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Effect of Coolant Temperature on Progressive Freeze Concentration of Refined, Bleached and Deodorised Palm Oil based on Process Efficiency and Heat Transfer

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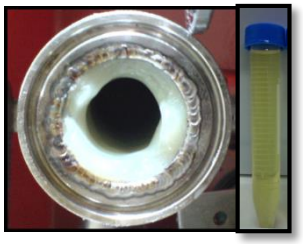
Graphical abstract



Product of stearin in solid and olein in liquid

Abstract

In this paper, raw material of refined bleached and deodorised palm oil (RBDPO) was separated into olein and stearin by applying progressive freeze concentration (PFC) as an alternative method to replace the conventional fractionation process using coil crystallizer. Overall heat transfer coefficient (U) was analyzed from the process by varying different coolant temperature values since both of them are closely related each other. In this case, heat transfer efficiency depends strongly on crystal of stearin formed on the inner wall of cooled surface and is explained theoretically from that angle. At optimum flowrate, operation time and initial iodine value (IV), the best results were observed at 28ºC of coolant temperature where high IV of olein and effective partition constant (K) obtained were 55.8 wijs and 0.27 respectively. Meanwhile, the highest U obtained at coolant temperature 28ºC same as result for process efficiency at 392.9183 W/m².K and time at 55 min.

Keywords: Progressive freeze concentration; palm oil; refined bleached and deodorised palm oil; iodine value and heat transfer

1.0 INTRODUCTION

Palm oil is resourceful edible oil and crucial as a vegetable oil besides coconut oil, palm kernel, or olive oil which is used as raw material for food or non-food industries. The oil palms (Elaeis) are very well-known worldwide because of the benefits to be used as raw material for a variety of products such as soap, frying oil, biodiesel, edible fats in the confectionery, powder detergent, ice cream, mayonnaise, pomades, candle and etc. Different types of oil can be extracted from the oil palm fruit and the refinery such as crude palm oil, RBDPO, stearin, olein, fractionated palm olein and palm mid-fraction. There is no doubt that many industries especially in Malaysia search and try to find a new technology to enhance the productivity and quality of oil with low cost. These issues also will be driven by continuing increase in the population in major consuming nation, especially in Asia [1].

There are three types of fractionation, which are dry fractionation, detergent fractionation, and solvent fractionation. Among the three fractionation processes, dry fractionation is the most common process used, which involves crystallization of olein at its melting temperature (16-24°C). Dry fractionation is mostly used in Malaysia's refinery industries because of its own advantages in the process compared to detergent and solvent fractionation. Particularly, solvent and detergent fractionation so far need or required greater capital investment than dry fractionation [2] and dry fractionation only requires crystallizer, filter and washing unit.

A typical suspension freeze concentration (SFC) process is composed of three processing units which are ice nucleator, a recrystalliser and ice crystal separator. Ice nucleator normally used in SFC is scraped surface heat exchanger (SSHE) to generate ice nuclei and to maintain high heat transfer by scraping the ice layer formed. The limited size of ice crystals formed from ice crystal scraping need an additional step in order to increase the size of ice crystals thus increase the process complexity. Furthermore, due to the large surface area of small ice crystals, the product obtained is not highly pure and increase the difficulty to separate it from mother liquor [3]. Apart from that, SSHE that is normally used in SFC process is the most

expensive type of heat exchanger, which is leading to high capital cost [4].

On the contrary, improved method of freeze concentration (FC) which is PFC is discovered by Matthews and Coggeshall in 1959, in which a single ice crystal is formed on the cooled surface [5]. PFC is applying the same concept with SFC but the major difference between these two methods is the size of ice crystal formed. The solution that needs to be concentrated flows over a cooled surface, which causes crystallization process, occurs on the surface. Further growth of ice crystal is producing ice crystal in layer form. The large size of ice crystal resulting in lower surface area and less impurities is trapped at the ice-liquid interface. The separation of ice occurs when concentrated solution is collected and flushed while the ice crystal adheres to the surface making the separation process easier [6]. Since the process is involving less unit operations, hence it is expected that the process to be much simpler and lowering initial investment compared with previous method.

