Operation and Optimization of an Oscillatory Flow Continuous Reactor

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Oscillatory flow reactors (OFRs) are a novel type of continuous reactor, in which tubes fitted with orifice plate baffles have an oscillatory motion superimposed upon the net flow of the process fluid. The combination of baffles and the oscillatory motion creates a flow pattern conducive to efficient heat and mass transfer while maintaining plug flow. Unlike conventional tubular reactors, where a minimum Reynolds number must be maintained, tube-side mixing is independent of the net flow, allowing long residence times to be achieved in a reactor of greatly reduced length-to-diameter ratio. We have evaluated a pilot-scale OFR as a method for continuous production of sterols in an ester saponification reaction. The OFR achieved the required product specification, in a residence time one-eighth that of a full-scale batch reactor. To better understand the effect of the process variables on the reactor performance, the OFR was modeled using a tanks-in-series residence time distribution, combined with the saponification reaction kinetics, giving a "flow-conversion" model which predicted conversion based on flow and feed parameters. This simple model has led to a number of insights into the optimal operation of the OFR, one of which was that the OFR could give desired conversion selectivity at a lower reaction temperature without significant alteration to other process variables. Also, the current product specification could be surpassed, if necessary. These predictions were verified by experiment. A full-scale OFR design based on these results would be less than one-hundredth the volume of a full-scale batch reactor, assuming the same production rate and product quality. The construction of such an industrial-scale OFR for the saponification reaction is now being considered by an industrial collaborator.

Introduction

Oscillatory flow reactors (OFRs) consist of tubes fitted with equally spaced orifice plate baffles. A through flow is applied to the reactor, and an oscillatory fluid motion is superimposed on the entire volume of the fluid in the reactor, such that the interaction of the fluid with the baffle geometry generates highly effective mixing within each interbaffle cavity, as well as along the length of the reactor as a whole.

Figure 1 shows a typical configuration of an OFR. A single-hole orifice baffle type is shown, but multiple-hole baffles can also be used.[1] The choice of a baffle design is usually a compromise between minimizing frictional losses and maximizing the mixing effect. The baffles are uniformly spaced, in a range of 1−2 times the tube diameter apart. The fractional baffle open area, S, is usually in the range of 0.2−0.5, depending on the orifice diameter. Figure 1 shows a "standard" configuration in which baffles are spaced at 1.5 tube diameters, with the baffle orifice diameter half the tube diameter, giving $S = 0.25$.

To characterize mixing in the reactor, two dimensionless numbers are defined, namely, the net flow Reynolds number, Re_n, and the oscillatory Reynolds number, Re_o.

$$Re_o = x_o \omega d\rho/\mu \tag{1}$$

$$Re_n = \rho du/\mu \tag{2}$$

For a sinusoidal oscillation (most common) x_o is the center-to-peak amplitude, ω, the frequency of oscillation,
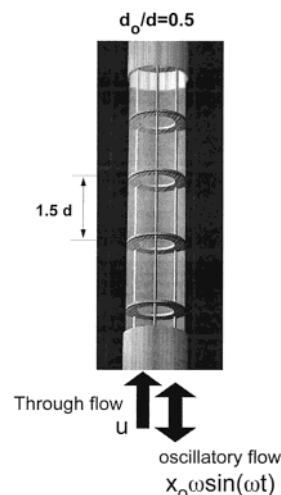

Figure 1. Typical OFR configuration.

d, the tube diameter, ρ, the density, and μ, the viscosity. Re_o is similar to the conventional number for steady flows, except that the velocity term used is the maximum velocity of a sinusoidal oscillation. In contrast to steady flows where transition to turbulence occurs at around $Re = 2100$, flow separation in oscillatory flows occurs for values of Re_o of order 50−100.[2] The net flow Reynolds number, Re_n, describes the externally imposed net flow, where u is the superficial fluid velocity corresponding to the throughput.

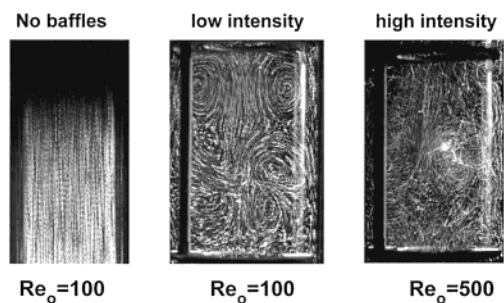To relate the oscillatory flow to the net flow, a *velocity ratio* is defined as

Figure 2. Flow patterns obtained in an OFR.

