The Electrocatalytic Hydrogenation of Soybean Oil

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Soybean oil has been hydrogenated electrocatalytically at a moderate temperature, without an external supply of pressurized H_2 gas. In the electrocatalytic reaction scheme, atomic hydrogen is produced on an active Raney nickel powder cathode surface by the electrochemical reduction of water molecules from the electrolytic solution. Adsorbed hydrogen then reacts with an oil's triglycerides to form a hydrogenated product. Experiments were carried out at 70°C with a flow-through electrochemical reactor operating in a batch recycle mode. The reaction medium was a two-phase mixture of soybean oil in a water/t-butanol solvent containing tetraethylammonium *p*-toluenesulfonate as the supporting electrolyte. In all experiments the reaction was allowed to continue for sufficient time to synthesize a brush hydrogenation product. The effects of oil content, applied current, solvent composition, and supporting electrolyte concentration on the efficiency of hydrogen addition to the oil and on the chemical properties of the hydrogenated oil product were determined. The electrohydrogenated oil is characterized by a high stearic acid content and a low percentage of total *trans* isomers, as compared to that produced in a traditional hydrogenation process.

KEY WORDS: Current, current efficiency, electrochemical, electrolyte, hydrogenated, Raney nickel, soybean oil.

The hydrogenation of the unsaturated fatty acid constituents of an edible oil's triglycerides is carried out to produce a more oxidatively stable product and/or to change a normally liquid oil into a semi-solid or solid fat with melting characteristics designed for a particular application. Most commercial oil hydrogenation plants use batch reactor equipment and Raney or supported nickel catalyst. The reaction is carried out at a high temperature (typically 150–225°C) and a hydrogen gas pressure in the range of 10–60 psig (1). These conditions are required to solubilize sufficiently high concentrations of hydrogen gas in the oil/catalyst reaction medium, so that the hydrogenation reaction can proceed at acceptably high rates. The hydrogenation rate and product distribution has been shown to be dependent mainly on temperature, pressure, agitation rate, and catalyst type and loading (2,3). Unfortunately, high reaction temperatures promote a number of deleterious side-reactions, including the unfavorable production of *trans* isomers and the formation of cyclic aromatic fatty acids (4).

This paper describes a new low-temperature electrocatalytic process for the hydrogenation of edible oils, where an electrically conducting catalyst (*e.g.*, Raney nickel) is used as the cathode in an electrochemical reactor. The reaction medium must contain both oil and a water- or proton-containing solvent. Atomic hydrogen is generated on the catalyst surface by the electrochemical reduction of solvent species from the adjacent electrolytic solution. The electrogenerated hydrogen then reacts chemically with unsaturated fatty acids in the oil by the following reaction sequence:

$$H_2O + e^- \rightarrow H_{ads} + OH^- \quad [1]$$

$$2H_{ads} + R\text{-}CH{=}CH\text{-}R' \rightarrow R\text{-}CH_2\text{-}CH_2\text{-}R' \quad [2]$$

An unwanted side reaction, which consumes current but does not effect the product yield, is the formation of H_2 gas by the combination of two adsorbed hydrogen atoms:

$$2H_{ads} \rightarrow H_2(\text{gas}) \quad [3]$$

All electrochemical reactors must contain two electrodes, a cathode (for reduction reactions) and an anode at which one or more oxidation reactions occur. For a water-based electrolytic solution, the anode reaction is often the oxidation of H_2O:

$$1/2H_2O \rightarrow 1/4O_2 + H^+ + e^- \quad [4]$$

By proper design and operation of an electrocatalytic hydrogenation reactor, the bulk pH of the reaction medium remains unchanged with time due to mixing of the alkaline solution near the cathode with acid generated at the anode.

In an electrocatalytic reaction scheme, hydrogen is generated *in situ* directly on the catalyst surface, so high operating temperatures and pressures are not required. By maintaining a low reaction temperature, it may be possible to minimize unwanted isomerization reactions, thermal degradation of the oil, and other deleterious reactions. By passing a high current through the catalyst (*i.e.*, maintaining a high concentration of atomic hydrogen on the catalyst surface), the hydrogenation rate of the oil may be kept high, even at atmospheric pressure and a low or moderate reaction temperature. Such an electrocatalytic method circumvents the need to produce, store, compress, and transport H_2 gas. Additional advantages of an electrocatalytic hydrogenation process are: i) The concentration of hydrogen on the catalyst metal surface can be easily controlled by adjusting the applied current (or electric potential), which may lead to improved product selectivity; ii) the catalyst will be cathodically polarized during reactor operation, resulting in less corrosion and lower concentrations of nickel ion contaminants in the oil product; and iii) there is little free hydrogen gas present, thus the risk of explosion and fire is reduced.