PFC process in this study involved the generation of solid stearin on the wall of the crystalliser as a single solid layer where the crystallization that occurs is contributed by the water temperature in water bath. The circulation flowrate gives an impact of heat transfer. This system involved several units that would contribute to heat transfer to generate the solid crystal of stearin as a layer on the wall of the coil crystalliser (CC).

Theoretically, the formation of crystals consists of three stages, where the solution at the first stage would go through an initial chilling process and to reduce the natural temperature of solution. Meanwhile in the second stage, the solution receives a temperature low enough for nucleation to occur and at the same time due to the exothermic nature of the process fusion heat will be released from the formation of solid stearin nuclei. Then, the solid stearin crystal nuclei will continuously grow until the actual temperature setting is reached. In natural process, this stage is in an unsteady state, which takes a shorter time. Further on, in the third stage, the bulk temperature changes slowly and as a consequences solid stearin crystal would form continuously with increasing concentration of mother liquor [7].

In this study, the quality of palm oil and process efficiency was determined by iodine value (IV) and effective partition constant, *K*. The *K* of the product could be influenced by the type of fractionation process, cooling rates and temperature of fractionation [8]. The quality of oil was measured based on the degree of unsaturation or double bonds presence in oils and fats, as indicated by the IV. It also reflects the ease of oxidation of oils and fats [9]. The chain length of fatty acids, trans fatty acid content, saturation ratios, and the position of fatty acids in glycerol backbone are changed depending on the value of IV [10]. Other than that, the IV is an important attribute of many specifications trades of quality oil, and in some country, it is an element of the legal definition of food product [11].

The present paper aims to study and evaluate the effect of coolant temperature towards process efficiency and heat transfer of PFC process to separate olein from RBDPO. In particular, this technique has never been applied in RBDPO application. By applying FC concept where water components usually crystallise, in this research stearin at melting point 44-58°C will be crystallise instead and concentrated olein at melting point 22-24°C is obtained [12].

2.0 EXPERIMENTAL

2.1 Material

RBDPO was used as a sample solution throughout the experimental work. Meanwhile, pure water used in waterbath, functioning as a medium for cooling and heating process.

2.2 Equipment

The crystalliser is known as coil crystalliser (CC), which could provide higher productivity and efficiency for progressive freeze concentration process through its higher surface area. For this study, the CC was made of stainless steel, as illustrated in Figure 1, and it was used to carry out the separation process of RBDPO. The crystallizer has three layers or stages and also fitted with stainless steel flanges, thus the chamber could easily be split into two to collect the products from the process. In addition, nine temperature detectors, thermocouple type K, are engaged in each stage of the crystallizer in order to determine the temperature profile which later are displayed by PicoLog recorder software through connected computer.

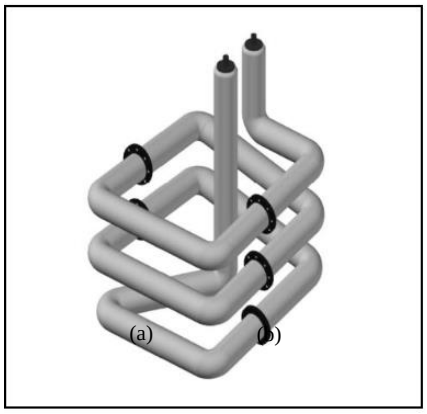


Figure 1 Coil crystalliser (CC)

2.3 Experimental Procedure

To start the operation, RBDPO obtained from a local palm oil refinery was pumped by a peristaltic pump into the CC until the crystalliser is completely filled. Then, it was immersed in a refrigerated water bath. The CC was connected to a peristaltic pump by a silicone tube and the RBDPO was circulated in the system for a period of an hour. Before starting the circulation at the desired temperature, the CC containing RBDPO must first be heated at a temperature of 70°C to avoid the presence of unwanted crystal and also to remove previous thermal history [9].

After that, the process proceeds to the designated reduced temperature, usually from 28°C until 24°C, which corresponds to the melting point of stearin and olein, which are about 48 °C and 20°C, respectively [9]. The circulation flowrate of RBDPO of approximately 2800 mL/min in the CC would cause the stearin layer to be formed on the inner wall surface of the CC and leaving behind a more purified olein. The process was also carried out with varied of time at 40 to 60 min and constant for initial iodine values of palm oil at 52.5 wijs as usually used in the refinery of palm oil.