$$\psi = \frac{Re_o}{Re_n} = \frac{x_o \omega}{u} = \frac{2\pi f x_o}{u} \qquad (3)$$

Usually, it is necessary for the oscillatory flow to be dominant for the full effect of the vortex cycle to be realized; i.e., the flow should be fully reversing. For this criterion to be satisfied, ψ should be, in practice, above 2. In typical operation the value of ψ would be in the range of 2–6, which has been demonstrated to ensure a good approximation to plug-flow behavior.[3]

The *key feature* of oscillatory flow mixing is that the intensity of mixing can be precisely controlled by altering the frequency and amplitude (for a given baffle geometry). This allows a wide range of mixing conditions to be achieved, from "soft" mixing, where Re_o is in the range of 50–500, to the most intense, corresponding to mixed flow (a single continuous stirred tank reactor, CSTR),[2] where Re_o is over 5000. Figure 2 shows the nature of the mixing obtainable. In the first image, no baffles are present in the tube, and no radial mixing occurs (which is undesirable). The second image shows typical vortex formation, which occurs when baffles interact with the oscillatory flow, and the third image shows how the mixing intensity is increased by increasing the oscillation intensity (reflected by the higher value of Re_o).

Background

The main advantage of an oscillatory flow reactor (OFR) is that it offers a means to perform reactions which require long reaction times (of order hours) in a reactor of greatly reduced length-to-diameter ratio. This is achieved while maintaining plug-flow residence time distribution (RTD) characteristics, effective mixing, and high heat- and mass-transfer rates.[4,5] Such requirements are difficult to achieve (for long reaction time processes) in tubular reactors relying on throughput alone to achieve mixing. These features make it possible to consider performing certain reactions continuously, which previously were only possible in batch. In the speciality chemicals industry, for example, stirred tank batch reactors are often employed, which are inefficient and often inherently unsafe because of the large inventories of hazardous and/or volatile liquids. The replacement of these batch processes with more efficient continuous ones and the consequent reduction in the reactor volume improve safety by reducing inventories. This is the primary incentive for speciality chemical companies to investigate the feasibility of novel reactors such as the OFR.

One example of such a speciality chemical process is the production of sterols by ester saponification, which is currently performed exclusively in batch mode using large stirred tank reactors. Typically, a reactor of order

50 m³ is used, operating at 115 °C and a pressure of 2 bar (gauge), on a 24 h batch cycle. Only 2 h of the cycle are actually allotted for the reaction, and therefore simply replacing this reactor with a continuous one, operating at the same mean residence time of 2 h at a throughput of 2.1 m³/h (50 m³ in 24 h), would require a 4.2 m³ reactor, which represents a 12-fold reduction in volume. Despite this obvious advantage of continuous operation, historically there has been no significant incentive for the industry to change to continuous production. This has recently changed, however, because of stricter health and safety regulations (COMAH), and now there is a specific need to improve the inherent safety of the saponification process. Unfortunately, the usual types of continuous reactors are generally considered to be unfeasible for such a long reaction time, and no continuous processes for this saponification reaction were thought to exist, until now.

In cooperation with a major U.K. sterol producer, we have evaluated the feasibility of replacing the batch saponification with a continuous OFR. The rationale was that a continuous saponification reactor would be inherently safer, because the inventory of solvent at high temperature would be greatly reduced. Further safety improvements could be achieved if the reactor could be operated at lower temperature. Another issue was product quality, because batch product quality is often variable, leading to rejected batches. It was hoped that the OFR could achieve a more consistent product. In terms of the reactor volume, a further reduction could be achieved if the OFR could be operated at a shorter residence time than the batch "reaction time", i.e., if the process could be changed from being mixing-limited, as it would be in such a large batch reactor, to being a *kinetically* limited one.

The viability of these requirements have been investigated using a pilot-scale OFR in the Department of Chemical Engineering, University of Cambridge. The study has comprised a proof of concept phase, a reactor characterization study comprising saponification experiments, and a modeling phase in which a reactor flow-conversion model was developed. This paper reports primarily on how the modeling and the characterization study were used to optimize the reactor performance.

Materials and Methods

Experiments were performed in a purpose-built laboratory-scale continuous OFR system. This comprised a feed section, a reactor section, and a product section housed in a self-contained fume enclosure. The reactor was constructed from two 316-grade stainless steel shell-and-tube vessels, mounted on an oscillator base unit. The tube side was a 24 mm i.d., 0.7 mm wall tube, 1.4 m in length, with each tube fitted with a baffle insert assembly using 23.5 mm o.d. baffles with 12 mm diameter orifices spaced at 1.5 tube diameters (36 mm). The two vessels were connected by a U bend, giving an overall reactor path length of 2.9 m, with a total of 80 interbaffle cavities. The reactor volume was approximately 1.3 L.

