ISSN-2230-7346
Journal of Global Trends in Pharmaceutical Sciences

COMPARITATIVE STUDY OF DIFFERENT CRUDE AND REFINED REHEATEDEDIBLE OILS AVAILABLE IN TIRUPATI

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ARTICLE INFO

Key Words

Edible oil, Reheating, toxic compounds, Acid value.

Access this article online
Website:
https://www.jgtps.com/
Quick Response Code:

ABSTRACT

Edible oil is an important constituent in food preparations. Edible oil is used in different types of cooking purposes. Due to its cost the edible oils are reheated. Due to this reheating there is a change in physicochemical properties which can be determined by different parameters like viscosity, density, acid value, saponification value, peroxide value. In our study we had taken different edible oils of both crude and refined oils. Physicochemical changes like darkening of oil, unpleasant smell, viscosity, acid value, saponification value, peroxide value of ground nut oil, sunflower oil, Palm oil of both crude and refined oils are increasing from 1^{st} heating to 6^{th} heating. Physicochemical parameters are increasing because of formation of toxic compounds. Increased number of frying leads to changes in the physical chemical parameters. During repeated frying of oil nutritional value, quality of the oil also decreases. Intake of food items with repeated heated oil leads to several health implications like cancers in (breast, lung, colorectal, prostate, pancreatic cancers), hypertension, diabetes mellitus, atherosclerosis, myocardial infarction, Alzheimer's disease, rheumatoid arthritis etc. It is suggested to better avoid the reheating of edible oil in cooking purposes.

INTRODUCTION

One of the important dietary components in our daily life is vegetable oil. In our daily food consumption edible oil is nearly used in all types of food preparations including frying, baking, sautéing, dressing, marinating, and extrusion cooking. They are obtained from oil seeds (i.e. groundnut oil, sunflower oil and coconut oil), food legumes (i.e. peanut), nuts (i.e. almond) or the soft substances of fruits (i.e. olives) triglycerides serves as main sources of fat and composed of three fatty acids and one molecule of glycerol (1). Edible oils are used in day to day life for our delicious food preparations because they give good taste and flavor to the food items. Edible oil gives energy, essential fatty acids for human nutrition They also act as a carrier for fat soluble vitamins and for fatty acids which are not synthesized by the body (2). Due to good taste, flavor and cheap price deep fried food is widely sold and available. Repeated heated cooking oil produces unwanted hazardous and cytotoxic compounds which cause harmful health problems to the human body (3). In presence of high temperature and oxygen cooking oil undergoes thermal oxidationprocess and generate cytotoxic products and free radicals and targets on the protein, lipids, DNA molecules. In proteins it undergo degeneration and cause cell and tissue injury , in DNA it cause DNA damage and in lipids it will undergo lipid peroxidation to form 4-hydroxy nonenal, aldehydes and other volatile compounds and this will interacts with membrane protein leads to pathological changes at different parts of the body such as Atherosclerosis, Myocardial infraction, Alzheimer disease, Diabetes mellitus, Cancer (4). Due to intake of bulk fried food results in cancerslike colon, rectum, breast, kidney and pancreas(5). Cancer is due to reduction in omega-3 and omega-6 fatty acids (6). Cancer causing agents like Free radicals, aldehydes, alkanes, 4-hydroxy nonenal, Polycyclic aromatic hydrocarbons (PAHs) (benzopyrene, dibenzo anthracene, benzoanthracene, dibenzo anthracene, heterocyclic amide (1,7). Physical changes that takes place during frying are darkening of colour, off flavor, increased viscosity (8). During repeated frying chemical changes like oxidation, hydrolysis, polymerization, isomerization which leads to lipid peroxidation (9). Lipid peroxidation also implicated in the pathogenesis of hypertension and atherosclerosis (10). When the frying time increases it leads to the formation of oxidized polymerized species which deteriorates the quality of oil (11). Fried food absorbs the oxidative products like hydro peroxide and aldehydes which will affect the quality of edible oil. At high frying temperature antioxidant activity decreases (12). We are focused on the changes that takes on reheating of edible oil and health effects occurs with intake of edible oil.