Throughout the process, nine different points of the CC were fixed with thermocouples to measure the temperature of

the coolant, RBDPO, and the wall of the crystallizer. The measured data were displayed on the computer after being detected by PicoLog recorder for easier monitoring process. After the desired temperature, the circulation was stopped and the purified olein was drained out, leaving behind stearin layer, which has to be melted or thawed, and its purity was then analysed. Before starting the successive experiment, the CC was flushed with hot water. The temperature of the water bath was then increased to 48°C to melt stearin completely and detached it from the wall of the crystallizer. The experimental setup is shown in Figure 2.

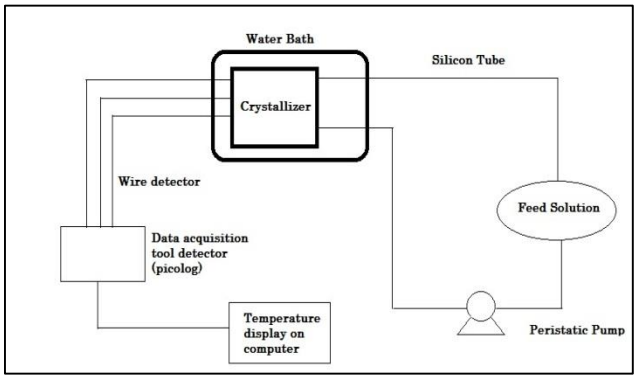


Figure 2 Experimental setup

2.4 Analytical Determination of RBDPO

Theoretically, the mechanism of concentration process occurs as a result of the exclusion of solute molecules from the advancing crystal front, the interface between the crystal of stearin and concentrate of olein phases [13]. Effective partition constant (K) acts as an index for separation effectiveness [14]. K is calculated based on Equation (1).

$$(1 - K) \log (V_L/ V_o) = \log (C_o/C_L) \quad (1)$$

where V_o and C_o are the initial volume of solution and initial solute concentration of solution, respectively. The final volume of solution and final solute concentration of the solution are represented by V_L and C_L respectively. The value of K varies between 0 and 1 where zero (0) indicates complete freeze concentration process [15]. The efficiency of each concentration process depends on the concentration increment of the concentrate in relation to initial solution concentration. As a rule, decrement solute in crystal produce higher solutes remained in concentrate.

The quality or another process efficiency of palm oil depends on the IV values of olein and stearin. The IV of olein and stearin were measured using MPOB Test Method p3.2-2004 and p4.2:2004 [16], respectively. The p3.2-2004 method is technically equivalent to ISO 3961:1996, while MPOB Test Method p4.2-2004 originated from AOCS Official Method Cc3-25 [17]. IV is calculated by the following Equation (2):

$$\text{IV} \left(\frac{g}{100g}\right) = \frac{12.69C\,(V_1 - V_2)}{m} \quad (2)$$

where,
C= Concentration of sodium thiosulfate solution (mole L^{-1});
V_1= Volume (mL) of sodium thiosulfate solution for the blank test;
V_2= Volume (mL) of sodium thiosulfate solution for the sample;
m = Mass (g) of the sample.

■3.0 RESULTS AND DISCUSSION

3.1 Determination of Iodine Value (IV) and Effective Partition Constant (K) for Coolant Temperature Effect

The determination of IV is crucial as it gives the measurement of unsaturated oil and saponification value, which is used to obtain the average molecular weight of the constituent fatty acid. Lower fatty acid in the palm oil gives better quality and high IV olein [18]. Table 1 summarized the statistical evaluation of K and IV value in olein as process efficiency under the influence of coolant temperature. The data collected was a little bit different from the standard reference of dry fractionation for olein result collection, but it is still acceptable by referring to the trend when the data is plotted. Usually the standard reference data collected from Palm Oil Research Institute of Malaysia (PORIM) for IV olein are 56.0- 57.0 wijs [11, 19, 20]. The less satisfactory data might be related to the final product that was not filter pressed due to high cost of filter pressed.

Meanwhile, coolant temperature is a main factor in this FC and PFC system since the process is closely related to temperature. Figure 3 shows the variation of coolant temperature with K and IV value of olein for disclose the trend of PFC process performance and efficiency. The coolant temperature range selected varies between 29 to 24ºC based on the experimental values of coolant temperature used in previous research by Berger *et al*. and Chong and screening tests for RBDPO itself [11, 12]. The figure shows a result for IV for olein; as the coolant temperature decrease, the IV of olein gives a high quality and efficiency trend increases exponentially from 54.8 wijs up to 55.8 wijs and K decreases from 0.62 to 0.27. The results seem quite satisfactory in the concentration limit and confirmed its capabilities in high production.

