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ORIGINAL RESEARCH ARTICLE

EFFECT OF PERIOD AND STORAGE CONDITIONS ON ESSENTIAL OIL YIELD AND COMPOSITION OF EUCALYPTUS CITRIODORA LEAVES

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ABSTRACT

Fresh eucalyptus citriodora leaves were harvested and kept under shade and sun for a period of four weeks. The effect of storage conditions and period of storage on the oil yield, oil composition and extraction pattern of the oil were investigated. It was observed that for the leave kept under sun there was significant decrease in the oil yield from 0.38% in the 1st week to 0.11% in the 4th week. However, for leaves kept under shade the oil yield slightly decreased from about 0.40% in the 1st week to 0.36% in the 4th week. The results further revealed that the storage condition and period of storage had no effect on the extraction pattern of the oil with about 72% of the oil extracted within 40 minutes of extraction time after induction period of 24 minutes. Two mathematical model equations were developed for the prediction of oil yield as a function of storage time for both conditions. The models predicted that for leaves stored in the shade and sun the expected oil yield would be 34.5% and 1% respectively. Physiochemical analysis of the oils revealed that the properties of the oil were not affected by both the period and condition of storage except the colour which changed from pale yellow to light brown. These results imply that the leaves should best be kept under shade before production in order to preserve its oil content and physiochemical properties.

1.0 Introduction

Essential oils are volatile, natural base products, which are found in spices, aromatic and medicinal plants. The Extraction of essential oils is well known from old ages when pure essential oil and crude extract of essential oil bearing plants, herbs and grasses were in use for various medicinal and fragrances, flavors, preservatives and insect repellants purposes (Weiss, 1997; Panda, 2000).

Eucalyptus plant is mainly grown in tropical and sub-tropical regions because of its resistance to many pest and adaptability to various climatic conditions. The plant has up to 700 species and can grow as high as 40 m tall in an altitude of 600m. The yield of eucalyptus oils from leaves depends on the species and varies from 0.10 to 9.0 % on dry weight basis. Oil content also

fluctuates with season. For instance, E. citriodora leaves collected in Pakistan in April-May contained 0.9% oil than 1.3% in December (Rafique and Chaudhary, 1996).

In terms of toxicity, eucalyptus oils have been categorized as GRAS (Generally classified as safe) by the United States Environmental Protection Agency (US EPA) due to the high LD 50 of their major components for rats (Batish et al., 2008). The physiochemical properties of essential oil are primarily responsible for its biological activities (Barbosa et al., 2016; Vuong et al., 2015). Some of these activities include antimicrobial (Saleem et al., 2016; Mekonnen et al., 2016; Luis et al., 2016; Said et al., 2016), herbicidal (Tomaz et al., 2014), anticancer (Vuong et al., 2015), and insecticidal activities (Karemu et al., 2013; Bossou et al., , 2015).

Many research works have been published on different factors affecting eucalyptus oils yields: tree age (Andrade and Gones, 2000), leaf age (Goguadza, et al, 1986; Silvestre et al, 1997), altitude (Manian and Gopalakrishinan, 1995), season (Rafique and Chaudhary, 1996), harvest time (Doran et al, 1995) and fertilizer application (Maffeis, et al, 2000). However, scanty information is available on effect of storage of eucalyptus citriodora leaves under different conditions on the oil yield and its physiochemical properties. The objective is to provide detail information on the best period and condition of which the leaves should be stored so as to preserve the oil yield and its physiochemical properties. It will also serve as an important resource to the commercial producers in the essential oil industry regarding the best period and condition of leave storage.

2. Materials and Methods

2.1 Material Sourcing and Preparation

Fresh eucalyptus citriodora leaves were obtained from National Research Institute for Chemical Technology (NARICT) plantation in Zaria, Nigeria. The leaves were removed from its stalk and freed from any foreign materials presence. 20kg of the leaves were weighed into 8 places (portions) for eight batches operation and kept four (4) portions under shade (Figure 1). The remaining four (4) portions were kept in the sun (Figure 2).

