

The use of diffusion theory for the assessment of mixing in a rotary coating pan

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The effectiveness of a coating pan in treating all the contained tablets identically depends upon its being a good mixing device; the mechanics of particle motion within such a pan are therefore of interest. One measure of the mixing rate in rotary drum mixers is the coefficient of self diffusion, a parameter difficult to measure in a coating pan. Three methods of measuring such a coefficient are reported and compared: linear sampling, total compartmenting of the bed, and high speed ciné-photography. The third method is the best, and a technique is described for treating the pictorial data to obtain quantitative results.

ONE of the more readily usable parameters in the kinetics of mixing processes in rotary drum mixers is the effective diffusion coefficient. To measure this, the mixer is initially loaded with two, usually equal, batches of particles, the batches being distinguished from each other by any property which can be assumed to have no influence on the mixing rate. The progressive interchange of the particles of the two halves of the initial charge is followed, and by the application of known solutions for the differential equation expressing Fick's second law of diffusion,

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2},$$

under the appropriate boundary conditions, the diffusion coefficient D for the mixing process can be calculated. Here c is the concentration, t is the time, and x is the linear measure of distance from the initial position of the interface between the two halves of the initial batch.

Such an approach has been used by Kaye & Sparrow (1964), who examined the sideways mixing of streams of particles as they flowed down an inclined plane. This was an attempt to isolate the surface diffusion mechanism in a rotary drum mixer from the convective mixing and shear mixing which, according to Lacey (1954), all contribute simultaneously to the mixing process in a rotary drum.

The rate of mixing in the axial direction in a long rotating cylinder was studied by Hogg, Cahn, & others (1966). These authors showed that the concentration along the mixer axis closely followed the appropriate solution of Fick's equation. In a later paper, Cahn, Fuerstenau, & others (1966) considered the mechanism by which random axial motion is generated in the shear zone of a drum mixer. Such random motion is a necessary presupposition if the use of diffusion theory is to be appropriate. They considered this a sufficiently important question to justify the simulation of the diffusion process by a Monte Carlo method on a computer (Cahn & Fuerstenau, 1967).

To study the mechanics of particle movement within a coating pan, and relate them to the overall performance, a measure of the prevailing rate

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of particle mixing is needed. Although the diffusion coefficient is an obvious choice, it is subject to some restrictions and disadvantages. The solution to the case of diffusion across a boundary from one semi-infinite cylinder to another is readily available from standard texts, e.g., Jost (1960).

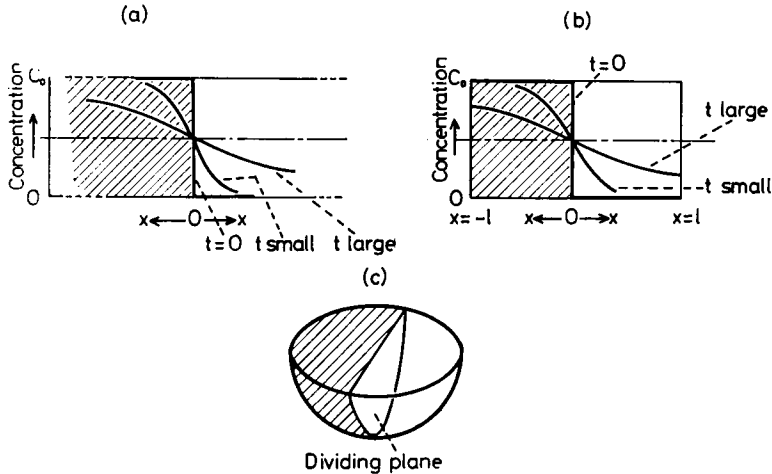


FIG. 1. (a) diffusive mixing in a semi-infinite system. Eqn: $c(x,t) = \frac{1}{2}c_0 \operatorname{erfc} \frac{x}{2\sqrt{Dt}}$. (b) diffusive mixing in a system of finite length. Eqn: $c(x,t) = \frac{1}{2}c_0 \sum_{\text{all } n} \left\{ \frac{\operatorname{erf}(2n-1) \pm x}{2\sqrt{Dt}} \right\}$. (c) the geometry of the mixing system formed by the two-coloured bed in a coating pan.

