Catalytic Transfer Hydrogenation of Low-erucic-acid Rapeseed Oil over a Ni-Ag\textsubscript{0.15}/SBA15 Catalyst

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Abstract: The kinetics of CTH of low-erucic-acid rapeseed oil using ammonium formate as a hydrogen donor over a Ni-Ag\textsubscript{0.15}/SBA15 catalyst were studied. Then, a kinetic model for the hydrogenation of low-erucic-acid rapeseed oil was established, and it was found that the reaction rate constants of hydrogenations of 9c-18:1 and 12c-18:1 oleic acid were 0.1262 and 0.0148, and the catalytic selectivity of linoleic acid was 2.04. For the catalyst loading of 0.23\%, the hydrogenation temperature was 80°C, the ammonium formate concentration was 0.32 mol/50 mL, and the low-erucic-acid rapeseed oil was hydrogenated in 90 min; it was also found that the iodine value of low-erucic-acid rapeseed oil was 80 g I\textsubscript{2}/100 g, the oleic acid content was 65\%, and the trans fatty acids (TFAs) content was only 6.7\%. Therefore, catalytic transfer hydrogenation (CTH) may be widely used in the modification of oils and fats.

Key words: catalytic transfer hydrogenation, low-erucic-acid rapeseed oil, kinetics, oleic acid, TFAs

1 Introduction

In November 2018, the US Food and Drug Administration approved a new qualified health claim: a oleic acid content higher than 70\% in vegetable oils can reduce the risk of cardiovascular disease.

According to the World Health Organization study, the incidence of cardiovascular disease among the residents of the Mediterranean basin is low and this is related to the long-term consumption of vegetable oils rich in oleic acid in this region\textsuperscript{1}. During the hydrogenation of oils, anti-nutritional factors such as trans fatty acids (TFAs) are produced that are harmful to human health. It was reported that TFAs can increase the incidence of coronary heart disease and cardiovascular disease in humans\textsuperscript{2}. In the human body, the content of low-density lipoprotein (LDL) increases due to the ingestion of TFAs\textsuperscript{3}. In 2010, the World Health Organization (WHO) recommended reducing the content of TFAs in food production. In September 2018, Canadian government completely banned the addition of artificial trans fatty acids in food.

In the hydrogenation processing of vegetable oils, the TFAs content can be reduced by modifying the composition of the catalyst and the hydrogenation process\textsuperscript{4}. In the hydrogenation process, a large amount of trans fatty acids is obtained due to high temperature. It was shown that soybean oil hydrogenated under supercritical conditions has a low trans fatty acid content\textsuperscript{5}. Catalytic transfer hydrogenation results in a minimal degree of isomerization of fatty acids and lower trans fatty acid content, representing a novel hydrogenation process\textsuperscript{6}. Based on their roles in the catalytic process, the components of the catalyst were divided into the main catalyst, cocatalyst and substrate\textsuperscript{7}. Typically, Al\textsubscript{2}O\textsubscript{3}, TiO\textsubscript{2} and SBA15 are used as the substrates in the preparation of the catalyst. The SBA15 substrate promoted the dispersion of the catalyst and the adsorption of hydrogen due to its large specific surface area and pore size\textsuperscript{8}. Moreover, SBA15 has good pore wall thickness and hydrothermal stability, and did not collapse easily under high temperature roasting conditions\textsuperscript{9}. Therefore, SBA15 is a good choice as a substrate for the preparation of supported metal catalysts.

