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ORIGINAL PAPER

High Oleic and Low *Trans* Fatty Acid Formation by an Electrochemical Process

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Abstract Electrochemical hydrogenation employing a mediator of formate/formic acid resulted in partial hydrogenation of vegetable and soybean oil at 20–40 °C and ambient pressure when palladium supported on alumina was employed as a catalyst. An oleic acid content of 48% with a corresponding iodine value of 81 for the vegetable oil hydrogenated at 20 °C was obtained. The total *trans* fatty acid content and especially the 18:1 *trans* fatty acid were found to increase with the reaction temperature and time. Nonetheless, relatively low total *trans* and 18:1 *trans* fatty acid (7 and 3.8%, respectively) contents were found when the vegetable oil was partially hydrogenated to achieve an iodine value of 112.

Keywords Soybean oil · Electrochemical hydrogenation · *Trans* fat · Formate

Introduction

Trans fatty acids which are mainly produced due to edible oil hydrogenation, have been associated with promoting coronary heart diseases. The consumption of *trans* fatty acids results in enhanced levels of low density lipoproteins (LDL) and lowered levels of high density lipoproteins (HDL) [1–3]. The enhanced LDL level in blood serum is viewed as a good predictor of cardiac heart diseases. In addition, high amounts of *trans* fatty acids consumption is associated with causing systemic inflammation and endothelial cell dysfunction [1]. Due to the adverse health effects associated with *trans* fatty acids, there has been many activities in the research community to minimize its formation. Researchers have investigated interesterification, blending of oils, fractionation, and even breeding plants and using genetic manipulation so that the oils have a high oleic acid content. One way to minimize the formation of *trans* fatty acid during hydrogenation of edible oils is to lower the temperature of the reaction. Relatively recently, researchers [4–6] have shown that formic acid can be used to hydrogenate vegetable oil at 100 °C and atmospheric pressure, which is considerably lower than the high temperature (150–225 °C) and high pressure (10–60 psig) employed in commercial processes.

Yusem and Pintauro [7] have shown that an electrochemical process can be employed to produce low *trans* fatty acid containing hydrogenated vegetable oil. In their process, application of current results in the formation of protons at the cathode. A hydrogenation catalyst (nickel) serves as the site for hydrogenation of the oil by adsorbed protons. An et al. [8, 9] introduced a solid polymer electrolyte reactor which previously used in the hydrogenation of organic compounds. List et al. [10] prepared partially hydrogenated soybean oils at palladium/cobalt or palladium/iron cathodes. For iodine values ranging from 90–100, the C18:1 *trans* fatty acid content was found to be in the range of 6.4–13.8%.

Mondal and Lalvani [11–13] have also employed an electrochemical method which is different from the ones

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described above [4–6] in that formate ion serves as the primary source of hydrogen. However, unlike the work by Arkad et al. [4], Weiner et al. [5] and Naglic et al. [6] in which relatively very large quantities of formic acid were used (in the absence of any applied current), formate ion was regenerated at the cathode of an electrolysis cell. Thus, the formate ion mediated hydrogenation of vegetable oil. This work was carried out in the presence of a nickel hydrogenation catalyst.

The current paper examined the role of palladium as a catalyst in the process described by Mondal and Lalvani [11–13]. In addition to the amount of *trans* fatty acids minimization, research was directed at enhancing the amounts of oleic acids formation.

Experimental

Two types of soybean oils, namely vegetable oil (high linolenic acid, 7.2%) and soybean oil (low linolenic acid, 1.2%) were studied. The fatty acid profile of the oils is given in Table 1. All reactions were carried out in a 150-mL laboratory-scale stirred batch glass reactor with a length-to-diameter ratio of 2.95. The reactor was double layered and equipped with a water inlet for continuously charging water into the reactor and a water outlet for releasing water to control the reaction temperature. The anode chamber contained 8 mL of analyte and it was separated from the cathode chamber by a fine glass frit. The anode and the cathode comprised of platinum mesh of surface area of 2.25 and 7.5 cm^2, respectively. A magnetic stirrer was employed to provide agitation and disperse the oil in the electrolyte solution.