Fig. 1 (a) shows the initial conditions and the progress of diffusive mixing with time for this case. Where the diffusional transfer is between cylinders of finite length, the solution becomes more complex, but still exists and is tractable. Because the diffusion equation is linear, any linear combination of solutions is a solution, which allows "reflection" at the plane barrier at the end of a cylinder to be used as a mathematical device to satisfy the condition of zero diffusion rate at such a boundary. Fig. 1 (b) shows the curves for this situation. The solution is a rapidly-converging series, especially for small values of t . The summation represents the reflection and super-positioning of the concentration wave.

The coating pan, however, is represented by Fig. 1 (c), and the explicit solution will be so complex that the best approach is probably a successive approximation type of numerical analysis by computer for any specific case. The question then arises whether the simple equation of Fig. 1 (a) will be applicable to conditions within the split load of a coating pan during times which are short compared with the time taken for mixing to progress through the entire charge. Experimentally, this paper shows that it is applicable, and that a value of the effective diffusion coefficient can be obtained. As soon as reflection occurs from the non-plane boundary, the simple theory breaks down.

Experimental

THE COATING PAN AND ACCESSORIES

The apparatus initially used consisted of a standard enamelled coating pan, 25 cm in diameter, mounted on and driven by an Erweka unit type KU 1 (Erweka Apparatebau G.m.b.H; Frankfurt). The speed and axial tilt of the pan could be controlled. In later experiments a transparent pan became necessary, and one was constructed from a 10 litre single-necked flask (Q.V.F. Ltd., Stone, Staffs). A large circular hole was cut in the base of the flask, which was then fastened by its buttress-ended neck to a plate which could be mounted in the rotary drive unit. The inside of the neck was filled with Polyfilla so that the inside surface of the flask was uninterruptedly spherical.

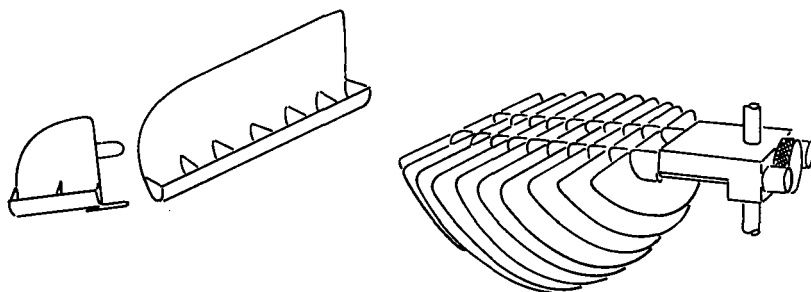


Fig. 2. The linear sampling device and the pan-dividing vanes.

Ancillary equipment included a linear compartmental sampling device and a set of pan-dividing vanes, both shown in Fig. 2. The sampler was made by bending a sheet of brass into a J-shape. Small semicircular pieces of brass were then soldered into the hook of the J to divide it into eight small compartments. A sample of the pan contents could then be taken along a straight line. The set of pan-dividing vanes was cut from thin stainless steel sheet; the completed set consisted of 12 plates located 14.5 mm apart, each one of a profile to fit the pan closely in the position in which it was fixed. The vanes enabled multicoloured beds of particles to be set up, and also enabled mixing rate determinations to be made.

The vanes were supported from outside the pan and observations could be made of particle transfer from one zone to another as the pan was rotated with the vanes held stationary. A second set was made to fit the glass pan.

Some of the ciné-photography was done with a Bolex H 16 reflex camera (Paillard S.A., St. Croix, Switzerland). This was an excellent camera for normal speed work, but it had a maximum speed restriction of 64 frames/sec. The higher speed work was done with a Fastax 16 mm high speed camera, Model WF 3 (Revere-Wollensak Div., Minnesota Mining and Manufacturing Co., Rochester, N.Y.) which could operate at speeds between 150 and 8000 frames/sec. Illumination was provided by two or more Phillips Photolita 500 W lamps, and for colour work, Ektachrome EF type 7242 film was used, which had a speed of 125 ASA. A zoom lens

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was used to vary the field of view without changing any other variable, and this gave the incidental advantage of a great reduction in perspective effects. Thus the view of the moving surface of the particles was practically an orthographic projection.

