Australian Journal of Crop Science AJCS

AJCS 13(06):954-965 (2019) ISSN:1835-2707
doi: 10.21475/ajcs.19.13.06.p1705

Response surface methodology optimization of oil extraction from oil palm meal (OPM) with hydrous ethanol and its pilot-scale application with recirculation of extraction solvent

Krit Somnuk[1*], Jarernporn Thawornprasert[1], Pin Chanjula[2], Gumpon Prateepchaikul[1]

[1]Department of Mechanical Engineering, Faculty of Engineering, Prince of Songkla University, Hat Yai, Songkhla, Thailand, 90112
[2]Department of Animal Science, Faculty of Natural and Resources, Prince of Songkla University, Hat Yai, Songkhla, Thailand, 90112

***Corresponding author**: **krit.s@psu.ac.th**

Abstract

In the small-scale dry oil extraction from palm fruit used by palm oil mills, mixed crude palm oil (MCPO) is extracted using a single screw press. The oil palm meal (OPM) by-product can be used as alternative feed for ruminants. Three parameters in further solvent extraction of oil are the ethanol-to-dried oil palm meal (DOPM) ratio (4.9–30.1 $g.g^{-1}$), the extraction time (0.2–18.8 min), and the speed of stirrer (48–552 rpm). These parameters were optimized to for maximum oil yield by response surface methodology (RSM) while employing hydrous ethanol as the solvent. In our laboratory-scale oil extraction, the maximal experimental yield was 10.27 wt.% under the recommend condition: 20.1 $g.g^{-1}$ ethanol-to-DOPM ratio, 11 min extraction time, and 300 rpm speed of stirrer. Moreover, prototype-scale oil extraction was tested with recirculated miscella for effects of the number of cycles on oil extraction from fresh DOPM. The results showed approximately 17.4 wt.% oil yield from 5000 g DOPM at the conditions recommended based on laboratory-scale experiments. In this study, recirculated miscella was used to extract oil from fresh DOPM. The first four cycles of oil extraction were almost as efficient as with fresh ethanol. Moreover, the protein content in the defatted oil palm meal (DFOPM) improved with oil extraction, because residual oil in DOPM was removed. The DFOPM should have less rancidity and longer shelf-life than DOPM.

Keywords: Recirculation; Oil palm meal; Solvent extraction; Hydrous ethanol.
Abbreviations: ANOVA_analysis of variance; DFOPM_defatted oil palm meal; DG_diglyceride; DOE_design of experiment; DOPM_dried oil palm meal; EDOPM_extracted oil of dried oil palm meal; FFA_free fatty acid; GC–FID_gas chromatograph–flame ionization detector; HHV_higher heating value; *k*_number of variables; MCPO_mixed crude palm oil; MG_monoglyceride; MS_ mean square; OPM_oil palm meal; Y_o_ yield of oil; PKC_palm kernel cake; _PKM palm kernel meal; PNS_palm nut shell; PPF_palm press fiber; *p*-value_indicator of statistical significance; RM_recirculated miscella; RPF_roasted palm fruit; RPO_refined palm oil; RSK_rubber seed kernels; RSM_response surface methodology; R^2_coefficient of determination; SEM_scanning electron microscopy; SS_sum of squares; TG_triglyceride; TLC/FID_thin layer chromatography with flame ionization detection; *Y*_response variable; vol.%_percentage by volume; wt.%_percentage by weight.

Introduction

In Thailand, oil palm (*Elaeis guineensis*) is an economic crop providing energy and food. Most palm oil factories are located in southern Thailand (Prasertsan and Prasertsan, 1996). Regarding fuel uses, crude palm oil (CPO) is used as a raw material in producing biodiesel. Approximately 0.97 million tonnes of biodiesel from CPO was blended with petroleum diesel and distributed through the local gas stations, in the years 2013-2017 (Office of Agricultural Economics, 2018). In terms of food crops, CPO at approximately 2 million tonnes is the leading vegetable oil in Thailand, and 1.09 million tonnes of refined palm oil (RPO) was produced in the years 2013-2017 (Office of Agricultural Economics, 2018). Palm oil is a relatively inexpensive vegetable oil, the more expensive edible oils including soybean, rapeseed, and sunflower oils (Norhaizan et al., 2013). CPO can be extracted from palm fruit by two methods, namely by wet or dry extraction (Azeman et al., 2015). Commercial large scale palm oil producers use wet extraction, for which the palm fruit are first treated with steam. The palm fruit are then extracted using a double screw press. The free fatty acid (FFA) content in CPO from this wet method is below 5 wt.% (Norhaizan et al., 2013). Regarding dry extraction, the palm fruit are first heated with hot air, followed by extraction in a single screw press, in small-scale palm oil plants (Mba et al., 2015). With high around 8–12 wt.% FFA, mixed crude palm oil (MCPO) (Jansri et al., 2011) can be used as animal feed. A by-product from the dry method is the oil palm meal (OPM), which is normally just used to feed ruminants (Lunsin, 2016). The hard palm nut shell (PNS) is not an appropriate ingredient in animal feed, instead the PNS should be separated from OPM for use as fuel, because it has

a high heating value. Such small-scale operation is not highly capital-intensive in terms of the initial and the maintenance costs (Owolarafe et al., 2007). After mechanical extraction, high residual oil content remains in the OPM, because of the low efficiency of such oil extraction. In summary, the dry method using screw press has poor efficiency of oil extraction, with the solid waste carrying residual oil that can be solvent extracted (Zakaria and Harvey, 2012). Rancidity tends to increase with high residual oil in animal feed (Baker and Davies, 1996; Mercier et al., 1998; DeRouchey et al. 2004), and the residual oil in OPM can be extracted with a solvent to address these problems.

