

# Experimental and Theoretical Investigation of Purification in a Column Crystallizer of Material with Impurities of the Eutectic Forming Type

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A theoretical analysis of the operation of a Schildknecht type of column crystallizer is presented and simplified to three special cases. Experimental determinations were made of concentration profiles established during the purification of benzene with cyclohexane as impurity under steady state, batch conditions of operation. Benzene containing 34 to 500 ppm. cyclohexane was obtained by processing feed material with 5,000 to 30,000 ppm. cyclohexane. It was found that experimental data obtained under conditions of normal operation are well correlated by theoretical expressions if it is assumed that the crystals are not pure but contain cyclohexane at the level of 34 to 100 ppm. It was further established that the resistance to mass transfer between the liquid which adheres to the crystals and the wash liquid is negligible in comparison to backmixing effects as represented in the model by an eddy diffusion term. Smooth operation of the column was obtained at crystal rates below a limiting upper value and at oscillation frequencies above a lower limit.

Fractional solidification (32) provides a means of purifying a variety of materials for the production of primary standards, pharmaceuticals, monomers, and semiconductors. Column crystallization (2, 16, 21) provides a convenient means of carrying out fractional solidification and providing countercurrent contacting of crystalline material with reflux liquid formed from the crystal melt. Column crystallization has been used to process a variety of materials (10, 12, 30) including systems forming solid solutions (16, 20, 24) as well as materials of the eutectic forming type (10, 16, 23). Products frequently are in excess of 98% purity (8, 10, 29, 30) and materials containing impurities of the order of 100 ppm. and less have been obtained (16, 25). The process is suitable for the laboratory (21, 22, 23) and for large industrial applications (9, 17, 18, 29). Separations may be carried out on a batch basis (16, 23, 24, 31) or with continuous feed and product removal (18, 25).

Since the conception of the process by Arnold (5) in 1951, effort in developing the process has been directed primarily toward improvements in operating and controlling the equipment and the process in order to increase the purity and the yield of the desired product. Theoretical analysis of the operation of column crystallizers has been attempted by Powers (20) and others (3, 4, 31). However, tests of the mathematical models have been very limited in scope and have never been carried out in a systematic manner in order to establish the dominant mechanism which determines the extent of separation and purification that is achieved.

Therefore the purpose of this contribution is to present an extension of previous theoretical analyses and describe the results of laboratory experiments which establish the dominant mechanisms of purification in a column crystallizer of materials with negligible or slight solid solubility.

## PROCESS DESCRIPTION

Column crystallizers of the Schildknecht type have been described in the literature (2, 21) but a brief discussion of the process is presented here for the convenience of

those who are not acquainted with the subject and in anticipation of the development of a mathematical model. A schematic diagram of a column crystallizer is presented as Figure 1. The column is formed by two concentric tubes with a spiral in the annular space. The column consists of freezing, purification and melting sections.

The primary separation of the components is accomplished by fractional solidification of material in the freezing section of the column. Although such solidification may theoretically produce crystals of high purity, many factors such as slight solid solubility, adsorption and occlusion or adherence of the mother liquor tend to yield an effective solid phase (crystals plus adhering liquid) with relatively high impurity content.

As the effective solid phase is augered up the column out of the freezing section and through the purification section, it is contacted with melt in a countercurrent fashion. In this manner the impure adhering mother liquor

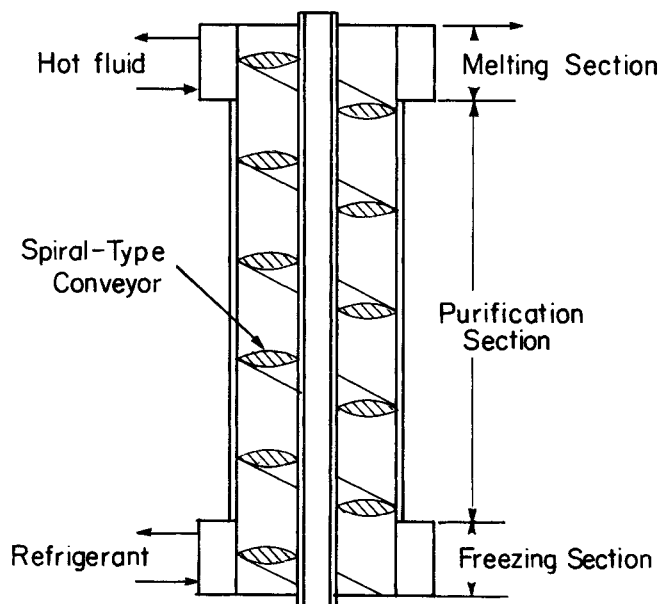


Fig. 1. Schildknecht type of column crystallizer operated at total reflux.

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is washed from the crystals. The wash fluid is produced in the melting section as a result of melting of the relatively pure crystals.

## THEORY

The qualitative description presented in the previous section provides the basis for mathematical analysis of the process. The developments outlined below are similar to those previously presented by Powers (20) except that incorporation of the effects of backmixing (eddy diffusion) is accomplished in a more rigorous manner and the possible presence of impurity in the crystal phase is taken into account.

