REVIEW

Reactors for Hydrogenation of Edible Oils

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Due to the characteristics of the hydrogenation of edible oil, by far the most common type of reactor has been the batch-type slurry hardener. Although continuous reactors offer several advantages compared to batch reactors, they are seldom used in the industry. This review paper describes the most commonly used full-scale reactors, both batch and continuous. Several different laboratory reactors are also described. The experimental results obtained from those reactors indicate that it is possible to achieve selectivites and reaction rates in a continuous reactor as high as in a slurry batch reactor.

KEY WORDS: Continuous, hydrogenation, industrial reactors, laboratory reactors, vegetable oils.

The hydrogenation of edible oils is quite complex. It is a three-phase process with hydrogen in the gas phase, a liquid oil and a solid catalyst, usually nickel. To achieve high reaction rates, not only an active catalyst but also good mass transfer conditions between the gas and the liquid and between the liquid and the catalyst are needed. Another problem with liquid-phase oil hydrogenation is that the oil to be hydrogenated varies in composition, depending upon source, quality and previous treatment. It is advantageous if the reactor can alternate easily between different products. It is difficult to find a reactor that can fulfill all of these expectations.

The hydrogenation reactor (often referred to as a hardener or converter) is usually a slurry reactor that is run batchwise. Most of the other steps in the manufacturing process are continuous, but there are still many reasons to use a batch reactor. It is easier to control the reaction in a batch process than in a continuous process, and this makes it possible to reach high selectivities and to adjust the process to different demands on the product and to a varying feedstock. The technology associated with hydrogenation in batch reactors is well proven. There are also several advantages with a continuous reactor, apart from the fact that it will be better integrated with the other parts of oil processing. It is possible to use a fixed-bed catalyst, which eliminates the need to filtrate the catalyst from the product. Higher reaction rates per reactor volume may be achieved, and less energy is needed. The possibility of using continuous processes has also increased with a more uniform quality of the starting oils, an improved ability to remove poisons from the feed, better information available for design of flow reactors and the better catalysts that are available. Continuous systems are in use, especially for production of general-purpose shortenings, where the products are sold in large volume.

General papers about commercial reactors for edible oil hydrogenation have been written by Albright (1,2), Allen (3) and Hastert (4).

INDUSTRIAL REACTORS

Batch reactors. The most important things to consider when designing a batch reactor are good contact between the oil and the gas, the energy required for the agitator, the compressor for the hydrogen, the pump for recirculating the oil, and good temperature control. The rate-determining step is often transfer of hydrogen from the gas phase to the liquid phase. As a result, the reactor has to be able to sustain a high level of mass transport. The internal mass transfer of triglycerides in the pores of the catalyst is probably important for the selectivity and the degree of isomerization of the product. Albright (2) states that the rate of isomerization increases compared to the rate of hydrogenation with higher temperatures, lower hydrogen pressures, decreased agitation, and generally with an increased catalyst load.

The most common batch reactors for edible oil hydrogenation are described by Allen (3). The first hydrogenation processes were either of the Wibuschewitsch type, where oil is sprayed into an atmosphere of hydrogen, or of the Normann type, where hydrogen is sparged into the oil. Both systems are still in use. The Normann system is favored in the U.S., while the Wibuschewitsch system is used more frequently elsewhere. The latter system supposedly gives better transfer of hydrogen to the oil.

There are two major kinds of batch reactors: recirculation reactors, where large volumes of hydrogen are recycled, and the newer dead-end reactors, where only as much hydrogen is supplied to the system as needed (possibly with internal recirculation).

The recirculating reactor is a tall, cylindrical vessel that usually holds 10,000–30,000 kg of oil (Fig. 1). It contains heating coils and a distribution device in the bottom of the reactor for dispersing the injected hydrogen. Hydrogen is continuously withdrawn from the headspace, purified and then sparged back into the reactor. The system needs a blower or compressor to circulate the hydrogen. It is necessary to purge 3–4% of the hydrogen to avoid accumulation of nitrogen and other impurities. The hydrogen is usually kept in large low-pressure tanks, and the pressure in the reactor is usually slightly above atmospheric pressure.

The dead-end converters hold about 5,000–30,000 kg of oil (Fig. 2). These reactors are somewhat shorter than the recirculating reactors because they need less headspace. They are designed for pressures between full vacuum and 7–10 bar. The agitators are often of the turbine type with two or more impellers, where the top impeller sucks hydrogen from the headspace into the liquid. There are also impellers with flat paddles set vertically around a horizontal disc. When a batch is started, the converter is kept under vacuum to deaerate and dry the oil during the initial heat-up period, before the hydrogen is admitted. Hydrogen is let into the system when the charge has reached the desired operating temperature. The charge is partly cooled in the reactor at the end of the hydrogenation.

