

## COMPARISON OF A-TOCOPHEROL WITH SOME NIGERIAN SPICES AS NATURAL ANTIOXIDANTS IN STORED CRUDE GROUNDNUT OIL

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**ABSTRACT:** *The antioxidant effects of *M. myristica*, *M. tenuifolia* and *A. danielli* in comparison to  $\alpha$ -tocopherol in crude groundnut oil stored at room temperature for 24 weeks were studied. Crude groundnut oil samples mixed with 200 ppm each of the grits (0.5 mm) and n-hexane extracts of the three spices were compared to two controls: untreated crude groundnut oil and crude groundnut oil mixed with 200 ppm  $\alpha$ -tocopherol and all were stored for 24 weeks at room temperature. Samples were tested for free fatty acid and peroxide value after the fourth and eighth week of storage and afterwards, every two weeks, until the 24<sup>th</sup> week. At the 24<sup>th</sup> week, % FFA was highest (3.67 %) in untreated (control 1) samples and lowest in  $\alpha$ -tocopherol-treated (control 2) samples. Again, peroxide value was highest (33.59 meq/kg) in untreated (control 1) samples and lowest in  $\alpha$ -tocopherol-treated samples. Both the spice grits and their extracts significantly reduced peroxide value and % FFA better than the untreated samples, with the spice extracts performing better than their grits. The study suggests that crude groundnut oil can be stored with 200 ppm grits and n-hexane extract of the three spices and better results might be achieved at even higher concentration.*

**KEYWORDS:** groundnut oil, antioxidant, *M. myristica*, *A. danielli*, *M. tenuifolia*

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## INTRODUCTION

Groundnut oil is expressed from the roasted seeds of *Arachis hypogaea* L., commonly known as groundnut, peanut, or earth nut. The economic importance and global trade of the seeds and oil of groundnut have been on the increase since the past century, contributing a significant percentage, 7–10% of the total vegetable oil globally (Kukoyi, *et. al.*, 2014). Nigeria is one of the major growers of groundnut seeds globally and a fourth of the groundnut seeds produced is processed into edible oil (Kukoyi, *et. al.*, 2014). A large percentage, 78% of the fatty acid in groundnut oil is unsaturated, with a large proportion being made up of oleic (18:1) and linoleic (18:2) fatty acids. Its' unsaturated fatty acid portion contributes to a healthy diet as it has been shown to contribute to decreased serum cholesterol (Kris-Etherton, *et. al.*, 2001). The sterols in groundnut oil inhibits cancer growth (Awad, *et. al.* 2000) and may offer protection from colon, prostate and breast cancer. Groundnut oil makes a good frying oil due to its' high smoke point of 229.4°C with little off-flavor development (Sander, 2002). It also finds various food applications as raw material in the production of spread, shortenings, salad dressings, hydrogenated fat, flavor compound bases,

confections, sauces, etc (Sander, 2002; Young, 1996). It is applied industrially as a major raw material in the production of paint, lubricants, insecticides, leather and furniture polish, etc (Kukoyi, *et. al.*, 2014). Groundnut oil is however, a source of allergen to people sensitive to it. However, refining process can remove most or all of the allergenic proteins (Sander, 2002). Groundnut oil gradually loses its' nutritional, physical and chemical attributes as well as its' economic value during storage. Room temperature storage of groundnut oil exposes it to oxygen and high temperature which accelerates the rate of hydrolytic and oxidative rancidity unlike those stored under cold conditions. Room temperature storage of groundnut oil is a common practice in Nigeria. Oxidation is an important determinant of groundnut oil rancidity and it can be delayed or prevented with the use of antioxidants.

Natural antioxidants are preferred over synthetic antioxidants as they are easily assimilated and are additional sources of nutrients and bioactive components in the diet (Embuscado, 2015). The exploration of spices as natural antioxidant has been on the increase. Spices are primarily used to add flavor and aroma to foods (Adegoke and Falade, 2016), but have been discovered to have hypoglycaemic, hypolipidaemic, antibacterial and antioxidant effects (Jaiyeoba, *et. al.*, 2017; Lawal, *et. al.*, 2017; Dauda and Adegoke, 2014) among others.

The *M. myristica* and *M. tenuifolia* plants belong to the same family and genus, Annonaceae and *Monodora*. *M. tenuifolia* plant is a hardy tree of about 10 to 17 m height that grow in the tropical rainforest including Nigeria. Both *M. myristica* and *M. tenuifolia* have the same English name, African nutmeg, but both are differentiated locally with *M. myristica* known locally as *ehuru*, *ariwo* and *awerewa* by the Igbo, Hausa and Yoruba speaking tribes of Nigeria. *M. tenuifolia* is locally identified as *Ehuru ohia*, *ehinawosin* and *Uyenghen* by the Igbo, Yoruba and Ijaw tribes of Nigeria (Njoku, *et. al.*, 2012; Ekeanyanwu and Njoku, 2013). The fruits are edible and have aromatic, oblong- and ovoid-shaped seeds which yield edible oil and also used as spice. While the seeds of *M. myristica* are slightly bigger and about 2cm long and 1.5cm wide with a seed coat that is smooth and thick when dried, *M. tenuifolia* seeds are narrower and about 2.5cm long and 1.5 cm wide with thin seed coats that appear rough when dried Burkill (1985). Fifty-three phenolics that were linked to the antioxidant effectiveness of the two seeds have been identified in both spices, with *M. myristica* having higher total phenolic content than *M. tenuifolia* (Akinwunmi and Oyedapo, 2014; Tchobo, *et. al.*, 2014). *A. danielli*, common name, African cardamom, genus, *Aframomum* and family, Zingiberaceae is a tree of about 3 -4 m height. It is known in Igbo speaking tribe of Nigeria as *urima*. It grows concealed beneath the covers of huge tropical rainforest trees with green flower and flask-shaped fruits having seeds that are aromatic, olive-brown coloured and about 3mm in diameter (Adegoke, *et. al.*, 2016). The major constituents of the seed oil of *A. danielli* are 1, 8-cineole (59.8%), Eugenol (51.1%),  $\alpha$ - pinene (14.1-15.2 %),  $\beta$ -pinene (13.2%) and  $\alpha$ -terpineol (9.3%) (Olusunde, *et. al.*, 1998; Adegoke and Krishna, 1998). The antibacterial and antioxidant properties of *A. danielli* have been studied in various food systems (Lawal, *et. al.*, 2017; Adefegha, *et. al.*, 2017).