APPARATUS AND INSTRUMENTS REQUIRED

Edible oils of different crude and refined oils were collected from Tirupati. Crude oils were collected from different oils situated in different areas in Tirupati and refined oils were collected from market.

Physicochemical Parameters

These physicochemical parameters namely

- Colour
- Odour
- Density
- Viscosity
- Acid value
- Saponification value
- Peroxide value

These properties are used to assess the quality and functionality of oils.

Colour: On reheating of different edible oils like Sunflower refined, Sunflower crude, Ground nut refined, Ground nut crude, Palm oil refined, Palm oil crude colour changes from Light Yellow to Orange colour.

Odour: On reheating of different edible oils pungent smell occurs.

Density: Densities of oil samples were measured by a Relative Density (R.D) bottle with a capacity of 10 ml according to the following formula

$$Density\ (\rho) = \frac{Mass\ of\ the\ sample\ (M}{Volume\ of\ the\ bottle\ (V)}$$

The uncertainty in density measurements was ±0.0001g/cm³.The density was determined for temperature ranging from 20°c to 50°c, with a degree's step increase. The density of vegetable oils is dependent on their fatty acid composition, minor components and temperature. The difference in the density of oils may be due to the refined and unrefined character of the studied oils. The density of vegetable oils linearly decreases with increase in temperature. Oils with the density of lower values are highly appreciable to consumers. The low density is obtained by more rigid π bonding between C-C bonds and becomes more strenuous. The relative density of oil at any temperature compared to water at a specified temperature is known as the mean molecular weight diminishes andalso the degree of unsaturation increases (2).

Viscosity: The viscosity of oils and their blends was determined by BROOKFIELD DVII + Pro viscometer at a constant shear rate at constant temperatures which were controlled by a microprocessor assisted water bath using spindle S51. Viscosity of oil is a measure of the oil's resistance to shear. High viscosity implies a high resistance to flow while a low viscosity indicates a low resistance to flow. Changes with temperature, decreasing temperature increases viscosity. The rise in temperature enhances movement of molecules and reduces intermolecular forces so the layers of liquid easily passes over one another and thus contribute to reduction in viscosity. Oil viscosity also depends upon molecular structure and decrease with the unsaturation of fatty acid. It may be due to double bonds that make bonding more rigid and rotation between C-C bonds becomes more strenuous (2).

Acid Value

Procedure

- Cooled oil sample is taken into a 250 ml conical flask
- To this add 50 ml to 100 ml of freshly neutralized hot ethyl alcohol and about one ml of phenolphthalein indicator solution was added.
- Then the mixture was boiled for about five minutes and titrated while hot against standard alkali solution was shaken vigorously during the titration.
- Then oil/fat weight was taken for the estimating the strength of the alkali used for titration. So that the volume of alkali required for the titration does not exceed 10 ml.

Calculation

$$ACID\ VALUE = \frac{56.1 x V x N}{W}$$

V = Volume in ml of standard potassium hydroxide or sodium

N = Normality of the potassium hydroxide solution or Sodium hydroxide solution

W = Weight in g of the sample (13, 14, 15)

Saponification Value

Procedure

- The saponification value (SV) is defined as the weight of Potassium Hydroxide in milligrams that need to saponify one gram of oil.
- It is determined by taking 1.0gram of oil sample into a conical flask to which 25ml of 0.5N alcoholic KOH is added and heated under a reserved condenser for 30-40 min to ensure that the sample was fully dissolved.
- After cooling the sample, Phenolphthalein indicator was added and titrated with 0.5NHCl until a pink end point was reached.
- A blank was determined with the same time conditions.

Calculation

$$\text{Saponification value} = \frac{(B - T) x N x 56.1}{W}$$

Where,

T=ml of HCl required by oil sample

N=Normality of HCl

W= Weight of oil in grams (13,16,18)

Peroxide value

- Procedure
- 1g of oil or fat is taken into a clean dry boiling tube and 1g of powdered potassium iodide and
    - 20mL of solvent mixture was added.
- The tube was placed in boiling water then the liquid was boiled for 30

seconds and allowed to boil vigorously for not more than 30 seconds.

