Soybean Oil Treatment Using the Dissolving Curve Equation of Hydrogen

Xingzhen Zhang1†, Tianyu Dong1†, Zeyu Wu1, Yuequn Jing2, Dianyu Yu†*, Hongwei Zhang1*, Jing Du1, and Liqi Wang3

1 School of Food Science, Northeast Agricultural University, Harbin, 150030, CHINA
2 School of Mathematics, Jilin University, Changchun, CHINA
3 School of Computer and Information Engineering, Harbin University of Commerce, Harbin, 150028, CHINA

Abstract: The solubility of hydrogen in n-hexane was determined using a homemade reactor. The solubility of hydrogen in soybean oil was established using the Peng-Robinson (PR) equation of state and the van der Waals mixing rule. The curve equation established a linear relationship between the solubility of hydrogen in oil and the number of moles of hydrogen in the reactor. Under the optimal temperature and catalyst, the relationship between the hydrogen consumption of the hydrogenation of oil and fat and the TFAs formed in the oil was determined. When the reaction pressure exceeded 3.0 MPa, the hydrogenation of oil was consumed. The amount of hydrogen, the rate of hydrogenation, and the change in the TFAs all stabilized. Therefore, the pressure of the general hydrogenation reaction should not exceed 3.0 MPa. This result provides a quick and simple method for controlling TFAs in oils and fats for industrial applications.

Key words: hydrogenated soybean oil, PR equation of state, solubility of hydrogen, hydrogen consumption, TFAs

1 Introduction

Oil hydrogenation technology is an important means of fat modification1. The conventional hydrogenation process is a typical solid-liquid-gas-phase system. In the oil hydrogenation process, hydrogen is dissolved in oil first, and then the liquid phase hydrogen gas and oil react together with a catalyst. Therefore, hydrogen diffuses into the surface of the catalyst in a three-phase system. Hydrogen is first adsorbed by the center of the catalyst activity. Then, adsorbed hydrogen encounters the adsorbed oil fatty acid chains to produce hydrogenated fats2,3. Hydrogen solubility in oil is the saturation hydrogen concentration in oil. The transfer of hydrogen in oils and fats plays a crucial role in the solubility of hydrogen in oils and fats. Meanwhile, the oil supply of hydrogen during the hydrogenation has an important impact on the hydrogenation reaction rate4 and trans fatty acids (TFAs). Chen et al.5 and Chávez et al.6 studied the vapor-liquid equilibrium of hydrogen and hydrocarbon as well as the impact of vapor-liquid equilibrium on the hydrogenation reaction, and the results showed that the solubility of hydrogen has an important influence on the reaction. In particular, hydrogen solubility is important for the hydrogenation of oils and fats.

Currently, hydrogen solubility in oil or an organic solvent measurement method has experimental and theoretical designs. The measurement process of the experimental determination method is complicated, and the theoretical calculation makes it easier to determine the solubility of hydrogen. Saajanlehto et al.7 measured and modeled the solubility of hydrogen in heavy oil system conditions at 498-598 K and 2.0-11.0 MPa. Browning et al.8 studied the vapor-liquid equilibrium of the hydrogen, vacuum gas oil, and middle distillate fractions. Ronze et al.9 measured the solubility of hydrogen in a straight run gas oil and hydrogen solubility simulation in a straight run gas oil using Hysys software. Li et al.10 measured the hydrogen and carbon dioxide solubility in toluene using an experimental device, and gas-liquid equilibrium experimental data were correlated to experimental values by calculating the Peng-Robinson equation of state. Hrnčíř et al.11 studied the solubility of hydrogen in hydrogenated pyrolysis oil and pyrolysis oil under different pressure conditions. The results showed that the solubility of hydrogen in bio-oil increases with an increasing pressure and temperature. Rojas et al.12 predicted a thermodynamic model of hydrogen solubility in pyrolysis oil by using the Peng-Robinson equation of state and proved the importance of the equilibrium model choice for vapor liquid equilibrium (VLE) calculations.
The increase in the hydrogenation reaction rate is mainly related to the diffusion of organic solvents and the solubility of hydrogen. In the process of oil hydrogenation, the solubility of hydrogen in oil is mainly a function of temperature and pressure. Therefore, increasing the temperature and hydrogenation pressure will increase the solubility of hydrogen in soybean oil. However, because the hydrogenation reaction is exothermic, the effect of increasing reaction temperature on the rate of hydrogenation reaction is small. At the same time, a hydrogenation temperature that is too high can easily lead to a significant increase in the content of TFA in hydrogenated oils and fats. Therefore, in industrial production, properly increasing the reaction pressure properly can improve the solubility of hydrogen in oil, ensure the effective hydrogen adsorbed on the catalyst surface, and reduce the formation of trans isomers. Yan et al. reported that the hydrogenation reaction rate of levulinic acid would be significantly accelerated by increasing the partial pressure of hydrogen. King et al. studied the hydrogenation of vegetable oils and fats, and the results showed that the hydrogenation products were affected by different hydrogenation conditions. Congxin et al. studied Pt/SAPO-11 oil-step catalytic hydrogenation reaction intermediates and the trends of final products of alkanes over reaction pressure, and other reaction conditions. The results showed that the probability of occurrence of the hydrogenolysis reaction increased. The hydrodeoxygenation reaction of oil was promoted by increasing the pressure. However, too high of a reaction pressure will reduce the service life of the equipment and increase the cost.