A variable-frequency, variable-amplitude oscillator was used, in which a pair of stainless steel pistons (24 mm diameter) provided fluid oscillation at the base of the reactor vessels. Piston motion was controlled by a "nutating" cam mechanism driven by an electric motor, which operated the pistons exactly 180° out of phase. Additionally, the cam assembly provided control of the
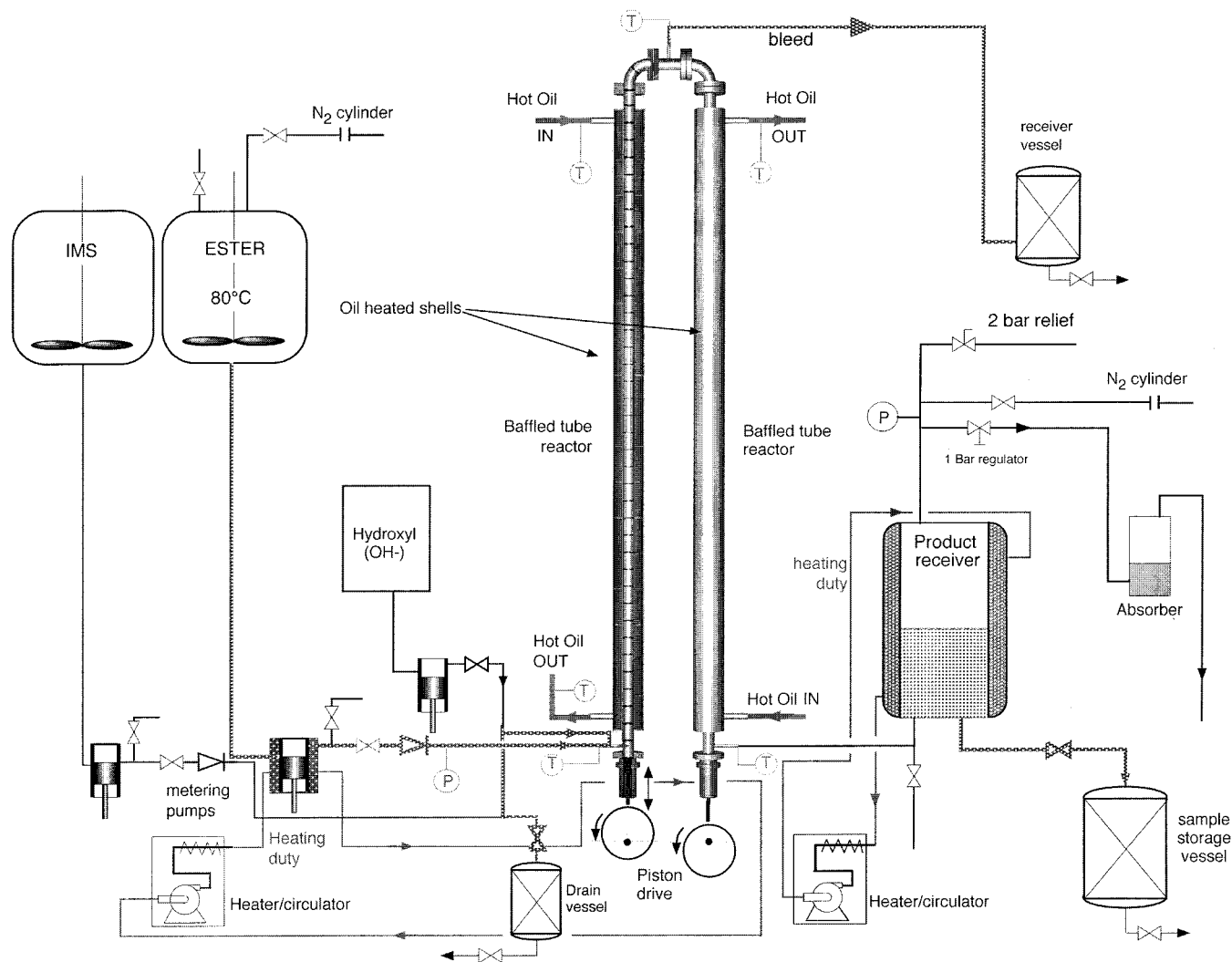
Figure 3. Flowsheet of the OFR saponification reactor system.

amplitude by linear translation of the cam assembly to predefined positions. In practice, frequencies in the range of $1-10$ Hz and amplitudes in the range of $0.5-15$ mm could be achieved, corresponding to Re_o values in the range of $100-1000$ (assuming a viscosity similar to that of water).