- The contents were transferred quickly to a conical flask which contains 20mL of 5% potassium iodide solution.
- The tube was washed twice with 25mL water each time and collected into the conical flask.
- Above solution was titrated against N/500 sodium thiosulphate solution until yellow colour disappears.
- To this 0.5mL of starch solution was added and shaken vigorously and titrated carefully till the blue colour just disappears.
- A blank titration should be done.

Calculation

Peroxide value (milli-equivalent peroxide/kg sample) =

W

Where,

S = mL $Na_2S_2O_3$ (Test-Blank) and

N = normality of $Na_2S_2O_3$ (13, 16, 18)

Table No.1: Colour of Edible Oils

Heating's	Ground nut Oil(crude)	Ground nut oil (refined)	Sunflower oil (crude)	Sunflower oil (Refined)	Palm oil (crude)	Palm oil (Refined)
Non heated	DeepYellow	LightYellow	LightYellow	Colour less	Creamellow	LightYellow
1st heated	DeepYellow	LightYellow	LightYellow	Colour less	Yellow	LightYellow
2nd heated	DeepYellow	LightYellow	LightYellow	Colour less	Yellow	LightYellow
3rd heated	LightOrange	Yellow	LightYellow	Transparent Light Yellow	Yellow	LightYellow
4th heated	Orange	LightOrange	Yellow	Transparent LightYellow	Yellow	Yellow
5th heated	Orange	LightOrange	Yellow	LightYellow	Yellow	LightOrange
6th heated	Orange	Orange	Yellow	LightYellow	Yellow	Orange

Table No.2: Density of Edible Oils

Heating's	Ground nut Oil(crude)	Ground nut oil (refined)	Sunflower oil (crude)	Sunflower oil (Refined)	Palm oil (crude)	Palm oil (Refined)
Nonheated	1.0424 g/ml	1.0252 g /ml	1.0308 g/ml	1.0304 g/ml	1.0284g/ml	1.0128 g/ml
1st heated	1.0484 g/ml	1.4688 g/ml	1.0612 g/ml	1.0108 g/ml	1.0556g/ml	1.0476 g/ml
2nd heated	1.3312 g/ml	1.5488 g/ml	1.0624 g/ml	1.0244 g/ml	1.073 g/ml	1.0668 g/ml
3rd heated	1.426 g/ml	1.5384 g/ml	1.0652 g/ml	1.0372 g/ml	1.08 g/ml	1.07 g/ml
4th heated	1.452 g/ml	1.5348 g/ml	1.0688 g/ml	1.036 g/ml	1.088 g/ml	1.0738 g/ml
5th heated	1.481 g/ml	1.588 g/ml	1.0766 g/ml	1.034 g/ml	1.095 g/ml	1.076 g/ml
6th heated	1.497 g/ml	1.6052 g/ml	1.0856 g/ml	1.039 g/ml	1.099 g/ml	1.077 g/ml