In this study, the hydrogen solubility in n-hexane was measured using a homemade autoclave system to verify the reliability of the system. Under certain conditions, the use of hydrogen solubility in soybean oil was measured in the system, the solubility data were correlated by the Peng-Robinson equation of state and the Van der Waals mixing rules, and a curve equation of hydrogen solubility in soybean oil was established. Using the solubility curve equation in soybean oil, the linear relationship between the solubility of hydrogen in oil and the number of moles of hydrogen in the reactor was established by measuring the number of moles of hydrogen in the reactor. In the presence of catalysts, hydrogenation of soybean oil was conducted. After the end of the reaction, the relationship between the hydrogen consumption of oil hydrogenation in the hydrogenation reaction and TFAs generated in oil was established to determine the pressure of the oil hydrogenation reaction and provide a fast basis for determining the TFA content in oil.

2 Materials and Methods

2.1 Materials

Hydrogen (purity ≥ 99%), and nitrogen (purity ≥ 99%) were obtained from Harbin City Gas Co., Ltd. n-hexane of analytical reagent grade was purchased from Sinopharm Chemical Reagent Co., Ltd.; soybean oil (acid value: 0.12 mg KOH/g, iodine value: 132 g I/100g oil; peroxide value: 3.6 mmol/kg; color: Lovibond color groove 133.4 mm: R1.1, Y10) was purchased from ninety-three oil industry Group Co., Ltd. provided; 5% Pd/C catalyst was obtained from Shaanxi New materials Co., Ltd. Ruike; and trans oleic acid methyl ester and trans-linoleic acid methyl ester were supplied by Sigma, USA.

2.2 Experimental device for solubility determination

A schematic view of the experimental apparatus is shown in Fig. 1, which mainly consists of two high-pressure balancing systems and measurement systems. The high pressure system consisted of a high pressure reaction vessel, an oil bath and the temperature and pressure indicators, and the autoclave was made of a stainless-steel material, volume with a 150.4 mL. At settled temperature and pressure conditions, gas and liquid gas reaches autoclave-liquid equilibrium. The reaction fluid was stirred using a magnetic stirrer in an autoclave, and the fluid in the system was used to maintain a balanced state. The measurement system was composed of sampling flash tanks, flash tank, pressure and temperature gauges, and other components. The volume of the sample canister was 60.5 mL, and the volume of the flash tank was 330.5 mL.
2.3 Equipment reliability experiment
To ensure the reliability of the experimental data, experimental hexane was chosen as the based material at a temperature of 373 K, and a pressure of 1.0-8.0 MPa. Under these experimental conditions, the solubility of hydrogen in n-hexane was measured and compared with the reference value.

2.4 Determination of the H₂ solubility in soybean oil
Soybean oil (50.0 g), a 0.5 mol/L sodium formate solution (60 mL) and EDDAB (1.0 g) were added to the electrolyte in the cathode chamber of the reactor, and then N₂ (9 MPa) was fed into the sealed reactor. When the pressure reached stability after 30 min, N₂ was discharged from the reactor, pressurized H₂ was fed into the reactor, and the reaction mixture was agitated for 30 min at 300 rpm. The electrolyte was discharged from the bottom of the reactor into the sampling tank, and then the electrolyte was moved into the evaporation tank in which the H₂ dissolved in the electrolyte was evaporated. The electrolyte in the evaporation tank was cooled to room temperature and moved to the collecting tank. The solubility of H₂ in electrolyte was calculated by the sum of the volume of the evaporation tank and the sampling tank minus the volume of electrolyte. The solubility of H₂ in electrolyte under different conditions was measured by changing the experimental conditions.

