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Review article

A Mini Review of Materials Used as Improvers for Insect and Arthropod Pest Repellent Textiles

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Abstract

Nowadays, concern regarding health epidemics has led to extensive development of functional textiles that have protective functions. Clothing and fabric with insect repellent finishes is a promising and effective way to protect body from insect bites that may carry pathogens. In recent years, several innovations in insect repellent textiles, in particular on new materials and techniques to improve the efficacy and durability of the fabric, have been developed. This review discusses issues and challenges associated with insect repellent finishes. This review also focuses on materials used as improvers in insect repellent textiles whether at the production stage (used in modification fiber or yarn) or in the formulation for post-treatment. The techniques of synthesis, the rates of release, the durability, and insect repellency have also been highlighted in this review. This review offers valuable input to scientists who work in the field of functional textiles and especially from countries with insect-borne disease problems.

1. Introduction

Arthropods like insects can bite and leave an unpleasant as well as uncomfortable feeling to humans. In addition, insects may carry parasites and pathogens that can spread diseases to humans after biting them [1-3]. For example, *Aedes aegypti* mosquito carries virus that can cause disease like dengue which normally causes a serious infection to around 50 to 100 million of people every year, worldwide [4]. Insects and other arthropods like fleas from rats, prairie dogs and rodents can spread

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disease such as plague, which can cross from the animals to humans [3, 5-7]. For instance, biting midges can cause disease such as plague, which can cross from the animals to humans [3, 5-7]. For instance, biting midges can cause visceral filariasis and Oropouche fever, while mosquitoes transmit pathogens related to dengue, yellow fever, malaria, Chikungunya and Zika fever [5-7].

One of the best ways to protect our body from biting insects is the use of insect repellents [1, 8]. Repellents function as a protective layer, when applied to the skin or clothes, can reduce the risk of getting a disease carried by these insects. The mode of action of a repellent is through formation of a vapor barrier on skin or clothes that emits an unbearable odor that can kill or repel the insects from coming in contact with human [1, 6, 8, 9]. Insects like mosquitoes and ticks are often attract to heat, movement, visual cues, skin odor and carbon dioxide exhaled by human, and repellents can interfere with insects' sensory organ such as taste or smell, disrupting their ability to find humans [6].

Ideally, insect repellents must possess several important characteristics like low toxicity, low skin permeation, non-irritative to humans, being unscented or having pleasant scent, long lasting, broad action against varieties of insects, non-oily and non-greasy to skin, as well as water and sweat resistance [4, 9]. Besides, they must also have high chemical stability, good adherence to substrates and be not detachable by abrasion, and be non-reactive to plastics, synthetic fibers, acrylic and glass [4, 9]. In addition, the repellents should not leave a stain on fabrics and clothes, and low cost [4, 9].

The active agents for insect repellents can be divided into two categories, namely natural or synthetic compounds [7, 10]. The natural-based insect repellents are usually from natural oils, and many are extracts or essential oils of plants such as lemon eucalyptus, thyme, garlic, peppermint and lemongrass [6, 11]. Meanwhile, examples of synthetic-based insect repellents commonly used are *N,N*-Diethyl-meta-toluamide (DEET), picaridin, permethrin and IR3535 [6, 7, 9]. There are several insect repellent formulations and products available in the market, which appear in the form of aerosols, pump sprays, lotions, creams, powders, grease sticks, wristbands and clothing [3, 12].

Among all, protective clothing is the most effective and convenient as it can cover most parts of human body [3, 13]. Due to increase public awareness of health and hygiene issues, functional textiles with insect repellent finishes have received great attention. Functional textiles are usually prepared by incorporating natural and synthetic insecticidal or repellent substances into or onto textiles [3]. Functional textiles are used in home textiles, bed nets, tent camping gear and military uniforms [14]. The types of textiles used for these purposes are both natural and synthetic [14]. Natural fabrics are sourced from plant and animal fibers such as cotton, silk and wool whereas synthetic fabrics are usually synthesized from plant cellulosic fibers, such as rayon, and petroleum-based fibers, such as polyester and nylon [14].

Physical, mechanical, biological and chemical treatment processes are usually involved in textiles finishing [14]. Insecticides and repellent substances can be imparted onto textiles at different manufacturing stages such as at fiber preparation, yarn, fabric and garment [4]. Normally, there are four main techniques applied in preparation of insect repellent textiles, namely absorption, incorporation, polymer coating and microencapsulation [1, 8]. The absorption technique, for example, usually employs post-treatment in which the fabrics are sprayed with or dipped into (pad dry cure) repellent agent. Meanwhile, the incorporation technique is usually done as pre-treatment during the manufacturing or dyeing process. Moreover, polymer coating and microencapsulation are quite similar in technique. In the case of polymer coating, a layer of polymer was coated onto the textile fiber. On the other hand, microencapsulation is a popular technique employed to increase the efficacy and stability of repellent agents. It involves containing the agents in a capsule that can provide protection from the immediate environment [1, 8].

