Dense–Bed Column Crystallizer: An Experimental and Analytical Study

The performance of a continuous, vertical, dense-bed column crystallizer is evaluated by comparing experimental and predicted column temperature profiles and by examining product composition at various operating conditions. The separation obtained in an eutectic system is modeled by a component conservation equation coupled with a nonadiabatic energy balance equation. The critical parameter in the analysis is the axial dispersion coefficient and the measured values of this coefficient were similar in magnitude to those reported for ice washing columns. The dense-bed column apparently achieves nearly ideal plug flow.

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SCOPE

For a variety of reasons the concept and practice of conducting crystallization inside a column in a manner somewhat analogous to distillation is of considerable interest. Primary impetus is to attain higher purity product than can be achieved in a single stage of conventional crystallization; however, much lower energy requirements (compared to distillation) and avoidance of high-speed mechanical separation equipment are additional incentives. The objective of this investigation is to study experimentally and to model mathematically the performance of a novel, vertical, dense-bed continuous crystallizer.

The concept of a column crystallizer is to form a crystal phase, internal or external to the column, and force the solids to flow countercurrently against a stream of enriched reflux liquid formed by melting the crystals. A temperature gradient is thus established in the column through which crystals are forced. In principle, such a device can be operated continuously or batchwise on either eutectic or solid-solution systems. A limitation inherent in column crystallization is the difficulty of controlling the solid phase movement since, unlike most distillations, the phases have similar densities, causing gravitational separation to be generally ineffective.

Based on reported successes of dense-bed columns with external crystal formation (McKay, 1965) and rotating spiral devices with internal crystal formation (Powers, 1963), it was anticipated that high separation efficiency could be achieved in short dense-bed columns having provision for internal crystal formation. Column crystallizers that operate with low porosity crystal beds, that is, dense-beds, normally have crystals formed outside the column. The crystal slurry is then forced by various means into the column at the end opposite the melter, hence the name—end-feed. Although the purification zone of the end-feed column has the desirable feature that it contains no mechanical internals, the feed, heating, and cooling orientation somewhat limits the separation power and applicability of the device. Only a limited supply of reflux liquid can be supplied for crystal washing without melting the contents of the column. Columns that employ center feeding similar to a distillation column typically use screw conveyors to move crystals. A lean (high porosity) crystal slurry must be maintained to avoid conveyor stoppage problems; however, reflux control is possible and high quality product has been achieved in spiral conveyor columns with high length to diameter ratios.

A laboratory column crystallizer that included an integral crystal forming section at the top of the column and positive means for controlling movement of the dense crystal bed was designed for this investigation. Liquid feed enters the middle of the column, mother liquor flows overhead from the freezer at the top of the column, and purified product is withdrawn as a liquid melt from the heater at the base of the column. Acetamide-water solutions were utilized as test material. The production of highly refined acetamide from feed solutions containing varying concentrations of water was an objective of the experimental program.

The analytical model presented in this study departs from former approaches in several respects. Previous investigators of continuous center-feed columns have utilized extraction models to describe isothermal enrichment section behavior and have not been concerned with nonadiabatic operation of the enrichment section or other sections of the column. In this investigation, a nonadiabatic, plug-flow axial dispersion model was employed to describe freezer, heater, stripping, and enrichment sections. Interphase transport terms are not considered in this study since axial dispersion and bulk flow effects have been shown by others to dominate column performance. An iterative numerical technique was employed to solve the resultant second-order differential equation.

CONCLUSIONS AND SIGNIFICANCE

A dense-bed column crystallizer with center feed was successfully operated with continuous product drawoff. Movement of the crystal bed was controlled with a porous, reciprocating piston. Product quality was insensitive to feed rate, product rate, and feed composition, indicating a high degree of separation power in the column. Refined acetamide of greater than 99.9% purity was consistently produced from feed concentrations of 97.0 to 90.0% acetamide containing water as a contaminant. The dense-

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bed crystallizer column developed for this study has considerable scale-up potential since mechanically it is less complex than spiral conveyor columns yet exhibits similar performance.