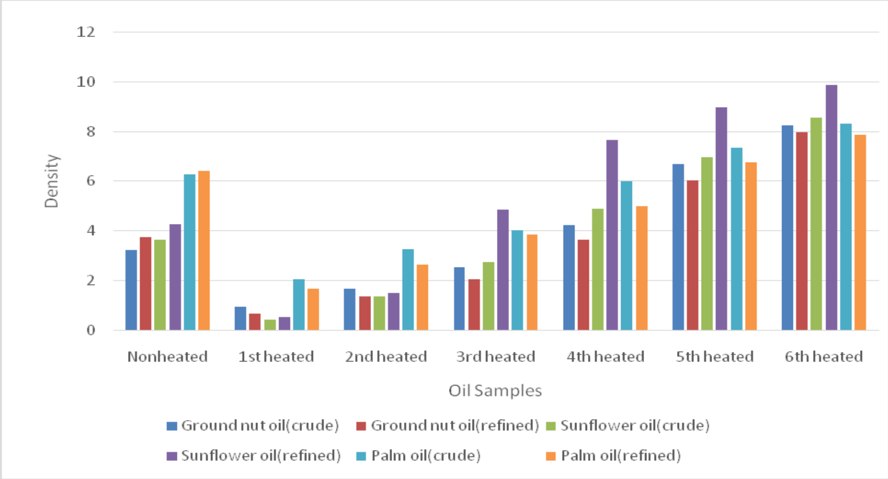

Fig 1: Graph of Density for different edible oils

Table No.3: Viscosity of Edible Oils

Heating	Ground nut Oil(crude)	Ground nut oil (Refined)	Sunflower Oil(crude)	Sunflower oil (Refined)	Palm oil (crude)	Palm oil (Refined)
Non heated	234dyne/s/cm²	212 dyne/s/cm²	182 dyne/s/cm²	175 dyne/s/cm²	442dyne/s/cm²	420 dyne/s/cm²
1st heated	265 dyne/s/cm²	242 dyne/s/cm²	201 dyne/s/cm²	192 dyne/s/cm²	458 dyne/s/cm²	432 dyne/s/cm²
2ndheated	346 dyne/s/cm²	339 dyne/s/cm²	265 dyne/s/cm²	244 dyne/s/cm²	592 dyne/s/cm²	555 dyne/s/cm²
3rd heated	482 dyne/s/cm²	469 dyne/s/cm²	292 dyne/s/cm²	276 dyne/s/cm²	689 dyne/s/cm²	661 dyne/s/cm²
4th heated	600 dyne/s/cm²	592 dyne/s/cm²	392 dyne/s/cm²	388 dyne/s/cm²	784 dyne/s/cm²	743 dyne/s/cm²
5th heated	796 dyne/s/cm²	746 dyne/s/cm²	498 dyne/s/cm²	472 dyne/s/cm²	920 dyne/s/cm²	906 dyne/s/cm²
6th heated	844 dyne/s/cm²	820 dyne/s/cm²	582 dyne/s/cm²	540 dyne/s/cm²	991 dyne/s/cm²	980 dyne/s/cm²

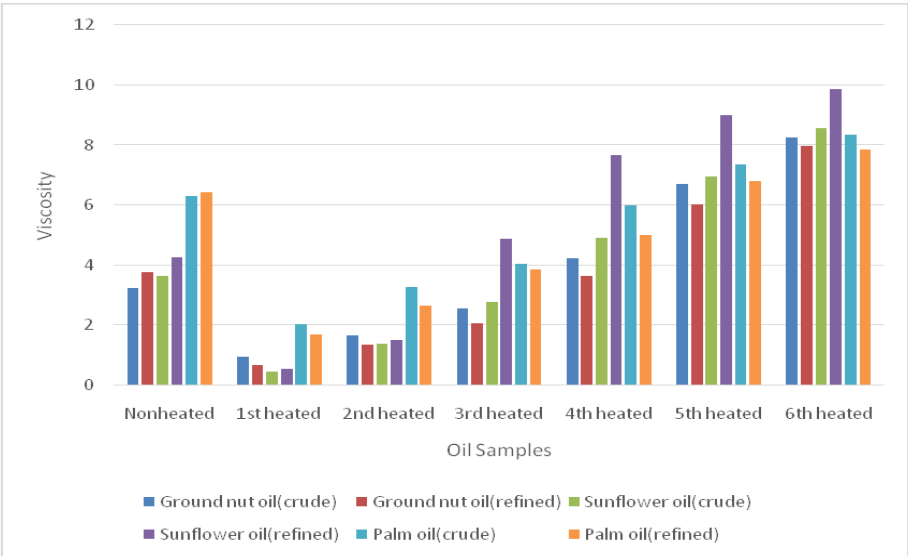

Fig 2: Graph of Viscosity for different edible oils

Table No.4: Acid Value of Edible Oils

Heating's	Ground nut Oil(crude)	Ground nut oil(refined)	Sunflower oil (crude)	Sunfloweroil(Refined)	Palm oil (crude)	Palm oil (Refined)
Non heated	0.61 mg/g	0.91 mg/g	0.96 mg/g	1.88 mg/g	0.96 mg/g	1.68 mg/g
1st heated	11.34 mg/g	1.12 mg/g	0.434 mg/g	2.65 mg/g	2.206 mg/g	1.76 mg/g
2nd heated	11.94 mg/g	20 mg/g	0.518 mg/g	2.34 mg/g	2.44 mg/g	1.79 mg/g
3rd heated	12.50 mg/g	11.29 mg/g	0.603 mg/g	2.45 mg/g	2.93 mg/g	1.86 mg/g
4th heated	12.65 mg/g	1.32 mg/g	0.682 mg/g	2.48 mg/g	2.97 mg/g	1.91 mg/g
5th heated	12.67 mg/g	1.34 mg/g	0.672 mg/g	2.49 mg/g	3.01 mg/g	1.93 mg/g
6th heated	12.69 mg/g	1.36 mg/g	0.687 mg/g	2.51 mg/g	3.05 mg/g	1.97 mg/g