MANUFACTURE OF UNIFORM PARTICLES

Lactose B.P. (2 kg), of mean particle size $94\ \mu$, and a size range of 30 to $170\ \mu$, was placed in the pan which was rotated at 30 rev./min with its axis at 30° to the horizontal. A 12% aqueous solution of polyvinylpyrrolidone (Plasdone) was sprayed in with a hand atomizer until the critical liquid content was attained, and granule growth began. Rotation was continued until spherical particles of diameter about 7.5 mm formed the bulk of the pan contents, when hot air was blown in to dry the surfaces of the particles. The pan contents were then sieved into three fractions: (a) $<6.7\ \text{mm}$ (b) 6.7–8.0 mm and (c) $>8.0\ \text{mm}$. Fraction (b) was dried overnight at 35° and stored. Fraction (a) was grown to larger size by adding lactose powder and more Plasdone solution. Fraction (c) was ground and re-used. The spherical particles of the correct size were coated with 12% Plasdone in chloroform solution to give them resistance to abrasion; dyes were incorporated in the final coat to give spheres which differed from one another only in colour.

The bulk density of about 8 kg of the particles was determined. The value obtained was 0.722 g/ml.

MEASUREMENT OF DIFFUSION COEFFICIENT

Three methods have been used to measure the amount of mixing taking place in the pan in terms of an effective diffusion coefficient.

(a) Half the pan was charged with granules of one colour, and the other half with granules of another but of the same diameter. The division between the charges was vertical and generally at right angles to the axis of rotation of the pan, i.e., the division was between the front and the back of the pan looking in through the pan mouth. The pan was rotated and after a number of revolutions was stopped. Eight samples of about 20 granules were taken, with the linear sample device, along a central line from back to front of the pan. The number of each colour of granule in each compartment was determined and the rotation of the pan restarted.

(b) The pan was operated with the dividing vanes in position and held stationary, starting with a bed divided in two colours as before. Mixing on the rolling surface of the bed still took place under these conditions, but without impactive mixing at the toe of the bed because the granules did not converge there. Pan rotation was stopped periodically, the pan was rotated backwards about 45° so that the bed surface returned to the horizontal; the vanes then divided the bed completely into sections. Each section was withdrawn by suction into a conical flask connected to a vacuum pump, counted and returned to the pan into the section from which it had been withdrawn.

(c) Both the above methods interfered to some extent with the mixing process within the pan, since it was necessary to stop the pan movement to remove and to return the sample of the granules. In the third method

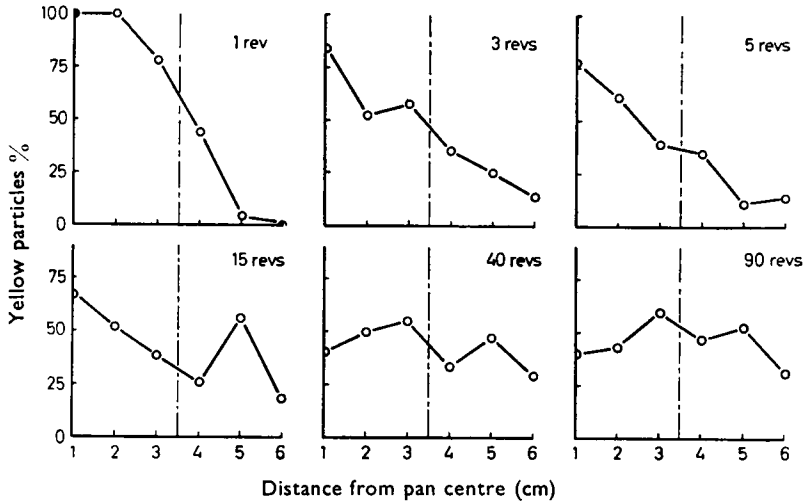


FIG. 3. Diffusive mixing results for method (a): periodically arresting the pan to take a linear sample.

