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Theory of Spherical Agglomeration. II. Layering/Crushing Process in a Rotating Conical Drum*

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Abstract

A theoretical study is made of the so-called layering/crushing agglomeration process in a rotating conical drum under steady-state continuous flow conditions. A particular application is the separation of bitumen from the solid particles in oil sands, where the nonwetting liquid is a bitumen-solvent mixture and the wetting liquid is water. It may be assumed that the water is completely imbibed by the agglomerating granules (particles), so that the system consists of granules suspended in the nonwetting liquid. In the layering/crushing process, the granules are divided into two nonoverlapping size distributions, the small crushed granules and the large granules on which the layering takes place. The agglomeration process therefore becomes a complicated example of three-phase flow. The three phases are the continuous nonwetting liquid and the two granular phases. The steady-state mass balance equations for the two groups of granules in the rotating conical drum can be integrated approximately. The mean velocity of the layered (large) granules parallel to the axis of the cone is directed from apex to

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base whereas the corresponding velocity of the crushed (small) granules is in the opposite direction.

INTRODUCTION

In the spherical agglomeration process, a high concentration of dispersed granules (agglomerates) with a changing size distribution is agitated in a fluid environment consisting of two immiscible liquids, preferentially wetting and nonwetting with respect to the solid material of the granules. Since the wetting liquid is almost entirely absorbed in the interstices between the individual solid grains of the agglomerating granules, the latter may be regarded as suspended in the continuous nonwetting liquid. There are several stages in the agglomeration process. (i) Nucleation, in which primary particles of a powder feed combine to form nuclei or seeds. (ii) Coalescence, which involves rapid growth of granules by combination of a number of nuclei. (iii) Layering, when the agglomerates reach a certain size. Larger agglomerates grow by coalescing with much smaller particles, which may be primary feed, nuclei, or pieces of broken (crushed) agglomerates, hence the description layering/crushing. In this paper we are mainly concerned with the layering/crushing processes.

Recently (in Part I) the authors (*1*) studied the steady-state spherical agglomeration process in a rotating conical drum, used to separate bitumen from solid particles in oil sands. The wetting liquid was water and the nonwetting (suspending) liquid a bitumen–solvent mixture. From the continuity equation for the flow of granules suspended in the bitumen–solvent liquid and experimental results, some general conclusions were reached concerning the agglomerating granules in the conical drum. In a second paper the spherical agglomeration process was treated theoretically as multiphase flow by one of the authors (*2*). Most theories of multiphase flow are confined to two phases, where one of the phases consists of discrete particles and agglomeration is absent (*3, 4*). Because of the agglomerating process, it is convenient to treat the granules in a small mass range as a separate phase. The equation of continuity (mass balance relation) for these granules includes a source term which describes the different stages in the agglomeration process listed above.

As it is a formidable task to solve the "microscopic" equations of motion giving the paths of the individual granules, the technique of so-called volume averaging (or an equivalent technique) is usually employed to investigate the mass-balance and momentum-balance equations of multiphase flow theory (*5, 6*). Parameters occurring in these equations of motion relate to averages over a volume which is large compared with the volume of a single granule but small on the scale of macroscopic inhomogeneities. Each phase, whether dispersed or continuous, is regarded as filling the whole volume of the physical system, so that the phases behave as interpenetrating continua. This would appear to raise difficulties when the granules over a small mass range are treated as a continuous phase. However, a consequence of size segregation is that in general each small mass range will occupy a fraction of the whole volume. Clearly, the mass range cannot be too small unless the size distribution is very narrow or the size segregation is pronounced.

In the layering/crushing process, where the sizes of the larger (layered) and smaller (crushed) granules do not overlap, it is possible to treat the agglomeration as three-phase flow. The three phases are the continuous suspending liquid and two discrete phases, the layered and crushed granules. We shall only examine here the mass balance equations for these three phases and shall return to the much more difficult problem of the momentum balance equations in a later paper. Although many authors have studied the latter equations for multiphase flow in the absence of agglomeration, differences in their interpretation exist (*3*) and the presence of agglomeration creates additional difficulties.

