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REVIEW ON THE POTENTIAL USE OF WASTE COOKING PALM OIL IN THE PRODUCTION OF HIGH OLEIC PALM OIL VIA ENZYMATIC ACIDOLYSIS

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Abstract

Production of structured lipids (SL) or tailor made fats provides an opportunity for cheap oils and fats to be utilized for the synthesis of high added value products. Much attention is being paid to SL due to their potential biological functions, industrial applications, and nutritional perspectives. The paper reviews the potential of waste cooking palm oil (WCPO) as an alternative substrate for the production of structured lipids especially for the production of high oleic palm oil. Utilization of waste cooking oil for biodiesel productions is well explored. However, WCPO utilization properties are still lacking scientific investigation due to limited chemical and physical functionalities. Therefore, enhancement of WCPO into high quality oil offers an alternative to be utilized for wide range of applications, thus reduce the environmental effect causes by its disposal problems. The paper also reviews and discusses the production of structured lipid via enzymatic acidolysis.

Keywords: Waste cooking palm oil, acidolysis, high oleic palm oil

1.0 INTRODUCTION

Developed countries are the major worldwide contributors to the increasing amount of waste cooking oils. Management of these oils face a significant challenge as their disposal problems and the possibility to contaminate water and land resources [1]. Disposal of waste cooking oils is relatively difficult due to their physical form. Usually waste cooking oils exist in a liquid or semi-solid form and solid waste regulations prohibit any liquid waste to be disposed in landfills. If open burning method is employed, black smoke will be produced. The most often and simplest method used especially by food industries is pouring the waste into drain or watercourse. At the end, the waste will results in pipes clogging and destruction towards waste water and septic system. With the mushrooming of fast food centres and restaurant in Malaysia, it is expected that extensive amounts of waste cooking oils will be discarded inappropriately.

Modified fats, known as structured lipids (SL) or "tailor-made fats", can be produced either chemically or enzymatically. Cheap oils and fats can be used as raw materials for the synthesis of high added-value products. In the field of edible oil and fats, the interest on lipase-catalysed production of SL presenting specific functional properties has greatly increased due to the benefits of enzymatic route relatively to chemical processes [2]. Lipases are vital tools for SL syntheses by modifying fatty acids composition and positional distribution of glycerol backbone in triglycerides. Acidolysis is one of the methods to produced structured lipid either by chemical or enzymatic reaction.

Chemical acidolysis catalysed by metal alkoxides which is a simple and inexpensive process. However, it is not proficient of modifying specific positions due to the random nature of the reactions [3]. Thus, enzymatic acidolysis offers a promising incorporation of desired acyl group into a specific position of triacylglycerol (TAG), while chemical catalysis is incapable to retain this region-specificity. According to Siew and Saw [3], the 2–position of the triglycerides in palm oil is specifically occupied by an unsaturated fatty acid, mainly oleic acid. Hence, 1,3-specific lipase (hydrolyse TAG atC1 and C3 glycerol bonds) are more favourable for positional specific modification of used-waste cooking palm oil; retaining the oleic acid at the 2-position of the triglycerides [4, 5]. The advantages of enzymatic reactions are selectivity, ease of product recovery, mild reaction conditions, little or no unwanted side reactions process, easy control over the process and less waste disposal [6, 7].

Xu [8] emphases on several advantages of lipase as biocatalysts for production of structured lipid are; i) efficiency of lipase under mild reaction conditions, ii) reduce environmental pollution, iii) utility in "natural" reaction systems and products, iv) availability of lipases from wide range of sources, v) the use of lipases for the production of particular biomolecules, and vi) ability to improve lipases by genetic engineering and in special situations.

For these reasons, many nutritional and specific structured lipids have been produced enzymatically and lot of studies were published. In addition, many general reviews have also been published on lipases and their applications in oil and fat modification since 1990 [8]. However, none of them focusing on the utilization of WCPO for the production of specific lipids. In this report, the potential of WCPO for enzymatic acidolysis for the production of high oleic palm oil is discussed with a brief over view of the application of high oleic oil.

Palm oil and palm olein have moderate contents of oleic acid [2, 9]. Oleic acid can be incorporated into oils to increase the oleic content of the mother oil, offer desirable properties related to both health benefits and stability characteristics [2]. Vegetable oils with enhance oleic acid content have receive great attention for use in a variety of industrial applications [10]. According to Corbett [11] high oleic canola oil has twice oxidative stability compared to regular canola oil. Besides stabilizing the oil, high oleic oil are also exploited in the preparation of snack food including potato chips frying as well as spraying to enhance mouth-feel and palatability. Not only restricted for food purposes, high oleic oils have been utilized for non-food applications. Ramli *et al.* [11] indicated that palm oil is among the vegetable source of this monounsaturated fatty acids due to the abundance amount of palm oil available in Malaysia compared to soybean and sunflower oil. Vegetable-based high oleic oil can be further process via transesterification to produce products such as polyurethane [13]. Other non-food applications include cosmetic formulation and use as vegetable lubricant.

2.0 SUBSTRATE: WASTE COOKING PALM OIL

Malaysian Palm Oil Industry Performance report has stated that Malaysia has produced 17.7 million tons of palm oil on 4.5 million hectares of land, as the second largest producer and largest exporter of palm oil in the world [14]. According to Sebayang *et al.* [15] about 40% of palm oil is made for cooking oil, margarine, special fats, and oleochemicals. Many types of vegetable oils are used for frying purposes including sunflower oil, olive oil, canola oil coconut oil, and palm oil. However, in Malaysia, palm oil has been widely used as cooking oil compared to other oils (canola oil, peanut oil, sunflower oil, soybean oil, olive oil). Generally, waste cooking oil is generated from vegetable oil that used at high temperature especially for frying or food preparation purposes. In general, vegetable oils will undergo oxidation and degradation process as the oil are heated in the extended of time which then produce oxides. Some of the oxides produced such as hydro peroxides, epoxides and polymeric compounds have scientifically shows an adverse health effects including growth retardation and organ damage [16].

