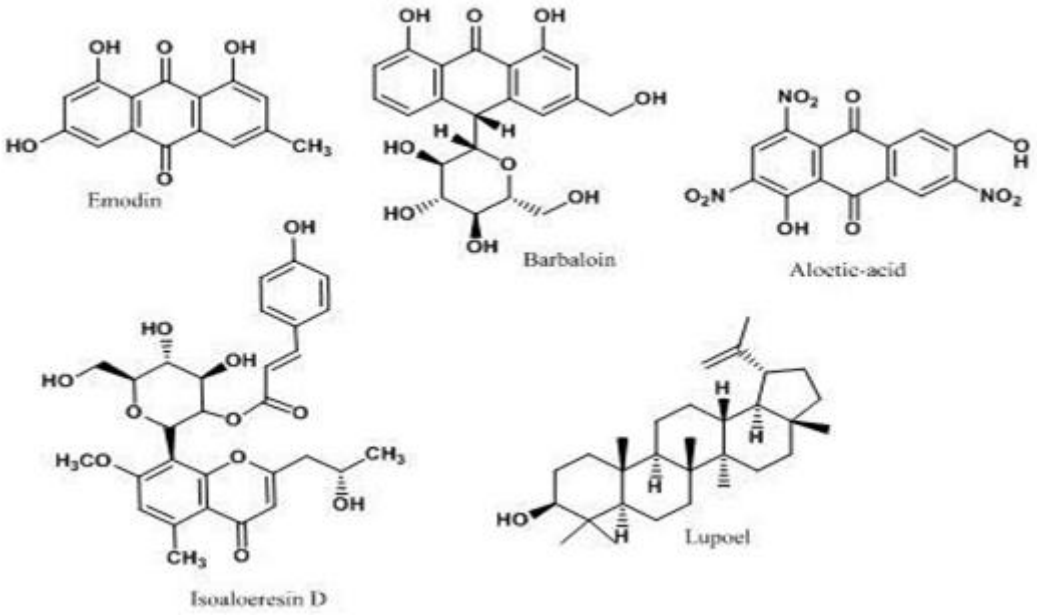

Figure 4
Active agents of aloe vera

ONION- ALLIUM CEPA

Onion consists of fibre, flavanoids and sulphur compounds. When plant gets damaged or is stressed, it generates an antimicrobial agent called allicin. The antibacterial property of onion has been applied in textile industry by different researchers to improve the microbial resistance of fabric. Cotton fabric was pre treated with oxygen plasma and grafted with onion pulp extract at 70 °C for different dilution. The zone of inhibition values obtained was proportional to the grafting time of pulp onion extraction, as suggested by Chen[34]. The Broad spectrum activity of onion has been reported by Škergetet al.,2009[35] after the antibacterial and antifungal activity of onion skin extract was tested against *E.coli, P.fluoresences, B.cereus* and *A.niger*. The reports revealed that the antimicrobial activity of the edible part of onion was comparatively very less than the other parts.

CLOVE OIL

Clove oil (eugenol) got its name from *Eugenia aromaticum.* The Main ingredient of clove oil is eugenol 72 - 90% and this component is responsible for the aroma. The effect of using DMDHEU (Dimethylol Dihydroxy Ethylene Urea) as cross linking agent along with clove oil as antimicrobial agent on cotton fabric was reported bySarkar[36] who found that the wash durability got improved by using cross linking agent. Zone of inhibition of 17 and 47 mm was observed against pathogens at 0.5 % and 1 % of oil respectively, with cross linking agent (Dimethylol Dihydroxy Ethylene Urea).

EUCALYPTUS OIL

Considering the cleansing property and antimicrobial property of eucalyptus oil, it is used for the development of functionalized textile to produce user friendly bacteria resistant fabric. It has been already reported for its antimicrobial activity against *Mycobacterium tuberculosis*, *Candia albicans* and viruses. The main component is Eucalyptol (1,8-cineole) which has both antibacterial and antifungal activity. Antibacterial property of two different eucalyptus leaves such as *E.cinera* and *E.odorata* leaf extracts were applied on wool and cotton fabric by Ben Fadhel*et al*., 2012[37]. Using acetone- water combination, the flavanoids extracted from leaves were treated on fabric and tested by AATCC 90 against *E.coli* and *S.aureus*. *E.odorata* was found to be more efficient on wool than cotton. Washing durability was also reported: gradual decrease in antimicrobial activity was observed after a number of laundering cycles but wool exhibited better wash durability than cotton.