The mathematic formulation circumvents problems associated with utilizing extraction models for mechanistic studies or design. Only the sensitivity of column performance to bulk flows, external heat flux, and the magnitude of axial dispersion coefficient is included. In extraction models, one must estimate the ratio of adhering liquid-to-solid, the interfacial area, and the mass transfer coefficient—all of which are difficult to establish. The nonadiabatic, plug flow, axial dispersion model described in this investigation requires only one system parameter, the liquid axial dispersion coefficient, which is an experimentally measurable quantity.

Comparison of experimental temperature profiles with computer data utilizing the axial dispersion coefficient as a free parameter yielded coefficients of 0.2 to 0.3 cm^2/s. These values are about an order of magnitude less than the axial dispersion values of 1.3 to 3.5 cm^2/s reported for columns with oscillating spiral conveyors, and they are slightly larger than coefficients reported for nonpulsed ice washing columns. As anticipated, the dense-bed column crystallizer exhibits near plug flow characteristics for both solid and liquid phases.

Crystallization purification processes depend upon the ability to selectively solidify and recover, in a refined state, a single constituent from a solution. In many organic and inorganic systems, the phase behavior is eutectic, and a pure component can be generated as a solid phase by crystallization from solution. The high degree of separation power in a single contact stage plus low energy requirements compared with distillation provide the attractiveness of crystallization as an industrial separation technique.

Column crystallizers and ancillary equipment are designed in a multitude of ways to achieve controlled solid phase movement, high product yield, and efficient heat addition and removal. Column crystallizers have been systematized into either end-feed or center-feed types. The Phillips style column (McKay, 1965; May, 1969) design is called an *end-feed unit* because crystals are formed in external equipment and the slurry is introduced into the column at the top. This type of column has no mechanical internals to transport solids and instead relies upon hydraulic force to move the solids phase into the melting zone. Impure liquid is removed through a filter directly above the melter. Another commercial design is the spiral-conveyor (Schildknecht, 1963) column. In this device, a rotating or an oscillating and rotating spiral is used to convey the crystals. This column is classed as a center-feed column crystallizer since liquid feed is introduced between the two enriched streams leaving the column, and the crystals are formed within the unit. A horizontal version of the center-feed crystallizer, which successfully utilizes scraper-conveyors to transport the crystals from the cold to hot zones, has been reported recently by Brodie (1971).

Schildknecht (1961) reported the development of a laboratory crystallization column which can compete with zone refining in its ability to purify materials from eutectics and solid solutions and which potentially can be operated continuously. Schildknecht and co-workers were primarily concerned with production of ultra-pure materials, and they concentrated their efforts at that time on development of laboratory columns operated at total reflux conditions. The subsequent open literature regarding column crystallization technology can be divided roughly into investigations of end-feed and center-feed columns. Center-feed column investigations, which are of prime interest in this study, are summarized in Table 1. Reviews of column crystallizer literature and patents can be found in Zief and Wilcox (1967), Albertins (1969), and Player (1969).

Powers (1963) presented the first mathematical analysis of the center-feed Schildknecht column. He examined total reflux behavior for both binary eutectic and solid solution systems. Powers employed steady state mass balance equations and a unidimensional differential countercurrent extraction model to describe the purification process. Subsequent students of Powers experimentally and theoretically investigated the first-order character of the equations proposed by Powers. Albertins (1969) conducted total reflux tests with the system cyclohexane and benzene and extracted mass transfer and axial dispersion coefficients from appropriate plots of experimental data. Gates (1970) extended Albertins' results; both reported dispersion coefficients of the same magnitude as pulse extractors. Henry (1970) conducted continuous flow tests on the eutectic system cyclohexane and benzene, determined the relative importance of interphase mass transfer and axial dispersion groups, and concluded that the axial dispersion group is dominant. Ritter (1969) used a plug-flow, axial dispersion model to describe washing mechanism in continuous ice purification columns. The washing operation was modeled successfully with an axial dispersion formulation; the dispersion coefficients obtained serve as one basis of comparison for the results of this study.