Numerous studies have shown that low hydrogen overpotential electrically conducting catalysts (*e.g.*, Raney nickel, platinum and palladium black on a carbon rod, and Devarda copper) can be used to electrocatalytically hydrogenate a variety of organic compounds, including benzene and multi-ring aromatic compounds, phenol, ketones, nitrocompounds, dinitriles, and glucose (5–12). These reactions were carried out in both batch and semi-continuous flow reactors. In most cases the reaction products were similar to those obtained from a traditional chemical catalytic scheme at elevated temperatures and pressures.

The apparatus, procedures, and results of preliminary soybean oil hydrogenation experiments are presented below. The reaction was carried out in a flow-through electrochemical reactor with a Raney nickel catalyst powder packed-bed cathode. A two-phase reaction medium consisting of oil, a

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water/t-butanol solvent, and a supporting electrolyte salt of tetraethylammonium *p*-toluenesulfonate (hereafter denoted as TEATS) was employed in all experiments. This solvent/supporting electrolyte system has been used previously in the electrocatalytic hydrogenation of benzene (5). The effects of solution composition and applied current on the performance of the electrocatalytic reaction and on the chemical properties of the hydrogenated oil product were determined.

EXPERIMENTAL PROCEDURES

Electrochemical reactor. The oil hydrogenation was carried out in the packed-bed flow-through electrochemical reactor shown schematically in Figure 1. The cylindrical reactor (20 cm^2 in cross-sectional area and 15.0 cm in length) was constructed of glass with a fine porosity glass frit separating the anode and cathode compartments of the cell. The anode was a 4.0 cm diameter DSA (Dimensionally Stable Anode) expanded titanium grid coated with IrO_2. At the anode, water in the reaction medium is oxidized to oxygen and protons (Eq. [4]). The cathode consisted of approximately 37 grams of finely divided Raney nickel powder (Catalyst Type BK111, active catalyst received in water; Degussa Corp., Ridgefield Park, NJ). The solution was pumped in the upward direction, pressing the nickel catalyst against the glass frit and forming a packed bed ≈0.9 cm in thickness. The direction of flow also minimized pH excursions of the electrolytic solution. A nickel wire mesh placed at the upstream end of the nickel bed served as the current collector. Electrical contact to the collector was provided by a nickel rod. Glass beads were placed above and below the electrodes to reduce the void volume in the reactor and to ensure a uniform flow distribution through the cell. The reactor was operated in a batch recycle mode by insertion into a circulation loop, which normally consisted of a variable-speed magnetic-drive gear pump (Model 120, Micropump Corp., Concord, CA) and a holding tank immersed in a constant-temperature water bath (see Fig. 2). A magnetic stirrer in the holding tank provided sufficient agitation to fully disperse the oil in the water/alcohol solvent. All hydrogenation experiments were carried out under constant current conditions (either 0.20, 0.30, or 0.40 Amps) with a Hewlett-Packard Model 6294A DC power supply (Hewlett-Packard, Palo Alto, CA). The current was measured with a multimeter (Model 169, Keithley Instruments, Cleveland, OH), and the total electrical charge passed during