Several studies have been done on the stability of stored groundnut oil using various natural antioxidants. However, studies on the storage of groundnut oil using *Monodora myristica* (African nutmeg, *ehuru*), *Aframomum danielli* (*Urima*), *Monodora tenuifolia* (*Efi*) which are underutilized but proven to have very good antioxidant potentials are yet to be done. This study investigated the storage quality preservation potentials of *Monodora myristica* (African nutmeg, *ehuru*), *Aframomum danielli* (*Urima*), *Monodora tenuifolia* (*Efi*) in crude groundnut oil stored at room temperature for a period of six months.

## MATERIALS AND METHODS

### Materials

This research was carried out between July, 2015 and February, 2016. The spices used in the study, *Monodora myristica* (African nutmeg, *ehuru*), *Aframomum danielli* (*Urima*), *Monodora tenuifolia* (*Efi*) and fresh groundnut seeds were purchased in their dried forms without any pest infestation or damage from the local retailers at a local market, Eke Onunwa in Owerri, Imo State, Nigeria. The  $\alpha$ -tocopherol (Vitamin E) (1000 IU) capsules were bought from a pharmacy shop. All reagents used were of analytical grade. Authentication of the spices, *Monodora myristica*, *Aframomum danielli* and *Monodora tenuifolia* (Plate 1) was done at the Taxonomy Unit of the Department of Botany, University of Nigeria, Nsukka.

### Sample Preparation

#### Preparation of Crude Groundnut Oil

Shelled and cleaned raw groundnut seeds (2 kg) were spread in a single layer on a baking sheet and toasted in a preheated oven at 177<sup>0</sup>C for 25 minutes or until the nuts turned golden brown. The nuts were stirred every 10 minutes to avoid scorching and ensure effective toasting. The nuts were cooled, skinned and milled to yield groundnut paste. The paste was mixed with 2 litres of water and boiled at 100<sup>0</sup>C for 30 minutes. The oil that rose to the surface was skimmed off, boiled at 100<sup>0</sup>C for 5 minutes to remove residual moisture, cooled and filtered to remove impurities. The groundnut oil was packed in glass container and stored in a cool place until it was used.

#### Preparation of spice grits.

A hundred grams, each of *M. myristica*, *A. danielli* and *M. tenuifolia* seeds were each sterilized with 50 mls methanol, air-dried for 30 minutes to remove residual methanol and then roasted in a preheated oven at 177<sup>0</sup>C for 5 minutes and dehulled to remove the seed coats. *A. danielli* having thin seed coat did not require roasting and dehulling. The prepared seeds were each milled in a hammer mill and passed through a No 32-mesh sieve to yield their respective grits of 0.5mm particle size. The spice grits were appropriately packaged in covered plastic containers, labeled and stored at 4<sup>0</sup>C until used.

### Preparation of Spice extracts.

The spices were ground using a laboratory mill and sieved through a fine mesh to produce the powder. Extraction was done using n-hexane (Viridi, *et al*, 2003). About 20g each of the powder of *M. myristica*, *A. danielli* and *M. tenuifolia*, were respectively placed in the soxhlet extractor unit. The extraction with n-hexane was carried out at 40°C for 12 hours. The solvent with extract was filtered with Watman no.1 filter paper and centrifuged for 5 min at 5,000 rpm. To purify the extracts, the extraction solvent was evaporated using rotary evaporator at 40°C. The solvent-free extracts were packaged in sample vials, labeled and stored at 4°C until use.

### Experimental Design

The study was fitted into a 2 spice forms × 3 spice types design. The crude groundnut oil was divided into eight portions with each sample containing 100g of crude groundnut oil. Sample labeled GO served as the negative control, being untreated crude groundnut oil. Sample labeled GVE was the positive control contained 0.02g (200ppm) alpha-tocopherol (Vitamin E) [34]. Samples labeled PMMg, PADg and PMTg contained 0.02g of the grits of *Monodora myristica*, *Aframomum danielli* and *Monodora tenuifolia* respectively. Samples labeled PMMe, PADe and PMTe contained 0.02g of the n-hexane extracts of *Monodora myristica*, *Aframomum danielli* and *Monodora tenuifolia* respectively. Each mixture was prepared in duplicates. All the samples were kept in the carefully labeled and covered plastic bottles. Initial FFA and Peroxide values of the samples were determined before storage at 28°C. Also measured were moisture content, relative density, saponification number and iodine value. The crude groundnut oil samples were tested for rancidity after the fourth and eighth week of storage and afterwards, every two weeks, until the twenty fourth week using free fatty acid (FFA) and peroxide value (PV) indicators.