2.5 Hydrogen mole fraction
Determination of the H₂ solubility in soybean oil was shown as a mole fraction using the following calculated formula:

\[ x = \frac{n_H}{n_H + n_I} \]

where \( x \) is the mole fraction of H₂ in the electrolyte under the designed conditions, and \( n_H \) and \( n_I \) are the quantity of evaporated H₂ and electrolyte, respectively.

2.6 The relationship between the mole fraction of hydrogen in the reactor and the solubility in soybean oil
The linear relationship between the solubility of hydrogen in oil and the number of moles of hydrogen in the reactor was established by measuring the number of moles of hydrogen in the reactor under the condition at 413 K using the solubility curve equation in soybean oil.

2.7 The effect of the reaction pressure on hydrogenation reaction process
Soybean oil (50.0 g) and a Pd/C catalyst 0.23 (w/w) were added to a 300.0 mL high pressure reactor. The reaction kettle was sealed, the reactor was subjected to a test leak by passing N₂, and then the system was evacuated to an absolute vacuum. The hydrogen pressure of the reactor reached a certain pressure. The reaction kettle was placed in an oil bath at a certain temperature, and after the temperature was constant, the magnetic rotor was turned on at a rotation speed of 300 r/min. After 2 h of reaction, the product was centrifuged several times to separate the catalyst to obtain a hydrogenated oil.

The pressure before the reaction was measured to determine the solubility in the soybean oil, and the number of moles of hydrogen in the reaction kettle was determined according to the solubility in the soybean oil. After the reaction was completed, the number of moles of hydrogen remaining in the reactor was determined by the same method. The effect of pressure on the hydrogen consumption of hydrogenated fats and oils and the formation of TFAs in fats and oils during hydrogenation was established.

2.8 Determination of TFAs in oils
The method of fatty acid methyl esterification was described by Yu et al. The TFAs of hydrogenated fats and oils were analyzed via gas chromatography, a CP-Sil-88 strong polarity capillary gas chromatography column (100 m × 0.25 nm × 0.2 μm), and a hydrogen flame ionization detector (FID). The additional parameters were as follows for fatty acid standards and samples: Carrier gas: (N₂), flow rate 30 mL/min; Gas: (H₂), flow rate 30 mL/min; Combustion gas: (Air), flow rate 300 mL/min; determination of fatty acid standards and samples using inlet temperature 533 K; detection temperature 533 K; column pressure 281.7 kPa; column temperature 706 K, 5 K/min temperature rise, hold for 12 min; trans oleic acid and trans linoleic acid standards and samples were measured using detection temperature of 523 K, an inlet temperature 523 K, column temperature 473 K, 5 K/min temperature rise, hold for 10 min, the split ratio for both analyses was 100:1, and the injection volume was 1 μL.

2.9 Determination of the iodine value of oil
The iodine values (IVs) were determined according to the method reported previously by Liu et al. A prepared potassium iodide (KI) solution (10% solution) was prepared and tested by dissolving 100 g of reagent grade KI in 1000 mL of deionized water, as noted in the Reagents. Salicylic acid (1.25 g/L) may be added to preserve the indicator. The mixture was then titrated with a 0.1 N Na₂S₂O₃ solution, adding it gradually and with constant and vigorous shaking. The titration was then titrated with 0.1 N Na₂S₂O₃ solution, almost disappeared. 1–2 mL of starch indicator or solution was added and the titration was continued until the blue color had just disappeared.

3 Results
3.1 Determination of hydrogen solubility in n-hexane
With n-hexane as the base material, the changes in hy-
drogen solubility in different pressure ranges were investigated and compared with the reference values of standard theory\textsuperscript{20}. According to Fig. 2, as the pressure increased, the solubility of hydrogen in \textit{n}-hexane increased rapidly at a pressure of 2.0 MPa, and the hydrogen mole fraction was 0.026, when the pressure reached 8.0 MPa, and mole fraction of hydrogen increased to 0.079. The results were compared with the reference value and the experimental and reference values were close\textsuperscript{21} indicating that this assay provides a reliable laboratory equipment and a feasible method for determining the hydrogen solubility in soybean oil.