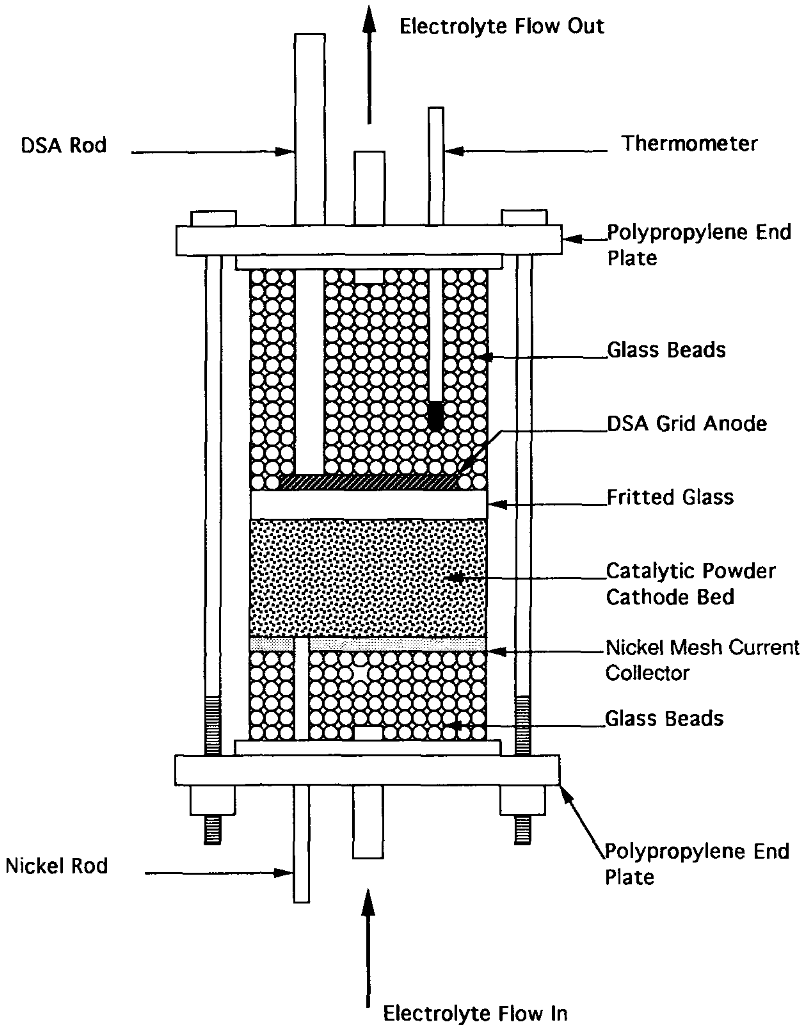

FIG. 1. Schematic diagram of the flow-through electrocatalytic hydrogenation reactor.

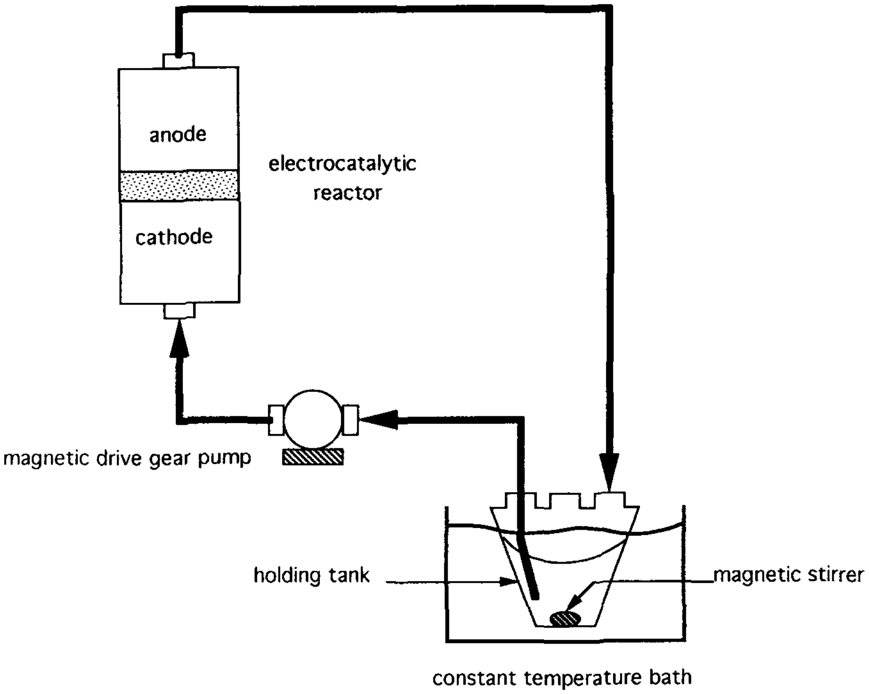

FIG. 2. Diagram of the batch recycle circulation loop.

an experiment was counted on a digital coulometer (Model 640, The Electrosynthesis Company, Inc., Amherst, NY).

