Benjamin Tawiah[1, 2], William Badoe[1, 2], *Shaohai Fu[1]

Advances in the Development of Antimicrobial Agents for Textiles: The Quest for Natural Products. Review

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[1]**Key Laboratory of Eco-Textiles, Jiangnan University, Ministry of Education,** Wuxi, Jiangsu Province 214122, China.

[2]**Department of Textiles, Kwame Nkrumah University of Science and Technology** Kumasi, Ghana.

*Email: shaohaifu@hotmail.com

Abstract

The antimicrobial finishing of textiles has attracted research attention lately due to demands for a healthy lifestyle. As a result, several synthetic and natural antimicrobial agents for textiles have been developed over the years. Recently research into antimicrobials agents of natural origin have become more popular due to their enormous therapeutic potential and effectiveness in the treatment of infectious diseases while mitigating the side effects of the synthetic antimicrobials. Research into these natural biocides for textiles has seen increasing consumers awareness for two reasons, namely the potential negative impact of synthetic biocides on health versus the benefits of natural biocides, and the increasing rate of microbial resistance to most natural biocides. The immense literature on natural biocides suggests the preparedness of the research community and industry in addressing the environmental and health challenges associated with synthetic antimicrobial agents in response to the new consumer demands. This review focuses on the advances in natural antimicrobial agents and various methods of their application. Literature suggest that natural antimicrobial agents have chalked some success in terms of efficacy and wash durability, with minimal effect on the tensile strength of fabrics.

Key words: *antimicrobial finishing, synthetic antimicrobial agent, natural antimicrobial agent, textiles, essential oils, dyes.*

Introduction

Fibres have been widely used in all areas of human activity as costumes, tents, tarpaulins, awnings, blinds, parasols, shower curtains, mattress ticking sails, waterproof clothing, etc. and in medical environments [1, 2]. However, natural fibres are prone to microorganism colonisation due to their large surface area and excellent moisture retention ability. Consumers are therefore justified in their increasing demand for the protection of fibres against microbes.

Antimicrobial finishing for textiles dates back to the ancient Egyptians, who applied herbs and spices to preserve mummy wrapping [3]. Today many different synthetic and natural compounds such as triclosan, quaternary ammonium, polybiguanides, N-halamines, silver and chitosan have been developed to impart antimicrobial properties to textiles [4 - 7]. Estimates have shown that the production of antimicrobial textiles was in the magnitude of 100,000 tons worldwide in the year 2000 [4]. Antimicrobial textile production has soared over the years, making it one of the fastest growing sectors in the textile industry [8]. A market survey report released recently by *Textile Intelligence* forecasts the global market for antimicrobial agents for all end uses, including textiles, to grow by nearly 12% per annum between 2013 and 2018 [9]. This projected growth coupled with the increasing demand for safe antimicrobial products is bound to advance the cause of cleaner and environmentally friendly antimicrobial textiles.

Antimicrobial agents are usually applied at the finishing stages of textile production, while in some cases biocide can be incorporated into synthetic fibres during extrusion. Bacterial resistance to biocides, their toxic effects on households and the environment, inadequate activity and poor durability on textiles have, however, become important issues of concern [4, 10, 11]. As a result, certain synthetic antimicrobial agents such as triclosan have been banned by a number of leading retailers and governments in Europe, especially those with the potential to cause skin irritation, and non-biodegradable and bioaccumulation effects [2]. Due to the high level of consumer awareness about clothing safety, many kinds of eco-friendly antimicrobial agents such as peroxy acids, chitosan and its derivatives, specific dyes and regenerable active *N*-halamine have been developed for textiles [12 - 15].

Lately investigation into natural antimicrobial agents, such as extracts from *neem*, *aloe vera, eucalyptus*, *capsaicin*, etc., have received great attention due to their eco-friendliness [10, 13, 16]. It has been found that natural antimicrobial agents exhibit broad-spectrum activity against bacteria, fungi and viruses, but they still has some limitations in terms of efficacy and durability [17 - 19], hence the quest for suitable methods of application of these natural biocides to impart high efficiency and durability.

Requirement of antimicrobial agents for textiles

Antimicrobial agents improve resistance against microorganisms, increase fabric durability and protect textiles against the colonisation of odour forming bacteria [20, 21]. This addresses hygiene needs in clinical and sensitive environments by minimising the chances for the microbial colonisation of textiles [22]. Antimicrobial treatments can also reduce the frequency of laundering, which gives the potential for significant savings in water usage and energy consumption and reduces the need for chemical materials in textile care [10, 13]. A good antimicrobial agent should be effective against a broad spectrum of bacterial and fungal species and must also exhibit low toxicity to consumers and the environment while remaining allergy and irritation-free [11]. Textiles treated with an antimicrobial agent should not only meet the standards of cytotoxicity, irritation and sensitisation tests, but should also satisfy the requirement as suggested by [2, 23 - 25] as follows:

a) Withstand repeated washing during its lifespan.
b) Should not have a negative effect on the quality (e.g. physical strength and handle) or appearance of the textile.
c) Be compatible with chemical materials for textile finishing.
d) Should not kill the resident flora of non-pathogenic bacteria on the skin of the wearer.
e) Comply with the statutory requirements of regulatory agencies.
f) Should not produce side effects for the manufacturer, user and environment.
g) Easily applicable and resistant to disinfection/sterilization.

