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Abstract of thesis

Except from virgin oils, crude oils cannot be consumed directly or incorporated to foods applications without refining. In fact, deacidification process is among the major steps in edible oil refining, whereas traditionally the separation of free fatty acids and triglycerides has been performed using chemical refining, largely consisting of neutralization and deodorization, which generates large amounts of waste-water and other by-products. On the other hand, in recent years the growing demand for more environmental-friendly processes has led to an increasing interest in physical refining process, whereas free fatty acids and other compounds are removed by steam distillation in the last step of the process, rather than using chemical neutralization. Thus, separating free fatty acids from triglycerides is crucial for the edible oil industry. With that said, molecular distillation with falling film was used in this study as deacidification process, aiming to elucidate the separation of fatty acids and triglycerides using model mixtures, both experimentally and in simulations.

Initially, the author presented a General Introduction in the thesis, vastly including up-to-date information related to the subject of this study, as well as the research goals. Next, in the first chapter, the author extensively performed model experiments on the separation of free fatty acids from triglycerides by molecular distillation, so that to comprehensively understand the principles of this separation process, whereas a binary mixture of oleic acid, as a free fatty acid, and refined soybean oil, as a triglyceride, were used. In those experiments, the

evaporation temperature tested ranged between 100 and 160 °C, meanwhile the condensation temperature, feeding rate, and vacuum pressure were kept constant. As evaporation temperature increased, free fatty acid content in retentate decreased, while free fatty acid in distillate content remained approximately 100 % (w/w). Since triglycerides have an extremely low vapor pressure compared to oleic acid, their fraction in the distillate is negligible. The results of this chapter demonstrated the influence of feeding rate and evaporation temperature on the free fatty acid separation mechanism. The retentate fraction increased with evaporation temperature but decreased with feeding rate. For a feeding rate of 5.0 x10^{-3} kg h^{-1}, the retentate fraction reached a plateau at 110 °C, while for higher feeding rates of 3.0 x10^{-1} kg h^{-1}, a plateau was found at 150 °C. Free fatty acid content in the feeding had no effect on the separation process; however, because the free fatty acid fraction in this study was limited, a higher free fatty acid fraction may have an influence on the fraction and free fatty acid content of retentate The driving force could be the vapor pressure, and triglyceride was difficult to evaporate due to its low vapor pressure. In distillate, the triglycerides fraction was assumed to be negligible. The modeling in the following chapter could be used to clarify this separation mechanism.

In the second chapter, after performing distillation experiments the author performed quantitative analyses using gas chromatography of the fatty acids present either in the original compounds, refined soybean oil and oleic acid, as well as in the retentate and distillate obtained by molecular distillation under various processing conditions, especially varying the condenser temperature, so that to determine the efficiency of separating free fatty acids and triglycerides using molecular distillation. The author clarified that the fatty acid composition of soybean oil in this study was similar to previous reports, which contained linoleic acid, oleic acid, palmitic acid, stearic acid, and arachidic acid. Moreover, the presence of oleic acid in the reagent oleic acid was also confirmed, and the remaining composition could include linoleic acid, palmitic acid, and myristic acid. The experimental results also indicated that the fatty acid composition in soybean oil and in the retentate, as well as fatty acid composition in oleic acid and in the distillate were considerably similar. Myristic acid, which is more easily evaporated and does not exist in soybean oil, was only found in the distillate, so that indicating that myristic acid was completely removed from the feeding mixture. Other fatty acids, such as palmitic acid, linoleic acid, and oleic acid, also supported this hypothesis.

In the third chapter, in order to elucidate the separation mechanism involved with molecular distillation, the author used the mathematical modeling approach, whereas the transport law was used to select the mass conservation as the major parameter, whereas Langmuir equation and Raoult's law were employed to describe the evaporation flux during the separation process. In addition, other experimental data, such as the evaporator length, were also required, so that to determine the coefficient of molecular distillation for the governing equation. As for the results of modeling study, the proposed governing equation could simulate the amount of free fatty acids in the retentate, with good precision. The author also reported that the modeling was revealed as effective in predicting the remaining mass fraction of volatile compounds in the retentate. To that end, the simulation required vapor pressure and the initial mass fraction of volatile compounds. To calculate the coefficient of molecular distillation, only one set of experimental data is required. In order to validate the model further, the author performed simulations using experimental data of the oil deacidification process by falling film molecular distillation, obtained from various literatures, whereas the author found good agreement and significant correlation, when comparing the simulated data using the model proposed in this study to those data reported in previous literature. However, some limitations were observed as well. The simulated mass fraction of volatile compound in the retentate was slightly lower than the experimental data when multiple volatile compounds evaporated at the same time.

In the general conclusion, the author reported about the suitability of falling film type-molecular distillation system for the separation of free fatty acid from refined vegetable oil, largely consisting of triglycerides. The evaporation temperature had a significant influence on the separation mechanism, whereas the initial free fatty acid content in feeding had a negligible influence. Both, the free fatty acid content obtained by chemical analysis, and the free fatty acid content obtained by mass balance indicated that the free fatty acid separation efficiency was proportional to evaporation temperature and reached a plateau at 140 °C. Moreover, the governing equation proposed by the author highlighted the capability to simulate free fatty acids content in retentate at any evaporation temperature, using only one experimental data. The equation could be used to simulate up to 27 % (w/w) of free fatty acids content in the feeding mixture. Further research relating to various feeding rate and vacuum pressure may be required, so that to further improve this model for more diverse simulations.

Abstract of assessment result

【Review】

In the first chapter, the applicant investigated the separation of free fatty acids and triglycerides initially prepared in a model mixture, aiming to clarify the mechanism behind falling film molecular distillation, based on both experimental and simulation approaches. The applicant focused particularly in the effect of evaporation temperature on on free fatty acids separation, as it was strongly related to other experimental parameters, such as condensation temperature, operating pressure, and wiper rotational speed. In the second chapter, the applicant proposed a mathematical model, aiming to predict the actual mass and mass fraction of each component present in the retentate and in the distillate. The applicant reported that a satisfactory and accurate level of agreement between the experimental and simulation results was achieved in the present study. In the third chapter, the applicant validated the model previously proposed, based on comparisons with previously literature reporting about oil deacidification with the falling film molecular distillation, which is similar to the system used in this study. Moreover, the model proposed in this study was designed to simulate the deacidification process, but it might also be useful for simulating the separation other volatile compounds, such as tocopherol or squalene. The dissertation is highly evaluated as an achievement that reveals significant academic improvements on fatty acid removal in fat and oil refining based on experiments and model analysis, proposes a mathematical model for fatty acid separation, and clarifies the effectiveness of the model, thereby providing fundamental knowledge useful for future development in the field of fat and oil production.

【Result】

The final examination committee conducted a meeting as a final examination on January 18, 2023. The applicant provided an overview of dissertation, addressed questions and comments raised during the questionand-answer session. All the committee members reached a final decision that the applicant has passed the final examination.

【Conclusion】

Therefore, the final examination committee approved that the applicant is qualified to be awarded Doctor of Philosophy in Food Innovation.