To make this reason more reliable and acceptable, according to Kawamura; [21, 22] and Berger [23], α crystals formed when the crystallization was conducted below 24°C and appeared as very unstable dotted spherulites under the microscope and its lifetime depends on the temperature, while β crystals, which appeared as very stable dendritic spherulites, were formed at temperature above 26°C.

Table 1 Data of process on coolant temperature for olein

Temperature (ºC)	Flowrate (mL/min)	Time (min)	Initial iodine value (wijs)	K-Value	IV of Olein (wijs)
29	2800	60	52.5	0.5200	55.20
28	2800	60	52.5	0.2715	55.84
27	2800	60	52.5	0.2999	55.63
26	2800	60	52.5	0.3690	55.37
25	2800	60	52.5	0.5693	55.00
24	2800	60	52.5	0.6221	54.89

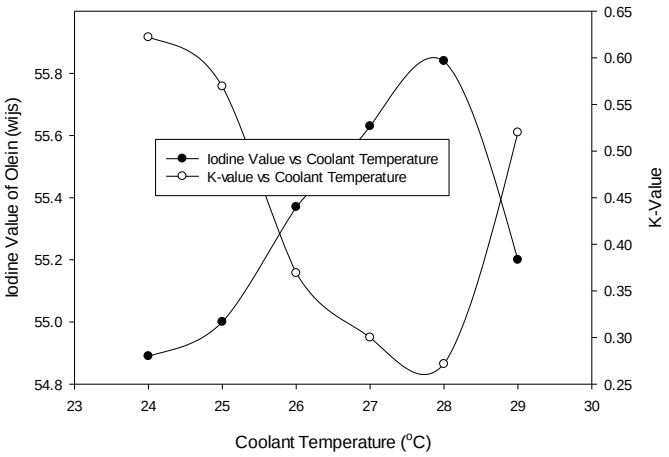


Figure 3 IV and K efficiency of olein

The figure also can be observed that the separation process is more effective at 28ºC since K was obtained is small and IV of olein quality efficiency is high. This implies that more soluble solids remained in the concentrate and higher purity of ice crystals was obtained. Lower K value (0.27) indicates that the ice crystal layer is slow in acquiring the solutes. The higher retention of RBDPO solution might be due to the fact that its chemical structure facilitates the retention of olein (unsaturated fatty acid) and stearin (saturated fatty acid) by integrating it more easily between the two molecules, thereby facilitating the growth of crystals of stearin in the cooling wall. The flow of slurry or RBDPO creates an optimal supersaturation and rates of nucleation and growth. It also enhanced the heat transfer between RBDPO and the cooling surface, and at the same time gives the optimal rates of crystal growth to produce a compact layer of suitable morphology and sufficient purity of stearin, thus achieving high IV [24].

Coolant temperature has strong influence on the crystal formation rate [25]. As the temperature difference between coolant and solution increases, the crystal formation rate increases and larger amount of the crystal of stearin formed. Particularly, the highest rate formation of crystal at 24ºC, it has been noted that the soluble solid trapped together and will strongly build the triglyceride (palmitic acid) bone in accordance to the different melting point because the speed of moving solids in solution becomes too fast to overtake the solids outward movement and promote solute inclusion in the stearin crystals [26]. This situation makes the inner wall of CC covered with crystal solids and gives products of low quality and impurities, and led to loss of palm olein (i.e. low yield of olein) [27]. IV of olein quality efficiency achieved was up to 55.8 wijs for this parameter and it is quite satisfactory in the quality standard of olein process limit.

3.2 Overall Heat Transfer

The method used in the present study to determine U。 is by measuring the variation of solution temperature. The solution and coolant temperature is a function of time, T_m (t), during chilling or freezing process period which can be determined experimentally by averaging the solution temperature values of inlet, outlet, and middle cycles. If the temperature of the solution in crystallizer decreases from T_{s1} to T_{s2} , where the temperature are different between the time interval in range 60 min, the heat removed is too long while it can take only about 30 min to reach T_{s2} where the stearin completely to be a solid. The situation also makes the stearin and olein mixed together and become a solid.