Figure 1: Eucalyptus citriodora leaves kept under shade

Figure 2: Eucalyptus citriodora leaves kept under the sun

2.2 Extraction of Essential Oil

The pilot plant for the extraction process is shown in Figure 3. The plant consists of cylindrical tank still, condenser, steam generation and cooling water units. 20 litres of water was charged into the boiler section (steam generation unit) of the tank still which is located at the bottom

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and occupies 1/5 of the tank height. The boiler section is separated from the oil extraction chamber by a stainless steel weir mesh. The extraction was first carried out using fresh leave as control before using stored leaves. 20 kg of the fresh leave was then charged into the tank still from the top and tight properly to avoid steam leakage. The gas burner located at the bottom of the tank still was then ignited and timed immediately. The burner fuel- to- air ratio was adjusted until blue flame was obtained implying steady energy supply.

Figure 3: Essential oil extraction pilot plant

The steam produced from the boiler passed across the packed bed of leaves and extracted the oil which was conveyed to the condenser. The cooling water entered the condenser from the overhead high density polyvinyl chloride (PVC) tank by action of gravity at the rate of 1 litre per minute. The flow of steam-oil mixture in the condenser and cooling water was counter current for effectiveness. The condensate (essential oil + warm water mixture) was collected in a transparent glass separating flask at various time intervals and separated. The volume of oil (see Figure 4) and corresponding warm water collected at various times were measured and recorded. Sodium sulphate was then added to the oil and allowed to stay overnight followed by filtration to remove any traces of moisture and suspended impurities.

Figure 4: Essential oil extracted

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After 1st week of storage, same extraction procedure mentioned above was repeated using leaves kept under sun (SUN) and shade (SHD) separately. In each case, the cumulative oil collected, extraction time, steam requirement and energy consumption were recorded. The same procedure was repeated for 2nd, 3rd and 4th week of storage for both sun and shade storage. The percentage yield of the extracted oil was calculated using Equation (1).

$$\text{Oil Yield } (\%) = \frac{\text{Weight of oil}}{\text{Weight of leaves}} \text{ x } 100 \qquad (1)$$

2.3 Determination of Physiochemical Properties of the Oil

The physiochemical properties of the extracted essential oil such as refractive index, specific gravity, solubility in ethanol and ester value were determined according to the methods described elsewhere (Galadima, 2004 and UNIDO, 1983)

3. Results and Discussion

3.1 Effect of Extraction Time on Oil Volume

The effect of extraction time on the quantity of essential oil extracted from eucalyptus citiriodora leave kept under both shade and sun are presented in Figures 5 and 6 respectively. In Figure 5, it can be seen that throughout the storage period (1-4 weeks), the volume of the oil extracted increases with increase in extraction time until it reached extraction time of 20 minutes after the induction period before declining. The induction period is the time between ignition of the burner and the first drop of the steam plus oil mixture collected. This period is generally dependent on the energy supply, volume of the water in the boiler and loading density of the leaves and was found to be about 24 minutes.

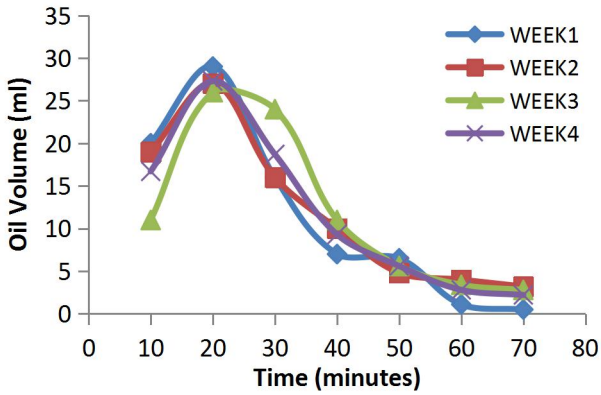

Figure 5: Extraction pattern of essential oil as a function of time for leaves dried under shade

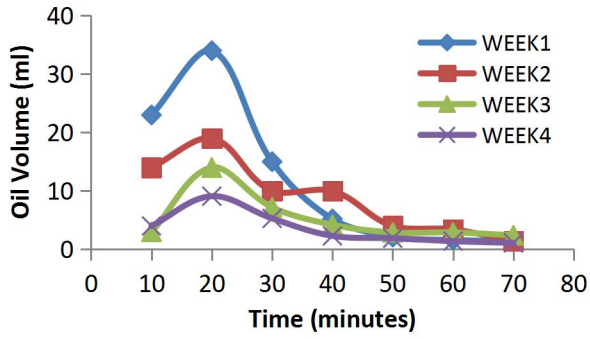

Figure 6: Extraction pattern of essential oil as a function of time for leaves dried under sun