Various synthetic and natural based-insect repellents have been utilized to treat textiles. However, according to United States Environmental Protection Agency (EPA), to date only permethrin has been registered for factory manufacture of insect repellents, clothing, and it has been mainly used in the US for military uniforms [3]. The Centers for Disease Control and Prevention (CDC) reported that permethrin is a highly effective insecticide and repellent; hence, the clothes that have been treated with it can repel and kill ticks, chiggers, mosquitoes, and other biting and nuisance insects and other arthropods [2]. Permethrin is poorly absorbed through the skin and works on contact, thus the insect must directly touch the surface of treated item for it to be effective [6, 15].

Prior to the present work, several review articles focused on insect repellents and insect repellent textiles have been reported. Table 1 displays a list of selected review articles that focused on insect and arthropod repellents published from 2010 to 2020. Nevertheless, a great deal of ongoing research over the last five years on materials and techniques that can improve the efficacy and durability of textiles with insect repellent finishes have added valuable inputs to existing knowledge. In addition to the best of our knowledge, there have not been many reviews that highlight the materials used to improve formulations and to modify fabrics for insect repellent textile purposes. In the present review, extensive lists of materials utilized as improvers in insect repellent textile published in research articles dated from 2015 to 2020, which were obtained from online database/search engine such as Google Scholar, PubMed, Scopus, Springer Link and Science Direct, were compiled and discussed. In addition, this review also addressed some issues and challenges encountered in the preparation of textiles with insect repellent finishes.

2. Issues and Challenges in Textile with Insect Repellent Finishes

Although synthetic based-repellents exhibit excellent insect repellency, their usage particularly at high concentrations is of great concern due to their negative effects on human. For example, DEET can be toxic and cause allergic reaction to the skin, especially in the case of children [9]. In addition, the usage of synthetic based-repellents can significantly affect the environment and cause possible resistance by insects [23, 24].

Due to the drawbacks of synthetic-based repellents, natural based-repellents have become popular among consumers as alternatives. Several studies have reported that natural based-repellents showed comparable insect repellent properties to those of synthetic-based repellents [25]. Nonetheless, most of natural repellents such as essential oils are less lasting or have poor longevity compared to synthetic repellents [3, 4]. Essential oils usually can last for a few minutes to hours due to their volatile nature [3, 4]. For instance, Müller *et al*. [26] reported that even though lavender essential oil showed good mosquito repellency rate (93% for indoor and 53% for outdoor), its repelling properties only lasted for about 2 hours. As a result, it needs to be applied frequently. Moreover, some insect repellents whether the synthetic-based (DEET, permethrin) or natural-based repellents (essential oils) have low solubility in water [27]. Therefore, direct use of these repellents can leave stain on textile products [27].

Besides that, synthetic fibers like polyester and polypropylene have poor ability to absorb moisture and thermoplastic in nature, thus can cause discomfort when used as clothing materials. On the other hand, although natural fibers are comfortable, they are not durable. As the natural fibers have hydrophilic nature, whereas the repellent substances are usually oleophilic in nature, thus it is such a great challenge for scientists to bind the repellent on fabric due to weak binding (prevent strong binding) [3, 18]. In addition, textile is often subjected to repeated washing. Therefore, most of the repellent substances are varnished following multiple washings. The repellent can leach out during washing since they do not have any affinity to textiles or fixed properly on the textiles [3, 27].

Table 1. List of selected review articles with their focus related to insect and arthropods repellents

No.	Title	Specific Focus	Reference
1	Environmentally responsive and anti-bugs textile finishes-Recent trends, challenges, and future perspectives	Processes and treatments involved in textile finishing and their drawbacks, environmental problem due to excessive use of dyes in textile finishing and the trends in textile finishes	[14]
2	Trends in insect repellent formulations: A review	Natural and synthetic substances used for production of insect repellents, the existing and new type insect repellent formulations and methods used to evaluate the safety, efficacy and toxicity of the insect repellents	[19]
3	A review on: Mosquito repellent methods	Methods to control mosquitoes using chemical and non-chemical repellents and their mechanisms, the advantages and disadvantages using natural and synthetic chemical repellents and safety measure when using insect repellents	[16]
4	Microencapsulation of essential oils application in textile: A review	Techniques and materials used for microencapsulation of essential oils and its benefits application on textiles	[17]
5	Mosquito protective textiles - A review	Issues and challenges in incorporation of insecticidal and repellent agents on textile and the use of microencapsulation technique to improve longevity and efficiency of insecticidal and repellent agents on textiles	[18]
6	Methods of imparting mosquito repellent agents and the assessing mosquito repellency on textile	Textile materials used in mosquito repellent studies, types of mosquito repellent textile product, techniques to impart the repellents on fabric and methods to assess the efficacy of mosquito repellent textiles	[1]
7	Current and future repellent technologies: the potential of spatial repellents and their place in mosquito-borne disease control	History, role and future approaches of repellent technologies in prevention of mosquito-borne illnesses, and comparison of the characteristics of spatial and contact repellents	[19]
8	Mosquito repellents: An insight into the chronological perspectives and novel discoveries	General characteristics of mosquito repellents, factors that influence mosquito biting behaviour, past and novel repellents available, importance of repellent formulations and their future prospective	[20]
9	Polymer-based drug delivery systems applied to insect repellent devices: A review	Synthetic and natural insect repellents and their targets, and polymeric carrier systems and their applications for insect repellents	[21]

3. Materials Used as Improvers in Insect Repellent Textiles

To date, a great number of strategies and techniques have been employed in pre- and post-production process of fabrics in order to fix and improve the efficiency as well as durability of insect repellent in functional textiles.