MASS BALANCE (CONTINUITY) EQUATION IN LAYERING/CRUSHING AGGLOMERATION

Choosing a continuous distribution of granule sizes, the continuity equation (also called the population balance equation) for granules of mass m is

$$\frac{\partial n(m)}{\partial t} + \nabla \cdot (n(m)\mathbf{v}(m)) + \frac{d}{dm}[G_1(m)n(m)] = S(m) \qquad (1)$$

where $n(m)dm$ is the number of granules/unit volume in the mass range $m,m + dm$; $\mathbf{v}(m)$ is the velocity of a granule of mass m; $G_1(m)$ is that part of the growth function which is not directly due to agglomeration or its opposite comminution; and $S(m)$ is the source term. The complete growth function which we denote by $G(m)$ is defined as the rate at which granules grow beyond their mass m, i.e., it equals dm/dt. The last term on the left-hand side of Eq. (1) was derived by Hulburt and Katz (*7*) from a statistical mechanical treatment (see also Ramkrishna and Borwanker (*8, 9*) and Ramabhadran and Seinfeld (*10*)). In spherical agglomeration, a source of variation with time in granular mass is entrapment of the suspending (nonwetting) liquid in the pores between the solid grains of the granules and this would account for the growth term $G_1(m)$. Such entrapment may be appreciable in the coalescence stage when the granules are still small but its significance dimishes as the granules grow.

Precisely $S(m)dm$ is the rate of change with time due to agglomeration in the number of granules/unit volume in the mass range $m,m + dm$. We shall assume that the agglomerates have reached a sufficient size where coalescence can be neglected and consider only the layering/crushing process in which the material to be layered is provided by crushed pieces and by added powders. The source term $S(m)$ may then be written as

$$S(m) = -\frac{d}{dm}[G_2(m)n(m)] - B(m)n(m) + \int_m^{m_0} B(\tilde{m})n(\tilde{m})\nu(\tilde{m})p(m,\tilde{m})d\tilde{m} + C(m) \quad (2)$$

where the different terms on the right-hand side have the following meanings. The first term accounts for layering, the next two terms crushing, and the last term the addition of powder. $G_2(m)$ is the growth function due to layering. On combining with $G_1(m)$, we have

$$G(m) = G_1(m) + G_2(m) = dm/dt \quad (3)$$

$B(m)$ is the fraction of granules of mass m that are crushed in unit time. The integral term in Eq. (2), when multiplied by dm, is the rate of production of the number of granules in mass range $m,m + dm$ from the crushing of all granules larger than m (*11*). The upper limit of integration m_0 ($> m$) is the mass of the largest granule that is crushed. $p(m,\tilde{m})dm$ is the probability of producing a daughter granule in the mass range $m,m + dm$ upon crushing of a parent granule of mass $\tilde{m}$, and $\nu(\tilde{m})$ is the avreage number of daughter granules. If more than two daughters are produced as a result of the crushing of the mass $\tilde{m}$, then $\nu(\tilde{m}) > 2$. $C(m)dm$ is the rate (number/unit time) at which particles of fine powder in the mass range $m,m + dm$ are added to unit volume of the agglomerating system. The quantities $n(m)$, $\nu(m)$, $G_1(m)$, and $S(m)$, which are examples of the volume averages mentioned in the Introduction, all depend on their position in the agglomerating apparatus and also on time t in the unsteady state. It is convenient to introduce an effective fraction of granules crushed:

$$B_e(m) = B(m) - \frac{1}{n(m)}\int_m^{m_0} B(\tilde{m})n(\tilde{m})\nu(\tilde{m})p(m,\tilde{m})d\tilde{m} \quad (4)$$

and also an effective source term:

$$S_e(m) = S(m) - \frac{d}{dm}[G_1(m)n(m)]$$

$$= -\frac{d}{dm}[G(m)n(m)] - B_e(m)n(m) + C(m) \qquad (5)$$

so that the last term of the left-hand side of Eq. (1) has been absorbed into the source function. (In Part I (*1*), the growth term $G_1(m)$ and the integral in Eq. (4) were omitted. Thus $G(m)$ should include $G_1(m)$, and $B(m)$ should be interpreted as $B_e(m)$ in Part I.)