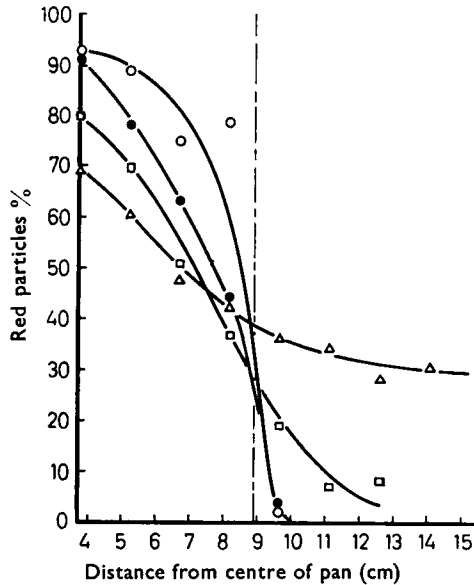


FIG. 4. Diffusive mixing results for method (b): running the pan with the dividing vanes in position, counting all the particles in each compartment periodically. ○ 5 revolutions, ● 10 revolutions, □ 40 revolutions, △ 100 revolutions.

a two coloured bed was prepared and mixed by rotation of the pan. The process was followed by ciné-photography. Subsequently, frames of the film were projected and a tracing made of the convoluted interface between the granules of the two colours. The percentage of each colour at any

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distance from the mean interface position was then determined by measuring the lengths of the intercepts made by a line parallel to the mean interface position, and at the required distance from it.

All three methods give a measure of the composition, as a colour percentage, along a line from the back to the front of the pan, as a function of number of revolutions. The composition distribution starts as a sudden discontinuity and gradually progresses through a series of sigmoid curves to a straight line at the 50% level.

Results and discussion

Method (a). The results are shown in Fig. 3, in which the graphs are similar in form to those of Fig. 1 (b). The scatter of the points is quite large, $\pm 10\%$ would be a conservative estimate, because of the sampling error. One compartment of the linear sampling device takes a sample of about 20 particles from the bulk. If the true proportion of one colour, p , in the bulk is 50%, the variance to be expected in a sample of n particles, where $n = 20$ is $np(1-p) = 5$ with a standard deviation $\sqrt{5} = 2.23$. Since the proportion of one colour is estimated from a sample containing about 10 particles of that colour, this represents an error of about $\pm 20\%$, a severe limitation on the method. Previous authors have not encountered this problem because they worked with ballotini of such small particle diameter that samples of 500 particles were taken by suction through a hypodermic needle. Any attempt to increase the sample size in a coating pan means that quantities of particles are being withdrawn large enough to be a significant proportion of the total available.

Method (b) overcomes the sampling difficulty since the entire bed is divided and each section counted individually. The results for this type of experiment are shown in Fig. 4. The difficulty of this technique is that

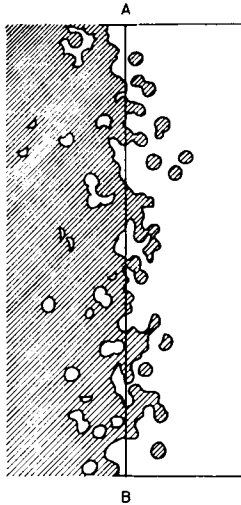


FIG. 5. Method (c): tracing of one frame of the ciné film. AB is a typical line on which the intercepts are measured and summed.

the pan must be operated with the vanes in position: any attempt to insert them to split the bed into slices causes more mixing. Thus the bed is mixing under rather different conditions. However, the effect of the vanes should be minimal because coating pans operate within the speed range classified as "low" by Carley-Macaulay & Donald (1962). Mixing in this range occurs only in the cascading surface layer: particles in the bulk of the bed are held by their neighbours and are carried round in an arc of a circle by the pan rotation. With the vanes in position, the movement of most of the bed surface was not impeded. But it is still true that in attempting to remove the sampling errors, the mechanism of particle movement is modified.