Antimicrobial agents purported to be eco-friendly have been marketed commercially by some companies, for example, Ultrafresh (Thomsan Research Associates), 2, 4, 4′-Trichloro-2′ – hydroxyldipenylether (Tinosan AM 110, Ciba Specialty Chemicals) [26], Sanitized AG (Clariant) [27], Ecosy (Unitika), Utex (Nantech Textile Company Limited) and Vantocil IB (Zeneca) [28] etc. However, a cursory look into the chemistry behind these purportedly natural biocides shows they are not entirely natural [29, 30]. Today wholly natural antimicrobial agents are rarely found on the market due to their poor efficacy and durability.

Natural antimicrobial agents

Research into natural products has demonstrated significant progress in the discovery of new compounds with antimicrobial activity from natural products. Among the known sources of natural compounds with valuable antimicrobial activity are medicinal plants, marine and terrestrial organisms, including fungi and bacteria [26, 31 - 36]. Nonetheless there is still vast fauna and flora that could provide antimicrobials for textiles. Several thousands of natural products with the potential to act as antimicrobial compounds still await further examination. Plants as a major source of natural antimicrobials in nature have received a lot of research attention lately [37 - 39]. Materials extracted from different parts of plants such as bark, leaves, roots and flowers containing common coloring materials such as tannin, flavonoids and quinonoids with strong antimicrobial properties, have been investigated [39, 40].

Extracts from plants

Several studies have shown that natural materials such as alkaloids, saponins, terpenoids and phenolic compounds possess strong antimicrobial activity [40 - 48]. Many of these natural antimicrobial agents are normally accumulated as secondary metabolites in plant cells, but their composition and concentration vary in different parts of plants [49]. Leaves have been found to contain the highest antimicrobial activity and are generally preferred for therapeutic purposes [50]. Perumal Samy et al., [51] found that vinyl hexylether and 2-methylnonane isolated from a *Lonicera involucrata* (*Twinberry Honeysuckle*) leaf had inhibitory activity against *E. coli*, and TIR-01(10,13-dimethoxy-17-(6-methylheptan-2-yl)-2,-3-4,-7,-8,-9,-10,-11,-12,-13,-14,-15,-16,-17-tetradecahydro-1-Hcyclopenta and TIR-05 (3-(2,4-dimethoxyphe-nyl)-6,7-dimethoxy-2,3-dihydrochro-men-4-one) extracted from the *twinberry honeysuckle* root also possess good antibacterial property against *E. coli* [18, 51- 58].

Gupta et al., [59] investigated the antimicrobial property of tannin-rich extract of the *Quercus infectoria* (QI) plant. They found that the use of copper sulphate and alum as a mordant together with the leaf extract had the potential to aid antimicrobial efficacy as well as durability. When the same extract was applied without the mordant, the antimicrobial activity was good but its durability was compromised because the antimicrobial activity decreased drastically after a few washes. They concluded that copper sulphate and alum could help improve the durability of antimicrobial activity of certain plant extracts in textiles.

Sathianarayanan et al., [60] extracted antimicrobial agents from *Ocimum sanctum* and the rind of *Punica granatum*, and compared the washing durability of fabrics treated with direct application, microencapsulation, resin cross-linking and their various combinations. Antimicrobial activities were evaluated using the AATCC-147 test method. Except direct application, all other treatments showed excellent antimicrobial activity as well as good wash durability. While a small decrease in the tensile strength and crease recovery angle was observed for resin treated and microencapsulated fabrics, no significant changes were observed in the combined processes [60].

Essential oils

Essential oils are a mixture of a variety of aromatic compounds which can give cologne and provide protection from a broad spectrum of microbes [61 - 63]. Due to their pleasant fragrance, essential oils containing different pharmaceutical effects have for centuries been applied on textiles to fulfill the psychological and emotional needs of humans [49, 64 - 66]. The application of essential oils for antimicrobial effect on textiles have increased in recent times because they are highly efficient when applied using the right technology [62, 65].