STEADY-STATE FLOW CONDITIONS

Consider the steady state when $n(m)$ is independent of time t in Eq. (1). On multiplying Eq. (1) by m and integrating with respect to m, we obtain

$$\nabla \cdot \int mn(m)\mathbf{v}(m)dm = \int mS_e(m)dm \qquad (6)$$

where we have interchanged the orders of the operation ∇ and integration with respect to m. Suppose, for simplicity, that the feed has negligible velocity compared with the velocity of the granules. Then the left-hand side of Eq. (6) is the net rate at which granular mass leaves unit volume, and in steady-state conditions this should equal the net rate at which mass of powder is being provided, i.e.:

$$\nabla \cdot \int mn(m)\mathbf{v}(m)dm = \int mC(m)dm \qquad (7)$$

If no feed is added, then the right side of Eq. (7) equals zero. Subtracting Eq. (7) from Eq. (6) and making use of Eq. (5), it follows that

$$\int m \frac{d}{dm}[G(m)n(m)]dm = -\int mB_e(m)n(m)dm \qquad (8)$$

The particles in the agglomerate occur in three separation size groups, the larger (layered) granules (mass m'), the smaller (crushed) granules (mass m''), and powder particles (mass m''') where $m' > m'' > m'''$. The integral on the left-hand side of Eq. (7) extends over the ranges of m' and m'' whereas that on the right-hand side of Eq. (7) only over the range m'''.

We are assuming that no overlap occurs in the ranges of m', m'', and m'''. The degree of entrapment of suspending (nonwetting) liquid within the granules decreases as the granules grow by agglomeration. We shall ignore entrapment in the larger (layered) granules, i.e., we assume $G_2(m'') = 0$ and hence $G_1(m'') = G_2(m'')$. It is convenient to combine all terms in $S_e(m)$ which differ from zero only in the range of m'' by introducing

$$D(m) = B_e(m) + \frac{1}{n(m)} \frac{d}{dm} [G_1(m)n(m)] \tag{9}$$

$D(m)$ is dominated by $B(m)$, so that $D(m) > 0$. Thus we postulate that $G_2(m) = 0$ unless m lies in the range of m', $D(m) = 0$ unless m is in the range of m'', and $C(m) = 0$ unless m is in the range of m'''. Equation (8) can now be written as

$$\int m' \frac{d}{dm'} [G_2(m')n(m')]dm' = -\int m''D(m'')n(m'')dm'' \tag{10}$$

It is possible to carry out an integration of Eq. (1) in the steady-state condition for a rotating conical drum agglomeration apparatus. Let z denote distance along the axis of the conical drum and $A(z)$ the cross-sectional area at z which is occupied by the agglomerating charge. Also let $v_z(m)$ be the component of the velocity vector $\mathbf{v}(m)$ parallel to the axis z of the drum and let an overbar denote an average over the cross-sectional area $A(z)$. Then we can prove that (*1, 2*)

$$\frac{d}{dz} \overline{(n(m)v_z(m))} + \frac{1}{A(z)} \frac{dA(z)}{dz} \overline{n(m)v_z(m)} = \overline{S_e(m)} \tag{11}$$

If the origin on the z axis is chosen at the true (geometrical) apex of the cone, then a reasonable approximation is $A(z) \sim z^2$, in which case Eq. (11) becomes

$$\frac{d}{dz} \overline{(n(m)v_z(m))} + \frac{2}{z} \overline{n(m)v_z(m)} = \overline{S_e(m)} \tag{12}$$

A good approximation is $\overline{n(m)v_z(m)} \simeq n(m)\overline{v_z(m)}$ if $n(m)$ is very nearly constant in any cross-sectional normal to the z axis. The solution of Eq. (12) is then

$$\overline{v_z(m)} = \frac{1}{z^2\overline{n(m)}} \int_{z_0}^{z} z^2\overline{S_e(m)}dz \tag{13}$$

where $z = z_0$ marks the apex of the operating conical drum. For $m = m'$ or $m = m''$, we may assume that $\overline{n(m)} = 0$ at $z = z_0$. (In Eq. 29 of Part I, the lower limit of integration should be z_0.)