To ensure that saturated oil detected at the conclusion of an electrolysis was produced *via* electro-generated H_{ads}, the Raney nickel was washed *in situ* with soybean oil prior to an electrolysis to deplete the catalyst of adsorbed hydrogen. First, water was removed from the catalyst by circulating three 200-mL aliquots of t-butanol through the reactor. Next, 200 mL of a 60:40 wt% soybean oil/t-butanol solution at 70°C was pumped through the reactor (under zero current conditions) for 12 hr. The oil solution was emptied from the reactor/recycle loop apparatus, and the catalyst was rinsed numerous times *in situ* with 200-mL portions of t-butanol until no oil was detected in an alcohol washing.

In a typical oil hydrogenation electrolysis, 100 mL of a water/t-butanol/TEATS electrolyte solution containing a known weight of soybean oil (either 24, 36, or 48 grams) was introduced into the reactor/recycle loop. The reaction mixture was pumped through the reactor at 20 mL/min. A constant DC current was applied for a specific period of time (between 4.5 and 17.5 hr, depending on the weight of oil in the reaction medium and the applied current) to reduce (theoretically) ≈25% of the double bonds in the initial oil. At the conclusion of an experiment, the two-phase electrolyte was withdrawn from the reactor and the Raney nickel catalyst bed was washed seven times *in situ* with 200-mL volumes of t-butanol (each washing lasted 1 hr). The electrolyte, oil, and butanol wash solutions were combined, and the alcohol was removed under vacuum by rotary evaporation. An aqueous solution of Na_2SO_4 and 100 mL of n-hexane were then combined with the oil/water/TEATS supporting electrolyte mixture in a separatory funnel. The top layer of hexane and oil was washed three times with water to remove all traces of TEATS salt. After vacuum evaporation of the n-hexane, the oil sample was analyzed for its chemical properties. With this extraction procedure, 95–100% of the initial oil was recovered at the conclusion of an electrolysis.

Materials. The two-phase electrolytic reaction medium was prepared from reagent-grade chemicals and deionized and distilled water. Refined soybean oil, provided by Capital City Products Co. (Columbus, OH) was used in all experiments. The electrolyte solution consisted of water, t-butanol cosolvent (Aldrich Chemical Co., Milwaukee, WI), soybean oil, and the hydrotropic salt tetraethylammonium *p*-toluenesulfonate (Aldrich Chemical Co.). t-Butanol was chosen as the cosolvent because this alcohol will not be oxidized when it contacts the anode. The use of TEATS as the supporting electrolyte in organic electrochemical syntheses is not new; the presence of this salt has been shown to improve the performance (energy efficiency) and/or product yield in such diverse electrochemical reactions as the electrohydrodimerization of acrylonitrile to adiponitrile (13) and the aqueous-phase electrocatalytic hydrogenation of benzene to cyclohexane (5). In the present study, the primary purpose of the TEATS was to increase the electrical conductivity of the water/alcohol solvent. Both t-butanol cosolvent and TEATS contributed to a lowering of the interfacial surface tension of the dispersed oil droplets, as evidenced by the visual observation of finely dispersed oil droplets in the holding tank.

Determination of chemical properties. Methyl esters of the initial and hydrogenated oil samples were prepared according to AOCS method Ce 2-66 (14). The esters were analyzed by gas chromatography (GC) (AOCS method Ce 1-62) in a Hewlett-Packard Model 5890 chromatograph with a flame ionization detector and a 1/8-inch stainless steel packed column (6 feet in length) containing 3%

SP-2301/2% SP-2300 on 100/120 Chromosorb W AW (Supelco Inc., Bellefonte, PA). The GC was operated isothermally at 200°C with an injector and detector temperature of 250°C. The carrier gas was N_2 at a flow rate of 20 mL/min. Confirmation of fatty acid peaks was achieved by comparison with the retention times of authentic samples available commercially. The weight of each fatty acid was determined from the relative peak areas and the total grams of oil extracted from the electrolyte. The iodine value for the oil was determined directly from the GC fatty acid composition (AOCS method Cd 1c-85). The total isolated *trans*-isomer content of hydrogenated oil samples was determined by infrared spectrophotometry (Mattson Instruments FTIR, Madison, WI), as described by AOCS method Cd 14-61. The free fatty acid content of the oil was determined by base titration (AOCS method Ca 5a-40), and the nickel content in the oil product was found by atomic absorption spectrophotometry (AOCS method Ca 15-75). In the present study, no attempt was made to determine changes in the minor constituents of the oil (*e.g.*, phospholipids, pigments, and alcohols) over the course of an electrolysis.