Method (c) ensures that there is no interference with the mixing mechanism, since all observation is photographic. Sampling presents a difficulty, in that only the surface layer of the bed is visible. Were a group of 20 particles counted, they would cover an area so large that the concentration change across it would be large. The method used was to trace a few frames of the film at convenient times from the start of the experiment, by projecting the picture onto tracing paper (cf. Fig. 5). The mean interface position was established by planimeter measurement. Lines were drawn at 2 mm intervals parallel to the mean interface, and the intercepts on each line made by regions of each colour were determined. The concentration of each colour as a function of distance from the interface was then plotted for each of a number of times of mixing. Some of these curves are shown in Fig. 6, and their quality is better than that of the curves of Figs 3 and 4. The points on the curves from the first two methods are so scattered that any form of derived plot is of doubtful value. In Fig. 7 the curves of Fig. 6 are plotted on probability paper; as expected, they give straight lines, since they are of the general form e^{-x^2} . Because the equation relating concentration to distance is:

$$C = \frac{c_0}{2} \left[1 - \operatorname{erf} \left(\frac{x}{2\sqrt{Dt}} \right) \right]$$

where c_0 is the initial concentration difference between the two halves of the system, then

$$\frac{dc}{dx} = -\frac{c_0}{2} \cdot \frac{2}{\sqrt{\pi}} \cdot \exp \left[-\frac{x^2}{4Dt} \right] \cdot \frac{1}{2\sqrt{Dt}}$$

and for $x = 0$,

$$\frac{dc}{dx} = \frac{c_0}{2\sqrt{\pi Dt}}$$

This gives the slope of the curve of concentration against distance, at the initial interface position, as a function of time. This slope is unaffected by plotting on probability paper, and the slope of the entire line, if it is straight, on a probability plot will be $c_0/2\sqrt{\pi Dt}$. Thus, if the slope is measured at successive intervals of time, a plot of the slope against $1/\sqrt{t}$, or alternatively of (slope)² against $1/t$, should be a straight line so long as D is constant. Constancy of D is a test of whether the simple diffusion law is applicable.

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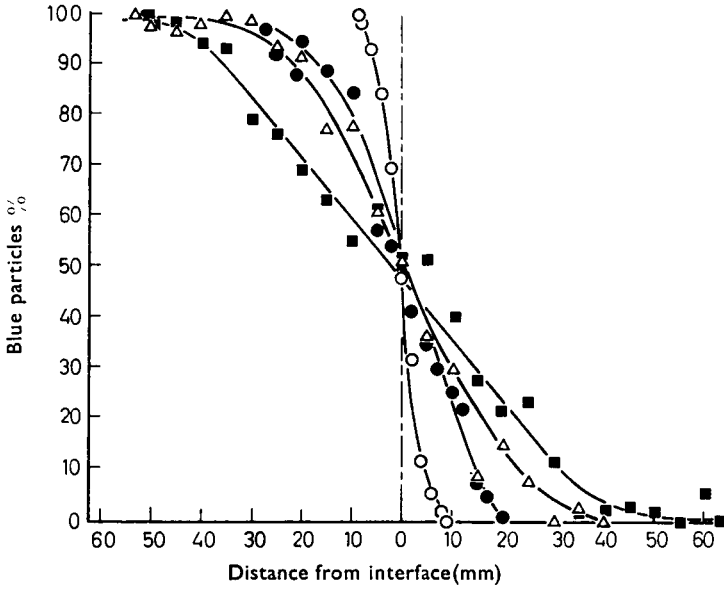


FIG. 6. Diffusive mixing results for method (c), calculated from a number of frames similar to Fig. 5. ○ 2, ● 4, △ 8, ■ 12 sec.