Table 1 Characteristics of vegetable oil and soybean oil

Characteristics	Vegetable oil	Soybean oil
Fatty acid (%)		
C16:0	10.2	9.98
C18:0	4.1	4.97
Trans C18:1	0.24	0.10
C18:1 (C9)	22.45	24.34
C18:1 (C11)	1.53	1.36
Trans C18:2	0.29	0.46
C18:2	50.86	55.71
Trans C18:3	0.37	0.32
C18:3	7.23	1.18
C20:0	0.31	0.40
Total (%)	97.58	98.82
Unsaturated (%)	82.07	82.59
Trans (%)	0.90	0.88
Iodine value	129.3	123.4
Condition at 4 °C	Liquid	Liquid
Color	Pale yellow	Pale yellow

A water–oil emulsion was prepared by adding 45 mL of oil to 105 mL of water containing 0.26 g of didodecyl diethyl ammonium bromide (DDAB), a food grade emulsifier. A mediator (hydrogen donor) consisting of 5.1 g of sodium formate and 1.5 mL formic acid, and 0.3 g palladium hydrogenation catalyst (Acros, New Jersey, USA) were added to the mixture. The anode compartment contained 0.25 g of formate, 0.8 mL of formate acid and 0.5 g of KCl dissolved in 7.2 mL of water. A Dynatronic power supply was employed to apply the desired current to the reactor.

After the experiment, the emulsion was left to sit in a beaker for about 30 min which resulted in the separation of the oil and water phases. The oil phase was subjected to a centrifugation which resulted in the separation of water and the solid catalyst. After filtering the oil again, 20 mL of *n*-heptane were added to it. After evaporation of the *n*-heptane, the oil samples were analyzed for its composition.

Product Analyses

Fatty acid compositions of the initial oil and hydrogenated oil products were analyzed by a gas chromatography (Agilent 6890, Palo Alto, CA, USA) equipped with a flame ionization detector (AOCS method Ch 21–94) [14]. A capillary column (SP-2560, Supelco Inc., Bellefonte, PA, USA) of 100 m × 0.25 mm with 0.2 μm film thickness was used. The oven temperature of GC was programmed at 140 °C for 5 min, increased to 220 °C at 4 °C/min, and maintained at 220 °C for 15 min. The 1 μL sample was injected into the injection port with the split ratio of 100:1 at 220 °C. Helium gas was used as a carrier gas at 1 mL/min. The temperature of a detector was 250 °C. The fatty acids were identified by comparing with the GC retention time of a reference fatty acid.

Only 18-carbon fatty acids are of interest for edible oil hydrogenation. The percent of total *trans* isomers in the initial oil and hydrogenated oil products was determined by Fourier transform infrared spectroscopy (AOCS method Cd 14–95) [14]. The overall degree of oil hydrogenation is typically quantified by the iodine value (IV), defined as centigrams of iodine that react with 1 g of oil (I_2 reacts with the double bonds in unsaturated fatty acids).

Calculation of Iodine Value

The iodine value for oil samples was determined directly from the GC fatty acid composition (AOCS method Cd 1c–85) [14], using the following formula:

$$\begin{aligned}\text{Iodine value} = {} & [(\%\text{C18:1} + \%\text{trans C18:1}) \times 0.86] \\ & + [(\%\text{C18:2} + \%\text{trans C18:2}) \times 1.732] \\ & + [(\%\text{C18:3} + \%\text{trans C18:3}) \times 2.616].\end{aligned}$$

Statistical Analysis

Samples were prepared in duplicate. Analysis of variance was used to analyze data using SAS 9.1 (SAS Institute Inc., Cary, NC, USA) at $\alpha = 0.05$.

Results and Discussion

Reproducibility of Fatty Acid Analysis

The reproducibility of the fatty acid compositions by GC was determined by running four replicates of the references for stearic, oleic, linoleic, and linolenic acids. The coefficients of variation for measuring stearic, oleic, linoleic, and linolenic acid were 1.3, 1.3, 1.4, and 1.4%, respectively. The low coefficients of variations show that the method for the fatty acid analysis was reproducible.

Premixing of Reagents

The influence of the presence of a glass frit between the anode and cathode compartment, the addition of hydrogen donor, and the application of electric current were studied and shown in Table 2.

When the reaction was conducted at 20 °C for 24 h in a reactor where the anode and cathode compartments were separated by a glass frit and the electric current was applied, the iodine value of vegetable oil decreased from 129.3 to 80.7 (37.6% hydrogenation, Exp. 1). However, when the same experiment was conducted without a glass frit (Exp. 2), almost no hydrogenation was observed. This might be due to the following reasons: (a) the recombination of protons (H^+) and hydroxide ions (OH^-) which are formed at the cathode and anode, respectively, (b) the oxidation of formate ions ($HCOO^-$) at the anode to CO_2 resulting in a loss of the hydrogen donor, and (c) the competitive adsorption of hydroxide ions on the catalyst surface.