RESULTS AND DISCUSSION

In organic electrochemical syntheses, the effectiveness of the primary electrode reaction is often gauged by the determination of the reaction current efficiency. In the present system, this quantity is a measure of the amount of electro-generated hydrogen that combines with an oil's unsaturated triglycerides. The current efficiency is computed from the change in total moles of double bonds in the oil (as determined from the GC fatty acid profiles of initial and final-product oil samples) and the total charge passed in an electrolysis (as noted by the coulometer reading):

$$\text{Current efficiency (\%)} = (\Delta \text{ moles of double bonds})(2 \text{ equiv/mole})F/C \qquad [5]$$

where F is Faraday's constant (96,487 C/equiv.) and C is the total coulombs passed in an experiment. For the present reaction system, a current efficiency below 100% indicates the fraction current consumed by the H_2 gas evolution reaction (Eq. [3]).

The effect of oil content and applied current on the current efficiency for oil hydrogenation is shown in Figure 3. The current density abscissa axis in this Figure is equal to the applied current divided by the cross-sectional area of the Raney nickel bed exposed to the anode (20 cm²). In general, the current efficiency results are high (50–80%), demonstrating the relative ease of adding atomic hydrogen to an oil's double bonds at a moderate solution temperature. The results in Figure 3 also show that the current efficiency for hydrogen addition to the oil (at a given concentration) decreases with increasing current density, as is often the case in an electrocatalytic hydrogenation (11,12). This is due to an imbalance in the atomic hydrogen production and consumption rates; at high current densities the H_{ads} electrochemical generation rate exceeds the rate at which hydrogen combines with the double bonds of the oil reactant. Excessive amounts of unreacted H_{ads} on the catalyst surface promote the H_2 gas evolution reaction. The results in Figure 3 suggest that a current efficiency of near 100% (*i.e.*, the maximum electrical efficiency for the reaction) could be achieved at a current density of ≈5 mA/cm². Such a low current density experiment was not performed because the net production rate of hydrogenated oil (given by the product of the current efficiency and current density), which controls the size and capital cost of an electrocatalytic reactor will be lower at 5 mA/cm² than that realized experimentally at 10–20 mA/cm².

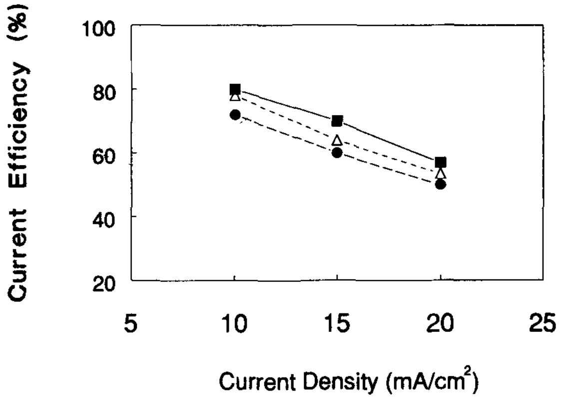

FIG. 3. The effect of current density and electrolyte oil content on the current efficiency for oil hydrogenation. Reaction temperature, 70°C; solvent composition, 50:50 vol% t-butanol/water; ■, 24 g oil; △, 36 g oil; and ●, 48 g oil.

At a given current density, the results in Figure 3 show that the reaction current efficiency decreases slightly with increasing electrolyte oil content. In organic electro-syntheses where the reactant and product(s) are completely soluble in the reaction medium, an increase in reactant concentration usually results in an increase in product current efficiency (5,12). For the two-phase electrolytic solution used in the present study this does not appear to be the case, for reasons not well understood at this time. The highest hydrogenation current efficiency (80%) was achieved with a current density of 10 mA/cm² and 24 grams of soybean oil in 100 mL of solvent/supporting electrolyte.