Although significant research has shown the mode of action of essential oils and their combinations for preservatives or antibiotics [67 - 79], the exact mechanism/mode of action of their synergies or purified components on microorganisms when applied in textiles have not been explicitly described [80 - 83]. Presently the generally accepted mechanism of antimicrobial action is the sequential inhibition of the common biochemical pathway, protective enzymes and the destruction of the cell wall of the microbe enhancing quick uptake of the antimicrobial agent as well as cell lysis [84 - 88]. For example, the synergistic effect of carvacrol and some hydrocarbon monoterpenes (such as α-pinene, camphene, myrcene, α-terpinene and *p*-cymene) showed good antimicrobial properties because the hydrocarbons interacted with the cell membrane of the microbes and facilitated quick penetration of carvacrol into their cells [89 - 91]. In a similar work carried out by Pei et al., [92] and confirmed by other researchers on the synergic effects of eugenol/carvacrol and eugenol/thymol, they intimated that carvacrol and thymol disintegrated the outer membrane of *E. coli*, making it easier for eugenol to enter the cytoplasm [94, 95]. The advantages of synergy with various essential oils is that it reduces the concentration needed to yield the same antimicrobial effect when compared with the sum of the purified components [93 - 96].

However, the synergistic effects of essential oils for textile applications, to the best of our knowledge, have not been reported, even though essentials oils have been extensively applied.

Matan et al., [97] showed that thyme essential oils have the ability to inhibit bacterial growth when applied using the pad-dry-cure approach. Walentowska et al., [66] also applied thyme essential oil to increase the resistance of ligno-cellulose textiles to bacteria and mould action. However, other application techniques such as microencapsulation, nanotechnology and cross-linking have been reported [98, 99]. Peppermint oil, with its main component being menthol, is another essential oil with strong antimicrobial activity. It has been reported to exhibit high fungistatic and fungicidal activity against mould growth on rubber wood and other substrates [100 - 103]. Moreover essential oils from clove, cinnamon and several other oils have also been used as antimicrobial agents for bio-medical applications and in the food industry [50, 75, 104-106]. Excessive application of essential oils in linen-cotton blends have been found to affect the tensile strength of fabrics [107], hence the need for compromise between the biocidal efficiency and tensile strength.

Natural pigments

Natural dyes have been widely used in textile coloration since ancient times [19, 108 - 111]. One of the most significant benefits of natural dyes is the eco-friendliness, i.e., they do not create any environmental problems at the stage of production and maintain stable ecological balance [112]. Furthermore natural dyes have an inherent antimicrobial property which is believed to be very potent [110, 113-115]. Natural dyes extracted from different parts of plants including bark, leaves, roots, fruits or seeds, and flowers contain different colouring materials such as tannin, flavonoids and quinonoids [115, 116]. Some natural dyes come from microorganisms such as fungi, algae and bacteria. These dyes do not only offer rich and a wide range of colors, but also possess antimicrobial properties, are environmentally friendly and can be used in low-cost treatments with the additional benefit of colouring in a single step [40].

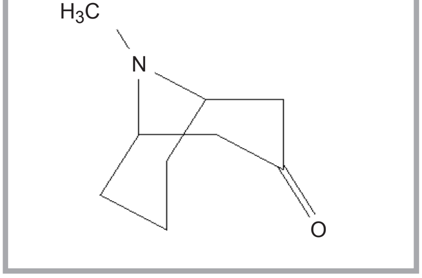

Figure 1. *Chemical structure of granatonine.*

As a result, research into the use of non-toxic, antimicrobial and eco-friendly natural dyes for textiles, preferably natural fibre products, has increased in recent times [117 - 119].

Çalis et al., [120] showed the antimicrobial activity of natural dyes extracted from *Rubia tinctorum*, *Allium cepa*, *Punica granatum* and *Mentha sp*. They found that dye extract of *Punica granatum* was effective against all the bacteria tested except *Escherichia coli* and *staphylococcus epidermidis*. The textile material impregnated with the natural dyes above recorded a high rate of inhibition against bacteria [20]. Mirjalili et al., [121] demonstrated a one-step dyeing process using walnut shells to impart colour and a antimicrobial property to polyamides. Natural dyed polyamide fabrics exhibited good wash fastness and excellent antibacterial activity against two well-known pathogenic bacteria *S. aureus* and *E. coli* when the fabric was treated with a mordant [121]. Jafari et al., [122] also showed that the dyeing of soybean protein fibres with madder, weld and walnut seed husk could exhibit excellent antimicrobial effects against Gram-positive bacteria.

The use of fruits for dyeing and antimicrobial finishing is not a new phenomenon, however, rarely do we find literature directly linking the antimicrobial effects of fruits on microbes especially in textiles, with the exception of *pomegranate* rind, which have been reported recently [123 - 127]. The rind of *pomegranate* is a rich source of tannins, phenolic content, alkaloids, etc. *Granatonine* is the main colouring component found in fruit rind in the form of N- methyl *granatonine* (***Figure 1***).

The extract of *pomegranate* rind has been used in the dyeing and bactericidal treatment of cotton fabric [128, 129]. It offered a wide range of shades with significant antimicrobial activity when copper and iron were used as mordant [123, 128]. When *pomegranate* rind was used without the mordant (copper, iron), few shades of colours were developed. Moreover the antimicrobial activity of the dyed cotton was satisfactory but not durable enough to withstand repeated washing. The mordant (copper and iron) helped to fix the dye into the fabric, while improving the antimicrobial activity of the fabric because they already possess some level of antimicrobial properties [128, 129].