One of the advantages with the dead-end reactor is the deaeration and dehydration of the charge at the beginning of the hydrogenation. It is also possible to control the process better than with a recirculating reactor

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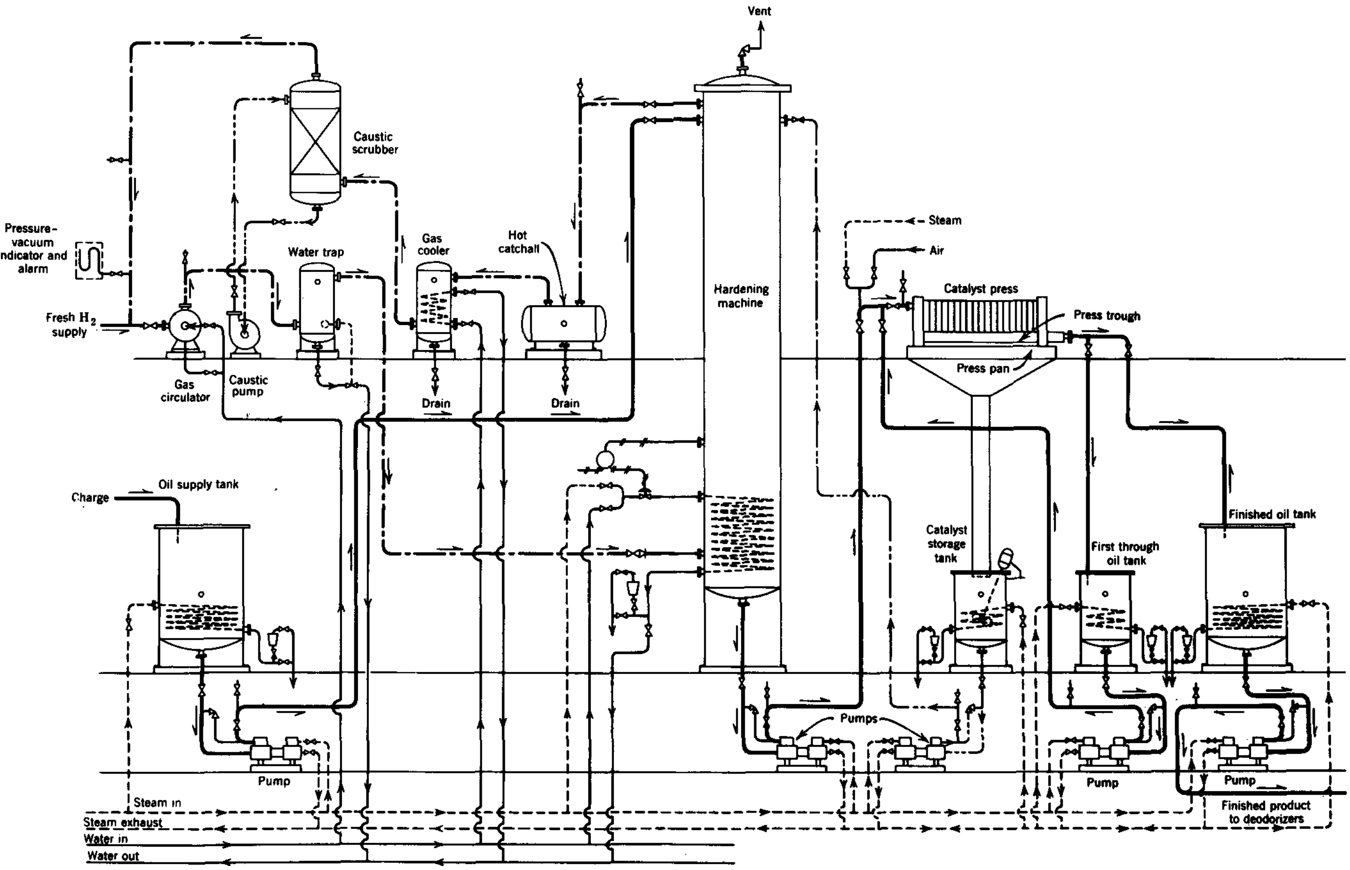

FIG. 1. Hydrogenation plant employing hydrogen recirculation (courtesy of Foster Wheeler Corp., Clinton, NJ).