In developing a mathematical model it is reasoned that the ultimate degree of purification obtained is determined by the processes which occur in the purification section. Therefore equations are developed to describe this section only.

In the mathematical analysis the effective solid phase is considered as a discontinuous phase passing counter-currently to a continuous phase, the reflux liquid. This is illustrated in Figure 2a in which the presence of the solid spiral is ignored. The physical situation is further simplified by considering the flow of the reflux liquid and effective solid streams to be uniform and unidirectional as illustrated in Figure 2b.

A mass balance on the impurity is made for a system consisting of the reflux liquid which occupies a differential length of the column. For purposes of simplification it is considered that the rate of mass transfer between liquid streams per unit interfacial area,  $M_t$ , is proportional to the difference in impurity content of the adhering ( $Y_a$ ) and reflux ( $Y_r$ ) liquid.

$$M_t = \rho K(Y_a - Y_r) \quad (1)$$

where  $K$  is the overall liquid phase mass transfer coefficient and  $\rho$  is the density of the liquid phase. In addition it is assumed that impurity transfer by eddy diffusion in the reflux liquid,  $M_d$ , is proportional to the gradient of impurity content in that stream.

$$M_d = -\rho D \frac{dY_r}{dz} \quad (2)$$

where  $D$  is the effective coefficient of eddy diffusion and  $z$  is the location in the purification section. The resulting differential equation is

$$A \frac{d\left(D\rho\epsilon \frac{dY_r}{dz}\right)}{dz} + \frac{d(Y_r V_r)}{dz} + KaA\epsilon\rho(Y_a - Y_r) = 0 \quad (3)$$

where  $A$  is the cross-sectional area of the column available for mass transport,  $\epsilon$  is the volume fraction of liquid, and  $a$  is the area for mass transfer from the adhering liquid to the reflux liquid per unit volume of total liquid in the annular space.

Another independent differential equation is obtained from an impurity balance around one end of the purification section under conditions of steady state and total reflux.

$$D\rho\epsilon A \frac{dY_r}{dz} + Y_r V_r - Y_a V_a - XL = 0 \quad (4)$$

where  $L$  is the crystal rate and  $X$  is the weight fraction impurity in the crystals.

A total mass balance on the same system reveals that the sum of the crystal and the adhering liquid rates is equal to the reflux liquid rate as given by Equation (5).

$$V_r = V_a + L \quad (5)$$

In order to simplify the mathematical analysis of the

operation of the column crystallizer it is assumed that  $V_r$ ,  $V_a$ ,  $L$ ,  $D$ ,  $\epsilon$  and  $Ka$  are constant throughout the length of the purification section. Subject to these assumptions, Equations (3) to (5) are combined to yield Equation (6) for the impurity distribution in the purification section of the column.

$$\alpha \frac{d^2 Y_r}{dz^2} + \beta \frac{dY_r}{dz} + Y_r - X = 0 \quad (6)^*$$

where

$$\alpha = RD/Ka \quad (7)$$

$$\beta = \frac{R(R+1)L}{Ka\rho A\epsilon} + \frac{D\rho A\epsilon}{L} \quad (8)$$

$$R = V_a/L \quad (9)$$

The general solution to Equation (6) is obtained by standard techniques for solving ordinary differential equations for the case in which  $X \neq f(z)$ :

$$Y_r(z) = C_1 \exp(s_1 z) + C_2 \exp(s_2 z) + X \quad (10)$$

where

$$s_1, s_2 = \frac{-\beta \pm \sqrt{\beta^2 - 4\alpha}}{2\alpha} \quad (11)$$

and  $C_1$  and  $C_2$  are constants to be evaluated from boundary conditions.

In order to simplify the comparison of theoretical developments with experimental results three special cases were considered. If it is assumed that the crystals are indeed pure (that is  $X = 0$ ), Equation (10) becomes

$$Y_r(z) = C_3 \exp(s_1 z) + C_4 \exp(s_2 z) \quad (12)$$

On the other hand, if it is assumed that the crystal has a constant impurity content,  $X$ , but that eddy diffusion has a negligible influence on the concentration profile in comparison with mass transfer between the adhering and reflux liquid, the general case reduces to

$$Y_r(z) = C_5 \exp(-\gamma z) + X \quad (13)$$

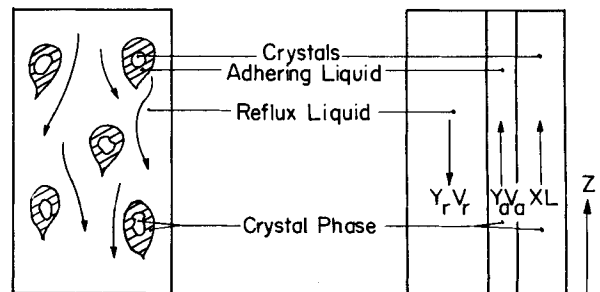
where

$$\gamma = \frac{\rho Ka A \epsilon}{R(R+1)L} \quad (14)$$

Finally, if  $X \neq 0$  and the resistance to mass transfer between the reflux liquid and the adhering liquid has negligible influence on the concentration profile in comparison with eddy diffusion (backmixing), the following expression is obtained:

$$Y_r(z) = C_6 \exp(-\phi z) + X \quad (15)$$

where



A. Physical Situation

B. Simplification of the Physical Situation

Fig. 2. Schematic of solid phase and the reflux liquid in counter-current contact in the purification section.