The primary reactor variables investigated were the mean residence time (τ) dictated by the flow rate, the tube-side mixing conditions dictated by the velocity ratio (ψ), the reaction temperature dictated by the reactor shell heater temperature, and the feed molar ratio dictated by the flow rate of NaOH relative to the other component feed rates. The molar ratio is defined as the number of moles of NaOH relative to the total hydrolyzable moles in the feed mixture. In practice, the molar ratios used were in the range of $0.6-1.05$.

Process Operation

The reactor was operated at reaction temperatures in the range of $115-85$ °C and at residence times in the range of $5-20$ min which corresponded to flow rates of between 15 and 4 L/h. It should be noted that these residence times are significantly less than the 2 h used in the industrial batch reaction. This range of times was found to be sufficient in practice to achieve the desired conversion in the OFR and, furthermore, is very close to the times predicted by the kinetic data. This result has been reported in a previous paper.[6]

The flowsheet of the laboratory-scale OFR is shown in Figure 3. As shown in the diagram, the ester feed mixture is preheated to 80 °C in a feed vessel (5 L) by means of an electrical hot plate. A mixture of 80% (mass basis) ethanol and 20% water was contained in an unheated second feed vessel. A 47% (by mass) sodium hydroxide solution was contained in a 1 L glass vessel (at ambient temperature). Steady-state flow rates for all of the feeds were ensured by the use of piston-type metering pumps. The protocol was to premix the solvent and NaOH streams approximately 250 mm before the reactor feed point, to allow for preheating by means of line heaters. The preheated ester stream was fed separately, directly into the reactor through a narrow bore tubing passing through a T junction, with the tube extending into the space of the first interbaffle region at the base of the reactor vessel.

The reactor product stream was collected in a 400 mL, jacketed Pyrex pressure vessel. The product vessel was also used to control the pressure in the reactor, via a 2.6 barg nitrogen gas supply fed into the top of the product vessel. A constant production rate was obtained by adjusting a liquid outlet valve to maintain a constant level (by eye) in the product vessel. Samples for off-line analysis were taken by diverting the product stream into 200 mL sample jars at specified times. The analysis method consisted of a titration to determine the unreacted OH content, a solvent extraction procedure to
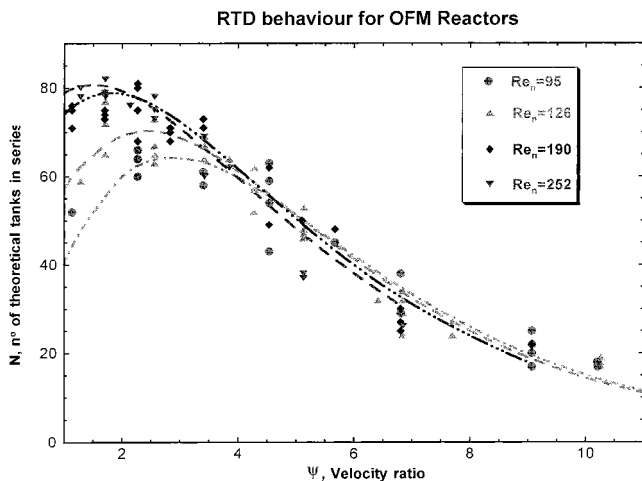
RTD behaviour for OFM Reactors

Figure 4. RTD behavior in an oscialltory flow reactor.

obtain the sterol alcohol fractions, and a composition analysis of the extracted sterol fraction by a gas chromatograph.

Modeling

To better understand the reactor behavior and to be able to simulate reactor operation, a *flow-conversion* model was developed using the well-established technique of combining reaction kinetics and RTD information.[7] The function of the model was to predict conversion of the main reactants in the ester hydrolysis reaction, as a function of reactor mixing conditions and operating variables. The model consists of two sections: a *fluid mechanics model* and a *chemistry model*.

For the fluid-mechanics model, a standard tanks-in-series approach was used as the basis for the reactor flow-conversion model. The main advantage of this approach is the simplicity with which the kinetics can be incorporated. The premise of the model is to represent the baffled tube as a series of ideal CSTRs, the number of which defines the RTD characteristics. This number, N, has been found experimentally in a previous study on the OFR[3] to be a function of both the oscillatory and net flow conditions, as shown in Figure 4.