In the layering/crushing process discussed here we can apply Eq. (13) separately to the two nonoverlapping size distributions (m' and m''). Making use of Eq. (9), we can write Eq. (5) as

$$S_e(m) = -\frac{d}{dm}[G_2(m)n(m)] - D(m)n(m) \tag{14}$$

where m does not include m'''. Since $G_2(m'') = 0$ and $D(m') = 0$, it follows from Eqs. (13) and (14) that

$$\overline{v_z(m')} = -\frac{1}{z^2\overline{n(m')}} \int_{z_0}^{z} z^2 \frac{d}{dm'}[G_2(m')n(m')]dz \tag{15}$$

and

$$\overline{v_z(m'')} = -\frac{1}{z^2\overline{n(m'')}} \int_{z_0}^{z} z^2\overline{D(m'')n(m'')}dz \tag{16}$$

These equations yield an important result. Since $D(m'') > 0$, the right-hand side of Eq. (10) is always negative and it follows from Eq. (16) that $\overline{v_z(m'')} < 0$, i.e., the mean axial velocity of the smaller crushed granules is directed from the base to the apex of the conical drum. In contrast, assuming that the integrand on the left-hand side of Eq. (10) does not change sign over the range of m', then $\overline{v_z(m')} > 0$. The mean velocity of flow parallel to the axis of the cone of the larger (layered) granules is toward the base of the conical drum. These conclusions are consistent with the results of two of the authors who observe that in a continuous run of their rotating conical vessel, any mixture of large and small granules will tend to segregate with the large granules at the base and the small ones at the apex (*12*). Also, Sugimoto (*13*) has described granulation experiments where fine powder, fed into the middle of a rotating cone, segregates into fine and coarse granules at the apex and base, respectively.

SOME QUALITATIVE FEATURES OF $\overline{n(m)}$ AND $\overline{S_e(m)}$ IN STEADY-STATE FLOW

In discussing the form of $\overline{n(m)}$ (strictly $\overline{n(m)\Delta m}$ over a small mass range Δm), it is convenient to display explicitly the dependence of $n(m)$ on distance along the axis z and so we write $\overline{n(m)} = \bar{n}(m,z)$. (Similarly, we shall write $\overline{S_e(m)} = \bar{S}_e(m,z)$.) In Part I (*1*), schematic plots of $\bar{n}(m,z)$ were drawn for the agglomeration process in the rotating conical drum, which included both coalescence and layering/crushing. These were suggested by available information on granule distribution obtained from measurements and also visual observations. Figures 1 and 2 show corresponding schematic plots of $\bar{n}(m,z)$ for the layering/crushing process in

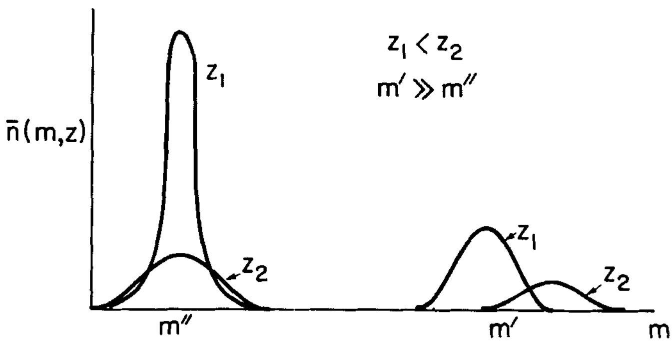

FIG. 1. Schematic representation of $\bar{n}(m,z)$ in conical drum. Constant z contours in $(\bar{n},m)$ plane for layered granules (mass m') and crushed granules (mass m'').

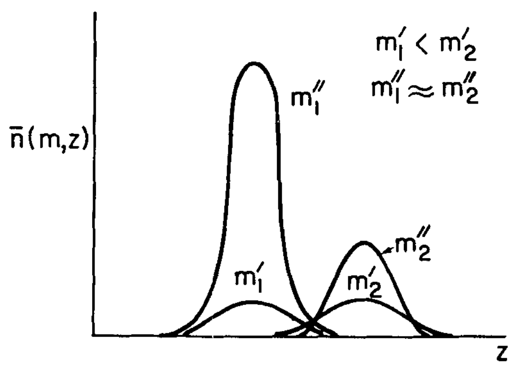

FIG. 2. Schematic form of $\bar{n}(m,z)$ in conical drum. Constant granular mass contours in $(\bar{n},z)$ plane for layered (m') and crushed (m'') granules.

