

MIXING OF SEGREGATION PARTICLES

by

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CHAPTER 1

INTRODUCTION

Solids mixing is a common processing operation widely used in a variety of industries. It is extensively employed in the manufacture of ceramics, fertilizers, detergents, glass, pharmaceuticals, animal feeds, and in the powder metallurgy industry.

Three principal mechanisms are known for the process of solids mixing: convective, diffusive and shear mechanisms (Lacey, 1954). However, the extent of mixedness as a function of time is generally difficult to predict because satisfactory theories capable of describing the mechanisms have not been fully developed.

The phenomenon of solids mixing has been analyzed largely based on a linear diffusion mechanism (see e.g., Lacey, 1954; Otake et al., 1961; Hogg et al., 1966; Chen et al., 1973). Such a simple approach is applicable only to nonsegregating particle systems, i.e., highly idealized systems.

The behavior of systems where the particles differ in properties and characteristics (segregating particle systems) is a matter of practical concern in industrial operations. The occurrence of demixing (segregation) is extremely important, since it can markedly alter product quality. Despite this importance, the simultaneous occurrence of mixing and demixing in segregating particle systems has received relatively little attention.

In recent years, solids mixing in a fluidized bed has been the subject of many experimental and theoretical studies. The simplicity of design, the ease of application in continuous systems, and the turbulent motion of particles are attractive characteristics associated with a

fluidized bed, which make it an effective device for solids mixing. The phenomenon of axial solids mixing in fluidized beds has been described by many investigators (see e.g., Rowe and Partridge, 1962; Kunii and Levenspiel, 1968; Schugerl, 1969). However, only few reports are available on the radial solids mixing in fluidized beds despite of its importance especially when heat transfer is involved. Correlations for estimation of the radial solids mixing are lacking.

Fluidization research has been focused largely on fine particles ($< 500 \mu\text{m}$). Relatively little attention has been given to the fundamental behavior of particles larger than $500 \mu\text{m}$. Nevertheless, there are some applications of fluidization where the use of large particles can be advantageous (e.g., Cox, 1958; Anon., 1961; Brown et al., 1972; Vogel et al., 1975; Wilson and Gillmore, 1975). In general, correlations for estimation of the solids mixing in large particle beds are rather sparse.

The present study has been conducted to investigate:

- (1) Axial mixing of segregating particles in a motionless mixer.
- (2) Mixing of large particles in a two-dimensional gas fluidized bed
--nonsegregating particle system.
- (3) Mixing of large particles in a two-dimensional gas fluidized bed
--segregating particle system.

In the next chapter, an extensive review on the mixing of segregating particle systems is provided. In the third chapter, a modified coalescence-dispersion model is given for the axial mixing of segregating particles in the motionless mixer. In the fourth chapter, a stationary random walk model is presented for the progress of axial and radial mixings of the large nonsegregating particle system in the two-dimensional gas fluidized bed. In the fifth chapter, the stationary random walk model

is extended to a nonstationary one for the axial mixing of the large segregating particle system in the two-dimensional gas fluidized bed. Finally, major conclusions and recommendations for future research needs are summarized.

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CHAPTER 2

LITERATURE SURVEY

2.1 INTRODUCTION

Segregation is a process which produces separation or classification of particles. Mixing particles of different characteristics is generally accompanied by segregation, which prevents the attainment of a random state of mixing. A typical example is shown in Figure 1. As can be seen from the figure, the homogeneity improves rapidly during the early stage of mixing, however, as the mixing is prolonged, the homogeneity is decreased due to the inherent segregation tendency.

The behavior of segregating particle systems where the particles differ in properties and characteristics is a matter of practical concern in industrial operations. The occurrence of the demixing (segregation) is extremely important, since it can markedly alter product quality. Despite of this importance, the simultaneous occurrence of mixing and demixing in segregating particle systems has received relatively little attention.

In addition to surveying the existing literature on segregation, in the present review, an attempt is made to identify several research areas that are in need of study.

2.2 CAUSES OF SEGREGATION

While there is no absolute way to detect if a given particle system will segregate it is possible to stipulate a few conditions that tend to cause the segregation. Characteristics of particles which may

affect a mixing process are size, size distribution, shape, effective particle density, surface characteristics, surface conductivity, bulk density, flowability, angle of repose, and resistance to agglomeration (Van Denburg and Bauer, 1964). Obviously not all of these are independent variables. Some, such as bulk density and angle of repose, represent the composite effect of two or more of the variables. A completely random mixture can be obtained only when the net influence of these factors is negligible.

It has been shown that size difference is the most important segregation factor in conventional mixers (Stairmand, 1962; Van Denburg and Bauer, 1964; Valentin, 1965; Williams, 1965, 1975; Campbell and Bauer, 1966). However, when aerated suspensions are created during a mixing process, e.g., mixing in a fluidized bed, density difference may become predominant (Rowe and Nienow, 1975).

2.2.1 Size and Size Distribution

Most industrial segregation problems arise from the size and the size distribution. There is no ready way to relate the size distribution with the performance of a mixer, but in general, the narrower the size distribution the less the tendency to segregate.

Fowler (1959) reported that smaller particles had greater angle of repose. The angle of repose aids segregation by causing the smaller particles to roll in a direction opposite to the larger ones. Olsen and Rippie (1964) proposed a first-order kinetics for the rate of segregation in a particle system with size difference. The rate constant can be correlated with the size and the size distribution. Hill (1965) reported that the segregation tendency was decreased when the proportion of small

particles was increased and vice versa. Donald and Roseman (1962a, 1962c) concluded: the greater the size difference, the faster the segregation. The size effect is further illustrated by Fig. 2, which is a plot of the standard deviation at steady state vs. the diameter ratio of ash to sand in a horizontal drum mixer. The line connecting the experimental points has a slope of 3, indicating that the size ratio is an important factor in segregation.

2.2.2 Density

Rose (1959) examined the experimental data of Coulson and Maitra (1950) for the mixing of components with different densities in a inclined drum mixer. The data cover only the region where mixing effects are dominant. Syskov and Iyan (1960) reported the data on the extent of density segregation in ore-coal mixtures. Fischer (1960) has proposed that the role of density on the segregation is determined by the ratio of the supporting surface to the mass. Donald and Roseman (1962b) discussed the effect of density differences and concluded that once the density ratio exceeds 1.2 segregation starts to have an effect in a horizontal drum mixer. Rippie et al. (1964) studied the interaction between the density and the size in particle systems with both size and density differences.

Nicholson and Smith (1966) studied the axial mixing of particles differing in density in a fluidized bed. Nienow et al. (1972) reported the role of particle size and particle density in segregation of particles in a fluidized bed. They concluded that a fairly wide particle size difference can be tolerated without appreciable segregation while even a small density difference led to readily settling of the denser particles.