### 3.2 Hydrogen solubility in soybean oil

At 393 K, 413 K and 433 K, the solubility of hydrogen in soybean oil under different pressures was determined as shown in Table 1. The temperature was increased, and the hydrogen solubility in the soybean oil binary system showed a gradual upward trend, this is because the volume of the soybean oil expanded with an increasing of system temperature. Soybean oil macromolecules provided free hydrogen molecules in an extremely small gap, in favor of small molecules such voids are squeezed hydrogen\textsuperscript{22}, while the viscosity of the soybean oil was reduced as the temperature was increased, the stirring effect was increased, and the mass transfer resistance was reduced in favor of hydrogen into the liquid soybean oil; thus, the hydrogen solubility in soybean oil can be increased.

Additionally under certain temperature conditions, the hydrogen solubility in the soybean oil of a binary system increased as the pressure was gradually was increased, which is because the increase in the system pressure can increase the concentration of hydrogen in the system, the gas density increased, promoting the hydrogen dissolved in soybean oil; hydrogen concentration of the soybean oil surface increased as the hydrogen pressure increased, so the gas molecules coming into the surface were more than the gas molecules escaping from the level, so that the gas solubility increased. The changes were consistent with Morais et al.\textsuperscript{23} who studied the change in solubility of hydrogen in oleic acid.

### 3.3 Calculation of the hydrogen solubility in soybean oil by the equation of state

#### 3.3.1 Prediction of the critical nature and eccentric factors of soybean oil

Since soybean oil is a mixture of various fatty acid triglycerides, its critical nature and eccentric factor are difficult to be determined by experiment. For soybean oil, the critical temperature $T_{c}$ and pressure $P_{c}$ were estimated using the group contribution method. The critical temperature and critical pressure of the estimated soybean oil values are shown in Table 2. The eccentric factor was estimated to be 0.231 via the Chen equation\textsuperscript{24}. The hydrogen critical temperature, pressure and eccentric factor were taken from the literature\textsuperscript{25}.

#### 3.3.2 Hydrogen association soybean oil solubility data

The Peng-Robinson (PR) equation of state is one of the most commonly used to calculate equilibrium, based on its predecessors, the calculation of the volume was modified to improve the calculation accuracy\textsuperscript{26}. In this study, hydrogen solubility in soybean oil was estimated by the Peng-Robinson equation of state and the Van der Waals mixing

| T=393K |  | T=413K |  | T=433K |  |
|--------|--------|--------|--------|--------|--------|
| P(MPa) | x      | P(MPa) | x      | P(MPa) | x      |
| 0.5    | 0.0071 | 0.5    | 0.0089 | 0.5    | 0.0116 |
| 1.0    | 0.0122 | 1.0    | 0.0142 | 1.0    | 0.0172 |
| 2.0    | 0.0224 | 2.0    | 0.0264 | 2.0    | 0.0324 |
| 3.0    | 0.0355 | 3.0    | 0.0435 | 3.0    | 0.0512 |
| 4.0    | 0.0576 | 4.0    | 0.0716 | 4.0    | 0.0836 |
| 5.0    | 0.0648 | 5.0    | 0.0864 | 5.0    | 0.0934 |

Note: x is the experimental value of hydrogen solubility in soybean oil
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rules for hydrogen, with experimental values for comparison. The hydrogen solubility data in soybean oil were verified by the Peng-Robinson equation of state, and the PR equation and mixing rule formula as follows:

Peng-Robinson equation of state:

\[ P = \frac{RT}{V-b} - \frac{a}{V(V+b) + b(V-b)} \]  \hspace{1cm} (1)

\[ b = 0.07780 \left( \frac{RT_c}{P_c} \right)^{0.45724} \]  \hspace{1cm} (2)

\[ a = 0.45724 \left( \frac{RT_c}{P_c} \right)^{2.68992} \left( 1 + m \left( 1 - \frac{T}{T_c} \right) \right)^{2} \]  \hspace{1cm} (3)

\[ m = 0.3746 + 1.54226\omega - 0.26992\omega^2 (0 < \omega < 0.5) \]  \hspace{1cm} (4)

Van der Waals mixing rules:

\[ a = \sum x_i x_j a_{ij} \]  \hspace{1cm} (5)

\[ b = \sum x_i b_i \]  \hspace{1cm} (6)

\[ a_{ij} = \sqrt{a_i a_j} (1 - k_{ij}) \]  \hspace{1cm} (7)

Where \( a_{ij} \) and \( b_i \) are for the binary interaction parameters between \( i \) and \( j \), and \( k_{ij} \) is the binary adjustable parameter. The binary interaction parameter \( k_{ij} \) was evaluated by generalized least squares, and the results are shown in Table 3.