MODEL FORMULATION

The formulation of a washing model with sidewall heat flux used to describe operation of a continuous dense-bed crystallizer is presented in this section. Assumptions are listed along with their implications. Heat and mass balance equations are coupled through an equilibrium relationship and the resulting equation is solved numerically for the temperature profile in the column. Details of the model development can be found in Moyers (1971).

ASSUMPTIONS

1. There is no clearly defined adhering, liquid phase. This assumption eliminates the need for an additional equa-

Table 1. Column Crystallizer Investigations

Column operation (center-feed only)	Treatments	
	Theoretical	Experimental
Solid solutions		
Total reflux—steady state	1, 2, 4, 6	1, 4, 6
Total reflux—dynamic	2	—
Continuous—steady state	1, 4	4, 8, 9
Continuous—dynamic	—	—
Eutectic systems		
Total reflux—steady state	1, 3, 4, 7	1, 3, 6
Total reflux—dynamic	—	—
Continuous—steady state	1, 5, 10	5, 8, 9, 10
Continuous—dynamic	—	—

1. Powers 1963)
2. Anikin (1969)
3. Albertins and Powers (1969)
4. Gates and Powers (1970)
5. Powers and Henry (1969)
6. Schildknecht (1961)
7. Arkenbout and Smit (1968)
8. Betts et al. (1968)
9. McKay (1959)
10. Bolsaitis (1969)

tion describing impurity extraction in both the component mass balance and energy balance equation. Analysis of continuous column crystallization data by Henry (1970) revealed that extraction effects, if present, are swamped by the influence of mass axial dispersion. Likewise, Ritter (1969) has successfully employed plug-flow axial-dispersion models without extraction effects to describe the performance of dense-bed adiabatic countercurrent brine-ice wash columns. Extraction effects were lumped into the magnitude of the axial dispersion.

2. Thermal equilibrium exists between phases at any cross section. This assumption eliminates interphase mass and heat transfer, leaving only axial dispersion, bulk flow, and wall heat flux terms to describe the process. Yagi et al. (1963), in their investigation of dense-bed, end-feed columns, support this assumption. Barrera-Larrarte (1969) show that equilibrium between the phases is obtained in less than 20 s, a small portion of the total residence time.

3. The liquid phase is saturated at all locations in column. Mass and energy balance equations, therefore, are coupled through the use of the equilibrium relationship. In this study, the bulk liquid phase has been found experimentally to be saturated in the high-purity zone although at the cold end of the column the liquid tends to be slightly supersaturated.

4. The solids flux is ideal piston flow.

5. The liquid flux is piston flow modified by axial dispersion.

6. The thermal and mass axial dispersion coefficients are equal, and further, they are constant throughout the length of the column as are the porosity, liquid-density, and specific heats. This assumption is not valid for systems with large thermal conductivity but is reasonable for aqueous or organic mixtures.

7. Although not a general assumption, the equilibrium curve of the eutectic system considered in this study is linear in the composition range used.

FORMULATION

One-dimensional mass and energy balance equations describing column crystallization have been developed previously (Moyers, 1971). Applying the above assumptions to the general energy balance equation, the following equation results:

$$\frac{d}{dz}(\underset{\sim}{S}H_s + \underset{\sim}{L}H_L) = (D_{ax}\phi\rho C_p)^L \frac{d^2T}{dz^2} - \frac{U^w 4}{d}(T - T^w) \tag{1}$$

Allowing thermal equilibrium at any column cross section and assuming heat of mixing effects are nil and that specific heats of solid and liquid are equal, Equation (1) can be arranged to the following form:

$$\lambda_s \frac{d\underset{\sim}{L}}{dz} = (D_{ax}C_p\phi\rho)^L \frac{d^2T}{dz^2} - (\underset{\sim}{L}C_p + \underset{\sim}{S}C_p)\frac{dT}{dz} - \frac{U^w 4}{d}(T - T^w) \tag{2}$$

Addition of component mass balance equations for a differential column segment results in

$$(y - x)\frac{d\underset{\sim}{L}}{dz} = -\underset{\sim}{L}\frac{dy}{dz} + (\rho\phi D_{ax})\frac{d^2y}{dz^2} \tag{3}$$