2.2.3 Shape

Significance of the particle shape lies primarily in its effect on the flow characteristics of particles. Other properties being equal, particles of different shapes will assume different angles of repose and hence any process which involves rolling or sliding will likely to separate them (Williams and Shields, 1967). The smooth-surfaced, spherical or egg-shaped particles tend to flow readily. The rough-hewn fractured pieces are less mobile, while the interlocking filaments, rods and complex crystals often flow so poorly that they tend to set up a small-scale equivalent of a log jam (Fischer, 1960).

Another point regarding the shape is that the irregular configuration displays more surface area per unit volume. This can be of importance when the surface effect becomes predominant or an aerated suspension is created during a mixing operation (Fischer, 1960).

2.2.4 Electrostatic Charge

The electrostatic effect is a direct consequence of the sufficiently high surface area compared to the mass of particles. Particle streams that flow freely in the uncharged state may become charged by agitation or mechanical stirring. Fischer (1960) concluded, "Electrostatic forces alone may be too weak to cause the motion of particles on a large scale. But when these forces are superimposed on random dynamic forces, the relatively small electrostatic forces can produce drastic effect."

2.3 MEASUREMENTS OF SEGREGATION

Williams and Kahn (1973) concluded, "In spite of its industrial importance, very few attempts to measure the extent of segregation."

Over 30 different mixing indices have been reviewed by Fan et al. (1970, 1975). These mixing indices constitute the counter-parts of segregation indices, and thus can be used as measurements of segregation as well.

Danckwerts (1952) proposed a useful segregation index analogous to the "scale of segregation" used in the statistical theory of turbulence. Williams and Birks (1965) devised a test for the measurement of segregation in a mixture divided into two regions. Williams and Schields (1967) defined a coefficient of segregation for the system in which the mixture is separated into halves. Harris and Hilton (1970) suggested that the summation over the absolute values of deviation from the mean can be used as a measurement of segregation in a core-flow central filling hopper. Rogers and Clements (1971) devised a sampling scheme to detect segregation in a tumbling mixer. Lai et al. (1974) proposed the test statistic of the Mann-Whitney test can be used as an index of segregation without assuming that the population is a normal distribution.

2.4 MECHANISMS OF SEGREGATION

Brown (1939) has described two mechanisms of segregation:

- (1) Vibrational segregation, in which larger particles tend to rise to the surface when a mixture is vibrated.
- (2) Pouring segregation, in which larger particles tumble further down the free surface of a pile.

Ashton and Valentin (1966) have proposed two additional mechanisms:

- (3) Free flight segregation, which results from different trajectories of materials falling at an angle.

- (4) Percolation segregation, in which small particles can pass through the interstices between large particles.

The vibrational segregation has been studied by Stairmand (1962) and Williams (1965). According to them, a simple geometrical analysis will show that spherical particles segregate in this way if the size of the smaller particles is less than one-sixth that of the larger ones. Williams (1963) reported a single large particle in the bed of small particles can be segregated to the top on vibration. Suzuki et al. (1968a, 1968b) and Takahashi et al. (1968) studied the solids circulation patterns in a two-dimensional hopper-shaped vessel subjected to vertical sinusoidal vibrations. This circulatory motion has been considered to play an important role in the segregation of the particles.

Pouring segregation is characterized by the fact that the fines are concentrated in the cylindrical region around the vertical axis of a cone with the coarses on the outside when a mixture is poured on to a free surface. Lawrence and Beddow (1968) noticed, in their study of a powder segregation during die filling, an inner mound of fines-rich material in the bottom of the die and an excess of coarse particles in the periphery of the die cavity. This segregation was increased with the rate of die filling and the height from which the powder falls into the die. Tanaka (1971) reported that heavier particles tended to remain near the central portion while lighter particles tended to roll down the slope toward the bin wall when the bin was filled.

The free flight segregation is due to the fact that if particles are projected horizontally the distance they will travel is proportional to the square of the particle diameter (Williams and Kahn, 1973). Fowler (1960) studied the variables that affect the segregation of particles discharged from bins and hoppers. In order to obtain clarity on this

phenomenon, Opitz (1971) took some pictures for the free fall of a stream of raw meal.

The percolation segregation is the size, shape, or density segregation in the failure zone between two bodies of particles moving at different velocities under the influence of gravity and strain (Bridgwater and Ingram, 1971). Bridgwater et al. (1969), in an attempt to separate out the interparticle segregation from the mechanisms of particle mixing, considered the random walk movement of percolating particles through a packed bed of larger particles. Bridgwater (1971) demonstrated the effect of percolation on the macroscopic equipment performance by a model mixer where the convection and the interparticle percolation predominate. Campbell and Bridgwater (1973) devised an equipment to demonstrate the occurrence of percolation in failure zones. Scott and Bridgwater (1975) studied the strain-induced interparticle percolation under closely controlled conditions. The amount of percolation was found dependent upon the size of particles, the total strain, and the rate of strain.

2.5 SEGREGATION MODELS

Several attempts have been made to model the behavior of segregating particle systems (see Table 1). Some of these are purely empirical. Others are semi-empirical or mechanistic models for segregation in specific mixers.

Rose (1959) has proposed the equation

$$\frac{dM}{dt} = A(1 - M) - B\psi \quad (1)$$

where

M = degree of mixedness,

ψ = demixing potential,

A, B = constants.

The first term in the right-hand side represents the mixing effect and the second term represents the demixing effect and is based on a somewhat arbitrarily defined demixing potential. The demixing potential ψ is defined by

$$\psi = \pm \sqrt{1 - M} \quad (2)$$

Equations (1) and (2) can be solved to obtain

$$M = \eta \{1 - [B/A \pm (1 \mp B/A) \exp(-At/2)]^2\} \quad (3)$$

where η is another constant to account for imperfectness of the mixer.

It was pointed out (Danckwerts, 1959) that A must be greater than B if the expression is to be physically compatible.

Weydanz (1960) has proposed a model for a two-dimensional process in which the segregation effect is present in only one dimension. The main assumptions are that the rate of segregation is constant and that the volume interchanges in either direction are constant. The model can be expressed as

$$M = [\exp(-2A't) + \sigma_{\infty}^2 (1 - \exp(-B'A't))^2]^{\frac{1}{2}} \quad (4)$$

where

M = degree of mixedness,

σ_{∞} = standard deviation at infinite time,

A', B' = constants.

Faiman and Rippie (1965) have suggested that both mixing and demixing are of the first order processes and that the process may be considered analogous to a reversible steps which occur simultaneously. The model can be expressed as

$$\frac{dC_1}{dt} = -k_1 C_1 + k_2 C_2 \quad (5)$$

where

C_1 = concentration of mixed particles,

C_2 = concentration of unmixed particles,

k_1, k_2 = constants.