The combination of sodium formate and formic acid was used as a hydrogen donor. When the reaction was carried out in the absence of hydrogen donor, appreciable hydrogenation (29.4%) was observed (Exp. 3). It appears that hydrogen produced from the cathode accomplished the hydrogenation of oil—a finding reported earlier by List et al. [10]. However, the presence of hydrogen donor, formate/formic acid significantly enhanced the reduction of vegetable oil. The effect of the electric current on the hydrogenation of vegetable oil was studied. When no current was employed to the reactor and formate/formic acid was added to the reaction mixture, the iodine value decreased from 129.3 to 126.2 (Exp. 4). Previous research data show that only large quantities of formate/formic acid can accomplish significant oil hydrogenation when the reaction temperature is kept at 100 °C [4–6]. Similarly, no hydrogenation was accomplished in the absence of both current and a hydrogen donor (Exp. 5). This result suggested that the electric current was important to hydrogenate oil at low concentrations of the donor.

The influence of nitrogen gas purging on the hydrogenation of vegetable oil was studied and shown in Table 3. The extent of hydrogenation was 8.4% when nitrogen purging was employed. This was considerably greater than the corresponding hydrogenation of 4.2% obtained in the absence of nitrogen purging. The data suggest that dissolved oxygen in the reaction medium inhibits hydrogenation, possibly by the oxidation of formate to bicarbonate. Oxygen reduction at the cathode could compete with the electrochemically regenerated formate ion at the cathode.

The influence of the addition order of hydrogen donor and oil during reagent mixing was studied and shown in Table 4. In the case of soybean oil, the extent of hydrogenation was 11.9% when formate/formic acid were added last to the reaction mixture. However, the extent of hydrogenation increased to 16% when oil was added last to

Table 2 Effect of glass frit, hydrogen donor, and electric current on the hydrogenation of vegetable oil at 20 °C for 24 h

Exp.	Reaction condition					Iodine value	*Trans* fat (%)	Hydrogenation (%)
	DDAB	Pd	Formate/formic acid	Current (0.15 A)	Glass frit			
	Unreacted vegetable oil					129.3	0.9	
1	○	○	○	○	○	80.7	24.7	37.6
2	○	○	○	○	–	126.4	1.3	2.2
3	○	○	–	○	○	91.3	19.5	29.4
4	○	○	○	–	○	126.2	1.1	2.4
5	○	○	–	–	○	126.0	1.1	2.6

○, added condition to the experiment; –, not added

Table 3 Influence of N_2 purging on the hydrogenation of vegetable oil at 40 °C for 8 h

Exp.	Reaction condition					Iodine value	*Trans* fat (%)	Hydrogenation (%)
	DDAB	Pd	Formate/formic acid	Current (0.15 A)	N_2			
	Unreacted vegetable oil					129.3	0.9	
1	○	○	○	○	–	123.8	3.9	4.2
2	○	○	○	○	○	118.4	6.8	8.4

○, added condition to the experiment; –, not added

Table 4 Influence of the addition order of formate/formic acid and oil during premixing

Reaction conditions		Iodine value	*Trans* fat (%)	Hydrogenation (%)
	Unreacted vegetable oil	129.3	0.9	
Soybean oil	(H_2O, DDAB, Pd) + formate/formic acid + oil	108.6	10.8	16.0
	(H_2O, DDAB, Pd) + oil + formate/formic acid	113.9	7.7	11.9
Vegetable oil	(H_2O, DDAB, Pd) + formate/formic acid + oil	115.5	8.5	10.4
	(H_2O, DDAB, Pd) + oil + formate/formic acid	115.9	9.4	10.7

Table 5 Influence of different hydrogen donors on the hydrogenation of vegetable oil

Exp.	Reaction conditions				Iodine value	*Trans* fat (%)	Hydrogenation (%)
	Hydrogen donor	Temp (°C)	Initial pH	Reaction (h)			
	Unreacted vegetable oil				129.8	1.0	
1	Na-formate	45	7.8	8	121.7	5.6	5.9
		65	7.8	8	114.8	14.2	11.2
2	K-formate	25	7.8	8	115.3	11.1	10.8
		45	7.8	8	113.2	11.1	12.5
		65	7.8	8	113.4	18.7	12.3
3	Formic acid	25	1.7	8	129.0	1.0	0.2
		45	1.7	8	128.7	1.1	0.5
		65	1.7	8	128.0	1.7	1.0
4	Na-formate + formic acid	25	4	8	115.5	9.4	10.7
		45	4	8	113.5	10.2	12.2
		45	4	12	107.1	14.7	17.2
5	K-formate + formic acid	45	4	8	113.3	11.9	12.4
		65	4	8	113.6	20.6	12.1