\* In the previous analysis of column crystallization presented by Powers (20), it was assumed that the crystals were indeed pure ( $X = 0$ ) and the second order differential term was avoided by applying the transport equation approach originally suggested by Furry, Jones, and Onsager (11) in connection with their analysis of thermogravitational thermal diffusion columns.

$$\phi = \frac{L}{D\rho A\epsilon} \quad (16)$$

If it can be established that  $X \neq 0$ , Equations (13) and (15) provide a good basis for determining whether eddy diffusion in the liquid phase or resistance to mass transfer between the reflux liquid and the adhering liquid plays a dominant role in establishing the concentration profile in a column crystallizer. As a first approximation it can be assumed that the mass transfer coefficient group,  $Ka$ , and the coefficient of eddy diffusivity are relatively independent of crystal rate,  $L$ . (This is perhaps reasonable in a column crystallizer in which hydrodynamic factors are primarily determined by action of the rotating and oscillating spiral.) In such a case, Equations (13) and (14) predict that increasing the crystal rate will result in a decrease of the separation if resistance to mass transfer between adhering and reflux liquid is dominant. In direct contrast, if eddy diffusion is the primary factor in determining the distribution of impurity, it follows from Equations (15) and (16) that an increase in crystal rate should result in an increase in separation.

## EXPERIMENTAL APPARATUS AND PROCEDURE

### Equipment

A column crystallizer similar to the type developed by Schildknecht (21, 23) was used in this investigation. A schematic of the column illustrating sample tap location, etc., has been presented elsewhere (1). The column was an annulus defined by a 2.6 cm. I.D. glass tube on the outside and a 1.1 cm. O.D. stainless steel tube on the inside. In the annulus a stainless steel spiral, lenticular in cross section (27) was rotated so as to convey the crystals from the freezing section of the column, through the purification section, to the melting section. The lengths of the freezing section, the purification section, and the melting section were 8, 48, and 5 cm., respectively. The freezing section of the column was at the

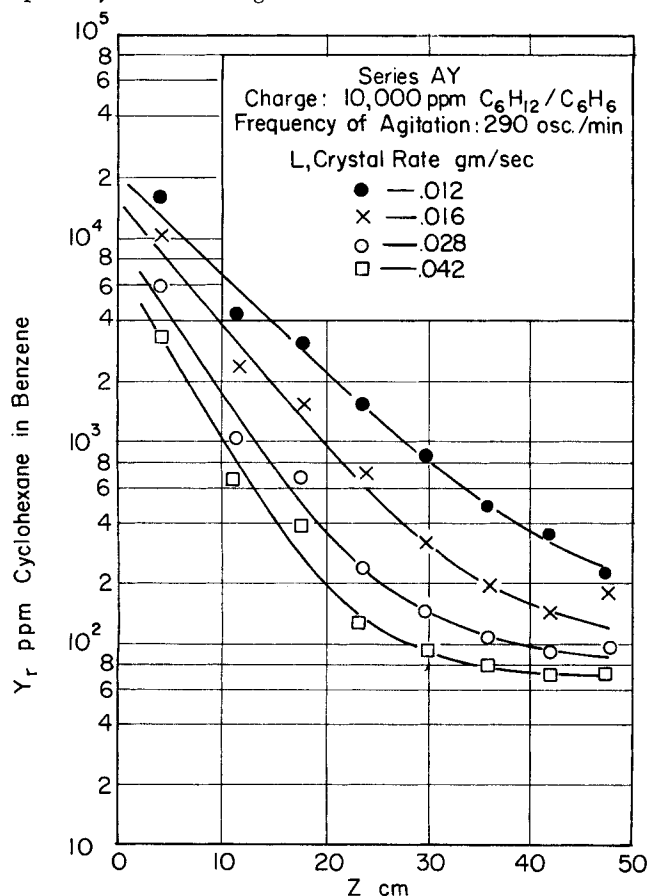


Fig. 3. Impurity content in the reflux liquid as a function of position in the purification section.

