Effect of Sludge Funneling in Gravity

Thickeners

The performances of cylindrical and conical thickeners are compared by means of calculations, using published data on the properties of a suspension. Experimental data on the operation of cylindrical thickeners indicate that sludge funneling at the bottom of such thickeners has a large effect and is a possible cause of observed discrepancies between theory and experiment.

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SCOPE

The analysis of gravity sedimentation and thickening processes has long been based on the assumption that the particle settling rate is a function of the concentration [that is, u=u(c)], this function being a characteristic of the slurry. This implies that all particle submerged weight is supported by liquid drag, whereas some interparticle (mechanical) support, transmitted from the bottom of the container, would be expected during the compression (thickening) of the sediment. In fact, in a continuous thickener at steady state, the operating conditions dictate a relation between u and c, which inevitably conflicts with the

assumption that another such relation is established by slurry characteristics. However, no experimental investigation yet reported has detected any appreciable mechanical support of particles, a result which is difficult to reconcile with theory.

In analyzing the thickening process, it is usually assumed that vertical plug flow exists in the thickener, whereas extensive funneling of the sludge as it approaches the offtake pipe has been observed. This effect is perhaps the cause of the above discrepancy between theory and experiment, and this paper reports a study of the effect of accounting for sludge funneling in thickener analysis.

CONCLUSIONS AND SIGNIFICANCE

The analytical results show that sludge funneling can have a large effect on thickener performance and is a possible explanation for the observed lack of mechanical support of particles. This indicates the need for further investigation into thickener design methods.

When gravity thickeners are analyzed, it is usually assumed that plug flow exists, that is, horizontal uniformity of all properties at each level. This assumption is applied to the analysis of the thickening zone, in which solid particles and liquid both move downward. In the clarification zone, above the thickening zone, the underflow stream (most of the particles and a small part of the liquid) separates from the overflow stream, and the flow is essentially two dimensional. The particles separated from the overflow stream enter the thickening zone as they settle onto the top of the sediment. If we neglect interaction between the two zones due to scouring of particles from the top of the sediment, all flow is downward in the thickening (concentration increase) zone.

The assumption of plug flow in the thickening zone is made so as to simplify the analysis, but it clearly introduces a degree of error. At the top of the sediment, the particles arriving are probably not distributed uniformly over the cross section, and at the bottom there is certainly some degree of funneling or ratholing of the sludge stream as it contracts toward the exit pipe. Rakes are normally used to assist the movement of sludge toward the offtake, and it is usually assumed that, as a result, sludge funneling only occurs over a small depth at the bottom of the thickener.

Few data on the effectiveness of rakes in promotiong radial sludge movement appear in the literature. However, Turner and Glasser (1976) found that rakes had little effect in this respect in a pilot scale thickener (2 m diameter) and observed funneling over most of the sludge depth. The purpose of this

article is to show by calculation that funneling can have a pronounced effect on thickener operation and that it provides a possible explanation for observed large deviations of cylindrical thickener performance from theory.

CONICAL AND CYLINDRICAL THICKENER THEORY

The theory of sludge thickening in conical and cylindrical vessels is considered first. The difference between the two vessel shapes is, of course, that in a cylindrical thickener the cross-sectional area is the same at all levels, while in a conical thickener the cross-sectional area varies with height. In both cases, horizontal uniformity is assumed at each level.

It is usually assumed that the sludge concentration achieved depends on the interparticle compressive stress (or effective pressure). However, it is recognized that this is not precisely true; variations in particle packing arrangement can have a significant effect, and there is also a time delay in the compression process which is usually neglected (Dixon, 1978). The compressive stress at a given level is due to the submerged weight of the particles (that is, allowing for liquid upthrust) in the sludge above, and, other things being equal, the greater the depth of sludge in the thickener, the higher the underflow concentration. (In the following, weight of particles means submerged weight, and gravitational force means the net gravitational force on the particles after allowing for upthrust.)