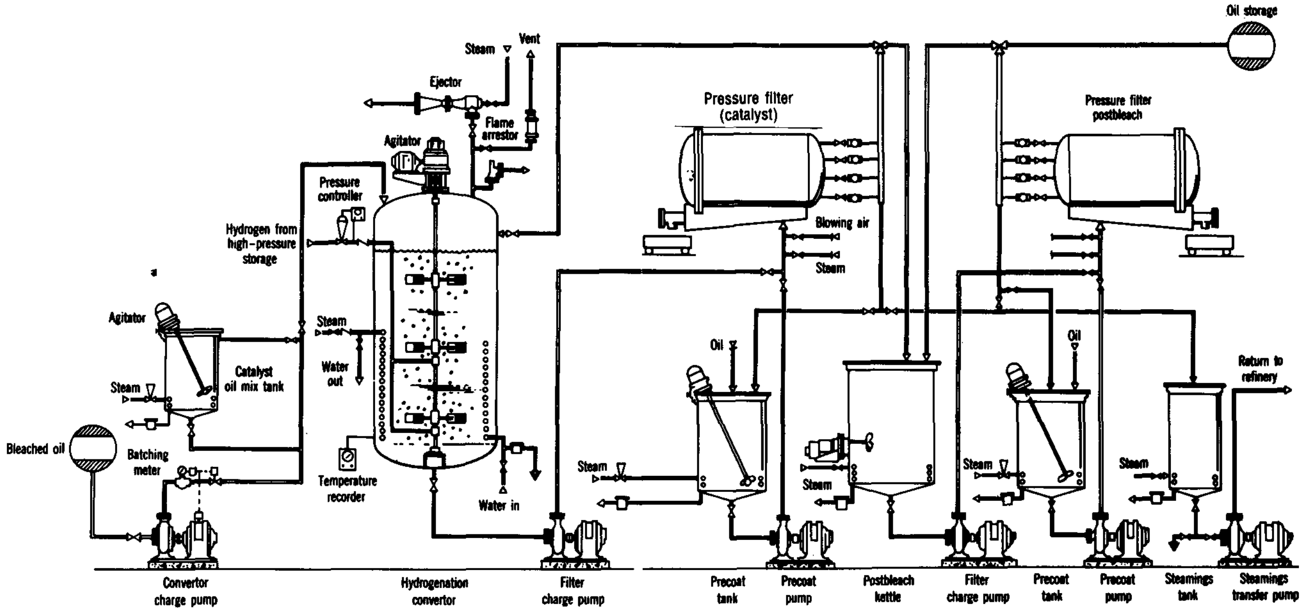

FIG. 2. Dead-end reactor (courtesy of Cherry-Burrell Process Equipment, a United Dominion Company).

because the reactions take place at a definite and constant temperature, and the iodine value (IV)-drop can be calculated from the pressure drop in the hydrogen supply tank. The selectivity can be better controlled because both temperature and pressure can be set. Finally, the equipment is simpler, less expensive and more easily maintained.

Continuous reactors. The difficulties with continuous reactors have been described by Albright (2). It is possible to find continuous flow reactors with low contents of isomerized products, but seldom with high selectivity ratios. The most common kinds of continuous reactors so far have been the tubular reactors (either straight or in U-bends) or flow reactors with well-stirred compartments. A cross-flow reactor has also been pointed out as useful for oil hydrogenation. Difficulties with flow reactors include: (i) maintaining uniform contact between the gas and the liquid; (ii) preventing back-mixing of the liquid phase; (iii) different residence times for gas and liquid, making the scale-up difficult; and (iv) obtaining residence times as long as those in a batch reactor. Short residence times are sometimes compensated for by the much higher reaction rates that may be achieved. Albright (1), Hastert (4) and Allen (3) describe several kinds of continuous reactors aimed at commercial use. Many of the processes are patented, and there is little to be found in the literature about their practical performance.

The Lurgi process consists of a vertical column with several reactor chambers where oil, hydrogen and catalyst flow concurrently from the bottom (Fig. 3). Between the chambers are trays with apertures that create a hydrogen flow through the oil and catalyst to improve the mixing. The selectivity obtained in the hydrogenation is not very high, and fairly long residence times would make it more difficult to change between products.

The Pintsch-Bamag reactor is also a column reactor with trays. The oil and catalyst enter the reactor at the top, while the hydrogen is fed at the bottom and bubbled upward through the sieve-type trays. Baffles on each tray prolong the residence times and prevent backmixing.

In the King-Dravo process, heated oil and catalyst are pumped through a pipeline reactor where hydrogen is introduced at spaced intervals (Fig. 4). The gas inlet provide the necessary agitation, together with the stirring caused by the flow velocity of the fluids in the reactor. The amount of hydrogen introduced to the reactor controls the degree of saturation of the product.

In the Procter and Gamble process, the reactor is divided into compartments with stationary baffles and an agitator on a common shaft. The compartments are separated by horizontal baffles with small clearances between these and the reactor walls, thereby eliminating backmixing between the compartments. The reactor is operated at 2–6 bar, and the reaction rate is quite high, up to 30 IV/min. The fast reaction resulting from the high agitation and the isothermal conditions is reported to produce less *trans* isomers than in conventional batch equipment, without any major loss in selectivity. Instead of mass transport of hydrogen as the rate-limiting step, the controlling step may be the transfer of triglycerides to the catalyst in this reactor. A similar reactor design has been reported by Nelles *et al.* (5).

The Buss loop reactor is a multistage horizontal autoclave reactor, with external recirculation of the oil and an external heat exchanger (Fig. 5). The oil/catalyst slurry is introduced through a Venturi nozzle at high velocity. According to Leuteritz (6) and Duveen and Leuteritz (7), this reactor has a high mixing intensity, as well as simple heat removal by an external heat exchanger. The reactor holds 2.5–20 tons of oil. Several Buss loop reactors have been installed, but most of them are operated batchwise.

