

Comparism of Repellent Produced From Grape and Orange Peels

^{1*}Adepoju, M.A., ¹Taiwo, A. A., ¹Ogunnaike A.E., and ¹Salami,A.A.

¹Department of Science Laboratory Technology,
Moshood Abiola Polytechnic, Abeokuta, Ogun State

Abstract

The activities of mosquito repellents produced from peels of orange and grape fruits were compared. The extracts of Citrus fruit peels were air-dried and powdered, soxhlet method was used for the extraction with diethyl ether as solvent. Three different repellents (Candle, Cream and liquid) were produced from each extract. Application of the products on human volunteers showed that the products repelled mosquitoes and other insects effectively. This study revealed that some phytochemical compounds found in the extracts were active ingredients which made the prepared repellents effective on all species of insects without having any harmful effect on the users. There were no significant difference in the results of physico-chemical parameters and repellent properties of the two studied citrus fruits.

Key Words; Production, Mosquito repellent, Orange, Grape, Peels.

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I. Introduction

Mosquitoes are commonly found in African countries, they are responsible for malaria, yellow fever and dengue fever, these diseases are very deadly if not properly treated. WHO reported about 15,000 deaths per annum in India, this embraced all ages. Mosquito control measures need to be put in place in urban and rural areas, the best way is to restrain their breeding by using non-hazardous chemicals (Neeraj Dhingra *et al.*, 2010). Mosquito repellents available in the market have formulations with remarkable safety profile, but their toxicity against the skin and nervous system can cause rashes, swelling, eye irritation and other serious problems to children (Neeraj Dhingra *et al.*, 2010). This led some researchers to bio-based natural mosquito repellents which is preferable to chemically prepared ones. Bio based mosquito repellent are based on bio active ingredients derived from plants. These bio based products play important role in controlling pest in areas where mosquito resistance, and environmental concerns limit the use of products. Some of these mosquito repellent are prepared from the following; Basil (*Ocimum basilicum*), oils of Castor, Cedar, Clove, Fennel, Citronella (Trongtokit *et al.*, 2005), Eucalyptus, Neem (Caraballo *et al.*, 2000), Rosemary and Catnip oil of *Nepeta* species which have nepetalactone, Celery extract (*Apium graveolens*) and *Solanum villosum* berry juice. These natural resources are good for the environment and also have good flavour (Muller *et al.*, 2008).

Mosquitoes are attracted towards man because of the presence of lactic acid and Carbon (IV) Oxide in human sweat. The attraction is caused by Chemo-receptors present in the antennae of mosquitoes which perceive the smell of the sweat. The special role of natural mosquito repellent is to mask human scent. Most plants contain compounds that can be used to prevent attack from plant eating insects. These can be categorized as repellents, feeding deterrents, toxins and growth regulators. Repellents from plant origin is not hazardous to human and domestic animals, they are easily biodegradable. Natural products are safe for human when compared to that of synthetic compounds (Patel and Oswal 2012). Hence, there is need to launch extensive search to explore eco-friendly biological materials for control of insect pests.

JUSTIFICATION

Reports from researchers showed that the smoke generated from mosquito coil is dangerous to health, this may lead to severe headache, nausea, respiratory impairment and vomiting, the condition may be severe with asthmatic patients. Therefore, it becomes imperative to produce mosquito repellents that are not harmful to man's health with the use of bio degradable product of plant to control mosquito bite and infections

II. Materials And Methods

SAMPLE COLLECTION AND PREPARATION

Fresh orange and grape fruits were purchased from popular markets in Abeokuta, Ogun State, Nigeria. The orange and grape fruits were washed in water, dried, peeled, and the peels were chopped with a knife and washed in water, the peels obtained from the fruits, were spread on a clean ply wood and air dried. The dried peels were grounded into powdery form using a manual grinding machine and stored in an air tight container.