Large amount of waste cooking oil are generated worldwide and this amount are varied depending on the amount of cooking oil consumed [17]. Estimated about 15 million tons of waste cooking oil are generated worldwide. Table 1 shows the amount of waste cooking oil generated from selected country. From the tabulated values, it shows that Malaysia has produced approximately 0.5 tonne of used-frying oils annually. This value is quite higher for the country with 27 million populations. In fast food industries alone, a single branch will serve fried food such as burgers, fried chicken, and French fries, contribute to the production of approximately 15 litres of waste cooking oil per day. Abdullah *et al.* [18] estimated that the total waste cooking oil generated from fast food franchises all over Malaysia reach several thousand litres a day. It is believe that the actual amount of waste cooking oil generated either worldwide or in Malaysia are far beyond the estimated value. Hence, waste cooking oil is readily available and possibly provides a constant feedstock for the demand of fatty acids and glycerol.

The increasing production of waste cooking oil from houses, restaurants, and industrial sources is a rising problem in the world. The disposal of waste cooking oil an important issue considered by many countries. Many developed countries strictly set policies on the disposal of waste cooking oil penalization through the water drainage [19]. For example, problems like drains blockage will appear resulting from the continuous process of pouring and throwing used-frying oil into kitchen sink. Sewerage system also may experience failure and affect the wastewater treatment plant causing the raise in processing cost. Even though some of these waste cooking oils were used for soap production, still the major portion of it is discharged into the environment [1]. According to Sanli *et al.* [20], 40% of sewerage system problems occurred in USA is caused by the waste cooking oil poured into the kitchen sink. Besides, this residue is also incorporated into food chain through animal feeds and become a possible cause of human health problems [21].

Table 1 Waste cooking oil production [25]

Country	**Quantity (million tons/years)**
United States	10.0
China	4.5
Europe	0.7 - 1.0
Japan	0.45 - 0.57
Malaysia	0.5
Canada	0.12
Taiwan	0.07

According to Enweremadu and Mbarawa [22], there is no specific or systematic method of processing waste cooking oil from households and most of the oil is thrown through home drains, ends up in wastewaters, and then discharged through surface waters, which ultimately caused water pollution. In addition, more than 80% of the oil is consumed at home, as the control of this disposal behaviour becomes a major problem due to the involvement of large volume [23]. Over the years, realisation on waste cooking oil utilization is mainly on the production of biodiesel. This insight of waste cooking oil exploitation for biodiesel production is generally from the argument of energy-crop programs compete with food crops that long termly will cause food shortages and price increase especially in developing countries [22]. Hence, the use of waste cooking oil for fatty acids enhancement will bring substantial environmental profits as it provides an alternative for final disposal of the oil previously discharged in to the environment as well as provide a solid foundation for exploration and exploitation on new utilization method of waste cooking oil [24].

3.0 CHEMICAL AND PHYSICAL PROPERTIES OF WCPO

Chemical and physical properties of waste cooking oil are mostly similar to fresh cooking oil [17]. However, relatively differ from source to source, depending on degree of heating, various physical and chemical changes of waste-cooking oil will occur [26]. Deep fat frying defines as simultaneous heat and mass transfer process ensued in food processing process where frying oil serves as heat transfer medium as well as plays a crucial role in flavour and texture characteristics [27]. In frying process, oil is heated in the occurrence of air and light at temperature of 160-200 °C for extended of time [17, 26]. Reports from Phiri *et al.* [28] indicate that a batch of frying oil use in fast-food business is discarded after 1 to 4 days of used depending on the operation hours. Common commercial practice indicates that frying oil should be discarded when the percentage of FFA reaches 1.0% or content of total polar materials exceed 24% [26]. However, there are no specific or precise measures to determine the fry-life of specific oil.

Cvengros and Cvengrosova [19] detected five common physical changes occurred in cooking oil after several frying processes including; increase in viscosity, increase in the specific heat, change in the surface tension, change in colour, and increase in the concentration of fat foam. Other than that, three common degradation reactions occurs are:

a) Hydrolysis (This process occurs due to the moisture content of fired food). FFA and glycerol are the products produced from this reaction [20].
b) Oxidation (this process occurs due to contact with oxygen). Oxidized monomeric, dimeric and oligo triglycerides and volatile materials such as aldehydes and ketone are several products form during the reaction. Unsaturated fatty acids react with molecular oxygen through a free-radical mechanism [26].
c) Polymerization: (The hydrolysis and oxidation processes occur at high temperature). The reaction produces ring structure of dimeric and polymeric triglycerides.

The combination of all the reactions results in the formation of undesirable products and increased the polar materials in the oil [26]. The change in the free fatty acids composition results from the double bond rapture during the frying process [29]. In addition, the saturation degree of oil also increases during the frying process [30]. The changes may influence some of the oil properties including heating content, iodine value, viscosity, and density of the oil depending on the number of frying. Therefore, these properties can be used to evaluate the quality of frying oil. Study conducted by Tynek *et al.* [31] has established a significant loss of the linoleic acid (C18:2) and a iodine value reduction of oil after heating arise from the intense thermo-oxidative transformations compared to heated oil containing food. The decrease in iodine value can be attributed to the destruction of double bonds by oxidation, polymerization, and scission [32]. Furthermore, the applied heat causes the oxidative rancidity causing an increase in the free fatty acids [33].

Nus *et al.* [34] successfully determined the composition of palm oil before and after the potatoes frying using column chromatography and high-pressure exclusion chromatography (HPSEC). The study had showed total polar content in the palm oil increased as the increase in number of frying. The continuous potatoes frying process has significantly increased the polymers, dimer and oxidized triacylglycerol contents. The results are demonstrated in Table 2. While Table 3 shows the description of fresh cooking oil and waste-cooking oil where both are originated from cooking palm oil. Study conducted by Abdul Halim and Kamarudin [35] showed higher water and fatty acid content of waste-cooking palm oil compared to fresh cooking palm oil. Abdullah [36] found higher acid value of waste-cooking palm olein compared to fresh palm olein. On the other hand, Alireza *et al.* [32] established a lower iodine value of used cooking palm oil than fresh cooking palm oil.