PILOT PLANT AND LABORATORY REACTORS

Moulton and Kwolek (8) have used a trickle-bed reactor for hydrogenation of soybean oil with a copper catalyst (Fig. 6). They activated the catalyst *in situ* in two different ways. Dry pretreatment started with purging with N_2, raising the bed temperature to 170°C and then introducing H_2. The hydrogen concentration was gradually

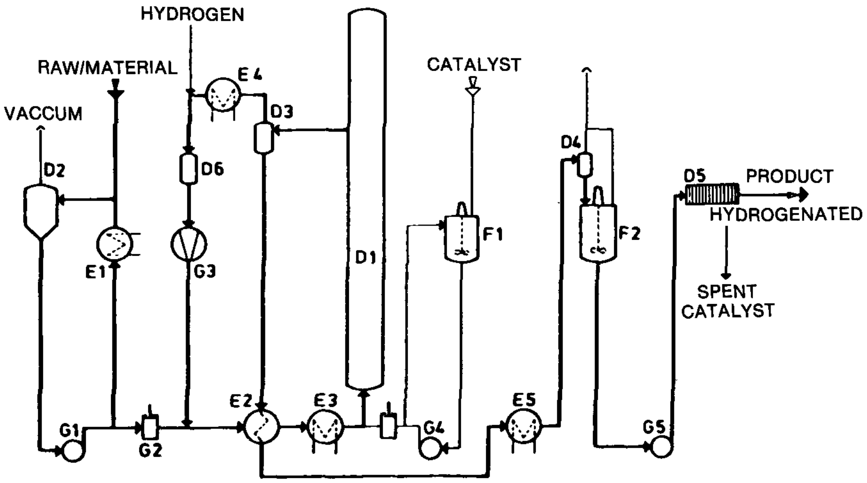

FIG. 3. Lurgi process (reprinted with permission from Ref. 4): D1: hydrogenator, D2: vacuum drier, D3: separating vessel, D4: flashing vessel, D5: catalyst filter, D6: intermediate vessel, E1–E5: heat exchangers, F1: catalyst slurry tank, F2: intermediate vessel, G1–G5: pumps and compressors.

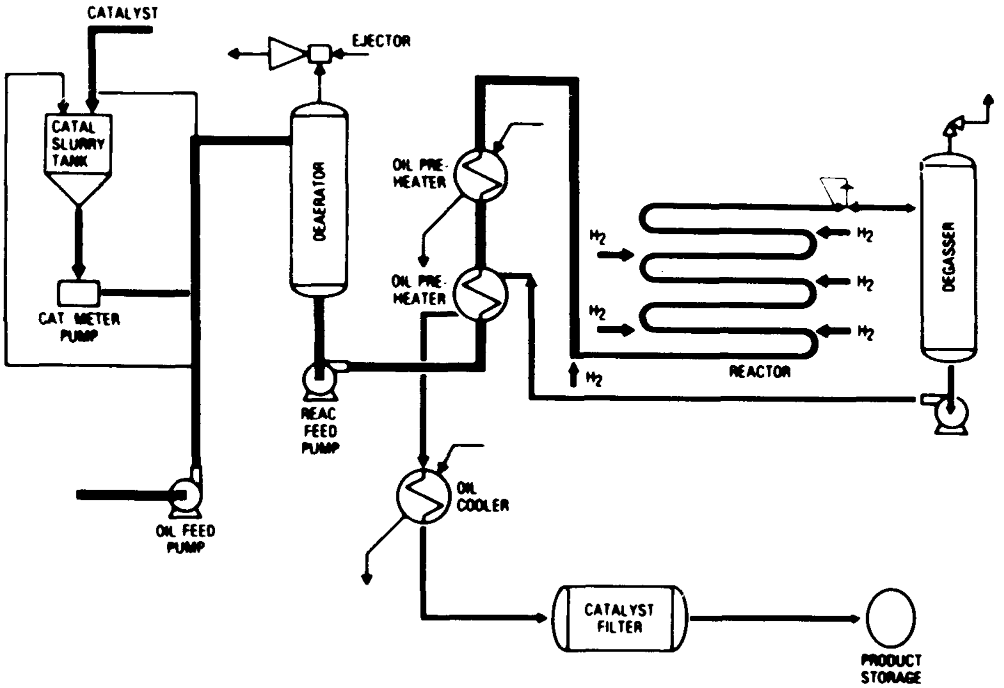

FIG. 4. King-Dravo process (reprinted with permission from Ref. 4). Abbreviations: CATAL and CAT = Catalyst; REAC = reactor.