Rumpf and Muller (1967) proposed that solids mixing can be described by a simplified second-order Kologoroff equation of the form

$$\frac{\alpha C(x,t)}{\alpha t} = -\frac{\alpha}{\alpha x} T(x,t) C(x,t) + \frac{\alpha^2}{\alpha x^2} D(x,t) C(x,t) \quad (6)$$

where

$C(x,t)$ = concentration of the key particles,

$D(x,t)$ = diffusion coefficient,

$T(x,t)$ = convection coefficient.

Pramoda (1969) based on the first-order kinetics proposed by Faiman and Rippie (1965), analyzed the overall functional relationship between the physical properties of segregating particle systems and an experimentally observed rate constants, k_o . Their analysis has led to the following correlation:

$$k_o = \left[\left(\frac{d_p}{d_s} \right)^3 - 1 \right] \left\{ 1 + D^* \left[\left(\frac{d_p}{d_s} \right)^3 - 1 \right] \left(\frac{\rho_s - \rho_p}{\rho_s + \rho_p} \right) \left[A'' \exp(-B''/v_o^2) + A''' \exp(-B'''/v_o^2) \right] \right\} \quad (7)$$

where

d = diameter of the particle,

ρ = density of the particle,

$A'', A''', B'', B''', D^*, v_o$ = constants.

Shinohara et al. (1970, 1972) have proposed a "screen" model for the particle segregation in filling hoppers. The model considers the

sieving process of small particles through the interspaces of larger ones to be analogous to the flow mechanism of particles from a hopper having a definite opening diameter. Thus, the relative mass flow rate of small particles passing through the interspaces of large particles per unit flowing area can be written as

$$F_{sr}' = \frac{6(1 - \varepsilon')}{\pi d_p^2} (\alpha' D') \quad (8)$$

where

d = size of the particle,

ε' = void fraction of the particle,

α', D' = complex functions related to flow rate.

The first term in the right-hand side represents the number of opening per unit area and the second term represents the mass flow rate per opening.

Tanaka (1971) proposed a "push-away" model for the segregation of particles differing in size in central filling hoppers. The model is schematically represented in Fig. 3, where H and L denote heavy and light particles, respectively. Sphere 1, 2, and 3 can not move unless coefficient of friction, μ , is smaller than the critical value, μ_c

$$\mu_c = (\mu_1/\mu_2) \tan\theta / (\mu_1/\mu_2) \quad (9)$$

Since $\mu_{CA} > \mu_{CB} = \mu_{CC} > \mu_{CD}$, L of model A can travel farther to the ends of the wall; L of model B and H of model C can travel to the same point of the way; model D will be stagnant. As results of these actions, three zones of different compositions can be obtained.

Fan et al. (1975), assuming that mixing and demixing can be described independently, presented a mechanistic model as shown in Fig. 4. The segregated state of type I represents a metastable state, and if it is acted upon by a mixing device the system will be converted into the segregated state of type II, which is stable. The process to be modeled

was the transition from the segregated state of type I to the segregated state of type II, which was visualized as proceeding through an intermediate state (mixed state). The first step was assumed to be a first-order rate process, and the second step was assumed to be an autocatalytic mechanism. The model can be expressed as

$$M = 1 - \exp(-k_1 t) - \frac{\alpha^* \{ \exp[\beta^* (\alpha^* + \frac{\sigma_\infty^2}{\sigma_0^2}) t] - 1 \}}{1 + \alpha^* \frac{\sigma_\infty^2}{\sigma_0^2} \exp[\beta^* (1 + \frac{\sigma_\infty^2}{\sigma_0^2}) t]} \quad (10)$$

where

- M = degree of mixedness,
- σ_0 = standard deviation at $t = 0$,
- σ_∞ = standard deviation at $t = \infty$,
- k, α^*, β^* = rate constants.

Baranov and Vorobev (1973) have proposed the following equation for the mixing of segregating particle system in a drum mixer

$$- \frac{dV'}{dt} = A^* V' + B^* \quad (11)$$

where

- V' = coefficient of variation,
- A^*, B^* = constants.

Bridgwater (1974) has proposed a percolation model for the segregation of particles in a packing bed. In his model, percolating particles are assumed to be spherical and to be small compared with the packing particles. The motion of the percolating particles thus consists of rolling down the surface (represented by a flat plane) of a packing particle, followed by free flight to the next packing particles as shown in Fig. 5. The model can be expressed as

$$mgl \sin \beta = \frac{1}{2} m \left(\frac{dl}{dt} \right)^2 + \frac{1}{2} I \omega^2 \quad (12)$$

where

m = mass of percolating particle,

I = moment of inertia,

W = angular velocity.

The time a percolating particle spends on a plane can be obtained by integrating equation (12) and from which the mean time it spends in the bed can be obtained by taking inclination distribution of the plane into consideration. A similar model, which takes plane length distribution into account, has been considered by Davidson (1959).

Gibilaro and Rowe (1974) formulated a model based on four physical mechanisms in a fluidized bed (Fig. 6): overall particle circulation, interchange between wake and bulk phases, axial dispersion, and segregation. The model at steady state can be expressed as

$$\frac{D}{H} \frac{d^2 C_B}{dz^2} + (W' + K' - 2K' C_B) \frac{dC_B}{dz} + q H (C_W - C_B) = 0 \quad (13)$$

and

$$W' \frac{dC_W}{dz} - q H (C_B - C_W) = 0 \quad (14)$$

where

C_B = concentration of the jetsam in the bulk phase,

C_W = concentration of the jetsam in the wake phase,

D = diffusion coefficient,

K' = segregation constant,

H = dimensionless bed height,

q = exchange rate,

W' = circulation rate.

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Table 1. Summary of segregation models

Investigators	Proposed model	Basic expression	Application
Rose (1959)	Mixing rate can be described in terms of the so-called mixing and demixing potentials.	$M = \gamma \{ 1 - \beta / A^{\frac{1}{2}} (1 + \beta / A) \exp(-At/2) \}^2 \}$	Batch mixer in general.
Weydanz (1960)	Mixing and demixing occur simultaneously in the vertical direction while only mixing works in the horizontal direction.	$M = [\exp(-2A't) + \sigma_{\infty}^2 (1 - \exp(-B'A't))^2]^{\frac{1}{2}}$	Horizontal drum mixer.
Falman and Rippie (1965)	Mixing and segregation are first-order rate processes.	$\frac{dC_1}{dt} = -k_1 C_1 + k_2 C_2$	Particle systems subjected to agitation and vertical sine wave motion.
Rumpf and Muller (1967)	Solids mixing can be described by a simplified second-order Kologoroff equation.	$\frac{\partial C(x,t)}{\partial t} = -\frac{\alpha}{\partial x} [\pi(x,t) C(x,t)] + \frac{\alpha^2}{2} [D(x,t) C(x,t)]$	Batch mixer in general.
Pramoda (1969)	Mixing and segregation are first-order rate processes.	$k_0 = \left\{ \left(\frac{d}{\sigma_s} \right)^3 - 1 \right\} \left\{ 1 + D^* \left[\left(\frac{d}{\sigma_s} \right)^3 - 1 \right] \left(\frac{f_s - f_0}{f_s + f_0} \right) \right\} \\ \times [A'' \exp(-B''/v_0^2) + A''' \exp(-B'''/v_0^2)]$	Particle system subjected to shear and/or vibration.