CHITOSAN

Chitin is one of the abundant polysaccharides found in nature, especially derived from marine shells and molusks. Chitin is made up of a linear chain of acetyl glucosamine groups, while chitosan is obtained by removing acetyl glucosamine group and by leaving the active amine group. CH_3-CO will be removed and NH_2 will be present (Figure 5). Antimicrobial activity of chitosan is connected with polycataionic nature, which will bind with negatively charged residues of molecules at the

surface of bacteria and subsequently inhibit the growth of bacteria. One main factor influencing activity is pH, at lower pH high chitosan activity. Water soluble chitosan oligomers is prepared by acid degradation for finishing polypropylene nonwoven fabrics to impart antimicrobial activity against *P.vulgaris, S.aureus* and *E.coli*.by Shanmugasundaram and Qin*et al*., 2006-- used Tetracycline drug on chitosan coated fabric and checked the antibacterial activity against Gram positive and Gram negative bacteria [38,39], and reported that the chitosan was a binder and thickener for pigment printing of polyester and polyester cotton blends and noted the reduction of 96% bacteria against *S.aureus* in 1 h. Chitosan which is reported for its multipurpose use has its disadvantages while applying on fabric. Chitosan is effective against microbes only at high concentration. Deposition of chitosan develops thick layer or film on the surface on the fabric, which reduces the air permeability. Another negative effect reported is after treating with chitosan the fabric become much stiff.

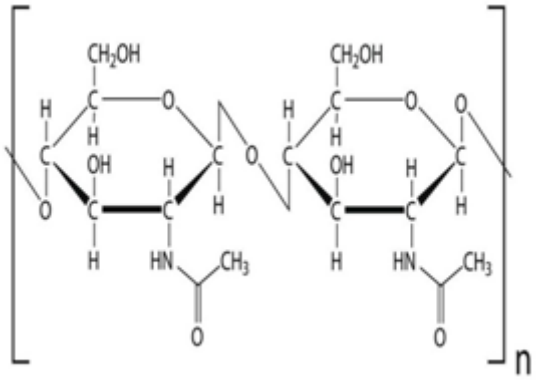

Figure 5
a) Chitin

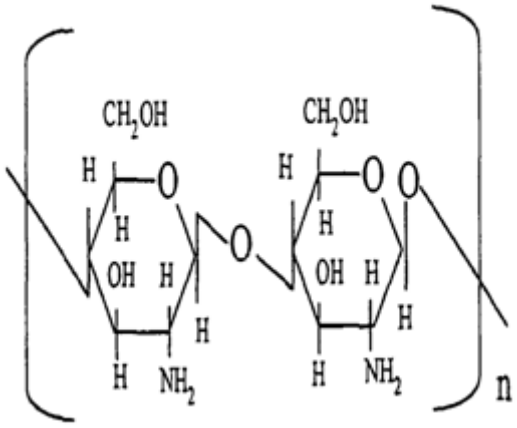

Figure 5
b) Chitosan

SERICIN

A Natural macromolecular protein, derived from *Bombyxmori*, Sericin consists of 25 - 30% of silk protein. Discharging of sericin during silk production causes water pollution. But it possesses various applications such as antibacterial, UV resistant, moisturizing properties. It has been reported that sericin 4 % (W / V) treated PET fabric showed 51% reduction of *S.aureus*[40].

OTHER NATURAL ANTIMICROBIAL AGENTS

Antibacterial activity of capsaicin coated wool fabric was reported byshahidi[26]. Capsaicin was coated on the wool fabric by sol-gel process and the antibacterial activity was tested by AATCC 90 method against *E.coli* and laundering method result was also reported. After repeated washing cycles, the antimicrobial activity slightly got reduced during each cycle. Herbal extract of *Blettilastriata* was immobilized on the polymer coated fabric to increase the wound healing property and tested against *E.coli* and *S.aureus*[41]. A study of microcapsule containing *Psidiumguajava* plant extract as antibacterial agent on cotton fabric was reported using AATCC147 method against *E.coli* and *S.aureus*. Leaf extract also showed antibacterial