However, the whole of the particle weight does not contribute to the bottom compressive stress. During passage downward, the particles move with a higher velocity than the liquid,

0001-1541-80-3777-0471-\$00.85. The American Institute of Chemical Engineers, 1980.

so that they are subject to an upward drag force which partly balances the gravitational force. Thus, the compressive stress acting at the bottom of the thickener depends on the weight of particles in the sludge per unit area and on the fraction of this weight which is supported by liquid drag.

If we assume, for both conical and cylindrical thickeners, horizontal uniformity at each level, and steady state operation, a force balance on a differentially deep layer of sludge yields, in the limit as the depth of the layer approaches zero (Dixon, 1977a,b)

$$\frac{d\sigma}{dx} = cg(\rho_p - \rho_l) - \frac{u}{K} \tag{1}$$

The first term on the right-hand side of this equation is the gravitational contribution to the rate of increase of compressive stress with depth, and the second term is the opposing contribution of the drag. As shown, the drag force is assumed to be proportional to the relative velocity u, which is Darcy's law. This is the expected drag relation in a thickener, because the flow Reynolds number is very low. For a given suspension, the permeability K, is usually assumed to depend on the particle concentration (decreasing as c increases), but it too is affected by particle packing arrangement.

In deriving Equation (1), inertial (acceleration) effects have been neglected. These effects are negligible when the sludge is in compression, which is the case presently being considered. In free settling, where there is little or no resistance to compression, inertial effects cannot always be neglected (Dixon,

Since the concentration achieved is assumed to depend on the compressive stress, there is a functional relation between σ and c, and Equation (1) can be rewritten as

$$\frac{dc}{dx} = \frac{cg(\rho_p - \rho_l) - u/K}{(d\sigma/dc)}$$
 (2)

where the compression coefficient $d\sigma/dc$ is a sludge characteristic which usually increases very rapidly as c increases.

To enable Equation (2) to be integrated to obtain the variation of c with x, a relation between u and c is needed. It is at this point that the analyses of conical and cylindrical thickeners diverge.

At steady state, in both cylindrical and conical thickeners, the volumetric flow rates of particles and liquid, and hence total sludge, are the same at all levels. However, the fluxes (or superficial velocities) are determined by dividing by the crosssectional area, which varies with depth in the conical case; that is

$$G_{\nu} = Q_{\nu}/A \tag{3}$$

$$G_{t} = Q_{t}/A \tag{4}$$

$$G_t = Q_t / A \tag{4}$$

The mean velocity of each phase is found by dividing the corresponding flux by the fraction of the cross-sectional area which is occupied by the phase. Thus, the mean particle velocity is given

$$v = G_{\nu}/c \tag{5}$$

and the mean velocity of the sludge is equal to G_t . The particle velocity varies with depth in both conical and cylindrical cases, since concentration varies with depth, but the fluxes only vary with depth in the conical case. The velocity of the particles relative to the mean velocity of the sludge is given by

$$u = v - G_t = G_p/c - G_t \tag{6}$$

In the case of a cylindrical thickener, G_p and G_t are the same at all levels, so that Equation (6) provides the necessary relation between u and c to allow Equation (2) to be integrated. [Separation of variables can be used as well as numerical integration (Perry and Chilton, 1973).] G_{ν} and G_{t} are determined by the thickener operating conditions. G_{ν} is determined by the feed rate of particles and the efficiency of clarification. (Ideally, all entering particles leave in the underflow stream.) G_t is determined by the volumetric sludge pumping rate.

In a conical thickener, Equations (2) and (6) are not sufficient, since G_{ν} and G_{t} are not constant owing to the cross-sectional area variation. From geometrical considerations

$$\frac{dA}{dx} = -2\sqrt{\pi} \tan(\theta/2)\sqrt{A} \tag{7}$$

(The minus sign is due to defining x as positive downward.) Equations (2) and (7) can be integrated numerically with respect to x, starting from the bottom of the thickener (where A and C are known, c being the desired underflow concentration), in conjunction with Equations (3), (4) and (6) and data on K and $d\sigma/dc$ as functions of c. [Separation of variables is not possible in this case, and one of the methods for numerical integration of ordinary differential equations must be used (Perry and Chilton, 1973). The writer uses the standard, fourth-order Runge-Kutta algorithm, with step size adjustment based on truncation error estimates (Ralston and Wilf, 1960).] The cylindrical case can be regarded as a special one for which $\tan (\theta/2) = 0$, so that dA/dx =0 for all x.