As a catalyst for hydrogenated oils, pure nickel had the characteristics of high catalytic selectivity and low cost\textsuperscript{10}. Raney-Ni has been the most widely used catalyst for hydrogenated oils, and has the Ni content above 90\%. This catalyst is generally active in the form of clusters and was easily deactivated during hydrogenation\textsuperscript{11}. However, Cu-Ni bimetallic catalyst easily leads to oxidative rancidity of vegetable oils due to hydrogenation process\textsuperscript{12}. The catalytic activity of the mesoporous SBA15 bimetallic loaded material was significantly higher than that of the Ni catalyst supported separately\textsuperscript{13}. It has been shown that Au metal...
can increase the activity of the catalyst while enhancing the interaction between Ni and SBA15. In the application of silver-nickel catalysts, because the active metal nickel tends to cluster on the support during the preparation of the supported catalyst, silver is beneficial for the prevention of the metallic nickel cluster. It was pointed out that in nickel-silver/SBA15 catalysts, an appropriate amount of silver increase the dispersion of metallic nickel. The addition of an appropriate amount of silver in the nickel catalyst can also effectively inhibit the formation of trans fatty acids. Ni-Ag catalyst was shown to exhibit a better selectivity for linoleic acid and lower isomer formation. Based on the study of the hydrogenation kinetics, a dynamic model was developed that represented the isomerization of linoleic acid in soybean oil under different reaction temperatures (from 160 to 250°C) and different reaction times (12-72 h). Kinetic studies were performed to compare the hydrogenation rates of different catalysts using the Runge-Kutta method to calculate the pathway for the conversion of linoleic acid to stearic acid. The hydrogenation kinetics were described by a simplified three-step model including linolenic acid. The mathematical model was used for the regulation of the hydrogenation process and to solve the problem of higher TFAs generated in the hydrogenation of fats and oils.

Thus, the kinetics of the hydrogenation of the oils were analyzed, and a kinetic model for the hydrogenation of the oils was established, providing theoretical guidance for application in practical production.

In this study, the Ni-Ag0.15/SBA15 catalyst was prepared, and it was found that the addition of the silver metal promoter promoted the dispersion of the main catalyst. The catalyst was applied to the hydrogenation of low-erucic-acid rapeseed oil under the CTH system. The effects of donor concentration, catalyst addition amount and hydrogenation temperature on the hydrogenation process were studied. The reaction rate of fatty acid during hydrogenation was examined to verify the consistency of the kinetic model of hydrogenation and the experimental values. The effects of the addition of metal Ag on the oleic acid isomerization and linoleic acid selectivity during the hydrogenation process were discussed.

2 Materials and Methods
2.1 Experimental materials
SBA15 mesoporous molecular sieve, (Nanjing Pioneer Nanotechnology Co., Ltd. China); nickel nitrate (six water), silver nitrate (Sinopharm Chemical Reagent Co., Ltd., China); low-erucic-acid rapeseed oil (Wuhan Zhongyou Hongda Technology Industry Co., Ltd., China); fatty acid composition in raw rapeseed oil: linolenic acid = 0.102, linoleic acid = 0.204, f_{16:1} = 0.482, f_{12:18:1} = 0.026, f_{18:1} = 0, f_{12:18:1} = 0, stearic acid = 0.0202; Iodine value: 110 g I\textsubscript{2}/100 g oil, oleic acid: 52%, erucic acid: ≤ 5%; ammonium formate solution (prepared in the laboratory).

2.2 Experimental methods
2.2.1 Catalyst preparation
Ni-Ag\textsubscript{0.15}/SBA15 was prepared by the coimpregnation method with the mass fraction of 12% Ni metal and silver nitrate as the main catalyst. Ni-Ag catalyst was shown to exhibit a better selectivity for linoleic acid and lower isomer formation. The overall mechanism of catalytic transfer hydrogenation (CTH) using ammonium formate as the hydrogen donor can be expressed as:

\[
\begin{align*}
\text{HCOONH}_4 & \rightarrow \text{NH}_4^+ + \text{HCOO}^- \\
\text{HCOO}^- + \text{H}_2\text{O} + \text{Ni-Ag} & \rightarrow \text{H} - \text{Ni-Ag} - \text{H} + \text{HCOO}^- \\
\text{H} - \text{Ni-Ag} - \text{H} + \text{R}_1 - \text{CH} = \text{CH} - \text{R}_2 & \rightarrow \text{R}_1 - \text{CH}_2 - \text{CH}_2 - \text{R}_2 + \text{Ni-Ag} \\
\text{HCOO}^- & \rightarrow \text{OH}^- + \text{CO}_2 \\
\text{CO}_2 + \text{H}_2\text{O} & \rightarrow \text{H}_2\text{CO}_3 \rightarrow \text{H}^+ + \text{HCOO}^- \\
\text{NH}_4^+ + \text{OH}^- & \rightarrow \text{NH}_3 + \text{H}_2\text{O}
\end{align*}
\]

The reaction vessel (150 mL) was preheated to 80°C, and then low-erucic-acid rapeseed oil (100 g), and a certain amount of the catalyst and an ammonium formate solution were added to the reaction vessel, and the mixture was continuously stirred at 200 rpm. After reaction for a period of time, the catalyst was separated from the oils and fats by centrifugation and the iodine value of the oils and fats and the content of each fatty acid component were determined.