**Plate 1:** A= *Monodora tenuifolia*; B = *Monodora myristica*; C = *Aframomum danielli*

### **Relative Density Determination**

Relative density of the crude groundnut oil samples was determined (A.O.A.C., 2002). The relative density bottle ( $MW_1$ ) was filled with distilled water and gently covered with the lid, the outside walls of the bottle was cleaned and weighed ( $MW_2$ ). The same procedure was conducted for the oil and relative density calculated thus:

$$\text{Relative density} = \frac{MO_1 - MO_2}{MW_2 - MW_1}$$

Where  $MO_1$  = Mass of relative density bottle used for oil;  $MO_2$  = Mass of relative density bottle and oil;  $MW_1$  = Mass of relative density bottle used for water;  $MW_2$  = Mass of relative density and water.

### **Moisture Content Determination**

Moisture content value of the crude groundnut oil samples was determined using 5g of the sample (A.O.A.C, 2002). Moisture content was calculated as:

$$\% \text{ Moisture content} = \frac{W_3 - W_2}{W_2 - W_1} \times 100$$

Where,  $W_1$  = Weight of porcelain dish;  $W_2$  = Weight of empty porcelain dish + sample before drying;  $W_3$  = Weight of porcelain dish + constant weight of dried sample.

### **Iodine Value Determination**

Iodine value of the crude groundnut oil samples were determined (A.O.C.S, 2004). A weighed sample of the oil, 0.5 g was put into a 500 ml stoppered conical flask, dissolved in 10 ml chloroform and 25 ml of Wij's iodine solution was added. A conical flask sealed by moistening the stopper with a little of 10% solution of potassium iodide was kept in the dark for 30 minutes and afterwards, 10 ml 15% potassium iodide and 100 ml water were added. The mixture was titrated with 0.1N Sodium thiosulphate; using 1-2ml of 1% starch solution as indicator until the blue-black colour was completely discharged. A blank determination was also conducted without the oil. Iodine value was calculated as below:

$$\text{Iodine Value, g I}_2/100\text{g} = \frac{(X-Y) \times N \times 12.69}{W}$$

Where X= Volume in ml of approximately 0.1N thiosulphate solution required for the blank, Y= Volume of ml of approximately 0.1N thiosulphate solution required for the test sample, W=Weight in grammes of sample, N= normality of thiosulphate solution, 12.69 = molecular weight of Iodine.

### **Saponification value Determination**

Saponification value of the crude groundnut oil samples were determined (A.O.C.S, 2004). Weighed quantity, 2g of the groundnut oil sample was added into a 300ml flask. Twenty-five mls (25ml) of 0.5N alcoholic Potassium hydroxide solution was added into the flask and heated to become colourless. The mixture was cooled and titrated with 0.5N HCl until the pink color just

disappeared using 0.5ml of 1% phenolphthalein solution as indicator. The titre value was recorded as X. A blank test was also determined.

$$\text{Saponification value (mg KOH per g of sample)} = \frac{X - Y \times N \times 56.1}{W}$$

Where: X= ml, of approximately 0.5N HCl in test sample (ml); Y= ml. of approximately 0.5N HCl in blank (ml); N=normality of acid (mmol/ml); 56.1 =Molecular weight of KOH (mg/mmol) and W= weight of sample in g.

### Free Fatty Acid Determination

Free fatty acid values of the crude groundnut oils were determined (A.O.C.S, 2004). Five (5g) of crude groundnut oil was weighed into a 250 ml Erlenmeyer flask, dissolved in 50ml of 95% ethanol and brought to boiling point. While it was still over 70<sup>0</sup>C, it was neutralized with 0.1N potassium hydroxide (KOH) and titrated with 0.5 N ethanolic. The end point was noted when the addition of a single drop of 0.1N KOH produced a permanent pink which persisted for at least 15sec.

$$\% \text{ Free fatty acid} = \frac{25.6 \times a \times N}{P}$$

Where: a = number of ml of the ethanolic KOH solution used (ml); N = exact normality of the ethanolic KOH solution used (mol/1000ml); p = weight of sample in g; 28.2= molecular weight of oleic acid (g/mol).

### Peroxide Value Determination

Peroxide value determination was carried out on the crude groundnut oil samples (A.O.C.S, 2004). Two (2g) of crude groundnut oil was weighed into a 250 ml glass-stoppered Erlenmeyer flasks, 30 ml of acetic acid-chloroform solution (2 volume glacial acetic acid and 1 volume chloroform) was added and the solution was swirled to allow dissolution. 1g of potassium iodide was added. The solution was left to stand for a minute with occasional shaking and 30 ml of distilled water was added. The solution was slowly titrated with 0.01 N sodium thiosulfate solution and 0.5ml of 1% starch indicator, with vigorous shaking until the yellow color just disappeared. The volume of the titrant used was noted. A blank determination without the oil was carried out and the volume used was noted. Iodine value was calculated as:

$$\text{Peroxide Value (mEq/kg of sample)} = \frac{T \times N \times 1000}{W}$$

Where: T = titre (ml), N= Normality of Na<sub>2</sub>S<sub>2</sub>O<sub>3</sub> (mEq/ml), W = Weight of sample (g); 1000 = Conversion of units (g/kg).

### Statistical Analysis

All determinations carried out were done in duplicates. Data generated were subjected to statistical analysis using SPSS 20 statistical software package. Analysis of variance (ANOVA) procedure

was used to analyze the data statistically. Means were separated using the Fisher's Least Significant Difference.

## RESULTS AND DISCUSSIONS

### Physicochemical characteristics of Crude Groundnut Oil before Storage

The peroxide value of the fresh crude groundnut oil sample was 0.49mEq/kg. Codex recommends an upper limit of 15 mEq/kg for peroxide value in crude edible oil beyond which oxidative changes become obvious (Codex, 2003). Peroxide value of oil sample indicates the quantity of peroxides formed as intermediate or final products of oxidation. The result is below the peroxide value of freshly processed palm oil of 2.6mEq/kg reported by Ngassappa and Othman, 2001.

