

## Research Article

# Vegetable oils deacidification using short path molecular distillation: Modeling and simulation

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### Abstract

Physical refining of vegetable oils aims to produce healthier oils, minimize excessive oil loss, and reduce production waste. Deacidification, or the removal of free fatty acids (FFA), is a crucial step in this process. This study aimed to develop a mathematical model that shows the deacidification behavior of cold-pressed camelina oil and lampante olive oil using short-path molecular distillation (SPMD) as a solventless and green technology. A mass conservation balance along with the Langmuir equation was used to describe the evaporation and separation mechanisms. The model's predictive capabilities were evaluated for oil samples at different feed flow rates ( $Q$ ) and evaporation temperatures ( $ET$ ), while other factors—including feed temperature, vacuum pressure, condensation temperature, and wiper speed—were kept constant. The group contribution method was applied to predict vapor pressure. The model predictions showed reasonable agreement with the experimental data, indicating that higher evaporation temperatures ( $ET$ ) and lower feed flow rates ( $Q$ ) led to greater deacidification efficiency. Furthermore, the proposed model can be used to determine FFA concentration at different longitudinal sections of the SPMD column.

**Keywords:** Short path molecular distillation (SPMD), Deacidification, Mathematical modeling, Simulation, Lampante olive oil, Camelina oil.

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## Nomenclature

$A_{1k}, B_{1k}, C_{1k}, D_{1k},$ $A_{2k}, B_{2k}, C_{2k}, D_{2k}$	Group contribution equations adjusted parameters	Greek symbols	
$D$	Distillate flow rate, mL/min	$\delta$	Film thickness, m
$ET$	Evaporation temperature, °C	$\mu$	Dynamic viscosity, N.s/ m <sup>2</sup>
$FFA$	Free fatty acids	$\nu$	Kinematic viscosity, m <sup>2</sup> /s
$H$	SPMD coefficient, kg <sup>1/2</sup> °C/ s <sup>2</sup>	$\pi$	The number pi, 3.14
$h$	Evaporation coefficient, s <sup>1/2</sup> (kg. kmol.°C) /m	$\rho$	Density, kg /m <sup>3</sup>
$J$	Evaporation flux, kg/(m <sup>2</sup> s)	<i>Subscripts</i>	
$L$	SPMD length, m	$i$	Component i
$M$	Average molecular weight of feed, kg/kgmol	$k$	Group k in i
$m$	Mass flow rate of feed, kg /m	$z$	Cartesian z coordinate
$m^o$	Mass flow rate of volatile compounds, kg /m		
$M^o$	Average molecular weight of volatile compounds, kg/kgmol		
$M_i$	Fatty acid molecular weight		
$N_k$	The number of group k in the molecule		
$P$	Total vapor pressure, Pa		
$P^o$	Vapor pressure of the volatile compound, Pa		
$Q$	Feed flow rate, mL /min		
$R$	Residue flow rate, mL /min		
$r$	Evaporator radius, m		
$s$	Cross-sectional area of falling film, m <sup>2</sup>		
$SPMD$	Short path molecular distillation		
$TG$	Triglycerides		
$v_z$	Feed velocity at direction z, m /s		
$wt\%$	Weight percent		
$Y_z$	Weight fraction of volatile compounds		

## 1. Introduction

Edible oils are refined so that disagreeable matters including oxidation compounds, free fatty acids (FFA), and phosphatides are eliminated. Deodorization or steam distillation, is a common physical refining process and serves as the final and most critical stage, typically occurring at around 240 °C [1,2]. However,

prolonged exposure to high temperatures can accelerate spoilage and rancidity [3]. It has been proved that removing free fatty acids (FFA) from oil improves its organoleptic properties [3–5]. A high FFA content can contribute to an undesirable odor in vegetable oils and is considered a crucial factor affecting both quality and process efficiency [1,2,6]. To meet edible oil standards, it is essential to reduce elevated FFA



levels [3,7]. The maximum allowable FFA content in cold-pressed and virgin oils is 2.0 wt% [8], while for refined olive oil, the recommended limit is 0.3 wt% [9].

Compared to triglycerides (TG), FFA are more volatile. Due to environmental concerns, recovering and recycling vegetable oil waste with high FFA content remains a significant challenge. Moreover, conventional deacidification methods have notable drawbacks, including excessive oil loss and the need for intensive wastewater treatment, especially at an industrial scale [5,10,11]. Regularly, FFA are primarily removed through alkali refining or alkali neutralization. The remaining FFA are further separated during the final steam deodorization stage. In the standard physical steam stripping deodorization process, high temperatures and low pressures are applied, allowing FFA molecules to evaporate along with other volatile and odorous compounds [7,12,13]. The financial aspect of vegetable oil production is substantially influenced by the deacidification process [4]. Enzymatic deacidification of vegetable oils, such as corn oil, has already been investigated [11,14,15]. Adsorption and solvent extraction are other techniques used for deacidification [16,17]. Additionally, deacidification of olive oil using charcoal or activated carbon provides an alternative method that avoids the use of chemical agents [18]. Membrane technology also presents new opportunities in the refining and deacidification of vegetable oils [19]. The elimination of free fatty acids (FFA) from palm oil using anion exchange resins is another viable alternative [20,21]. FFA can also be separated through physical refining processes, including SPMD [7,16]. Olive oil is usually treated by physical procedures [22].