EXTRACTION

1kg of the powdered sample was weighed into a 2L soxhlet apparatus containing 1000 ml of diethyl ether. The citrus oil extraction was done at temperature range of 50^oC to 70^oC for about 6 hours. The extract was left over night in order to cool down the temperature and to allow the remaining diethyl ether to evaporate.

REPELLENT CREAM PREPARATION

Petroleum jelly and paraffin wax were measured into the pot; the mixture was heated and allowed to melt. Lanolin, glycerin, vitamin E, mineral oil, Shea butter and Coconut oil were added and stirred thoroughly. The mixture was allowed to cool, fragrance and colour were added, and the resulting mixture was thoroughly mixed. When warmed the extracted oil was added to the mixture, thoroughly mixed before pouring into a container.

LIQUID REPELLENT PREPARATION

The oil from citrus fruit contains limonene, the extracted oil from the citrus rinds was used to prepare insecticide, and citrus oil is all that is needed to make insecticide spray. The peels were dried, grounded and was soaked in alcohol, the mixture was strained leaving the alcohol to get evaporated.

REPELLENT CANDLE PREPARATION

The candle was made from mixture of solid paraffin and stearic acid as hydrocarbon base. Solid paraffin and stearic acid were heated to a temperature of 700^oC until they melted, essential oil was added, this made the temperature dropped between 550^oc – 600^oc. Active ingredients Cucurmin were then added to separate formulations. The wick was inserted in the liquid wax, so as to obtain the required diameter. It was poured into a cup shaped mould, which was then filled with liquid wax. After cooling the solidified candle was removed from the mould.

PHYSICO-CHEMICAL ANALYSIS OF THE EXTRACTED OILS

SOLUBILITY

Four drops of extracted grape peels oil was added to the test tube containing 8 drops of water. The mixture was stirred thoroughly with glass rod. The mixture was allowed stayed for 5 minutes. Two separate liquid phases were observed. The pH of the water phase was measured to check for the solubility of the essential oil in water. The above experiment was also carried out on orange peels oil. (Hognadottir *et al.*, 2003).

BOILING POINT

5 ml of extract ed oil from grape peel is placed a small test tube, a capillary tube with sealed end is placed inside the test tube with the closed end upward. The test tube is clamped to a ring stand, and a thermometer was attached to it. A 250 ml beaker was half filled with water, and placed on the hot plate. The test tube containing the mixture and thermometer were carefully lowered into the beaker of water. The mixture was heated slowly on a hot plate. few bubbles were observed flowing out of the end of the capillary tube as the oil approaches its boiling point. When steady streams of bubbles were observed, the temperature at which the oil began to flow into the capillary tube was recorded. The above experiment was repeated for orange peels oil. (Hognadottir *et al.*, 2003).

SPECIFIC GRAVITY

A dry density bottle of 25 ml capacity was weighed to give initial weight of W₀. It was filled with water and reweighed to give (W₁). The water was drained and bottle was clean . It was then filled with grapefruit essential oil and weighed to give (W₂). The specific gravity was then determined from the following equation. The above experiment was carried out again using grape essential oil (Hognadottir *et al.*, 2003).

$$\text{Specific gravity} = \frac{W_2 - W_0}{W_1 - W_0}$$

REFRACTIVE INDEX

Abbe's refractometer was used for the determination of refractive index of the oils. There refractive index is denoted by n_D^{25} where n is the refractive index at 25°C taken with sodium light (D-line). The refractometer was calibrated with distilled water (RI of 1.3330 at 20°C/68°F and 1.3325 at 25°C/76°F) which has refractive index of $n_D^{29.5} = 1.3315$. The glass prism was wiped clean using cotton pad which was earlier moistened with acetone. A drop of grape peels oil was placed between the prisms of refractometer and allowed time for temperature equilibrium between the instrument and the sample. The telescope was rotated to bring the border line of total refraction other junction of cross-wire in the telescope. The refractive index was recorded at room temperature. The above experiment was repeated for orange peels oil (Maria *et al.*, 2012).