The free fatty acid value of the freshly expressed crude groundnut oil sample was 0.68%. FFA is an index of edible oil quality determined by the quality of raw material and processing (Mehmood, *et. al.*, 2012). The FFA of the freshly processed groundnut oil was within the Codex quality standard of 3.3% for vegetable oils (Codex, 2013) and lower than the results (2.2-2.5%) reported by Almeida, *et. al.* 2013. This could be as a result of proper raw material handling and high processing quality.

The iodine value of the freshly expressed crude groundnut oil sample was 80.97gI<sub>2</sub>/100g. Iodine value is the percent by weight of molecular iodine absorbed by oil. It is a standard for determining the degree of saturation and susceptibility to oxidation of an oil sample. The value increases as unsaturation increases (Dayritt, *et. al.*, 2007). The range of iodine value obtained was slightly lower than the value, 86-107gI<sub>2</sub>/100g recommended by the Food and Agriculture Organization for groundnut oil (Codex, 2013). The range of iodine value recommended for palm kernel and palm oil, 14.1-21gI<sub>2</sub>/100g and 50-55gI<sub>2</sub>/100g respectively are higher than that of groundnut oil, 86-107gI<sub>2</sub>/100g, signifying its high unsaturation level (Codex, 2003).

**Table 1: Physicochemical properties of the fresh crude groundnut oil before treatment and storage**

| Physicochemical Properties            | Crude Groundnut oil sample |
|---------------------------------------|----------------------------|
| Free fatty acid value(%)              | 0.68±0.113                 |
| Peroxide value(mEq/kg)                | 0.49±0.04                  |
| Iodine value (g I <sub>2</sub> /100g) | 80.97±0.0014               |
| Moisture content (%)                  | 0.024±0.70                 |
| Relative density                      | 0.911±0.00                 |
| Saponification number (mgKOH/g)       | 14.03±0.23                 |

The moisture content of the crude groundnut oil just after extraction was 0.024%. The standard Organization of Nigeria SON (2000) recommends a moisture content of not more than 0.29% in vegetable oils. Moisture content determines product quality. High moisture content in vegetable oil increases hydrolysis, higher FFA and hydrolytic rancidity (Dayritt, *et. al.*, 2007). Ngando, *et. al.*, (2011) found a positive correlation between moisture content and free fatty acid.

The result in Table 4.1 also shows that the saponification value of groundnut oil was 14.03 mgKOH/g. Saponification value was reported by Abayeh, *et. al.* (1998) to be inversely related to the average molecular weight of the fatty acids in the oil fractions. Oil fractions with saponification values of 200 mgKOH/g and above had been reported to possess low molecular weight fatty acids. The relative density of groundnut oil was 0.911. These were closely related to the Codex standard (Codex, 2003) range of 0.912-0.920, 0.891-0.899 and 0.899-0.916 for groundnut oil,

### **Comparative antioxidant effect of *M. tenuifolia*, *A. danielli*, *M. myristica* and $\alpha$ -tocopherol on the free fatty acid value of stored crude groundnut oil**

In Fig. 1, the antioxidant effectiveness of 200 ppm *M. tenuifolia* in comparison to  $\alpha$ -tocopherol in reducing free fatty acid content of the groundnut oil samples were as follows:  $\alpha$ -tocopherol (2.04%) > *M. tenuifolia* n-hexane extract (2.21%) > *M. tenuifolia* grits (2.31%) > untreated crude groundnut oil (3.67 %). Only the untreated groundnut oil had FFA values above the Codex recommended limit of 3.3% in edible oil. The antimicrobial and antioxidant action of *M. tenuifolia* have been shown by Njoku, *et. al.*, (2012) and Ajayi, *et. al.* (2015). Antioxidants are important in oil to delay the onset of the oxidation induction stage or stop the action of the free radicals present in the oil.

In Fig 2, the antioxidant effectiveness of 200 ppm *A. danielli* in comparison to  $\alpha$ -tocopherol in reducing free fatty acid of room temperature stored groundnut oil after 24 weeks was in the order:  $\alpha$ -tocopherol (2.04%) > *A. danielli* n-hexane extract (2.96 %) > *A. danielli* grits (3.44 %) > untreated crude groundnut oil (3.67 %). Only the untreated groundnut oil and the samples treated with *A. danielli* grits had FFA value above the upper limit of 3.3% for FFA in vegetable oil set by Codex (2003). Although, *A. danielli* grits and n-hexane extracts reduced the FFA of the samples better than in untreated groundnut oil samples with significant difference,  $\alpha$ -tocopherol had better performance. This is contrary to the study by Adegoke and Krishna (1998) where 200ppm *A. danielli* methanol and diethyl extracts had better antioxidative properties than butylated hydroxyl toluene and  $\alpha$ -tocopherol in peanut oil under 30 days accelerated storage at 65°C. Different solvents have varied bioactive extraction effectiveness in different medium as a result of their different polarity. N-hexane has a relative polarity of 0.009 as compared to methanol with a relative polarity 0,762 (Reichardt and Welton, 2010). The antioxidant effectiveness of *A. danielli* powder was shown in the preservation of sensory and physicochemical attributes of soymilk stored for 6 months (Dauda and Adegoke, 2014) and in the reduction of microbial count in groundnut paste stored for 20 days (Bello, *et. al.*, 2020).

Fig. 3 shows that 200 ppm *M. myristica* in comparison to  $\alpha$ -tocopherol performed in the following order in room-temperature stored groundnut oil after 24 weeks:  $\alpha$ -tocopherol (2.04%) > *M. myristica* n-hexane extract (2.22 %) > *M. myristica* grits (2.49%) > untreated crude groundnut oil (3.67%). Only the untreated groundnut oil had FFA value above the recommended upper limit of 3.3% for FFA in vegetable oil set by Codex (2003). A study (Eze-Steven, *et. al.*, 2013) demonstrated the ability of *M. myristica* to reduce TBA and acid values of palm oil stored in room temperature for three weeks.