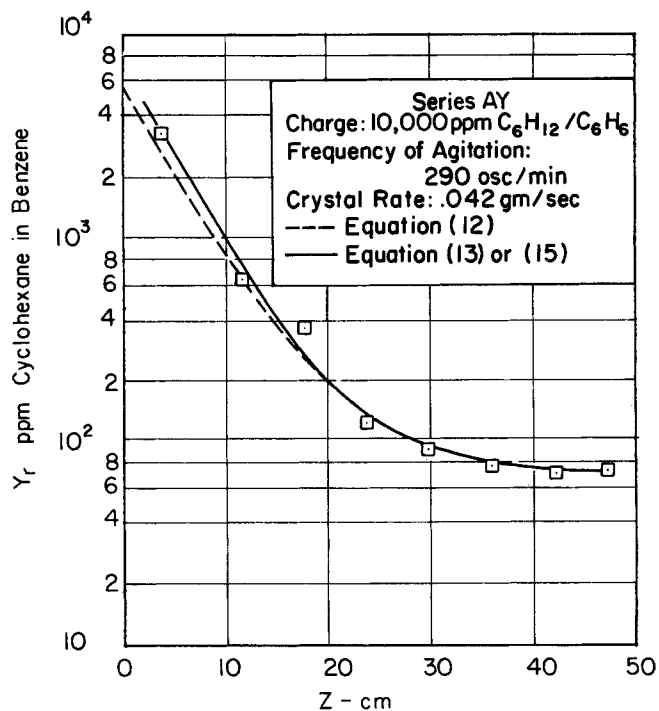


Fig. 4. Correlation of experimental concentration profile with several semi-theoretical equations.

lower end of the annulus and was enclosed in a concentric jacket through which a refrigerant was circulated. The energy necessary to melt the crystals was supplied by an electrical resistance heater located in the top of the column, that is in the melting section.

The spiral in the annulus was used to convey the crystals through the column as well as agitate the crystal mass. For this purpose both rotary and oscillatory motions were imparted to the spiral by a specially designed mechanism. Both the rate of rotation and the rate of oscillation could be varied independently from 0 to 440 cycles/min.

In order to minimize changes in the crystal rate through the purification section due to heat transfer with the surroundings, the purification section was insulated on three sides by Styrofoam slabs 1 in. thick. The entire column was also placed in a constant temperature air bath maintained at 5.5°C., the melting point of pure benzene and the approximate temperature of the purification section of the column.

### Procedure

The column was cleaned and rinsed with feed mixture. A liquid charge of 260 cc. was placed in the column. The rotation rate and amplitude of oscillation of the spiral were set at 59 rev./min. and 1 mm., values which yielded smooth operation. Circulation of coolant through the jacket of the freezing section was initiated at temperatures equal to or slightly below the freezing temperature of the feed mixture. The coolant temperature was decreased at a rate of 6 to 20°C./hr. until the desired crystal rate was established. If this procedure was not followed operational difficulties were often encountered; if the coolant temperature was initially established at the value required for steady state operation, the initial freezing rate would be too great and the column would plug. If a higher initial coolant temperature were used and maintained at a constant value, the crystallization rate would decrease to zero as the concentration of impurities built up in the freezing section and lowered the freezing point of the mixture.

The crystals served to cool the entire contents of the column as they were transported toward the melting section during startup. Heat was supplied to the melting section to maintain constant crystal level in the melting section of the column. Under steady state conditions, the power input to the heater was measured and was used to calculate the flow rate of crystals through the purification section (1). Once the crystal stream was established throughout the length of the column and if the crystal flux was less than 0.13 g./sq. cm.-sec., then there was no danger of spontaneous plugging of the column. That is, the operation of the column was stable and reliable.

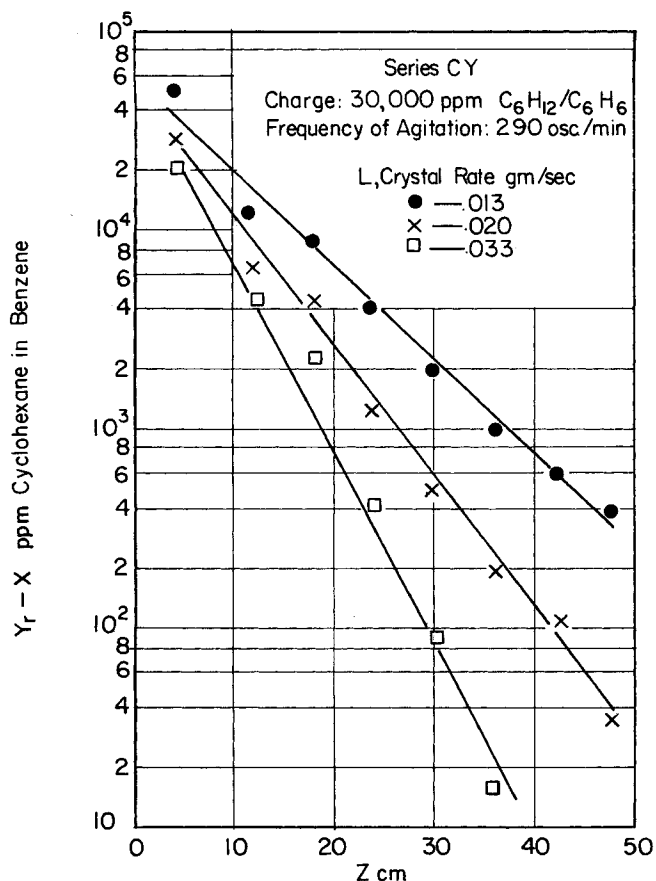


Fig. 5. Modified plot of the impurity content of reflux liquid as a function of position in the purification section of the column.