Regarding oil extraction with a solvent, Santos et al. (2013) evaluated the oil yield from five vegetable species: castor oil plant, physic nut, soybean, groundnut, and crambe, using hexane and ethanol solvents (Santos et al., 2013). The results showed that hexane was significantly more efficient than ethanol. *Jatropha curcas* had the highest 48.57 wt.% and 25.57 wt.% oil yields when using hexane and ethanol, respectively. Clearly, the oil yield from *Jatropha curcas* extraction on using hexane had 62% higher efficiency than with ethanol solvent. Reshad et al. 2015 studied oil extraction from rubber seed kernels (RSK) using Soxhlet apparatus for solvent extraction. Response surface methodology (RSM) with factorial central composite design (CCD) was employed to optimize two parameters: extraction time (3–12 h) and ratio of RSK-to-solvent (0.03–0.09 $g.mL^{-1}$) with various solvents (hexane, methanol, and ethyl acetate). RSK with approximately 1 mm average particle size was extracted for 4 h using polar and non-polar solvents at solute–solvent ratio 0.064. The highest 46.01 wt.% oil yield was obtained with hexane, while the lowest 32.67 wt.% yield was with methanol (Reshad et al., 2015). Similar results are described by Sayyar et al. 2009 and Somnuk et al. 2017. However, considering toxic contaminants in ruminant feed, ethanol is superior to hexane. The residual ethanol in the final product is easier to remove and less harmful than residual hexane. Moreover, extraction with hexane as solvent is expensive, complex and poses health and safety hazards, due to flammable and explosive solvent vapors (Zakaria and Harvey, 2012). Therefore, organic solvents such as hexane are not suitable for food processing, and furthermore, handling organic waste is costly (de Melo et al., 2015; Filly et al., 2015; Kurnin et al., 2016; Elgharbawy et al., 2018). Ethanol (ethyl alcohol) is more environmentally friendly than toxic solvents such as hexane and methanol. In Thailand, ethanol is produced by fermentation from cassava and molasses (Silalertruksa and Gheewala, 2012). The purity of ethanol can be improved by removing the residual water in hydrous ethanol (95 vol.% purity) using a molecular sieve to obtain anhydrous ethanol (99.9 vol.% purity) (Wei-Cheng et al., 2014). Thus, the production cost of anhydrous ethanol is higher than that of hydrous ethanol. An alternative environmentally friendly solvent such as ethanol is needed for oil extraction from OPM. To the best of our knowledge, little research has directly focused on the optimization of oil extraction from dried palm oil meal (DOPM) with hydrous ethanol, to produce animal feed. Therefore, the objective of this work was to optimize three key parameters: ratio of ethanol-to-DOPM, extraction time, and speed of stirrer, in laboratory-scale oil extraction from DOPM of oil with hydrous ethanol, by response surface methodology (RSM). A 5-level 2-factor central composite design (CCD), is employed to optimize these three parameters. Moreover, prototype-scale tests of oil extraction are reported in this work, with recirculation of miscella (RM), i.e. reusing recovered extraction solvent in the next batch extraction. The number of cycles of oil extraction from fresh DOPM is assessed when RM extraction was repeatedly run until the efficiency had decreased significantly.

Results and Discussion

Oil extraction from DOPM in laboratory scale

Fig. 1 shows oil palm meal (OPM, Fig. 1A), OPM after comminution, approximately 2 mm particles of dried oil palm meal (DOPM, Fig. 1C) after drying, and defatted oil palm meal (DFOPM, Fig. 1D). OPM is solid fibrous waste from extraction of mixed crude palm oil (MCPO) by mechanical screw pressing of palm fruit. DOPM fibers are short and dark brown. After oil extraction the color of DFOPM was pale brown. According to Table 1, the crude fiber fractions were 34.88 wt.% and 12.03 wt.% in DOPM and DFOPM, respectively. The protein content in DOPM was 8.39 wt.%, and 13.00 wt.% in DFOPM. The protein content in the latter is a much higher because residual oil in DOPM was removed. Thus, the DFOPM has improved protein content for feed of ruminants. Regarding the use of solid wastes from small-scale palm oil production, fuel energy in biomass was studied. Approximately 19098.7 $kJ.kg^{-1}$ for DOPM, and 19063.3 $kJ.kg^{-1}$ HHV for DFOPM were found. The HHV of both DOPM and DFOPM is similar to softwood biomass, approximately 19000 $kJ.kg^{-1}$ HHV (Demirbaş, 1997). The ash contents were 10.53 wt.% for DOPM and 5.75 wt.% for DFOPM, with about 58.7% decrease from oil extraction. The ash content of DOPM was higher than of typical woody biomasses, such as hazelnut shell, softwood, hardwood, tea waste, wood bark, corncob, or olive husk; but lower than those of wheat straw and tobacco leaf (Demirbaş, 1997). The ash from DOPM does not have sufficient nutrients for use as fertilizer. However, such ash has been used as fertilizer for palm trees, and it was highly alkaline with pH approximately 11 (Tay and Show, 1995). The ash from burning DOPM can be used as a partial replacement for cement in a concrete mix (Munir et al., 2015). The lignin contents were 30.39% in DOPM and 30.36 wt.% in DFOPM, so this was not significantly affected by solvent extraction. For instance, Li et al., (2012) reported that ethanol organosolv pretreatment with dilute sulfuric acid as a catalyst was studied in order to enhance enzymatic saccharification of moso bamboo. Increasing acid content in the liquid mixture played a very important role in catalyzing the removal of hemicellulose and lignin (Li et al., 2012). Thus, lignin cannot be extracted with pure ethanol as used in oil extraction. These lignin contents of approximately 30 wt.% in DOPM and DFOPM are similar to softwood, tobacco stalk, and spruce wood (Demirbaş, 1997). Similar level was reported by Medina et al. (2015), namely 28.89% lignin yield from the oil palm empty fruit bunches was achieved using a sequential acid/alkaline treatment. Furthermore, Watkins et al., (2015) reported that the lignin yields from different biomass resources after a formic acid/acetic acid treatment and peroxyformic acid/peroxyacetic acid treatment of alfalfa fibers, pine straw, wheat straw, and flax fibers were 34%, 22.65%, 20.40%, and 14.88%, respectively (Watkins et al., 2015). The experimental design matrix and laboratory scale results are summarized in Table 2.

Table 1. Physical properties and chemical compositions of DOPM, DFOPM and hydrous ethanol.