Table 2 Composition of different compounds in the palm oil at different numbers of frying [34]

Compound	Fresh cooking palm oil	WCPO
Water content (% wt)	0.8	0.95
Acid (mg KOH/g)	0.1	7.25
FFA (%)	0.13	0.2
Iodine value (g I_2/ g oil)	57.27	52.81

Table 3 Description of fresh cooking oil and WCPO [32, 35]

No. Frying	TPM	TGP	TGD	OTG	DG	NOTG
	Weight = mg/100 mg of oil					
0	9.3 ±0.1	0.1 ±0.0	1.0±0.3	1.1 ±0.2	6.7±0.2	93.7±0.1
60	18.2 ±0.1	1.6 ±0.0	5.0 ±0.1	5.3 ±0.1	6.1 ±0.1	82.8±0.5
90	26.4 ±0.3	18.2 ±0.1	7.9 ±0.1	8.3 ±0.1	6.2 ±0.1	73.6 ±0.3

TPM: Total Polar Molecules
TG: Triglycerides
DG: Diacylglycerol
TGP: TG Polymers
TGD: TG Dimers
OTG: Oxidized TG
NOTG: Non-oxidized TG

The peroxide value of oil indicates the rancidity in saturated fats and oils. Fan *et al.* [36] reported the peroxide value of palm olein after five days of frying process has increased from 3.40 to 34.55 meq/kg. On the other hand, the peroxide value of WCPO gained by Wannahari and Nordin [37] is 30 meq/kg. Report published by Wen *et al.* [38] specifies the peroxide value of waste cooking oil collected from a public restaurant in East China University of Science and Technology, Shanghai, China is 23.1 meq/kg. Tomasevic and Siler-Marinkovic [39] obtained the peroxide value of waste cooking sunflower oil ranged from 13.2 to 20.1 meq/kg. Chemical analysis of restaurant grease and animal fat samples conducted by Canakci [40] shows the average peroxide value of 66 meq/kg.

The acid number or acid value of oil is particularly corresponding to their ester amount, which indicates the quantity of free fatty acids (FFA) and mineral acids present in the sample [41]. Chhetri *et al.* [1] have reported the acid value for waste cooking canola oil is approximately 2.10 mg KOH/g which comparatively higher than the fresh canola oil. Abdullah [18] has reported the acid value for WCPO was 7.25 mg KOH/g. WCPO has higher acid value compared to fresh palm oil due to the increase in FFA content during the continuous frying process. Aladedunye and Przybylski [29] justified that the change in the FFA composition results from the double bond rapture during the frying process.

Iodine value defines the degree of unsaturation or double bonds of oils and fats [42]. It also specifies the ease of oxidation of oils and fats. Generally, frying process decreases the content of unsaturated fatty acids in frying oil and fat. Study conducted by Tynek *et al.* [31] has found a significant loss of C18:2 (linoleic acid) fatty acid and a fall in iodine value of oil after the frying process due to intensive thermo-oxidative transformations. The decrease in the iodine value is related to the destruction of double bonds by oxidation, polymerization, and scission. Alireza *et al.* [32] observed the reduction on iodine value from 57.27 to 52.27 gI_2/100g after 5 days of frying process for the study on the effect of frying process on fatty acid composition and iodine value of palm olein. Same trend of iodine value reduction was also found in the study conducted by Fan *et al.* [36], where iodine value of palm olein decreased from 51.2 to 44.6 gI_2/100g after 5 days of frying.

4.0 FATTY ACIDS PROFILE

The physical and chemical properties of waste cooking oil are almost similar to the fresh cooking oil and relatively diverse from source to source. However, water content and FFA content in waste cooking oils are comparatively higher than the fresh edible oil due to the effect frying process. According to Azócar *et al.* [43], waste cooking oil is known for high acid value due to the higher level of FFA. During the frying process, oil is heated in the presence of air and light at the temperature range from 160 to 200 °C for relatively long period of time [17]. Thermolytic, oxidative, and hydrolytic are three common reactions occurred throughout the frying process and resulting in the physical changes of the oil: increase in viscosity and specific heat, change in surface tension, colour, and rise the concentration of fat formation [26].

Study conducted by Taufiqurrahmi *et al.* [44] found WCPO consists mainly of 28 % oleic acid and 21.47 % of palmitic acid. Abdul Halim and Kamaruddin [35] showed that the FFA content of WCPO is higher compared to the fresh cooking palm oil. Knote and Steidley [30] stated that, the total concentration of stearic and palmitic acid increase significantly after frying process. The study also revealed the increase in oleic acid (unsaturated fatty acid) concentration as well as the reduction in concentration of linoleic acid

(18:2) and a- linolenic acid (18:3) after the frying process with the formation of other fatty acid such as palmitoleic (16:1) and arachidic (20:0) in waste cooking oil. Table 4 illustrates the comparison of fatty acids composition between the fresh cooking palm oil and WCPO.

5.0 ENZYMATIC ACIDOLYSIS

Enzymatic acidolysis involves the strategy of acyl exchange of oils with excess of fatty acid. The strategy includes the substitution of fatty acid residues specifically at the 1,3-positions of the oils via 1,3-position-specific lipase, making the residues at the 2-position unchanged. Ferreira-Dias *et al.* [2] described that acidolysis reaction as a reaction consists of fatty acyl group is exchanged through a combination of hydrolysis of a donor ester with esterification of released alcohol moiety with another free fatty acids (Figure. 1). Hypothetically, these reactions produced the mixtures of triacylglycerols (TAGs) and targeted structured TAGs as well as fatty acids. The fatty acids can be removed after the reaction by molecular distillation or alkali extraction. Thus, if all the acyl groups at the 1, 3-positions are replaced with the desired one, eventually the target structured TAG species with specific structure can be attained with relatively high purity. Examples of specific TAG obtained by lipase-catalysed modification of TAGs are human milk substitutes, low calories TAGs, TAGs enrich omega-3 polyunsaturated fatty acids (PUFA), and cocoa butter substitutes.