The advancement in materials studies have opened wide array of materials that exhibit unique properties for application in many fields including insect repellents and textiles. A number of researchers have studied the feasibility of a variety of materials as improvers to enhance the efficacy and durability of repellent-treated textiles. These materials can be utilized in insect repellent formulations as walls or membranes that can protect the repellent agents from oxidation caused by heat, light, moisture or external substances in post-treatment application. Besides, the materials can also be incorporated directly onto fabric fibers during pre-treatment process to obtain the desire properties. The efficacy of repellent-treated fabrics is usually measured by recording how efficiently the product provides knock-down or repellency effects on biting insects, controls the release of repellents active agents and resists external factors such washing and heating [27]. Table 2 summarizes the materials used as improvers that are highlighted in this review.

3.1 Alginate

Alginate is a naturally occurring polysaccharide extracted from brown seaweed and it is mainly composed of α-(1→4)-linked l-guluronic acid (G) and β-(1→4)-linked d-mannuronic acid (M) [28, 29]. Commercially, alginate can be found in the form of sodium salts. Alginate is commonly used as wall material in encapsulation systems due to its interesting properties such as nontoxicity, biocompatibility, biodegradability and reproducibility [28-30]. Alginate has been utilised in the agriculture, food and pharmaceutical industries as carriers, thickeners and texture-providing materials [28, 29].

Insect repellents have been incorporated into alginate in order to improve the efficacy of insect repellent textiles. For example, Sumithra [31] assessed the ability of sodium alginate to increase the repelling efficiency oo mosquito *Anopheles*. The sodium alginate microcapsule was developed through an ion gelation process and effectively encapsulated extracts of herbal plants (*Amanakku, Avaram* and *Amman pacharisi* oils). The microcapsules were imparted on selected fabrics by pad-dry-cure technique. They observed that fabric made of 100% cotton showed higher mosquito repellent efficiency (84%) compared to other fabrics that were made of 68% of cotton mixed with 32% of polyester, poly Lycra or core spun Lycra. They also observed that encapsulation of herbal plant extracts in sodium alginate microcapsules retained the efficiency of the repellents on the fabric, with 52% of mosquito repellency preserved even after 25 industrial washes.

In another study, Geethadevi and Maheshwari [32] investigated the efficacy of sodium alginate, *Acacia arahica* gum and *Moringa oleifera* gum to encapsulate and increase the durability of thyme, cypress and grape fruit oils on bamboo/tencel blended fabric. The thyme, cypress and grape fruit oils were loaded in the microcapsules at a mixture ratio of 2:1:1. Prior to treatment of the fabric with repellent formulation, the fabric was immersed in 8% citric acid to act as a cross linker. It was noted that the sodium alginate and natural gum capsules had successfully improved the mosquito repellency of the fabric by up to 84%. Besides, the capsules, especially those that incorporated *Moringa oleifera*, were able to protect the oil for a longer duration as the fabrics still showed around 44 to 60% of mosquito repellency even after 30 washes. They also reported that except from one person, no allergic reaction was recorded for fabrics treated with sodium alginate.

Muttakin *et al*. [33] prepared alginate microcapsules loaded with DEET and Kalamegh (*Andrographis paniculata*) extract and studied its repellency effect on *Culiseta longiareolata* mosquitoes when the microencapsulates were imparted on woven and knitted fabrics. They observed that the fabrics imparted with the microcapsules formulation showed higher washes durability than fabric that was applied directly with DEET. The woven and knitted fabrics treated with DEET and Kalamegh microcapsules formulations exhibited around 86 and 83% mosquito repellency, respectively, even after 30 cycles of wash. These mosquito repellency values were higher compared to the fabrics treated directly with 5 and 10 mg/l DEET that showed around 60-73% and 53-73%, respectively, of mosquito repellency after 30 washes.

3.2 Cellulose

Cellulose is a major component present in plants and can be classified as the most abundant natural polymer on the earth [34]. Cellulose is composed of D-glucopyranose units that are linked together by β-(1 → 4)-glucosidic bonds [30, 34]. Due to its abundance and biodegradability, cellulose and its derivatives have been subjected to intense research on many fields [35].

