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Microencapsulated citronella oil for mosquito repellent finishing of cotton textiles

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ABSTRACT

Microcapsules containing citronella essential oil were prepared by complex coacervation and applied to cotton textiles in order to study the repellent efficacy of the obtained fabrics. Citronella released from treated textiles was indirectly monitored by the extractable content of its main components. Repellent activity was assessed by exposure of a human hand and arm covered with the treated textiles to *Aedes aegypti* mosquitoes. Fabrics treated with microencapsulated citronella presented a higher and longer lasting protection from insects compared to fabrics sprayed with an ethanol solution of the essential oil, assuring a repellent effect higher than 90% for three weeks. Complex coacervation is a simple, low cost, scalable and reproducible method of obtaining encapsulated essential oils for textile application. Repellent textiles were achieved by padding cotton fabrics with microcapsules slurries using a conventional pad-dry method. This methodology requires no additional investment for textile finishing industries, which is a desirable factor in developing countries.

1. Introduction

Mosquitoes are insects of major public health concern because many species are vectors of diseases. *Aedes aegypti* is the primary vector of urban yellow fever and dengue, the most important arboviral human infection worldwide. Infected specimens are widely distributed in Argentina due to the traffic of people from affected neighbouring regions and to adequate climatic conditions.[1] From1998 to the middle of 2007, a total of 4718 suspected cases of Dengue Fever were reported in Argentina.[2] A significant outbreak has been taking place in several Argentine provinces since February 2009 with more than 20 000 clinical cases of Dengue fever, including five cases of Dengue Hemorrhagic Fever, and four deaths up to April 2009.[3]

In recent years various essential oils have been reported as mosquito repellents due to their eco-friendly and biodegradable nature,[4,5] in particular, the essential oil extracted from *Cymbopogon nardus* (citronella), one of the main aromatic crops grown in subtropical regions of Argentina such as Mesopotamia, Chaco and Formosa. Citronella has been found to possess effective repellent activity against *A. aegypti* females for 2 h after the direct application of pure oil on the skin of human subjects.[6]

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A more durable effect has been found with synthetic repellents such as DEET, the most widespread mosquito repellent. Recent studies have shown that a formulation with 80% DEET provides almost 14 h of protection against *Aedes aegypti* while a 7% formulation provides protection for less than 4 h.[7]

The incorporation of insect repellents in textiles seemed an alternative way to provide longer lasting protection, although their direct application to fabrics has proven to be inefficient in prolonging the repellence for long enough. Microencapsulation technology has enabled an increase in the durability of the desired effect in versatile textile finishing. A microcapsule provides both space for storing a certain amount of a functional agent and a protective shield from the effects of sunlight, moisture and oxygen. The release of the functional agent occurs by diffusion through the capsule wall and/or rupture of the microcapsules. Therefore, the use of microcapsules could provide a durable repellent finish that could be applied to a multitude of fibres.[8]

Fabrics with repellent activity have been previously achieved by polymer coating methods,[9] by encapsulation of DEET in chitosan microcapsules,[10] and by inclusion of DEET and permethrin in cyclodextrins grafted to textile substrates.[11] However, to the best of our knowledge, the repellent efficacy of cotton fabrics with microencapsulated citronella oil finishes has not been investigated.

The aim of this work is to study the repellency to female *A. aegypti* of fabrics treated with gelatin-arabic gum microcapsules containing citronella oil. These microsystems have been selected due to the low cost and the biocompatible nature of the ingredients involved, which is most desirable in fabrics intended for human use. Microcapsules containing citronella oil were characterized and applied to cotton textiles. The residual mosquito repellency of treated fabrics was determined by means of an in vivo assay and was compared to the repellency of fabrics sprayed with non-encapsulated citronella.

2. Materials and Methods

2.1. Materials

Citronella essential oil (Fornasari Ltd., Buenos Aires, Argentina) was used as mosquito repellent agent. The fabric used was a bleached 100% cotton plain weave with a specific weight of 140 g/m^2. Type A gelatin (Rousselot Argentina Corp., Buenos Aires, Argentina) and arabic gum (Van Rossum Ltd., Buenos Aires, Argentina) were employed as wall materials. Glutardialdehyde 25% w/w (Merck & Co. Inc., Hohenbrunn, Germany) was used as hardening agent. All chemicals were reagent-grade.

2.2. Preparation and characterization of microcapsules

Microencapsulation of essential oil was performed by complex coacervation as previously described.[12] Briefly, an o/w emulsion was prepared by homogenization of 1.5% w/v gelatin with citronella essential oil for five minutes at 18 500 rpm with a high shear homogenizer Heidolph 900 DIAX (Schwabach, Germany). Temperature was maintained at 40 °C while 1.5% w/v arabic gum solution was added drop-by-drop (5:1 w/w oil to total polymer ratio). Coacervation was accomplished by dilution with distilled water at a rate of 20 ml/min. Temperature was then reduced to below 10 °C and the pH was adjusted to 8 with sodium hydroxide. 0.6 ml of a 25% w/v glutardialdehyde aqueous solution, corresponding to 0.03 mmol glutardialdehyde / g protein, was slowly added upon mechanical stirring with a pitched blade impeller Heidolph RGL 500 (Schwabach, Germany). The suspension of gelatin-arabic gum microcapsules containing citronella was kept upon magnetic stirring at room temperature overnight. Size and morphology of microcapsules were determined by optical microscopy (OM) using a Zeiss Axioskop 40 microscope with a Nikon coolpix 8800 camera (Göttingen, Germany). Microcapsules slurry was spray dried in an AS Niroatomizer mobile minor unit (Soeborg, Denmark) using an inlet temperature of 120 °C and an outlet temperature of 90 °C. A free-flowing powder was obtained and was further analyzed by scanning electron microscopy in a Philips SEM 505 (Philips/FEI, Eindhoven, Netherlands).

2.3. Textile treatment and evaluation

Cotton fabrics were treated either with microencapsulated citronella or by spraying with an ethanolic solution of citronella. In the first case, cotton fabrics were padded twice through an aqueous finish bath containing citronella microcapsules (16% solid in water), nipped to obtain a wet pickup of 100%, and dried in the tenter frame at 100 °C for three minutes. In the second case, a 10% w/w solution of citronella oil in ethanol was prepared and sprayed onto fabrics until the samples were entirely wet.

All samples were cut into 10 cm × 10 cm pieces as specimens and stored for up to 44 days indoors at 22 ± 4 °C and 65 ± 5% relative humidity. On each day of a predefined series of time intervals, samples were taken and tested by two independent methods. First, a judge smelt the swatch after scratching an 'x' on the specimen with a plastic stick. Samples were measured on a positive and negative scale about the presence of the fragrance.[13] Second, a solvent extraction of citronella essential oil from textiles was performed as previously described.[14] Briefly, three specimens of each sample were incubated with ethanol for 24 hours at room temperature in closed vials with magnetic stirring. The three main components of citronella: citronellal, citronellol and geraniol were determined in the ethanolic extracts by gas chromatography (GC) coupled to a flame-ionization detector (Thermoquest, Milan, Italy) using an HP-5 column in the following conditions: 80 °C initial temperature, 8 °C/min temperature gradient, and 200 °C final temperature. Textiles sprayed with the ethanolic solution of citronella were extracted with ethanol as described and analyzed by GC in order to determine the initial content of citronella. The aforementioned GC conditions were also used to characterize the composition of the original citronella essential oil.