### Table 2: The critical properties of the H\textsubscript{2} and soybean oil.

|        | \( T_c / K \) | \( P_c / MPa \) | eccentric factor \( \omega \) |
|--------|---------------|-----------------|-----------------|
| soybean oil | 978.8         | 0.33            | 0.231           |
| H\textsubscript{2} | 33.2          | 1.30            | -0.22           |

### Table 3: Two element interaction parameters in the Peng-Robinson equation.

| Temperature (K) | 393 | 413 | 433 |
|-----------------|-----|-----|-----|
| \( k_{ij} \)   | 0.376 | 0.423 | 0.515 |

### Table 3.

3.3.3 Calculation of the hydrogen solubility in soybean oil

The binary interaction parameters obtained in Table 3 were introduced into the PR equation of state, and the hydrogen solubility in soybean oil was calculated by MATLAB at different temperatures and pressures, as shown in Fig. 3. The solubility of hydrogen in soybean oil can be determined by the model, within a certain pressure range. The trend of the hydrogen solubility data obtained by the model fitting with temperature and pressure is equal to the current measurement results, which shows that the fitting effect of the model is better. This result is consistent with the prediction of the solubility of hydrogen in petroleum fractions studied by Aguilar-Cisneros et al.

### 3.4 The relationship between the molar fraction of hydrogen and its solubility in soybean oil

From the curve equation of solubility in soybean oil, the pressure directly affects the solubility of hydrogen in soybean oil in the reaction vessel. At 413 K, the relationship between the molar fraction of hydrogen in the reactor and the solubility in soybean oil is shown in Fig. 4. There is a certain linear relationship between the solubility of hydrogen in oil and the mole number of hydrogens in the reactor. With an increasing of hydrogen pressure, the solubility of hydrogen in oil increases, and the mole number of hydrogens in the reactor increases simultaneously. The fitting equation between the solubility of hydrogen and the mole number of hydrogens in the reactor is \( y = 7.5428x + 0.0862 \), where \( R^2 = 0.9822 \), indicating the fitting degree. This can be used to calculate the mole number of hydrogens in the reactor. This method is consistent with a previous study on determining of the solubility of hydrogen in soybean oil.
3.5 Changes in the amount of hydrogen in the hydrogenation of soybean oil

At 413 K temperature and a certain pressure, according to the relationship between the molar fraction of hydrogen in the reactor and the solubility of soybean oil, the molar number of hydrogen in the reactor before and after the reaction, the hydrogen consumption of oil hydrogenation, the iodine value of oil and the TFA content in hydrogenated oil are shown in Table 4. With an increase in the hydrogen pressure, the hydrogen consumption increased when hydrogenating soybean oil, and the hydrogenation reaction rate of soybean oil gradually increased (Table 4 and Fig. 4).

When the hydrogen pressure ranged from 1.0 MPa to 2.0 MPa, the hydrogenation reaction rate of hydrogenated soybean oil increased rapidly; and pressure increased slowly from 2.0 MPa to 3.0 MPa. The hydrogen consumption of soybean oil is slow, and the hydrogenation reaction rate increase slowly. When the pressure exceeded 3.0 MPa, the hydrogen consumption of soybean oil hydrogenation tended to be stable, and the hydrogenation reaction rate remained unchanged.

This was mainly attributed to the increasing hydrogen pressure, the amount of hydrogen dissolved in soybean oil increased, the amount of hydrogen spreading to the catalyst surface increased per unit time, the effective concentration of hydrogen adsorbed on the catalyst surface increased, the amount of hydrogen consumption of hydrogenated oils increased, and the hydrogenation reaction rate increased, with an accelerating the hydrogenation reaction, which is consistent with the conclusion of Morais et al.\textsuperscript{23}

Table 4 The main data of hydrogenation reaction under different pressure conditions.