Table 4 Fatty acids composition of fresh cooking palm oil and waste-cooking oil [44, 45]

Fatty acids	Carbon number: double bond	Fresh Cooking Palm Oil (%)	WCPO (%)
Oleic acid	C18:1	49.482	28.64
Palmitic acid	C16:0	36.768	21.47
Linoleic acid	C18:2	11.747	13.58
Stearic acid	C18:0	-	13.00
Palmitoleic acid	C16:1	-	7.56
Myristic acid	C14:0	0.849	3.21
Linolenic acid	C18:3	0.539	1.59
Lauric acid	C12:0	0.230	1.1
Arachidic acid	C20:0	0.161	0.64
Heptadecanoic acid	C17:0	-	0.59
Arachidonic acid	C20:4	-	0.37
Eicosadienoic acid	C20:2	-	0.29
Others		0.219	8.04

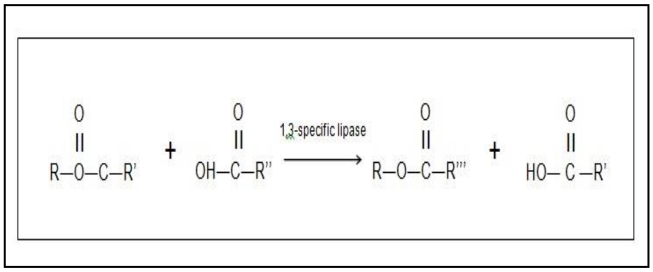

Figure 1 Enzymatic reaction of acidolysis

Carrı´n and Crapisten [22] have stated the drawbacks of chemical-based partial hydrogenation and transesterification of vegetable oil in food preparations including the increase in saturated fatty acids concentration as well as trans-fatty acid isomers. The trans isomers which known to have similar physical properties to saturated fatty acids, however have shown adverse health implications more than saturated fatty acids. The drawbacks have triggered the upsurge interest towards lipase-catalysed reactions due to great activity under mild reaction conditions, high specificity, and selectivity towards their natural substrates [5]. Therefore, enzymatic acidolysis is an appropriate method to incorporate a desired acyl group onto a specific position of triacylglycerol as to produce structured lipids by overcomes the flaws of chemical catalysis. Chemical catalysis does not reflect the regiospecificity due to random nature of the reaction compared to lipase-catalysed reactions that offer improved products quality as well as easy downstream process due to the absence of by-products formation. Gupta *et al.* [23] also have emphasized the advantages of lipase mediated modification of oils and fats possess specific reactions with moderate reaction conditions. According to Aravindan *et al.* [24] acidolysis processes are widely implemented in biotechnological

Rafael and Roberto [50] have emphasized the advantages of enzymatic acidolysis process including the high positional selectivity and specificity, low cost and fewer side products, initiated by the inevitable existence of acyl migration which can be minimized by controlling the reaction conditions such as enzyme load, water content, substrate ratio, reaction temperature, and reaction time. Therefore, enzymatic acidolysis has been preferred as a suitable method to enriched vegetable oils with regard to the modification of oils rich in high value polyunsaturated fatty acids such as linoleic acid, oleic acid, arachidonic acid, eicosapentaenoic acid, and docosahexaenoic acids. Enzymatic acidolysis has been implemented widely in variety of consumer products mainly in oleochemical industries. In addition, the applications of structured lipids by acidolysis process have attracted much attention and this has effect in the growth of interest in producing modified lipid [51]. Table 5 shows the applications of enzymatic acidolysis in several productions of consumer products.

Table 5 Application of enzymatic acidolysis

Application	Reference
Low caloric and dietetic Triacylglycerol	52-55
Human milk fat substitution	59-63
Cocoa butter substitutes	67, 68, 122
Plastic/ coating material	46, 68
Frying oil	51, 70, 72, 73

Amongst vegetable oils, virgin olive oil is a remarkable substrate for the production of TAGs with medium chain fatty acids due to its high oleic acid (55–83 wt%) and moderate linoleic acid (3.5–21 wt%) contents as located mainly at *sn*-2 position in TAGs. The structured TAGs gained from enzymatic acidolysis of virgin olive oil with medium chain fatty acids catalysed by *sn-1,3* specific lipases will present oleic acid at the sn-2 position, while medium chin fatty acids will be esterified at the external position as this provides better absorption for human consumption [52]. Covas *et al.* [53] reported that dietary ingestion of virgin olive oil have physiological benefits including supressing certain types of cancer, lowering serum cholesterol levels, enhancing liver function, and reducing the effects of aging as well as heart disease. Other types of oils such soybean and fish oils are successfully incorporated with medium chain fatty acids, caprylic and capric respectively which offer nutritional and therapeutic benefits [54, 55].

Generally, human milk fat comprises oleic acid (30-35 %) palmitic acid (20-30 %), linoleic (7-14 %), and stearic acid (5.7-8 %). Unlike vegetable oils and cow's milk fat, 65 to 70 % of total palmitic acid (major saturated fatty acid) in human milk fat is esterified at *sn*-2 position of TAGs, while the unsaturated fatty acids are located at the external positions [56, 57]. In addition, fatty acids profile in human milk fat has essential influence on intestinal absorption and digestibility of infants especially the presence of palmitic acid at *sn*-2 position provide an effective absorption of palmitic acid as *sn*-2 monoacylpalmitate [2, 58]. Sahin *et al.* [59] indicated the use of vegetable oils and cow's milk will result in deficient of calcium and fatty acid absorption caused by the formation of insoluble calcium soap with the released of fatty acids by the action of the *sn-1,3* specific pancreatic lipase. Therefore, many studies regarding the production of human milk substitute have been established by enzymatic acidolysis catalysed by *sn-1,3* lipase from different sources including lard, tripalmitin, palm oil, butterfat, and palm stearin with free fatty acids [59, 60, 61, 62, 63].

Cocoa butter substitutes are very important enzymatic produced structured lipid especially in food industry for chocolate making as it has unique composition that gives desired physical properties to manufactured product including snap, melting properties, and gloss [64]. Cocoa butter is essentially formed by TAGs with saturated fatty acids (palmitic and stearic acids), located at the external positions, and a monosaturated fatty acids (oleic acid) at *sn-2* position [2]. This specific structure is responsible for the distinctive rheological and sensory properties of cocoa butter. In addition, cocoa butter melts over narrow temperature range from 27 to 33°C as this melting behaviour provides a cooling effect in the mouth [65]. Due to the high cost and fluctuations in the supply as well as the demand, the industry has come out with alternatives with similar TAG composition via lipase-catalysed reaction either interesterification or acidolysis of plant oil with fatty acids. Ciftci *et al.* [66] conducted the study on acidolysis of pomace oil with mixture of palmitic and stearic acid using *n*-hexane as solvent for the production of cocoa butter-like fat in packed-bed reactor.