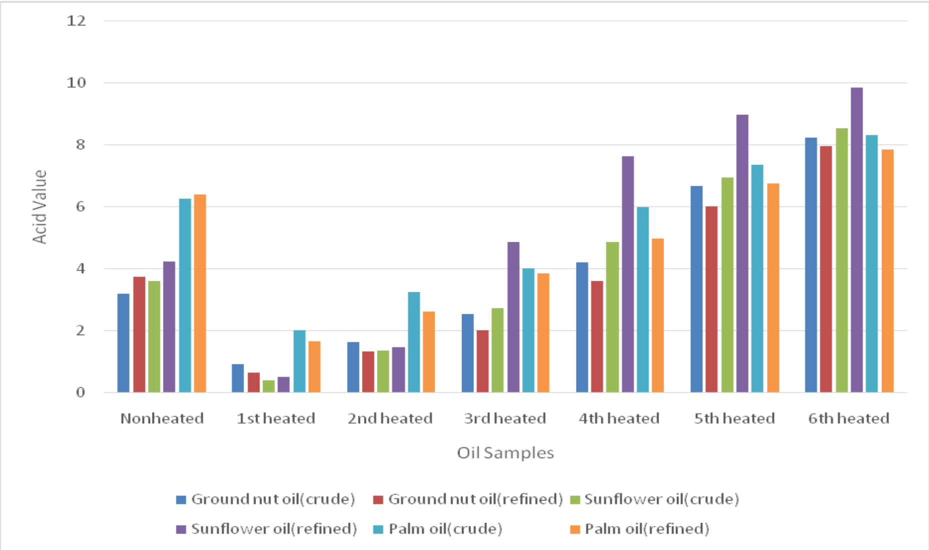

Fig 3: Graph of Acid Value for different Edible Oils

Table No.5: Saponification value of Edible Oils

Heating's	**Ground nut Oil(crude)**	**Ground nut oil (refined)**	**Sunfloweroil (crude)**	**Sunflower oil (Refined)**	**Palm oil (crude)**	**Palm oil (Refined)**
Non heated	241.22 mg/g	187.31 mg/g	63.34 mg/g	72.99 mg/g	137.37 mg/g	129.09 mg/g
1st heated	243.33 mg/g	193.21 mg/g	91.47 mg/g	78.41 mg/g	145.98 mg/g	156.82 mg/g
2nd heated	247.61 mg/g	196.33 mg/g	96.43 mg/g	99.53 mg/g	153.82 mg/g	155.85 mg/g
3rd heated	249.84 mg/g	211.43 mg/g	109.78 mg/g	147.18 mg/g	170.71 mg/g	159.85 mg/g
4th heated	252.44 mg/g	215.71 mg/g	138.74 mg/g	159.85 mg/g	189.79 mg/g	165.88 mg/g
5th heated	254.33 mg/g	220.65 mg/g	187.62 mg/g	232.84 mg/g	194.84 mg/g	171.91 mg/g
6th heated	260.89 mg/g	255.76 mg/g	260.29 mg/g	254.56 mg/g	257.87mg/g	252.41 mg/g

Table No.6: Peroxide value of Edible oils

Heating	Ground nut Oil(crude)	Ground nut Oil(Refined)	Sunflower Oil(Crude)	Sunflower Oil(Refined)	Palm oil (crude)	Palm oil (Refined)
Non heated	3.21 mg/g	3.74 mg/g	3.63 mg/g	4.25 mg/g	6..28 mg/g	6.42 mg/g
1st heated	0.92 mg/g	0.65 mg/g	0.42 mg/g	0.53 mg/g	2.03 mg/g	1.67 mg/g
2nd heated	1.65 mg/g	1.34 mg/g	1.36 mg/g	1.48 mg/g	3.26 mg/g	2.63 mg/g
3rd heated	2.54 mg/g	2.04 mg/g	2.75 mg/g	4.87 mg/g	4.03 mg/g	3.85 mg/g
4th heated	4.23 mg/g	3.63 mg/g	4.89 mg/g	7.65 mg/g	5.99 mg/g	4.98 mg/g
5th heated	6.69 mg/g	6.02 mg/g	6.96 mg/g	8.98 mg/g	7.36 mg/g	6.78 mg/g
6th heated	8.24 mg/g	7.98 mg/g	8.55 mg/g	9.87 mg/g	8.33 mg/g	7.86 mg/g