Table 1. Continued

Investigators	Proposed model	Basic expression	Application
Shinohara et al. (1970, 1972)	Sieving process of small particles through the interspaces of larger ones could be simulated by the flow mechanism of particles from a hopper.	$P_{sr}' = \left[\frac{6(1 - \xi_d)}{\pi d_d^2} \right] (\alpha' D')$	Filling hopper.
Tanaka (1971)	The composition of segregating zones is determined by the push-away action of a two-dimensional 3 spheres model.	$\mu_c = (\mu_1/\mu_2) \tan \theta / (\mu_1/\mu_2)$	Filling hopper.
Gelves-Arocha (1973)	Mixing and demixing can be described independently. Mixing is assumed to be a first-order rate process and segregation is compatible with an autocatalytic mechanism.	$M = 1 - \exp(-k_1 t) - \frac{\alpha^* \left\{ \exp \left[\beta^* \left(\alpha^* + \frac{\sigma_{\infty}^2}{\sigma_0^2} \right) t \right] - 1 \right\}}{1 + \alpha^* \frac{\sigma_{\infty}^2}{\sigma_0^2} \exp \left[\beta^* \left(1 + \frac{\sigma_{\infty}^2}{\sigma_0^2} \right) t \right]}$	Kenics mixer or other motionless mixers.
Baranov and Vorobev (1973)	The dependence of mixing rate on the material non-uniformity is linear in character.	$-\frac{dv'}{dt} = A^* v' + B^*$	Horizontal drum mixer.

Table 1. Continued

Investigators	Proposed model	Basic expression	Application
Bridgwater (1974)	The motion of percolating particles consists of rolling down the surface of a packing particle followed by free flight to the next packing particle and so on.	$mg \sin \beta = \frac{1}{2} m \left(\frac{dl}{dt} \right)^2 + \frac{1}{2} I \omega^2$	Packed-bed mixer.
Gibilaro and Rowe (1974)	The motion of particles in fluidized bed can be described by three modes of mixing and a mode of segregation.	$\frac{D}{H} \frac{d^2 C_B}{dz^2} + (W' + K' - 2K' C_B) \frac{dC_B}{dz} + qH(C_W - C_B) = 0$ $W' \frac{dC_W}{dz} - qH(C_B - C_W) = 0$	Fluidized bed mixer.

2.6 REDUCING SEGREGATION

While several methods have been suggested for reducing segregation in the mixture, none of them can be said entirely satisfactory.

Willemsley (1960) reported that the density segregation can be counteracted by having the heavier particles larger. Bourne (1965) suggested that segregation can be overcome by some additional operations, e.g., grinding or granulation, before or during mixing, however, this adds to the cost of the whole operation. Johnson (1968) studied the effect of coating on the segregation tendency of particles. Silicon coating was found to increase the rate of segregation while rosin and magnesium stearate coatings to decrease the rate of segregation.

In many cases segregation can be reduced by introducing into mixtures an additive that will more seriously affect the process or end product. Hogg (1971) suggested that wet mixing may be helpful when particles are extremely troublesome in the dry state. Klein and Wilcox (1972) reported that the addition of deagglomerators can produce a powder with superior properties. Pieces of hard rubber were found to be the most successful deagglomerator in the drum mixer while a rapidly revolving bar was most successful in the twin-shell mixer.

Fischer (1960) reported that radial segregation in a mixer can be prevented if the particles were charged in alternate layer instead of completely separated. Donald and Roseman (1962b) analyzed the effect of loading on the segregation in a horizontal drum mixer and concluded that the segregation can be reduced by increasing the speed of the mixer. Williams and Kahn (1973) also draw the same conclusion from their study of a inclined drum mixer. Lloyd et al. (1970) reported that at a properly selected speed a "pseudo-random" mixture can be achieved in a horizontal

drum mixer.

Hersey (1975) introduced the ordered mixing concept. The random distribution mechanisms are destroyed by the ordered mixing but this can be beneficial when segregation is a problem. Ordered mixtures can be achieved through particle interactions, e.g., adsorption, chemisorption, surface tension, frictional, electrostatic or any other form of adhesion.

2.7 SELECTION OF A MIXER

Homogeneity of a mixture which can be obtained depends primarily on the choice of a mixer; a suitable mixer may produce reasonably good mixtures even for components having strong segregation tendency while an unsuitable mixer may produce poor mixtures for components which segregate only slightly.

Solids mixers are available in a wide variety of design but potential users often have little quantitative data on which to base their selection. Ashton and Valentin (1966) and Miles (1970) have shown that most industrially available mixers can ultimately produce a mixture close to random for nonsegregating particle systems. This is however not true for segregating particle systems. The quality of the final mixture is determined by the mixing mechanisms in the mixer employed.

Three principal mechanisms are known to be active in the solids mixing: diffusive, shear and convective mixings (Lacey, 1954). It has generally been believed that the convective mixing is the one which minimizes the segregation (Harnby, 1967). In the cases where the diffusive mixing prevails, the segregation tends to be severe. Segregation caused by the shear mixing is relatively minor but can become important if bulk

density of a mixture is reduced. Williams (1968) classified mixers into two groups: segregating mixers, which rely mainly on the diffusive or the shear mixing, and nonsegregating mixers or, strictly speaking, less-segregating mixers, which rely mainly on the convective mixing.

Horizontal drum mixers are typical segregating mixers. Performance of such mixers can often be improved by fitting baffles or deflectors (see e.g., Hill, 1965) to elevate the charge pass a normal flow angle so that particles are thrown and do not roll down the surface. Donald and Roseman (1962b) concluded that the design of a tumbling mixer should allow frequent change in the particle path direction and had no tendency to set up axial gradient. These two points of mixer design have been incorporated into both the Y-shaped and the diamond mixer. Williams (1968) reported that the improvement can also be made by means of a shaft with projecting vanes, fitted along axis of a drum mixer.

Nonsegregating mixers included ribbon blender, nauta mixer, lodge mixer, etc. In the ribbon blender there is some segregation due to the rolling of particles down the inclined surface, and some due to the shearing, but main mixing action is the convection. The nauta mixer and the mixers with mechanical stirrers, such as the lodge mixer, are designed to produce the convection within the body of particles rather than the shear.