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However, similar pattern of extraction was observed for leaves dried under sun (Figure 6) with maximum oil extracted within 20 minutes before declining. It was also observed that about 72% of the oil was extracted within 40 minutes for dried leaves in the shade and 45% of the oil also in 40 minutes for the leaves dried in the sun. This implies that there was significant reduction in the oil extracted from the leaves dried in the sun when compared with the leaves under shade and fresh leaves (67%) as reported by other researchers (Mu'azu, et al., 2009). It also evident from these graphs (Figures 5 and 6) that storage of the leaves under both conditions (sun and shade) had no effect on extraction pattern of the oil but rather the oil yield.

3.2 Effect of Storage period and condition on Oil Yield

It was observed that during storage of the leaves under both sun and shade there was significant reduction in the weight from 20 kg of fresh leave to 8 kg for storage under shade and 7kg under sun in the 4th week as shown in Table 1.

Table 1: Effect of storage period on eucalyptus leave

Storage period (week)	Weight (kg)	
	Sun	Shade
0	20	20
1	12	14
2	9	10
3	8	9
4	7	8

This reduction in leave weight is primarily due to evaporation of both moisture and oil from the leaves and is more pronounced on the leaves kept under sun which also increases with time of storage in both cases. The effect of storage time (weeks) and condition (shade and sun) on oil yield were studied as shown in Figure 7. As can be seen from Figure 7, there was slight significant changes in the oil from 1st week of storage (0.40%) through the 4th week (0.36%) for the leaves kept under shade. This implies that the reduction in leaves' weights as shown in Table 1 were basically due to evaporation of the moisture not the oil.

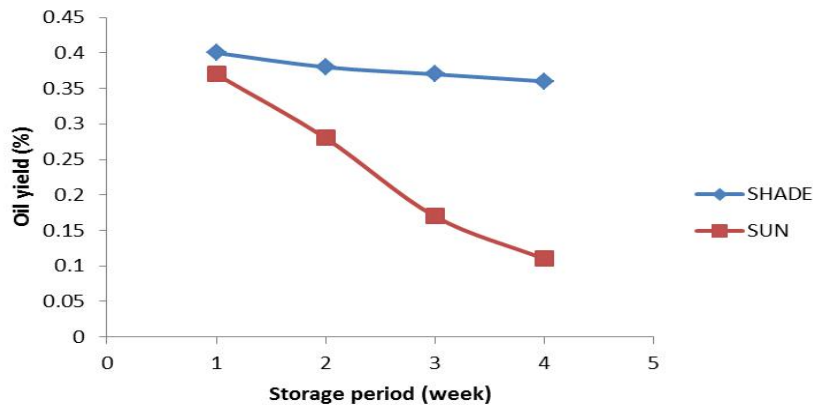

Figure 7: Effect of storage period and condition on oil yield

In the case of leaves kept in the sun also shown in Figure 7, significant reductions in the oil yield were observed from 0.38% in the 1st week to 0.11% in the 4th week. This also implies that

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storage of the leaves in the sun reduces both oil and the moisture significantly. It also evident that in the 1st week of storage the oil yield remained relatively constant in both cases (0.38-0.40%) in spite reduction in the leave weight. This explains the fact that the reduction in the weight was due to evaporation of the moisture not the oil and was being held as unbound moisture or free moisture while the oil is bound moisture held within the leave's matrix (Mu'azu et al., 2009)

Comparison of the oil yield obtained from sun and shade storage in the 4th week with the oil yield from the fresh leaves, the results revealed reduction in oil yield by 70.3% for sun and 10% for shade indicating that storage under the shade preserve more oil than sun. It is worth mentioning that the oil yield obtained from this plant species (0.579%) is far lower than the yield obtained from same plant species (1-1.2%) grown in India as reported elsewhere (Manika et al., 2012). The difference could be due time of harvesting, maturity, tree age, leaf age and fertilizer application (Maffeis, et al., 2000)