The antimicrobial properties of pigment from microorganisms have also been investigated [130, 131]. Pigment from the fungi *Monascus purpureus* was extracted using ethanol and applied for dyeing wool fibres [131]. Excellent colour fastness with good antimicrobial properties was obtained when alum and stannic chloride were used as mordents. Kim et al., [132] also used *protease* extract from bacteria (*Rhizopus oryzae*) for dyeing wool and silk, and the result showed that by increasing the *protease* extract, the colour of the fabric was enhanced with a good antimicrobial property.

The antibacterial properties of pigments obtained from other fungal species, namely *isaria farinosa, emericella nidulans, fusarium verticillioides* and *penicillium purpurogenum* were studied [133 - 135]. The results showed that the dyed fibres had antimicrobial activity with excellent colour fastness [136]. The extraction, characterisation, and use of pigment produced by the bacteria *Serratia sakuensis* has also been investigated. A novel red pigment was produced by solid-state cultivation of bacterial isolate obtained from garden soil, and used for dyeing silk, wool and cotton fabrics. The dyed fabrics demonstrated good colourfastness and antibacterial activity [137]. Similarly Sharma et al., [138] applied pigments obtained from the fungi *Trichoderma virens*, *Alternaria alternate* and *Curvularia lunata* to wool and silk fibres without a mordant. The fibres showed good rubbing fastness with promising antifungal activity.

Chitosan and its derivatives

Chitosan and its derivatives appear to be the most effective natural antimicrobial agent on the market. Deacetylated chitosan derives its antimicrobial properties from the polycationic sites that are able to bind to negatively charged residues of macromolecules at the cell surface of the bacteria, which subsequently inhibit the growth of bacteria [13, 139 - 141].

Several mechanisms of action and modes of inhibition of chitosan have been suggested [142, 143], meanwhile there is no generally accepted mode of action because the dissimilar physical states and molecular weights of chitosan and its derivatives render distinctive modes of antibacterial action [144]. According to Khor et al., [145] low molecular weight water-soluble chitosan and ultrafine nanoparticles could penetrate the cell wall of bacteria and combine with its DNA, thereby inhibiting the synthesis of mRNA and DNA transcription, while high molecular weight water-soluble chitosan and solid chitosan including larger size nanoparticles interact with the cell surface and alter cell permeability [146]. Others have suggested a situation where the particles interact with the cell and form an impermeable layer around it, thus blocking the transport of essential solutes into the cell [144, 147 - 149]. Irrespective of the mode of action of chitosan, it is highly effective when applied at higher concentration and at a relatively lower of pH 6.5 [13].

Besides the biocidal properties of chitosan on textiles, it also has several other advantages for coloration because the amine group present readily reacts with dyes for successful dyeing/printing [13, 150, 151]. Chitosan is mostly applied by the traditional pad-dry-cure process using a chitosan/citric acid mixture mainly on cotton fabrics, even though other techniques have been used to impart antimicrobial property to fabrics. The use of binders with chitosan for imparting antimicrobial activity to fabrics has also been reported [152 - 154]. In the case of the latter, it can be applied to all manner of fabrics due to the presence of the binder.

The use of chitosan and its derivatives on fibres seems to be the more realistic prospect since this product does not provoke any immunological response [155].

Methods of application for natural antimicrobial agents

Natural antimicrobial agents can be applied to textile by different methods such as pad-dry-cure, coating, spray and foam techniques. It can also be applied directly by adding the antimicrobial agent into the spinning fibre solutions [25, 156 - 158]. Some of the well-acclaimed methods for application of natural products for impacting antimicrobial finishing to fabrics are as folows [159 - 162]:

- Direct application techniques.
- Nanotechnology.
- Insolubilisation of the active substances in/on the fibre.
- Treating the fibre with resin, condensates or cross-linking agents.
- Microencapsulation of antimicrobial agents with the fibre matrix.
- Fibre surface modification.

Direct application technique

Direct application is one of the oldest methods of imparting antimicrobial agents to fabrics. This method is simple but usually cannot achieve satisfactory durability due to the poor attractive force between the fibres and antimicrobial agent, for example, herbal extracts applied directly to textile by pad-dry-cure or [163, 164]. Pad-dry-cure is the conventional technique for applying finishing agents to most textile materials. It is a simple and easy technique where fabric is immersed for about 5 - 10 minutes in an aqueous treating solution. The fabric is padded through squeeze rolls to give a specified wet pick-up, reported as a percentage weight of fabric (% o.w.f.), after which it is dried and cured for a specified time at a specified temperature [165]. Unlike the pad-dry-cure method, the exhaust process involves loading fabric into a bath, originally known as a batch, and allowing it to reach equilibrium with a solution, suspension or dye. Exhaust dyeing enables molecules to move from the solution onto the fibres until it is completely exhausted, after which the fabric is rinsed to remove any excess solution/dyestuff [166, 167]

These two methods are fundamental to any other method for imparting permanent finishes to fabrics regardless of the technology involved.