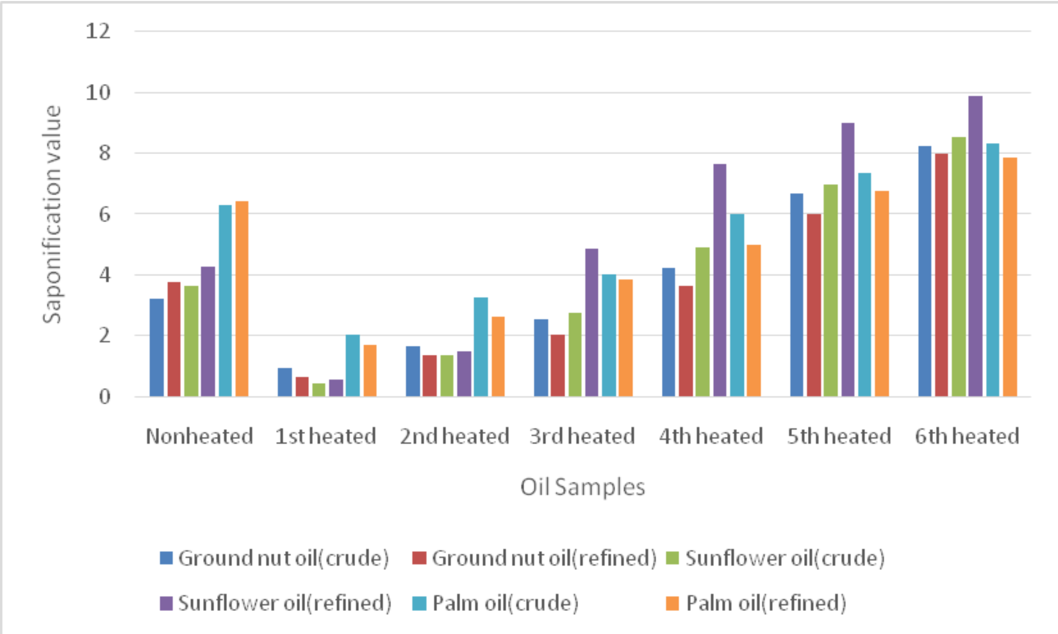

Fig 4: Graph of Saponification Value for different edible oils

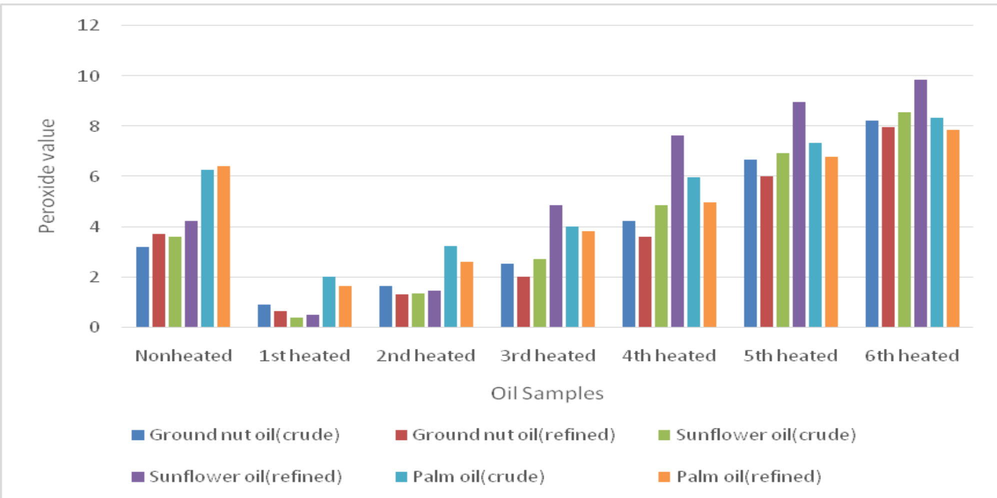

Fig 5: Graph of Peroxide Value for different edible oils

DISCUSSION

Colour: On reheating of different edible oils like Sunflower refined and crude, Groundnut refined and crude, Palm oil refined and crude colour changes from Light Yellow to Orange colour. Whenrepeated reheating is done leads in color change and formation of toxic compounds like polycyclic aromatic hydrocarbonsPAH's (benzopyrene, dibenzoanthracene, benzoanthracene), hetrocyclic amides, 4-hydroxytrans-2-nonenal(HNE) epoxides, polar dimmer leads to increase in viscosity.

Density: Density is one of the important physical parameters used to assess the quality of edible oils. The results for the densities of the different edible oil are shown in Table no1. The fig no -1 shows the comparison between reheating of crude and refined of different edible oil samples. The results tabulated are done at room temperature in which crude oils have shown more density compared to refined oils. 1st heated sunflower refined oil has shown least density (1.0108 g/ml) whereas 6th heated groundnut refined oil has shown more density (1.6052g/ml). Whereas in case of oil with lower density like first heating of sunflower refined oil are highly appreciable to the customers. Density is more for 6th heated groundnut refined oil because it undergoes hydrogenation process for the free fatty acids i.e., while heating bonds will break to satisfy the valency of hydrogens which leads to increase in weight. The graphical representation of densities also been presented in graph no 1. The graph also shows that there is no significant increase in the non-heated edible oils whereas we can find the steep height in the density of the ground nut oil(refined).later in the second heating the ground nut (refined) and palm oil crude have shown the steep increase in the graph compared to the other edible oils used.

Viscosity