Camelina (*Camelina sativa L.*) is gaining increasing global recognition [23–25] as an oil seed crop with a vast record of farming. The camelina oil market is steadily expanding [26]. Camelina oil

is emerging as an important vegetable oil, containing a significant amount of unsaturated fatty acids, including 30 to 40 wt% alpha-linolenic acid and 15 to 25 wt% linoleic acid. Its unique fatty acid profile positions camelina oil as a potentially suitable option for a wide range of bio-based markets [27–33].

Olive oil is renowned for its high oleic acid content, exceptional thermal stability, and distinctive taste and aroma [34]. Due to its high content of monounsaturated fatty acids and the presence of beneficial compounds such as squalene, vitamins, sterols, and antioxidants like tocopherols, olive oil continues to experience high demand [35,36]. In the Mediterranean basin, virgin olive oil is the main oil source and its economic impact is significant [37,38]. Olive oil production in the 2021/22 crop year was over 3 million metric tons [39]. Iran is among the 24 major olive-producer countries [40]. Virgin olive oils are classified and evaluated based on their acidity, and when the FFA content exceeds 3.3 wt%, the oil is classified as lampante olive oil. Lampante olive oil, being a non-edible type, must undergo refining to become suitable for consumption.

Short-path molecular distillation (SPMD) is recognized as a physical separation procedure with applications in the food, pharmaceutical, and petrochemical industries. Recently, research in this area has been grown progressively [41,42]. The separation of high boiling point and heat-sensitive constituents is a primary application of SPMD [43,44]. One of the key uses of SPMD in the food industry is vegetable oil refining [41,42]. The SPMD process involves brief exposure of the feed to high temperatures and high vacuums on the evaporator surface, which is positioned close to the condenser [45].

Some value-added compounds in the food, chemical, and pharmaceutical industries, such as omega 3, vitamins, essential oils, bio-compounds, fragrances, and flavors are heat-sensitives [43]. These compounds are prone to damage when

exposed to thermal stress, resulting in thermal decomposition, polymerization, and side reactions that negatively affect the quality of the final product. By significantly reducing the operating pressure, the distillation temperature and duration of thermal stress on the materials can be minimized, leading to a significant improvement in product quality [41,46,47].

In short-path molecular distillation (SPMD), the feed flows along the wall of the evaporator as a thin fluid film from the supply point to the discharge outlet. The residence time in the system is very short, and the period of heat stress is minimal. An innovative wiping mechanism inside the evaporator ensures ideal mixing and even distribution of the feed as it flows downward. This continuous movement brings volatile molecules to the film surface. Thus, the volatile molecules vaporize more effectively. Heating in the evaporator is achieved using a heat transfer medium, such as thermal oil or steam [42]. A distinctive feature of this technology is the condensation of the vaporized molecules on the condenser surface, which is located inside the SPMD chamber. The 'short path' of the vaporized molecules to the condenser results in minimal pressure loss. Therefore, in short-path molecular distillation (SPMD), distillation occurs at significantly lower temperatures compared to thin film evaporators. This method uses the mildest process parameters [48]. The effect of gas pressure on the evaporation and condensation rates has also been reported [49].

SPMD is an environmentally friendly technology in which volatile molecules are evaporated from a hot surface and move towards an adjacent condenser. The distance is highly evacuated, ensuring that the mean free path of the evaporated molecules is equal to or greater than the gap between the evaporator and the cold surfaces, minimizing energy consumption and reducing thermal degradation [50,51]. The mean free path, which can be determined by the kinetic

theory of gases, is a key factor in the design of SPMD. In a study, the mean free paths of molecules were [52]. When the vacuum pressure in the distillation gap is reduced to below 10 mbar, undesired intermolecular collisions are largely prevented. By conducting distillation under decreased pressure, the boiling points of volatile compounds are lowered, allowing for the recovery of high boiling point chemicals that might otherwise undergo thermal degradation. This process ensures that sensitive compounds are preserved during distillation [6,41,42]. In a study, SPMD conditions were optimized to increase oleic acid purity in a fatty acid mixture [53]. In another study, SPMD parameters were optimized for treating olive pomace oil to remove squalene while preserving tocopherol content [54]. Additionally, several studies have focused on tocopherol recovery from soybean oil deodorizer distillate [55,56].

SPMD designs could be classified as falling film or centrifugal. In centrifugal type, the feed is conducted to the center of a rotating evaporator. In contrast, the falling film SPMD contains a cylindrical evaporator with an internal condenser. The feed enters at the top of the system [6] and flows downward under the influence of gravity, forming a thin film on the evaporator surface [42]. A high-viscosity feed demands a low feeding rate to generate a thin and homogeneous film. Wiper blades make working with high viscosity feeds more practical [57]. Depending on the design and manufacturer, the relative position of the condenser and evaporator can differ. In some models, the evaporator is located at the center, surrounded by the condenser. In some designs, the condenser is placed at the center, surrounded by the evaporator. The typical SPMD gap ranges from 10 to 50 mm [52]. The application of SPMD has been steadily expanding in recent years. It can also be used for the pre-refining of oils with high FFA content, reducing it to practical levels before



processes like steam distillation or alkali neutralization.