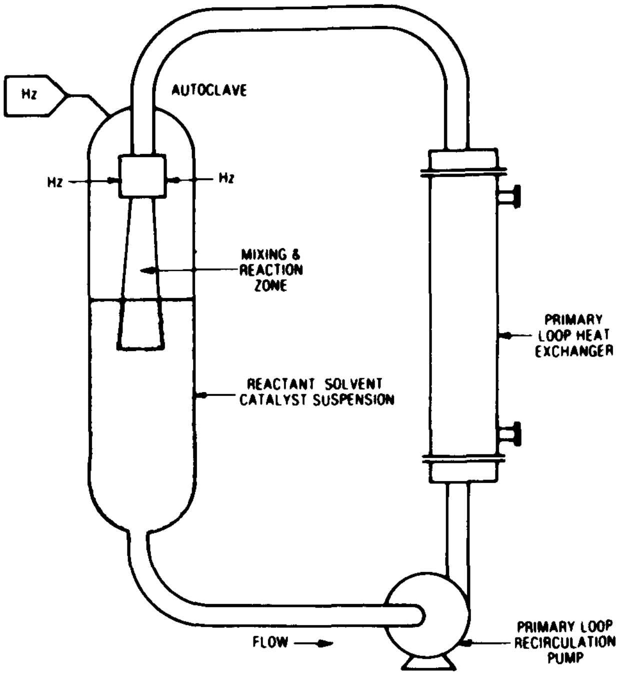

FIG. 5. Buss loop reactor (reprinted with permission from Ref. 4).

increased, while the temperature was controlled. The temperature was then lowered to 100°C, and the pressure was raised to at least 4.5 bar. Moisture was formed and collected before the oil was let into the system. Additional moisture was often formed when the oil was introduced, and the experimental runs were started when the oil product was water-free. The "wet" pretreatment also started with purging the system with N_2. The oil was then introduced to the system, and after that the amount of hydrogen in the inlet was slowly increased in steps of 10% each. The operating conditions were 110–153°C and 3.1–6.2 bar. The authors compared their results with batch hydrogenations. The selectivites found in these comparable experiments were similar, but the temperature needed to be lower for the continuous reactor. One problem with trickle-bed reactors is that they often have poor radial heat distribution, especially in production-scale units (9). Channeling may also occur, as well as incomplete wetting of the bed.

Heldal *et al.* (10) have hydrogenated soybean oil in trickle-bed reactors with palladium-on-polystyrene, palladium-on-carbon and extruded nickel catalysts. At the selected operating conditions (hydrogen pressure 4.5 bar and temperature 80–170°C for the palladium catalysts, 2.4–4.5 bar and 140–260°C for the nickel catalysts), they found that both the specific isomerization (S_i, %*trans*/ΔIV) and the linolenate and linoleate selectivities were lower than for hydrogenations performed in comparable batch reactors. They claimed that these low selectivities for the trickle-bed reactor may be caused by the large interfacial area that facilitates the mass transport of the hydrogen. They also stated that the pores of the catalysts used for the experiments might have been too small, thereby hindering the transport of the triglycerides inside the pores.

The continuous hydrogenation of soybean oil has been studied by Koritala *et al.* (11,12) and Snyder *et al.* (13).

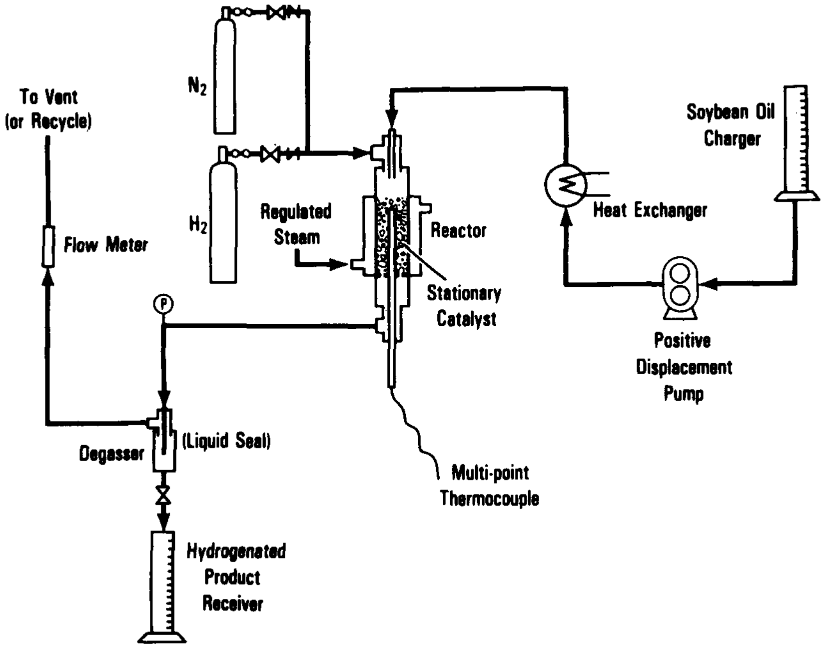

FIG. 6. Trickle-bed reactor (reprinted with permission from Ref. 8).