Fig. 4 which compared the performance of 200 ppm each of the three spices with  $\alpha$ -tocopherol in reducing the FFA value of crude groundnut oil after 24 weeks storage had the following order:  $\alpha$ -tocopherol (2.04%) > *M. tenuifolia* extract (2.21%) > *M. myristica* extract (2.22%) > *M. tenuifolia* grits (2.31%) > *M. myristica* grits (2.49%) > *A. danielli* extract (2.96%) > *A. danielli* grits (3.44 %) > untreated crude groundnut oil (3.67%). Only the untreated groundnut oil and groundnut oil treated with *A. danielli* grits had FFA values above the recommended limit of 3.3%. These results are consistent with those obtained by Omer and Abu, (2007) in traditionally produced groundnut oil after three months of storage and also those obtained by Bamidele and Adebayor, (2013).

From the curve in Figure 4, untreated groundnut oil had the shortest induction time at the 8<sup>th</sup> week which is time taken for the oil to resist oxidation indicated by a sharp rising curve, while the oil samples treated with spices and  $\alpha$ -tocopherol showed almost parallel curves, indicating higher resistance to oxidation (Sahin, 2019). High FFA value above the recommended limit indicates oil rancidity. Though the lipases in fresh oil seeds are inactivated by heat during processing, FFA builds up in stored lipids due to auto catalyzed hydrolysis by the residual lipases and the action of excess moisture. Vegetable oil with high degree of unsaturation, like groundnut oil (Ahmed and Young, 1982) are easily prone to rancidity (Kim and Min, 2008), however, the low residual moisture content of the groundnut oil as shown in Table 1 might have resulted to the low FFA rise in the stored samples.

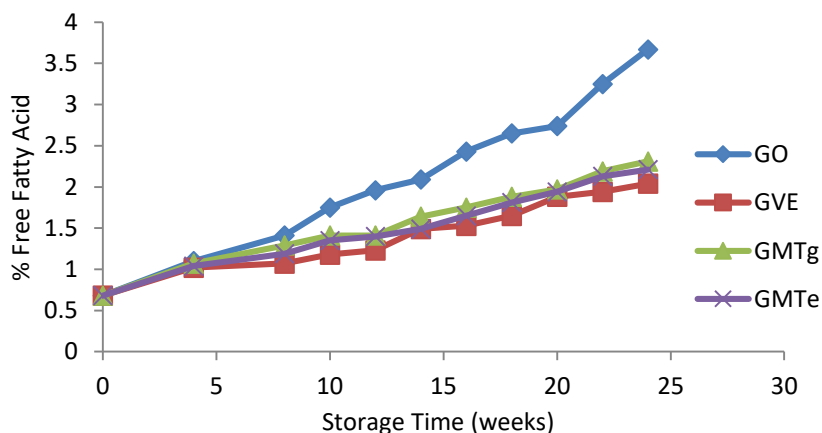


Fig 1: Comparison of the antioxidant effect of the grits and extracts of *M. tenuifolia* with  $\alpha$ -tocopherol on the % FFA of crude groundnut oil stored for 24 weeks at room temperature  
 GO =Control 1(Untreated crude groundnut oil); GVE=Control 2(Crude groundnut oil and  $\alpha$ -tocopherol); GMTg = Crude groundnut oil + *M. tenuifolia* grit; GMTe= Crude groundnut oil + *M. tenuifolia* extract;

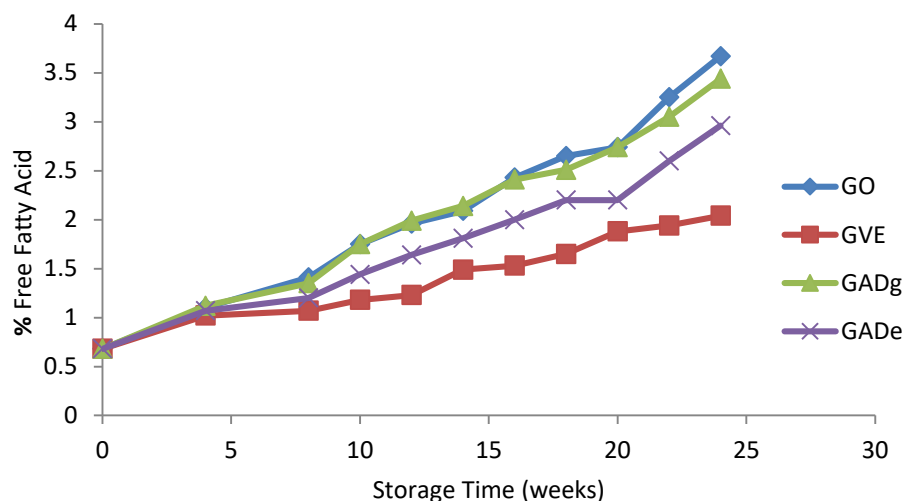


Fig 2: Comparison of the antioxidant effect of the grits and extracts of *A. danielli* with  $\alpha$ -tocopherol on the % FFA of crude groundnut oil stored for 24 weeks at room temperature

GO =Control 1(Untreated crude groundnut oil); GVE=Control 2(Crude groundnut oil and  $\alpha$ -tocopherol); GADg = Crude groundnut oil + *A. danielli* grit; GADe= Crude groundnut oil + *A. danielli* extract;

The spice extracts had the highest reducing effect on FFA of the crude groundnut oil than the spice grits. Extraction ensures maximum availability, synergy and concentration of anti-oxidants from plant materials (Odukoya, *et. al.*, 2005). The spice grits and powder should be used at higher concentrations to achieve maximum antioxidant effectiveness.