Osborn and Akoh [67] have used beef tallow as substrate for the production of cocoa butter substitute via enzymatic acidolysis using *Rhizomucor miehei* (IM 60) lipase. Coatings are applied on food products for multiple of reasons. Edible coating commonly prepared from proteins, polysaccharides, as well as lipids. Unlike proteins and polysaccharides, lipids are effectively facilitating a moisture barrier by averting or minimizing the transport of permeable gases, moisture, and aroma between the food and its external environment with improved surface appearance [68]. In addition, lipids also play a crucial role as protecting encapsulated materials from moisture absorption. Sellapan and Akoh [68] conducted the study on the production of edible coating lipids via enzymatic acidolysis of tristearin with lauric and oleic acid acids. The study showed that the synthesized coating lipid has improved characteristics in preventing moisture absorption than cocoa butter along with emphasizing the effect of operating condition such as temperature, substrate mole ratio, water content, time, enzyme load, and enzyme reusability. On the other hand, acidolysis is useful for the production of plastic fats and oils especially in shortening and margarine manufacture where the chemical properties of original fat and fatty acids inherent properties are unaffected [67]. Carrı´n and Crapiste [46] performed the modification of sunflower oil with palmitic-stearic acids via lipase catalysis in order to gain healthier alternative hydrogenation products.

Modification of fatty acid composition of vegetable oils has been accomplished for many years as to attain three quality aspects: i) nutritional value ii) oxidative value and iii) functional properties [69]. For example, high oleic oils have greatly received attention due to nutritional properties including minimizing the cardiac related diseases. Through the years, enzymatic acidolysis has been implemented as a fast and easiest method for plant oil modifications with mild reaction conditions, specificity of enzymatic action and better acceptability, and products with better defined chemical structure and composition [70-71]. Lai *et al.* [51] indicated that the unique combination of structural lipid where medium chain fatty acids located at *sn-1, 3* positions, and essential long chain fatty acid at *sn*-2 the glycerol backbone provides a quick energy effect, and other health benefits. Liew [72] accomplished the incorporation of oleic acid with palm olein at optimum temperature of 50 °C, 10 % (w/w) lipase loading and substrate concentration of 1:2 (palm olein: oleic acid molar) where final oleic acid content has increase up to 60% by using *T. Lanoginosa and Alcaligenes sp* lipase. Liew [72] has stated that high oleic content in palm oil; more than 50% will inhibit the olien solidification in temperature climate. On the other hand, Fajardo *et al.* [73] incorporated palm oil with *n-3* polyunsaturated fatty acids (PUFA) to obtain average EPA and DHA incorporation of 20.8% and 15.6% for 24 hours incubation using IM60 from *Rhizomucor miehei* and QLM from *Alcaligenes sp.* The study on lipase-catalysed acidolysis of palm olein and caprylic has been conducted by Lai *et al.* [51] in continuous pack bed reactor where the study achieved the incorporation degree of caprylic acid up to 30.5% by using Lipozyme IM 60.

6.0 STRUCTURAL LIPID

Natural fats exist in a wide variety of functional, nutritional, and organoleptic properties which particularly depend on the composition of saturated and polyunsaturated fatty acids, length of fatty acids chain, and the distribution of different fatty acids in TAG with respect to the *sn-1*, *sn-2*, or *sn-3* positions 2. Hayes [74] expresses structured lipid as TAG containing the mixtures of short (C_1-C_4), medium (C_6-C_{12}), and long (C_{14} and higher) chain type of acyl group that restricted to either the 1(3) – or 2-position on the glycerol backbone. According to Xu [8], novel TAG can be obtained by several method including incorporation of new fatty acids, changing the natural position of fatty acids profile, and synthesis of completely new TAG. These structured lipids (SL), also known as modified lipids, or "tailor made fats" can be produced by chemical or enzymatic reactions.

Example of structured lipid obtained by lipase-catalysed modification of fatty acids composition in triacylglycerol (TAG) are human milk substitutes, low calorie TAGs, cocoa butter substitutes, and omega-3 polyunsaturated fatty acid enriched TAGs as well as other specific fatty acid. Therefore, cheap or used/waste oils and fats could possibly be used as raw materials for the synthesis of high value added products with enhanced and broad utilization properties. A lot of studies have successfully incorporated vegetable oils with various fatty acids derivatives including oleic, linoleic, palmitic, caprylic, and pentadecanoic [3, 75, 76, 77, 78, 79].

The advantages of SL in nutrition, health, and food applications cannot be overemphasized. Currently researchers have concentrated on improving the techniques of SL synthesis for maximum incorporation degree with optimum reaction conditions. Yankah and Akoh [77] have described the industrial validity of any fats or oils are restricted by the non-random distribution of fatty acids resulting to impart a given set of physical, chemical, and functional properties. In addition, natural fats have known to have low variability desired especially in food industry. Keskin *et al.* [80] indicated that natural oils (mainly TAG) are differ in their physical and nutritional properties, hardly meet the nutritional recommendations or display the desirable physiochemical properties. Hence, synthesis of SL with desired fatty acid has provides an alternative solution to enhance functional properties of natural fats to meet a nutraceutical need including nutritional, therapeutic/medical, low caloric value, or fortification [66, 67, 81].

Yankah and Akoh [77] stated that reformation of fat to inhibit absorption or to achieve a lower caloric density is the principle of low-calorie fats due to the metabolism and absorption of fatty acids depend on their position in triacylglycerol. Therefore, the physiological effects of the desired fatty acids as well as the positional orientation on the glycerol backbone must be initially identified. With respect to the desired positional distribution of the fatty acids as *sn*-1, *sn*-2, or *sn*-3, the enzyme selection is the main determining factor. Nagachinta and Akoh [79] have reported that the absorption of eicosapentaenoic acid and DHA was higher when they were predominantly in the *sn*-2 position than they were randomly distributed on 3 positions of TAG. Study conducted by Lopez-Lopez *et al.* [58] revealed that lower fat absorption in formulated infant milk than human milk is due to the differences in stereospecific structure of TAG of formulated milk and human milk fat.