They used a continuous slurry reactor (coiled tube) for high-pressure hydrogenation with copper-chromite catalysts and for hydrogenation with nickel and sulfur-promoted nickel catalysts (Fig. 7). For pure nickel, the linolenate and linoleate selectivity ratios (LnSR and LoSR) were approximately 1.5 and 5–15, respectively, while the specific isomerization was 0.4–0.7. For the sulfur-promoted nickel, the LnSR and LoSR were higher (3 and 10–17, respectively), and the specific isomerization was also higher (approximately 1.5). Koritala *et al.* (11) used a high pressure of hydrogen (80 and 320 bar) for the copper-chromite catalyst to overcome the low activity of the catalyst and to eliminate the formation of conjugated dienes. The LnSR was high, 7–9 in all their experiments, while specific isomerization was 0.82–0.94. They concluded that, by changing the operating conditions, it was possible to prepare salad oil as well as shortening and cooking oils. Snyder *et al.* (13) found that a high hydrogen pressure reduced the amount of conjugated dienes in the product, while the linolenate and *trans*-isomer selectivities were unaffected.

Rusnac *et al.* (14) have used a slurry column equipped with static mixers. The reactor could be operated under different conditions with respect to phase velocities and

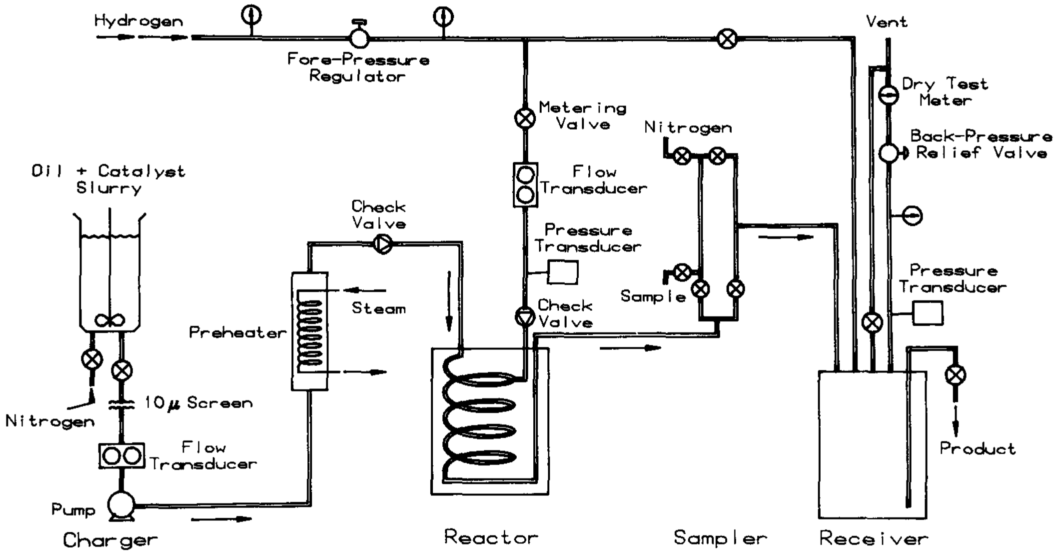

FIG. 7. Continuous slurry reactor (reprinted with permission from Ref. 11).

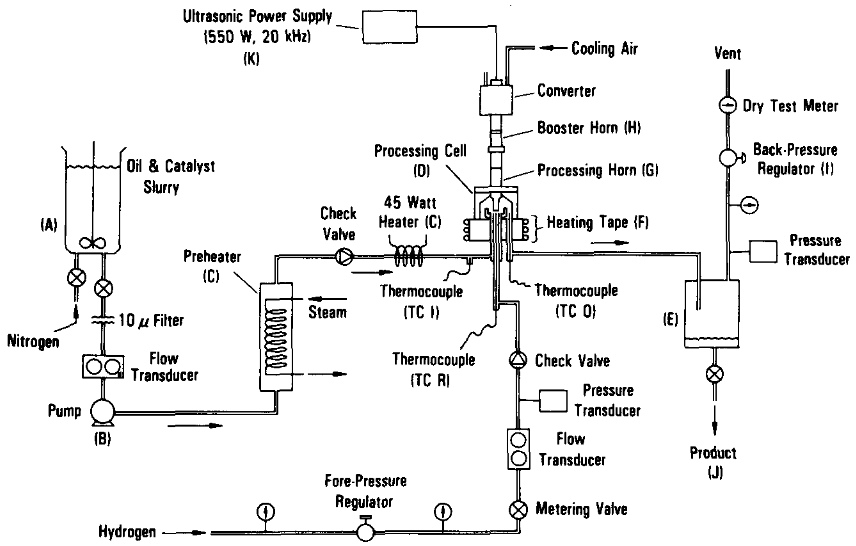

FIG. 8. Ultrasonic reactor (reprinted with permission from Ref. 16).

temperature, without major effect upon the gas-liquid interfacial area. The static mixers decrease the mean bubble diameter and increase the gas holdup, leading to a high gas-liquid mass transfer. They presumed that the liquid-solid mass transfer was limiting the reaction rate.