Türkoğlu *et al*. [25] investigated the effect of encapsulation of limonene and permethrin on cotton fabric repellency against *Culex pipiens* mosquito. In this study, the repellent agents were encapsulated with ethyl cellulose via a coacervation method at four different ratios. The results implied that the encapsulation yield of repellent agents and the surface of the capsules varied depending on the polymer to repellent agent ratio. It was found that optimum capsule formula for application on textile was 4 to 1, as this gave a smoother surface and better particle size distribution. It was noted that mosquitoes tended to stay away from cotton fabric impregnated with limonene and permethrin capsules, and that mortality rates of mosquitos were 40.8 and 53.9%, respectively. The researchers reported that the capsules were durable as they still remained on the fabric after washing. Although the efficacy of the treated cotton fabrics decreased with increasing washing, the fabrics still showed mosquito repellency at a certain rate even after 20 washing cycles.

Thite and Gudiyawar [36] reported the use of hydroxypropylmethyl cellulose as a coating material to improve mosquito repellency of insect repellents for application on cotton fabric. For this study, the insect repellents used were synthetic repellent DEET and three natural repellents: lemongrass, citronella and Tulsi oil. The repellent microcapsules were applied on cotton fabric using both microencapsulation and resin cross link methods. It was noted that fabric treated with DEET and lemongrass oil microcapsules showed high mosquito repellency as compared to citronella oil and Tulsi oil microcapsules. It was reported that even though there was no significant difference in mosquito repellency on fabric with and without resin, the application of resin provided durability to the insect repellent finishes. In addition, although significant difference was observed in mosquito repellency on cotton fabric before and after washing, the durability of DEET and lemongrass oil were still excellent due to good adherence and penetration of these microcapsules on the fabric.

Muñoz *et al*. [37] developed a novel citriodiol-loaded electrospun ethyl cellulose nanofibrous mat for application in insect repellent textiles. In this study, two types of citriodiol-loaded ethyl cellulose mats were developed, namely monolithic nanofibers and core-enriched nanofibers. The nanofibrous mats was then incorporated as a core between layered polyester fabrics, and its effectiveness to control *A. aegypti* mosquitoes was evaluated. The researchers also revealed that core-enriched ethyl cellulose nanofiber mats exhibited more prolonged mosquito repellency than monolithic nanofiber mats. They observed that the core-enriched nanofiber mat acted as reservoirs for citriodiol, and thus were able to increase the repellency time. A core-enriched ethyl cellulose nanofiber mat with 36 h of deposition time was reported to be the most effective and was able to retain 100% of mosquito repellency for 34 days before decay to 50% in 47 days.

Hassan and Sunderland [38] applied ethyl cellulose to encapsulate permethrin and imidacloprid, and they found that the application of microcapsules that contained a mixture of permethrin and imidacloprid at a ratio of 3:1 to wool fabric exhibited excellent repellent efficiency and durability to washing. They reported that similar insect-resistance behavior was detected on unwashed and washed wool treated with the ethylcellulose capsules. No visible damage and 100% of *T. bisselliella* larvae mortality was observed on the treated wool fabric. On the other hand, only 5% of *T. bisselliella* larvae died and moderate damage was observed on blank-treated control fabric.

3.3 Chitosan

Chitosan is the second most abundant natural polymer and is derived from chemical deacetylation of chitin [39]. The polymer has been utilized in many areas, such as agriculture, medicine, pharmaceuticals and functional foods due to its biodegradability, biocompatibility, and insecticidal and antibacterial properties [40, 41].

Kala *et al*. [42] prepared chitosan nanocapsules containing lemongrass oil through the ion gelation technique. Acrylate was incorporated in the chitosan nanocapsules as a thickener and fabric binder to obtain chitosan nanogel before impregnation on cotton fabric to achieve long-lasting and wash-durable mosquito repellency. The results showed an improvement in wash durability and retention of chitosan capsules on fabrics following the utilization of acrylate as binder. The cotton fabrics treated with lemongrass oil in ethanolic solution and encapsulated in chitosan nanocapsules without binder showed a significant decline in mosquito repellency after 15 washes. In contrast, the cotton fabric impregnated with chitosan nanogel exhibited around 75% of *A. aegypti* mosquito repellency even after 15 cycles of washing. In addition, no toxicity and abnormality were detected on the bodies of female Swiss albino mice after application with the chitosan gel formulation.

In another study, Gogoi and co-workers [43] coated wool fabric with lemongrass oil to impart a multifunctional finish. Before fabric coating, an oil in water dispersion was prepared by adding nonionic detergent to a mixture of water and lemongrass oil. Chitosan was used as a crosslinking agent to retain the lemongrass oil on wool fabric for a longer duration. They reported that the combination of lemongrass oil and chitosan performed well, improving the antimoth properties of wool fabric. For instance, wool fabric treated with 20% lemongrass oil and containing 0.25% of chitosan as crosslinking agent recorded a high moth mortality rate (80%) and the surface of fabric appeared undamaged.