ACID VALUE

10 g of the extracted grape oil was weighed into a dried conical flask. 50 ml of ethyl alcohol was added into the conical flask and 3 drops of phenolphthalein was added to it. The solution was titrated against 0.1N KOH until the end point is reached, (pink colour). The volume of KOH used in the titration was recorded. (Maria *et al.*, 2012). The above process was repeated with for orange peels oil. The acid value was calculated with the equation:

$$AV \text{ (mg KOH/g sample)} = \frac{56.1 \times V \times N}{m}$$

Where, V – Titre volume of standard potassium hydroxide solution (ml);

N – Normality of the potassium hydroxide solution, m – Mass of oil sample (g)

SAPONIFICATION VALUE

2g of grape peels oil was weighed into a 250 ml conical flask, to which 50 ml 0.5 M alcoholic KOH was added. The mixture was constantly stirred for 1 hour this was followed by reflux. 3 drops of phenolphthalein indicator was added and titrated with 0.5 M HCl until the pink coloration disappeared. (Maria *et al.*, 2012). The procedure was repeated for orange peels oil. The saponification value was calculated with the equation:

$$SV \text{ (mg KOH/g sample)} = \frac{[(S-B) \times M \times 56.1]}{\text{Sample Weight}}$$

Where, S = sample titre value (mL), B = blank titre value (mL), M = molarity of the HCl

III. Results And Discussion

The physicochemical properties of grape and orange peels are presented in Table 1. The specific gravity of the two samples are similar (0.791 grape and 0.811 orange), this is similar to the result presented by (Fashola *et al.*, 2016), they reported 0.84540 for grape and 1.8429 for tangerine. (Barkatullah *et al.*, 2012) also reported a close value of 0.79g/ml for *Skimmia laureola* oil. In the other hand, higher value of 1.995g/ml was reported by (Olagunju 2013) for mango seed oil. The refractive index for the two samples were 1.43 and 1.512 respectively, these are also close to the value by (Fashola *et al.*, 2016), 1.473 grape and 1.476 tangerine. (Akinhanm *et al.*, 2008) also observed lower refractive index of 1.201 for cashew seed oil. In the same vein, (Adelaja 2006) also reported lower refractive index (1.13) for coconut oil. Refractive index shows the stability of oil during thermal treatment and the level of saturation of oil (Olagunju, 2013).

The boiling points ranges from (171°C to 179°C) the two sample are insoluble in water but soluble in organic solvent (Schumann *et al.*, 2005). The boiling point of a liquid varies, this depends on the environmental pressure (Olagunju, 2013).

The acid value for the samples observed for orange peels were 17.3 mg KOH/g while 15.33 mg KOH/g for grape, the observed value is lower than acid value (31) reported for edible oil (Siekmann 2005). The result obtained in this result is higher than 0.39 – 0.86 mg KOH/g oil and 0.16 – 0.60 mg KOH/g oil reported for groundnut oil and palm oil respectively by (Babatunde and Bello, 2016). Acid value is an important physicochemical property index of oil which determines the edibility and suitability of oil for industrial use such as paint (Akubugwo *et al.*, 2008). This value is used to measure the extent of glycerides in the oil, which have been decomposed by lipase and other physical factors such as light and heat (Demian, 1990).

Saponification value observed is 180.3-190.56 mg KOH/g which is greater than saponification value observed for edible oil, palm kernel oil 0.2503, pea nut oil 0.1925, palm oil 0.2000 (Gunstone *et al.*, 2016). The value observed for saponification is close to value reported for soap making oil coconut oil 191.1, olive oil 135.3, soy bean oil 135.9 castor oil 128.6 (Hognadottir and Russell, 2003). Saponification value shows the average molecular mass of various fatty acids in oil samples. The lower value of saponification indicate molecular weight of fatty acids is lower and has lower limit of use in industry (Denniston *et al.*, 2004). The

saponification value put forward the use of oil in production of liquid soap, shampoos and lather shaving creams (Oderinde *et al.*, 2009).