the reaction mixture. This observation suggests that there was involved a competitive adsorption between formate/formic acid and oil. Since the amount of formate/formic acid employed in hydrogenation is relatively much lower than that of oil, the overall reaction can be limited by the amount of formate/formic acid. However, almost no influence of the addition order of oil and formate/formic acid on the hydrogenation of vegetable oil was observed. Naglic et al. [6] suggested that hydrogenation with a hydrogen donor in aqueous medium proceeds in a complex three-phase system (oil–water-solid catalyst). The reaction involves competitive adsorption between water and formate/formic acid on the identical active sites of the catalyst surface. Both sodium formate and water are hydrogen donors in the reaction.

The influence of different hydrogen donors on the hydrogenation of vegetable oil was studied and is shown in Table 5. The hydrogen donor is a very important component in the reaction. Sodium formate, potassium formate, formic acid, sodium formate with formic acid and potassium formate with formic acid were added to the reaction mixture at different temperatures to choose the best

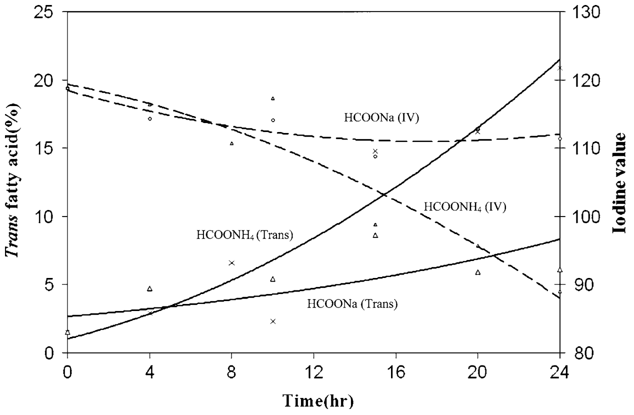

Fig. 1 Influence of sodium formate (HCOONa) and ammonium formate ($HCOONH_4$) on *trans* fatty acid contents and iodine value of soybean oil during hydrogenation at 20 °C

hydrogen donor. The use of sodium formate and potassium formate resulted in a significant extent of hydrogenation (greater than 10%). However, the use of sodium formate and potassium formate resulted in the increase of *trans* fatty acid contents (Exp. 1, 2). The data also suggest that the increase of temperature led to the increase of hydrogenation. It has been shown that the catalytic transfer hydrogenation is a second order reaction and the rate of reaction depends on the concentration of hydrogen donor [9]. When formic acid only was used as a donor (Exp. 3), very low hydrogenation was observed. The degree of hydrogenation in the presence of formic acid may be due to the decomposition of formic acid in the presence of a metal. It is also possible that at the low pH 1.7 used in the reaction, the predominant electrochemical reaction was hydrogen evolution but very little regeneration of the mediator. The use of potassium formate and sodium formate with formic acid (Exp. 4, 5) resulted in a similar extent of hydrogenation. As the reaction time increased (Exp. 4), both the extent of hydrogenation and the *trans* fatty acid contents increased.

The influence of hydrogen donors consisting of sodium formate and ammonium formate on the *trans* fatty acids and iodine value of soybean oil during hydrogenation is shown in Fig. 1. When sodium formate was employed, the hydrogenation extent (i.e. decrease in iodine value) increased as the reaction time increased. However, the use of ammonium formate accomplished much greater hydrogenation (25%) than sodium formate after 24 h of reaction. *Trans* fatty acid contents increased with time and it was a direct function of the extent of hydrogenation. The possible reason that the hydrogenation reaction proceeds slowly in the presence of sodium formate might be due to the formation of alkali salts in the reaction mixture (Na_2CO_3 and $NaHCO_3$) which affects adsorption of the reactants and leads to palladium hydride formation on the active sites. In addition, the sodium carbonate releases CO_2 that causes frothing in the system and is highly undesirable when it is neutralized to adjust for acidic pH for waste treatment.