PFC system also will produce a trend of temperature profile based on the design of the crystalliser. The temperature profile could be observed by the temperature detector of the acquisition tool, called PicoLog. For this study, there are about two trends of temperature profile for heating and cooling process but for this paper only cooling process for further discussion.

Figure 4 shows the temperature for cooling process, it can be seen that at stage 2, Tsm suddenly drops down and goes up again, which indicates the release of fusion heat to depicts stearin nucleation occurring at that stage. It occurs in a very short period and facing unsteady heat transfer compared to the stable temperature in stage 3 where it is almost at its freezing point. In stage 4, U values can be determined by measuring the solution concentration increment in a definite time.

In order to determine the temperature distribution and heat flow, an approach through a basic heat transfer according to heat flow across a tube was employed, which is between inside and outside following thermal resistance [28]. The general information about the detailed of calculation was depicted in Equation (3).

$$R=\left[\begin{matrix}Thermal\ resistance\\ of\\ inside\ flow\end{matrix}\right]+\left[\begin{matrix}Thermal\ resistance\\ of\\ tube\ material\ flow\end{matrix}\right]+\left[\begin{matrix}Thermal\ resistance\\ of\\ outside\ flow\end{matrix}\right] \quad (3)$$

In this study, the PFC process involved cooling and freezing which is to generate a layer of solid stearin crystal on the inner wall of the CC. Therefore, there is a resistance, R, involved between the material flowing inside the CC and the coolant outside the CC. The R could then be expressed by Equation (4) with assuming R through the stainless steel wall to be negligible for this study.

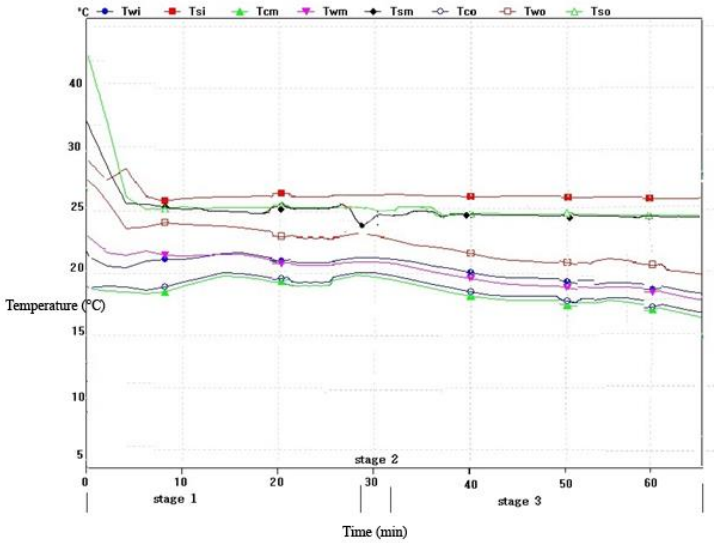


Figure 4 Temperature profile of the cooling process in the PFC system from PicoLog data

$$R = \frac{1}{A_i h_g} + \frac{x}{k_i A_m} + \frac{1}{A_o h_o} \quad (4)$$

Where A_i = inside surface area of CC, m^2; A_o = outside surface area of CC, m^2; $A_m = \frac{A_o - A_i}{In\left(\frac{A_o}{A_i}\right)}$ = logarithmic mean area, m^2; h_g = heat transfer coefficient for RBDPO,W/($m^{2.o}$C); [29] h_o = heat transfer coefficient for water, W/ ($m^{2.}$ °C); k_i = thermal conductivity of tube material, W/(m. °C); R = total thermal resistance from inside to outside flow, °C/W and x = thickness of medium wall, m.

Meanwhile, this system, A_m would be calculated using different equation considering area of layer of solid stearin crystal surface. Hence, Equation (5) is produced:

$$A_m = 2\pi L \frac{x}{In\left(\frac{r}{r-x}\right)} \quad (5)$$

where r = radius of the stainless steel tube for the CC, m ; L = length of the stainless steel tube for the CC, m and x= thickness of CC wall.