Nanotechnology

The application of nanotechnology is ideal for improving the antimicrobial activity and wash durability of textiles [168, 169]. Hasabo et al., [99] prepared nanoscale particles from *Azadirachta indica* (neem) extract and used them to treat cotton fabrics. Such fabrics showed excellent antimicrobial activity against *Staphylococcus aureus* and *Escherichia coli.* They also demonstrated that the antimicrobial activity of neem nanoparticle treated fabrics has antimicrobial efficiency up to 25 washes, whereas the activity of the fabrics containing neem extract remained only up to 10 washes. *Azadirachta indica* herbal extract nanoparticles were prepared by the coacervation process, followed by crosslinking with glutaraldehyde and application on cotton using the traditional pad-dry-cure method (***Table 1***) [99].

Table 1. *Antibacterial assessment of nanoparticles of neem extract using the AATCC 100 test method [99].*

Fabric treatment	Antimicrobial activity (Bacterial reduction, %)	
	S. aureus	*E. coli*
Neem extract	98.73	86.84
Nano particles of neem extract	100	91.48

The antimicrobial activity of propolis has been investigated since the late 1940s [170]. The propolis extract has been suggested to inhibit the growth of various strains of Gram-positive bacteria, including *Streptococcus* and *Bacillus* [171]. The use of nanopropolis particles has been suggested to show strong efficacy in the range of 85.2 - 100% according to antibiotic susceptibility tests using *Staphylococcus aureus* and *Streptococcal pharyngitis* [172 - 178]. Nanopropolis has been tried clinically [179, 180], but no such trials have been reported in textile applications.

Microencapsulation

Microencapsulation is a process by which droplets of liquid or particles of solid are covered with a continuous film of polymeric material [181 - 183]. This technology has become one of the most promising techniques of imparting functional finishes to textiles. Microencapsulation is more advantageous compared to the conventional processes in terms of economy, energy saving, eco-friendliness and controlled release of substances [184]. However, if the use of a binder during the process of its application to textiles is not carefully controlled, it can affect the handle of the textile. The capsules are applied to fibres as dispersion with a binder using padding, spraying, impregnation, and exhaust or screen-printing techniques [155].

Major interest in the microencapsulation process is currently shown in the application of durable fragrances [65, 185], phase-change materials [186, 187], dyes [118, 188, 189], fire retardants, counterfeiting, antimicrobial agents, and cosmetic textiles [186]. Many techniques have been developed for producing microcapsules, such as phase separation, spray-drying, solvent evaporation as well as interfacial and *in-situ* polymerisation [188, 190 - 196].

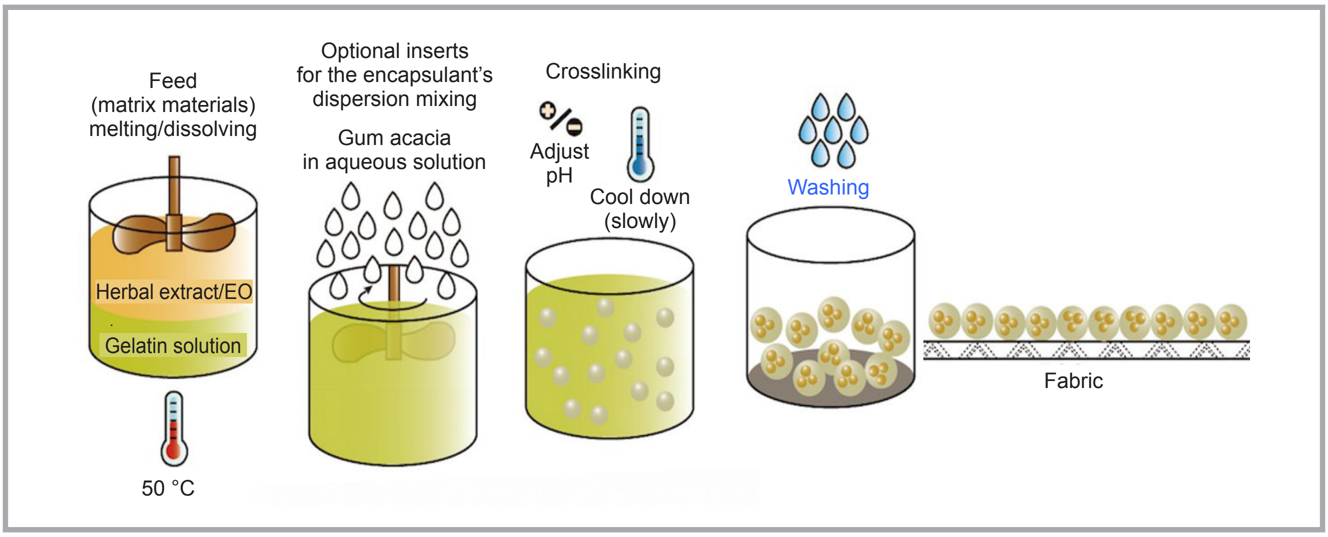

Figure 2. *General scheme for preparation of microencapsulation by complex coacervation using gelatin and gum acacia.*

