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Microencapsulated essential oils : from fragrant fabrics to repellent textiles

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INTRODUCTION AND OBJECTIVES

Functional textiles have been developed to satisfy the needs of comfort and safety of consumers. The most studied applications include insects' repellency, antimicrobial finishing, phase change materials, fire retardants, and aromatherapy.

The need for long-term durability of functionality has brought about the introduction of microencapsulation techniques. In particular, essential oils have been microencapsulated using different wall materials to protect them from oxidation and control their release (Hong 2000). Some of them have been reported as effective mosquito repellents (Trongtokit 2005, Sorge 2009), which is a relevant application especially for some developing countries. In Argentina, for example, Aedes aegypti mosquitoes infected with dengue virus are nowadays widely distributed due to the traffic of people from affected neighboring regions and to adequate climatic conditions. Thus, the development of repellent textiles is a good option as a complementary strategy for the control of vector populations.

The aim of this work is to develop repellent textiles with a durable functionality using microencapsulated essential oils as finishing treatments. Microcapsules of lemon essential oil were obtained by complex coacervation with gelatin and arabic gum and applied to cotton fabrics by conventional textile procedures. The optimized methodology was applied to repellent essential oils such as citronella and citriodiol and the repellent efficacy of cotton fabrics was evaluated by an *in vivo* assay. These controlled release microsystems have been selected due to the low cost and grass status of the ingredients involved, which is desirable for textiles intended for human use.

MATERIALS AND METHODS

Preparation and characterization of microcapsules

Microencapsulation of lemon and citronella essential oils as well as the natural pesticide citriodiol was performed by complex coacervation as previously described (Leclercq 2009). Briefly, an o/w emulsion was prepared by homogenization of 1.25% w/v type A gelatine, 0.85% w/v arabic gum, and 8.5% w/v essential oil for five minutes at 9600 rpm and 50º C. Microcapsules were obtained by coacervation accomplished by dilution with distilled water and hardened with glutardialdehyde. Size and morphology of microcapsules were determined by optical microscopy and scanning electron microscopy (SEM) after spray drying of the slurry.

Textile treatment and evaluation

Bleached 100% cotton plain weaves were padded twice through an aqueous finish bath containing microcapsules, nipped to obtain a wet pickup of 100%, and dried in a tenter frame at 80ºC for 3 minutes. Microcapsules containing lemon essential oil were applied with or without the addition of auxiliary products, such as acrylic and polyurethane binders, at different concentrations. In this case, fabrics were also cured at 130°C for 35 seconds in a scale pin stenter.The presence of microcapsules on textiles was assessed by SEM and the fragrance intensity was evaluated by three trained judges and quantified by gas chromatography.

Repellency test procedure

The repellent activity was evaluated with three sets of gloves manufactured from treated textiles as follows: i) untreated fabrics (UF), ii) fabrics padded with microencapsulated citronella oil (MC-citronella), and iii) fabrics padded with microencapsulated citriodiol oil (MC-citriodiol). Samples were hung in the open-air during the period of the assay except for those used for the shell life tests which were kept in semi-hermetic bags.

The repellent activity was evaluated by inserting a human hand and arm covered with the gloves into a test chamber (50 x 50 x 50 cm), based on cage tests described in bibliography (Kweka 2008). The covered arm was kept for five minutes in the test chamber containing approximately 200 Aedes aegypti adult females. The number of insects landing was counted independently by two observers. The same subjects' uncovered arm and hand were also inserted as control. The trials were conducted in quintuplicate in five different cages at 26±1 ºC and 80±5%RH with a 5 minute waiting period between replicates.

The percentage of insects landing on treated fabrics (IL) was calculated against untreated fabrics using the following equation:

$$\%IL = \left(\frac{MTF}{Muh}\right)100$$

where *MTF* and *Muh* correspond to the mean number of mosquitoes landing on treated fabrics and uncovered hand respectively.

RESULTS AND DISCUSSION

Microencapsulation parameters were optimized to obtain uniform microcapsules in the 10-30 µm range containing each of the three essential oils (Fig.1).