Harnby (1967) compared the performance of four types of mixers for segregating particles. The impeller type mixer showed a superiority over the others in regard to both the efficiency and the quality of mixtures. The results can be explained by the mixing mechanisms in the mixers as shown in Table 2. Lynch and Ho (1972) compared rotating-shell mixers with fixed-shell rotating agitator type mixers and made general recommendations for selection.

King (1964) proposed a multiple mixer system for alleviating the segregation problems. The major mixing of this system is carried out in larger batches and a small final mixer is used to eliminate the segregation which may have risen during transportation of the mixture. Cook et al. (1974) evaluated a nauta mixer for preparing multicomponent mixtures. The results indicate the difficulties may exist with multicomponent mixtures that each of the components behaves independently, each approaching the required specification at different rates.

2.8 MIXING OF SEGREGATING PARTICLE SYSTEMS IN CONTINUOUS MIXERS

Williams and Rahman (1971) concluded, "Though many papers dealing with the batch mixing of particles have appeared there seems to be little movement toward the study of continuous mixing systems involving particles."

Poole et al. (1965) reported experimental data on the mixing of metal powders with ceramic oxides in a continuous ribbon bladed mixer. Sugimoto et al. (1966) studied the phenomenon of segregation in a continuous horizontal drum mixer and proposed a model to predict the period and the scale of fluctuation in the discharge rate and also the composition of different segregation zones. On another occasion, Sugimoto et al. (1967) argued that small and large particles might have quite different residence time distributions in the continuous drum mixer and substantiated their argument by the experimental investigation. Williams and Rahman (1970) studied the effect of operating variables on the residence time distribution in a continuous inclined drum mixer. They concluded that in many cases the undesirable segregation can be reduced by carefully controlling the operating conditions. Harwood et al. (1975) evaluated several industrial continuous mixers for dry powders. They concluded

that a desired mixer was the one which develops continuously the convection current without creating a free-fall surface across which segregation can take place.

2.9 MIXING OF SEGREGATING PARTICLE SYSTEMS IN FLUIDIZED BEDS

While gas fluidized beds have been known as excellent mixers for nonsegregating particle systems, relatively few attempts have been made to evaluate them as mixers for segregating particle systems.

Leva (1962) reported that the segregation in a fluidized bed can be reduced by introducing a stirrer or by carefully shaping the bed. Nicholson and Smith (1966) studied the axial mixing of particles differing in density in a fluidized bed and proposed a first-order rate equation to describe the progress of mixing in the short mixing time. Nienow et al. (1972) reported the role of particle size, particle density, and air rate on the segregation behavior in a gas fluidized bed. They concluded that a fairly wide particle size difference can be tolerated without appreciable segregation while even a small density difference led to readily settling of the denser particles. Gibilaro and Rowe (1974) formulated a model based on four physical mechanisms: overall particle circulation, interchange between wake and bulk phases, axial dispersion, and segregation. Experimentally observed equilibrium concentration distributions can be satisfactorily predicted from this model.

2.10 CONCLUDING REMARKS

Mechanisms of segregation are closely related to those which give rise to the diffusive mixing. Both processes are favored by the conditions which lead to maximum opportunity for particles to move as individuals. Thus, for materials which tend to segregate, it may be necessary to sacrifice some advantages of the diffusive mixing in order to achieve acceptable final mixtures.

Batch mixing of segregating particle systems has long been a problem. Segregation occurs not only during the mixing but also during the subsequent handling. The possibility of using a continuous mixing system should be considered to eliminate the intermediate storage and minimize the subsequent handling. Such a system should include not only a mixer (or a number of mixers) but also a feeder, a storage vessel or vessels, a recycle loop, and other auxiliary units.

Since there is no easy way to relate the particle characteristics with the mixer performance at this stage, there is no substitute for experimental investigation. Only by trial and error can one select a suitable mixer and establish an optimum operation to obtain the best possible homogeneity.

According to Vogel (1965) and Matthee (1967), the still unknown general differential equation of mixing and segregation may be formulated by determining the functional relationship among dimensionless groups. Much work needs be done before it will be possible to identify all the relevant dimensionless groups and the form of the interrelation among them.

TABLE 1. Role of mechanisms involved in various mixers (Harnby, 1967).

Type of mixer	Mechanisms		
	Rolling or Diffusive	Shear	Convective
Rotocube (without impeller) (with impeller)	Major	Minor	—
	Major	Minor	Minor
V-Mixer	Major	Minor	—
Nautamix	—	Minor	Major
Ribbon blender	—	Major	Major