Phase separation is a process based on the simultaneous desolvation of oppositely charged polyelectrolyte induced by media modifications. As illustrated in ***Figure 2***, the dispersed phase of dense coacervates made up of concentrated polyelectrolyte and a dilute equilibrium phase is dependent on the pH, ionic strength and polyion concentrations [197 - 199]. Gelatin, a collagen hydrolysis product, is used in complex coacervation. It is associated with different polysaccharides to neutralise opposite charges and thereby form a stable complex. Usually the gelatin is positively charged, and coacervation is induced by anionic colloids [200, 201].

Sathianarayanan et al., [60] used herbal extract as core material and gum acacia as wall material to prepare microcapsules. Such treatment of fabrics has been reported to be durable [202]. Emissive fabrication of chitosan microcapsules encapsulating essential oils in the presence of bio-surfactant has been studied [183, 203 - 205]. This method proved reliable and effective due to the antimicrobial activity of both chitosan and essential oil. Also the prolong bioactivity of the fabric has been suggested to be as a result of the slow release of the antimicrobial agents out of the polymer reservoir [60, 188].

Cross-linking method

Cross-linking happens when a cross-linker makes intermolecular covalent bridges between the polymer chains [13, 206, 207]. Cross-linkers include glutaraldehyde, genipin, glyoxal, dextran sulfate, 1, 1, 3, 3-tetramethoxypropane, oxidized cylclodextrins, ethylene glycoldiglycerylether, ethyleneglycoldiglycidylether (EGDE), and diisocyanate [208 - 210]. The approaches to cross-linking are chemical, radiation, and physical methods [115, 211, 212]. Chemical cross-linking happens when the cross-linker makes intermolecular covalent bridges between polymer chains [213, 214]. In radiation cross-linking, heat or a catalyst are not needed, thus no additional toxic chemical is introduced into the system [213, 215]. Radiation polymerisation has been utilizsed by researchers to obtain Interpenetrating Polymer Networks (IPNs) for drug delivery applications, but their applications in textiles have not been reported [216 - 219]. In physical cross-linking, however, a bond is obtained by using cross-linkers that establishe ionic interactions between polymer chains [220]. Chemical cross-linking has really excellent durability with no adverse effect on the tensile strength or handle of the fabric [60]. Physical cross-linking, although not as permanent as chemical cross-linking, possesses good thermal and antimicrobial activity, and is environmentally friendly [60, 221, 222].

To achieve the cross-linking effect of herbal products on fabrics, the herbal extract is mixed with a non-formaldehyde base resin usually with $MgCl_2$ as catalyst [115]. The fabric is coated by the traditional pad-dry-cure/exhaust process to achieve the cross-linking effect. Joshi et al. [13] used glyoxal as a cross-linking chemical to enhance the application of *Azadirachta indica* extract on polyester/cotton blend fabrics. The antibacterial properties of the fabric were retained after several washing cycles, which clearly demonstrated that carefully selected resins have the potential to enhance the cross-linking yield of bioactive plant parts irrespective of the substrate. Similarly citric acid was used as a cross-linking agent in the antibacterial finishing of cotton with green tea leaf extracts to increase durability against washing [220]. Also cotton fabrics were treated with two different cross-linking agents, i.e. Butane tetracarboxylic acid (BTCA) and Arcofix NEC (low formaldehyde content). Cotton fabrics treated with the cross linker above showed broad-spectrum antimicrobial activity against Gram-positive and Gram-negative bacteria as well as fungi [3, 223 - 225].

Combination of microencapsulation and cross-linking

Microencapsulation is a physico-chemical approach for textile finishing. The synergy of microencapsulation and cross-linking has great potential for the application of natural antimicrobes to textiles [5, 74, 89, 226, 227]. Herbal extracts of *pomegranate* rind and *Ocimum tenuiflorum* were used as core materials with acacia gum as the wall material for microencapsulation and non-formadehyde resin as the cross-linking agent [228]. As can be seen from ***Table 2***, cotton fabrics treated using the above-mentioned methods showed good and relatively lasting antimicrobial activity compared to the untreated fabrics.

The compound prepared showed excellent antimicrobial activity against both Gram-positive and Gram-negative bacteria [60]. In a similar study conducted by Banupriya et al., [229], it was found that extracts of *Michelia alba* have high antimicrobial efficacy against all types of Gram-negative bacteria when applied on cotton fabrics using cross-linking and microencapsulation processes. Several other plants have been reported to exhibit excellent bioactive functions [48, 63, 116], some of which have been applied extensively in medicine, but their use in textile applications has not been reported yet due to the lack of suitable methods of application.