In SPMD, the key process factors include evaporation temperature, feed flow rate, feed temperature, wiper speed, and vacuum pressure [41,58,59]. To scale up the process, it is essential to describe the main phenomena at a specific scale (e.g., laboratory or pilot scale) using dimensional analysis. Given the significant role of mathematical modeling and simulation in characterizing the studied procedures, their impact on scaling up activities is undeniable [60]. Modeling and simulation of the transport phenomena occurring in SPMD has been a challenging objective. To derive the governing equations that represent the processes in SPMD treatment, it is essential to have a comprehensive understanding of the behavior of the treating compound molecules under different conditions [42]. Mathematical modeling and simulation are highly beneficial in this regard [44]. Furthermore, several studies have been conducted focusing on the mathematical modeling of SPMD [6,42,61]. Due to the design and operational similarities, analyzing and understanding wiped film evaporator technology proves useful in simulating and modeling SPMD. Successful mathematical modeling of SPMD requires incorporating parameters that account for both thermodynamics and transport phenomena. Vaporization enthalpy, vapor pressure, heat capacity, thermal conductivity, and density are examples of the essential properties required in SPMD modeling. However, it is often challenging to obtain the necessary properties [42]. Regarding mass balance, the primary objectives in SPMD modeling and simulation involve determining the compositions and flow rates of the distillate and residue streams.

Establishing the governing equations is a crucial step in this process, often leading to a set of partial differential equations. Solving these equations requires significant computational effort.

However, the complexity of the model can be significantly reduced by making certain assumptions. An SPMD model could incorporate the kinetic theory of gases to describe the evaporation rate and vapor phase behavior. For instance, Xubin *et al.* applied this theory to develop a model that predicts the concentration and temperature profiles of a binary mixture during the SPMD process [62]. Lutusan *et al.* conducted several studies in which they investigated the mass and energy balances of various compounds [63]. Vapor phase properties, thin film thickness, and mean free path are critical factors influencing SPMD performance. To develop an accurate simulation of the SPMD process, a deep understanding of the molecular behavior of different compounds under various conditions is essential [42]. The general mass and heat transfer within the evaporating film of SPMD has been studied [63]. The kinetic theory of gases can be used to describe the behavior of vapor phase molecules. Through conduction and diffusion mechanisms, the heat and mass transfer rates are influenced by the driving forces acting at the liquid-vapor interface [64]. Additionally, even minor deviations in the process parameters can lead to significant changes in the properties of the resulting phases. Therefore, it is crucial to develop accurate models that can help determine the optimized process conditions [65]. Compared to FFA, the vapor pressures of TG are much lower, meaning that TG does not evaporate within the concerned temperature range [66]. In this case, the volatile components (i.e., FFA) constitute a smaller fraction of the total mass. As a result, the non-volatile molecules, including TG, have a greater influence on the behavior of SPMD [67].

The aim of this study was to develop a mathematical model to predict the changes in FFA content during the deacidification of vegetable oils via SPMD, utilizing mass balance and evaporation rate equations. The simulated results were compared with experimental data

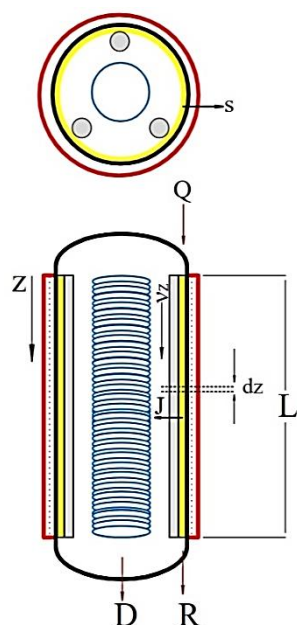
from the deacidification of cold-pressed camelina oil and lampante olive oil. Additionally, the developed model was used to determine the FFA content changes in the treated oils throughout the SPMD process. For vapor pressure calculations, the group contribution method was applied.

## 2. Mathematical model

Mass conservation balance and the Langmuir equation were employed to describe the changes in FFA content during the SPMD process. The schematic diagram and real photo of the studied SPMD chamber are presented in Fig. 1. To establish the mass balance, a volumetric element

with a thickness of  $dz$  and a cross-sectional area of  $s$  was selected. The following modeling assumptions were made:

1. Steady-state conditions,
2. Constant film thickness ( $\delta$ ) throughout the hot surface of the evaporator,
3. Negligible re-evaporation of condensed molecules from the condenser surface,
4. Using rectangular coordinates ( $\delta$  is small relative to the system diameter),
5. FFA content expressed as the dominant fatty acid of the oil. That is, the FFA of the camelina and olive oils samples were assumed as linolenic and oleic acids, respectively.



**Fig 1.** Schematic diagram and photo of the SPMD chamber: (D) distillate flow rate, (L) evaporator length, (Q) feed flow rate, (R) residue flow rate, (S) cross-sectional area at  $z$ , (J) evaporation flux, (S) film thickness, ( $v_z$ ) feed velocity at direction  $z$ .