It is convenient to further rearrange Equation (2) to

$$\frac{dc}{dx} = \frac{cg(\rho_p - \rho_l)}{d\sigma/dc} \left(1 - \frac{u}{u_t}\right) \tag{8}$$

 u_t equals $cg(\rho_p - \rho_l)K$ and is the terminal settling velocity of the suspension, a function of concentration. It is the velocity with which the particles settle (relative to the average velocity) if they are falling freely in the absence of a compressive stress gradient. It is the relative velocity at which gravitational and drag forces are balanced at any given concentration and could also be called the no compression settling velocity.

During thickening the particles do not fall freely; part of the weight of each layer acts on the layer below, causing compression. Thus the particles fall at less than the terminal velocity, making dc/dx positive in accordance with Equation (8). The fact that u must be less than u_t places a limitation on the possible operating conditions of the thickener.

For the case of a cylindrical thickener, Equation (6) can be converted to a straight line relation by multiplying throughout by c. The settling flux S is defined to be cu and is the particle flux relative to the convective particle flux cG_t . Equation (6) then becomes the equation for the thickener operation line

$$S = G_p - cG_t \tag{9}$$

which has slope $-G_t$, and ordinate intercept G_p , both of which are fixed by the thickener operating conditions. The abscissa intercept equals G_p/G_t , which is equal to the underflow concentration c_u . The terminal settling flux S_t is defined similarly as cu_t and is a characteristic of the sludge, being a function of concentration. The requirement that u be less than u_t is equivalent to Sbeing less than S_t , and so the operating line must lie below the terminal or no-compression settling flux line throughout the thickening zone.

In the case of a conical thickener, the operating line (S vs. c) is not straight, and it cannot be drawn before Equations (2) and (7) have been solved because the cross-sectional area at which each concentration occurs is not known. However, as in the cylindrical case, the operating line must lie below the no-compression line throughout the sediment.

COMPARISON OF CYLINDRICAL AND CONICAL PERFORMANCE

Only a few data are available on compressibility and permeability of sludges. Smiles gave data on some Australian red mud, and these will be used for illustration. For this material $g(\rho_{\nu} \rho_l$) equals (Smiles, 1975) 20.90 Mg/m^2s^2 , and the compressibility and permeability data (Smiles, 1976) are fitted by

$$\sigma = a \ 10^{-D}$$
, where $a = 98.1 MPa$ (10)

and

$$u_t = e \ 10^D D^{-f}$$
 where $e = 3.70 \ nm/s$, and $f = 6$ (11)

for the range D = 6 to 3 (c = 0.167 to 0.333). Equation (10) can be used to determine the relation between bottom concentration and depth in a static sediment (as obtained at the completion of batch thickening), which is

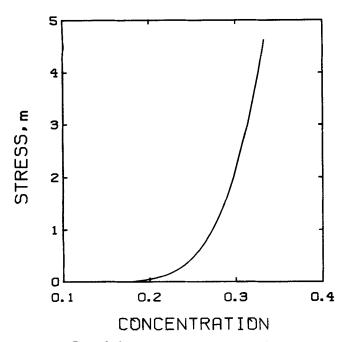


Figure 1. Compressive stress vs. concentration.

$$X = a \ 10^{-D} (D + (\ln 10)^{-1}) / g(\rho_{\nu} - \rho_{t})$$
 (12)

This gives the minimum possible sludge depth required to produce a given underflow concentration in a continuous thickener, corresponding to an infinitely small particle throughput rate.

According to Equation (10), σ is zero at zero concentration (although this involves extrapolation outside the range of the data). One would expect σ to be zero up to the nonzero concentration at which the flocs just touch, that is, up to the critical concentration. However, the critical concentration is very difficult to identify experimentally owing to the ease of compression of sludges when the flocs first come into contact, and, owing to the ease of compression, it is not necessary to identify the critical concentration accurately because the early stages of compression require only a very small depth. In the calculations reported below, the critical concentration was taken as 0.1, and Equations (10) and (11) were assumed to hold down to this value.

