TABLE II

It is envisaged that β-acetate (**X a, b**) is formed quantitatively and preferentially in respect of α-acetate on steric grounds by the acetic acid addition. This β-acetate in presence of LTA undergoes decarboxylation (11) and the radical formed is converted into carbonium ion. Now the acetate ion interacts with the carbonium ion to form 1,2-diacetoxypenta- and heptadecane (**IX a, b**). The formation of **XI a, b** is supposed to be by addition of $\dot{C}H_3$ at the β-position and as the secondary radical is resonance stabilized by the adjacent carboxylic group the attack of carboxymethyl radical takes place which on subsequent dehydration results in **XI a, b**.

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✻Optimization of Industrial Vegetable Oil Hydrogenators

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ABSTRACT

A method to optimize the operation of industrial vegetable oil hydrogenators is proposed. The hydrogenation data from an operating plant are used to fit a simple mathematical model which is then used to select values for temperature and hydrogen pressure such that a desired product is obtained in minimum hydrogenation time. Methodology is suggested whereby any nonoptimal operation can be changed to optimal operation in a few trials.

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Nomenclature: E = activation energy, kcal/kg mol; IV = iodine value; k_L - reaction rate constant for linoleate hydrogenation, sec^{-1} $atm^{-0.5}$ (kg cat/100 kg oil)$^{-1}$; k_O = reaction rate constant for oleate hydrogenation, sec^{-1} atm^{-1} (kg cat/100 kg oil)$^{-1}$; k^o = preexponential factor; k_B = hydrogen mass transfer coefficient, kg mol sec^{-1} atm^{-1} (kg oil)$^{-1}$; [L] = linoleate concentration, kg/100 kg oil; m = catalyst concentration, kg/100 kg oil; N = hydrogen mass transfer rate, sec^{-1} (kg mol/kg oil); [O] = oleate concentration, kg/100 kg oil; pH_2 = hydrogen pressure in liquid phase, atm; pH_2^* = hydrogen pressure in gas phase, atm; R = reaction rate, sec^{-1} (kg/100 kg oil); R_g = gas constant, kcal kg mol^{-1} K^{-1}; [S] = stearate concentration, kg/100 kg oil; s = linoleate to oleate ratio; T = absolute temperature, K; x_i = optimization variables. Greek symbols: ϵ = tolerance; θ = hydrogenation time, min. Subscripts: d = desired value; L = linoleate, O = oleate; S = Stearate; t = trial value; o = initial value.

INTRODUCTION

Modeling and computer simulation of vegetable oil hydrogenation has attracted a lot of attention over the years. Presently, models with various degrees of sophistication are available. Design of a full-scale unit on the basis of laboratory data alone can seldom be considered optimal, due to the inherent empiricism in the hydrogenation models. In the present work, a methodology is suggested whereby the data from an operating unit can be used to optimize its performance in a few steps.

OPTIMIZATION METHODOLOGY

A simplified reaction scheme was earlier shown to fit the hydrogenation data from industrial hydrogenators (1,2). A simple procedure to estimate the parameters in the model was presented (2). The simplified nature of the model and the subsequent estimation of the model parameters from plant data necessitate that the search for optimum operating variables be restricted to the immediate neighborhood of the prevailing operational practice. The possibility of the actual optimum lying far outside such a restricted domain, however, cannot be ruled out. The search for this optimum can be carried out in steps by a procedure logically similar to the evolutionary operation (3) as follows.

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Present plant data are used to estimate the parameters for the methematical model. Using this simulation model and a suitable numerical search technique, optimal operating conditions are obtained in the vicinity of the prevailing conditions. The restricted optimum policy is implemented and new hydrogenation data collected. These data are used to reestimate the model parameters. Another optimization is carried out restricting the search to the neighborhood of the last implemented operating policy. A few cycles of simulation, restricted optimization and implementation are expected to lead to the ultimate optimum. If properly carried out, this technique will overcome the dangers of trespassing the unexplored domain of operation at any time. Besides a simple mathematical model and parameter estimation procedure, a fast converging search technique for obtaining a restricted optimal policy is necessary for successful execution of the scheme. These are discussed below.