3.4 Cyclodextrin

Cyclodextrins, also known as macrocylic oligosugars, are products from bacterial degradation of starch [44]. Due to their unique truncated cone structures, cyclodextrins have hydrophobic inner cavities and hydrophilic outer surfaces that enable interaction with various guest molecules to form noncovalent inclusion complexes [45, 46]. Cyclodextrins have wide ranges of application, such as in the pharmaceutical and food industries, and in recent years have been used in environmental chemistry especially in insect repellent formulations. The performance of DEET and eucalyptol encapsulated into β-cyclodextrin using a kneading method as tick (*Hyalomma marginatum*) repellent was evaluated by İnceboz *et al*. [47]. In this study, the microcapsules were fixed on cotton fabric with the aid of cross-linking agent. It was observed that the optimum complex concentration for thermal tolerance was 1:3 and 1:2 for β-cyclodextrin : eucalyptol and DEET, respectively. It was reported that application of eucalyptol and DEET complexes improved the tick repellent activities of the fabric. They noted that fabric treated with eucalyptol complexes showed more stability and higher repellent effect on tick than DEET. They observed that the repellency effect of DEET-impregnated fabric decreased and gradually resembled untreated fabric as the number of washing cycles increased. Meanwhile, the repellency effect of s eucalyptol-impregnated fabric remained stable after 24 h with only a few ticks that contacted the fabrics showing vital activity.

Meanwhile, Bezerra *et al*. [48] developed cyclodextrin complexes that contained citronella oil for application on wool fabric. The cyclodextrin complexes were fixed on fabrics via esterification with the help of butane-1,2,3,4-tetracarboxylic acid (BTCA). They reported that the inclusion of citronella oil in the cyclodextrin produced complexes with average size of 3.014 μm. No health problem was recorded when using the fabric treated with citronella oil complexes as the cyctotoxicity test showed high cell viability (96.394%). The inclusion of citronella oil successfully prolonged the release of oils from the complexes coated on wool fabric. The inclusion of citronella oil also improved the efficiency and increased the wash durability of the wool fabric as some of complexes was observed on the fabric even after 2 cycle of washing.

Specos *et al*. [49] assessed the influence of β-cyclodextrin citrate impregnated on polyester (PET) net substrates in improving the mosquito repellency of citriodiol. In this study, the citriodiol was loaded in the β-cyclodextrin citrate via two methods, namely pipette dripping and impregnation of fabric in a plastic bag. It was found that impregnation of PET nets using the Foulard method enabled the attachment of more than 15% w/w β-CD and its permanence on the PET substrate was noted even after 10 washing cycles. Application of citriodiol/β-CD complexes on the PET nets delayed the release of the repellent agent, which resulted in longer lasting and reloadable mosquito repellent nets. Unlike untreated PET nets, a significant repellency effect of citriodiol/β-CD complex on *A. aegypti* mosquitoes was observed after ten days of application. The treated PET nets continued to show higher mosquito repellency after 17 days until no significant in repellency was observed on both untreated and treated nets after 27 days. In addition, the results also showed that even though both methods exhibited similar release behavior, plastic bag impregnation seemed to be a more suitable method for citriodiol inclusion in β-cyclodextrin citrate.

3.5 Gelatin

Gelatin is a natural polymer derived from partial hydrolysis of collagen, which is the most abundant protein component found in the skin, tendon, cartilage and bones of animals [51, 52]. It has biodegradable and biocompatible properties, and can be commercially obtained at low cost [51, 52]. For that reason, gelation has been employed and fabricated for various application [51].

Teli and Chavan [53] integrated citronella essential oil into gelatin based microcapsules and evaluated the influence of microcapsule drying techniques in enhancing mosquito repellency effect. They observed that due to application of the least heat, freeze drying was the most suitable drying technique, with more citronella essential oil encapsulated in the microcapsules (95.37%) compared to spraying (82.21%) and oven drying (58.23%) techniques. Cotton fabric treated with microcapsules obtained from three different drying techniques demonstrated almost similar mosquito repellency effects (89±1%) and strong aroma after 30 min. They reported that even though the repellency effect and aroma of microcapsules decreased after 10 washes, the microcapsules dried via the freeze drying technique still exhibited higher mosquito repellency (61.38%) than via spraying (57.65%) and oven drying (49.25%) techniques.

In addition, gelatin has also been used in combination with other materials as an improver in insect repellent textiles. For example, Rana *et al*. [54] prepared marigold flower and nirgundi leaf extract microcapsules by mixing gelatin and gum acacia as wall materials. The microcapsules were imparted on cotton fabric with the help of citric acid as binder and their efficacy on *Anopheles stephensi* female mosquitoes as well as the durability to washing and sun-drying were evaluated. It was found that cotton fabric with marigold and nirgundi extract microcapsules exhibited around 94 to 96% of mosquito repellency after 60 min. They observed that although the mosquito repellency of treated fabric gradually decreased after each cycle of washing and sun-drying, the fabrics were durable up to 15 wash cycles and 3 h of exposure as it still showed good mosquito repellency (around 50%).