Feed is maintained at a constant temperature and enters the evaporator, continuously flowing along the internal wall of the evaporator. The mass flow rates of the residue and distillate, denoted as  $R$  and  $D$  (in kg/h), respectively, are

considered. The overall mass balance can be written as follows:

$$Q = R + D \quad (1)$$

Due to the heat transfer from the hot surface of the evaporator, more volatile compounds, mainly FFA, evaporate to the adjacent cold condenser surface. The evaporation flux can be expressed by Langmuir equation as Eq. 2

$$J = hP \sqrt{\frac{M^0}{ET + 273.15}} \quad (2)$$

where J is the evaporation flux, h is the evaporation coefficient, P is the vapor pressure,  $M^0$  is the average molecular weight of the volatile compounds [6,68].

Assuming an ideal mixture of two compounds, the relative enrichment of the condensate will be  $(P_1 / M_1) \div (P_2 / M_2)$  instead of  $P_1 \div P_2$  [50]. The primary volatile compounds of feed are assumed to be FFA. Thus, the vapor pressure is dependent on the weight fraction of FFA (Y), average molecular weight of feed (M), average molecular weight of FFA ( $M^0$ ), and vapor pressure of the FFA ( $P^0$ ).

$$P \frac{m^0}{M^0} = P \frac{m}{M} \quad (3)$$

$$P = Y \frac{M}{M^0} P^0 \quad (4)$$

By substituting Eq. 4 in Eq. 2.

$$J = h Y \frac{M}{M^0} P^0 \sqrt{\frac{M^0}{ET + 273.15}} \quad (5)$$

By considering the element shown in Fig. 1, the mass balance can be set as Eq. 6.

$$\rho^0 v_z Y_z - \rho^0 v_{z+dz} Y_{z+dz} - 2 \pi r h Y_z \frac{M}{M^0} P^0 \sqrt{\frac{M^0}{ET + 273.15}} dz = 0 \quad (6)$$

Dividing by dz and following differentiation we get

$$\rho \frac{d(v_z Y_z s)}{dz} = -h \frac{2 \pi r M P^0}{\sqrt{M^0 (ET + 273.15)}} Y_z \quad (7)$$

Because the volatile molecules are a small fraction of the feed, velocity (v) and the elemental surface (s) are assumed to be constant. So,

$$\rho \frac{dY_z}{dz} = -h \frac{2 \pi r M}{v s \sqrt{M^0}} \frac{P^0}{\sqrt{(ET + 273.15)}} Y_z \quad (8)$$

Using h, r,  $\rho$ , v, s, M, and  $M^0$ , the H value is defined as

$$H = h \frac{2 \pi r M}{\rho v s \sqrt{M^0}} \quad (9)$$

Regarding the fact that all terms in Eq. 9 are constant and are dependent on the SPMD design and the feed characteristics, H can be assumed as the SPMD coefficient ( $\text{kg} \sqrt{^\circ\text{C}/\text{s}^2}$ ). By integrating Eq. 8 throughout the evaporation chamber we have:

$$\int_{Y_0}^{Y_z} \frac{dY_z}{Y_z} = -H \frac{P^0}{\sqrt{(ET + 273.15)}} \int_0^z dz \quad (10)$$

Eq. 10 is to be solved with the following boundary conditions:

$$\text{B.C. 1: at } z = 0; Y_z = Y \quad (\text{initial FFA content}) \quad (11)$$

$$\text{B.C. 2: at } z = L; Y_z = \bar{Y} \quad (\text{final or residue FFA content}) \quad (12)$$

$$Y_z = Y_0 \exp\left(-H \frac{P^0}{\sqrt{(ET + 273.15)}} z\right) \quad (13)$$

As previously mentioned, linolenic acid and oleic acid, the predominant FFAs in camelina and olive oils, were selected for vapor pressure calculations. Vapor-liquid equilibrium data is essential for designing processes and equipment, especially when dealing with mixtures containing trace amounts of compounds that need to be separated through distillation. Empirical or analytical models are beneficial for

accurately determining the required properties, including vapor pressure. A widely used approach for calculating physicochemical properties is the group contribution method. For predicting vapor pressure, the group contribution method proposed by Ceriani and Meirelles was applied. It can be used to determine the vapor pressure of key fatty compounds found in the edible oil industry. This method is particularly valuable for assessing distillation loss during the treatment of edible oils [69]. As Eq. 14 displays,

$$\ln P_i^\circ = \sum_k N_k \left( A_{1k} + \frac{B_{1k}}{ET^{1.5}} - C_{1k} \ln ET - D_{1k} ET \right) + \left[ M_i \sum_k N_k \left( A_{2k} + \frac{B_{2k}}{ET^{1.5}} - C_{2k} \ln ET - D_{2k} ET \right) \right] + 0.001(3.4443 - \frac{499.3}{ET^{1.5}} - 0.6136 \ln(ET) + 0.00517 ET) \quad (14)$$

**Table 1.** Parameters of Eq. 14.

Group	A <sub>1k</sub>	B <sub>1k</sub>	C <sub>1k</sub>	D <sub>1k</sub>	A <sub>2k</sub>	B <sub>2k</sub>	C <sub>2k</sub>	D <sub>2k</sub>
CH	-117.5	7232.3	-22.7939	0.0361	0.00338	-63.3963	-0.00106	0.000015
CH	8.4816	-10987.8	1.4067	-0.00167	-0.00091	6.7157	0.000041	-0.00000126
COOH	8.0734	-20478.3	0.0359	-0.00207	0.00399	-63.9929	-0.00132	0.000001
CH=cis	2.4317	1410.3	0.7868	-0.004	0	0	0	0

Using the experimental data including initial and final FFA contents, ET, the vapor pressure at ET (i.e., P<sup>0</sup>), and the SMPD apparatus length (L), the actual H value can be calculated as follows:

$$H = \frac{\sqrt{ET + 273.15}}{L P^\circ} \ln \frac{Y_0}{Y_L} \quad (15)$$

Regarding the fact that TG is not evaporated at the operating ETs, the distillates are assumed to be almost entirely FFA.