THE MATHEMATICAL MODEL

A simulation model of industrial vegetable oil hydrogenation was presented earlier (1,2) based on a simplified reaction scheme. The governing equations are as follows:

$$-R_L = k_L^0 \exp(-E_L/R_g T) \, [L] \, pH_2^{.5} \, m \qquad [1]$$
$$R_O = -R_L - k_O^0 \exp(-E_O/R_g T) \, [O] \, pH_2 \, m \qquad [2]$$
$$PH_2 = PH_2^0 - N/k_B \qquad [3]$$
$$N = -(R_O + 2R_L)/28200 \qquad [4]$$

where R = rate of formation, k^0 = preexponential factor, E = activation energy, R_g = gas constant, T = reaction temperature, P_{H_2} = hydrogen pressure in the oil phase, pH_2 = hydrogen pressure in the gas phase, [L] = linoleic concentration in the slurry, [O] = oleic concentration in the slurry, m = catalyst concentration, k_B = gas to liquid mass transfer coefficient, and N = hydrogen mass transfer rate.

The subscripts L and O stand for linoleic and oleic acids, respectively. Linolenic acid is usually present (if at all) only in small quantities compared to linoleic and oleic acids and reacts out in first few minutes of hydrogenation. The linolenic acid present in initial samples is added to linoleic acid using the relation, [L] = 1.5104 [Ln] based on iodine values ([Ln] = linolenic acid concentration).

The procedure for solving these equations and estimating the parameters, k_L^0, E_L, k_O^0 and E_O has been discussed elsewhere (2). Estimated parameters and operating conditions

for some industrial hydrogenators chosen for optimization are given in Table I.

OPTIMIZATION OBJECTIVE

In the present work, hydrogenation time has been chosen as the objective function to be minimized. The calculation scheme seeks to select values for operating variables such that a given batch of oil is hydrogenated to a desired product in minimum operational time. It is believed that lowering the hydrogenation time will bring down the operational cost.

OPERATING VARIABLES

In a hydrogenation unit, reaction temperature, hydrogen pressure, agitation intensity and catalyst concentration are the variables which can be manipulated to achieve a desired product quality.

Temperature

Batch hydrogenators are usually equipped with coils for exchange of heat. These are used to bring the batch of oil initially to reaction temperature by circulating steam through them. The same heat exchange facility can be used to extract heat from the reactor during reaction. It is, therefore, possible to impose externally any temperature/time profile on the reactor.

A time variant temperature profile is generally encountered in batch hydrogenation. In the present work, the temperature/time relationship is assumed to be of the form:

$$T(t) = x_1 + x_2 t + x_3 t^2 \qquad [5]$$

Such a polynomial approximation is similar to Rayleigh's method of optimization with continuous variables (4). The quadratic dependence allows the temperature to remain constant, increase or decrease monotonically or pass through a maximum or a minimum depending on relative values of x_2 and x_3. Generally the profiles in use are monotonically increasing or passing through a maximum (2). A higher order expression for temperature/time relationship was not considered worth the complexity it would involve in optimization.

Hydrogen Pressure

The usual plant practice is to maintain hydrogen pressure

TABLE I

Operating Conditions and Estimated Parameters for Different Plants

Plant	I	II	III	IV
Oil hydrogenated	Cottonseed	Soybean	Soybean	Soybean
Batch size (tons)	5.5	5.5	5.0	5.5
Temperature range (C)	145-183	165-210	150-200	155-200
Hydrogen pressure (atm)	1.7	1.7	2.3	2.4
Catalyst concentration (kg/100 kg oil)	0.048	0.048	0.048	0.045
Stirrer speed (rpm) and nos. of impellers	42 2	42 2	70 2	—
Hydrogenator type	Gas recirculation	Gas recirculation	Dead end	Gas recirculation
$k_L^0 \times 10$	0.3374	0.4029	0.4116	0.3747
E_L	2828.0	2878.0	2670.0	2600.0
$k_O^0 \times 10$	0.1726	0.0991	0.1799	0.1556
E_O	4605.0	5215.0	5539.0	5553.0

constant during hydrogenation. It was, however, felt that variation in pressure with time would offer some additional leverage to achieve a desired selectivity. The continuous variation in pressure with time was expressed as:

$$P_{H_2}^o = x_4 + x_5 t + x_6 t^2 \qquad [6]$$

This quadratic expression allows flexibility similar to that in the variation of temperature.