The equilibrium curve for an eutectic system in the high purity range is approximated by the linear relationship

$$T = Ay + B \tag{4}$$

Substituting the equilibrium relationship into Equation (3) and employing the saturated liquid phase assumption yield

$$(T - B - xA)\frac{d\underset{\sim}{L}}{dz} = -\frac{\underset{\sim}{L}dT}{dz} + (D_{ax}\phi\rho)^L \frac{d^2T}{dz^2} \tag{5}$$

Equations (5) and (2) can be solved for $d\underset{\sim}{L}/dz$ and equated; thus, the energy balance and component mass balance equations are combined into the single equation below:

$$A'\frac{d^2T}{dz^2} + B'\frac{dT}{dz} + C'T = D' \tag{6}$$

Using second-order numerical approximations for the temperature derivatives, Equation (6) can be rearranged as follows:

$$A^*T_{n+1} + B^*T_n + C^*T_{n-1} = D^*\Delta Z^2 \tag{7}$$

The coefficients in Equation (7) contain dependent and independent variables and system parameters. This equation can be solved for the column temperature profile utilizing auxiliary expressions defining liquid and solid flux at various positions in the column, including the feed point.

An enthalpy balance can be made at the feed location assuming no discontinuity in the temperature profile. The feed is assumed to enter the column into a differentially small segment located at the feed point and may be saturated or superheated. The resulting equation can be arranged into a form similar to Equation (7):

$$a'T_{f+1} + b'T_f + C'T_{f-1} = d' \tag{8}$$

Equations (7) and (8) form a tridiagonal system and can be solved explicitly for temperature by Gaussian elimination methods. To solve Equations (7)and (8), values of the liquid flux must be known at all positions. Conversely, in order to solve for liquid flux, the temperature at all mesh points including the end points is required. Terminal temperatures were selected to conform to material balance and product purity demands. The solution procedure was to first select an overall column temperature profile, including end points, numerically solve Equation (2) for liquid flux (starting with known terminal flux of liquid from the freezer) using a modified Euler second-order predictor-corrector method, and then utilize the stored liquid flux values to solve Equation (7) for a new temperature profile. The procedure is repeated until the solution converges.

COMPUTATIONS

To test the mathematical model, anticipated experimental conditions were simulated. Parameters such as physical properties and heat transfer coefficients are generally available and column configuration is fixed by design. Concentration of impurity in the crystal phase, however, must be obtained experimentally. Both product and exit impure liquor composition must be specified to solve Equation (6). The product composition will approach the residual solid impurity level very closely in a properly designed column.

Figure 1 illustrates the typical variation of liquid and solid fluxes present in a dense-bed column. As expected, a jump change in the liquid flux and to a lesser extent in the solid flux occurs at the feed location. A moderate amount of melting occurs in the purification section of the column. Solids are contained inside the column; consequently, solid flux is zero at both ends of the column. Liquid flux must satisfy both overhead impure liquor rate and melted product rate and must change sign in the melter section.

Column temperature profiles were computed for various

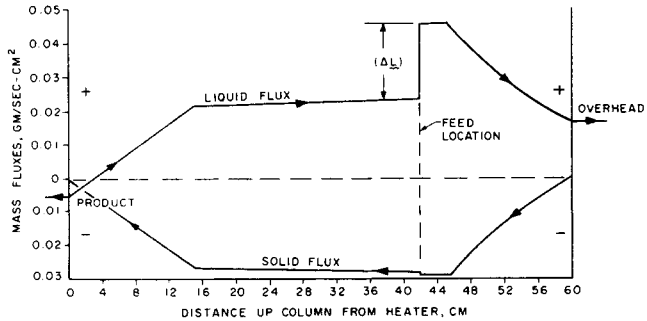

Fig. 1. Computed liquid and solid mass fluxes in dense bed column crystallizer.