Property	DOPM	DFOPM	Hydrous ethanol
Higher heating value (kJ.kg^{-1})	19098.7	19063.3	
Ash (wt.%)	10.53	5.75	
Protein (wt.%)	8.39	13.00	
Crude fiber (wt.%)	34.88	12.03	
Element (wt.%)			
Carbon	46.59	47.54	
Hydrogen	5.92	5.61	
Nitrogen	1.63	1.02	
Sulfur	0.14	0.144	
Oxygen	31.51	36.59	
Lignin (wt.%)	30.39	30.36	
Property and price of solvent			
Purity (%, min)			95 (vol.%)
Density @30^{o}C (kg.L^{-1})			0.8
Boiling point (^{o}C)			78.15
Price (USD.kg^{-1})			0.53$^{(a)}$

Note: $^{(a)}$ refer to Bank of Thailand (2017)

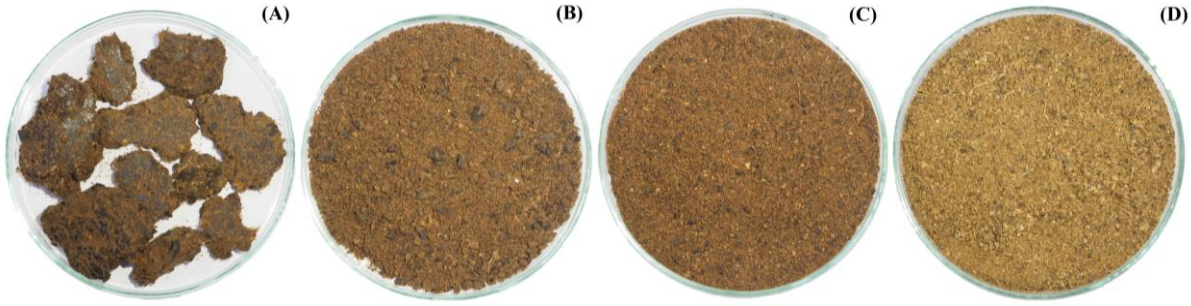

Fig 1. Raw materials and products of each processing step; (A) oil palm meal, (B) OPM after comminution process, (C) approximately 2 mm particles of dried oil palm meal after drying, and (D) defatted oil palm meal.

Table 2. Experimental design matrix and results for oil extraction from DOPM.

Run	*R* (g.g^{-1})	*T* (min)	*S* (rpm)	Yield of oil, Y_o (wt.%)
1	17.5	9.5	300	10.10
2	17.5	9.5	300	10.04
3	17.5	9.5	300	10.15
4	17.5	9.5	300	10.05
5	17.5	18.8	300	10.35
6	17.5	0.2	300	8.59
7	25.0	15.0	450	10.86
8	25.0	4.0	450	10.51
9	4.9	9.5	300	6.57
10	30.1	9.5	300	11.01
11	25.0	15.0	150	9.83
12	25.0	4.0	150	10.27
13	10.0	15.0	450	9.65
14	10.0	4.0	450	8.27
15	10.0	15.0	150	8.57
16	10.0	4.0	150	6.62
17	17.5	9.5	552	10.82
18	17.5	9.5	48	9.51

Note: *R* is ratio of ethanol-to-DOPM, *T* is extraction time, and *S* is speed of stirrer.

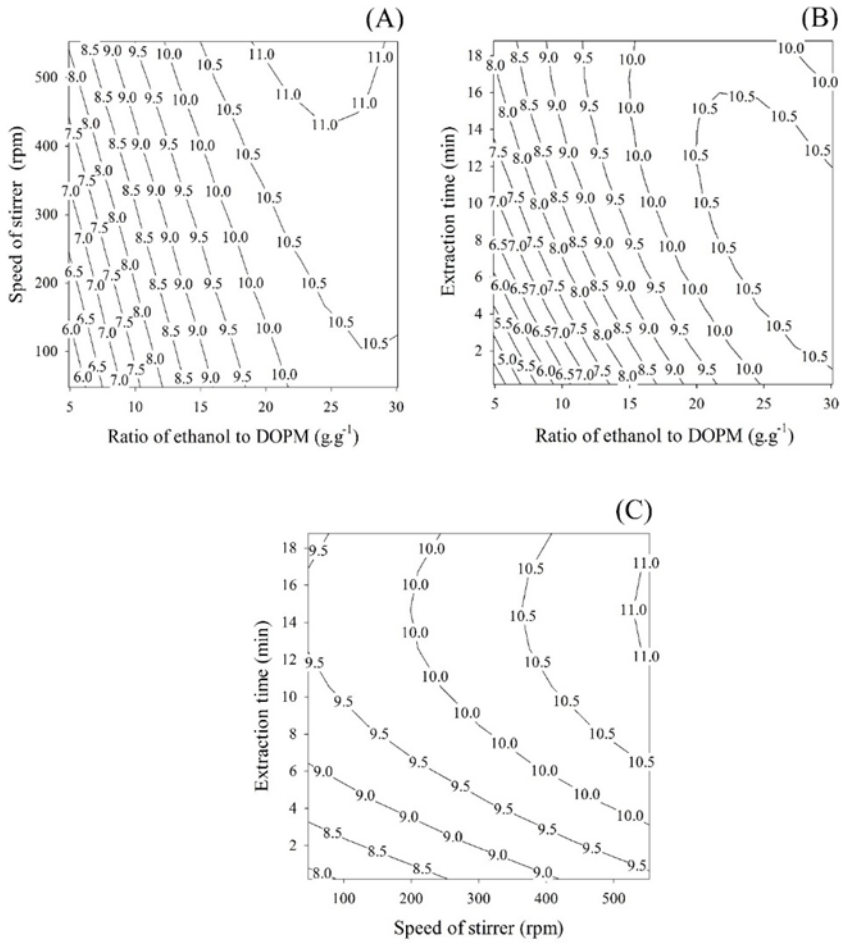

Fig 2. Contour plots of oil yield extracted from DOPM by using hydrous ethanol; (A) ratio of ethanol-to-DOPM and speed of stirrer, (B) ratio of ethanol-to-DOPM and extraction time, and (C) speed of stirrer and extraction time.

Table 3. Coefficients, *p*-values, and ANOVA for the fitted model.