### Comparative antioxidant effect of *M. tenuifolia*, *A. danielli*, *M. myristica* and $\alpha$ -tocopherol on the peroxide value of stored crude groundnut oil

In essence the results for peroxide value in Fig. 5 shows that the highest performance at the 24<sup>th</sup> week in comparison to  $\alpha$ -tocopherol followed the order of:  $\alpha$ -tocopherol (16.15meq/kg) > *M. tenuifolia* extract (17.96meq/kg) > *M. tenuifolia* grits (23.39 meq/kg) > Untreated crude groundnut oil (33.59 meq/kg). The untreated groundnut oil samples were the first that had peroxide value (19.10 meq/kg) above the recommended limit of 15meq/kg at the 20<sup>th</sup> week, while  $\alpha$ -tocopherol was the last (16.15 meq/kg) to exceed that limit at the 24<sup>th</sup> week. *M. tenuifolia* extract showed considerable antioxidant effectiveness in stored beef patties (Ugwuona, *et. al.*, 2019). The antioxidant effectiveness of *M. tenuifolia* was also demonstrated in other studies (Njoku, *et. al.*, 2012; Eze-Steven, *et. al.*, 2013).

The result in Fig. 6 shows the effect of 200 ppm of *A. danielli* on the peroxide value of crude groundnut oil stored for 24 weeks at room temperature. The highest performance at the 24<sup>th</sup> week in comparison to  $\alpha$ -tocopherol was as follows:  $\alpha$ -tocopherol (16.15 meq/kg) > *A. danielli* extract (21.24 meq/kg) > *A. danielli* grits (26.22 meq/kg) > Untreated crude groundnut oil (33.59 meq/kg). The peroxide value of groundnut paste during a 20-day storage study was significantly reduced with 0.6% (6000 ppm) *A. danielli* powder (Bello, *et. al.*, 2020). The peroxide values obtained in the study after 24 weeks storage are much lower than those obtained in soya oil stored for 20 days

with *A. danielli* diethyl ether and methanol (Jaiyeoba, *et al.*, 2017). *A. danielli* had better reducing effect at 200ppm than butylated hydroxytoluene (BHT) on the peroxide value of mayonnaise stored for 60 days (Etti, *et al.*, 2012).

The result shown in Fig. 7 illustrates the effect of 200 ppm of *M. myristica* on the peroxide value of crude groundnut oil stored for 24 weeks at room temperature. The highest performance in comparison to  $\alpha$ -tocopherol was as follows:  $\alpha$ -tocopherol (16.15 meq/kg) > *M. myristica* extract (16.41 meq/kg) > *M. myristica* grits (17.33 meq/kg) > Untreated crude groundnut oil (33.59 meq/kg). *M. myristica* had a reducing effect on the acid value of palm oil stored at room temperature for three weeks (Eze-Stevens, *et al.*, 2013). *M. myristica* powder (1000 ppm) and methanolic extract (500 ppm) reduced the peroxide value of accelerated oven stored soya oil for 24 days (Womeni, *et al.*, 2013).

From Fig. 8, 24 weeks, the highest antioxidant effects of 200 ppm of the spices on the peroxide value of the groundnut oil samples in comparison to  $\alpha$ -tocopherol is shown thus:  $\alpha$ -tocopherol (16.15 meq/kg) > *M. myristica* extract (16.41 meq/kg) < *M. myristica* grits (17.33 meq/kg) < *M. tenuifolia* extract (17.96 meq/kg) < *A. danielli* extract (21.24 meq/kg) < *M. tenuifolia* grits (23.39 meq/kg) < *A. danielli* grits (26.22 meq/kg) < Untreated crude groundnut oil (33.59 meq/kg). All the samples had values above the recommended limit for peroxide value in edible oil (Codex, 2013). From the curve in Figure 8, untreated groundnut oil also had the shortest induction time, which is time taken for the oil to resist oxidation indicated by a sharp curve at the 16<sup>th</sup> week, while the oil samples treated with spices and  $\alpha$ -tocopherol showed more flatter curve, indicating higher resistance to oxidation (Sahin, 2019). Compared to all the spices and their forms, *Mondora myristica* extracts yielded the highest antioxidant effects on the peroxide value of groundnut oil samples. The range of peroxide values obtained in the stored groundnut oil samples were close to the values reported by Makeri, *et al.* (2011) in oils of two groundnut varieties stored at room temperature for 9 weeks.

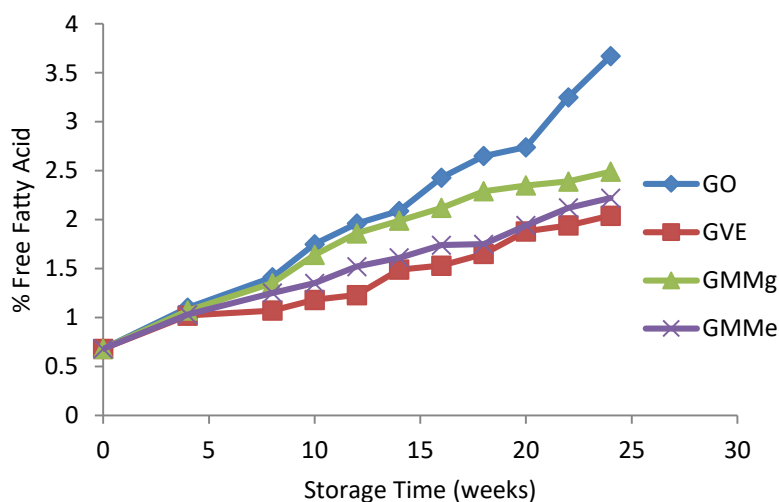


Fig 3: Comparison of the antioxidant effect of the grits and extracts of *M. Myristica* with  $\alpha$ -tocopherol on the % FFA of crude groundnut oil stored for 24 weeks at room temperature

