DEVELOPMENT OF ECO-FRIENDLY AROMATIC MOSQUITO REPELLENT FROM *Pelargonium radula* (Jeremin) STYPTIC OIL

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INTRODUCTION

Essential oil has been the subject matter of many investigations due to its eco-friendly and bio-degradable nature. Geranium oils obtained from *Pelargonium* sps. is one of the various essential oils that are used as mosquito repellent. A thorough study has been performed on the geranium plant species known as *Pelargonium radula* to produce geranium oil. However, stability is one of the issues related to essential oil formulations. Incorporation of essential oil in a control-release formulation could solve the problem and it also offers several advantages. In this work, the objective of this research was to produce a new controlled-release delivery system for mosquito repellent based on the incorporation of geranium oil into solid lipid particles (SLP).

METHOD

The experimental work started with the extraction step where geranium oil was extracted using steam distillation method. The active compounds in the oil were determined using GC and GC-MS. Repellency activity of geranium oil was then evaluated using human as volunteers and adult female *Aedes aegypty* as vectors. Additional analyses that were carried out were anti-microbial assay and skin irritation potential test.

Next, ultrasonic-solvent emulsification technique was used to produce SLP loaded with geranium oil. The SLP acts as a controlled release delivery system. Two different sizes of SLP (nano: SLN and micro: SLM) were produced by exposing the system to different length of ultrasonication times. The SLP was characterised in terms of particle size, charge surface, morphology, encapsulation efficiency, active ingredients release patterns and *in vivo* penetration.

The SLP which was stable, provided excellent controlled release property and with minimal transdermal penetration was included in the final product formulation. The effectiveness of the mosquito repellent cream containing geranium oil SLP was determined by laboratory repellent bioassay.

RESULTS

The geranium extract was obtained from steam distillation extraction process and the yield obtained was 2.54% or 0.76 g/g. The main chemical component of the geranium plant was geraniol (20.52%) followed by citronellol (20.31%). The geranium oil obtained possessed an aromatic scent (sweet rose like odor). The repellency activity results showed that geranium oil has a median effective concentration value (EC_{50}) of 0.0051 mg cm^{-2} compared to the repellency activity of DEET which has an EC_{50} value of 0.0005 mg cm^{-2}. Anti-microbal analysis showed that geranium oil has good anti-microbacterial characteristic especially on *Staphylococcus aureus* (straining, 1.22 mm). Contact with geranium oil also did not result in any adverse effects such as rashes or sensitivity to human skin.

Geranium oil was successfully loaded into solid lipid particles (SLP) using ultrasonic solvent emulsification technique. Two types of SLP were produced which were the SLN (567.24 nm) and SLM (1.37 µm). The geranium oil-SLP formulations were found to be most stable at pH 6.5 to pH 7 where the value of the zeta potential was -50 mV to -80 mV. The structure of the geranium oil-SLP was spherical and smooth when seen through transmission electron microscopy (TEM). The geranium oil-SLP formulations also have high encapsulation efficiency (EE %) around 89 to 99 % (w/w).

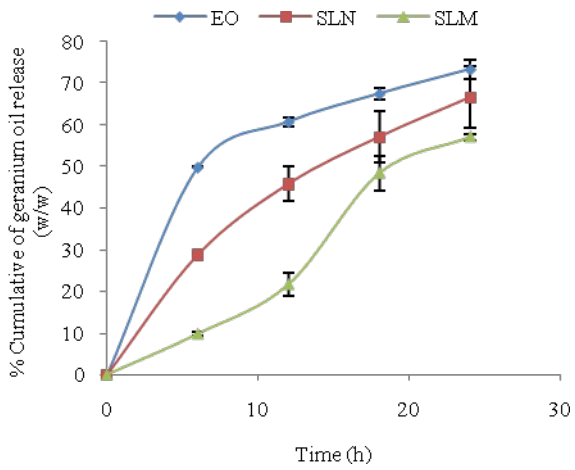

Figure 1. Release profile of geranium oil from (◊) unencapsulated geranium oil (EMU), (□) geranium oil loaded solid lipid nanoparticles (SLN) and (Δ) geranium oil loaded solid lipid microparticles(SLM)