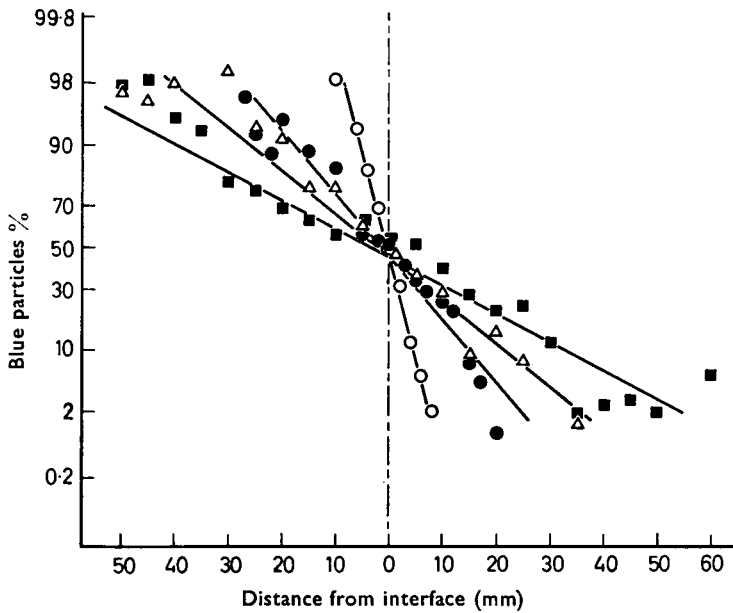


FIG. 7. The curves of the type shown in Fig. 6, plotted on probability paper to give a series of straight lines of slope $1/2\sqrt{\pi Dt}$. ○ 2, ● 4, △ 8, ■ 12 sec.

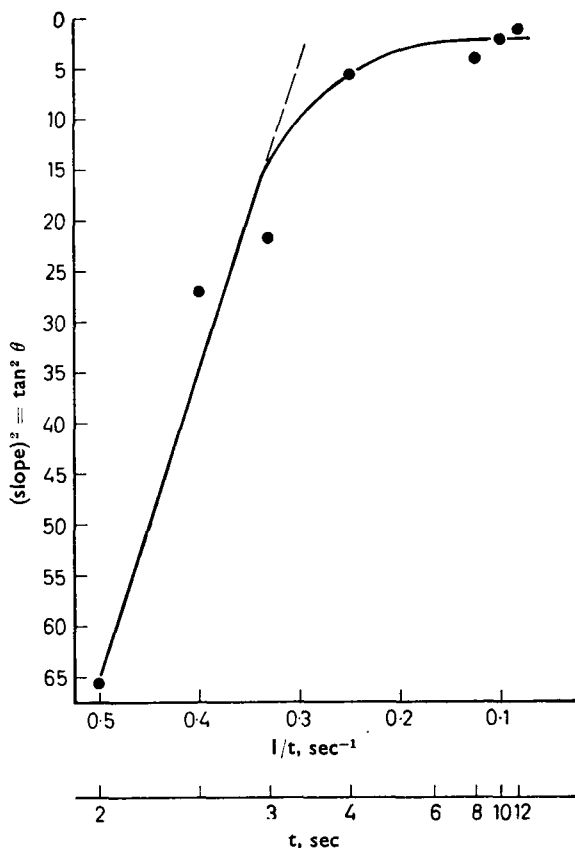


FIG. 8. A plot of the $(\text{slope})^2$ from curves of the type shown in Fig. 7 against the reciprocal of the time. The linear portion of the graph gives the time over which the simple equation of Fig. 1(a) is applicable.

Such a plot is made in Fig. 8, and it is seen that for short times there is a straight line relation. The value of D for the straight part of the curve is $0.9 \text{ cm}^2 \text{ sec}^{-1}$. The main criticism of the third method is that it is based only upon observation of the surface layer of the bed, and it is the surface value of D that is being measured. The entire series of measurements for the third method occupied only about half a revolution of the pan, and represents the time taken for the initial dividing plane between the two colours of the charge to be completely destroyed. It is thus the period over which the top end of the bed surface is being supplied with unmixed particles.

CONCLUSION

Three methods of determining the effective diffusion coefficient between the two differently-coloured halves of a batch of particles in a rotary coating pan have been tested. The values obtained are independent of

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the boundary conditions which must be applied in solving the diffusion equation for any particular physical situation, provided that the elapsed time, measured from the start of the mixing process, is short.

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