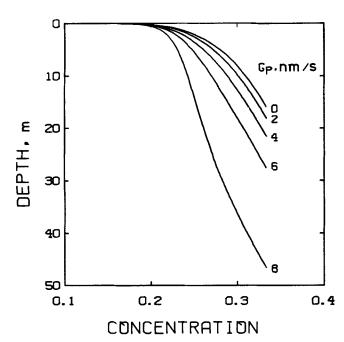


Figure 3. Concentration vs. depth, Figure 2 operating conditions and zero flux.

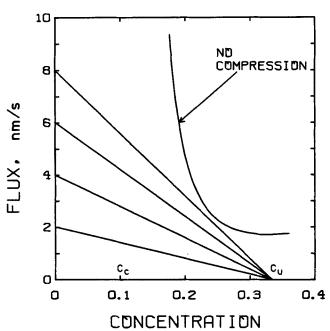


Figure 2. Settling flux plot, including four cylindrical operating lines.

According to Equation (12), the static depth required to compress from 0 to 0.1 is only 4.9 μ m. However, the difficulty of compression rapidly increases, as shown in Figure 1. [In this figure, and following ones, stresses and pressures are expressed in terms of head of submerged particles, by dividing by $g(\rho_{\nu} - \rho_l)$.]

Figures 2 to 4 show the results of calculations for a cylindrical thickener at four particle fluxes and a fixed underflow concentration of 0.333. These define a pencil of operating lines, as shown in Figure 2. The variations of concentration with depth in each sediment are shown in Figure 3. [The sediment starts at the critical concentration, at the top. In the clarification zone above, the average concentration is lower and not shown in the figure. With transient resistance to compression neglected, the particles jump to the critical concentration as they settle onto the top of the sediment (Dixon, 1978).] It is noted that the depth of sediment required increases as G_{ν} increases; the closer the operating line is to the no-compression settling flux line, the

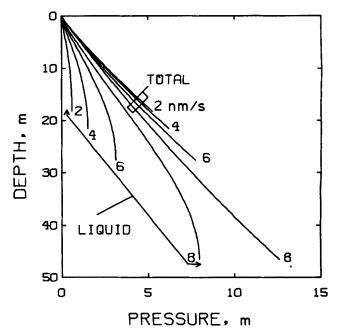


Figure 4. Total and liquid excess pressures vs. depth, Figure 2 operating conditions.

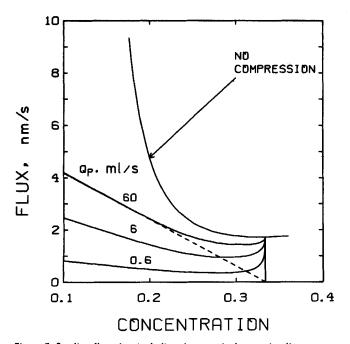


Figure 5. Settling flux plot, including three conical operating lines. $c_u = 0.333$, 60 deg cone, 200 cm² bottom area.

greater the fraction of particle weight which is supported by liquid drag, and the greater the sediment depth required for the chosen concentration. [It will also be noticed that the chosen underflow concentration is such that even in the zero flux limit, the required depth $(16\,\mathrm{m})$ is greater than is normally available in a thickener.]

Figure 4 shows the variations with depth of the weight of particles per unit area (the total stress) and the liquid excess pressure (the amount by which the liquid pressure exceeds the hydrostatic pressure corresponding to the depth) for each of the fluxes. At each level, the difference between the total stress and the excess pressure is the particle compressive stress. The spatial gradient of the excess pressure equals the drag force on the particles per unit volume of suspension. The higher the particle flux, for fixed underflow concentration, the greater the proportion of particle weight which is borne by drag, as seen in Figure 4. However, even at the highest flux, which is very close to the

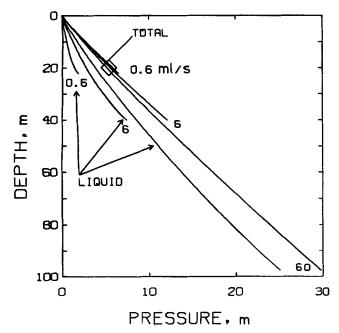


Figure 7. Total and liquid excess pressures versus depth, Figure 5 operating conditions.