Coefficient			For hydrous ethanol		
			Value		*p*-value
β_0			-0.23443		0.7474383
β_1			0.60245		0.0000005
β_2			0.41659		0.0000321
β_3			0.00587		0.0023066
β_4			-0.01036		0.0006849
β_5			-0.00016		0.0658556
β_6			-0.00859		0.0000356
β_7			-0.00804		0.0053792
ANOVA					
Source	SS	MS	F_0	F_{signif}	DOF
Regression	29.14	4.163	66.62	0.00000014	7
Residual	0.625	0.06250			10
LOF Error	0.617	0.08818	34.3565	0.00729284	7
Pure Error	0.00770	0.00257			3
Total	29.77				17
R^2 = 0.979, and $R^2_{adjusted}$ = 0.964					

Note: R^2 is coefficient of determination, $R^2_{adjusted}$ is adjusted coefficient of determination, *p*-value is an indicator of statistical significance, DOF is degrees of freedom, SS is sum of squares, and MS is mean square.

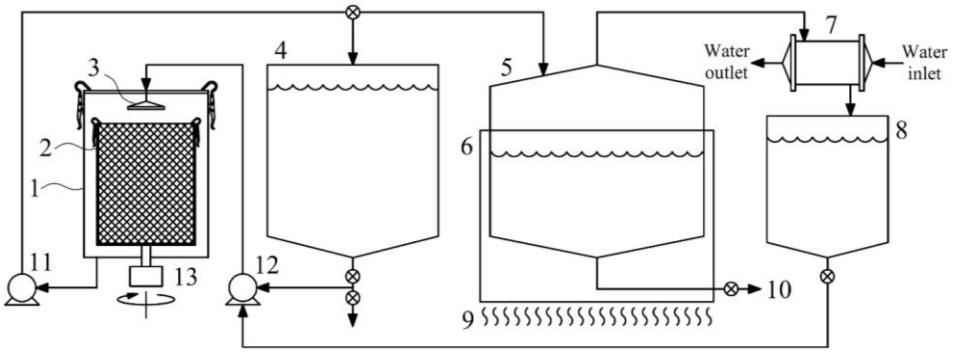

Fig 3. Schematic diagram of prototype-scale oil extraction from DOPM**.**
(1**:** extraction tank, 2**:** rotating drum stainless steel sieve, 3**:** spray head, 4**:** miscella tank, 5**:** distillation tank, 6**:** hot water tank, 7**:** shell and tube heat exchanger, 8**:** solvent tank, 9**:** heat source, 10**:** EDOPM outlet, 13**:** motor, and 11, 12**:** circulating pumps**).**

Table 4. Operating conditions for and composition in EDOPM from oil extraction with hydrous ethanol.

Parameter or component	Optimal condition	Recommended condition
Condition		
Ratio of ethanol-to-DOPM ($g.g^{-1}$)	23.2	20.1
Extraction time (min)	11	11
Speed of stirrer (rpm)	552	300
Yield of oil		
Predicted oil yield (wt.%)	11.24	10.50
Actual experiment yield (wt.%)	10.81	10.27
Composition of EDOPM		
Free fatty acid (wt.%)		1.19
Triglyceride (wt.%)		13.57
Diglyceride (wt.%)		2.36
Monoglyceride (wt.%)		82.88
Ester (wt.%)		-
Higher heating value ($kJ.kg^{-1}$)		35111.3
Density at 30°C ($kg.L^{-1}$)		0.910

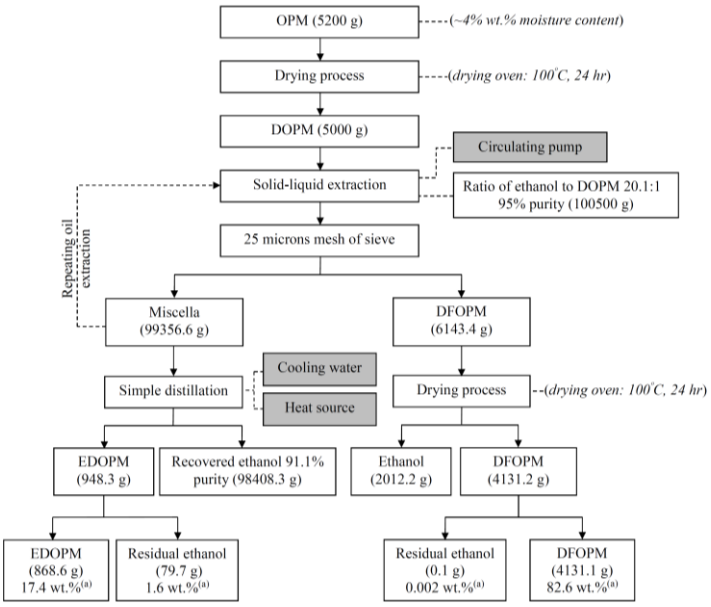

Fig 4. Material balance of oil extraction from DOPM. (Note: [(a)]Yield of products (wt.%) = the weight of product (g) / the weight of DOPM (g). The yields are relative to 100 wt.% of DOPM.).

Table 5. The fatty acid profile in EDOPM.

Common name	Fatty acid	Content (wt.%)
Caprylic acid	C8:0	0.95
Nonanoic acid	C9:0	0.43
Capric acid	C10:0	1.26
Undecanoic acid	C11:0	0.09
Lauric acid	C12:0	18.32
Tridecanoic acid	C13:0	0.09
Myristic acid	C14:0	7.36
Pentadecanoic acid	C15:0	0.11
Palmitic acid	C16:0	40.50
Palmitoleic acid	C16:1	0.22
Heptadecanoic acid	C17:0	0.13
Stearic acid	C18:0	4.63
Oleic acid	C18:1	11.44
Linoleic acid	C18:2	0.94
Alpha linolenic acid	C18:3	0.02
Arachidic acid	C20:0	0.38
Paullinic acid	C20:1	1.02
Behenic acid	C22:0	0.11
Erucic acid	C22:1	0.00
Lignoceric acid	C24:0	0.17
Nervonic acid	C24:1	0.00

Note: The EDOPM was obtained from oil extraction at recommended operating conditions.