Table 1 lists the chemical properties of the final oil products corresponding to the data points in Figure 3. No observable trends between any of the measured properties and the electrolyte oil content or applied current were identified. The degree of fatty acid saturation from these experiments (as quantified by the product IV value) was comparable to a traditional chemical catalytic brush hydrogenation (15). The 18:0 selectivity in the Table refers to the relative increase in the stearic acid content of the product oil per unit change in IV value. The 18:3 selectivity was calculated from the relative decrease in linolenic acid per unit change in IV value. For the brush hydrogenation of soybean oil by means of a high-temperature chemical catalytic technique, the 18:0 and 18:3 selectivities were computed from fatty acid profile data in the literature (product oil IV value of 110) and were found to be 0.50 and 2.7, respectively (15). The electrocatalytic process produces an oil with more saturated fat and approximately the same triene content as that synthesized by a traditional nonelectrochemical method. The lowest 18:0 selectivities were obtained for 10 mA/cm², suggesting that less saturated oil is produced when the rate of formation and surface concentration of H_{ads} are low.

The total *trans* isomer content of the electrochemically

TABLE 1

Chemical Properties of Electro-Hydrogenated Soybean Oil for Variation in Electrolyte Oil Content and Current Density[a]

Exp. conditions (wt. oil; current density)	Fatty acid profile (wt. %)						IV Value	18:0 Selectivity (%)	18:3 Selectivity (%)	% Free fatty acid[b]	% Total *trans*
	C16	18:0	18:1	18:2	18:3	C20					
Reference oil	10.5	4.1	24.2	53.4	7.3	0.2	132	—	—	0.06	2.5
24 g; 10 mA/cm^2	10.1	5.6	46.5	34.8	2.2	0.4	106	1.39	2.67	0.97	9.9
24 g; 15 mA/cm^2	10.3	5.9	42.6	38.4	2.1	0.3	109	1.91	3.01	1.99	12.1
24 g; 20 mA/cm^2	10.2	6.4	37.2	41.3	4.0	0.4	114	3.13	2.45	2.17	8.3
36 g; 10 mA/cm^2	10.2	5.5	46.2	35.2	2.1	0.4	106	1.39	2.74	1.29	11.7
36 g; 15 mA/cm^2	10.2	7.1	38.3	40.4	3.1	0.4	111	3.48	2.68	3.75	8.3
36 g; 20 mA/cm^2	10.1	5.2	39.8	40.6	3.5	0.4	114	1.46	2.78	0.70	7.7
48 g; 10 mA/cm^2	10.1	5.1	44.6	37.2	2.1	0.4	108	1.07	2.96	0.75	9.9
48 g; 15 mA/cm^2	10.3	6.5	39.7	39.3	3.5	0.4	111	2.84	2.48	3.96	8.8
48 g; 20 mA/cm^2	10.3	5.7	36.6	42.8	3.8	0.4	116	2.42	2.85	1.44	7.9

[a] Reaction temperature, 70°C; solvent composition, 50:50 (v/v) % t-butanol/water; TEATS concentration, 1.0 M.

[b] % Free fatty acid given as % oleic.

TABLE 2

Current Efficiencies and Chemical Properties of Electro-Hydrogenated Soybean Oil for Variations in the Electrolyte TEATS Concentration[a]

Exp. conditions (TEATS concentration)	Fatty acid profile (wt. %)						IV Value	Current efficiency (%)	18:0 Selectivity (%)	18:3 Selectivity (%)	% Free fatty acid[b]	% Total *trans*
	C16	18:0	18:1	18:2	18:3	C20						
Reference oil	10.5	4.1	24.2	53.4	7.3	0.2	132	—	—	—	0.06	2.5
0.5 M	10.0	5.6	38.1	42.0	3.3	0.4	114	54	2.11	3.03	2.80	6.7
1.0 M	10.2	7.1	38.3	40.4	3.1	0.4	111	64	3.48	2.68	3.75	8.3
1.5 M	10.1	7.7	36.5	41.0	3.9	0.4	113	59	4.50	2.37	13.71	6.2
2.0 M	10.2	6.0	35.4	43.8	3.7	0.4	116	48	2.99	3.01	2.49	5.3

[a] Reaction temperature, 70°C; solvent composition, 50:50 (v/v) % t-butanol/water; initial oil content of electrolyte, 36 g; current density, 15 mA/cm^2.