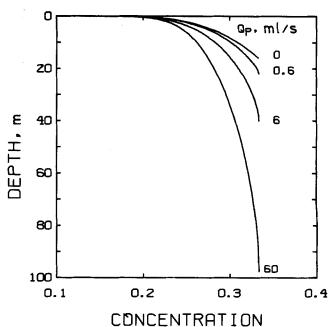


Figure 6. Concentration vs. depth, Figure 5 operating conditions and zero flux.

limiting value corresponding to an operating line which is tangential to the no-compression line, only about 63% of sediment weight is supported by drag. The reason for this is easily seen from Figure 2. Even at a high flux, the operating line cannot be close to the no-compression line over the whole of the sediment concentration range $(c_c \text{ to } c_u)$. At the lowest flux, only a small part of the particle weight is supported by drag, so that the liquid excess pressure line is far below the total pressure line.

Figures 5 to 7 show the results of calculations for thickening the red mud to 0.333 concentration in a cone with 60 deg included angle and a bottom area of 200 cm², at particle throughput rates of 0.6, 6.0 and 60 ml/s. Based on the bottom area, these rates correspond to fluxes of 30 to 3 000 μ m/s, which are very much greater than the limiting flux for cylindrical thickeners at the same underflow concentration (9 nm/s, Figure 2). However, the fluxes are smaller at higher levels, of course, since the area is larger.

Figure 5 shows the operating lines obtained, which are seen to be of quite complex shape, and to pass, on average over the concentration range, much closer to the no-compression line than any cylindrical operating line. At the largest of the three throughput rates the operating line passes very close to the no-compression line in the vicinity of c_u , which cannot occur in the cylindrical case. As a result, a large fraction of particle weight in the sediment is supported by drag and a corresponding small fraction by interparticle stress. Eighty-five percent of sediment weight is supported by drag (Figure 7), compared to 63% in the cylindrical case at close to the limiting flux.

It will be noticed in Figure 5 that the operating lines are nearly straight at the low concentration end (the top of the sediment). As shown by the dashed line in the figure, the straight portions intersect the abscissa at the underflow concentration, when extrapolated. At the top of the sediment the concentration changes very rapidly with depth (Figures 3 and 6) so that little area variation occurs over the low concentration end of the operating line. In this range, the operating line is close to the cylindrical line corresponding to the area at the sediment top. However, at higher concentrations (lower levels), the area reduction causes the relative velocity to be higher than it would be if the cylindrical operating line were followed, and the operating line bends toward the no-compression line.

It will also be seen in Figure 5 that the operating lines are very steep at their c_u ends (the bottom of the thickener). This is because, at the bottom of the thickener, the operating line is

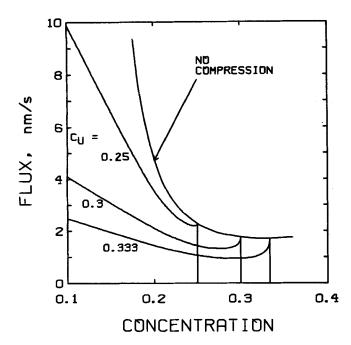


Figure 8. Settling flux plot, including three conical operating lines. $Q_{\nu} = 6$ ml/s, 60 deg cone, 200 cm² bottom area.

tangential to the cylindrical operating line corresponding to the bottom area, which is steep because the area is small. Differentiating Equation (9) with respect to c, and using Equations (3) and (4), we get

$$\frac{dS}{dc} = \frac{d(Q_p - cQ_t)/A}{dc} = -\left[(Q_p - cQ_t)/A^2 \right] \frac{dA}{dc} - Q_t/A$$
$$= -\left(S/A \right) \frac{dA}{dc} - G_t \tag{13}$$

Since S equals zero at the bottom, the slope of the operating line is $-G_t$, which is the slope of the cylindrical operating line. The conical line deviates from the bottom cylindrical operating line at lower concentrations (that is, above the thickener bottom) because the area increases as the concentration decreases (S/A > 0, dA/dc < 0).