the rotating conical drum. Constant z contours in the $(\bar{n},m)$ plane are drawn in Fig. 1 for two different values of z, which are distinguished by the suffixes 1 and 2, where $z_1 < z_2$. Clearly the plots of $\bar{n}(m,z)$ for the two sizes of granules (m' and m'') are completely separate along the m axis, since there is no overlap. Also $n(m'',z)$ may exceed $n(m',z)$ by orders of magnitude but the curves in Fig. 1 are not drawn to scale. In Fig. 2, constant m contours are drawn in the $(\bar{n},z)$ plane, again schematically. Curves for two masses $m'_1 < m'_2$ of the larger granules and two masses $m''_1 \simeq m''_2$ of the smaller granules are shown. In that section of the conical drum where layering/crushing takes place, the mass of the layered granule must be increasing with z and hence the number $\bar{n}(m',z)$ must be decreasing. The crushed granules need not grow in size as z increases since they are not agglomerating. That their mean velocity parallel to the axis is directed toward the apex suggests that their number decreases with an increase in z. One characteristic that seems reasonable is that the plots of $\bar{n}(m',z)$ and $\bar{n}(m'',z)$ along the z axis should cover approximately the same interval of the axis.

Let us first examine $\bar{S}_e(m,z)$ in the case where powder feed is absent, i.e., $C(m) = 0$. Then from Eqs. (6) and (7)

$$\int m\bar{S}_e(m,z)dm = 0 \tag{17}$$

Also, noting that $D(m'') > 0$ and $G_2(m'') = 0$, by Eq. (14), $S_e(m'') < 0$. Relation (17) is consistent with $S_e(m') > 0$, which we shall assume here and which, since $D(m') = 0$ by Eq. (14), implies that $G_2(m')n(m')$ decreases as m' increases. (It is observed experimentally that $G_2(m')$ $(= dm'/dt)$ actually increases with m', hence the decrease of $n(m')$ with increasing m' must be more marked.) Using these results, schematic plots of $\bar{S}_e(m,z)$ as a function of m for two different fixed z can now be drawn. As illustrated in Fig. 3, $\bar{S}_e(m,z)$ consists of a negative portion for the smaller granules (m'') and a position portion of the larger granules (m'). In view of the weighting factor m in Eq. (17), the area under the negative portion in Fig. 3 is very much larger than the area under the positive portion, but these areas are not drawn to scale. Making use of Figs. 2 and 3, characteristic plots of $\bar{S}_e(m,z)$ against z for given granular masses are shown in Fig. 4, again not drawn to scale. The curves in Fig. 4 are consistent with the results that $\overline{v_z(m')} > 0$ by Eq. (15) and $\overline{v_z(m'')} < 0$ by Eq. (16). The general profiles in Figs. 3 and 4 still apply if $C(m) \neq 0$ and $S_e(m') > 0$.

CONTINUITY EQUATION FOR THE SUSPENDING LIQUID PHASE

Let ρ_c be the (constant) density of the suspending (nonwetting) liquid, $\mathbf{v}_c$ its velocity, ε the fraction of the total volume occupied by the liquid, and

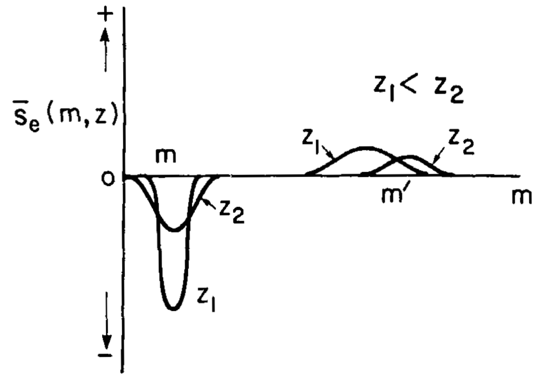

FIG. 3. Schematic plot of $\bar{S}_e(m,z)$ as a function of m for different fixed z. m' and m'' equal masses of layered and crushed granules, respectively.