activity against *S.aureus* but it was ineffective against *E.coli*. Konrai, Henna, Papaya were used as antimicrobial agent on jersey knitted fabric by three methods such as direct, exhaust and micro encapsulation against *E.coli* and *S.aureus*. Micro encapsulated fabric retained antibacterial activity after 15 wash cycles. The antibacterial activity of *Seabuckthorn* leaves extract checked against *E.coli* and *S.aureus* was found to be 12.4 mm and 16.7 mm respectively[42]. The antibacterial activity of azuki beans water extract including green, black and red showed good activity against *S.aureus, Aeromonashydrophila* and *Vibrio parahaemolyticus*. There is no activity by the white bean extract. These results suggested that polyphenols including proanthocyanidins in coloured beans were responsible for the activity[43]. The combined effect of ginger oil and curry leaves was reported by Krishnaveni *et al*., 2017 [44] finish has been imparted by pad-dry-cure coating and tested against group of Gram positive and Gram negative bacteria and there was no negative effect on physical properties such as absorbency, stiffness and strength. *Glycyrrhizaglabra* (Yasthimadhu) applied on organic cotton fabric by pad-dry-cure coating in combination with non polar solvents as an antibacterial and antifungal agent, reported the washing durability of active agent up to 12-18 washes for different solvents against *S.aureus*[45].

IMPACT OF ANTIMICROBIAL AGENTS ON HEALTH AND ENVIRONMENT

The Environmental impact of antimicrobial textiles relates to many factors such as substrate material, antimicrobial compound production, textile treatment production and finally the mode of disposal[46,4] and is a critical goal of concern.[7] Production and disposal of antimicrobial agents have significant influence on environmental aspects, beginning from the substrate selection to final disposal. If the disposed antimicrobial is not removed from the effluent, they may end up in the aquatic body. The Removal of synthetic agents through waste water treatment for triclosan is one option. In case of quaternary ammonium compounds (QACs), which is defined as "hard antimicrobial agent', the degradation of QACs is generally considered poor[48]. Aerobic condition treatment is a good option for triclosan removal, and its estimated degradation occur in 41 minutes and its half life in water is about 10 days [46,49]. Continuous exposure of antimicrobials develops drug resistance in microbes, due to their alteration in genetic makeup. Resistant strains are able to propagate and spread as they will not be killed by normal antibiotic[50,51]. Safety of antimicrobial agents attains much attention. Use of natural antimicrobial agents with potential antimicrobial spectrum and eco friendly nature are in high demand. The mode of action (leaching / bound), nature of active agent, concentration on textile, skin sensitivity and irritation level are the main factors of consideration while checking the bio efficacy of antimicrobial agents on textiles.

CONSTRAINTS

Different types of synthetic antimicrobial agents are available in market and they are reported for their better spectrum activity and durability. At the same time, noted side effectsare reported. Use of these agents were banned in many countries. Considering all these problems, now days research focuses on the development of natural antimicrobial agents with less side effects. The main problem is the availability of natural agents in required quantity and compared to synthetic it can withstand less number of wash cycles. But the implementation of natural agents as antimicrobial agents is a cost effective method when it is obtained from natural sources.

FUTURE PROSPECTS

- Research is needed to enhance the durability of natural antimicrobial agents on fabric after repeated washing.
- Combination of natural antimicrobial agents may be experimented to obtain a broad spectrum activity.

CONCLUSION

The textile industry is the second largest polluter next to oil industry. As the world has started marching towards sustainability, it has become necessary for the textile industry to stop using hazardous substances and switch to natural materials. Regulations related to the manufacture of textile products have been made stringent by many countries around the world and this has compelled the manufacturers to look for safer alternative materials. Natural materials are always on the top compared to synthetic materials owing to their friendliness towards the user and the environment. Synthetic antimicrobial agents are durable, have broad spectrum activity and are highly efficient but the problem is their hazardous nature and the associated human health issues and environmental pollution. Antimicrobial agents derived from natural sources are safe for human and the

environment but the spectrum of activity and efficiency are not as good as the synthetic ones. Still, a combination of these natural antimicrobial agents would be as efficient as the synthetic ones. To achieve this, more research work is needed in the field. Hence, natural antimicrobial agents derived from plant sources would be of prime importance in the future. We are in an urge to protect and conserve the natural ecosystem of the earth thereby restoring the global sustainability.

CONFLICT OF INTEREST

Conflict of interest declared none.

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