The mathematical analysis of the operation of the column crystallizer was restricted to steady state, batch-by-batch operation and therefore it was essential to develop a reliable criteria for determining when the column had come to steady state. For this purpose, samples of the reflux liquid were taken as a function of time at one location in the purification section of the column and analyzed for impurity content. The column was operated until the impurity content in the reflux liquid at the particular sample tap no longer changed with time. In general, the time required for the column to reach steady state ranged from 3 to 7 hr. after startup. The longer times were required at lower crystal rates.

After establishment of steady state conditions in the column, samples were taken from each of eight samples taps and analyzed using an F and M Research Gas Chromatograph Model 5750 with flame ionization detector. With calibration, the resolution was about 1 ppm. A temperature profile was also determined.

#### Reagents

The major impurity in the benzene used in this investigation was found to be cyclohexane and therefore no further purification of feed material was made [except in preparing primary standards for calibration of the chromatograph (1)]. Reagent grade cyclohexane was used as the impurity in the charge to the column. No purification of this material was attempted as it was the minor component. Charges were made up in lots of 3,000 or 6,000 cc. and were analyzed by chromatograph to insure that the composition was within 5% of the nominal 5,000, 10,000 and 30,000 ppm. cyclohexane in benzene feed mixtures.

#### Results

Typical results are presented as Figures 3, 4, and 5. Data for a total of 30 composition profiles determined at steady state under a variety of operating conditions were obtained (1).

#### ANALYSIS OF RESULTS

The experimental results presented in Figure 3 can be well represented by any of the three sets of equations re-

sulting from simplification of the general analysis [Equation (12), Equations (13) and (14), Equations (15) and (16)]. For example the dashed line on Figure 4 is of the form of Equation (12);

$$Y_r(z) = 0.0057 \exp(-0.193z) + 0.000087 \exp(-0.00442z)$$

By applying Equations (7) through (9) and (11) together with an experimentally determined value of void fraction  $\epsilon$ , and an estimate of the interfacial area term,  $a$ , (1) it is possible to calculate values of the eddy diffusivity,  $D$ , and liquid phase mass transfer coefficient,  $K$ . The results are

$$D = 3.5 \text{ sq.cm./sec.}$$

$$Ka = 7.9 \times 10^{-6} \text{ sec.}^{-1}$$

$$K = 1.9 \times 10^{-7} \text{ cm./sec.}$$

The calculated value for  $D$  is found to be in the range of those measured by Jones (13) and Smoot and Babb (26) in pulsed liquid extraction columns operated in a manner similar to the column crystallizer. However,  $K$  is found to be four orders of magnitude smaller than those reported in the literature (6, 14, 15, 28). In view of this, it is concluded that this model is not compatible with the physical phenomena.

As the result of these calculations it was decided that  $X \neq 0$ . The solid line on Figure 4 resulting from the application of either Equation (13) or Equation (15) correlates data from Figure 3 well when  $X = 70$  ppm. The choice for this value for  $X$  is not arbitrary. It is justified by the fact that for sufficiently high values of  $z$  the impurity content in the reflux liquid is essentially constant at 70 ppm. Hence according to Equation (6) it follows that in this case  $X = 70$  ppm.

For  $X \neq 0$ , either Equation (13) or Equation (14) suggests that a plot of  $\log(Y_r - X)$  vs.  $Z$  should yield a straight line. For example, a set of data for which  $X = 100$  ppm. were fit to a straight line using the technique of least squares. The resulting agreement is illustrated on Figure 5. Note that the same value of  $X = 100$  ppm. was used in obtaining straight line correlations from three different runs. [Similar plots were prepared using data from Figure 3 and other runs (1).]

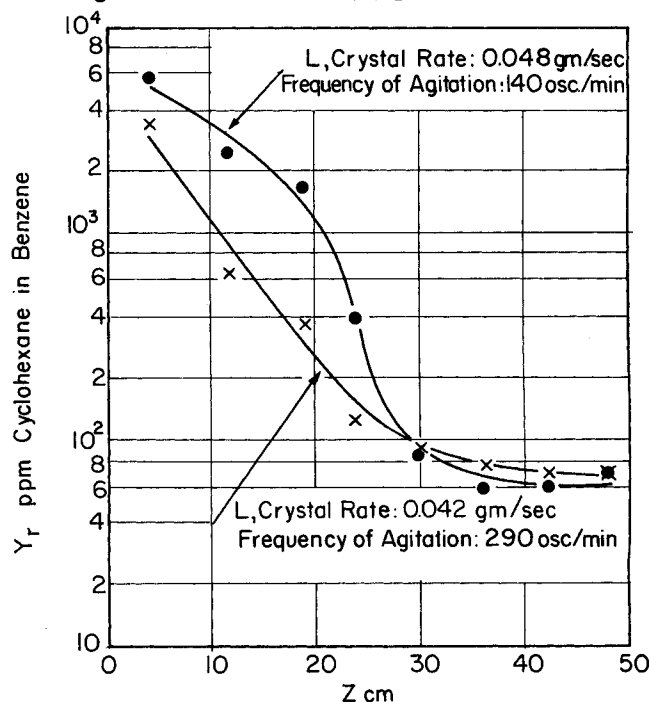


Fig. 6. The effect of the intensity of agitation on the impurity distribution in the reflux liquid in the purification section at a high crystal rate.