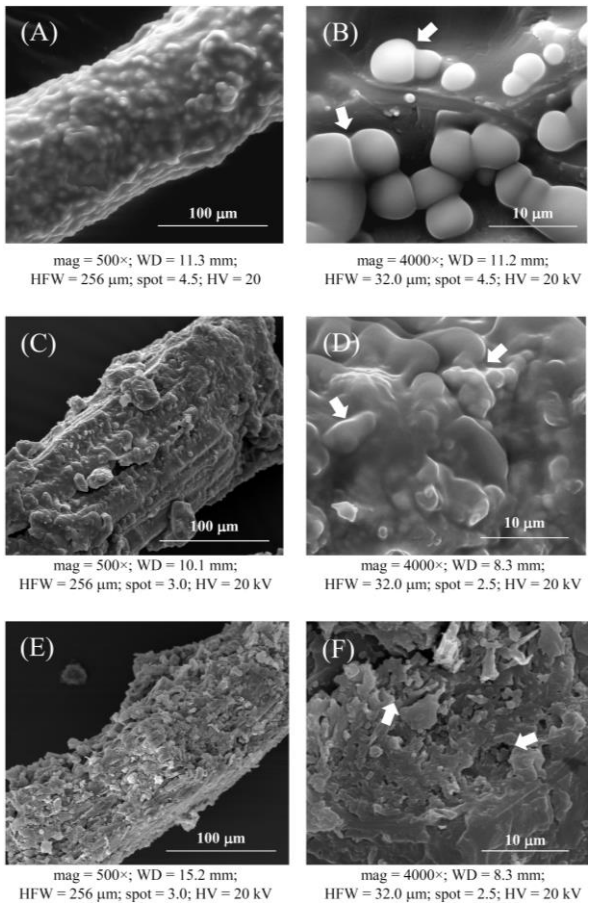

Fig 5. Scanning Electron Microscopy images; roasted palm fruit at magnifications of (A) 500×, (B) 4000×; (C) oil palm meal at magnifications of 500× and (D) 4000×; and (E) defatted oil palm meal at magnifications of 500× and (F) 4000×.

Table 6. Factor levels for the manipulated variables in oil extraction experiments.

Independent variable	Coded level				
	-1.682	-1	0	+1	+1.682
R : Ratio of ethanol-to-DOPM ($g.g^{-1}$)	4.9	10.0	17.5	25.0	30.1
T : Extraction time (min)	0.2	0.4	9.5	15.0	18.8
S : Speed of stirrer (rpm)	48	150	300	450	552

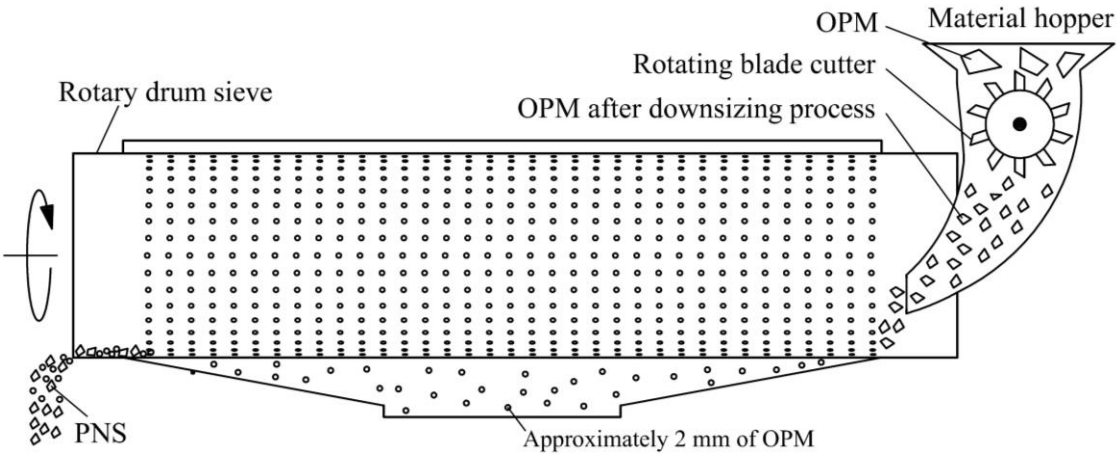

Fig 6. Cross-section of a palm nut shell–oil palm meal separator**.**

Fitted model for oil extraction in laboratory-scale

Regression analysis was used to fit models from eighteen experiments with a *Microsoft Excel add-in tool*, as detailed in Table 3. The relationship between oil yield (Y_o) and the three independent variables (ratio of ethanol-to-DOPM (*R*), extraction time (*T*), and speed of stirrer (*S*) was fitted with the quadratic model in Eq. (1). The goodness of fit was assessed from the coefficient of determination (R^2), the adjusted coefficient of determination ($R^2_{adjusted}$), the probability of error (*p*-value), and the coefficients in the model, as reported in Table 3. The *p*-value is mainly a measure for the statistical significance of each term in the fitted model, with smaller value indicating higher significance. The least *p*-value 0.0000005 occurred for the term $\beta_1 R$ when hydrous ethanol was used, while the next ranked 0.0000321 was for the term $\beta_2 T$. The analysis of variance (ANOVA) of the fitted model for ethanol extraction is shown in Table 3.

$$Y_o = \beta_0 + \beta_1 R + \beta_2 T + \beta_3 S + \beta_4 RT + \beta_5 RS + \beta_6 R^2 + \beta_7 T^2 \quad (1)$$

Where; Y_o is the yield of oil; *R* is the ratio of ethanol-to-DOPM; *T* is the extraction time; *S* is speed of stirrer; and β is a coefficient.