Viscosity one of the important physical parameter used to assess toxic substances formed during reheating of oils. The result of various viscosities has been shown in Table 3. The comparison between the different oils (heated and reheated) have been discussed in Fig 2. When compare reheated oils of refined and crude oils viscosity values is more for crude edible oils than refined oils like 6th heated groundnut crude(844 dyne/s/cm²) has more value than 6th heated ground nut refined(820 dyne/s/cm²)and the 6th heated sunflower oil crude(582 dyne/s/cm²) value is higher than6th heated sunflower oil refined(540 dyne/s/cm²)and 6th heated palm oil crude(991 dyne/s/cm²) value is higher than 6th heated palm oil refined(982dyne/s/cm²) .when we compare both nonheated and reheating of different edible oils of crude and refined highest viscosity value is for 6th heated palm oil crude(991dyne/s/cm²) and least is for fresh sunflower oil refined(175 dyne/s/cm²). Viscosity of oil increases withincreased number of fryings. Viscosity is more for 6th heated palm oil crude(991 dyne/s/cm²) because when repeated reheating is done leads changes in colourand formation of toxic compounds like polycyclic aromatic hydrocarbons. PAHs(benzopyrene, dibenzoanthracene, benzoanthracene), Heterocyclic amides, 4hydroxy-trans-2-nonenal(HNE) epoxides, polar dimmer leads to increase in viscosity.

Acid value

Acid value indicates the condition and edibility of the oil. The result of various acid values havebeen shown in Table 4. The comparison between the different oils(heated and reheated) have been discussed in Fig 3. Acid value is expressed as the amount of KOH (in milligrams) necessary to neutralize free fatty acids contained in 1.0g of oil. An increase in acid value is results in development of objectionable flavours and odours. According to Diop A etal (17) acid value increases after frying our results was also similar after repeated heating of edible oil. The acid value of 1st heated sunflower refined oil was lowest (0.434 mg/g) followed by 2nd heated sunflower refined oil (0.518 mg/g).Thehighest acid value was found in 6th heated ground nut crude (12.69 mg/g). Acid value of oils indicates the amount of free fatty acid present in the oil. %FFA content is a conventional expression of the percentage mass-fraction of total fat. Decrease in acid value reduces its decomposition to volatile compounds (aldehydes, ketones) which causes rancidity in oil.Thus,the stability and shelf life of the oil was increased. When acid value increases decomposition of volatile compounds (aldehydes, ketones)which causes increase in rancidity of oil and shelf life and stability of oil decreases and viceversa.