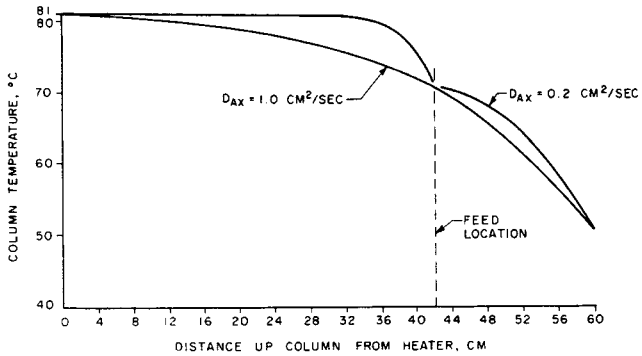

Fig. 2. Computed column temperature profiles.

values of axial dispersion coefficient since this is the only free parameter in the model. Figure 2 shows typical computed temperature profiles and the extreme sensitivity of temperature to the magnitude of the axial dispersion coefficient. A cusp at the feed point is apparent at low axial dispersion values; however, the temperature profile was not measured between the freezer and feed point or inside the freezer in this investigation, and the predicted cusp was not confirmed experimentally. It is apparent from Figure 2 that near plug flow conditions are necessary to achieve efficient separation in short dense-bed column crystallizers.

EXPERIMENTAL PROCEDURE

The crystallizer column configuration utilized for this investigation is illustrated on Figure 3. The solid phase is constrained inside the column by screens below the heater and on the bottom of the porous piston. Both the piston and scraper are connected to the same shaft; hence, they rotate and reciprocate as a unit. The feed pumps, column feed tank, and the majority of the piping were installed inside a heated enclosure since the freezing point of the test solution was well above ambient conditions. Details of apparatus can be obtained in patent by Moyers (1974).

The binary eutectic system, acetamide and water, was used as the test system. Acetamide purification was chosen for test purposes primarily because acetamide melts at 81°C; therefore, hot water served as both heating and cooling medium. Equilibrium data for the acetamide water system were obtained from Mullin (1961) and Timmermans (1950).

Samples were analyzed using a dual column Hewlett Packard No. 5750B research chromatograph equipped with a thermal conductivity detector and Parapak Q columns. A Moseley Model 7128A strip chart recorder and a Hewlett Packard No. 3370A digital integrator were utilized for analyzing samples. Samples were placed in glass sample bottles partially filled with preanalyzed anhydrous methanol (Matheson Coleman and Bell-Analyzed Reagent A.C.S.). The methanol was dehydrated to about 150 ppm water by Linde No. 3A molecular sieves. Product samples were obtained directly from the column by inserting a syringe needle through the septum in the sample port at the bottom of the column and then injecting the liquid through a septum in the top of a sample bottle. The chromatograph was operated in the programmed mode because low temperatures were required for accurate water and methanol resolution and because higher temperatures were necessary to reduce acetamide evolution time. Detailed description of experimental procedures and analysis methods can be found in Moyers (1971).

A properly operating column contains a densely packed nearly opaque crystal phase throughout the length of the column. To shorten the lengthy startup times, the column was charged with reagent grade acetamide (containing about 1% water) rather than feed stock, and a dense crystal bed was carefully formed inside the column by slowly cooling the contents. Feed was then initiated, and the base slowly heated to the product melting point. After the base temperature reached 81°C or above, product was withdrawn at a constant rate, and conditions were controlled so that crystal inventory remained constant. Steady state was assumed to be achieved when the water content of the product decreased and remained at a low level. The overall material balance of the column was consistent with the impure feed stock. Temperature measurements were obtained from thermistor probes positioned along the column for use in comparing with computer profiles.

EXPERIMENTAL RESULTS

A summary of overall column performance is shown on Table 2. These data were obtained after steady state conditions were achieved. Startup data are indicated on Figure 4 and show the composition of the product taken from the base of the column as a function of time. Steady state conditions were assumed to prevail when product water concentration decreased to and remained at a constant value.