[b] % Free fatty acid given as % oleic.

saturated oil was found to be significantly lower than the 20% reported in the literature (15) for a high-temperature chemical catalytic brush hydrogenation process. We believe this is due to the lower operating temperature of the electrocatalytic method. In light of a recent health effects study, which showed that *trans* isomers could raise blood levels of harmful, low-density lipoprotein cholesterol and lower protective high-density lipoprotein levels (16), a major focus of future experiments will be directed toward lowering the reaction temperature below 70°C and increasing the extent of hydrogenation to synthesize a low-IV margarine-type product with a low percentage of *trans* isomers.

The percentage of free fatty acids in the hydrogenated oil product was always greater than that in the starting oil, presumably due to triglyceride hydrolysis in the alkaline electrolyte region of the cathode bed (Eq. [1]). Higher solution flow rates through the electrocatalytic reactor and/or the use of a buffered aqueous phase should minimize this problem (such experiments were not carried out in the present study).

The effects of t-butanol/water solvent composition and TEATS concentration on the hydrogenation current efficiency and chemical properties of the oil product are presented in Tables 2 and 3, respectively. In these experiments the initial oil content of the two-phase reaction medium was 36 grams, and the applied current was 300 mA (the apparent current density was 15 mA/cm^2). The current efficiency results indicate a modest improvement in H_{ads} addition to the oil (current efficiencies of 64–70%) when the electrolyte contains 1.0 M TEATS and between 0 and 50 volume percent t-butanol. As was the case for the chemical property data in Table 1, no discernable trends exist between electrolyte composition, percent total *trans* isomer content, and stearic and linolenic fatty acids selectivities. In general, the electrocatalytically hydrogenated triglyceride is characterized by a high stearic content and a low percentage of fatty acids in the *trans* isomer form. The free fatty acid content of the oil product in these experiments is also similar to that in Table 1. The high percentage of free fatty acids for the 1.5 M TEATS and 75:25 t-butanol/water experiments is attributed to electrolyte channeling through the Raney nickel powder cathode bed, which created dead (stagnant) volumes of high-pH electrolyte in the cathode compartment.

Because water was always present in the two-phase electrolyte, nickel catalyst corrosion could have occurred, in which case dissolved nickel would be present in the hydrogenated oil product. To determine whether nickel corrosion is a potential problem, samples of hydrogenated

TABLE 3

Current Efficiency and Chemical Properties of Electro-Hydrogenated Soybean Oil for Variations in Solvent Composition[a]

Exp. conditions (vol. % t-butanol/water)	Fatty acid profile (wt. %)						IV Value	Current efficiency (%)	18:0 Selectivity (%)	18:3 Selectivity (%)	% Free fatty acid[b]	% Total *trans*
	C16	18:0	18:1	18:2	18:3	C20						
Reference oil	10.5	4.1	24.2	53.4	7.3	0.2	132	—	—	—	0.06	2.5
0:100	10.4	7.5	39.6	38.7	3.1	0.3	109	70	3.71	2.48	2.91	7.7
25:75	10.2	6.5	39.7	39.3	3.5	0.4	111	65	2.84	2.48	2.46	7.9
50:50	10.2	7.1	38.3	40.4	3.1	0.4	111	64	3.48	2.68	3.75	8.3
75:25	10.1	6.9	35.4	42.9	3.9	0.4	115	52	4.00	2.64	8.26	8.0

[a] Reaction temperature, 70°C; current density, 15 mA/cm^2; initial oil content of electrolyte, 36 g; TEATS concentration, 1.0 M.

[b] % Free fatty acid given as % oleic.

oil product from each electrolysis were analyzed for dissolved nickel by atomic absorption spectrophotometry. In general, the nickel concentration in the oil was low, with nickel levels of 0–0.5 ppm in 10 hydrogenated oils, 1.4–4.6 ppm in 4 samples, and 15.1 ppm nickel in 1 oil product (the experiment in Table 3 with no t-butanol cosolvent). There were no identifiable correlations between the reactor operating conditions, electrolyte formulation, and dissolved nickel concentration.

ACKNOWLEDGMENTS

This work was sponsored by the U.S. Department of Energy, Advanced Industrial Concepts Division, through a National Renewable Energy Laboratory subcontract (XD-0-10103-1). The authors also thank Capital City Products Company for providing the soybean oil used in this study.

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[Received September 19, 1991; accepted February 27, 1992]