Gelatin mixed with arabic gum microcapsules was prepared and its ability to improve mosquito repellency and durability of cotton fabric was assessed by Specos *et al*. [55]. In this study, both citronella essential oil and citridiol were encapsulated in the mixture of gelatin at the ratio of

Table 2. Summary of materials used as improvers in insect and arthropod pest repellent textiles

Material	Repellent Agent	Method	Remarks	Reference
Alginate	Herbal plant extracts	Microencapsulation of the plant extracts and impartment using pad-dry-cure methods	-100% cotton showed high mosquito repellency (84%). Able to retain the mosquito repellency of fabric for about 52% after 30 washes	[31]
Alginate, Acacia arabica gum & *Moringa oleifera* gum	Thyme, cypress and grape fruit oils	Microencapsulation of mixed oils in the individual capsules and fixation on fabric with citric acid	-Improved fabric repellency of about 60 to 84% -Able to protects oils even after 30 washes (60-44% repellency)	[32]
Alginate	DEET and Kalamegh extract	Microencapsulation of repellent and impartment to textile	-Higher wash durability. Exhibited 86 and 83% mosquito repellency after 30 washes	[33]
Ethyl cellulose	Limonene and permethrin	Microencapsulation of repellents and impartment using pad-dry-cure methods	-Mosquito stayed away from fabric and mortality rate for permethrin and limonene was 53.9 and 40.8%, respectively -Increased wash durability, still showed repellency after 20 washing cycles	[26]
Hydroxypropyl methyl cellulose	DEET, lemongrass oil, citronella oil and Tulsi oil	Microencapsulation and resin cross link method	- Fabric treated with DEET and lemongrass oil microcapsules showed high mosquito repellency -Application of resin improved wash durability of treated fabric	[36]
Ethyl cellulose	Citriodiol	Citridiol loaded on electrospun ethyl cellulose nanofibrous and incorporation between two polyester layers	-Prolonged mosquito repellency up to 100% repellency for 34 days	[37]
Ethyl cellulose	Permethrin and imidacloprid	Microencapsulation and impregnation on fabric	-Excellent moth repellency on wool fabric -Improved wash durability, washed and unwashed fabrics showed similar resistance behavior	[38]
Chitosan	Lemongrass oil	Encapsulation of lemongrass oil and impregnation on fabric	-Improved wash durability with utilization of binder in chitosan nanocapsules, exhibited 75% mosquito repellency after 15 series washes	[42]

Table 2. Summary of materials used as improvers in insect and arthropod pest repellent textiles (continued)

Material	Repellent Agent	Method	Remarks	Reference
Chitosan	Lemongrass oil	Coated with fabric with lemongrass oil dispersion and incorporated chitosan as crosslinking agent	-Addition of chitosan as crosslinking could improve antimoth properties. -High mortality rate and undamaged fabric	[43]
β-cyclodextrin	DEET and eucalyptol	Encapsulation of repellents and impregnation to fabric	-Improved tick repellent activities. - Eucalyptol complexes showed more stable and higher repellent effect on tick more than DEET - Repellency effect of eucalyptol-impregnated fabric remained stable after 24 h	[47]
β-cyclodextrin	Citronella oil	Encapsulation of repellent and impregnation to fabric	-No cytotoxicity effect -Prolonged release of oil and increase wash durability	[48]
β-cyclodextrine	Citriodiol	Encapsulation of repellent and impregnation to fabric	-Improved wash durability and longer lasting citriodiol	[49]
β-cyclodextrin	Citronella oil	Encapsulation of repellent and impregnation to fabric	- Prolonged release of citronella oil from up to 660 and 360 min on cotton and polyester, respectively	[50]
β-cyclodextrine	Cedar oil	Encapsulation of repellent and impregnation to fabric	-Improved effectiveness and prolonged moth repellent activities more than 2 months	[24]
Gelatin	Citronella oil	Encapsulation of repellent and impregnation to fabric	-Freeze drying gave sample good encapsulation -Exhibited high mosquito repellency even after washes	[53]
Gelatin and gum acacia	Marigold flower and nirgundi leaf extracts	Encapsulation of repellent and impregnation to fabric	-Exhibited more than 94% mosquito repellency after 60 min -Fabric durable up to 15 wash cycles and 3 h exposure	[54]

Table 2. Summary of materials used as improvers in insect and arthropod pest repellent textiles (continued)

Material	Repellent Agent	Method	Remarks	Reference
Gelatin and Arabic gum	Citronella oil and citridiol	Encapsulation of repellent and impregnation to fabric	- Cotton fabric impregnated with citridiol microcapsules exhibited 100% *A. aegypti* for more than 30 days	[55]
Acacia gum	Eucalyptus and cedarwood oils	Encapsulation of repellent and impregnation to fabric	- Durable to rubbing and washes -Exhibited mild and faint aroma after rubbing and washes -100% mortality of silverfish after 18 h of exposure	[57]
Silver nanoparticles	*Moringa* extract	One pot synthesised	-Immobilization with high nanoparticles enhanced durability -100% repellency to *C. pipiens* until 6 days. Repellency decreased to 50-90% after 12 days	[58]
Copper Metal Organic Framework	-	Incorporated directly on fabric surface and loading with repellent	-Cu-BTC increased DEET loaded for more than 60% -Controlled release DEET up to 168-228 hours	[59]
Titanium Metal Organic Framework	-	One-pot synthesis	-Exhibited 100% *C. pipiens* repellency with light -Good washing resistance after 5 wash cycles	[60]
Poly(methyl methacrylate)	Camphor oil	Miniemulsion polymerisation method	-Controlled release of camphor oil and maintained functionality for longer time -Landing time of mosquitoes decreased after 20 wash cycles	[61]
poly(epsilon-caprolactone)	Permethrin	Interfacial polymerisation and spray to fabric	-Good resistance to washing and heating -Cotton had better permethrin adherence than polyester	[62]
Urea-Formaldehyde	Ylang-ylang oil	Encapsulation of repellent and impregnation to fabric	-Good adherence to cotton fabric -Exhibited 80% of mosquito repellency with 60 min exposure	[63]
Kaolinite	-	Coated on fabric	-Improved mortality rate of moths and reduced weight loss -Path bath method at room temperature more effective for moth proofing than exhausted method	[64]