Figure 4 also shows a correlation fitted[3] to these data which relates N to both the oscillatory and bulk flow conditions (Re_n and Re_o) and has the general form

$$N = \frac{41.5 + aRe_n - bRe_n{}^2}{1.36}\{[(\psi + 1.8 - cRe_n{}^{-d}) + 1]e^{-0.35(\psi + 1.8 - cRe_n{}^{-d})}\} + 1 \quad (4)$$

where $a-d$ are fitted constants. Although empirical, this equation is usable within the operational limits of the reactor for predicting both the trends and absolute values of N to within a margin of ± 3 N values, which is an error of $\pm 4\%$ in practice. With this equation for predicting the RTD behavior of the reactor (for a given set of flow conditions), the conversion of the reactor can be determined by incorporating the reaction kinetics.

It should be noted that the ester saponification reaction system is extremely complex, with hundreds of ester components present in the feed material, which comes from a natural source. To simplify the process, only three main reactions were considered in the chemistry model. Reactions in which two steryl esters, denoted as E_1 and E_2, react with the base, B, to produce
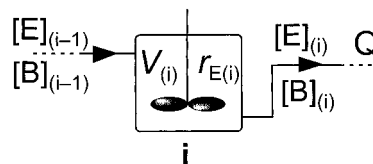

Figure 5. Theoretical CSTR for the tanks-in-series model.

Table 1. Ester Hydrolysis Reaction Scheme

reaction	rate equation	k value
steryl ester 1: $E_1 + B = S_1$ (undesired)	$r_1 = k_1[B][E_1]$	$k_1 = 44.5e^{-4570/T}$
steryl ester 2: $E_2 + B = S_2$ (desired)	$r_2 = k_2[B][E_2]$	$k_2 = 985e^{-5785/T}$
aliphatic ester: $E_3 + B = A$	$r_3 = k_3[B][E_3]$	$k_3 = 133000e^{-7614/T}$

corresponding sterols, S_1 and S_2, as well as an aliphatic ester, denoted as E_3, producing an aliphatic alcohol, A (for reasons of commercial confidence, the exact names of the components are not given), were considered. The kinetics of these reactions were provided by the collaborating company and are shown in Table 1. The Arrhenius-type equations for the rate constants for each reaction are also shown.

The model expresses the species rate equations in terms of a material balance applied to a generic (ith) CSTR in the RTD model of the reactor. Figure 5 shows the basic structure of the model, and the following equation shows the mass balance expressed in terms of a generic ester species, E.

$$[E]_{i-1}Q - r_{E(i)}[E]_iV_i = [E]_iQ \quad (5)$$

Q is the volumetric flow rate (m$^3 \cdot$s^{-1}), r is the rate of disappearance (mol$\cdot$m$^{-3} \cdot$s^{-1}), and V_i is the volume of the CSTR. This equation can be rewritten in terms of the ith CSTR residence time, τ_i,

$$r_{E(i)} = \frac{[E]_{i-1} - [E]_i}{\tau_i} \quad (6)$$

where τ_i is assumed to be equal for all of the CSTRs and, therefore, is given by τ/N, where τ is the mean residence time of the reactor and N is the total number of CSTRs (the model parameter). For the three esters in the chemistry model, the overall rate for the disappearance of the base species (OH$^-$) is given as

$$r_{B(i)} = r_{E_1(i)} + r_{E_2(i)} + r_{E_2(i)} \quad (7)$$

To solve this equation, the kinetics and the mass balance are combined to obtain expressions which give the ith species concentration as a function of the species rate constant, temperature, ith CSTR base concentration, and ($i - 1$)th species concentration. For a generic ester species, E, this expression is given as