If the particle throughput rate is held at 6.0 ml/s, but the underflow concentration is reduced to 0.3 and to 0.25 (that is, the sludge pumping rate is increased to 111 and to 133% of its previous value), a remarkable result is obtained (Figures 8 to 10). The operating line moves closer to the no-compression line (Figure 8), so that the fraction of sediment weight supported by drag increases (Figure 10). This is the opposite of the result for the cylindrical case, where keeping G_{ν} fixed and reducing c_{ν} moves the operating line away from the no-compression line, so that the fraction of the weight supported by drag decreases.

Decreasing c_u causes the operating line to move closer to the no-compression line in the conical case because of the increase in the average compressibility of the sludge. The more easily compressible the sludge, the greater the rate of change of concentration with depth (other things being equal), and hence the smaller the rate of change of area with concentration [dA/dc] in Equation (13)]. Since the bottom area is fixed, but not the top, it is easier to consider changes occurring with increase in height above the bottom. The operating line moves away from the no-compression line more rapidly, or toward it less rapidly, than it would otherwise because of the increase in area with height. However, the lower the sludge concentration, the less rapidly the area increases as concentration decreases, and the closer overall is the operating line to the non-compression line.

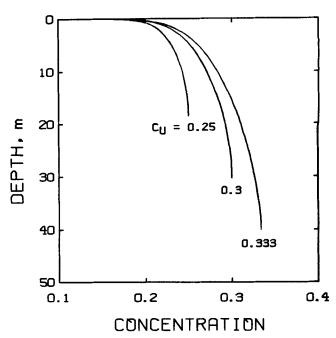


Figure 9. Concentration vs. depth, Figure 8 operating conditions.

EFFECT OF FUNNELING IN A CYLINDRICAL THICKENER

If sludge funneling occurs through a large part of the depth of sediment in a thickener, an obvious suggestion is that the thickener might perform more nearly according to conical than according to cylindrical theory. To follow closely the conical theory given above, the sludge would have to form a central conical core, horizontally uniform at each level, surrounded by completely stagnant sludge and negligibly affected by flow of liquid from the higher pressures in the core to the lower pressures in the surroundings. Clearly, a thickener will not conform precisely to this flow pattern, but nevertheless its performance could lie between those predicted by cylindrical and conical theories.

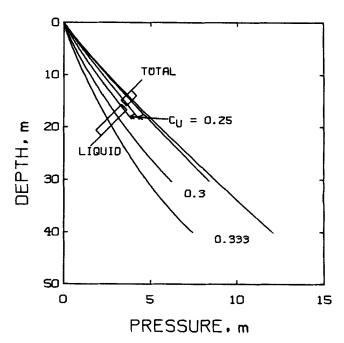


Figure 10. Total and liquid excess pressures vs. depth, Figure 8 operating conditions.

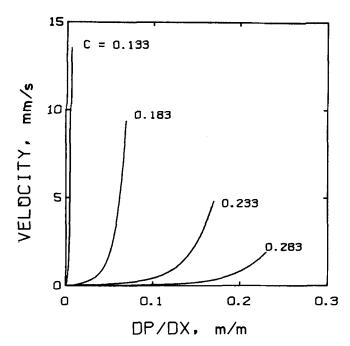


Figure 11. Apparent velocity vs. liquid excess pressure gradient from conical thickener data interpreted according to cylindrical theory.