However, R could be expressed in terms of overall heat transfer coefficient, U_o using Equation (6). The equation may be expressed as follows:

$$R = \frac{1}{U_o A_m} = \frac{1}{A_i h_g} + \frac{x}{k_i A_m} + \frac{1}{A_o h_o} \quad (6)$$

In order to calculate U_o, effect of coolant temperature gives the thickness of solid stearin on the inner wall of CC. The thickness is inserted in Equation (6) to calculate A_m and justify that the resistance affects the thickness. Other constant values are listed as follows:

L = 2.37 m
h_o = 0.58 W/ ($m^{2.o}$C)
h_g = 243.55 W/($m^{2.o}$C)
A_o = 0.308 m^2
A_i = 0.189 m^2
r = 0.0254 m

3.3 Heat Transfer Performance

Figure 5 illustrated the heat transfer performance or overall heat transfer in the best result for coolant temperature effect shows at 28ºC U was found to be 392.9183 W/m².K and time at 55 min. Compared to another values of U at different coolant temperature and time, U values at 28ºC seems to be the highest but the timing used need to be longer in order to give completely cooling the crystalliser as shown in Figure 4, which at stage 3 is a comfortable zone for cooling process occur. The suitable value for U is chosen at 55 min as it believed that longer time was needed to perform crystal of stearin formation in the wall of crystalliser.

This is in agreement with the findings from previous researchers, where it was found that for palm olein, short tempering period does not always allow adequate crystallization of RBDPO. It was also mentioned that 1 hour in insufficient, but 8 hours or more is adequate for complete crystallization [12] Nevertheless, this process has used completely different equipment and the condition of the equipment influences the operating condition for circulation or tempering time. Therefore, if longer circulation time was used in this process, more solid crystalline will be developed on the inner wall surface of the crystalliser.

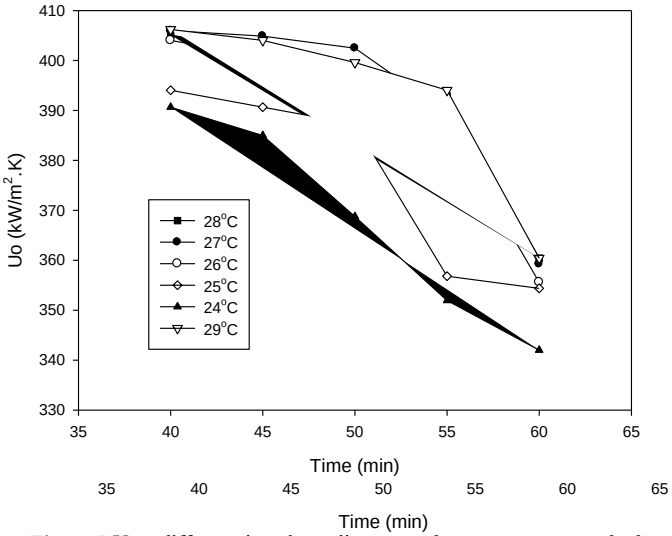


Figure 5 U_o at different time depending on coolant temperature applied

From Figure 5, it is revealed that the heat transfer from outside the CC wall into the solid stearin depends on the coolant temperature and for that reason the thickness of solid stearin also changes. Besides, the stearin crystal thickness has gradually increased over time. Meanwhile, at longer time, there is higher resistance, hence decreasing the overall heat transfer coefficient. Figure 5 also reveals that the best U_o is at the intermediate at time and coolant temperature of 55 min and 28ºC respectively. The reason of choosing this intermediate U_o instead of the higher value is because the process occurring at high overall heat transfer corresponds to a shorter time, thus providing a shorter period for stable crystal production, which is unfavourable.

4.0 CONCLUSION

Fractionation of RBDPO through PFC system is completely different from the conventional system, but the process still relies on the heat transfer performance with influence of coolant temperature. High process efficiency, high quality olein and high heat transfer have been observed at the temperature of 28°C. The K-value and IV of olein was obtained at 0.2715 and 55.8 wijs, respectively. Meanwhile, the best choice of U_o according to the time process was obtained at 55 min with heat transfer of 392.9183 W/m².K. The result conformed to the quality of palm based-standard reference, thus this process could serve as an attractive alternative for fractionation of RBDPO. All analyses support the objective of this study and give enough evidence for good efficiency and heat transfer performance of olein in the PFC system.

Acknowledgement

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