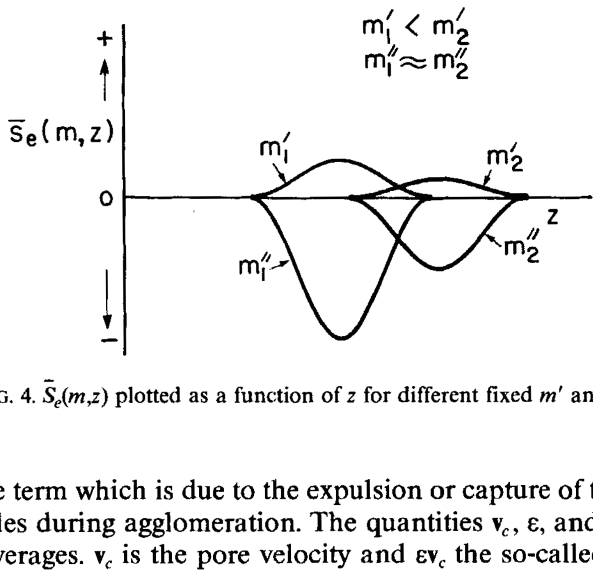

FIG. 4. $\bar{S}_e(m,z)$ plotted as a function of z for different fixed m' and m''.

q_c a source term which is due to the expulsion or capture of the liquid by the granules during agglomeration. The quantities $\mathbf{v}_c$, ε, and q_c are local volume averages. $\mathbf{v}_c$ is the pore velocity and $\varepsilon\mathbf{v}_c$ the so-called superficial velocity. (In the experiments on extraction of bitumen from oil sands, some bitumen–solvent mixture, which constitutes the suspending liquid, is trapped in the granules.) In steady-state flow, the mass balance equation is

$$\nabla \cdot (\rho_c \varepsilon \mathbf{v}_c) = q_c \tag{18}$$

The left-hand side is the net rate at which the suspending liquid mass is departing from unit volume of the agglomerating system. Thus q_c is the rate of charge with time in the mass of suspending liquid being added to unit volume of the system due to expulsion or capture of this liquid by the granules in the agglomeration process. Let us assume that the rate of increase of granular mass $G_1(m)$, introduced in Eq. (1), is due to the entrapment (or occlusion) of the suspending nonwetting liquid. Then

$$q_c = -\int n(m'')G_1(m'')dm'' \tag{19}$$

assuming that $G_1(m') = 0$. If v_{cz} is the z-component of the velocity $\mathbf{v}_c$ and an overbar again denotes a cross-sectional average in the conical drum, then we derive from Eq. (18) the equation corresponding to Eq. (12) for granules, namely:

$$\frac{d}{dz}\,(\overline{\varepsilon v_{cz}}) + \frac{2}{z}\,\overline{\varepsilon v_{cz}} = \bar{q}_c/\rho_c \tag{20}$$

with solution

$$\overline{\varepsilon v_{cz}} = (\overline{\varepsilon v_{cz}})_{z=z_0} + \frac{1}{\rho_c z^2}\int_{z_0}^{z} z^2\bar{q}_c(z)dz \tag{21}$$

In the experiments with the conical drum on bitumen separation from oil sands by the agglomeration method (*12, 14*), the oil sands are fed in at the apex and the suspending (bitumen–solvent) liquid at the base. The drum is designed so that the bulk of the suspending medium flows from the base to the apex of the conical drum under gravity, so that the first term on the right-hand side of Eq. (21) is negative. With change in agglomerate size, the so-called residual saturation of the suspending medium (the equilibrium amount remaining with the agglomerate bed after gravity drainage) is illustrated in Fig. 5. This shows that as the granules grow when they travel from apex to base during agglomeration, they are expelling the nonwetting liquid, i.e., $\bar{q}_c(z) > 0$, and therefore $G_1(m'') < 0$. Furthermore, $\bar{q}_c(z)$ decreases with an increase in z, as shown schematically in Fig. 6. It follows from Eq. (21) that the cross-sectional average superficial velocity $\overline{\varepsilon v_{cz}}$ increases in magnitude from the base to the apex $z = z_0$, which is to be expected since the area $A(z)$ is least at $z = z_0$. Equation (21) may be used in the region of coalescence as well as in that of layering/crushing.