2.2.2 Determination of fatty acids
Methyl esters of fatty acids were prepared according to the literature. The compositions of the samples were analyzed by gas chromatography (GC) using a flame ionization detector (FID) and a CP-Sil-88 (100 m × 0.25 mm × 0.2 μm) column (Agilent Technologies, Palo Alto, CA, USA). The carrier gas was nitrogen at a flow rate of 1 mL min\textsuperscript{-1}. The hydrogen and the combustion-supporting gas (air) were provided at the flow rates of 30 mL min\textsuperscript{-1} and 380 mL min\textsuperscript{-1}, respectively. The injection temperature and the measured temperature both were 260°C. The pressure prior to columnization was 81.7 kPa, the column temperature was 170°C, the split ratio was 100:1, and the sample size was 1 μL.

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2.2.5 Catalyst selectivity

The linoleic acid selectivity ($S_L$) of the catalyst and the trans isomer value ($S_t$) of oleic acid are calculated as follows:\(^2\)

$$S_L = \frac{k_{S_0} + k_{S_1}}{k_{S_2} + k_{S_3} + k_{S_4}}$$

$$S_t = \frac{\text{trans C18:1 wt} \%}{IV_0 - IV}$$

In the formula, trans C18:1 wt% was the mass percentage of trans oleic acid, IV<sub>0</sub> was the initial iodine value of the rapeseed oil, and IV was the final iodine value of the rapeseed oil.

2.2.6 Dynamics model

According to the reaction pathway of the main fatty acid isomers in the hydrogenation process of rapeseed oil, the hydrogenation kinetics model of rapeseed oil was determined as shown in Fig. 1. Only a small amount of linolenic acid is present in rapeseed oil. Therefore, in the initial stage of the hydrogenation reaction, only the main fatty acid isomers in the process of the hydrogenation of linoleic acid into stearic acid in oils and fats are studied. The isomers of oleic acid are 9c-18:1, 12c-18:1, 9t-18:1 and 12t-18:1.

The seven differential equations for these isomers are solved by the steepest descent method and the fourth order Runge-Kutta method\(^3\):

$$\frac{df_{18:3}}{dt} = -k_{11} f_{\text{linoleic}}$$

$$\frac{df_{\text{linoleic}}}{dt} = k_{11} f_{18:3} - (k_{21} + k_{22} + k_{23} + k_{24}) f_{\text{linoleic}}$$

$$\frac{df_{9c-18:1}}{dt} = k_{22} f_{\text{linoleic}} + k_{41} f_{9c-18:1} - (k_{32} + k_{42}) f_{9c-18:1}$$

$$\frac{df_{12c-18:1}}{dt} = k_{21} f_{\text{linoleic}} + k_{32} f_{12c-18:1} - (k_{31} + k_{32}) f_{12c-18:1}$$

$$\frac{df_{12t-18:1}}{dt} = k_{24} f_{\text{linoleic}} + k_{34} f_{12t-18:1} - (k_{32} + k_{34}) f_{12t-18:1}$$

$$\frac{df_{9t-18:1}}{dt} = k_{21} f_{\text{linoleic}} + k_{41} f_{9t-18:1} - (k_{31} + k_{41}) f_{9t-18:1}$$

$$\frac{df_{12t-18:1}}{dt} = k_{23} f_{\text{linoleic}} + k_{33} f_{12t-18:1} + k_{34} f_{9t-18:1} + k_{33} f_{12t-18:1}$$

By determining the fatty acid content of rapeseed oil during hydrogenation, the rate constant for the transformation of fatty acids during the reaction of the oils and fats was obtained, and the agreement between the fitted values obtained by the model and the experimental values was verified. $f_i$ is the mass fraction of the substance, and $k_i$ is the kinetic rate constant of fatty acid during hydrogenation.

2.2.7 Statistical methods

All of the measurements were carried out in three parallel experiments. The mean values and standard deviations were calculated. The diagrams were drawn according to the data using Origin 8.5. Data analysis and statistical analysis were performed using the SPSS software. The kinetic parameters were analyzed using Matlab 2016.