Fibre surface modification

Altering the surface properties of fibres is also an interesting way of ensuring the strong adhesion of finishing agents to textiles. Surface modification methods, such as oxygen plasma treatment, ultrasound technology, UV radiation, surface bridging, enzyme treatment and several other techniques have been adopted lately in a quest to impart durable antimicrobial finishes to fabrics using natural products [230 - 233]. Several studies on the use of natural antimicrobial products and colorants combined with these techniques to impart durable finishing treatment have been reported [109, 132, 230, 231, 234].

Chemical modification

Chemical modification is used to impart a deliberate change in the composition or structure of fabrics leading to an improvement in fibre properties [235, 236]. In the pursuit to achieve durable antimicrobial finish with the natural biocide *berberine* (***Figure 3***), the surface of cotton fabric was modified with an anionic bridging agent to create electrostatic interactions with cationic natural dye, thereby enhancing the affinity of *berberine* to cotton fabric [132, 237].

In a similar development, cotton and wool fabrics were modified by pre-treatment with hydrogen peroxide and formic acid to increase the accessible areas [234]. Glyoxal was also used to enhance the application of leaf and seed extract on polyester/cotton blend fabrics [13, 238]. Antibacterial properties were retained after several wash cycles. Also citric acid was used as a cross-linking agent with green tea leaf extracts for antibacterial finishing of cotton fabric. The treatment above was reported to have significantly increased the antibacterial durability of the textile [109, 220].

The use of biological enzymes

Enzyme treatments are used extensively in the textile industry to catalyse chemical reactions due to their effectiveness in improving the wettability, dyeability, bleachability and other finishing processes without fibre devastation [239 - 242]. The biological enzyme *proteases,* capable of conducting *proteolysis*, was combined with some natural dyes possessing antimicrobial characteristics and applied on wool fibres [243, 244]. The effects of protease enzyme treatment on dyeing and antimicrobial yield studied demonstrated that the alkaline *protease* enzyme process enhanced the quantity of natural dyes exhausted while maintaining strong antimicrobial activity. This was attributed to the partial or complete damage of the cuticle in the wool, which facilitated easier penetration of natural dye molecules into fibres [243, 245 - 247]. Also antimicrobial peptides form part of a class of naturally occurring antimicrobial molecules which have good antimicrobial activity against a wide range of pathogenic microorganisms. Antimicrobial peptides are short (typically ranging from 12 – 100 amino acid residues in length), exhibit rapid and efficient antimicrobial toxicity against a wide range of pathogens, and constitute critical effector molecules in the innate immune system of both prokaryotic and eukaryotic organisms. Antimicrobial peptides exert their microbicidal effect via the disruption of the microbial cell membrane together with intracellular action [248, 249]. Laverty et al., [249] have done an extensive review of antimicrobial peptides and their mechanism of action.

The use of plasma technology

The use of physical modification techniques such as low temperature plasma treatment has also been exploited in the application of natural dye/antimicrobial agents [160, 230]. Ghoranneviss et al., [230] studied the dyeing properties of plasma-treated wool fibres with two natural dyes, namely madder and weld. From their findings, they concluded that the treatment of wool fibres with plasma

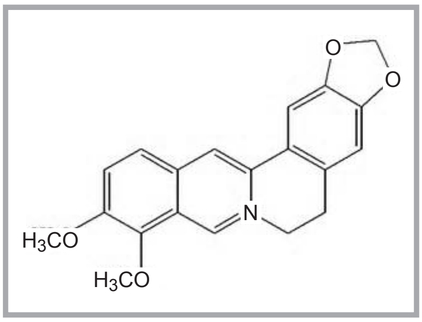

Figure 3. *Chemical structure of berberine.*

enriches the natural coloration process while enhancing the antimicrobial properties of the fibre. In a similar study conducted by Chen and Chang [231], plasma treatment was suitably introduced as a replacement for metallic mordant treatment, recording impressive results. They used low-temperature microwave plasma to treat cotton fabric prior to grafting with onion skin and onion pulp extracts in order to impart antimicrobial properties to cotton fabrics. They discovered that the functional groups produced on the fibre surface after the low temperature oxygen plasma treatment significantly increased the hydrophilic nature of cotton, thereby enhancing the grafting yield of both the onion skin and pulp extracts [135, 160].

The use of ultra sound and UV technologies

The use of modern fabric coloration technologies such as ultra sound and UV radiation in the application of natural dyes with antimicrobial properties has also been reported [115, 250 - 252]. Wool fibres were dyed with natural dye *lac (*secretion of *Kerria lacca* insect*)* using both conventional and ultrasonic techniques [232, 253]. In this study, ultrasonic dyeing recorded an impressive 47% increase in lac dye uptake, where the amount of dye extracted was 41% higher than by conventional methods of extraction. In a related study conducted by Kamel et

Table 2. *Antimicrobial activity of cotton treated with herbal extracts using various methods [26]; Sa – Staphylococcus aureus , Kp – Klebsiella pneumoniae*