According to Siew and Saw [3], the 2–position of the triglycerides in palm oil is specifically occupied by an unsaturated fatty acid, mainly oleic acid. Therefore, 1, 3–specific enzymes are more favourable for positional specific modification of waste-cooking palm oil; retaining the oleic acid at the 2-position of the triglycerides. Enzymatic reaction have known for its advantages on selectivity, mild reaction conditions, little or no unwanted side reactions or by-product isomerisation, ease of product recovery, easy control over the process and less waste disposal [6, 7]. In food industry for example, interseterification reaction has carried out at high temperature (up to 270 °C), under reduce pressure, for less than 2 hours, by using metal alkylates or alkali metal as catalyst. The interchange of acyl group proceeds at random where final products remain contaminated by residual catalyst and the formation of considerable amount of side products, with a subsequent low product yield [2].

The replacement of inorganic catalyst by lipases has been attempted due to the advantages of enzymatic route relatively to the chemical process. Lipase catalysed structured-lipid is frequently carried out in batch and continuous reactors, either with the presence of organic solvent or in solvent-free system. Ferreira-Dias *et al.* [2] have claimed the use of thermostable lipases is crucial for reactions that occur in solvent-free system, where at least one of the substrates has high melting point such as palm stearin (m.p. = 47-54°C). As for the reactions carried out at near-room temperature, the use of organic solvent is essential to dissolve the solid fats. However, this will increase the complexity of the system and costs related with solvent and downstream processing. Based on these facts, search for green processes have attracted the attention for search and advance study for lipases produced by thermophilic microorganisms. Table 6 shows several studies on lipase-catalysed acidolysis reaction of different oils/fats for production of structured lipids.

Table 6 Lipase-catalysed production of structured lipids

Product	Biocatalyst	Substrate	Reaction system	Result
High oleic acid palm olein [3]	Lipozyme TL IM	Palm olein + oleic acid (C18:1)	Solvent-free	56% C18:1
Modified palm olein with high PUFA [75]	Lipozyme TL IM	Palm olein + C18:2*n*-6 + C18:3*n*-3	*n*-hexane	24.6% C18:2n-6 3.74% C18:3n-3
HMS[76]	Lipase DF	TAG + C16:0 + C18:1	*n*-hexane	67.2% C18:1 67.8% C16:0
Nutritional low-calories fat [7]7	*Rhizomucor miehei*	Tristearin + C18:1 Tristearin + C8:0	*n*-hexane	55.2% C18:1 37.0% C8:0
HMS[79]	Novozyme 435	Palm olein + DHA[3] + ARA[4]	*n*-hexane	25.25g/ 100g DHA+ ARA 17.2g/ 100g DHA+ ARA (*sn*-2 position)
Modified sunflower oil [78]	Lipozyme RM IM	Sunflower oil + palmitic-stearic acids blend	*n*-hexane	-
Modified Terebinth fruit oil [84]	Rhizomucor miehei	Terebinth fruit oil + C8:0 + 18:0	*n*-hexane	19% C8:0 14% 18:0

Enzymatic intersesterification as a contrivance for fats modification has offers many advantages. In contrast to hydrogenation and fractionation, interseterification does not continually alter the fatty acid composition or the unsaturation level of starting mixture; but the inherent stability of the oil or blend remains predictable [77]. Even though chemical interesterification catalysed by metal alkoxides is a simple and inexpensive process, it is not proficient of modifying specific position due to random nature of this reaction [3]. Enzymatic interesterification also affords a milder processing conditions and the possibility of regiospecificity and fatty acid specificity. Hasnisa and Jumat [75] have justified the application of lipases for modification of oils and fats offer some benefits over the conventional catalysts including the synthesis of novel products and incorporation of desirable fatty acids at specific positions of the lipid as to improve functionality, metabolism, adsorption, clinical use, and nutritional properties. In addition, Senanayake and Shahidi [83] described the enzyme-mediated interesterification reaction is a suitable method for small scale modified structured-lipid production due to the milder reaction conditions with less side reaction.

Generally, acidolysis is the reaction involves the replacement of acyl group from the reacting acids to produce fatty acid esters. For example, the involvement of lipase in acidolysis reaction for the production of human milk substitution (HMS). Nagachinta and Akoh [79] prepared human milk substitution formula from palm olein enriched with docosahexaenoic (DHA) and arachidonic (ARA) using Novozyme as catalyst in *n*-hexane system. Good quadratic models were obtained for the incorporation of DHA and ARA at optimum reaction conditions (temperature: 58-60°C; substrate mole ratio to total fatty acids to palm olein: 12 mol/mol; reaction time: 18-24 hours). On the other hand, Esteban *et al.* [76] successfully produced HMS from incorporation of palm stearin with palmitic acid enriched with acylglycerols at *sn*-2 position and oleic acid enriched with fatty acids mixture at *sn*-1, 3 positions in presence of *n*-hexane at 37°C. Pacheco *et al.* [78] constructed the kinetic models for sunflower oil acidolysis reaction with palmitic-stearic acids blend using Lipozyme RM IM for the production of improved melting point, solid fat content, and oxidative stability of sunflower oil.

Lipase-catalysed acidolysis reaction for the production of healthier diet fatty acids has been well explored. Siew and Saw [3] have conducted the study on enrichment of palm olein with high oleic acid for production of high oleic oil (HOO). The study also emphasized on the health properties of HOO, particularly against coronary heart disease, high cholesterol level, and oxidation. Hasnisa and Jumat

[75] produced modified palm olein with linoleic acid and a-linolenic acid catalyst by Lipozyme TL IM for the production of enriched palm olein with polyunsaturated fatty acids to increase nutritional value. Yankah and Akoh [77] incorporated tristearin with oleic acid and caprylic acid to produce dietary fat with low calories using *Rhizomucor miehei* lipase in the presence of *n*-hexane. Kocak *et al.* [81] have characterized fatty acids and triacyglycerol composition of terebinth fruit oil for the synthesis of modified terebinth oil with high content of caprylic acid and stearic acid in *n-hexane* system using immobilized *sn*-1, 3 specific lipase from *Mucor miehei*. Caprylic acid was incorporated to reduce caloric value and stearic acid to surge the melting point of oil.