3 Results and Discussion

3.1 Process parameters

3.1.1 Effects of formate ion concentration

The effect of formic acid ion concentration on the iodine value of low-erucic-acid rapeseed oil during hydrogenation was studied by adding a 0.23 wt. % Ni catalyst at 80°C for 90 min, as shown in Fig. 2.

As observed from Fig. 2, the iodine value of low-erucic-acid rapeseed oil gradually decreases with increasing formate ion concentration. For the formic acid ion concentration of 0.16 mol/50 mL, the reaction proceeds slowly for 60 min. However, as the formate ion concentration increases, the reaction speed increases.

When the formate ion concentration is increased to 0.40 mol/50 mL, the final iodine value obtained at the end of the reaction is only marginally different from the formate ion concentration of 0.32 mol/50 mL\(^4\).

It was reported that when the formic acid ion concentra-

![Fig. 1](image-url)  
**Fig. 1** Kinetic model of hydrogenation of low erucic acid rapeseed oil.

![Fig. 2](image-url)  
**Fig. 2** Effect of the formic acid ion concentration on the iodine value of low erucic acid rapeseed oil during hydrogenation.

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trans fatty acid content are shown in value and effects of the hydrogenation temperature on the iodine a catalyst of 0.23 wt. min in a 0.32 mol/50 mL ammonium formate solution with 3.1.3  Effects of temperature  The hydrogenation of low-erucic-acid rapeseed oil was carried out for 90 min at 80°C and with a formate ion concentration of 0.32 mol/50 mL. The results for the effect of the catalyst loading on the iodine value is shown in Fig. 3.

As observed from Fig. 3, as the catalyst loading gradually increased, the iodine value tended to decrease. For the hydrogenation time of less than 60 min, the iodine value of the oils and fats decreased slowly, showing that the hydrogenation reaction was slow. However, for the catalyst loading of 0.23%, the hydrogenation time began to decrease sharply after 60 min, and the iodine value reached 80 after hydrogenation for 90 min.

When the catalyst loading was increased to 0.25%, the hydrogen atom loading on the catalyst was higher than the supply rate controlled by the formate ion concentration, and the hydrogenation effect was not obvious. Therefore, the loading of the catalyst employed in the hydrogenation process should be set at 0.23%.

3.1.3 Effects of temperature
Low-erucic-acid rapeseed oil was hydrogenated for 90 min in a 0.32 mol/50 mL ammonium formate solution with a catalyst of 0.23 wt.% Ni in 100 g of oil. The results for the effects of the hydrogenation temperature on the iodine value and trans fatty acid content are shown in Fig. 4.

As observed from Fig. 4, as the hydrogenation temperature was gradually increased, the iodine value in hydrogenated oils was also lowered. When the temperature was raised from 60°C to 80°C, the rate of decline in the iodine value was found to be higher, indicating that the rate of hydrogenation of unsaturated fatty acids was greater during this stage. The TFAs content in the hydrogenated oils increased from 4.6% at 60°C to 6.7% at 80°C. However, as the temperature continued to increase to 100°C, the trans fatty acid content increased significantly. Therefore, to reduce the TFAs content in hydrogenated oils, the hydrogenation temperature should be maintained at 80°C.

3.2 Kinetic analysis of the Ni-Ag0.15/SBA15 catalyst in the CTH of low-erucic-acid rapeseed oil
The hydrogenation reaction of vegetable oils was performed using Ni/SBA15 and Ni-Ag0.15/SBA15 as the catalysts. The fatty acid composition of the low-erucic rapeseed oil in the hydrogenation process was measured, and the rate constant of the reaction was calculated by an in-house computer code with the results shown in Table 1.

An examination of the data presented in Table 1 shows that for the reaction rate constant of the hydrogenation of rapeseed oil, since the hydrogenation of linolenic acid to linoleic acid was faster and the linolenic acid content in rapeseed oil was low, this process can be ignored.

In the process of the hydrogenation of linolenic acid into stearic acid in low-erucic rapeseed oil, the reaction rate of the formation of 9c-18:1 oleic acid ($k_{33}$) was much higher than that of the formation of 12c-18:1 ($k_{34}$). Additionally, the reaction rate constant of cis-oleic acid ($k_{23}$) was much greater than that of trans-oleic acid ($k_{21} + k_{22}$).