4:1. The reseachers also observed that both emulsified and encapsulated citriodiol provided higher protection against mosquitoes than citronella formulations. The cotton fabrics impregnated with citridiol microcapsules were durable and able to exhibit 100% of *A. aegypti* repellency for more than 30 days after application. Meanwhile, cotton fabrics treated with microencapsulated citronella essential oil presented more than 80% of mosquito repellency for two weeks.

3.6 Gum

A variety of gums have been studied as wall materials for application of insect repellent onto textiles. Plant gum can be employed by itself or blended with other polymers. For example, Karthigeyan and Premalatha [56] assessed the potential of gum acacia loaded with Justicia Adhatoda Vasica extract incorporated on bleached cotton fabric to control mosquitoes. They observed that the microcapsules exhibited small spherical shape and were bound firmly to the surface of cotton. The treated fabric showed around 80% mosquito repellency and was able to retain on the fabric even after 15 cycles of washing without losing its content.

On the other hand, Sharma and Goel [57] evaluated the effects of microenacapsulation of eucalyptus and cedarwood oils in acacia gum on their efficacy at repelling silverfish (*Lepisma saccharina*) insect and durability on woven fabric. They noted that microencapsulation had improved the fastness of the essential oils against rubbing and washing, with eucalyptus oil showing a better durability than cedarwood oil. The fabrics imparted with eucalyptus and cedarwood oil microcapsules exhibited strong and moderate aroma after 10 cycles of dry rubbing before this decreased to moderate and mild after 30 cycles of dry rubbing. Meanwhile, the fabric imparted with both of the microcapsule formulations exhibited faint aroma after 30 cycles of wet rubbing. Faint and mild aroma of cedarwood and eucalyptus oils was observed on fabrics after 10 wash cycles, respectively. In addition, the researchers also reported that all three silverfish insects were found dead in 3 days and 18 h of exposure to fabric imparted with cedarwood and eucalyptus oil microcapsules, respectively.

3.7 Metal-based materials

In recent years, materials made of metal have been synthesized and applied in modification or formulation of insect repellent textiles. Metal-based materials are gaining considerable interest due to their porous structure and high surface area. They also have repellent properties and can be made by green synthetic methods, which can reduce toxicity and negative environmental effects.

El-Sayed *et al*. [58] discussed the use of *Moringa* silver nanoparticles to modify fabrics for mosquito repellent application. The silver nanoparticles were synthesized by eco-friendly methods using *Moringa oleifera* leaf extracts as reducing and stabilizing agent. In this study, four type of fabrics, namely 100% cotton, 50% cotton (CO)/50% polyester (PET), viscose and linen were modified by the sol-gel technique in a bath containing hydrolyzed silane, anti-crease agent and some additives. They reported simultaneous reaction of the epoxy group in the hydrolyzed silane with functional groups in the fabric and silver nanoparticles that facilitated the immobilization of *Moringa* silver nanoparticles on the fabric surface, and thus enhanced the durability of the finished fabric. They noted that the modified fabrics showed 100% repellency to *Culex pipiens* for 6 days. The mosquito repellency of modified fabrics decreased by around 50% except for linen, which exhibited more durability and displayed 90% repellency after 12 days. No adulticidal effects were recorded on modified cotton and CO/PET, while some mortality was observed in linen and viscose fabrics after 24 and 48 h.

New hybrid materials consisting of metal ions and polydentate binding ligands as linkers have been utilized in textiles with insect repellent finishes. For example, Emam and Abdelhameed

[59] directly incorporated a metal organic framework based on copper-benzene-1,3,5-tricarboxylic acid (Cu-BTC) onto cotton, linen and silk fabrics to improve the controlled release and durability of DEET. They suggested that the porous Cu-BTC acted as trapping sites for DEET on the fabrics. It was observed that the porous Cu-BTC was able to increase the amount of DEET loaded on fabrics by more than 60%, with more DEET entrapped on the cotton fabric than other two studied fabrics. In addition, incorporation of Cu-BTC onto the fabrics controlled the release of DEET and was able to enhance the length of the release up to 168-228 h. It was found that release rate and percentage of DEET varied depending on the fabric type and Cu-BTC modification, with silk showing faster release than cotton and linen. The researchers also noted the in-situ incorporation of Cu-BTC affected fabric structure as the modification had changed the color of fabrics to bluish/greenish.