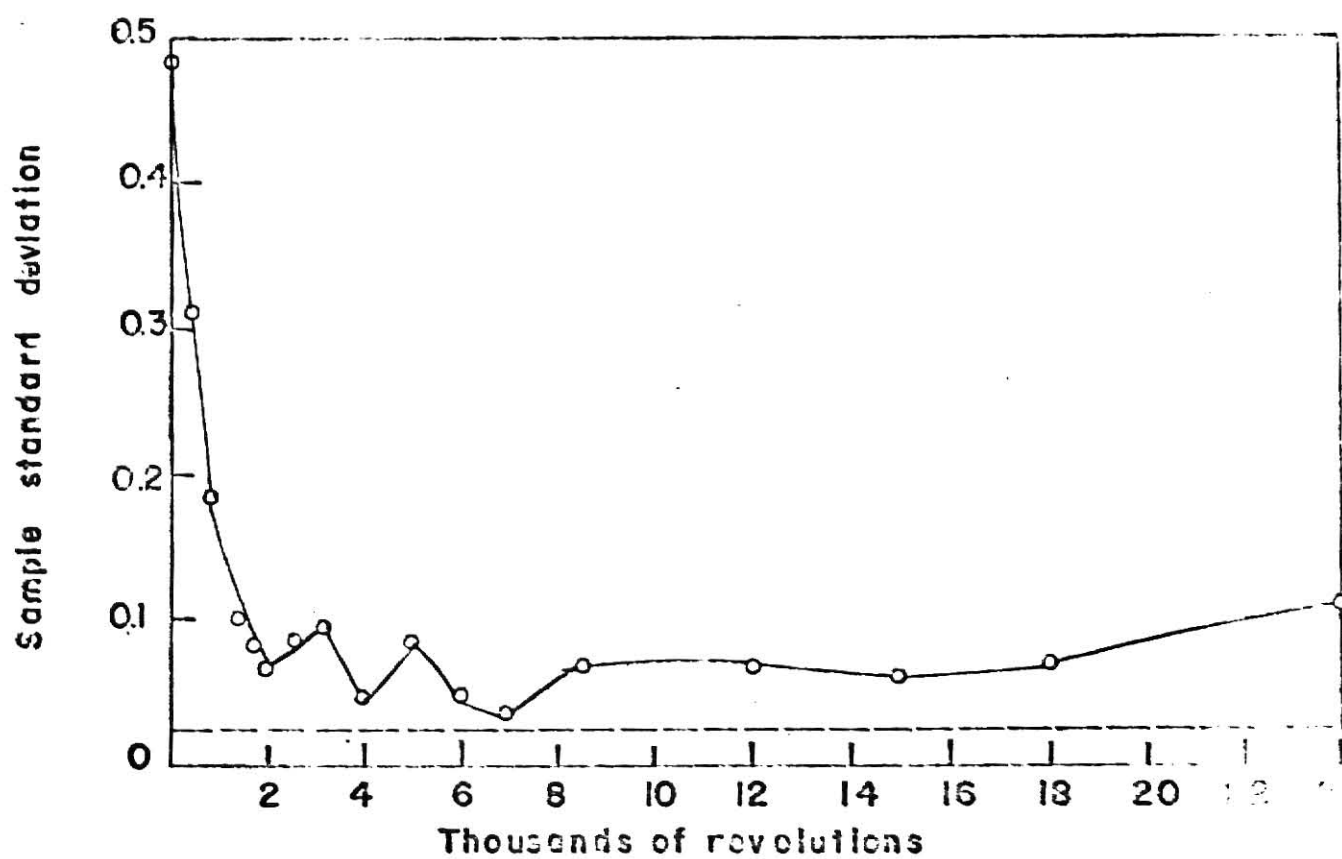


Fig.1 Typical example of mixing - demixing v.s. time
(Weidenbaum et al., 1955).

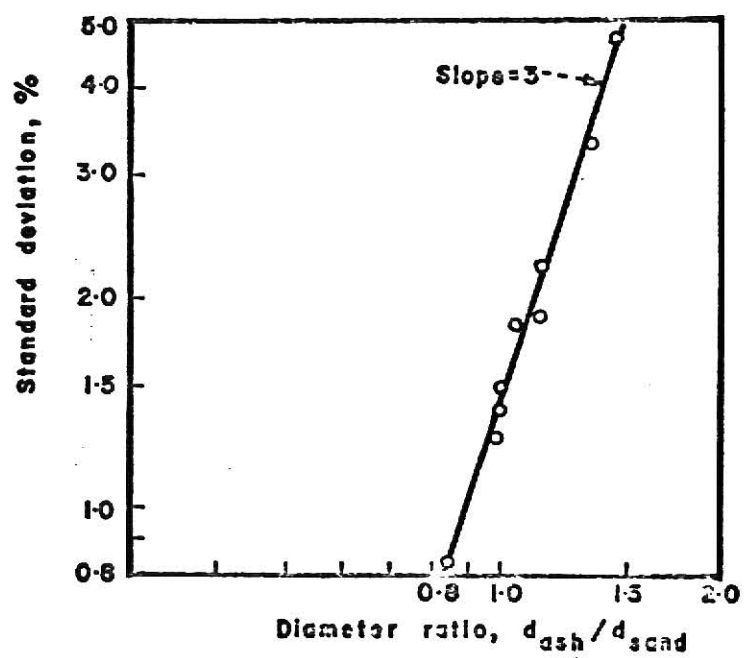


Fig. 2 Effect of size variations of soda ash-sand mix on the mixture homogeneity (Campbell et al., 1966).

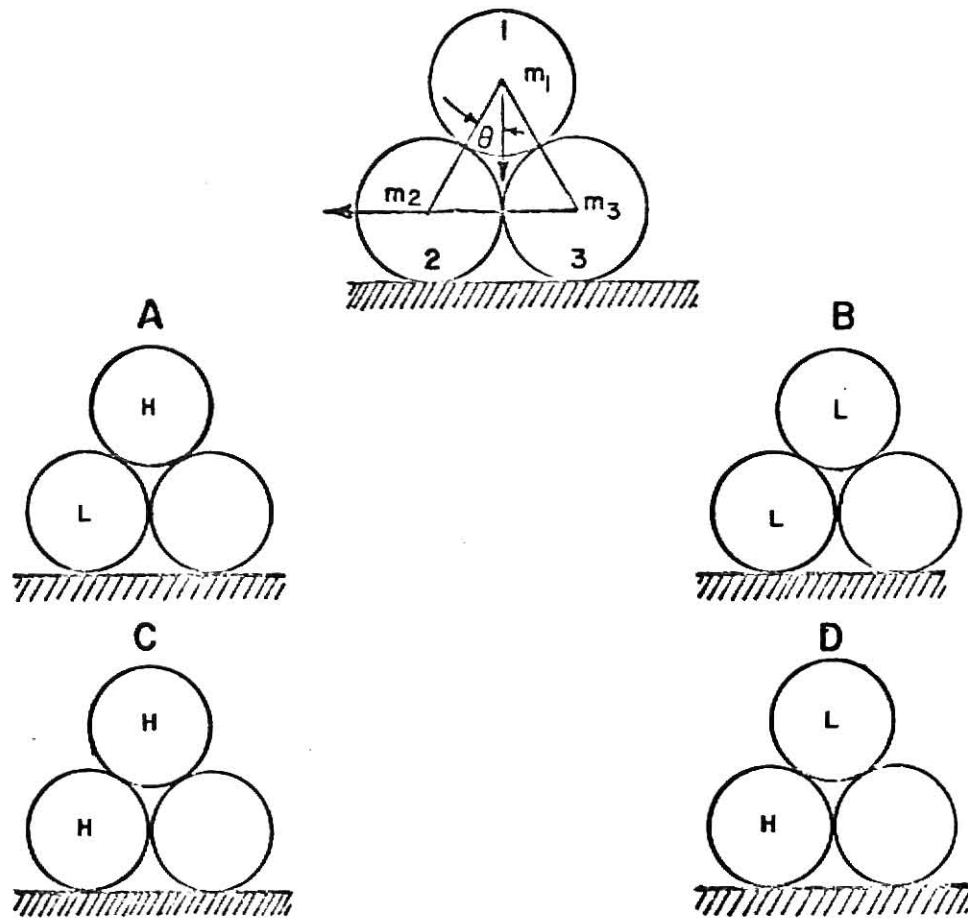


Fig.3 Typical two-dimensional model of combination of three spheres of equal diameter and possible combinations of heavy and light components (Tanaka, 1971).