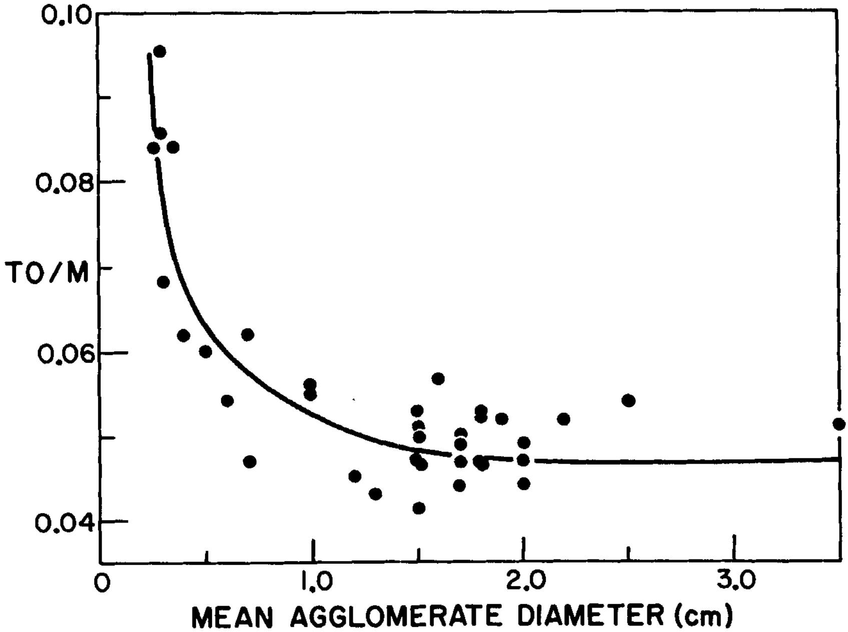

FIG. 5. Plot of ratio (suspending (nonwetting) liquid content to solids content) within a granule as a function of the granule diameter in experiments with the conical drum on bitumen separation from oil sands, described in Refs. *12* and *14*.

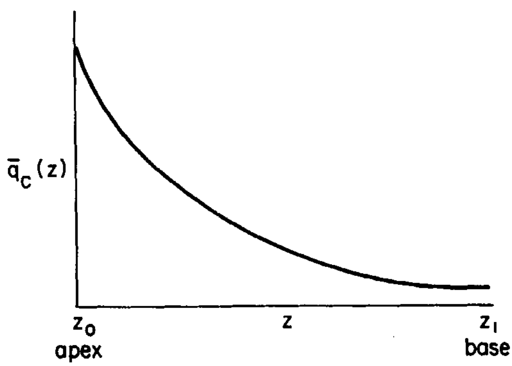

FIG. 6. Schematic plot of $\bar{q}_c(z)$ as a function of z in bitumen separation from oil sands (*12, 14*).

DISCUSSION

In this paper we have only considered the motion of the granules and suspending liquid parallel to the axis of the rotating conical drum. The velocity components of both granules and suspending liquid in a cross-sectional plane of the drum far exceed the corresponding velocities parallel to the axis. The latter are due to spiralling motions. As explained in Part I, the much larger velocity components normal to the drum axis are eliminated from the continuity (mass balance) equations under steady-state conditions by applying the divergence theorem to a cross-sectional slab of the conical drum. For our model of the layering/crushing process, we are able to separate the motion of the layered granules from that of the crushed granules. This yields a result, which although admittedly approximate, is an example of size segregation, a phenomenon which indeed is not well understood. It is striking that the use of the mass balance equation for the granules is sufficient to yield our predictions that the layered and crushed granules move on the average in opposite senses, parallel to the drum axis.

ERRATUM FOR PART I (*1*)

Some have already been mentioned in the text. Additional corrections are as follows:

Equation (24): should read $\int_{D_1}^{2^{1/4}D_1} N(D)dD$ = constant

Page 90, 6 lines from bottom of 2nd paragraph:
"*m* increases with *m*" should read "*m* increases with *z*"

The line following Eq. (29) should read:
"where charge is fed into the drum at position $z = z_0$"
in place of:
"where $\bar{v}_z(m,z)$ must be finite at the apex ($z = 0$) of the conical drum"

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