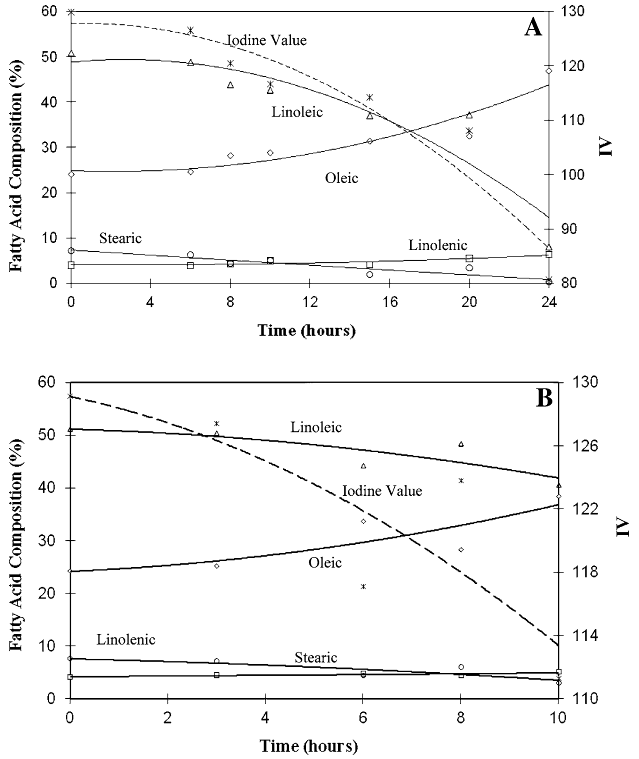

Fig. 2 Effect of reaction time on the iodine value and stearic, oleic, linoleic, and linolenic acid contents during the hydrogenation of vegetable oil at 20 °C (**a**) and 40 °C (**b**)

Reaction Time

The influence of reaction time on the iodine value and the fatty acid composition of the hydrogenated vegetable and soybean oil were studied. The experiments were carried out using a constant current of 0.15 A. The oil–water emulsion contained 0.26 g DDAB as an emulsifier and 0.3 g Pd/aluminum catalyst was added.

Figure 2 shows the iodine value and stearic, oleic, linoleic, and linolenic acid contents during the hydrogenation of vegetable oil at 20 and 40 °C, respectively. As the reaction time increased, the iodine value significantly decreased and the hydrogenation increased. The vegetable oil was hydrogenated up to 37.8% at 20 °C for 24 h of processing (Fig. 2a). The hydrogenation achieved 14% for 10 h at 40 °C (Fig. 2b), which is 3.9% greater than the corresponding hydrogenation at 20 °C (Fig. 2a). The fatty acid profile data show that oleic acid content increased from an initial value of 24.0 to 28.8% in 10 h at 20 °C (Fig. 2a). The corresponding oleic acid content of the oil at 40 °C was 38.4%. The linoleic acid content at 20 °C decreased sharply as the reaction time increased. However, the decrease in linoleic acid content at 40 °C was relatively modest. The stearic acid and linolenic contents did not significantly increase or decrease both at 20 and 40 °C. Thus, it appears the reaction rate constant for the reduction of linoleic to oleic acid is much more significant than the corresponding rate constant for linolenic to linoleic acid and oleic to stearic acid hydrogenation.

The *trans* fatty acid content profiles of vegetable oil during the hydrogenation at 20 and 40 °C are shown in Fig. 3. Total *trans* fatty acid contents increased as the reaction time and temperature increased. The *trans* fatty acid contents increased to 5.6 and 9.6% at the end of 10 h of hydrogenation at 20 and 40 °C, respectively. There was a strong inverse relationship between the iodine value and the *trans* fatty acid content. Significant increases in C18:1 and C18:2 *trans* fatty acid contents were observed when either the reaction time or the temperature of the reaction was increased.

Selectivity

Adsorption of the reactants onto the catalyst surface is important in controlling the selectivity and isomerization during hydrogenation. The hydrogen bond enables unsaturated fatty acids to contact the catalyst surface. The selectivity ratio of different fatty acids depends on geometric configuration and chemical and physical characteristics of the catalyst.

In this study, the preferential hydrogenation is to produce more monounsaturated acid with a minimum formation of saturated fatty acids and *trans* fatty acids. The selectivity of an individual component is defined as the change of the fatty acid content (%) per the change of the iodine value. The following is an equation used to calculate the selectivity:

$$S_j = \left| \frac{\left[\frac{j_{\mathrm{final}} - j_{\mathrm{inital}}}{j_{\mathrm{initial}}}\right]}{\left[\frac{\mathrm{IV}_{\mathrm{initial}} - \mathrm{IV}_{\mathrm{final}}}{\mathrm{IV}_{\mathrm{initial}}}\right]} \right| \qquad (1)$$

where, S is selectivity and j is the fatty acid composition (%) such as stearic, oleic, linoleic, and linolenic acid.