A reactor with an ultrasonic device has been used for hydrogenation of soybean oil, using both nickel and copper-chromite catalysts (15,16). It has been found that acoustical cavitation produces extreme localized heat and pressure under the proper conditions and may enhance chemical reaction rates. They used a continuous slurry reactor, where the slurry was preheated before entering the ultrasonic cell (Fig. 8). For the copper-chromite catalyst, the LnSR was lower than with similar conditions in a batch reactor, 5-7 instead of 10, and stearate was formed (LoSR 10-28). The amount of *trans* isomers was lower than in comparable runs with batch reactors. For the nickel catalyst, the LnSR and the specific isomerization were the same as in a batch reactor, but the LoSR was lower. The increase in reaction rate was by a factor of 20 when compared with nonultrasonic reaction in a stirred autoclave. Wan *et al.* (17) have also hydrogenated soybean oil with and without ultrasonic energy. In preliminary investigations, they had found only a 30% increase in hydrogenation rate when comparing continuous flow with and without ultrasonic energy.

Mukherjee *et al.* (18) studied several different kinds of catalysts in a vertical tube reactor (Fig. 9). The studied catalysts were Raney nickel, reduced nickel, reduced palladium, copper chromite and granulated alloys (Raney type) of Ni-Al, Cu-Al, Pd-Al and Cu-Cr-Al. The copper and copper chromite catalysts were (as usual) selective for linolenate hydrogenation. Palladium on a carrier, Raney nickel, Raney copper and Raney palladium were all very active. The former catalysts could be used for production of salad oils, and the latter for production of solid or semisolid fats. The stationary catalysts were in general very active; the IV was reduced by 40-60 IV-units within 2-4 min of hydrogenation. Some experiments were performed with hydrogenation of soybean oil dissolved in hexane. With granular Raney nickel and Raney copper catalysts and a high pressure of hydrogen, the rate of the IV-drop was as high as 50-60 IU-units/min.

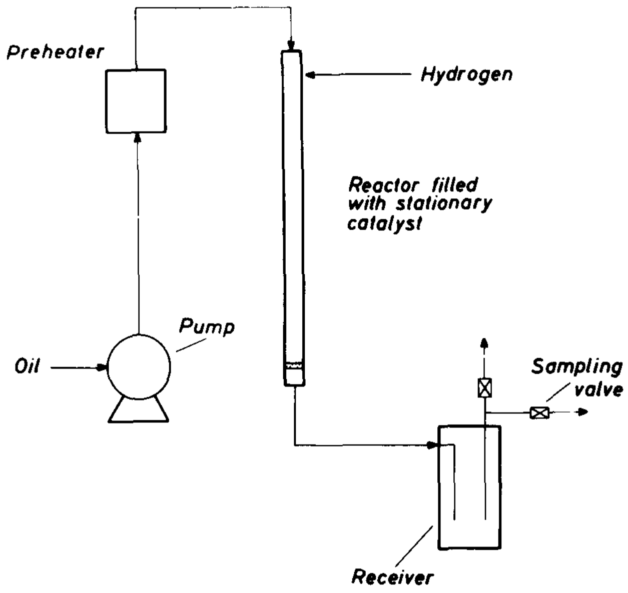

FIG. 9. Continuous vertical tube reactor with stationary catalyst (reprinted with permission from Ref. 18).

An isothermal concurrent flow reactor has been used for the hydrogenation of soybean oil with nickel and copper catalysts (19,20). To increase the dispersion of hydrogen into the oil, the reactor included a small premixer with static mixers. The operating conditions for the nickel catalyst were 150-190°C and atmospheric pressure; while for the copper catalyst, the temperature was somewhat higher, 175-215°C. With both catalysts, the

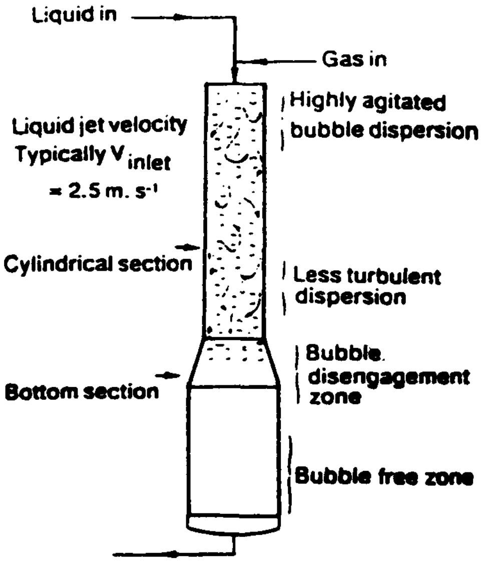

FIG. 10. Cocurrent down-flow contactor reactor (reprinted from Ref. 21, with permission from Pergamon Press, Ltd., Oxford, United Kingdom).

reaction rates were much higher than in batch hydrogenations. For the copper catalyst, the ΔIV/min was 3.2–4.8 for the continuous reactor, compared to 1.5–2.3 for the batch reactor. The selectivities were higher, and the amount of conjugated dienes was lower than for the batch reactor. The reaction rate for the nickel catalyst was 6–7 times higher in the continuous reactor than in a batch reactor at the same operating conditions (temperature, pressure, catalyst loading). Neither catalyst showed any change in LnSR with changes in operating conditions.