Product quality showed insensitivity to feed rate, reflux rate, and feed composition, indicating a high degree of separation power in the crystallizer. Acetamide purities exceeded 99.9% through feed concentration variations from 3 to 9 wt. % water. The product quality and separation efficiency obtained in the dense-bed column are shown on Table 2 and are similar in magnitude to those reported by Henry (1970) for an oscillating spiral-type column. Because of the high separation efficiency exhibited by the 30-cm column, the purification zone of the column was shortened to 15-cm for tests 30 and 32. A slight reduc-

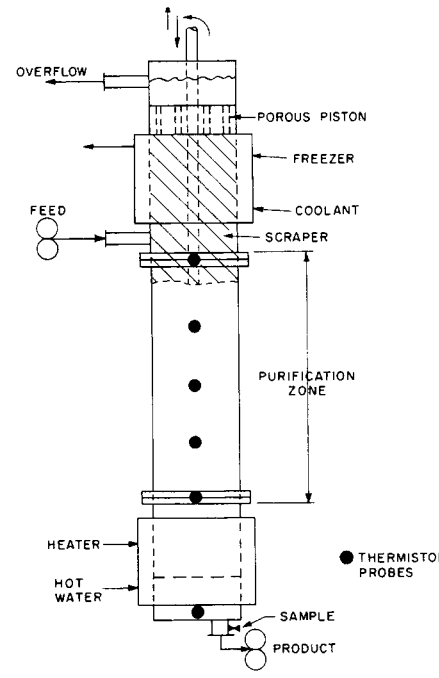

Fig. 3. Configuration of dense bed column crystallizer.

Table 2. Experimental Results for Continuous Column Operation

Test no.	Feed Rate, cc/hr.	Feed Composition, wt. % water	Product Rate, cc/hr.	Product Composition, wt. % water	Reflux ratio	Column performance Separation efficiency, %	Yield, %
23	210	4.2	70	0.020	3.7	99.5	35.0
25	450	5.1	50	0.050	5.5	98.4	11.7
26	202	4.4	70	0.070	2.4	98.9	36.2
27	225	8.9	40	0.120	8.6	98.6	19.5
29	328	2.9	68	0.090	4.5	96.7	21.3
30	304	3.4	64	0.150	8.7	95.7	21.8
32	330	4.0	150	0.260	3.3	93.4	47.5

Tests 30 and 32 were conducted in a 15-cm column; other tests were performed in a 30-cm column.

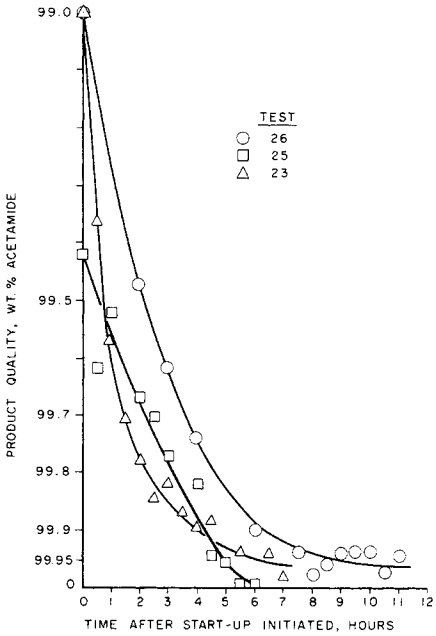

Fig. 4. Time required to achieve steady state operation.

tion in efficiency was measured although product purity remained near 99.9%.

Reported yields on Table 2 are scattered. The higher the feed purity in an euctectic system, the larger the potential product yield will be. In column crystallizers, it is possible (although not always practical) to attain near maximum theoretical yields. In this work the actual product yield for a given test was primarily limited by the cooling capabilities of the equipment.

Experimental temperature profiles were obtained from thermistor measurements in the purification zone of the apparatus for eventual comparison with computer profiles. Figure 5 shows typical column temperatures between the feed point and heater for feeds of approximately the same composition. A very sharp rise in temperature occurs below the feed point. This suggests that near plug flow conditions in the liquid phase are present since an abrupt rise occurs in the model at this location for low values of axial dispersion.

Figure 6 shows a comparison of experimental temperature profiles obtained in 15- and 30-cm columns for similar feed concentrations. The short column produced acetamide of essentially the same quality as the long column. Further reduction of column length would likely have a deleterious effect on performance.