Contour plots

Figs. 2A, 2B, and 2C show the relationships between the dependent (yield) and independent variables (ratio of ethanol-to-DOPM, extraction time, and speed of stirrer), presented as contour plots. The effects of the ratio of ethanol-to-DOPM and speed of stirrer on the oil yield is presented in Fig. 2A. The oil yield contour is of haft-elliptical shape for stirrer speed in the range from 440 rpm to 552 rpm and for the ratio of ethanol-to-DOPM from 19 to 29 $g.g^{-1}$. It is clear that the oil yield increased with each of these two parameters. The interaction of these two independent variables is also of importance. This condition is consistent with the report by Abed et al. (2015). They reported that the speed of stirrer improved mass transfer and eddy diffusion in the solvent at the particle surfaces. Moreover, increasing the ratio of solvent-to-solid will increase the oil yield, up to some limit, because the concentration gradient in liquid phase increases as dissolved substances are diluted, and this favors good mass transfer (Abed et al., 2015). Fig. 2B shows the effects of extraction time and ratio of ethanol-to-DOPM. High oil yield from DOPM takes place in the range of extraction time from 1 to 16 min and the ratio of ethanol-to-DOPM from 20 to 30 $g.g^{-1}$. Thus, oil could be extracted with a short extraction time when a high ratio of ethanol-to-DOPM is used. The interaction of extraction time and the ratio of ethanol-to-DOPM is also of importance. Fig. 2C shows the effects of extraction time and speed of stirrer in a contour plot. High oil yield is obtained with stirrer speeds over 520 rpm and extraction times from 12 to 18 min. The results show some interaction of these variables on the right side of the contour plot, although that is not highly significant for oil extraction from DOPM. Similar results were reported by Suganya et al., (2012), who optimized oil extraction from marine macroalgae *Ulva lactuca* and the high 500 rpm stirrer speed was beneficial. This is clearly revealed that mass transfer rate plays a major role during solvent extraction (Suganya et al., 2012). Udachan and Sahoo (2014) studied the parameters affecting solvent extraction of lactic acid from fermentation broth. They concluded that the extraction efficiency was little affected by speed of stirrer and extraction

time (Udachan and Sahoo, 2014). Maximizing the oil yield was done numerically using the fitted model with Excel solver *add-in tool*, and the results are shown in Table 4. These optimal conditions based on the regression model were then tested experimentally, and the oil yields were 10.50 wt.% and 10.27 wt.% from the model and experimentally, respectively.

Recommended operating conditions in laboratory-scale

The maximum yield of oil from extraction is the primary consideration. The maximum 10.81 wt.% oil yield was achieved when using hydrous ethanol under these conditions: 23.2 $g.g^{-1}$ ratio of ethanol-to-DOPM, 11 min extraction time, and 552 rpm speed of stirrer. The residual oil in DFOPM was analyzed using the Soxhlet apparatus with a standard procedure (ASTM D5369-93(2008)e1, 2018) using hexane, and 4.82 wt.% oil was found in the DFOPM. Regarding residual oil in diet of ruminants, Peixoto et al. (2017) reported that the total amount of oil in the diet of ruminants is less than 5%, and oils have a very important role in the energy metabolism of these animals because of the high content of fatty acids (Peixoto et al., 2017). Thus, the residual oil in DFOPM at 5% level is acceptable for animal feed. The estimated oil yield (wt.%) was substituted into Eq. (1) for the response variable (Y_o), and the three control variables (ratio of ethanol-to-DOPM ($g.g^{-1}$), extraction time (min), and speed of stirrer (rpm)) were determined using the Excel solver. These variables were constrained to stay within the experimental ranges in oil extraction. The final DFOPM was analyzed by Soxhlet extraction to find conditions with the residual oil in DFOPM close to 5 wt.%, and the operating conditions found were: 20.1 $g.g^{-1}$ ratio of ethanol-to-DOPM, 11 min extraction time, and 300 rpm speed of stirrer. This operating point is recommended to meet the requirements for total amount of lipid in the diet of ruminants, and this left the least amount of oil in the diet of ruminants as requested by Peixoto et al. (2017). The consumption of ethanol solvent decreased significantly from ethanol-to-DOPM ratio 23.2 $g.g^{-1}$ (optimal conditions) to 20.1 $g.g^{-1}$ (recommended conditions). The composition of extracted oil was 1.19 wt.% FFA, 13.57 wt.% TG, 2.36 wt.% DG, and 82.88 wt.% MG, as shown in Table 4. Regarding the oil quality, FFA content in oil is used to define its quality (Kardash and Tur'yan, 2005). Slightly over 1 wt.% FFA content in the EDOPM was measured, and this should be reduced to less than 1 wt.% level with acid-catalyzed esterification, followed by base-catalyzed transesterification if biodiesel is produced (Abbaszaadeh et al., 2012; Somnuk et al., 2013; Somnuk et al., 2019). Moreover, the oil extraction using ethanol without the requirement for solvent evaporation, which can be directly converted to ethyl esters to reduce the overall cost of biodiesel production (Larissa et al., 2017). The use of ethanol as solvent in prototype scale extraction with recirculation of extraction solvent is described next.

Oil extraction from DOPM in prototype scale with recirculation of miscella

Fig. 3 is a schematic diagram of prototype scale oil extraction from DOPM with recirculation of extraction solvent. The recommended conditions from laboratory-scale experiments were used in prototype scale. Approximately 5000 g DOPM was dumped into the rotating drum stainless steel sieve (approximately 16.3 L with #500 sieve number corresponding to 25 micron size), which was fitted inside a cylindrical extraction tank (400 mm diameter and 600 mm height). The lid of the extraction tank was closed and locked. Fresh ethanol in the solvent tank was fed by pump and sprayed on the DOPM in extraction tank. Two circulating pumps were turned on to circulate the ethanol through DOPM in the extraction tank, and the timer was started. The hydrous ethanol was kept circulating through the rotating drum stainless steel sieve until completion of oil extraction at 11 min duration, following the recommended conditions. The miscella in the extraction tank was then fed into the miscella recirculation tank by a pump. Most miscella had already flowed through the rotating sieve into the recirculation tank. The residual miscella in soaked DOPM was further centrifuged using the rotating drum stainless steel sieve, driven by a motor at 1425 rpm. The recirculated miscella (RM) in tank was reused in the next batch, and so on, to test it as an alternative for fresh ethanol in each extraction. For this, the DFOPM in the rotating sieve was dumped and fresh DOPM was then loaded into the drum sieve for the next cycle of oil extraction. The RM was then fed by pump and sprayed on the fresh DOPM. After the second cycle of oil extraction was complete, the RM was fed into the recirculation tank. Oil extraction batches were run using RM until the efficiency of oil extraction had decreased significantly. After the oil extractions, the clear yellow miscella was pumped into the distillation tank. It was heated with hot water to separate the solvent from the oil by simple distillation. The operator can collect the EDOPM from the bottom of distillation tank. Regarding solvent recovery, the ethanol vapor from distillation was condensed in a water-cooled shell-and-tube heat exchanger. The recovered ethanol can be used again as oil extraction solvent.