### 3. Experimental data

To validate the model, results from two published studies on the deacidification of cold-pressed camelina and lampante olive oils using SPMD were utilized [25,36]. Briefly, the trials were conducted using a pilot-scale SPMD unit made of borosilicate glass, designed by the Iranian

vapor pressure depends on evaporation temperature (ET). N<sub>k</sub> is the number of group k repetitions in the FFA chemical formula. M<sub>i</sub> is the molecular weight of compound i. A<sub>1k</sub>, B<sub>1k</sub>, C<sub>1k</sub>, D<sub>1k</sub>, A<sub>2k</sub>, B<sub>2k</sub>, C<sub>2k</sub>, and D<sub>2k</sub>, whose values for each group are given in Table 1, are factors attained by regression of the available experimental statistics. k denotes the groups available in the compound i (i.e., oleic or linolenic acids).

Research Organization for Science and Technology (Tehran, Iran). Figure 1 illustrates the structure of the SPMD chamber. The core component of the apparatus was the SPMD chamber (inner diameter: 0.12 m, length: 0.36 m; surface area: 0.14 m<sup>2</sup>), equipped with a Teflon roller-type wiper to ensure uniform oil distribution on the evaporator surface. A WiseCircu bath (WCR-P8 WiseCircu, Witeg Labotechnik GmbH, Germany) supplied a low-temperature flow to the cold trap and internal condenser. A 2 kW heating bath, operating with thermal silicone oil, was used to heat the evaporation section through its external jacket. A vacuum pump (Trivac D4B, Leybold, Germany) maintained a constant vacuum of 0.1 mbar throughout all experiments. The cold stream temperature and wiper speed were set at 0°C and 30 rpm, respectively. The SPMD system allowed



for adjustable feed flow rates ( $Q$ ) ranging from 0.50 to 3.00 mL/min.

#### 4. Results and discussion

The initial FFA contents of the cold-pressed camelina and lampante olive oil samples were 2.23 wt% and 10.11 wt%, respectively. The

lowest final FFA contents achieved were 1.13 wt% for cold-pressed camelina oil and 0.81 wt% for lampante olive oil. Table 2 [25,36] presents the calculated vapor pressures of linolenic and oleic acids at the reference temperatures studied. As observed, the vapor pressure of fatty acids increases significantly with rising ET.

**Table 2.** Oleic and linolenic acids vapor pressures at different ET.

Evaporation temp. (ET), °C	150	160	175	180	200
Linolenic acid vapor pressure, Pa	22.0	42.4	107.3	144.1	439.0
Oleic acid vapor pressure, Pa	16.6	33.0	85.9	116.0	356.0

Table 3 presents the mean H values at different flow rates (0.50, 1.75, and 3.00 mL/min). These

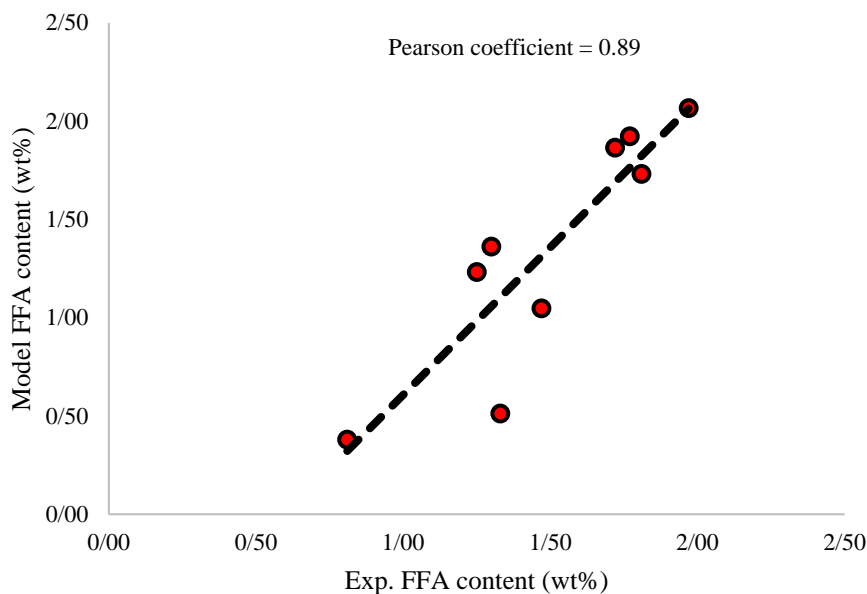
H values represent the arithmetic mean at each  $Q$ , calculated using Eq. 15.