A striking deviation of observed cylindrical thickener behavior from cylindrical theory was first reported by Fitch (1962), and has been verified by later workers (Kos, 1977; Flannery, 1978). In an experimental program lasting 2 yr, a laboratory thickener was operated over a wide range of conditions, and concentration and excess pressure variations with depth were measured. In no case was appreciable particle pressure observed; that is, the liquid pressure was always close to that required to support the whole weight of the particles. As shown above, these results are at variance with predicted cylindrical behavior. Even when operating close to the limiting flux for a given underflow concentration, an appreciable fraction of total submerged particle weight should be supported by interparticle stress, and the fraction should be greater at lower fluxes,

On the other hand, the results given above show that nearly the whole of the particle weight can be supported by drag in a conical thickener, especially at lower underflow concentrations. If the sludge concentration is sufficiently high (low sludge pumping rate) and the throughput sufficiently low (and the thickener is deep enough to accommodate the sediment), a low degree of drag support must be obtained, with or without funneling. However, it appears that laboratory investigations to date have not entered the appropriate operating range to give appreciable particle pressure, due to the effect of funneling.

Dahlstrom et al. (1973) quote typical underflow concentrations from red mud thickeners as having a maximum value of 0.15. According to Equations (10) and (12), the stress required to achieve this concentration is 20 Pa, corresponding to a static sediment depth of only 7.2 mm. If plug flow existed in a cylindrical thickener, the necessary sediment depth would not be more than two or three times the static depth, say 2 cm, even at close to limiting throughput. Clearly, the rakes could not move the sludge to the outlet pipe of a large thickener if so small a depth were present, and the sludge builds up so that it can funnel toward the outlet.

Thus it seems quite possible that in typical industrial thickeners, little of the sludge depth is effective in causing sludge compression because funneling is a necessary part of the sludge removal process. In deep cone thickeners, used for some years in U.K. collieries (Abbott et al., 1973), considerably greater depth than usual is available (about 7 m). Data of Dell and Keleghan (1973) indicate that the typical deep cone underflow concentration of 65 mass % (about 0.4 volume fraction) corresponds to a static depth of about 1.5 m. It seems that a significant fraction of sludge depth is effective in sludge compression in these thickeners, and data on the pressure and concentration profiles in them would be of interest (although they are usually operated with intermittent sludge withdrawal, and so not at steady state).

Kos (1977) used data from a laboratory scale continuous thickener to deduce the relation between drag and relative velocity for the material involved and found a non-Dareyan relation. He determined the liquid pressure gradient at a given level from the measured pressure distribution and calculated the relative velocity at the level from the concentration, using Equation (6) and the particle feed and sludge pumping rates. Instead of a linear relation between velocity and drag, as used in Equation (1), he found that the drag varied in proportion with a small power of the velocity (much less than unity).

This is an unexpected result, of course, since drag is normally found to vary with a power of velocity between one and two, the former corresponding to low Reynolds number flow which is expected in a thickener. Kos found little particle pressure acting in the sludge, and his drag vs. velocity relation could be due to funneling, especially in view of the fact that he did not use rakes. When it is assumed that no funneling is occurring, u is calculated from Equation (6) taking G_p and G_t as constants, calculated from Q_p , Q_t and the thickener cross-sectional area [Equations (3) and (4)]. However, if funneling occurs, the effective cross-sectional area decreases toward the bottom, where u values are lower. Thus, at low u values, the calculated values could be smaller than the true values, so that if the true relation is linear, the apparent u vs. drag relation is concave upward.

This is illustrated in Figure 11, where the data used to prepare Figures 5 to 7, plus results for some additional particle throughput rates, have been cross plotted. The apparent u was calculated without taking into account the cross-sectional area variation, the fluxes being based, at all levels, on the bottom area. The curves in Figure 11 have the same shape as those obtained by Kos and show funneling to be a possible cause of the apparent deviation from Darcy's Law which he found.

If funneling of sludge is a significant effect in thickeners, as the above results indicate, a complicating factor is thereby introduced into thickener design. The calculations reported above give only a qualitative indication of the effect of funneling by assuming that the sludge flow is actually conical, surrounded by stagnant sludge. The real sludge flow pattern is expected to depend on the thickener geometry, the speed of the rakes, the quantity of sludge in the thickener, the particle throughput rate and the rheological properties of the sludge. Given the necessary data on the sludge, the thickening process would be described by differential equations in at least two dimensions (for a circular thickener, assuming circular symmetry), which would require lengthy numerical solution. The flow pattern is also expected to be different in different diameter thickeners, so that scale-up of pilot thickener data will also be difficult. Further investigation is needed before a method can be developed for taking the effect of sludge funneling into account in thickener design.