Hydrogen Mass Transfer Coefficient, x_7 (=k_B)

In plant practice, mass transfer coefficient can only be varied by varying agitation speed. For implementation of the computational scheme of simulation, stirrer speed will have to be correlated to the corresponding k_B value which is then used to solve Equations 1-4. Instead, k_B itself is taken as a variable in the optimization. Optimum value of k_B can then be converted to equivalent stirrer speed. Suitable correlations exist for this purpose (5-9).

Catalyst Concentration, x_8 (=m)

The hydrogenation time decreases as the catalyst concentration increases. The highest possible concentration should, therefore, be chosen. It is thus not an optimization variable in the truest sense of the term. However, if the objective is to minimize operational cost, catalyst concentration may become an important variable. It has been retained in the present model for possible use of the technique in economic optimization.

CONSTRAINTS ON THE VARIABLES

The parameters k_L^o, k_O^o, E_L and E_O were estimated from the plant data. The reasonably good fit between calculated and measured concentration profiles is no guarantee that these parameters will simulate the operation if the operating variables such as T, pH_2^o, k_B, m were to vary far beyond the prevailing plant practice. It is, therefore, imperative to restrict the probe for the optimum set of variables x_i, (i = 1,2, ...,8), to a narrow region. The actual limits on these variables is, however, a matter of discretion. The upper (max) and lower (min) limits on T, pH_2^o, k_B, m are chosen considering the existing conditions in the plant. These are transformed to the explicit upper (h_i) and lower (g_i) limits on the optimization variables (x_i) as shown in Table II. x_1 can take any value between the allowable temperature range of the reaction (g_1 = T_{min}; h_1 = T_{max}). x_2 and x_3 are defined as given in Table II. θ_{avg} is the rough estimate of hydrogenation time (200 min in the present case). Such a choice of limits on x_1, x_2 and x_3 ensures that the reaction temperature will not exceed the lower and upper bounds of temperature at any time. Similar choice of limits is made for x_4, x_5 and x_6.

TABLE II

Values of g_i, h_i and δX_i for Optimization Variables

Variable x_i	Lower limit, g_i	Upper limit, h_i	δX_i
x_1	T_{min}	T_{max}	0.5
x_2	$\theta_{avg}^{-1}(g_1 - x_1)$	$\theta_{avg}^{-1}(h_1 - x_1)$	0.5×10^{-2}
x_3	$\theta_{avg}^{-1}(g_2 - x_2)$	$\theta_{avg}^{-1}(h_2 - x_2)$	0.3×10^{-2}
x_4	pH_2^o, min	pH_2^o, max	0.1×10^{-1}
x_5	$\theta_{avg}^{-1}(g_4 - x_4)$	$\theta_{avg}^{-1}(h_4 - x_4)$	1.0
x_6	$\theta_{avg}^{-1}(g_5 - x_5)$	$\theta_{avg}^{-1}(h_5 - x_5)$	1.0
x_7	k_B, min	k_B, max	0.4×10^{-7}
x_8	m_{min}	m_{max}	1.0