GO =Control 1(Crude Groundnut oil); GVE=Control 2(Crude Groundnut oil and  $\alpha$ -tocopherol); GMMg = Crude Groundnut oil + *M. myristica* grit; GMMe= Crude Groundnut oil + *M. myristica* extract;

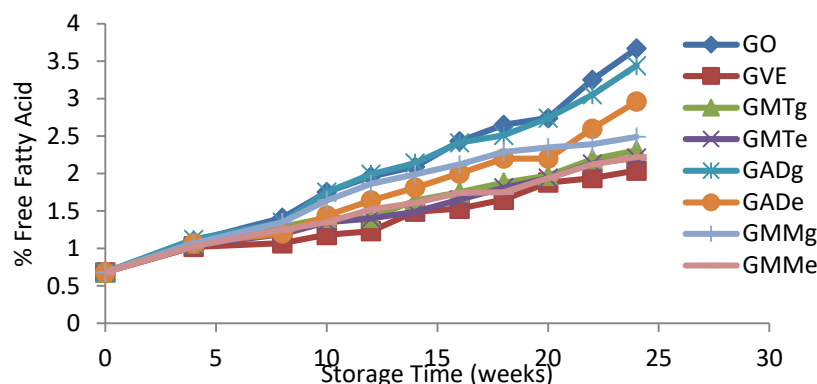


Fig 4: Comparison of the antioxidant effect of the grits and extracts of three Nigerian spices with  $\alpha$ -tocopherol on the % FFA of crude groundnut oil stored for 24 weeks at room temperature

GO =Control 1(Crude Groundnut oil); GVE=Control 2(Crude Groundnut oil and  $\alpha$ -tocopherol); GADg = Crude Groundnut oil + *A. danielli* grit; GADe= Crude Groundnut oil + *A. danielli* extract; GMMg= Crude groundnut oil + *M. myristica* grits; GMMe = Crude groundnut oil + *M. myristica* extract; GMTg= Crude groundnut oil + *M. tenuifolia* grits; GMTe = Crude groundnut oil + *M. tenuifolia* extract;

All the groundnut oil samples were within the Codex quality standard of 15mEq/kg (Codex, 2013) except untreated crude groundnut oil, crude groundnut oil + *M. tenuifolia* grits, crude groundnut oil + *A. danielli* extract and crude groundnut oil + *A. danielli* grits. This could be due to differences in antioxidant efficacy of the spices and high level of unsaturation in groundnut oil. High unsaturation level of groundnut oil as indicated by the iodine value in Table 4.1 makes it prone to oxidative deterioration during storage. Even though the spices at 200ppm all had significant effects in lower peroxide value of groundnut oil, higher antioxidant effect can be obtained at even higher concentration of about 400ppm or more (Makeri, *et. al.*, 2011).

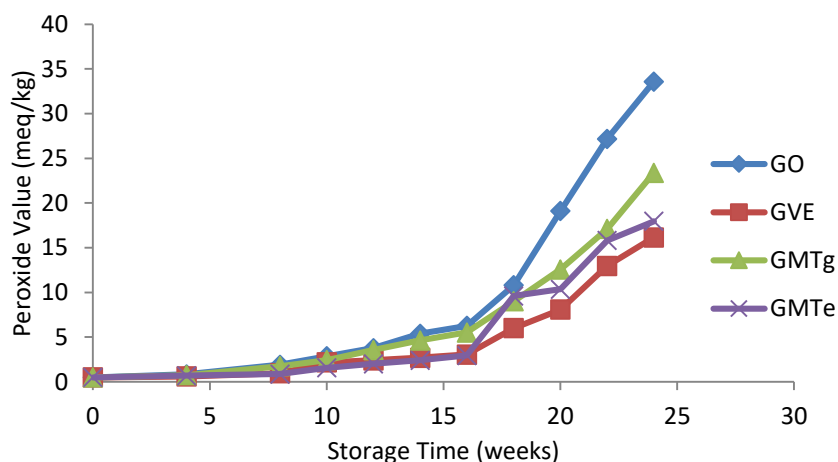


Fig 5: Comparison of the antioxidant effect of the grits and extracts of *M. tenuifolia* with  $\alpha$ -tocopherol on the peroxide value (meq/kg) of crude groundnut oil stored for 24 weeks at room temperature.

GO =Control 1(Crude Groundnut oil); GVE=Control 2(Crude Groundnut oil and  $\alpha$ -tocopherol); GMTg = Crude Groundnut oil + *M. tenuifolia* grit; GMTe= Crude Groundnut oil + *M. tenuifolia* extract;

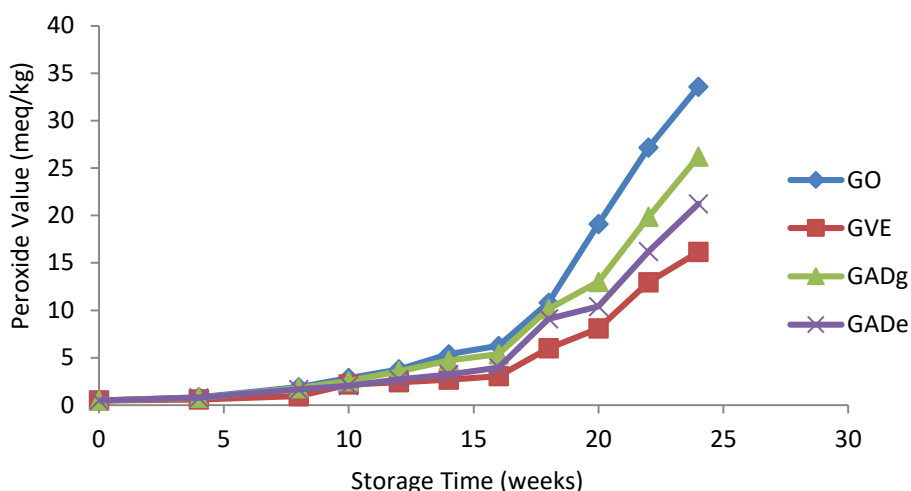


Fig 6: Comparison of the antioxidant effect of the grits and extracts of *A. danielli* with  $\alpha$ -tocopherol on the peroxide value (meq/kg) of crude groundnut oil stored for 24 weeks at room temperature.