**Table 3.** H mean values at each  $Q$ .

Feed flow rate ( $Q$ ), mL/min	0.50	1.75	3.00
H for camelina oil, $\text{kg } \sqrt[3]{^\circ\text{C}}$	0.244	0.203	0.104
H for olive oil, $\text{kg } \sqrt[3]{^\circ\text{C}}$	0.453	0.254	0.176

As shown, the H values decreased at higher  $Q$ , which may be attributed to an increased YL due to a lower heat transfer rate and reduced residence time at higher feed flow rates. According to Eq. 15, a higher YL value results in a lower H value. Additionally, the reduction in H values at higher  $Q$  is more pronounced for lampante olive oil, likely due to its higher initial FFA content.

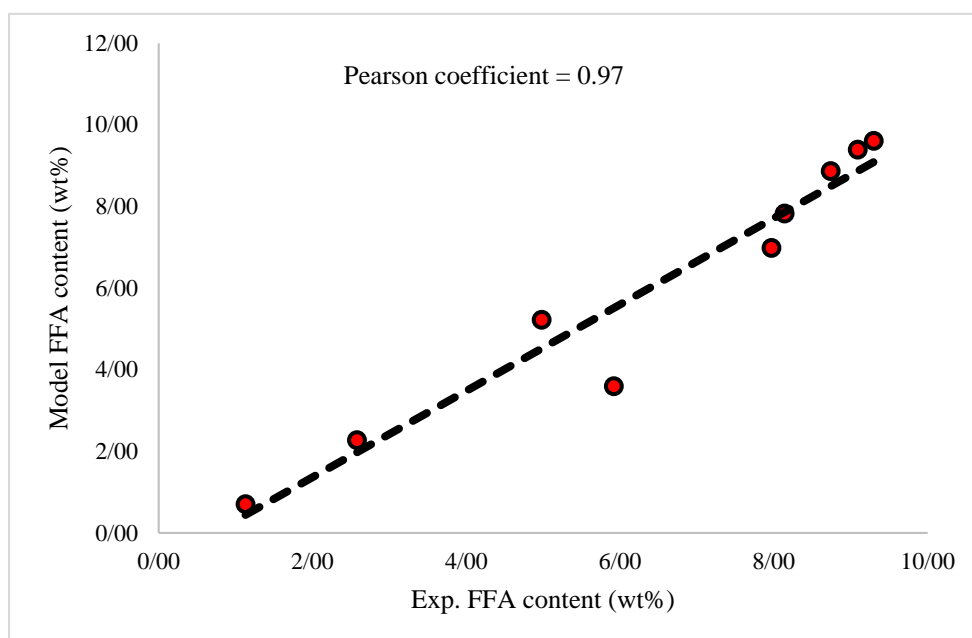
Figs. 2 and 3 illustrate the linear regressions of the experimental results versus the model's predicted outcomes for the oils. For camelina oil, which had a relatively lower initial FFA content (2.23 wt%), the Pearson correlation coefficient was 0.89, indicating a strong and adequate association.



**Fig. 2.** Model FFA content plotted vs. experimental FFA content for the cold-pressed camelina oil

For lampante olive oil, which had a significantly higher initial FFA content (10.11 wt%), a stronger agreement between the model and experimental data was observed, with a Pearson correlation coefficient of 0.97. The model-

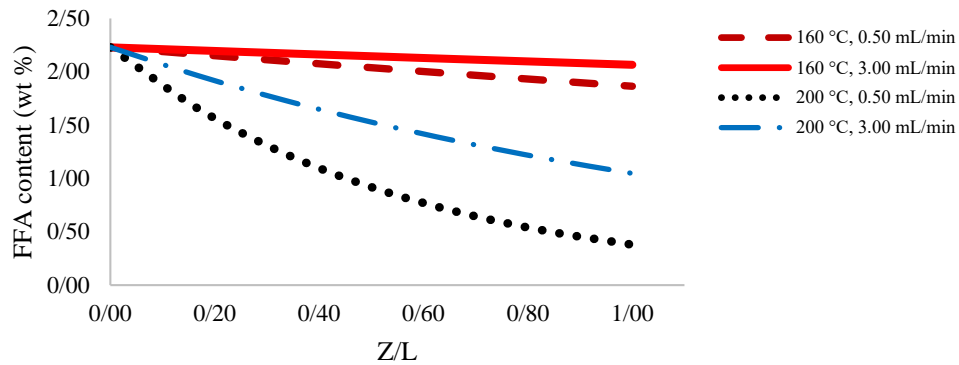
predicted FFA contents were calculated using Eq. 13, while vapor pressure calculations were performed using Eq. 14. The experimental FFA contents represent actual values reported in previous research studies [25,36].



**Fig 3.** Model FFA content plotted vs. experimental FFA content for the lampante olive oil.

In a similar study, the performance of SPMD in separating FFA from TG was examined. In that research, oleic acid was added to soybean oil in varying percentages. The deacidification efficiency of these binary blends was evaluated using both experimental and modeling approaches. While other SPMD process parameters, including Q, vacuum pressure, and condensation temperature, remained constant, ET varied between 110 and 160°C. Similar to the present study, the FFA content in the treated feed decreased as ET increased. A strong agreement between the model and experimental results was reported, with a Pearson correlation coefficient of 0.99 [6].