$$[E]_i = \frac{[E]_{i-1}}{1 + k_E T\tau[B]_i} \quad (8)$$

where the ith base concentration is the unknown. When the individual reactant concentrations are substituted and rearranged, the overall expression can be found as follows:
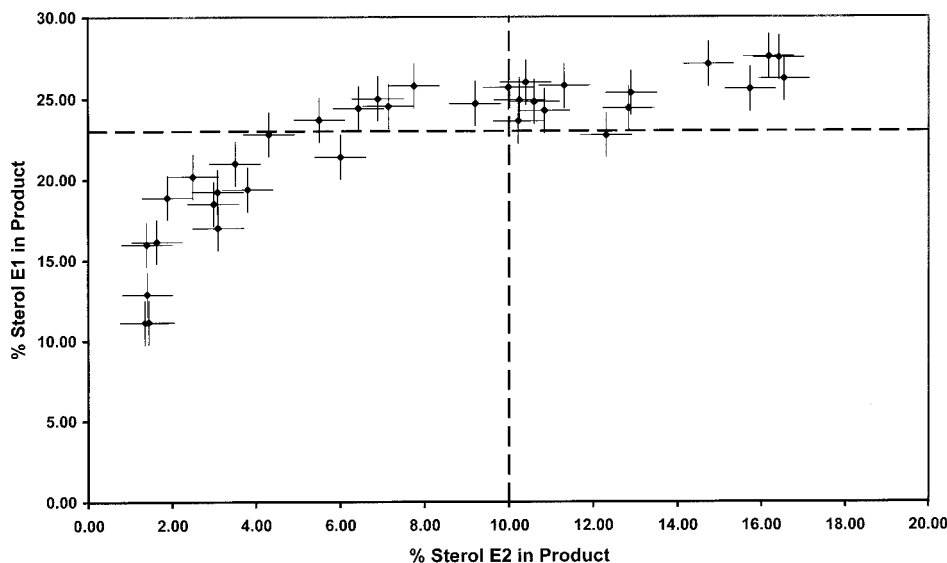
Figure 6. Results of experiments conducted at 115 °C.

$$[B]_i \left[\frac{1}{\tau} + \frac{k_1 T[E_1]_{i-1}}{1 + k_1 T\tau[B]_i} + \frac{k_2 T[E_2]_{i-1}}{1 + k_2 T\tau[B]_i} + \frac{k_3 T[E_3]_{i-1}}{1 + k_3 T\tau[B]_i} \right] - \frac{[B]_{i-1}}{\tau} = 0 \quad (9)$$

This equation is solved for each of the CSTRs, for $i = 1-N$, by determining its positive root. The model was implemented on an Excel spreadsheet by applying the equations for each successive CSTR (from $i = 1$ to N), using [E] values calculated from the previous CSTR $(i - 1)$ to obtain the ith values (for $i = 1$, the entry species concentrations are calculated from the feed conditions). The value for the base concentration $[B]_i$ obtained in the solution is then the value for the concentration of base leaving the ith CSTR, which is used for the next $(i + 1)$ CSTR, and the whole process is repeated up to $i = N$.

The inherent assumption in this model is that there is no segregation. The reaction in each theoretical stirred tank is assumed to be *not* mass-transfer-limited but *kinetically* limited. Another assumption is that the dispersion of ester into the aqueous medium is ideal. Although there is considerable evidence to suggest that this does, in fact, occur in oscillatory flow reactors,[4] it must be acknowledged that this is a simplification to the model and that the predicted conversion may not therefore exactly match the experimental conversion for a particular set of conditions. A good match, however, would indicate that mixing was very close to ideal (assuming, of course, that the kinetic data are reliable).

Results

The experiments were principally conducted at 115 °C for direct comparison with the industrial batch process. Figure 6 shows the results of all runs conducted at 115 °C, expressed in terms of the desired sterol S_2 vs the undesired sterol S_1. The runs were conducted on the pilot reactor at residence times from 10 to 20 min and molar ratios from 0.6 to 1.05. The data points in the upper left-hand area represent product that fulfills the current specification that greater than 23% (by mass) of the total product is S_2 and less than 10% is S_1. The error margins shown were due to the analytical method,
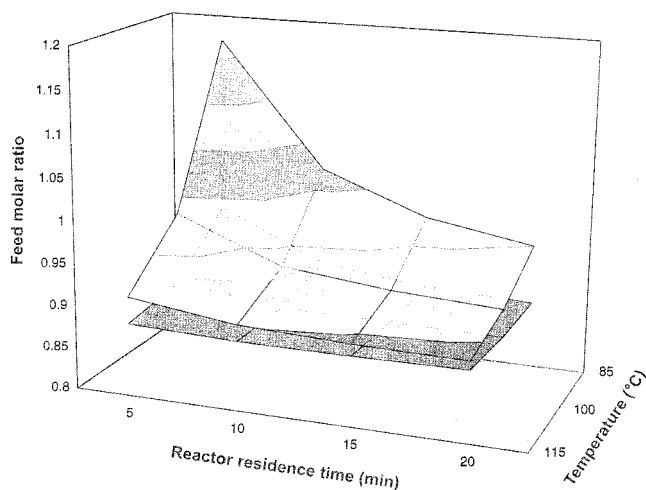

Figure 7. OFR model prediction.

described previously. From the figure it is clear that the reaction could be successfully performed in the OFR pilot reactor, with a subset of reaction conditions leading to a product which fulfills the current batch product specification.