The selectivities of stearic, oleic, linoleic, and linolenic acids during hydrogenation at 20 and 40 °C were determined and are shown in Fig. 4. The selectivity of stearic, oleic, and linoleic acid increased during hydrogenation at 20 °C (Fig. 4a). Whereas the selectivity of linolenic acid decreased with the reaction time, the selectivity of other fatty acids increased. The results suggest that the rate

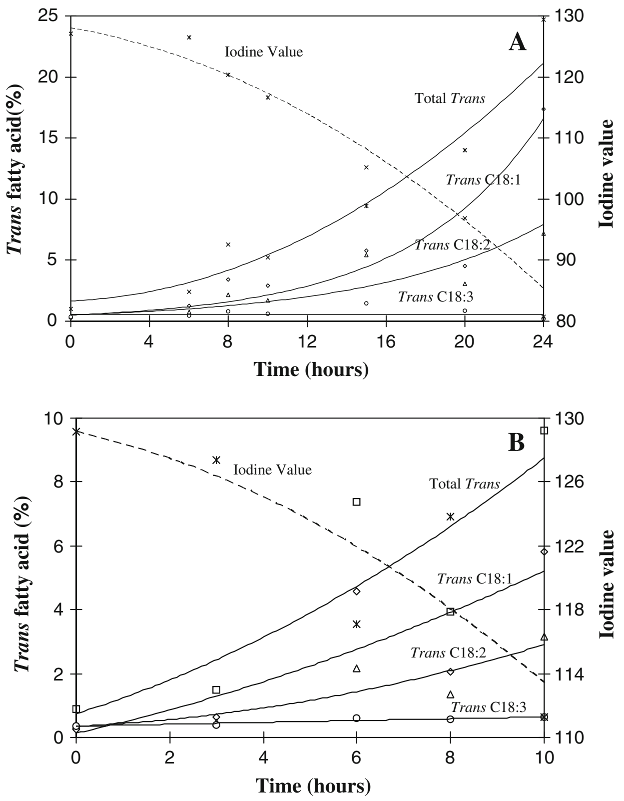

Fig. 3 Effect of reaction time on the iodine value and *trans* fatty acids during the hydrogenation of vegetable oil at 20 °C (**a**) and 40 °C (**b**)

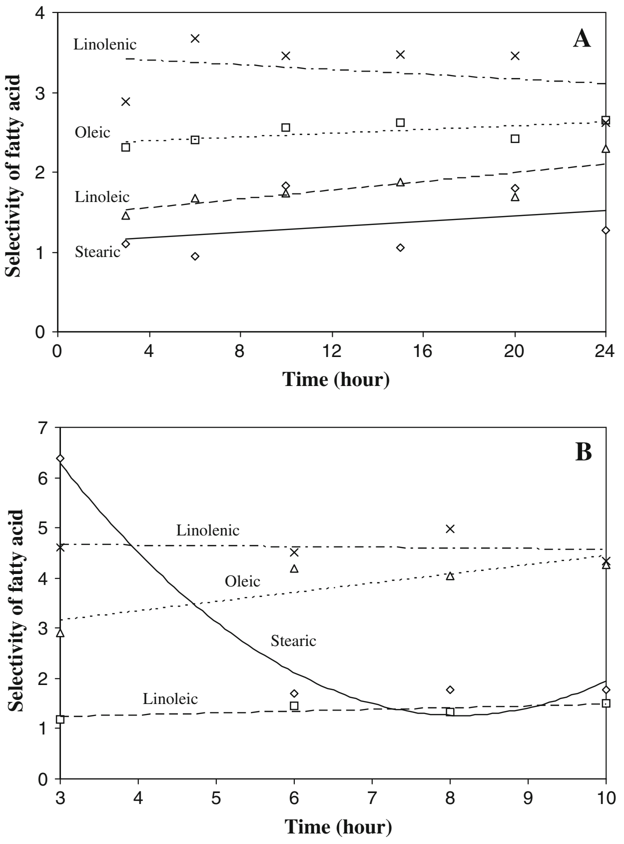

Fig. 4 Selectivity of stearic, oleic, linoleic, and linolenic acids during the hydrogenation of vegetable oil at 20 °C (**a**) and 40 °C (**b**)

constant for the formation of linolenic acid was the highest, whereas the stearic acid formation was the lowest. The selectivity of stearic acid was initially the highest and rapidly decreased with the reaction time during hydrogenation at 40 °C (Fig. 4b). The selectivity of oleic acid increased with the reaction time, whereas the selectivity of linolenic and linoleic acid remained relatively unaffected.