Saponification Value

Saponification value represents the number of milligrams of potassium hydroxide required to saponify 1 gm of fat. The result of various saponification values has been shown in Table 5. The comparison between the different oils(heated and reheated) have been discussed in Fig 4. Table 5 gives the saponification values of some edible oils and fig 4 shows the comparison of acid value between nonheated and reheating of different oil samples. It is a measure of the average molecular weight or chain length of all the fatty acids present. As most of the mass of a fat/tri-esters is in the fatty acids, the saponification value allows for comparison of the average fatty acid chain length. The long chain fatty acids found in fats have a low saponification value because they have a relatively fewer number of carboxylic functional groups per unit mass of the fat as compared to short chain fatty acids. According to Adel Imhemed Alajtal etal (18) saponification value increases after frying our results was also similar after repeated heating of edible oil. Saponification value is least for non-heated sunflower oil crude(63.34 mg/g) and highest saponification value is for 6th heated ground nut crude oil(260.89 mg/g). The lower the saponification value suggests the low molecular weight of fatty acids or lessnumber of ester bonds. Hence fat molecules did not interact with each other. The lower the saponification value lower the risk of resulting obesity, insulin resistance, inflammation, development of type 2 diabetes, hypertension, dyslipidemia, disorders of coagulation and atherosclerotic vascular disease.

Peroxide value

Peroxide value is used to measure the extent to oxidation reactions occurred during processing and storage. The result of various peroxide values have been shown in Table 6. The comparison between the different oils(heated and reheated) have been discussed in Fig 5. Detection of peroxide gives the initial evidence of rancidity in unsaturated fats and oils. Oils with a high degree of unsaturation are most susceptible to autoxidation. The double bonds found in fats and oils play a important role in autoxidation. The double bonds of an unsaturated fatty acid can be cleaved by free-radicle reactions in the presence of molecular oxygen. This reaction causes the release of malodorous and highly volatile aldehydes and ketones. This gives objectionable in taste and odour. Hence rancid food products cause immediate illness or harm, although rancidification can reduce the nutritional value of food by degradation of nutrients.Thebest test for autoxidation is determination of peroxide value,as peroxides are intermediates in the autoxidation reaction. Autoxidation is a reaction involving oxygen that leads to deterioration of oils and fats which form off-flavours and off-odour. Peroxide value, which is the concentration of peroxide in an oil or fats useful for assessing the extent to which spoilage has occurred. According to Adel Imhemed Alajtal etal (18) peroxide value increases after frying our results was also similar after repeated heating of edible oil. When we compare both nonheated and reheating of different edible oil we conclude that highest peroxide value is for 6th heated sunflower refined oil(9.87 mg/g)and least value is for 1st heated sunflower crude oil(0.42 mg/g). Finally, we found that 6th heated sunflower refined oil having high peroxide value hence it is having high degree of unsaturation it is more prone to oxidation hence it is better for cooking purpose. 1st heated sunflower crude oil (0.42 mg/g) having less peroxide value it is having less unsaturation. Oils exposed to both atmospheric oxygen and light showed a much large increase in peroxide value during storage.

CONCLUSION

Different crude and refined edible oils were collected from different areas in Tirupati. Physical chemical parameters like colour, viscosity, density, acid value, saponification value, peroxide valuewerechecked. On reheating of oil colour change occurs and however viscosity also increasing because formation of toxic compounds like polycyclic aromatic hydrocarbons. PAHs (benzopyrene, dibenzoanthracene, benzoanthracene), heterocyclic amides, 4-hydroxy-trans-2-nonenal(HNE) epoxides,polar dimmer. Acid value, saponification value, peroxide value also increases after frying. Reheating of oil concludes that while reheating of oil formation of toxic compounds, hydrogenation occurs which has serious impact on health so better avoid the reheating of oil in cooking purposes.

Acknowledgement:

The authors are thankful to Principal, Seven Hills College of Pharmacy, Venkataramapuram, Tirupati and Management, for their kind support for our research work.

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