To better understand the performance of the reactor, modeling was performed. The model described previously was used to predict the conversion of the desired ester species, E_2, and the undesired species, E_1, as functions of the residence time and molar ratio at fixed temperatures. Figure 7 summarizes the results of the modeling in the form of two surface plots. The upper surface represents the conditions above which the level of the undesired sterol, S_1, exceeds 40% conversion. The lower surface represents the conditions below which the level of desired sterol, S_2, would be lower than 95% conversion. These conversion constraints define a product that is considerably better than that which is currently accepted from the batch process.

The domain between the surfaces represents the conditions at which these product specifications would be achieved. Temperatures above 115 °C are not considered because they would require the use of higher pressures (to keep the solvent below its boiling point), thereby making the process more hazardous. The use of temperatures below 85 °C would result in practical
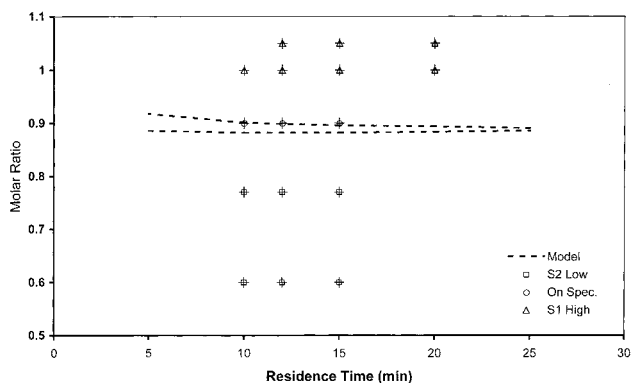
Figure 8. Comparison of experimental results with a predicted operating window at 115 °C.

problems, because the viscosity of the steryl stearate mixture would markedly increase, as the temperature approached the mixture's melting point. The lower residence time constraint of 5 min is primarily a practical one, because the length reduction advantage of the OFR would be lost below this value. Also, operation at lower residence times would entail the use of higher molar ratios to remain in the operating window. This would result in the use of greater amounts of base, which is undesirable for operational reasons. A 20 min residence time is chosen as the upper constraint because it is at the upper limit of operability at 115 °C.

The effect of temperature was investigated because, from the Arrhenius expressions for the kinetics of the formation of S_1 and S_2, given in the previous section, it is clear that the reaction constant, k_1, for the formation of S_1 will decrease more than k_2 as the temperature is decreased. Hence, the discrimination between the two species could be increased by operating at a lower temperature than that currently used. Figure 7 shows that the desired product specification can be achieved in the OFR at temperatures below 115 °C. It is clear that, as the temperature is decreased, the molar ratio operating window widens. This has significant consequences for the operation of an industrial OFR, because the product specification would be more easily achieved, and the process would not have to be as tightly controlled.

To evaluate the validity of this flow-conversion model, the experimental results at 115 °C were compared to the predicted operating window for that temperature. Figure 8 shows the experimental conditions used and the predicted 115 °C operating window (see Figure 7). The square data points represent the conditions at which the desired product specification was not achieved because of insufficient formation of sterol S_2. The triangular data points represent the conditions at which too much sterol S_1 was formed. The circular data points represent the conditions required for a product to fall within specification. The error bars represent the degree of accuracy with which the residence times and molar ratios were able to be maintained.

It is clear that the agreement between the predicted operating window and the experimental results is good. It should be noted that the operating window at 115 °C is relatively narrow when compared to the error margins. Hence, operating the process at this temperature makes it inherently difficult to maintain the molar ratio accurately enough to ensure that the product remains within specification. The modeling predicted that this problem would be greatly lessened by operating at a lower temperature (see Figure 7). Furthermore, the model predicts that product quality could be enhanced by lower temperature operation. This is illustrated in Figure 9, where the range of product compositions that are theoretically possible at 85 and 115 °C for molar ratios of 0.9 and 1.0 is shown. The curves shown are essentially operating lines. It should be noted that full conversion of S_2 cannot be achieved at substoichiometric molar ratios. It is clear that selectivity of S_2 over S_1 is significantly greater at 85 °C than at 115 °C. For example, at 85 °C, at a molar ratio of 0.9, 95% conversion of S_2 is achieved at 22% conversion of S_1, whereas at 115 °C for the same S_2 conversion, S_1 conversion would be 36%.