Results of oil extraction from DOPM in prototype scale

According to the lab-scale results, approximately 10.5 wt.% oil can be extracted from DOPM under the recommended conditions: 20.1 $g.g^{-1}$ ratio of ethanol-to-DOPM, 11 min extraction time, and 300 rpm speed of stirrer. Tables 4 and 5 show the conditions and compositions in EDOPM from oil extraction with hydrous ethanol, and the fatty acid profile in EDOPM, respectively. The EDOPM was found to contain 40.50% palmitic acid, followed by lauric, oleic, and myristic acids at 18.32, 11.44, and 7.36%, respectively. As an alternative to fresh ethanol, recirculation of miscella (RM) was tested for multiple cycles. The RM, as a solution of accumulated oil from DOPM, was repeatedly used to extract oil from fresh DOPM for up to six cycles. The accumulated oil was observed after each completed oil extraction, and the yellow color intensity of RM increased gradually with EDOPM concentration in RM as extractions were run. The final RM was separated by simple distillation. The oil contents from first to sixth cycle were 0.7 wt.%, 1.36 wt.%, 2.0 wt.%, 2.53%, 2.94 wt.%, and 3.07 wt.%., so that the accumulation of oil in each cycle decreased as 0.7%, 0.66%, 0.64%, 0.53%, 0.41, and 0.13%, showing a clear drop after the fourth cycle. Thus, the use of hydrous ethanol (95 wt.% purity) for oil extraction with recycling of miscella should be limited to four cycles, each with fresh DOPM. As the efficiency of oil extraction decreased with cycle count, also the residual oil in DFOPM exceeded the acceptable 5% level. The recirculation of miscella is worthwhile, because it reduces ethanol consumption and the energy consumed in distillation to separate the EDOPM. The

decreasing efficiency of oil extraction is also consistent with that reported by Neto et al., (2018). They concluded that the driving force in ethanol solvent decreased when the concentration of oil in miscella increased, and became zero at the saturation concentration (Neto et al., 2018).

Material balance of prototype-scale of oil extraction

Fig. 4 shows the material balance in prototype-scale oil extraction from DOPM using recirculation of extractant. The recommended conditions were used to process 5000 g of DOPM in prototype scale extraction using the rotating drum stainless steel sieve. After extraction, 99356.6 g of miscella and 6143.4 g of DFOPM were obtained. The DFOPM was oven dried at 100^{o}C for 24 hr, and ethanol recovery of 2012.2 g was attained. The residual ethanol at 0.1 g in DFOPM after drying was detected using GC–FID. Regarding the flow of miscella, the alternative solvent by recirculation of miscella (RM) was tested in multiple solid-liquid extractions. The RM was repeatedly used to extract oil from fresh DOPM. However, the material balances showed the weight of miscella from a single extraction batch compared with 100 wt.% of DOPM. Therefore, the first cycle of clear yellow miscella was pumped into the distillation tank. It was heated by hot water to separate the solvent from the oil by simple distillation. EDOPM at 948.3 g and recovered ethanol at 98408.3 g were obtained with the distillation process. The residual 79.7 g of ethanol (1.6 wt.%) was detected using GC–FID relative to 100 wt.% of DOPM. In summary, distillation gave as yield 868.6 g extracted oil of dried oil palm meal (EDOPM, 17.4 wt.% oil yield) and recovery of hydrous ethanol (91.1 wt.%) was 98408.3 g of the initial 100500 g of ethanol. Residual ethanol at 0.002 wt.% was detected in DFOPM after oven drying at 104^{o}C for 24 hr. Regarding savings in the costs of solvent and energy, the recovered ethanol can be used again in a new cycle of oil extraction. Moreover, the recirculation of miscella is worthwhile, because it reduces ethanol consumption and the energy consumed in distillation to separate the EDOPM.

Scanning electron microscopy images

Fig. 5 shows scanning electron microscopy (SEM) images of roasted palm fruit (RPF), dried oil palm meal (DOPM) and defatted oil palm meal (DFOPM). Before oil extraction from RPF by a mechanical screw press, the fiber in RPF appears to have larger oil droplets than in DOPM, as shown in Figs. 5A (at mag. 500×) and 5B (at mag. 4000×), respectively. Arrows point to the larger oil droplets. After the mixed crude palm oil milling by mechanical screw pressing, most of the crude palm oil had been extracted from palm fruit. DOPM fibers are short. It was dried to remove moisture for preparing fibrous DOPM for oil extraction with solvent. There may be some residual oil on the DOPM fibers in Figs. 5C (at mag. 500×) and 5D (at mag. 4000×). Arrows point to the residual oil droplets on DOPM fibers. After extraction with solvent, DFOPM appears to have smoother surfaces than DOPM, as shown in Figs. 5E (at mag. 500×) and 5F (at mag. 4000×). The oil drops were apparently extracted from DFOPM fiber surfaces (arrows). The oils were drawn out of the DFOPM by solvent extraction with ethanol.

Materials and methods

Raw materials

The OPM obtained from a small-scale palm oil producer in Southern Thailand was used to extract the residual oil with a solvent, as shown in Fig. 1A. Larger palm nut shell (PNS) particles are found along with OPM, seen in Fig. 1B. Before solvent extraction, the approximately 2 mm size PNS was first removed from OPM using a palm shell–oil palm meal separator (as described in the section of preparing fine OPM for oil extraction process), giving the cleaned intermediate product in Fig. 1C. The OPM was oven dried at 104^{o}C for 24 hr to ensure similar moisture in the raw materials across all experiments. The moisture content of fresh OPM is approximately 4 wt.%. The dried oil palm meal (DOPM) with particles smaller than 2 mm was used as the raw material for solvent extraction. Fig. 1D shows defatted oil palm meal (DFOPM) after solvent extraction. Commercial grade 95 vol.% ethanol (hydrous ethanol) was used to extract the oil. Table 1 shows physical and chemical properties of DOPM, DFOPM and hydrous ethanol.