GO =Control 1(Crude Groundnut oil); GVE=Control 2(Crude Groundnut oil and  $\alpha$ -tocopherol); GADg = Crude Groundnut oil + *A. danielli* grit; GADe= Crude Groundnut oil + *A. danielli* extract;

## CONCLUSION

*Monodora myristica*, *Monodora tenuifolia* and *Aframomum danielli* at a concentration of 200 ppm, all had antioxidant effects on crude groundnut oil stored at room temperature for 24 weeks. This was shown by their reduction effects on the free fatty acid value and peroxide value of crude groundnut oil during storage with significant differences from the control (untreated crude groundnut oil). The spices grits and n-hexane extracts had antioxidant effects that were comparable to  $\alpha$ -tocopherol (Vitamin E). In some cases, the spices performed better than  $\alpha$ -tocopherol. Solvent extraction increased the antioxidant effect of the spices, as the groundnut oil treated with spice extracts had lower peroxide value and FFA than those treated with the grits. Generally, all the spice forms had antioxidant potentials and can be used to delay the onset of FFA and peroxide development in groundnut oil during storage.

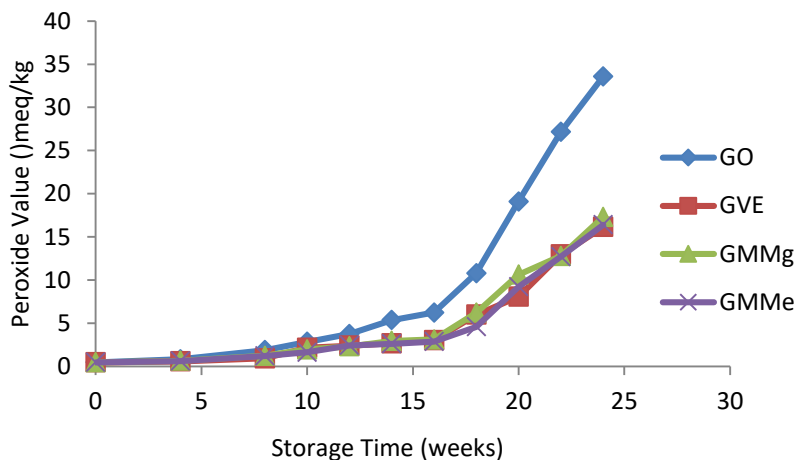


Fig 7: Comparison of the antioxidant effect of the grits and extracts of *M. myristica* with  $\alpha$ -tocopherol on the peroxide value (meq/kg) of crude groundnut oil stored for 24 weeks at room temperature.  
 GO =Control 1(Crude Groundnut oil); GVE=Control 2(Crude Groundnut oil and  $\alpha$ -tocopherol); GMMg = Crude Groundnut oil + *M. myristica* grit; GMMe= Crude Groundnut oil + *M. myristica* extract;

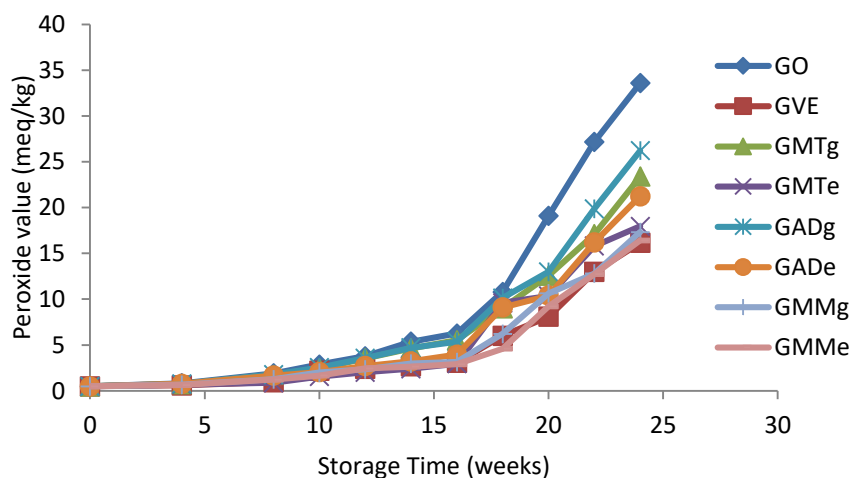


Fig 8: Comparison of the antioxidant effect of the grits and extracts of three Nigerian spices with  $\alpha$ -tocopherol on the peroxide value (meq/kg) of crude groundnut oil stored for 24 weeks at room temperature.  
 GO =Control 1(Crude Groundnut oil); GVE=Control 2(Crude Groundnut oil and  $\alpha$ -tocopherol); GADg = Crude Groundnut oil + *A. danielli* grit; GADe= Crude Groundnut oil + *A. danielli* extract; GMMg= Crude groundnut oil + *M. myristica* grits; GMMe = Crude groundnut oil + *M. myristica* extract; GMTg= Crude groundnut oil + *M. tenuifolia* grits; GMTe = Crude groundnut oil + *M. tenuifolia* extract;

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