	Growth under fabric	Qualitative study zone of inhibition, mm				Qualitative study bacterial growth reduction, %			
		Tulsi extract		Pomegranate extract		Tulsi extract		Pomegranate extract	
		Sa	*Kp*	*Sa*	*Kp*	*Sa*	*Kp*	*Sa*	*Kp*
Untreated (control)	Present	0	0	0	0	0	0	0	0
Direct treated	Present	12.5	6.6	12.6	7.1	99.9	95.3	99.9	90.8
Micro-encapsulated	Present	10.1	5.8	8.0	5.9	99.9	92.6	99.8	85.2
Cross-linked	Present	9.9	4.9	13.5	6.5	99.2	94.2	98.2	89.2
Micro-encapsulated & cross-linked	Present	10.2	5.4	10.1	8.0	99.8	94.5	99.7	87.1

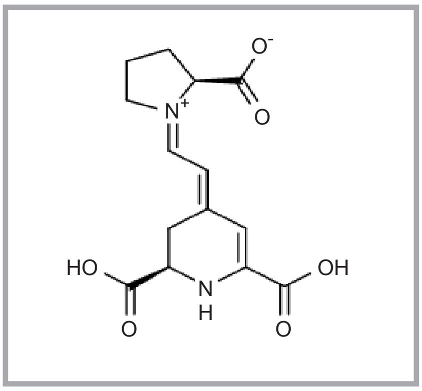

Figure 4. Chemical structure of indicaxanthin.

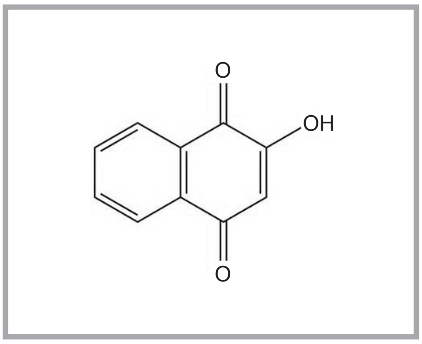

Figure 5. Chemical structure of lawsone.

al., [232] where cationised cotton fabric was dyed using conventional and ultrasonic methods, the ultrasonic dyeing technique recorded a 66.5% improvement in lac dye uptake compared with the conventional method. Also the antimicrobial property of the ultrasonic dyeing technique was much better than that of the conventional method. In a similar comparative study of the two processes conducted by Vankar et al., [254] using dye extract of *eclipta prostrata*, cotton fabric was dyed using both conventional and sonicator dyeing. The dyeing efficiency of sonicator dyeing was much higher than in the conventional method.

In a recent study conducted by Guesmi et al., [255] an assessment was made of the dyeing properties of modified acrylic fibres dyed with *indicaxanthin* (***Figure 4***) using both conventional and sonicator dyeing.

In line with results obtained in the previous venture, sonicator dyeing recorded higher dye uptake, better wash and light fastness, an excellent antimicrobial property with a short process time, and energy consumption compared with the conventional dyeing technique. A similar result was obtained by Vankar et al., [233] when they dyed cotton with *Symplocos spicata* plant dye extract using the two technologies.

Iqbal et al., [256] studied the effect of UV irradiation on henna-dyed cotton fabrics. In their research, *henna* and cotton were UV irradiated separately and then coloured. A second set of henna and cotton were not irradiated under UV light. An investigation into the properties of the two samples revealed that the colour strength and shade of fabrics dyed with irradiated henna were much higher compared to those of non-irradiated henna samples, which was attributed to the hydrolytic degradation of *lawsone* (the colouring matter in henna, see ***Figure 5***) upon irradiation, resulting in a considerable increase in the sorption of the coloured matter.

They further concluded that the UV irradiation of both henna dye and cotton is necessary for the attainment of an excellent colour strength and antimicrobial property [109, 256]

Future prospects

Consumers are now increasingly aware of a hygienic lifestyle, hence it is imperative that textile products should be finished with safe and effective antimicrobial agents. With the enhanced consumer awareness of eco-safety, research has now focused on the use of sustainable and environmentally friendly materials. In recent years, considerable attention has been given to products produced from non-food crops for use in the textile industry [13]. Based on the biocompatibility, biodegradability and non-toxicity of these natural products, in addition to their recently discovered properties such as insect repellency, deodorisation, flame retardancy, UV protection, and antimicrobial activity, these natural products are gaining popularity around the world for producing more appealing and highly functional value-added textiles.

The focus on plant derived antimicrobial agents with potent antimicrobial activities and their application to textiles has increased significantly considering the number of research publications on the topic over the past few years. The contribution of plant-based antimicrobial agents to green nanotechnology in recent years for the development of bioactive textiles has become a reality. The demand for antimicrobial textiles will be spurred primarily by the growing awareness of consumers of the importance of personal hygiene and the health risk posed by microorganisms.

Considering the level of research into so-called 'green technology' today, it is possible that antimicrobial finishing of textiles may be tagged as naturally or synthetically protected, which will offer consumers a greater choice in their quest to satisfy their health needs with respect to textiles.

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