Quantification of Product Quality

The desired product quality depends on the use to which the product is put. In the manufacture of vegetable shortenings, the purpose of hydrogenation is to obtain a product with as low an iodine value as possible without allowing the product melting point to exceed certain limits. Although iodine value is easily related to product composition, this is not the case with melting point. The melting point depends on the way linoleate, oleate and stearate are distributed in the triglyceride molecules. This is usually very difficult to determine. In the present work, the product quality is quantified in terms of its composition. The product must satisfy the conditions

$$[L_t] = [L_d] ; [O_t] = [O_d] ; [S_t] = [S_d] \qquad [7]$$

where [L], [O] and [S] are the concentrations of linoleic, oleic and stearic acids, subscripts t and d denote the simulated and desired values, respectively. The C-14 and C-16 saturated acids present in the oil are unchanged during hydrogenation (1) and hence are not included here. The overall mass balance, namely,

$$[L_t] + [O_t] + [S_t] = [L_d] + [O_d] + [S_d] = [L_o] + [O_o] + [S_o] \qquad [8]$$

eliminates one of the conditions in Equation 7. Subscript o denotes initial values. Therefore it is sufficient to have

$$[L_t] = [L_d] \text{ and } [O_t] = [O_d] \qquad [9]$$

For convenience, the conditions (Eqn. 9) are transformed to

$$IV_t = IV_d \qquad [10]$$
$$s_t = s_d \qquad [11]$$

where IV is the iodine value defined by Equation 12 and s is the ratio of concentrations of linoleic and oleic acids.

$$IV = 1.7321 [L] + 0.8601 [O] \qquad [12]$$
$$s = [L]/[O] \qquad [13]$$

It can be seen that conditions represented by Equations 10 and 11 are equivalent to Equation 9. These two independent constraints (Eqns. 10 and 11) introduce complexity in the optimization similar in nature to that often encountered in solution of boundary value problems.

FEASIBLE SET OF VARIABLES

The hydrogenation process can now be visualized as:
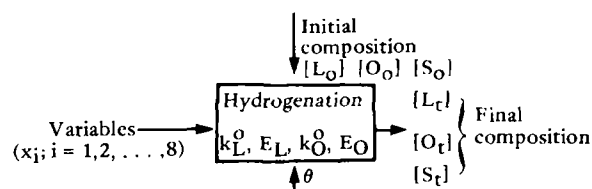
For a set of variables x_i (i = 1,2, ...,8), if hydrogenation is carried out for time θ, a product with composition [L_t], [O_t], [S_t] is obtained. For a given batch of oil x_i and θ are the input variables. In view of the constraints on [L_t] and [O_t] (Eqns. 10 and 11), any two of the variables, x_i and θ, will depend on the remaining ones. A feasible set of variables is the one which yields [L_t] and [O_t] satisfying Equations 10 and 11. Such a feasible set has to be obtained iteratively starting from a trial set because it is impossible to transform the constraints on [L_t] and [O_t] into explicit constraints on x_i and θ. The following procedure has been adopted in the present work to obtain a feasible set.

A trial set of values of optimization variables is arbitrarily

chosen. Hydrogenation is simulated for time θ such that $[L_t]$ and $[O_t]$ satisfy the constraint on iodine value, (Eqn. 10). Equation 13 is used to calculate s_t and the difference $(s_t\text{-}s_d)$ is utilized to drive the trial set to a feasible set. Fast and definite convergence on a feasible set is essential for a "reasonable effort" optimization scheme. This important aspect of the optimization is discussed below.

In order to explain the convergence procedure, the following nomenclature is used: δs = unit increase in s (arbitrarily chosen as 0.001); δx_i = average necessary change in x_i to increase s by δs keeping other variables constant; f = desired change in s_t in terms of number of units = $(s_d \text{-} s_t)/\delta s$.

The simulated output composition for a trial set of variables is used to calculate f. This change has to be achieved by suitably changing the value of one or more variables of the trial set. The possible change in any of these variables is limited because of the constraints on x_i values. It can be shown that for moving s_t closer to s_d, i.e., driving f to zero, x_i should be increased or decreased depending on whether $f/\delta x_i$ is positive or negative, respectively. Also, for $f/\delta x_i > 0$; $(h_i - x_i)/|\delta x_i|$ = maximum feasible change in s_t by altering x_i alone = u_i (say). Or, for $f/\delta x_i < 0$; $(x_i - g_i)/|\delta x_i|$ = u_i

$$[14]$$

Although any of the x_i can be varied to drive f to zero, it is desirable to use that variable which offers maximum change in s_t. If u_j = max $[u_i, i = 1,2,\ldots,8]$, then the corresponding variable x_i is varied. Two cases may arise:

Case I: $u_j > |f|$

The value of x_j in the old trial set is replaced by a new value as follows:

$$x_{j,new} = x_{j,old} + f \, \delta x_j \qquad [15]$$

The other values of x_i ($i = 1,2,\ldots,8$, $i \neq j$) remain unchanged. Using this new set of variables, the simulation program is run for a reaction time which is sufficient to achieve the desired IV (Eqn. 10). The new product composition is used to calculate a new value of f, namely f_{new}. If $f_{new}/f_{old} < 0$, then the root (value of x_j for which $|f| < \epsilon$, preassigned tolerance) is straddled. This means that the value of x_j for which $|f| < \epsilon$ lies between $x_{j,new}$ and $x_{j,old}$. A better estimate of x_j is then made using Regula-Falsi method (10). If $f_{new}/f_{old} > 0$, then value of x_j is changed to its maximum possible extent.

Case II: $u_j < |f|$

This condition implies that variation in a single variable x_j will not, in general, be sufficient to affect a change in the magnitude of f. In that case, x_j is first changed to maximum possible extent, i.e.,

$$\begin{aligned} x_{u,new} &= h_j & \text{if } f/\delta x_j > 0 \\ x_{j,new} &= g_j & \text{if } f/\delta x_j < 0 \end{aligned} \qquad [16]$$

The simulation program is run with the new set of variables and f_{new} obtained as in case I. If $f_{new}/f_{old} < 0$, it indicates that the root has been straddled and a change in another variable is not necessary. However, if $f_{new}/f_{old} > 0$, variation in another variable of the trial set is required. This variable, x_k is chosen corresponding to u_k where

$$u_k = \max [u_i, i = 1,2,\ldots,8, \qquad i \neq j] \qquad [17]$$

With this variable replaced, the simulation program is run as before to arrive at yet another f_{new}. The choice of new variables to be altered and a change from one trial set to another is continued until $f_{new}/f_{old} < 0$. Alteration in a maximum of two or three variables was found sufficient to reach this stage. The straddled root is estimated by Regula-

Falsi method as before.

The iteration procedure described above makes it possible to obtain a feasible set starting with any trial set. The simulated hydrogenation time for the feasible set is the value of the objective function.

It is essential to know δx_i ($i = 1,2,\ldots,8$) to implement the convergence scheme. An approximate estimate is adequate as these values are used only to get the starting points for Regula-Falsi method. Sensitivity analysis is carried out to obtain δx_i estimates. For this, any one variable (say x_j) is varied over its entire range keeping the rest constant. For each value of x_j, the simulation program is run until IV_d is reached. The product composition for each simulated hydrogenation is used to calculate s values. The (x_j, s) data so generated are used to obtain average δx_j. The procedure is repeated for other variables. The value of δs was chosen to be 0.001. As mentioned earlier, change in only two or three variables is necessary to obtain a feasible set from a trial set. δx_i is therefore estimated only for some dominant variables. Large values of δx_i are supplied for other variables so that these variables are not selected for changes during the implementation of convergence scheme (see Eqn. 14). The δx_i values are shown in Table II.

OPTIMUM FEASIBLE SET

The possibility of obtaining a feasible set of optimization variables and corresponding objective function paves the way for using a suitable optimization technique to move from one feasible set to another such that the objective function is minimized. The complex procedure of Box (4) was considered most suitable for this multivariate constrained optimization.

