Microencapsulation for improved mosquitoes’ repellent efficacy of cotton fabrics

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Abstract. In recent years, mosquitoes that can transfer viruses causing vector-borne diseases, such as dengue fever, chikungunya, Zika and West Nile virus have dramatically increased and reached Europe. In 2018, a higher number of 1503 human cases were reported in the EU/EEA and EU neighbouring countries. The current research was involved in the development of mosquito repellent cotton fabrics with natural essential oils and further improvement of mosquitoes repellent efficacy by microencapsulation of repellents on cotton Fabrics. The repellents efficacy for Anopheles spp. is calculated by using the results of WHO modified test method CTD/WHO PES/IC/96.1. Mosquito-repellency of the treated cotton fabrics against Aedes aegypti mosquito species were tested by using Y-tube Olfactometer. SEM images of treated cotton fabrics were also represented in this paper.

1. Introduction
A recent outbreak of West Nile virus in Central Europe (2018) demonstrates the need for increased research on repellents against invasive mosquitoes which carry the West Nile virus. In 2018 a higher number of 1503 human cases were reported in the EU/EEA and EU neighbouring countries [1].

Many infectious diseases are vector-borne including malaria, Zika virus, dengue and chikungunya virus, West Nile virus and plague, all of which are spread via the bites of infected mosquitoes, and are prevalent in subtropical regions to which South European countries belong. Recent studies indicate that Dengue viruses infect more than 50 million people per year and cause more than 20,000 deaths annually [2]. During the last 15 years Aedes albopictus (Asian tiger mosquito) was introduced into Europe and has spread to several countries, which increases the possibility of its transmission [3,4]. There is limited published data on the prevention of these mosquito borne diseases. Therefore, the development of repellents new targets and related technology becomes paramount importance. The current research was involved in the development of mosquito repellent cotton fabrics with natural essential oils and further improvement of mosquitoes repellent efficacy by microencapsulation of repellents on cotton fabrics.
2. Material and Methods
Scoured and bleached samples of woven cotton fabric were used in the current study. The first set of cotton fabrics was treated with immortelle oil, vibroactivated zeolites (VZ) and water glass, by using traditional Pad-Dry laboratory device [5]. In the continuous impregnation process, the cotton fabric wet pick-up of the treatment liquor was 100%, followed by drying at 120 °C for 2 min. The concentrations of repellents used in the treatment and their corresponding sample labelling code are listed in Table 1. The repellents efficacy for Anopheles spp. is calculated by using the results of WHO modified test method CTD/WHO PES/IC/96.1, which essentially is a chamber method, where mosquitoes are released onto treated fabric. In this method, the mosquito will be repelled to the bait through the gap provided. their effective mosquito repulsion i.e. their ability to migrate away is observed and calculated as Percentage Repellency (resp. efficacy). The observations were recorded over 10 and 30 minutes after mosquitoes were released [6].

| Sample code | Treatment with repellents |
|-------------|---------------------------|
| UN          | Untreated                 |
| VZ_20       | 20 g/l vibroactivated zeolite |
| I_5         | 5 g/l immortelle essential oil |
| I_5_WG_10   | 5 g/l immortelle essential oil + 10 g/l water glass |

The second set of cotton fabrics, was treated with immortelle oil microcapsules (ME), by using Pad-Dry impregnation system, the same system used for the first set samples. Microcapsules were produced in laboratory scale by coacervation phase separation technique, whereby immortelle essential oil (EO) (30%) was emulsified with 1% w/v chitosan and 0.025% w/v sodium alginate. Mosquito-repellency of the treated cotton fabrics against Aedes aegypti mosquito species were tested by using Y-tube Olfactometer [6]. The physical structure and elemental composition of cotton fibers were analyzed by Hitachi SU3500 scanning electron microscopy equipped with energy dispersive spectrometer detector (SEM-EDS). The imaging was made with 3 kV accelerating voltage. The samples were coated with a 10 nm thick layer of gold before analysis.

3. Results and discussion
Cotton samples used in the current research were conventionally treated with new natural mosquito repellents: Immortelle essential oil and micro- to nano- vibroactivated zeolite. Furthermore, waterglass were also used to incorporate with immortelle essential oil (see Table 1 and Figure 1).

Figure 2 shows SEM images of the surfaces of the cotton fibers after treatment with different mosquito repellents. It can be seen that untreated cotton shows fibrilar surface structure. Cotton fibers surface looks smoother and uniform when impregnated with immortelle oil, Vibroactivated zeolite or water glass. vibroactivated zeolite (VZ_20) and water glass (I-WG_10) particles are clearly seen on the fiber surface. The zeolites particles are uniform in the range from 5μm to 10μm. It is found that water glass (sodium silicates) particles are smaller and located only in the spaces between fibers.
Figure 1. Repellents efficacy of conventionally treated cotton fabrics against Anopheles spp mosquitoes.

Figure 2. SEM images of cotton fabrics: a) untreated, b) treated with vibroactivated zeolite, c) immortelle oil, and d) immortelle oil and water glass - signals detected by the backscattered electrons (BSE) (LUT).
Figure 3. Repellency for conventional finishing with immortelle oil (I) and microencapsulated immortelle oil (I_ME) on cotton samples against Aedes aegypti by using Y-tube olfactometer; ME-microcapsules.

Efficacy results for microencapsulated immortelle oil are high giving a 100% efficacy against Aedes aegypti (Figure 3), compared with the immortelle oil alone and against the same mosquito, only a 57% efficacy rate against Aedes aegypti was achieved but 70% to 80% against Anopheles spp.

The repellent with the greatest efficacy was cotton treated with immortelle oil and water glass and will be considered for future research work on this area. Further studies are being undertaken to optimize the concentration of immortelle oil to be used and to improve the efficacy of microencapsulation of immortelle oil for the controlled release of bioactive oils to achieve the long lasting of mosquito-repellency. This is a pilot study demonstrating initial findings on potential repellents against mosquitoes further work on synergistic effects of different repellents, fabric construction, repellent concentration, wash fastness properties and technology application still need to be assessed.

4. Conclusion
The natural bio repellent used in this paper, immortelle oil, showed good to excellent repellent efficacy, when applied using conventional technology or novel microencapsulation procedure. For Anopheles, the highest efficacy is obtained with immortelle oil and water glass but not so high for vibroactivated zeolite. Microencapsulated immortelle oil against Aedes aegypti showed the highest efficacy, compared with immortelle oil alone when applied using conventional technology for the same mosquito.

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