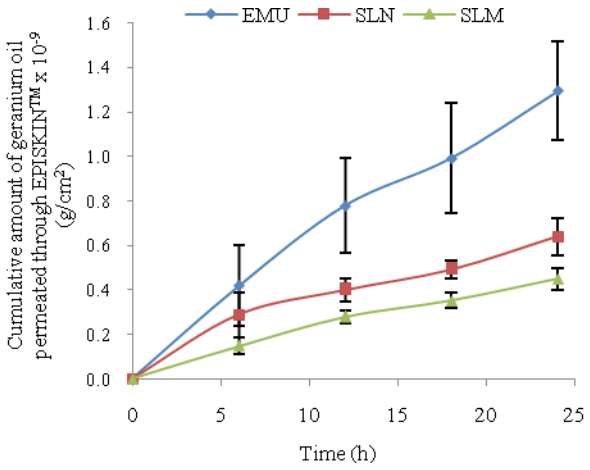

Figure 2. Cumulative amount of geranium oil permeated (g/cm^2) from unencapsulated geranium oil (EMU), geranium oil-loaded solid lipid nanoparticles (SLN) and geranium oil-loaded solid lipid microparticles (SLM) through EPISKIN™, as a function of time

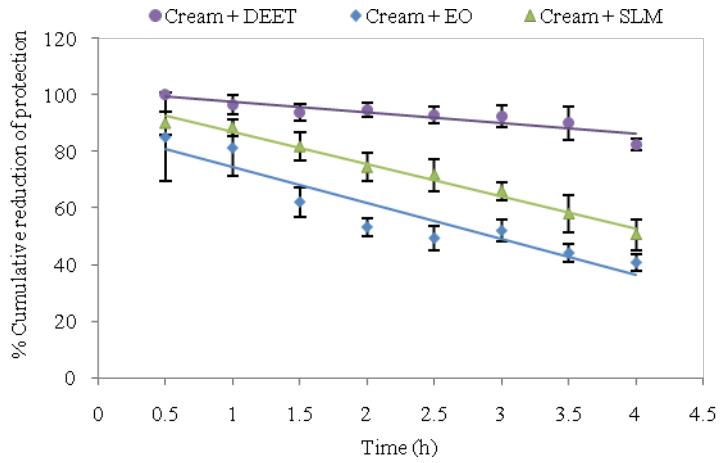

Figure 3. Percentage protection from mosquito bites based on cream product; (○) DEET-based repellent cream, (◊) geranium oil-based repellent cream, and (Δ) geranium oil loaded SLM-based repellent cream

Release profiles of unencapsulated geranium oil (EMU), geranium oil loaded solid lipid nanoparticles (SLN) and geranium oil loaded solid lipid microparticles (SLM) after 24 hour exposure to temperature of 35ºC are shown in Figure 1. The cumulative of oil release are as follows: EMU 73.32 % > SLN 66.57 % > SLM 57.04 %. This showed that SLM worked best in slowing the release of geranium oil.

Figure 2 shows the profile of transdermal penetration of the three formulations. The geranium oil-SLM formulations has the greatest capability in reducing the absorption of the geranium oil through the skin based on diffusion studies done on EPISKIN™ membrane (EMU: 1.29 % > SLN: 0.64 % > SLM: 0.45 %) (Figure 2).

Based on these results, geranium oil SLM was selected to be included in the final product formulation. Figure 3 shows that geranium oil-SLM formulations applied to based mosquito repellent cream formulation managed to improve the effectiveness of the protection from mosquito bites type *Aedes aegypty* (CREAM+SLM: 52.68 % > CREAM+EO: 40.78 %). Standard repellent DEET was also evaluated and as expected gave higher protection (CREAM+DEET: 82.48 %) compared to the natural geranium oil loaded SLM. Pearson correlation analysis indicated there was a significant relationship ($p < 0.05$) between the release profile of geranium oil from geranium oil loaded SLM, transdermal diffusion of geranium oil loaded SLM and percentage protection of mosquito bites of geranium oil loaded SLM. This proves that geranium oil loaded SLM is an effective natural mosquito repellent compared to unencapsulated geranium oil.