RESULTS AND DISCUSSION

Four different industrial hydrogenation units were examined and kinetic parameters were estimated from hydrogenation data. Table I gives the design and operating data along with the estimated kinetic parameters for all the four cases. Detailed investigation was undertaken for case I where effect of temperature/time profile (x_1 to x_3), pressure/time profile (x_4 to x_6), mass transfer coefficient (x_7) and catalyst concentration (x_8) on hydrogenation time was examined. To begin with, the mass transfer coefficient for hydrogen and catalyst concentration were held constant at the prevailing values and optimization was carried out to find the optimum temperature/time and pressure/time profiles. Temperature was allowed to vary in the range 375-525 K and hydrogenation pressure was varied from 0.7 to 2.7 atm. With the optimum temperature and pressure profiles (case Ia, Table III), the hydrogenation time was reduced from the present 165 min to 138 min. Optimization was then carried out allowing variable temperature but uniform pressure and also uniform temperature and pressure. These cases also offered a possible reduction in hydrogenation time (cases Ib and Ic, Table III). It would, therefore, seem that variation with time in temperature and pressure offers no specific advantage.

The industrial hydrogenators are seldom equipped with variable speed stirrers and, as such, plant data showing the effect of speed of agitators on hydrogenation were not available. However, to see if variation in agitation intensity can offer a significant saving in hydrogenation time over and above that obtained using temperature and pressure variation, k_B was explicitly allowed to vary between 50 and 200% of the existing value. The optimization routine did not suggest further reduction in hydrogenation time. The result may seem anomalous, considering that the hydrogenation rates are hydrogen mass transfer controlled and hence

TABLE III

Optimum Operating Conditions

Case	Optimum values of variables								Hydrogenation time (min)	
	x_1	x_2	$x_3 \times 10^2$	x_4	$x_5 \times 10^2$	$x_6 \times 10^4$	$x_7 \times 10^6$	x_8	Current	Optimum
Ia	519.0	0.4364	-0.3758	0.7485	0.1039	0.2098	0.5527	0.048	165.0	138.0
Ib	521.0	0.03554	-0.008528	0.9823	0.0	0.0	0.5527	0.048	165.0	137.0
Ic	520.0	0.0	0.0	1.0	0.0	0.0	0.5527	0.048	165.0	137.0
Id	511.0	0.0	0.0	1.1	0.0	0.0	0.5527	0.060	165.0	111.0
II	505.0	0.0	0.0	1.228	0.0	0.0	1.2360	0.048	195.0	185.0
III	468.0	0.0	0.0	2.168	0.0	0.0	4.6820	0.048	185.0	180.0
IV	462.0	0.0	0.0	2.451	0.0	0.0	4.2454	0.045	180.0	175.0

should increase with increased agitation, i.e., increased k_B. However, increased k_B and hence higher liquid-phase hydrogen concentration adversely affects the selectivity. This is evident because the linoleic and oleic hydrogenation rates are 0.5 and first order in hydrogen concentration respectively (Eqns. 1 and 2). The constraint on selectivity (Eqn. 11) disallowed variation in k_B although it was free to vary between 50 and 200% of the existing value. To study the effect of catalyst loading, a higher concentration (0.06) was used and optimization carried out. As expected, increased concentration resulted in a lower value of hydrogenation time (case 1d, Table III).

Based on the results obtained in case I, temperature and pressure were kept time-invariant (x_2, x_3, x_5 and x_6 were set equal to zero) while optimizing the remaining three cases. Mass transfer coefficient and catalyst concentration were held constant at the prevailing values. Table III summarizes the optimal values of temperature and pressure which resulted in minimum hydrogenation time.

The results presented in this work are for one cycle of simulation and optimization. The computational time for one cycle of parameter estimation and optimization together was 20 sec on a DEC 1090 computer system. Implementation of the one cycle optimum policy will generate fresh hydrogenation data. The model parameters can then be reestimated. After readjusting the bounds on the values of T, pH_2, k_B and m, optimization can be carried out to obtain an improved policy. A repeated application of this strategy would ultimately result in optimal operation of the plant.

The suggested model makes no distinction between cis and trans isomers of the fatty acid chains. These considerations can be incorporated in the simulation model. The optimization will, however, become much more difficult because of additional restriction on product quality, namely, a desired cis-trans break-up. Work in this direction is under way.

This evolutionary operation with the aid of computer simulation and optimization can be most useful in situations where prevailing operating practice must change. This is necessary when (a) feed stock quality or feed type changes (b) a new catalyst is introduced, (c) an oil-batch has to be hydrogenated to a different end point.

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