Preparing fine OPM for oil extraction process

A palm nut shell–oil palm meal separator was specially designed by Mechanical Engineering Department, PSU. This machine was constructed to separate the larger particle size palm nut shell (PNS) from OPM. Thus, particles coarser than 2 mm of PNS were held by a rotary drum sieve (#10 mesh) to remove most of the PNS in DOPM. The rotary drum sieve (265 mm in diameter and 1200 mm in length) was constantly rotated at 25 rpm. To prepare fibrous OPM for oil extraction with solvent, fresh OPM from the mechanical screw press was dumped into a material hopper and then chopped by a rotating blade cutter. The comminuted OPM was dropped on the rotary drum sieve, and the approximately 2 mm size OPM was discharged and collected. The PNS exited at the end of the rotating mesh sieve, as shown in Fig. 6. After this pre-processing, the small particle size OPM was used as raw material for solvent extraction of oil.

Laboratory scale oil extraction from DOPM

The experiments used laboratory scale batchwise oil extraction. First, 10 g of DOPM was loaded into a 400 mL beaker on a 0.0001 g resolution digital analytical balance (AL204 Mettler-Toledo, Switzerland). The required amount of ethanol for desired ethanol-to-DOPM (g.g^{-1}) ratio was then poured into the beaker. To prevent solvent evaporation, aluminum foil was wrapped over the beaker. DOPM in ethanol solvent was constantly mixed with a stirrer at 30^{o}C for the designated extraction time. The suspension was filtered through a filter paper (W. & R. Balston Ltd. Genuine Whatman No. 1) to separate the defatted oil palm meal (DFOPM) from the miscella (the solution of EDOPM dissolved in ethanol). The ethanol and EDOPM in miscella were then separated by simple distillation. The extracted oil was kept at 104°C for 6 h to ensure that no residual solvent remains in the EDOPM (Abdullah and Koc, 2013).

Analysis methods

The compositions of DOPM and DFOPM solid biofuels were analyzed using CHNS–O analyzer (CE Instruments Flash EA 1112 Series, Thermo Quest, Italy) under ASTM D 4239, (ASTM D4239-17, 2018) and EN 15104, (BS EN 15104:2011, 2018) for carbon (C), hydrogen (H), nitrogen (N), sulfur (S), and oxygen (O). The nutritional compositions were analyzed in terms of ash (AOAC 942.05, gravimetric method), protein (AOAC 984.13, Kjeldahl method), and crude fiber (ANKOM^{200} Fiber Analyzers). The fatty acid profiles were analyzed using a GC–FID (GC 6890, Hewlett Packard, USA). The percentages of lignin in DOPM and DFOPM were analyzed using a gravimetric method with a digital analytical balance (AT200, Mettler Toledo, USA). The residual ethanol in EDOPM and DFOPM was detected using GC–FID (GC 6850, Hewlett Packard, USA). The higher heating values (HHV) of the products DOPM, DFOPM, and oil, were measured using a calorific method (IKA ® Calorimeter System C5000 control, Germany). A thin layer chromatograph with flame ionization detection (TLC/FID, IATROSCAN MK-65, Mishubishi Kagahu Latron Inc.; Japan) was used to analyze the percentages of triglyceride (TG), diglyceride (DG), monoglyceride (MG), and free fatty acid (FFA) in the oil. The fiber surfaces of DOPM and DFOPM were imaged using a scanning electron microscope (SEM). The oil yield was calculated using Eq. (2) (Yunos et al., 2017)

$$Y_o = \left(W_o / W_d\right) \times 100 \quad (2)$$

Where; Y_o (wt.%) is the yield of EDOPM; W_o (g) is the weight of EDOPM; and W_d (g) is the weight of DOPM.

Experimental design and statistical analysis

The response surface methodology (RSM) approach, with 5-level and 3-factor central composite design (CCD), was used to optimize the oil extraction from DOPM. Multiple regression was used to fit second-order polynomials that predict the oil yield. A general second-order polynomial is shown in Eq. (3) (Rashid et al., 2018).

$$Y = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \beta_{ii} x_i^2 + \sum_{i=1} \sum_{j=i+1} \beta_{ij} x_i x_j + \varepsilon \quad (3)$$

Where; Y is the response variable; x_i and x_j are the uncoded independent variables; β_0, β_i, β_{ii}, and β_{ij} are the constant coefficients for intercept, linear, quadratic, and interaction terms, respectively; k is the number of variables; and ε is the error.

The three manipulated parameters, first studied in the batch mode, were ratio of ethanol-to-DOPM (R), extraction time (T), and speed of stirrer (S), which were to be optimized to maximize the oil yield from oil extraction. The axial parameter for rotatability is expressed in Eq. (4) (Abdelhafez et al., 2016). In this three-independent-variable case, the axial parameter (α_X) is 1.682 (for rotatable CCD). The five factor levels are coded as -1.682, -1, 0, +1, and +1.682 in the ranges of independent variables (ratio of ethanol-to-DOPM, g.g^{-1}; extraction time, min; speed of stirrer, rpm) in the experimental design, as shown in Table 6. The value 1.682 is chosen to fulfill the rotatability in the design of experiments. The central composite design had totally eighteen experiments, summarized in Table 2.

$$\alpha_x = \sqrt[4]{2^k} \quad (4)$$

Where α_x is the axial parameter for rotatability, and k is the number of variables.

Conclusions

Oil extraction from OPM produces defatted oil palm meal (DFOPM) as side product and this is promising as potential animal feed. The oil from OPM can be used in biodiesel production as a biofuel source, or as an additive in animal feed. In laboratory-scale oil extraction experiments, the yield was 10.27 wt.% under these recommended conditions: 20.1 g.g^{-1} ratio of ethanol-to-DOPM, 11 min extraction time, and 300 rpm speed of stirrer. The recommended conditions were used to process 5000 g of DOPM in prototype scale extraction experiments with recirculation or extraction solvent. The recirculation of miscella (RM) was used repeatedly to extract oil from fresh DOPM, reducing fresh solvent consumption in oil extraction and energy consumption in the distillation to separate the oil from the ethanol in RM. The residual oil in OPM was reduced by solvent extraction converting it to DFOPM, which should have less tendency to rancidity and longer shelf-life than OPM.

Acknowledgments

This work was supported by the Energy Policy and Planning Office of Thailand (EPPO); and Prince of Songkla University under Grant number NAT581217a.Special thanks go to a lecturer Assoc. Prof. Seppo Karrila, Ph.D. for correcting the English of this paper.

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