The selectivity of *trans* fatty acids are shown in Fig. 5. The selectivity of *trans* fatty acid increased as the reaction time increased. The selectivity of *trans* fatty acid at 40 °C was higher than the selectivity of *trans* fatty acids at 20 °C. The selectivity of *trans* C18:1 fatty acid was the highest both at 20 and 40 °C. The data clearly show that temperature has a profound effect on enhancing the formation of *trans* fatty acids, an observation which is in agreement with previous published data [6–8, 15]. As compared to the selectivity of *cis* fatty acids, the selectivity of *trans* fatty acids was much greater.

Current

The effect of the applied electric current on *trans* fatty acids and iodine value, and the fatty acid profile of vegetable oil and soybean oil during hydrogenation at 20 °C for 24 h are shown in Figs. 6 and 7. As the electric current increased from 0.08 to 0.13 A, the iodine value of the vegetable oil decreased and the hydrogenation increased (Fig. 6a). However, as the current increased further to 0.15 A, the hydrogenation decreased. As noted earlier, the iodine value decreased along with the increase of total *trans* fatty acid content (Fig. 6a). In the case of soybean oil hydrogenation, a similar relationship between the iodine value and the *trans* fatty acid content were observed (Fig. 6b). The maximum hydrogenation was observed for an experiment carried out using a current of 0.10 A. The fatty acid profile for the vegetable oil (Fig. 7a) shows that oleic acid content increased as the current increased from 0.08 to 0.13 A (44% of oleic acid). However, a further increase of the current led to the decrease of oleic acid contents. Both linoleic and linolenic acid contents are observed to decrease as the current decreased from 0.08 to 0.13 A; however, the opposite trend is observed at a higher current of 0.15 A. The fatty acid profile of soybean oil was very similar to that of the vegetable oil (Fig. 7). The data show that maximum oleic acid content is observed when hydrogenation is carried out at 0.10 A. It is interesting to note that stearic acid content did not increase substantially.

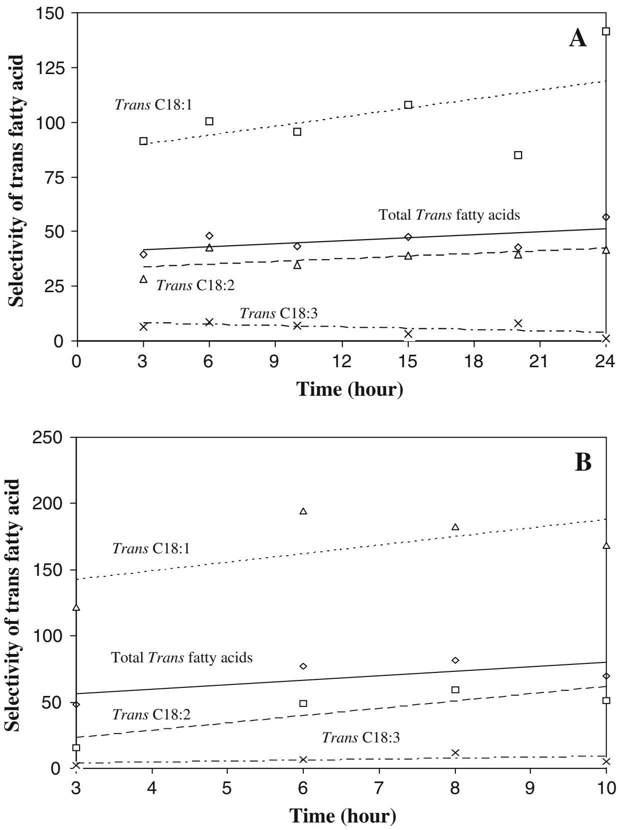

Fig. 5 Selectivity of *trans* fatty acids during the hydrogenation of vegetable oil at 20 °C (**a**) and 40 °C (**b**)