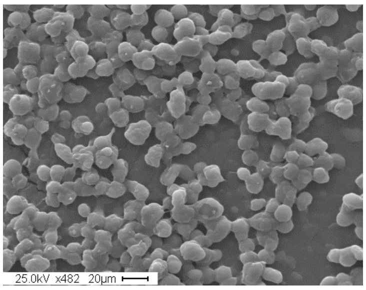

Figure 1 : SEM image of spray dried microencapsules containing citriodiol (magnified x 500)

Microcapsules containing lemon essential oil were used for the optimization of textile application. Once applied as previously described, they could be observed among the fibers (Fig.2, left) and the fragrance could be detected for at least 18 months (data not shown) upon scratching.

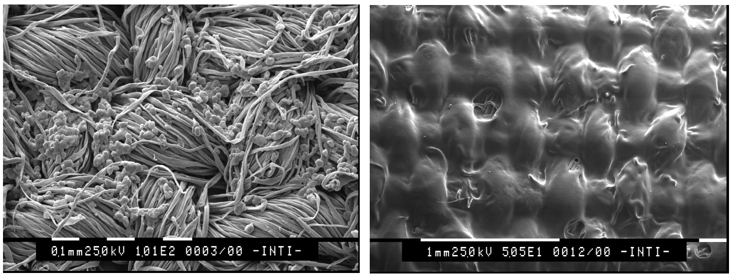

Figure 2 : SEM image of cotton fabric treated with microcapsules containing lemon oil without (left, magnified x 1000) or with (right, magnified x 500) the addition of binders

The use of binders, initially proposed as a way to improve the washing ressistance of treated textiles, resulted in decreased fragrance intensity and therefore lower durability. This may be due to a polymeric film formation over cotton fibers, which interferes with fragrance release (Fig.2, right).

Figure 3 shows the percentage of mosquitoes landing (%IL) as a function of time on gloves subjected to different treatments. Repellence was improved for both microencapsulated citronella and citriodiol, with respect to untreated cotton glove, while citriodiol provided the highest protection against mosquitoes. In fact, cotton fabrics treated with microencapsulated citronella presented an 80% repellency for two weeks, while repellency for fabrics treated with microencapsulated citriodiol was higher than 95% after more than one month (Fig.4).

Cotton samples padded with either citronella or citriodiol microcapsules were kept in Ziploc® bags and their repellency was evaluated every month to simulate the storage before distribution. Textiles treated with microencapsulated citronella started losing their repellency after 7 months, while citriodiol provided full protection even after 12 months-storage.

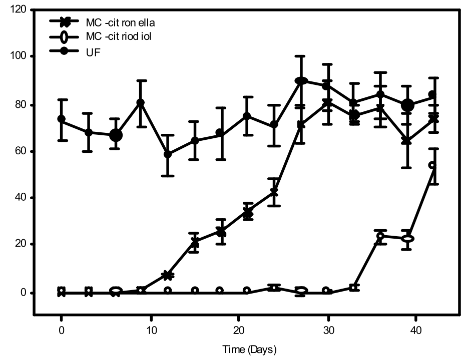

Figure 3 : variation of the percentage of insects landing on untreated or treated fabric referred to insects landing on a bare arm. (Bars show standard deviations for n = 5)

CONCLUSIONS

We have previously demonstrated that these repellent textiles based on microencapsulated essential oils obtained by complex coacervation resist only a few washing cycles (Miró 2010). Moreover, repellent effect has proven to be much shorter than fragrance durability of textiles treated with microencapsulated lemon oil. However, fabrics treated with microencapsulated citriodiol, which can provide full protection for at least a month, may have potential in some situations where protection is required with no need of washing such as disposable cloths for people working at risk zones.

Further investigations will be conducted to test other microencapsulated systems applied to different textiles substrates in order to improve laundering stability.

It must be stressed that the methodology described to develop this kind of repellent fabrics is simple, low cost, and reproducible and requires no additional investments for textile finishing industries, a desirable factor in developing countries.

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