7.0 HIGH OLEIC OIL

7.1 Oleic Acid

Oleic acid (cis-9-octadecanoic acid) is a monosaturated fatty acid present in animal and vegetable oils. It is an odourless and colourless fatty acid. It occurs naturally in greater quantities than other fatty acid derivatives [3]. Oleic acid is a group of omega-nine fatty acid, and considered as one of the healthier fatty acids in the diet. Oleic acid commonly abbreviated with lipid number of 18:1 cis 9 and has the formula $CH_3(CH_2)_7CH=CH(CH_2)_7COOH$ [85]. The term 'oleic' means related to or derived from oil or olive which predominantly olive oil composed of oleic acid. According to Siew and Saw [3], oils contain high oleic acid posse high premium over the normal oil. Table 7 shows oleic acid chemical and physical properties.

Oleic acid is has the ability to facilitate improved health as well as oxidative stability for lengthen the oil shelf life, flavour, durability and cold flow performance [87]. From the study conducted by Shannon [88], increasing oleic acid composition to more than 60% will enhance edible and industrial applications. The study also found that soy oil with high oleic acid content reduces the need for hydrogenation and eliminates trans-fats as well as improved soy-diesel, lubricants and hydraulic oils properties. A diet in which fat consumption is high in oleic acid, like olive oil, is related with lowering cholesterol levels, arteriosclerosis, and heart disease [89, 90]. Hence, high oleic acid-contain oils have a significant prospect to substitute higher saturated fresh or used edible oil by delivering certain functionality and nutritive values.

7.2 Application of Oleic Acid

Oleic acid is commonly found trapped in triglycerides, which are lipids that closely associated to oil secretion and production. Plasma triglycerides help the liver isolate fat and glucose and responsible for the excretion of skin oil. In plants, oleic acid functions to provide nutrition and sustenance. Sales-Campos *et al.* [91] have indicated that oil rich with oleic acid such as olive oil has beneficial effect on cancer, autoimmune and inflammatory diseases, as well as ability to aid wound healing. Lopez–Huentas [92] has claimed that the substitution of saturated fat with oleic acid or PUFA has been expected to reduce the risk of coronary heart disease by 20-40 % via low-density lipoprotein (LDL). Other literatures also reported other beneficial effect of oleic acid intake on reducing the risk factors for cardiovascular diseases including factors lead to thrombogenesis, insulin sensitivity, and in-vitro LDL oxidative susceptibility [93, 94].

Table 7 Properties of oleic acid [86]

Physical Properties	
Molecular formula	$C_{18}H_{34}O_2$ $C_8H_{17}CH=CH(CH_2)_7COOH$
Synonym	(Z)-9-Octadecenoic acid; cis-9-Octadecenoic acid; Red oil
Molecular weight (g/mol)	282.47
Melting point (°C)	13
Boiling point (°C)	360
Density (g/ml)	0.8634
Specific heat at 50 °C (J/g)	2.046
Vapour pressure at 175 °C	1 Torr
Hazardous Characteristics	
Overall toxicity	0
Flammability	1
Absorbed through skin	0
Destructive to skin/eyes	0
Self-reactive	No (Incompatible with strong oxidizing agent)
Sensitizer	No

0-None (very low); 1-Slight; 2-Moderate; 3-High; 4-Severe

Hulshof *et al.* [95] have recommended high oleic acid intake will be beneficial as it limits the intake of saturated fat. This can be accomplished by altering the dietary patterns such as using high oleic acid contain oil (olive oil) instead of using butter, or by using modern food technology to modify fatty acid profile of particular foods naturally rich in saturated fatty acids by incorporation with oleic acid. A study conducted by Lopez-Huertas [92] has showed the significant effects of long term consumption (1 year) of milks enriched with long chain omega-3 PUFA (EPA + DHA) and/or oleic acid on peripheral vascular disease (a type of cardiovascular disease) patient, reduced the total cholesterol to 9%, yet maintained the normal levels of LDL-cholesterol as well as potential atherogenic particles. The study also concluded that to reduce serum levels of total cholesterol and LDL-cholesterol, it is recommended to substitute the whole milk generally consumed by children with a fat-free milk preparation. Table 8 shows the summary of oleic acids industrial applications.

Table 8 Oleic acid industrial applications

Application	Reference
Health and Food Purposes	
i. Treatment for cancer, inflammatory dieses, and wound healing	91
ii. Dietary supplement: Low density lipoprotein (LDL) lower the risk of cardiovascular dieses	93, 92, 100
Non Food Purposes	
i) Biolubricant/ Synthetic fluids	96, 97, 98
ii) Surfactant	101, 102, 103, 104
iii) Plasticizer	105, 106, 107, 108, 109, 110
iv) Nanoparticles	111, 112, 113, 114, 115, 116, 117

The use of oleic acid not only limited to dietary and nutritional purposes. Oleic acid has been utilized for variety of applications including for non-food purposes. One of oleic acid application that gained much attention is the production of biolubricant via chemical modification of oleochemicals olefinic group [96]. Salih *et al.* [97] have conducted a study on improvement of thermostability of biolubricant derived from epoxidation of oleic acid (source from palm oil) via opening of the oxirane ring, and esterification for enhance performance and efficiency. In addition, Salimon *et al.* [96] have produced synthetic biolubricant basestocks through chemical modification of oleic acid with improved low temperature properties and oxidative stability. Garcia-Zapateiro *et al.* [97] deal with thermal, viscous, and tribological characterization of variety of estolides obtained from oleic and ricinoleic acids via different synthesis protocols, and amount of vegetables oils blends. The study aims to achieve and propose a promising formulation for wide range of lubricant application. Ozgulsun *et al.* [98] indicated that the use of environmentally acceptable vegetable-oil-based products such as biolubricants has many advantages including nontoxic, renewable, biodegradable, and lower production cost compared to other synthetic fluids.