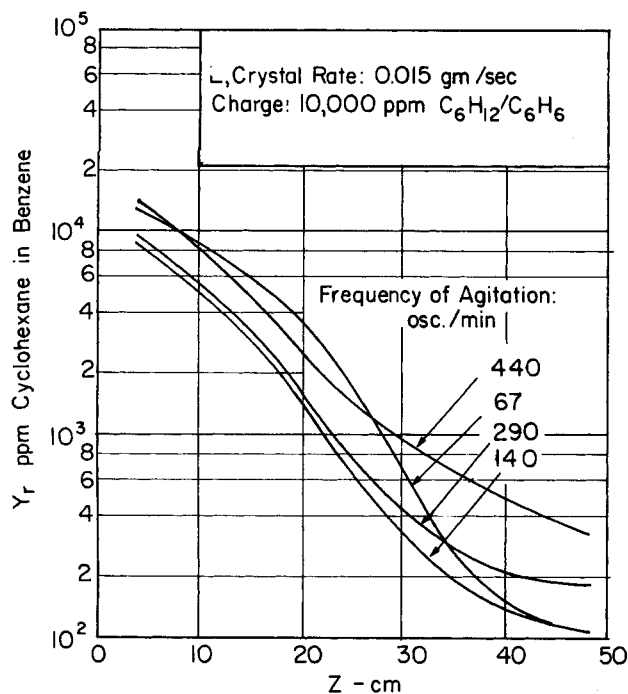


Fig. 7. The effect of the intensity of agitation of the impurity distribution in the reflux liquid in the purification section at a low crystal rate.

In formulating the mathematical model it was assumed that  $X \neq f(z)$ , that is the resistance to mass transfer within the crystals themselves is so high that the crystals reaching the top of the column are those that were formed in the freezing section and have passed through the purification section without change in composition. Experiments were carried out in an attempt to determine whether or not the empirically determined crystal impurity content,  $X$ , might vary between the freezing and melting section. It was reasoned that all things being equal, the degree to which the impurity within the crystals is transferred to the reflux liquid is a function of the residence time of the crystals in the purification section. The longer the residence time, the more nearly pure crystals are expected. As shown in Table 1, decreasing the residence time by a factor of more than two by increasing the crystal rate did not result in a significant change in the purity of the material obtained at the top of the column. Therefore it is concluded that the crystals apparently contain a small amount of impurity which remains constant throughout the length of the purification section for a given charge composition and a given intensity of agitation.

TABLE 1. EFFECT OF INCREASING THE CRYSTAL RATE ON THE PURITY OF THE MATERIAL OBTAINED AT THE TOP OF PURIFICATION SECTION

Crystal Rate g./sec.	Distance from Freezing Section	
	48 cm.	42.5 cm.
0.033	98	100
0.044	101	107
0.065	106	108
0.072	103	100

As pointed out at the end of the theoretical development, operation of a column at different crystal rates provides a means of establishing the dominant mechanism for mass transfer. Therefore, as illustrated on Figures 3 and 5, several series of runs were made in which the only variable was the crystal rate. In all cases for which the theoretical model is valid (straight lines are obtained as

in Figure 5) an increase in crystal rate resulted in an increase in separation, that is the total impurity content in the purification section was decreased (see Figure 3 and 5). Therefore it is concluded that the dominant mechanism in establishing the concentration profile is backmixing (eddy diffusivity); and therefore resistance to mass transfer between the reflux liquid and the adhering liquid is of relatively little importance.

Not all experimental profiles could be fitted as illustrated in Figure 5. One such example is presented in Figure 6. The deviation is the result of mechanical factors. At the higher crystal rate and lower level of agitation the crystals are not uniformly dispersed in the liquid and poor contacting results.

The influence of agitation frequency on separation is not easy to anticipate *a priori* nor to treat theoretically. As it was established that eddy diffusion severely limits the separation, one would predict on this basis alone that decreasing the agitation frequency would result in an increase in separation. The data presented in Figure 7 demonstrates that this prediction is valid at frequencies between 440 and 140 oscillations/min. Further decrease in frequency to 67 oscillations/min. results in a decrease in separation as a result of mechanical segregation of phases as discussed previously.

Comparison of Figures 6 and 7 reveals that the level of frequency at which a further decrease in frequency results in a decrease in separation depends on the crystal rate.

Results from all experiments with linear plots of  $(Y_r - X)$  vs.  $z$  on semilogarithmic coordinates yielded values of  $\phi = (L/\rho DA\epsilon)$  as determined from the slope. This term is very closely associated with the Peclet number,  $N_{Pe}$ , for packed beds.