The rate constant of hydrogenation of trans oleic acid to stearic acid ($k_{31} + k_{34}$) is smaller than the rate constant of hydrogenation of cis-oleic acid to stearic acid ($k_{23} + k_{33}$), and the rate constant of the hydrogenation of linolenic acid to oleic acid ($k_{21} + k_{22} + k_{23} + k_{34}$) is greater than the rate constant of the hydrogenation of oleic acid to stearic acid ($k_{31} + k_{32} + k_{33}$); this trend is consistent with the findings of Fernández et al. However, the fatty acid changes in hydrogenated oils, and cis-trans isomerization of oleic acid are an important focus of this study. The rate constant

![Fig. 3](image1.png)  
**Fig. 3** Effect of the catalyst concentration on the iodine value of low erucic acid rapeseed oil during hydrogenation.

![Fig. 4](image2.png)  
**Fig. 4** Effect of the reaction temperature on the iodine value and TFAs of low erucic acid rapeseed oil during hydrogenation.
of the isomerization of 9c:18:1 oleic acid to 9t:18:1 oleic acid \( k_{42} \) in hydrogenated oils was greater than the rate constant of linoleic acid hydrogenation to 9t:18:1 oleic acid \( k_{21} \). The rate constant for the isomerization of 12c:18:1 oleic acid to 12t:18:1 oleic acid \( k_{52} \) in hydrogenated oils is similar to the rate constant of the linoleic acid hydrogenation to 12t:18:1 oleic acid \( k_{24} \).

The reaction rate constant of the Ni/SBA15-catalyzed hydrogenation of linoleic acid to \( \text{cis} \)-oleic acid \( k_{22} + k_{32} \) was lower than that of the corresponding Ni-Ag0.15/SBA15-catalyzed hydrogenation reaction. The reaction rate constant of the Ni-Ag0.15/SBA15-catalyzed hydrogenation of linoleic acid to \( \text{trans} \)-oleic acid \( k_{21} + k_{24} \) was lower than the corresponding Ni/SBA15-catalyzed hydrogenation reaction. There were no significant differences in the reaction rate constants for the hydrogenation of 9c:18:1 oleic acid and 9t:18:1 oleic acid mutual transformation \( k_{31} \) after the Ni/SBA15 and Ni-Ag0.15/SBA15 catalyzed hydrogenation reaction. In the process of the catalytic hydrogenation of rapeseed oil, the reaction rate and reaction time are linked, and therefore there exists a linear relationship between reaction time and iodine value.

Therefore, fitted values and the corresponding experimental values for the relationship between the iodine value of low-erucic-acid rapeseed oil and various fatty acid contents in the hydrogenated oils after hydrogenation were calculated by the fourth-order Runge-Kutta method, with the results shown in Figs. 5 and 6.

In the initial stage of hydrogenation, a decrease in the iodine value of low-erucic-acid rapeseed oil is observed while the content of linoleic acid decreases rapidly.

When the iodine value of low-erucic-acid rapeseed oil was less than 90 g I\(_2\)/100 g, the content of linoleic acid gradually decreased. When the iodine value of low-erucic-acid rapeseed oil was less than 70 g I\(_2\)/100 g, the content of linoleic acid tends to be stable. In the initial stage of the hydrogenation reaction of oils, the total oleic acid content gradually increases with the decrease in the iodine value of low-erucic-acid rapeseed oil.

However, for iodine value lower than 90 g I\(_2\)/100 g, the total oleic acid content in the hydrogenated oils gradually decreases. This was because as the iodine value decreases, the oleic acid was hydrogenated to stearic acid, so that the oleic acid content gradually decreased and the stearic acid content slightly increased\(^{29}\). The changes in the fatty acid content of low-erucic-acid rapeseed oil shown in Figs. 5 and 6 indicate that the experimental values were consistent with the fitted values.

### 3.3 Catalyst selectivity

The S\(_{LO}\) of linoleic acid and the \( \text{trans} \) isomer value Si of oleic acid in the hydrogenated low-rapeseed oil were calcu-
lated using the relevant data listed in Table 1 with the results shown in Table 2. It is observed from Table 2 that the hydrogenation selectivity of Ni-Ag0.15/SBA15 catalyst for linoleic acid was 2.04, and the trans isomer value of oleic acid was 0.16. The selectivity (\(S_{LO}\)) and activity (Si) of the Ni-Ag0.15/SBA15 catalyst were superior to those of Ni/SBA15, and this change was consistent with the conclusion of Stanković et al.\(^{27}\).