The present study was conducted to determine the repellent activity of the extracted oil from orange and grape peels. The visual appearance of the repellent cream was pale yellow with acceptable odour. The spreadability of the two extract orange and grape was 10 secs because it has an easily spreadable property. The pH of the two extract ranged from 6.8-7.0 which is an indication of a neutral product. The irritancy test of the two extract was performed and there were no sign of redness and itching in area applied on some bear portion of the human body (hand, legs and face). The two cream made from the extracted oil were thermally stable at $45^{\circ}\text{C} \pm 1^{\circ}\text{C}$ and no phase separation was observed after 72 hours.

Generally, it was observed that repellent cream made from the extract of grape and orange peels showed a long lasting repellent effect up to 6 hours. Candles were prepared with the two extracts, properties of the prepared candle evaluated is presented in Table 3, the flames of the repellent candles started to repel mosquito 68 secs after it was lit, after the lighting of the candle the smoke produced from the two extract grape and orange stopped after 60 secs similarly the extract of the two samples (orange and grape) showed long lasting repelling effects for more than 6 hours. The insecticides produced were put in an air tight container to avoid oxidation of the content. It was tested on some insects (cockroach, mosquitoes and black ants) it was found to be effective and was able to kill the insects within a space of 120 secs, this was observed with product from grape and orange while for the mixture of orange and grape 108 secs was observed and this was shorter than time observed when they were used singly, this may be due to the high content of limonene in the grape and orange fruit extracted oil used as part of the raw materials in producing the insecticides. The pleasant odour added as a fragrance in the two extracts (orange and grape) were perceived at the range of 120 secs. The extract of orange and grape show a long lasting repellent effect of more than 6 hours as shown in Table 4.

Table 1: physicochemical analysis of grape and orange oil

Parameters	Grape peels	Orange peels
Specific gravity	0.791	0.811
Refractive index	1.453	1.512
Boiling point ($^{\circ}\text{C}$)	$171^{\circ}\text{C} - 173^{\circ}\text{C}$	$177^{\circ}\text{C} - 179^{\circ}\text{C}$
Solubility	Insoluble (H_2O)	Insoluble (H_2O)
Acid value(mg KOH/ g oil)	15.33	17.3
Saponification value (mg KOH/ g oil)	190.56	180.3

Table 2: Results of Evaluation of Repellent cream made from extract of orange and grape peels

Parameters	Orange peels extract	Grape peels extract	Grape and orange extract
Physical Appearance	Pale yellow	Pale yellow	Pale yellow
	Colouration with	Colouration with	Colouration with
	Odour	Odour	Odour
Spreadability	< 10 Secs	< 10 Secs	< 10 Secs
Irritancy Test	Non	Non	Non
pH	7.0	7.0	7.0
Thermal stability	Stable at $45^{\circ}\text{C} + 1^{\circ}\text{C}$ For 48 hrs.	Stable at $45^{\circ}\text{C} + 1^{\circ}\text{C}$ For 48 hrs	Stable at $45^{\circ}\text{C} + 1^{\circ}\text{C}$ For 48 hrs

TABLE 3: Evaluation of repellent properties of candle

Parameters	Orange	Grape	Mixture of Orange and Grape
Mosquito repellency	68 secs	68 secs	66 secs
Pleasant odour	120 secs	121 secs	120 secs
Smoke production	68 secs	66 secs	60 secs
Activity	> 6hours	> 6hours	> 6hours

Table 4: Evaluation of liquid repellent made

Parameters	Grape	Orange	Mixture of Orange and Grape
Mosquito Repellency	120 secs	120 secs	118 secs
Pleasant Odour	60 secs	60 secs	60 secs
Activity	> 6hours	> 6hours	> 6hours

IV. Conclusion

In conclusion, repellents produced from citrus fruits peel have proved effective as mosquito repellents. The results showed that both orange and grape extracted oils can provide substantial protection against mosquitoes and insects bite as the of synthetic based DEET .This lasted up to 6 hours per application.

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