| Pre-reaction pressure P(MPa) | Post-reaction pressure P(MPa) | Pre-reaction Hydrogen volume (mol) | Post-reaction Hydrogen volume (mol) | Hydrogen consumption (mol) | IV (I\textsubscript{2} g/100 g) | TFAs (%) |
|-----------------------------|-------------------------------|-----------------------------------|-----------------------------------|---------------------------|------------------|---------|
| 0                           | 0                             | 0                                 | 0                                 | 0                         | 132              | 1.23    |
| 1.0                         | 0.4                           | 0.0801                            | 0.0299                            | 0.0502                    | 105.64           | 21.84   |
| 1.5                         | 0.7                           | 0.1165                            | 0.0517                            | 0.0648                    | 98.76            | 20.46   |
| 2.0                         | 0.9                           | 0.1529                            | 0.0728                            | 0.0801                    | 92.55            | 19.54   |
| 2.5                         | 1.3                           | 0.1893                            | 0.1015                            | 0.0878                    | 90.26            | 18.25   |
| 3.0                         | 1.7                           | 0.2257                            | 0.1307                            | 0.0942                    | 85.91            | 16.81   |
| 3.5                         | 2.2                           | 0.2621                            | 0.1675                            | 0.0946                    | 85.22            | 16.45   |
| 4.0                         | 2.7                           | 0.2985                            | 0.2039                            | 0.0946                    | 84.83            | 16.30   |
| 4.5                         | 3.2                           | 0.3349                            | 0.2402                            | 0.0947                    | 84.50            | 16.30   |
| 5.0                         | 3.7                           | 0.3713                            | 0.2766                            | 0.0947                    | 84.17            | 16.29   |

Fig. 5 Effect of hydrogen pressure on hydrogen consumption and TFAs of oils and fats.

3.6 The effect of the reaction pressure on the hydrogenation reaction process

The effect of pressure on hydrogen consumption and TFAs during oil hydrogenation was established by using the data in Table 4. As shown in Fig. 5, the hydrogen concentration on the catalyst surface was the main reason for TFAs formation during soybean oil hydrogenation\textsuperscript{30,31}. The TFA content in the products of the hydrogenation reaction shows a significant downward trend in the pressure range of 1.0-3.0 MPa. Under certain other conditions, with an increasing hydrogen pressure, the solubility of hydrogen in oil will be increased. When the double bond of unsaturated fatty acids in oil is opened, the excited hydrogen will quickly bind to the unsaturated fatty acids, because the excited hydrogen concentration is high. Because of the high concentration of hydrogen in the excited state, unsaturated fatty acids do not easily recombine after opening the double bond, which results in a gradual decrease in the TFA content in oils and fats. This result is consistent with...
the conclusion that a sufficient hydrogen supply during the hydrogenation process is conducive to the reduction of TFAs formation. When the hydrogen pressure in the reactor exceeds 3.0 MPa, enough hydrogen has been dissolved in soybean oil to saturate the effective hydrogen on the catalyst surface, and the concentration of hydrogen in the excited state remains basically unchanged. Therefore, the probability of TFAs formation in the oil tends to balance. The pressure of the hydrogenation reaction should not exceed 3.0 MPa.

4 Conclusions

Measuring the hydrogen solubility in n-hexane proved that the test data from the homemade autoclave were reliable. Hydrogen was injected into the autoclave in the range of 0-5.0 MPa and 393 K-433 K. The amount of hydrogen in the autoclave was then measured, and the curve equation of the solubility of hydrogen in soybean oil was established. It was proven that the curve equation of the solubility of hydrogen in soybean oil fitted well with the actual value. The linear equation between the solubility of hydrogen in oil and the molar number of hydrogens in the reactor was established. R² = 0.9822, which fitted well with the actual value. Under 1.0-3.0 MPa pressure, with an increasing hydrogenation pressure, the hydrogen solubility in soybean oil and the hydrogen consumption of hydrogenated oil increased, the hydrogenation reaction rate increased, and the TFAs during the oil hydrogenation process decreased gradually. When the pressure was 3.0 MPa, the hydrogenation consumption of oil was 0.0942 mol, while that of TFAs was only 16.81%. It provides a simple calculation method for TFAs control in industrial production. When the reaction pressure exceeded 3.0 MPa, the hydrogen consumption in hydrogenated oils and TFA variations were stabilized. Therefore, the pressure of the hydrogenation reaction should not exceed 3.0 MPa.

Author Contributions

Conceptualization, X.Z. and T.D.; methodology, Z.W.; software, Y.J.; validation, H.Z. and D.Y.; formal analysis, H.Z.; investigation, J.D.; resources, L.W.; data curation, T.D.; writing—original draft preparation, X.Z.; writing—review and editing, D.Y.; visualization, H.Z.; supervision, J.D.; project administration, D.Y and L.W.; funding acquisition, D.Y and L.W. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest

The authors declare no conflict of interest.

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