$$N_{Pe} = \frac{L}{\rho DA\epsilon} d_p = \phi d_p \quad (17)$$

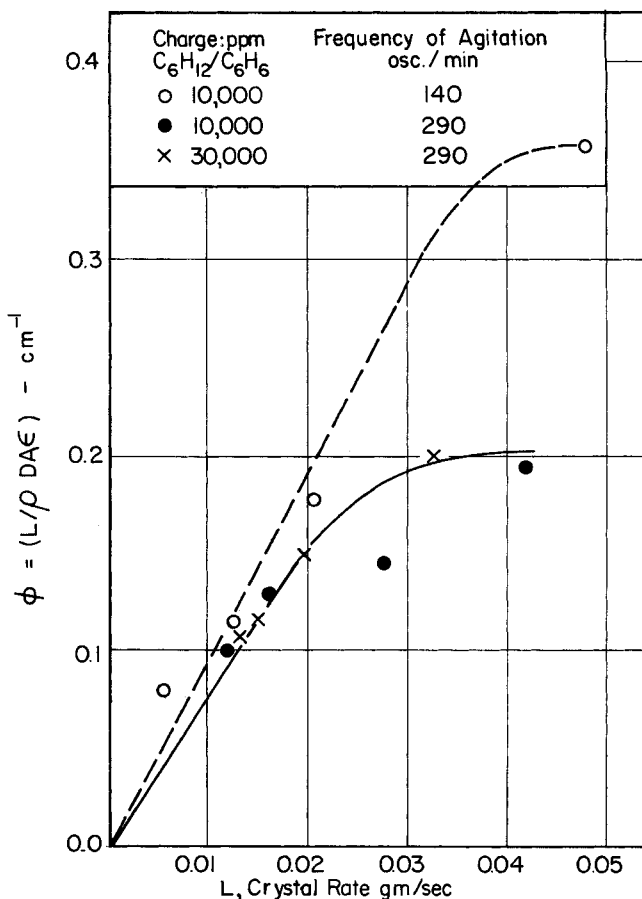


Fig. 8. Mass transfer factor as a function of the crystal rate.

where  $d_p$  is the mean particle diameter. In a similar manner, the crystal rate,  $L$ , is linearly related to the Reynolds number,  $N_{Re}$

$$N_{Re} = \frac{L}{A\epsilon\mu} d_p \quad (18)$$

where  $\mu$  is the fluid viscosity. Therefore a plot was made of  $\phi$  vs.  $L$  in a manner analogous to the treatment of data on mass transfer in packed beds (7, 19). The results are presented as Figure 8.

The intensity of agitation is the common parameter in the family of curves so obtained. (Data obtained with feed of different composition are represented as one curve.) From the results obtained at 290 oscillations/min. it is seen that as the crystal rate is increased,  $\phi$  increases to approach a constant value for a given level of agitation. (A dashed line of similar shape was drawn through the points for 140 oscillations/min.) These graphical relations are in qualitative agreement with the Peclet number-Reynolds number relationship for packed beds (7, 19). This functional agreement is further endorsement that the controlling mechanisms of mass transfer are bulk flow and eddy diffusion.

Note from Figure 8 that over a considerable range of crystal rate,  $L$ , the mass transfer factor,  $\phi$ , is a linear function of  $L$  and, to a first approximation, passes through the origin. Therefore, it follows from the definition of  $\phi$  that the coefficient of eddy diffusivity,  $D$ , is independent of crystal rate,  $L$ , as previously assumed. The development applied here assumed  $Ka$  to be negligible and therefore it is not possible to check the assumption that this grouping is likewise independent of  $L$ .

The effect of the charge composition on  $\phi$  is also illustrated in Figure 8. Mass transfer factors for both the 10,000 ppm. and the 30,000 ppm. charges at 290 oscillations/min. are shown as a single function of the crystal rate. It is noted that  $\phi$  is independent of the charge composition as required in simple eddy diffusion type of mass transfer postulated in the mathematical model.

## SUMMARY AND CONCLUSIONS

A theoretical and experimental investigation has revealed that most of the experimental results can be well correlated with a mathematical model which assumes that the crystals contain a small amount ( $\approx 100$  ppm.) of impurity and that eddy diffusion (back-mixing) plays the dominant role in determining the composition profile under batch, steady state conditions. Mechanical factors such as plugging and poor dispersion of the solid phase cause reduced separations and yield concentration profiles which are not in agreement with theoretical predictions.

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## NOTATION

$A$  = cross-sectional area of the column available for mass transport, sq.cm.  
 $a$  = area for mass transfer from the adhering liquid to the reflux liquid per unit volume of total liquid, sq.cm./cc.  
 $C_{1-6}$  = constants of integration  
 $D$  = effective coefficient of eddy diffusion, sq.cm./sec.

$d_p$  = mean crystal diameter, cm.  
 $K$  = liquid phase mass transfer coefficient, cm./sec.  
 $L$  = crystal stream rate, g./sec.  
 $M$  = rate of mass transfer per unit area, g./sq.cm.-sec.  
 $N_{Pe}$  = Peclet number, Equation (17)  
 $N_{Re}$  = Reynolds number, Equation (18)  
 $R$  = constant defined by Equation (9)  
 $s_{1,2}$  = defined by Equation (11)  
 $V$  = liquid stream rate, g./sec.  
 $X$  = weight fraction impurity in the solid phase  
 $Y$  = weight fraction of the impurity in the liquid phase  
 $z$  = location in the purification section, cm.