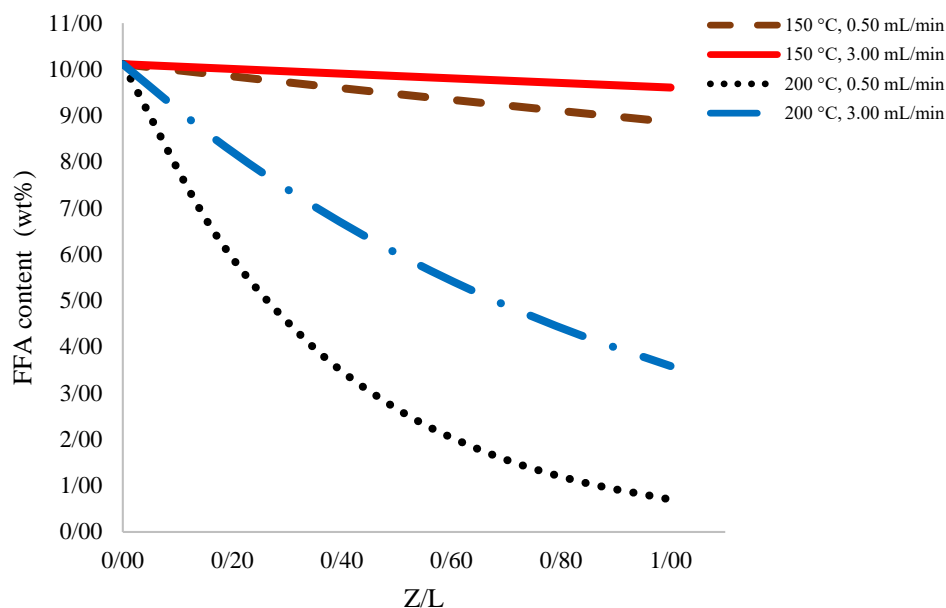
Figs. 4 and 5 illustrate the trends in FFA content reduction for the two oil samples throughout the SPMD evaporator column, at the minimum and maximum limits of the studied ET and Q ranges. For both treated oils, the decrease in FFA content through the SPMD evaporator is more pronounced at higher ET and lower Q. In SPMD, the vapor pressure difference serves as the driving force for evaporation and separation of constituents with higher volatility [41]. For vegetable oils, the vapor pressure difference between FFA and TG (the main compound) is significant.



**Fig 4.** FFA content changes of cold-pressed camelina oil throughout the SPMD evaporator column at ET of 160 and 200 °C and Q of 0.50 and 3.00 mL/min.

As shown in Table 2, FFA exhibits higher vapor pressures at higher ET. Increased vapor pressure results in a higher evaporation rate, leading to improved deacidification efficiency. In other words, at elevated ET, the enhanced vapor pressure of the evaporating molecules improves separation efficiency. A lower Q results in more

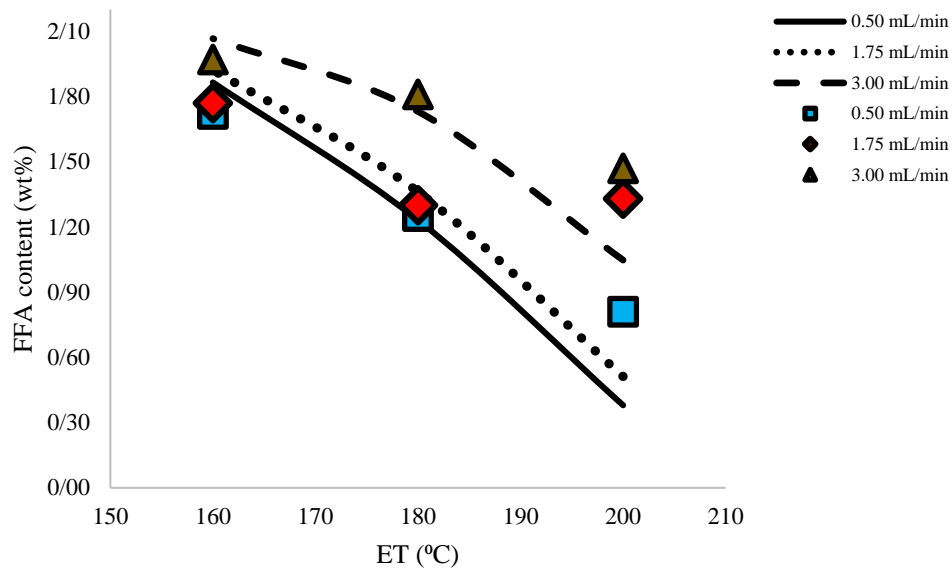
exposure time for the feed in the SPMD evaporator chamber, making heating more effective. This extended exposure allows volatile compounds more time to escape from the liquid phase, thereby significantly improving deacidification efficiency with a reduction in flow rate.