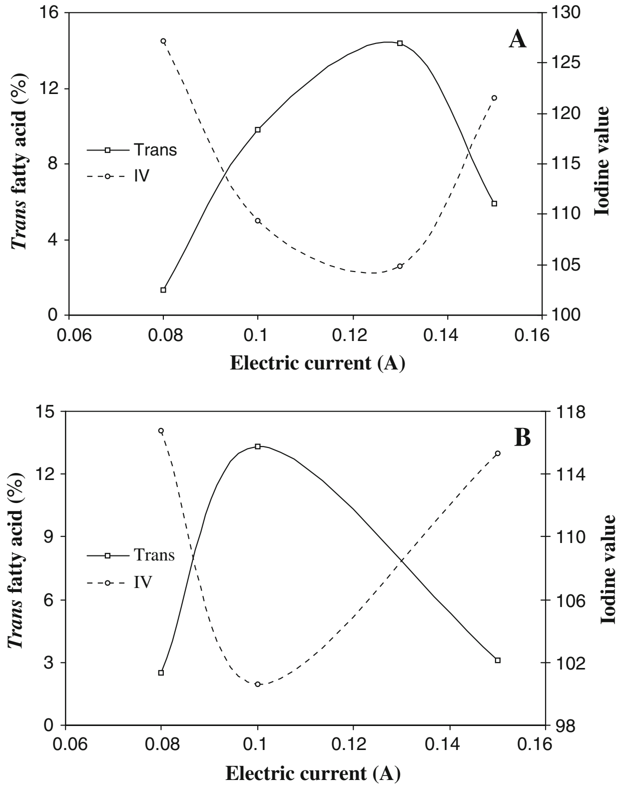

Fig. 6 Effect of the electric current on *trans* fatty acids and iodine value of vegetable oil (**a**) and soybean oil (**b**) during hydrogenation at 20 °C for 24 h

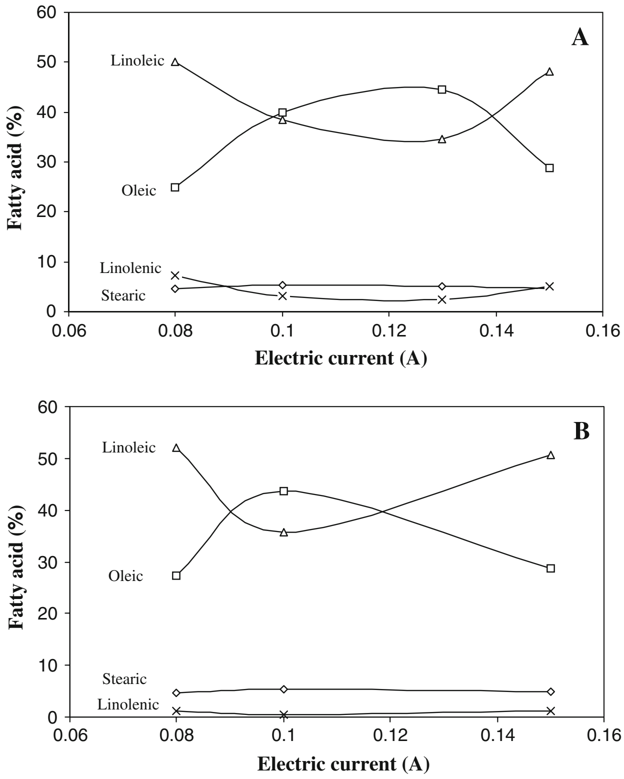

Fig. 7 Effect of the electric current on the fatty acid composition of vegetable oil (**a**) and soybean oil (**b**) during hydrogenation at 20 °C for 24 h

Table 6 Effect of the reuse of hydrogen donor solution on the fatty acid composition, iodine value, *trans* fatty acids and hydrogenation of soybean oil

	First run	Second run	Third run
Fatty acid (%)			
C16:0	10	10	10
C20:0	0.40	0.43	0.41
C18:0	6.88	7.33	7.81
C18:1	67.12	63.26	56.51
C18:2	13.44	16.88	23.1
C18:3	13.44	16.88	23.1
Iodine value	81.4	84	89.1
Trans fatty acids (%)	23.9	18.2	15.4
Hydrogenation (%)	33.4	31.3	27.1

At relatively low current densities, the hydrogenation of vegetable oil by formate/formic acid and its regeneration reactions are predominate. However, as the applied current increased, high amounts of hydrogen gas were formed due to the proton reduction at the cathode resulting in lowered faradic efficiency for formate regeneration and thus lowered hydrogenation extent. It is also possible that occlusion of a hydrogen gas film at the cathode limits the transport of bicarbonate and formate ions to and from the cathode.

Reusability of Hydrogen Donor (Mediator)

The reusablility of hydrogen donor on the hydrogenation of soybean oil was studied. Soybean oil was first electrochemically hydrogenated using formate/formic acid. The oil phase was separated and analyzed. The aqueous phase of the solution containing the palladium catalyst was added to a new batch of soybean oil and then electrochemically hydrogenated. All experiments were carried out at 20 °C for 24 h. The fatty acid composition and iodine value of the hydrogenated soybean oil are shown in Table 6.

The hydrogenation of soybean oil was achieved to 33.4% for the first use. Upon the reuse of the aqueous phase, the extent of hydrogenation decreased. The iodine value in the subsequent experiment increased slightly because some catalyst and a part of the mediator of formate were lost during the process. The experimental results suggested that the aqueous phase of the reaction mixture significantly retains its activity and could be reused for further hydrogenation.

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