Boyes *et al.* (21) have studied mass transport limitations and reaction kinetics in a cocurrent downflow contactor. The gas and liquid were mixed under turbulent conditions before contact with the fixed-bed catalyst, or mixed together with a catalyst for slurrylike conditions (Fig. 10). They found that the surface reaction rate was controlling the overall reaction rate because the mass transport was greatly enhanced in the reactor. The operating conditions were 1–4 bar, 130–210°C, palladium on stainless steel (fixed bed); palladium/charcoal or Harshaw NiDM3 (slurry).

Choi *et al.* (9) have hydrogenated soybean oil with a nickel/silica catalyst in a rotating packed-disc reactor (RPDR). This reactor facilitates hydrogen transfer into the liquid phase. It consists of three rotating discs, partly submerged in the liquid, with hydrogen in the headspace (Fig. 11). It was not very selective compared to a slurry reactor, perhaps due to the larger catalyst particles used. LoSR was higher at low pressure and at high rotating speeds. In slurry reactors, the selectivity usually decreases at higher rotating speeds (due to better mass transport of hydrogen), but, with the RPDR, the increased exposure time in the gas phase at low rotating speeds leads to a reaction that goes too far, e.g., it raises the production of saturates. Both the reaction rate and the selectivity increased with higher temperature.

Grau *et al.* (22) have constructed a slurry reactor with the aim of eliminating the induction times often encountered when the catalyst needs to be activated before use. The catalyst in the cup-and-cap reactor (CAC) is stored in a small container inside the reaction vessel, where it is activated and then kept at operating temperature and pressure until it is released into the oil (Fig. 12). This reactor is simple in operation, and the amount of catalyst added to the reactor is well controlled. Experiments showed that virtually no induction period was encountered when using the CAC reactor.

The mass transfer on the liquid side of a hydrogen-oil system has been determined in a series of papers by Ganguli and van den Berg (23–26). They have hydrogenated soybean oil with a homogeneous Ziegler-Natta catalyst (nickel diisopropyl salicylate and aluminum triisobutyl) in both a falling-film reactor and an agitated reactor. Their experiments lead to estimations of the liquid-side mass transfer coefficient in both reactors and of the interfacial area in the agitated tank.

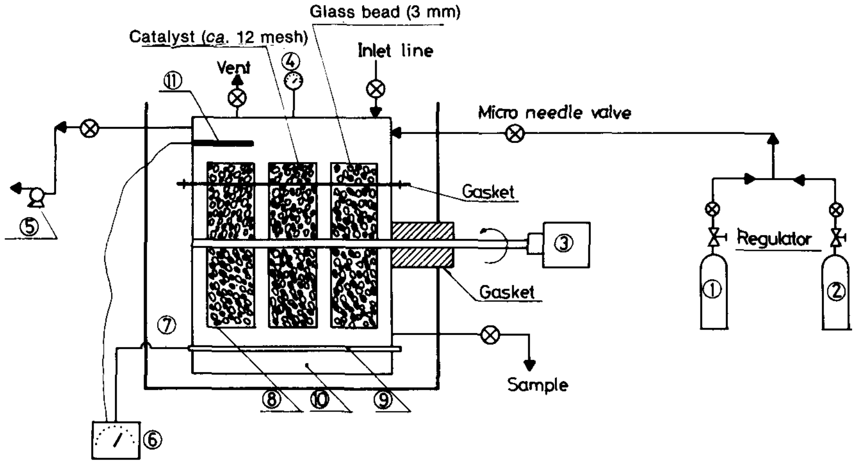

FIG. 11. Rotating packed disc reactor (reprinted with permission from Ref. 9): 1: N_2 cylinder, 2: H_2 cylinder, 3: variable speed motor, 4: pressure gauge, 5: vacuum pump, 6: temperature controller, 7: water bath, 8: rotating disk, 9: immersion heater (250 watt), 10: soybean oil, 11: thermowell.

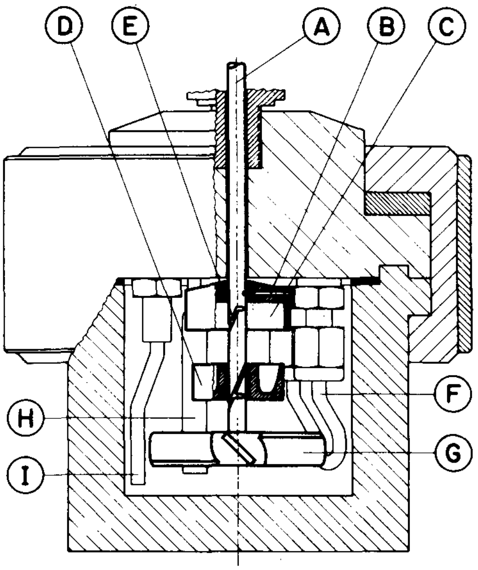

FIG. 12. Cup-and-cap reactor (reprinted with permission from Ref. 22, copyright 1987, American Chemical Society): A: shaft, B: screw, C: cap, D: cup, E: grooved sleeve, F: cooling element, G: agitator, H: thermoresistance wall, I: hydrogen inlet sampling tube.

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