To confirm the predicted advantages of lower temperature operation, a further series of experimental saponification runs were conducted at 100 and 85 °C. The conditions used were within the operating window shown in Figure 7. The products of all of these runs were found to surpass the specification, demonstrating that low-temperature operation of the OFR is indeed viable.
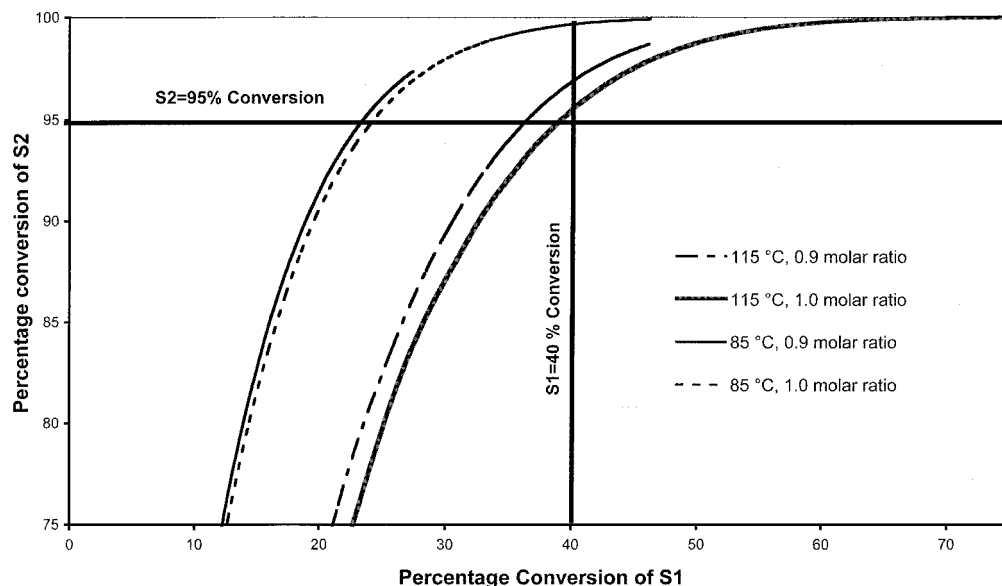

Figure 9. Effect of temperature and molar ratio on the product composition.

The effect of reducing the temperature is illustrated by the following example: the averaged composition of all products from experiments conducted at 115 °C at a residence time of 15 min and a molar ratio of 0.9 was 26.0% (of the total product) S_2 and 10.4% S_1, whereas at 85 °C the averaged result for the same conditions was 25.5% S_2 and 6.5% S_1. It is therefore clear from the experimental evidence that the advantages of lower temperature OFR operation predicted by the model can be realized.

Conclusions

The greatest incentive for industry to change the batch saponification plant to continuous processing is safety, because a large batch reactor containing solvent above its ambient temperature boiling point represents a major hazard. An equivalent continuous reactor would greatly lower the inventories of solvent. In this study it has been demonstrated that the reaction *can* be performed in an OFR. Furthermore, this can be achieved while operating at a temperature below the boiling point of the solvent, further improving safety.

For the OFR, there are larger operating windows at lower temperature, leading to greater flexibility of operation. The product specification is more easily achieved and can even be surpassed, if required.

The residence time we recommend for the industrial-scale operation of the OFR is 12 min, 1 order of magnitude less than the 2 h used in the batch reactor. This assumes that scale-up can be achieved without loss of mixing efficiency, a feature of OFRs for which there is already evidence to confirm this.[1] This shorter residence time, combined with the inherent 100% occupation time of a continuous reactor (rather than the one-twelfth batch saponification occupation time), would require a continuous OFR of only 4.1 m³, which is at least 2 orders of magnitude smaller than the 50 m³ batch reactor for the same production rate. Hence, solvent inventories would be greatly lowered by conversion of this batch process to continuous processing in an OFR.

In summary, it has been demonstrated that the performance of an oscillatory flow reactor can be predicted for a saponification reaction by modeling the behavior of the reactor by the tanks-in-series approach. The predictions of the model led to operation of the OFR at more optimal conditions, which will lead to improved and more consistent product quality and an inherently safer plant.

Notation

ψ = velocity ratio
μ = viscosity [Pa·s]
r = density of the reaction mixture [kg/m³]
τ = residence time [s]
ω = angular velocity of the oscillation [rad/s]
d = internal diameter of the reactor [m]
f = frequency of the oscillation [Hz]
$k_{1,2,3}$ = rate constants of reactions (1)−(3) [m³/mol·s]
N = number of ideal tanks
Q = volumetric flow rate [m³/s]
r = rate of disappearance [mol/m³·s]
Re_n = net flow Reynolds number
Re_o = oscillatory Reynolds number
S = fractional baffle open area
u = average fluid velocity [m/s]
V_i = volume of the ith ideal stirred tank [m³]
x_0 = center-to-peak amplitude [m]

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