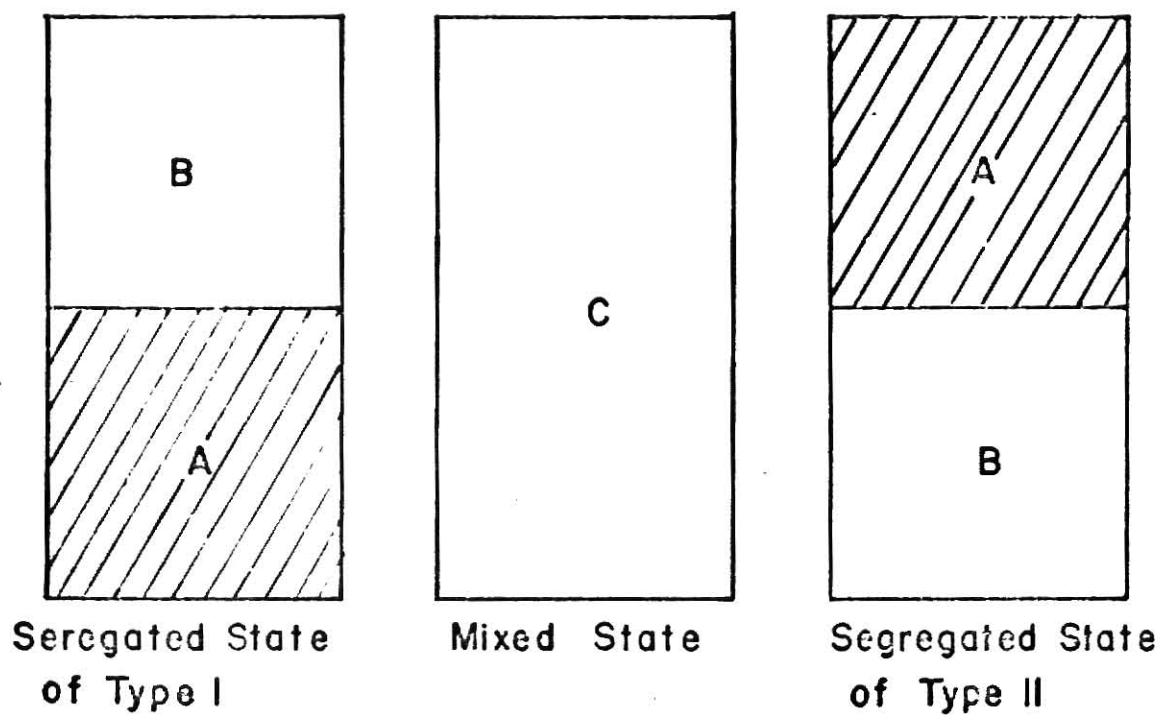


Fig.4 Representation of a Semi-Emperical Kinetic Model (Gelves-Arocha, 1973).

A = Small or light particles
B = Big or heavy particles
C= Mixture of A and B

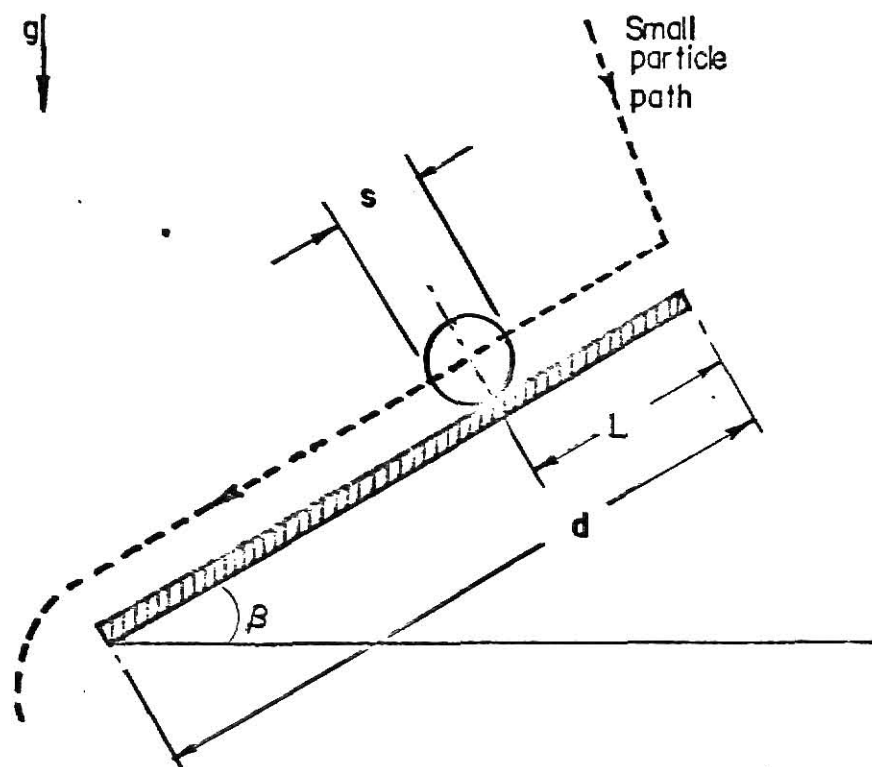


Fig.5 Particle motion on a plane (Bridgewater, 1974).