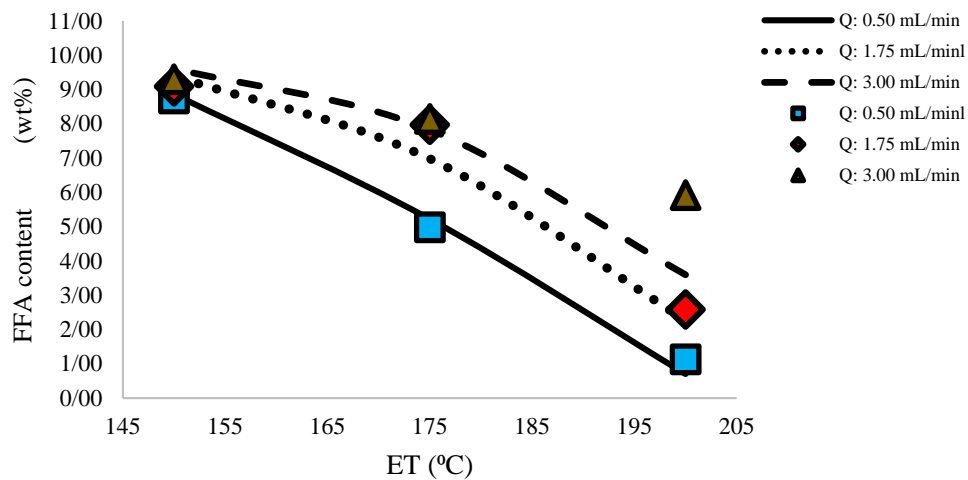
**Fig 5.** FFA content changes of lampante olive oil throughout the SPMD evaporator column at ET of 150 and 200 °C and Q of 0.50 and 3.00 mL/min.

Figures 6 and 7 present the model results alongside the experimental values for the changes in FFA content versus ET. According to Eq. 14,  $P^0$  is a function of ET. As indicated in Eq. 13, by applying the arithmetic mean value of H at each Q,  $Y_z$  is considered a function of both ET and z. Therefore, at constant Q and  $z = L$  (i.e., the SPMD system outlet),  $Y_L$  (the FFA content in the outlet stream) depends solely on ET. Higher H values correspond to higher evaporation rates in the evaporator's

falling film section. Therefore, the lower FFA content at lower Q is scientifically explainable. As shown, for both samples, the model demonstrated good agreement with the experimental results at lower ET. However, at an ET of 200°C, the experimental data tend to show some discrepancies compared to the model. One explanation for the more accurate predictions of the model at lower ET is that Eq. 14 predicts the vapor pressure of FFA more precisely at lower temperatures.



**Fig 6.** Comparison of experimental and model FFA content vs. ET at different Q related to the treated camelina oil. Lines and markers represent the model and experimental results, respectively.



**Fig 7.** Comparison of experimental and model FFA content vs. ET at different Q related to the treated olive oil. The lines and markers represent the model and experimental results, respectively.

## 5. Conclusions

This investigation aimed to develop a viable model to show the separation behavior of FFA from cold-pressed camelina oil and lampante olive oil using SPMD technology. The required

algorithm was established based on the mass conservation law and the Langmuir equation. By defining an SPMD coefficient (H) specific to the system design and the properties of the treated feed, the governing equations were derived. According to the results, the FFA content

throughout the evaporator column and the final FFA content at the outlet were strongly influenced by the applied ET and Q values. The analytical solution of the model's differential equations proved to be effective in describing the deacidification efficiency of SPMD. For both studied oils, the experimental and model results showed good agreement. The proposed model can be used for optimization and prediction purposes to scale up the oil deacidification process. Although the developed models primarily focused on FFA separation, the methodology can be applied to model the removal behavior of other volatile constituents, such as tocopherol or squalene recovery. Finally, it is recommended to develop similar deacidification modeling and simulation studies that incorporate qualitative factors, such as peroxide value.

### Conflict of interest

No conflict of interest has been declared by the authors.

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## مقاله پژوهشی

# اسید زدایی روغن‌های گیاهی با استفاده از تقطیر مولکولی مسیر کوتاه: مدل سازی و شبیه سازی

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### چکیده

تصفیه فیزیکی روغن‌های گیاهی به منظور به دست آوردن روغن‌های سالم‌تر، اجتناب از افت روغن و کاهش ضایعات تصفیه دارای کاربرد است. اسیدزدایی یا جداسازی اسیدهای چرب آزاد یک مرحله ضروری در تصفیه فیزیکی روغن‌های گیاهی است. هدف پژوهش حاضر توسعه مدل ریاضی بیان کننده رفتار اسیدزدایی روغن کاملینای پرس سرد و روغن زیتون لامپانت با استفاده از فناوری تقطیر مولکولی مسیر کوتاه (SPMD) به عنوان یک فناوری سازگار با محیط زیست (بدون استفاده از حلال) بود. از معادلات موازنه جرم و لانگمویر به منظور بیان مکانیسم‌های تبخیر و جداسازی استفاده شد. به منظور ارزیابی کارایی مدل از شدت جریان‌های مختلف خوراک و دماهای تبخیر متفاوتی بهره گرفته شد. سایر فاکتورها مانند دمای خوراک، فشار، دمای میعان و سرعت پخش کننده ثابت بودند. به منظور پیش‌بینی فشار بخار، از روش سهم گروهی استفاده شد. مقادیر پیش‌بینی شده به صورت منطقی با نتایج آزمایش‌های تجربی سازگار بود. دماهای تبخیر بالاتر و شدت جریان‌های پایین‌تر منجر به بازده‌های اسیدزدایی بالاتری شدند؛ همچنین از مدل پیشنهاد شده به منظور پیش‌بینی چگونگی تغییر محتوای اسیدهای چرب آزاد در طول ستون تقطیر کننده مولکولی مسیر کوتاه استفاده شد.

کلمات کلیدی: تقطیر مولکولی مسیر کوتاه (SPMD)، اسیدزدایی، مدل سازی ریاضی، شبیه سازی، روغن زیتون لامپانت، روغن کاملینا.