In the design of Phillips (McKay, 1965) and Brodie (1971) columns, considerable attention was given to formation of coarse pure crystals by providing an environment for slow crystal growth. In the dense-bed concept, the only concern is to form a solid crystalline phase that can be contained and manipulated in the column. Observation of particles initially formed at the feed zone in the absence of a dense crystal bed showed that very fine crystals were spontaneously formed. During continuous melt crystallization with a dense crystal bed in the column, discrete crystals were not observable. In fact, the bed gave the appearance of being consolidated. Despite a complete lack of care in forming crystals, separation efficiencies in the dense-bed column crystallizer designed for this investigation were 98 to 99%.

COMPARISON WITH MODEL

Rate data and terminal conditions from Test No. 25 were used to simulate crystallization column operation on the University of Delaware's SDS-9300 computer. Temperature profiles computed for several values of axial dispersion are indicated on Figure 7 and experimental data for Tests

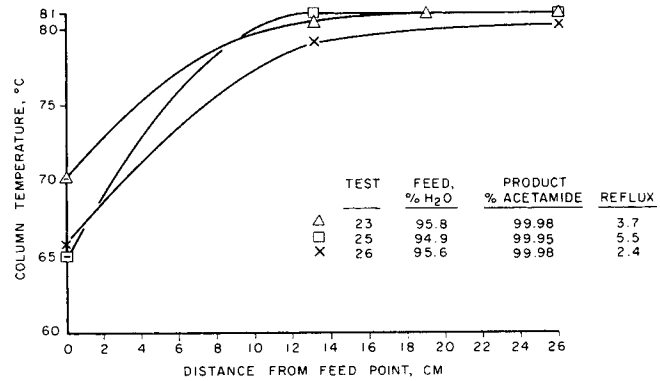

Fig. 5. Experimental temperature profiles—30.9-cm column.

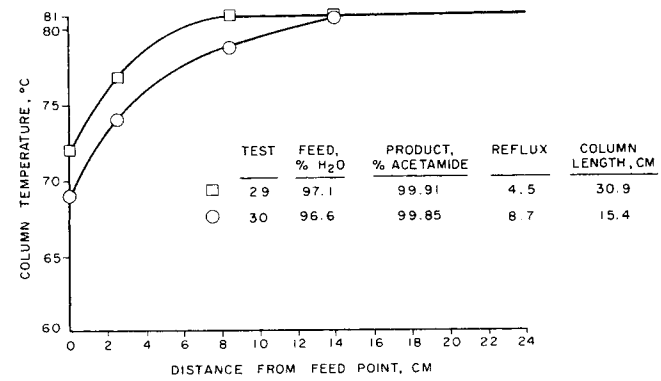

Fig. 6. Experimental column temperature profiles.

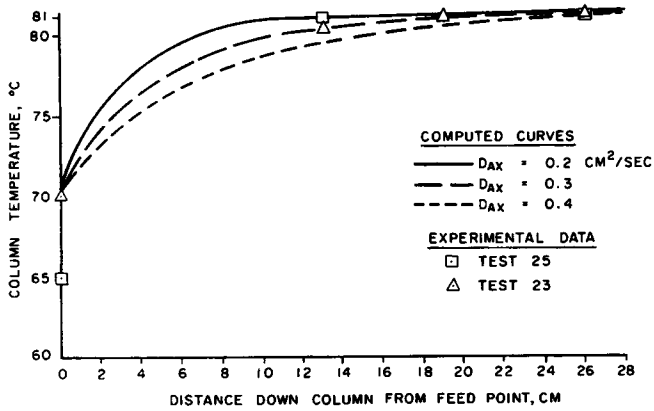

Fig. 7. Comparison of computed and experimental column temperature profiles.

TABLE 3. COMPARISON OF AXIAL DISPERSION COEFFICIENTS

Column type	Dispersion coefficient, cm/s	Reference
Spiral conveyor crystallizer	1.6-3.5	Albertins (1969)
Spiral conveyor crystallizer	1.3-1.7	Gates (1970)
Countercurrent ice wash column	0.025-0.17	Ritter (1969)
Dense-bed crystallizer	0.20-0.30	Moyers (1971)

23 and 25 are also shown. Experimental data are approximated quite well by utilizing an axial dispersion coefficient of 0.2 to 0.3 cm²/s in the nonadiabatic model. The thermistor measuring column temperature at the feed location is influenced by the close proximity of the cooler, causing the recorded temperature at the wall to be lower than the average column contents.