Figures 5 and 6 show that the content of stearic acid in the Ni-Ag0.15/SBA15 hydrogenated low-erucic rapeseed oil was lower than that in the Ni/SBA15 hydrogenated low-erucic rapeseed oil, which was consistent with the high \(S_{LO}\) value for the Ni-Ag0.15/SBA15 catalyst. The content of TFAs in the Ni-Ag0.15/SBA15 hydrogenated low-erucic-acid rapeseed oil was lower than that in the Ni/SBA15 hydrogenated low-erucic-acid rapeseed oil, which was consistent with the low Si values for the Ni-Ag0.15/SBA15 catalyst. This means that the addition of silver increases the dispersion of Ni on SBA15, increasing the specific surface area of the catalyst, so that the amount of hydrogen adsorbed by the catalyst increases, facilitating the conversion to the cis structure in the hydrogenation\(^{27}\).

3.4 **trans Fatty acids and oleic acid content of hydrogenated oils**

The **trans** fatty acid and oleic acid contents of different hydrogenated oils are listed in Table 3. Ni-Ag0.15/SBA15 catalyst was used to hydrogenate different vegetable oils, and the TFAs content of the hydrogenated low-erucic-acid rapeseed oil (CTH conditions) was 4.5% lower than that of the supercritical CO\(_2\) hydrogenated soybean oil. The oleic acid content was 14.73% higher than that of the supercritical CO\(_2\) hydrogenated soybean oil. This means that catalytic transfer hydrogenation of low-erucic-acid rapeseed oil can obtain hydrogenated oils with high oleic acid and low **trans** fatty acids contents.

It was necessary to control the formation of **trans** fatty acids and stearic acid as much as possible to increase the oleic acid content. Although the fatty acid composition of soybean oil and low-erucic rapeseed oil are different, the decrease in the reaction temperature led to a decrease in the **trans** fatty acid content\(^{16}\).

4 **Conclusions**

Using a bimetallic catalyst for the hydrogenation of low-erucic-acid rapeseed oil and the ammonium formate solution as the hydrogen donor, the hydrogenation temperature was lowered by 20°C relative to that of supercritical CO\(_2\) hydrogenation. Ni-Ag0.15/SBA15 hydrogenated low-erucic-acid rapeseed oil, the reaction of the production of linoleic acid from **cis**-structured oleic acid was higher than that of the Ni/SBA15 catalyst, and the reaction rate of the **trans**-isomerization of oleic acid was lower than that obtained using the Ni/SBA15 catalyst. A dynamic model of the hydrogenation of rapeseed oil was developed to describe the hydrogenation process. The results showed that the Ni-Ag0.15/SBA15 catalyst displays high hydrogenation selectivity to linoleic acid, low **trans** isomerization value of oleic acid, and achieves lower **trans**-fatty acid content for the hydrogenated low-erucic-acid rapeseed oil. For the catalytic transfer hydrogenation of low-erucic-acid rapeseed oil, the **trans** fatty acid content was 4.5% lower and oleic acid content was 14.73% higher than those of the supercritical CO\(_2\) hydrogenated soybean oil. Thus, catalytic transfer hydrogenation (CTH) is a promising hydrogenation method.

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| Catalyst          | \(S_{LO}\) | \(S_i\) | TFAs (%) |
|-------------------|------------|--------|---------|
| Ni/SBA-15         | 1.62       | 0.27   | 10.8    |
| Ni-Ag0.15/SBA-15  | 2.04       | 0.16   | 6.7     |

**Table 2** \(S_{LO}\) of linoleic acid and the trans isomer value Si of oleic acid in the hydrogenation.

| Raw material               | Temperature | Hydrogenation method | TFAs (%) | Oleic acid (%) |
|----------------------------|-------------|----------------------|----------|---------------|
| low erucic acid rapeseed oil| 80°C        | CTH                  | 6.7      | 65.00         |
| Soybean oil\(^{17}\)       | 100°C       | Supercritical CO\(_2\) | 11.2     | 50.27         |

**Table 3** Contents of trans fatty acids and oleic acid in different hydrogenated vegetable oils.
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