Oleic acid also has been exploited for the production of surfactants. Surfactants are surface-active compounds widely used for cosmetics, household detergents, and cleaners. Generally, the feedstocks are C_{12} and C_{14} fatty acids and alkanols of petrochemical origin, where C_{12} and C_{14} acids are obtained from coconut and palm kernel oil (laurics) which are supplied from South East Asia [101]. Study conducted by Dierker and Schäfer [101] signified the synthesis of surfactants with similar properties as from lauric oils by increased the hydrophilic properties of oleic acid by ionic and non-ionic group attached to the double bond and carboxylic group with improved water solubility and micelle formation. Kalhapure and Akamanchi [102] carried out the study aimed to meet the needs of pharmaceutical industry for safe and nontoxic surfactant from oleic acid via attachment through ester linkage. Xu *et al.* [103] studied the surfactant-free microemulsion composed of oleic acid as to discover the essential conditions for preparation of surfactant-free emulsion with long term dispersion stability. Kamogawa *et al.* [104] prepared the oleic acid/water emulsion in surfactant-free condition by sequential processing using midsonic-megasonic waves.

The use of natural plasticizers with low toxicity as well as good compatibility in several polymers application has gained more attention. Natural-based plasticizer is an alternative approach for conventional polymers including rubber, resin, plastics, and elastomers [105]. Extensive research is going in order to discover with improved functionality of alternative plasticizer for medical and other commodity of plastic materials. This includes epoxidized triglyceride originated from vegetable oils such as soybean oil, linseed oil, castor oil, sunflower oil, and fatty acid esters [106, 107]. Hence, oleic acid has been introduced as one of the raw materials for the synthesis of natural plasticizer. Pena-Serna and Lopes-Filho [108] conducted the study on effect of ethanol and glycerol concentration on structural and functional properties of zein-oleic acid films. Orellana-Coca *et al.* [109] have employed lipase-catalysed esterification, epoxidation, and perhydrolysis reaction for the production of epoxy alkylstearates (stabilizer for PVC, primary plasticizers) from oleic acid in solvent-free system. In addition, Yahya and Arof [110] studied the effect of oleic acid plasticizer on chitosan-lithium acetate solid polymer electrolytes for advance applications and development of thin and safe rechargeable lithium ion batteries.

Studies on magnetic nanoparticles have gained much more attention recently. The studies are subjected in biological applications such as magnetic drug delivery, cell separation and sorting as well as magnetic resonance imaging [111, 112]. Thach *et al.* [113] studied the effect of size of magnetite nanoparticles on the ability of drug loading by coated the nanoparticles with single layer of oleic acid, function as hydrophobic surface, as well as coated with double layer of oleic acid/sodium dodecyl sulphate for hydrophilic surface. Anwar *et al.* [114] deliberated a study on aqueous phase transfer of oleic acid coated iron oxide nanoparticles for pharmaceutical applications especially ferro-fluid. The iron oxide nanoparticles were coated with different oleic acid concentration (5-50%) and characterized by dynamic light scattering (DLS). Zhang *et al.* [115]

have identified the chemical structure of the surfactant adsorbed on the magnetic nanoparticles and conferred the model of oleic acid molecules coated on the nanoparticles surface. On the other hand, Gu and Chen [116] have selected oleic acid for carbon coatings on $Li_4Ti_5O_{12}$. The study has revealed that carbon coating by oleic acid (carbon source) has enhanced the ability of lithium ion diffusion in the electrode with capacity of 164.67 mA.h/g even after 50 cycles. In addition, Garland *et al.* [117] performed a study on oleic acid coverage in organic aerosol with the evidence of island formation in which oleic acid serves as an inert or reactive barrier to diffusion gases.

7.3 High Oleic Oils

Oleic acid is one of the most important and versatile oleochemicals derived from oils and fats [12]. Vegetable oils with enhance oleic acid content have receive great attention for use in a variety of industrial applications [10]. Ramli *et al.* [12] indicated that palm oil is among the vegetable source of this monounsaturated fatty acids due to the abundance amount of palm oil available in Malaysia compared to soybean and sunflower oil. High oleic canola oil has twice oxidative stability compared to regular canola oil [11]. Study conducted by Przybylski *et al.* [82] shows highest frying stability of high oleic canola oil from the measurement of polar components, oligomers and non-volatile carbonyl components formation.. Ray *et al.* [118] produced structured lipid composed of high 1,3-distearoyl-2-oleoyl-sn-glycerol content stearins via acidolysis of high oleic sunflower oil (HOSO) with stearic and palmitic acids. Ray *et al.* [118] also emphasized that HOSO comprises of high oleic acid levels (80%), mainly in the form of triolein (OOO) (>70%) and contained low solids content at room temperature. Even though HOSO does not contain enough solids for many applications but it can be structurally modified through enzymatic acidolysis as to increase the overall Sat-O-Sat (symmetric disaturated monounsaturated TAG) level. Not only restricted for food purposes, high oleic oils have been utilized for non-food applications.

Clemente and Cahoon [87] stated that high soybean oil with 85% oleic acid has an oxidative stability index value which is nearly 12-fold higher than conventional soybean oil. In addition, this property is crucial for the use of vegetable oil for lubricants, motor, and hydraulic oil [119,120]. Study conducted by Kinney and Clemente [10] showed that high oleic oil display superior properties in biodiesel formulation compared to the conventional soybean oil. Similar result has been found from study conducted by Tat *et al.* [121] regarding exhaust emissions from engine fuelled with biodiesel from high oleic soybean oil. Tat *et al.* [121] also established that biodiesel from high oleic soybean oil has enhanced oxidative stability with less emission of nitrogen oxides (NOx) as well as improved cold point properties due to lower content of palmitic acid (4% in high oleic oil compared to 12% in conventional soybean oil). Undoubtedly, vegetable oils with novel additional functionality can be generated by crossing of high oleic oils derived from biotechnology with altered contents of fatty acid alternatively from easy, fast, and cost friendly method via enzymatic acidolysis compared to breeding of mutant alleles.

8.0 CONCLUSION

The review showed the potential of cooking palm oil as an alternative substrate/ raw material for the production of structured lipid, namely the production of high oleic oil.

Nowadays, many researches are focusing on the production of structured lipid from edible oils. Therefore, this review examines the utilization of WCPO for the production of structured lipid as to reduce the environmental effects caused by its disposal problems via enzymatic acidolysis with less environmental effects. The enhancement of cheap oil into high quality oil offers an alternative method for its utilization for wide range of applications not only limited for food and nutraceutical industries. However, further development of enzyme technology for the application of structured lipid from oleo-chemical substances is essential. The search for more stable and effective low-cost process will allow high production at optimized reaction conditions, with great benefits in terms of utilization properties and environmental aspects.

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