NOTATION

The positive direction is taken as downward for all vector quantities.

= constant in red mud compression equation, ML-1T-2 a

= cross-sectional area, L²

= particle concentration, volume fraction

= critical particle concentration c_c

= underflow particle concentration C_{2}

D particle dilution = 1/c

constant in red mud permeability equation, LT^{-1}

constant in red mud permeability equation

f acceleration due to gravity, LT-2 particle volumetric flux, LT

sludge volumetric flux = volume-average velocity, LT^{-1}

permeability, M-1L3T

= particle volumetric flow rate, L3T-1

= sludge volumetric flow rate, L^3T^{-1}

= settling flux = cu, LT⁻¹

 S_t = terminal, or no-compression, settling flux = cu_t , LT⁻¹

= particle settling velocity, relative to G_t , LT⁻¹ u

= terminal, or no-compression, settling velocity, LT⁻¹ u,

= particle velocity, relative to the container, LT⁻¹

= vertical position, L x

X = total depth of sediment, L

θ = included angle of cone

= liquid density, ML-3 ρ_l

= particle density, ML-3 ρ_p

= interparticle compressive stress, based on total crosssectional area, ML⁻¹T⁻²

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Manuscript received June 7, 1979; revision received November 19, and accepted December 21, 1979.

An Introduction to the Nonlinear Theory of **Adsorptive Reactors**

Coupling between transient chemical reaction at equilibrium and a linear and isothermal chromatographic separation is studied when a nonlinear chemical mass action law is assumed to hold between the species. It is shown that the qualitative features of the chromatograms can be predicted by simple algebraic calculations. Two novel properties of the adsorptive (or chromatographic) reactor are demonstrated. The improvement of the theory, with regard to quantitative predictions, is discussed.

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SCOPE

An adsorptive or chromatographic reactor is defined as a transient chemical reactor in which the reactants and the products undergo a chemical reaction and a chromatographic separation simultaneously. The coupling between these two physicochemical phenomena gives rise to peculiar behaviors with respect to standard reactors: equilibrated reaction forced to completion, enhancement of selectivity, of chemical reaction rate, etc. Dinwiddie (1966) and Magee (1961) have outlined the potential uses of this reactor in industry. On the other hand, Langer et al. (1972) and Hattori et al. (1968) have pointed out that the chromatographic reactor is a good tool to determine kinetic schemes or to measure kinetic rates with a little amount of reactant. In biochemistry, the method is used to measure association constants of drugs and proteins (see, for example, Oster et al. 1976). In the field of separation science, Shiloh (1966) and Golden et al. (1974) have studied the competition between separation and precipitation, complexation or neutralization in ion exchange columns (see Klein, 1978).

Unfortunately, the analysis of the transient behavior of the reactor needs complicated models. Two kinds of models are found in the literature: complicated models, which account for

as many as possible physical processes and require tedious numerical procedures to be solved (Chieh Chu, 1971; Hattori, 1968) and simple models in which simplifying assumptions are made (first-order kinetic in Langer et al., 1972, linear chromatography in Kocirick, 1967, and Villermaux, 1978, instantaneous equilibrium in Magee, 1963, or Gore, 1967, analogy with semibatch reactor in Schweich et al., 1978).

The purpose of this paper is to give a first insight into what we think to be a general method which describes the behavior of the adsorptive reactor when the feed composition change is an instantaneous step function (the case of a feed pulse is not studied). It is assumed that dispersion and mass transfer processes are negligible and that the reaction is at equilibrium at every time and at any point inside the isothermal column. We use the same mathematical method as Tondeur (1969). It is very similar to the method of characteristics used by Rhee (1970 to 78) and to the concept of coherence developed by Helfferich (1967) and Helfferich and Klein (1970). After the development of theory, the transient esterification of acetic acid with ethanol and the dehydrogenation of cyclohexane to benzene are interpreted as illustrative examples.

Some possible generalizations are suggested, especially in the frequent case of nonlinear adsorption isotherms.