## Greek Symbols

$\alpha$  = constant defined by Equation (7)  
 $\beta$  = constant defined by Equation (8)  
 $\gamma$  = constant defined by Equation (14)  
 $\epsilon$  = volume fraction liquid  
 $\mu$  = viscosity of the liquid phase, g./cm.-sec.  
 $\rho$  = density of the liquid phase, g./cc.  
 $\phi$  = constant defined by Equation (16)

## Subscripts

$a$  = adhering liquid  
 $d$  = mass transfer by eddy diffusion as the result of a concentration gradient  
 $r$  = the reflux liquid  
 $t$  = mass transfer between countercurrently passing streams as the result of a concentration difference

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# Drying Polymers During Screw Extrusion

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Partial removal of dissolved or absorbed solvent from polymers can be accomplished during the process of screw extrusion. This paper shows how two simple transport models based either on an effective diffusivity or on an empirical mass transfer coefficient can be combined with the fluid mechanical equations which describe polymer flow during screw extrusion. In this way, the drying of solvent from a polymer during extrusion can be correlated in terms of the design and operating parameters of the extruder screw and a mass-transfer coefficient or an effective diffusivity. This approach is illustrated employing data obtained by using two extruders of different size and two different solvent-polymer systems. Furthermore, it is pointed out how the results can be used to predict the extent of drying during screw extrusion as well as how modifications in extruder-screw design can permit more extensive drying of polymers during the extrusion process.

During polymer manufacture it is often necessary to remove dissolved, volatile hydrocarbon impurities from the product. Frequently, this is carried out by drying with steam or hot gas. As an example, conventional processes for polypropylene manufacture may involve several stages of drying to remove dissolved solvents such as hexane, xylene, etc., from the product.

The work described here demonstrates that it is also possible to remove some dissolved volatile material from a polymer as it undergoes screw extrusion, a final step in preparing a plastic for packaging. Thus this extrusion, aside from its conventional role, can also serve to replace part of the usual drying operation thereby decreasing the requirement for ordinary drying equipment. This study develops a design method for predicting how much drying a given extruder can provide for a given solvent-polymer system. Alternatively, the results indicate how an extruder can be modified to provide additional drying capacity.

The approach begins by focusing attention on the fluid mechanics of polymer flow within a screw extruder. This provides knowledge of the geometrical configuration and residence time of the drying polymer in the extruder. With these results it is possible to formulate drying either in terms of an empirical mass transfer coefficient or in terms of an effective diffusivity and thereby arrive at equations which predict the extent of drying. These equations involve mechanical design parameters of the extruder as well as an effective diffusivity of solvent in polymer in the case of a diffusional model, or a mass-transfer coefficient in the

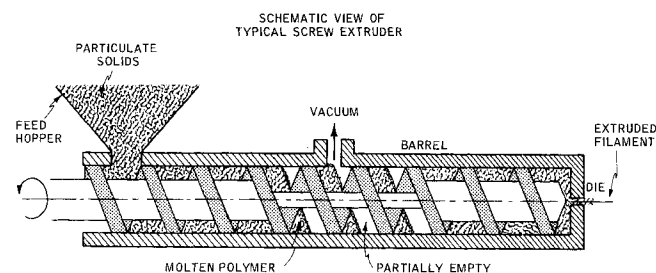


Fig. 1. Schematic view of typical screw extruder.

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Figure 2  
"UNWRAPPED" SCREW CHANNEL

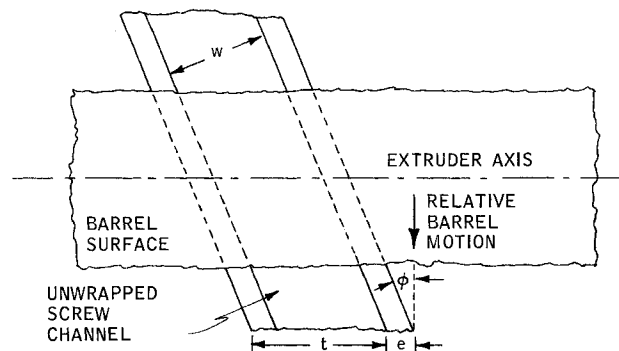


Fig. 2. "Unwrapped" screw channel.

case of the more empirical model.

## FLOW IN SCREW EXTRUDER

A conventional description of a screw extruder is that of a screw turning within a concentric cylinder. The polymer as a particulate solid is fed into a hopper at the low-pressure end; at the other end it passes through a die in the plastic state under high pressure. Heat generated by the shearing action within the extruder melts the polymer (heat can also be supplied by external sources). The relative motion of screw and barrel provides conveying action toward the die. Figure 1 depicts a typical screw extruder.

The fluid dynamical theory of isothermal screw extrusion has been presented in detail in many places (1 to 9). For the purpose at hand, only a cursory description and the results of the theory will be given.

The conveying action of an extruder is easily understood by considering the helical screw channel as unwrapped and laid flat as illustrated in Figure 2. With this convention, the barrel becomes a flat plate which moves over the screw channel as noted. The component of barrel motion parallel to the screw channel imparts a drag force to the molten polymer which wets the barrel. Thus, the fluid is conveyed towards the die against a pressure which is a maximum at the die. The net flow is conveniently and