Table 3 presents a summary of axial dispersion coefficients experimentally determined in various column purifiers. Although other column investigators used extraction models to describe purification mechanisms, they concluded that axial dispersion dominated. Coefficients determined for the dense-bed column developed for this investigation lie between those found for spiral conveyor columns and wash columns. Since the crystals are highly compacted in the dense-bed column, axial mixing is much less than other type columns, even though liquid fluctuations caused by pulsing of the overhead piston contribute to axial mixing. Thus, operation of a dense-bed, center-feed crystallizer is feasible and causes less axial mixing than dilute slurry crystallizers with oscillating conveyors.

DISCUSSION

The nonadiabatic model developed for this study can easily be used to rate or design column crystallizers if certain parameters are available. The key scale-up parameter that requires resolution is the axial dispersion coefficient which includes effects related to the amplitude and frequency of pulsing and to the size, shape, and packing of particles in the bed. In principle, the approximate magnitude of the dispersion coefficient can be determined from estimated particle size and published correlations of Peclet vs. particle Reynolds number; however, prediction of crystal characteristics in densely packed crystallizer columns is not reliable at present, and the actual degree of axial dispersion in a pulsed column is best determined from a combined analytical and experimental approach as outlined in this study.

ACKNOWLEDGMENT

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NOTATION

C_p = specific heat/unit mass
D_{ax} = axial dispersion coefficient
d = column diameter
H = enthalpy per unit mass
$\underset{\sim}{L}$ = mass flux of liquid with respect to stationary coordinates
$\underset{\sim}{S}$ = mass flux of solid phase with respect to stationary coordinates
T = absolute temperature
U = overall heat transfer coefficient
x = mass fraction transferring component in solid phase
y = mass fraction transferring component in liquid phase
z = column position, positive from bottom

$A, B, A^*—D^*, A'—D', a'—d'$ = locally defined coefficients

Greek Letters

ϕ = void fraction
μ = viscosity
ρ = density
λ = heat of melting per unit mass

Superscripts

L refers to liquid phase
W refers to column wall

Subscripts

S = solid phase
L = liquid phase
f refers to feed point

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On the Optimization of Distributed Parameter Systems with Boundary Control: A Counter Example for the Maximum Principle

A counterexample is given to the strong maximum principle for boundary control of a class of distributed parameter systems. The particular system deals with chemical reactors suffering catalyst decay and is in the class whose members are described by sets of first-order partial differential equations of the hyperbolic type. It is shown that an optimal control exists and that over any finite time interval in which the control is unconstrained the exit conversion from the reactor remains constant. It is further shown that for certain values of the parameters the optimal control policy violates the necessary conditions of the strong maximum principle for boundary control of distributed parameter systems.

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SCOPE

The maximum principle of Pontryagin (1962) has been used extensively in the optimal control of lumped parameter systems described by ordinary differential equations. A similar strong maximum principle was developed by Degtyarev and Sirazetdinov (1967) for the optimal control of distributed parameter systems described by a set of first-order partial differential equations and where the control was distributed. For the case of boundary control, where the control enters in the boundary conditions to the partial differential equations or where the control is a function of one independent variable only, Degtyarev and Sirazetdinov obtained a weaker form of the necessary conditions. Whereas in the case of distributed control, the hamiltonian must reach a maximum with respect to the control at the optimum, the boundary hamiltonian, in the case of boundary control, need only remain stationary with respect to the control at the optimum whenever the control is unconstrained. However, several authors have also stated the strong maximum principle as a necessary condition for optimality in the boundary control case.

In the present study, the validity of this strong maximum principle will be examined for a certain class of boundary control problems. The particular example under investigation relates to the optimal inlet temperature control of a tubular fixed-bed chemical reactor with slowly decaying catalyst.