Abdelhameed *et al*. [60] designed anti-mosquito textiles by introducing a composite of titanium-bearing metal organic framework (Ti-MOF) on cotton, linen and viscose fabrics. The studied fabrics were modified with hydrolyzed 3-glycidyloxypropyltrimethoxysilane before being coated with the Ti-MOF composites. In addition, to speed up the impregnating process and shorten the curing time, the fabrics were treated with dimethylol dihydroxyethylene urea (anti-crease agent) in the presence of magnesium chloride as catalyst. The researchers reported that the fabrics incorporated with porous Ti-MOF composites had improved mosquito repellency efficiency and durability of the studied fabrics. Compared to untreated fabrics which showed no mortality, the fabrics treated with 6 g/m^2 Ti-MOF composites exhibited 100% and more than 10% of *Culex pipiens* mosquito mortality after 24 h with light and in dark, respectively. They noted that the crystalline structure of cotton had improved crosslinking bonding with hydrolyzed 3-glycidyloxypropyltrimethoxysilane and dimethylol dihydroxyethylene urea, and thus higher amounts of Ti-MOF composites were able to be coated on its surface than in the case of linen and viscose fabrics. It caused more production of carbon dioxide from urea and resulted in higher mosquito repellency on cotton than linen and viscose fabrics. The modified fabrics also showed good washing resistance as they displayed good mosquito repellency even after 5 wash cycles.

3.8 Other materials

Other than materials that have been discussed earlier, some other polymers, organic- and inorganic-based materials have also demonstrated great potential as improvers in insect repellent textiles. For instance, Wang *et al*. [61] evaluated the effects of encapsulation of camphor oil on the release and mosquito repellent performance of cotton fabric. The camphor oil was loaded into poly(methyl methacrylate) via the miniemulsion polymerization method. The researchers also reported that poly(methyl methacrylate) effectively controlled the release of camphor oil and extended the functionality of cotton fabrics for a longer time. They observed that the landing time for the first mosquito on the cotton fabrics that were coated eight times was about 144.9 min and reduced to 123.9 nm after undergoing 20 washing cycles.

The encapsulation of insect repellent into polymeric nanocapsules consisted of polymer wall and liquid oily core can effectively improve the controlled release and durability on fabric. Forgearini *et al*. [62] prepared permethrin-loaded lipid core nanocapsules, which contained poly(epsilon-caprolactone) as the wall material. The poly(epsilon-caprolactone) nanocapsules were applied to cotton and polyester fabric and their ability to improve the durability of permethrin on fabric was evaluated. The fabrics treated with permethrin-loaded lipid core nanocapsules showed good durability and resistance to washing and heating. They also noted that the presence of poly(epsilon-caprolactone) in the nanocapsules was able to provide better permethrin adherence to cotton fabric than polyester fabric.

Reyes *et al*. [63] prepared mosquito repellent textiles via coacervation of microcapsules of ylang-ylang (*Cananga odorata*) essential oil within urea-formaldehyde shell. It was noted that ylang-ylang microcapsules exhibited good adherence to cotton fabric. The repellency of treated cotton fabric against adult female *A. aegypti* mosquitoes was reported to be improved, with more than 80% of repellency was observed within 60 min of exposure.

Clay mineral was also utilized in the insect repellent textiles. Jose *et al*. [64] assessed the effect of coating wool fabric with kaolinite to improve the moth proofing. In this study, the wool fabric was coated with kaolinite through exhausted method at high temperature and pad bath method at room temperature. They observed that the coating of wool fabric with kaolinite improved the mortality rate and reduced fabric weight loss. In addition, it was noted that the wool fabric coated with kaolinite at room temperature showed more effective moth proofing than exhausted method. The fabric coated with kaolinite at room temperature exhibited 70% of *Anthrenus verbasci* mortality and 1.52% weight loss. Meanwhile, the fabric coated using the exhausted method exhibited 56% *Anthrenus verbasci* mortality and 2.55% weight loss.

4. Conclusions

In this review, a wide range of materials that have been recently used as improvers in the preparation of insect repellent textiles are summarized. The materials have been utilized as wall materials to protect the repellent active agents from external factors. In addition, they can also be incorporated directly onto fabrics to serve as repellent agents or to trap insect repellent on fabrics. Some of these materials have improved the controlled release of repellent agents from fabric. Moreover, some of these materials were also able to improve the repellency performance and durability of treated fabrics for an extended time. Great efforts have been made in recent years to develop protective textiles that can help solve problems concerned with health epidemics caused by insect pathogens. Therefore, this review is relevant and beneficial to scientists working on innovative improvers for functional textiles. Despite their great potential to be used as improvers for insect repellent textiles, the materials have limitations. In the future, textile scientists should study the degradation of the improvers as well as the effects of weather conditions on improver effectiveness. These are the research gaps that should be studied in order to enhance the quality of insect and arthropod pest repellent textiles.

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