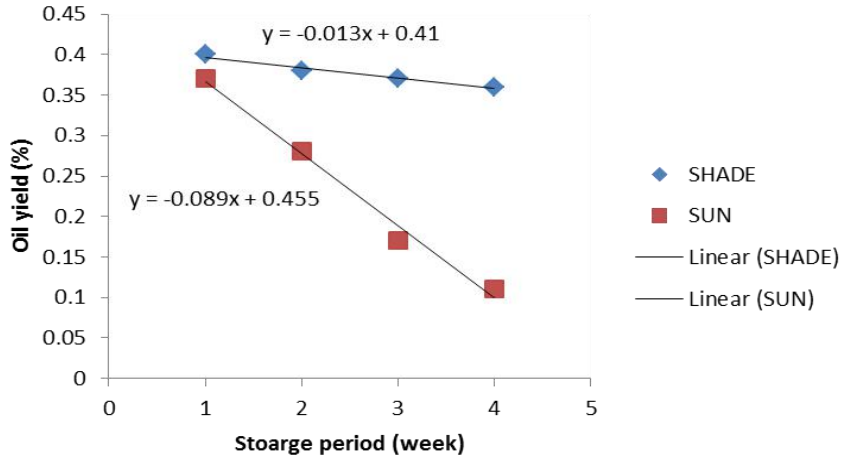

Figure 8: Linearization of effect of storage period and condition on oil yield

Linearization of Figure 7 resulted in Figure 8 and two model equations for prediction of oil yield as a function of time of storage of the leaves for shade and sun as shown in Equations (2) and (3) respectively.

$$Y = -0.013X + 0.41 \quad (2)$$

$$Y = -0.089X + 0.455 \quad (3)$$

In the above equations, Y represents oil yield and X storage period. For instance, if X=5 in Equation (2), then Y= -0.031(5) + 0.41 = 0.345 or 34.5%. Similarly, for X=5 in Equation (3), then Y= -0.089(5) + 0.455 =0.01 or 1%. In other words, when the leaves are kept under both shade and sun for a period of 5 weeks, the expected oil yield would be 34.5% and 1% respectively.

3.3 Physiochemical properties of the Essential oil

The physiochemical properties of the extracted essential oil presented in Table 2 and 3 for both sun and shade storage shows a very good correlation with the literature values as well as the oil extracted from the fresh leaves. These results clearly indicate that the properties of the oil were not significantly affected by the time of storage (1-4 weeks) considered in this study and storage condition (sun and shade).

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Table 2: Effect of storage period on physiochemical properties of the oil stored in the sun

Parameter	Storage period (week)					
	Fresh	1	2	3	4	Literature value
Colour	Pale yellow	Light brown	Light brown	Light brown	Light brown	Pale yellow
Appearance	Liquid	Liquid	Liquid	Liquid	Liquid	Liquid
Odour	Aromatic	Aromatic	Aromatic	Aromatic	Aromatic	Aromatic
Taste	Spicy	Spicy	Spicy	Spicy	Spicy	Spicy
Refractive index @25°C	1.4520	1.4519	1.4535	1.4565	1.4539	1.4511-1.4570
Specific gravity@25°C	0.88	0.91	0.89	0.94	0.92	0.89-0.93
Solubility (in 70% ethanol)	1:3	1:4	1:5	1:4	1:3	1:3-1:5
Ester value	15	17	16	16	14	12-60

Table 3: Effect of storage period on physiochemical properties of the oil stored in the shade

Parameter	Storage period (week)					
	Fresh	1	2	3	4	Literature value
Colour	Pale yellow	Light brown	Light brown	Light brown	Light brown	Pale yellow
Appearance	Liquid	Liquid	Liquid	Liquid	Liquid	Liquid
Odour	Aromatic	Aromatic	Aromatic	Aromatic	Aromatic	Aromatic
Taste	Spicy	Spicy	Spicy	Spicy	Spicy	Spicy
Refractive index @25°C	1.4525	1.4529	1.4553	1.4555	1.4540	1.4511-1.4570
Specific gravity@25°C	0.90	0.92	0.88	0.93	0.91	0.89-0.93
Solubility (in 70% ethanol)	1:4	1:3	1:4	1:5	1:3	1:3-1:5
Ester value	14	15	16	13	15	12-60

4. Conclusions

The following conclusions were made from this study.

1. The pattern of oil extraction was not affected by both the storage period and condition.

2. Storage of the leaves under shade slightly decreased the oil yield as compared with sun storage which significantly decreased the oil yield by about 25% on weekly basis.

3. The best condition to store the leaves is under shade to preserve the oil.

4. Model equations were developed for the prediction of oil yield as a function of time of storage for both sun and shade

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5. The physiochemical properties of the oil do not significantly change with both the condition and storage period.

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