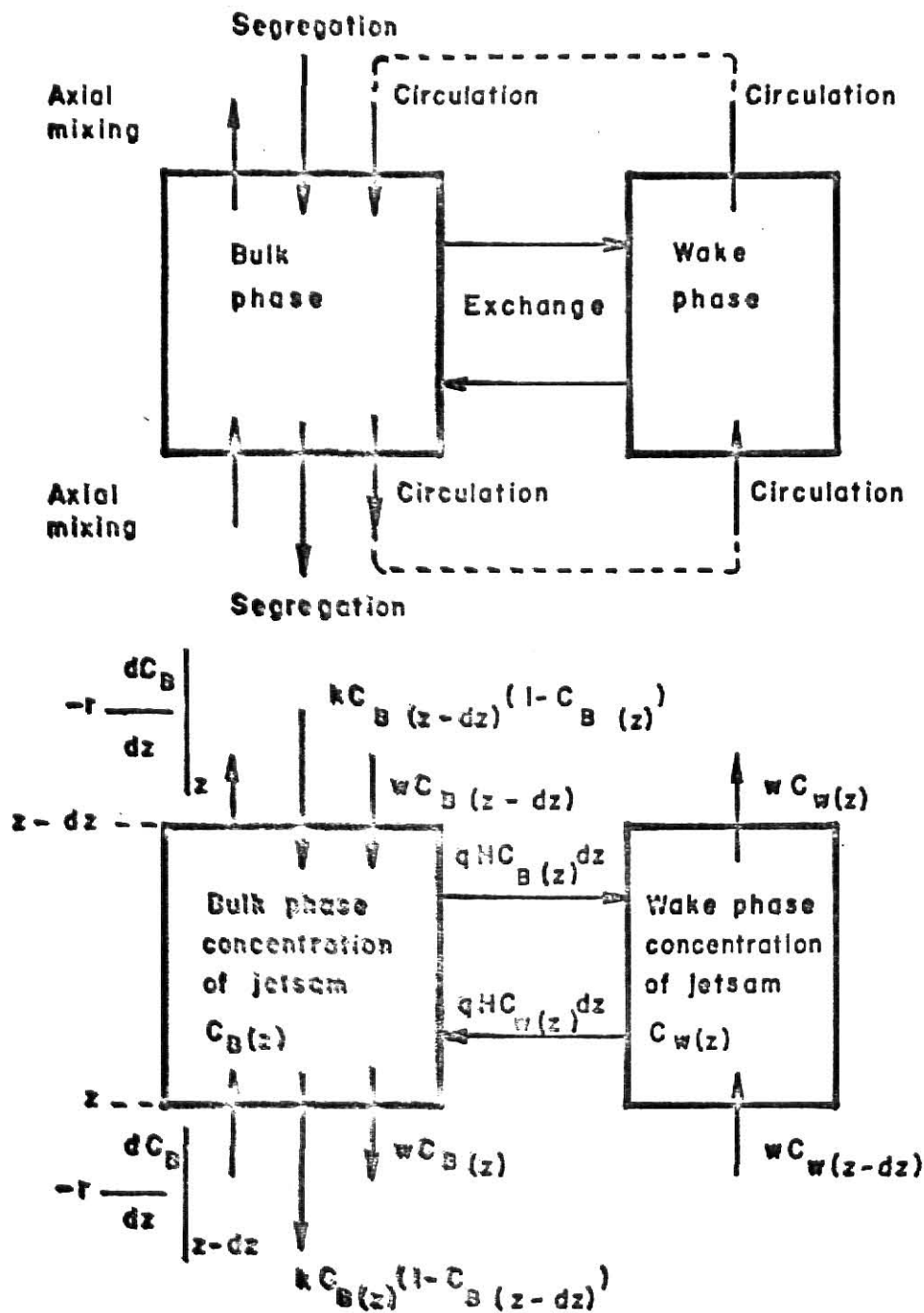


Fig.6. A model for a segregating gas fluidised bed (Gibilaro et al, 1974)

NOTATION

A, A', A'', A''', A^*	= constants
B, B', B'', B''', B^*	= constants
C	= concentration
\bar{C}	= mean concentration
$D(x, t)$	= diffusion coefficient
D^*, D'	= constants
d	= diameter of the particle
F_{sr}'	= relative mass flow rate of small particles passing through the interspaces of large particles per unit flowing area
g	= gravity
H	= total bed height
I	= moment of inertia
i	= total number of push away actions
K	= equilibrium constant
K'	= constant
k_o	= observed rate constant
k_1	= mixing rate constant
k_2	= demixing rate constant
k'	= constant
L	= length of the plate
l	= large particle
M	= degree of mixedness
N	= number fraction
n	= number of planes the percolating particles rolls down
q	= rate of exchange

s	= small particle
S	= cross-sectional area of a hopper
$T(x, t)$	= convection coefficient
t	= time
V'	= coefficient of variation
W	= angular velocity
W'	= circulation rate

Greek Letter

ρ	= density of the particle
ϵ'	= void fraction
β	= inclination of the plate
μ	= coefficient of friction
ψ	= demixing potential
η	= efficiency factor
σ	= standard deviation
σ_0	= standard deviation at $t=0$

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CHAPTER 3
A MODIFIED COALESCENCE-DISPERSION MODEL
FOR THE AXIAL MIXING OF SEGREGATING PARTICLES
IN A MOTIONLESS MIXER

3.1 INTRODUCTION

Solids mixing is a common processing operation widely used in a variety of industries. It is extensively employed in the manufacture of ceramics, fertilizers, detergents, glass, pharmaceuticals and animal feeds, and in the powder metallurgy industry.

Three principal mechanisms are known for the process of solids mixing: convective, diffusive and shear (Lacey, 1954). However, the extent of mixedness as a function of time is generally difficult to predict because satisfactory theories capable of describing the mechanisms have not been fully developed.

The phenomenon of solids mixing has been analyzed based on essentially linear diffusion models (see e.g., Lacey, 1954; Otake et al., 1961; Hogg et al., 1966; Chen et al., 1973). Such a simple approach is applicable only to nonsegregating particle systems, i.e., highly idealized systems.

The behavior of systems where the particles differ in properties and characteristics (segregating particle systems) is a matter of practical concern in industrial operations. The occurrence of demixing (segregation) is extremely important, since it can markedly alter product quality. Despite this importance, the simultaneous occurrence of mixing and demixing in segregating particle systems has received relatively little attention.

Some attempts have been made to formulate mixing rate expressions for segregating particle systems. Rose (1959) derived his expression by incorporating the so-called mixing and demixing potentials into a regular diffusion expression. While his model has been successfully employed in representing some experimental data, definition of the demixing potential is conceptually difficult to accept. It was pointed out (Danckerts, 1959) that the estimated parameters in the model are inconsistent with the theory behind the model. Weydanz (1960) proposed a model for a two-dimensional process in which the segregation effect is present in only one dimension. The main assumptions were that the rate of segregation is constant and that the volume interchanged in either direction is constant. Neither of these assumptions can be expected to be valid in anything other than highly limiting cases. Faiman and Rippie (1965) suggested that both mixing and demixing are first order processes and that the process may be considered analogous to a reversible chemical reaction in which both forward and reversible steps occur simultaneously. The rate expression thus derived can only describe the later stage of a mixing and demixing process. Fan et al. (1975), assuming that mixing and demixing can be described independently, presented a mechanistic model which is analogous to chemical kinetics of two complex reactions in series. The model can describe mixing curves (degree of mixedness vs. time plots) to some extent, however, it gives no concentration distributions which are extremely important in the design of multiple mixers and continuous mixing systems.

A modified coalescence-dispersion model has been developed in the present study. The concept of coalescence-dispersion was introduced by

Curl(1963) and has been employed by many investigators to study the effect of dispersed phase mixing on various reactions (see e.g., Spielman and Levenspiel, 1965; Kattan, 1967; Komasaawa et al., 1971; Zeitlin et al., 1972; Patterson, 1973). The validity of the present model has been tested against the available data (Gelves-Arocha, 1973). Relationships between parameters of the proposed model and physical properties of the system have been analyzed.

3.2 DESCRIPTION OF THE MODEL

In the present model, the entire length of the mixer is divided equally into a finite number of sections in series in the vertical direction. Each section in turn is divided equally into a finite number of cells in the horizontal direction. Each cell contains a specified number of particles. Schematic representation of the cells and sections is given in Fig. 1.

Mixing particles of the same size which may or may not have other similar characteristics is considered first. The following mixing mechanisms are assumed (see Fig. 2):

- (1) The convective mixing in the axial direction is induced by allowing random coalescence between cells in the adjacent sections, namely the i -th and the $(i+1)$ -th sections, two at a time. No coalescence is possible between cells in the same section. The number of particles in each cell and the number of coalescences per unit time (coalescence rate) are invariant with respect to time and position.
- (2) The diffusive mixing mechanism is induced by collision and redistribution of individual particles between coalescing cells.

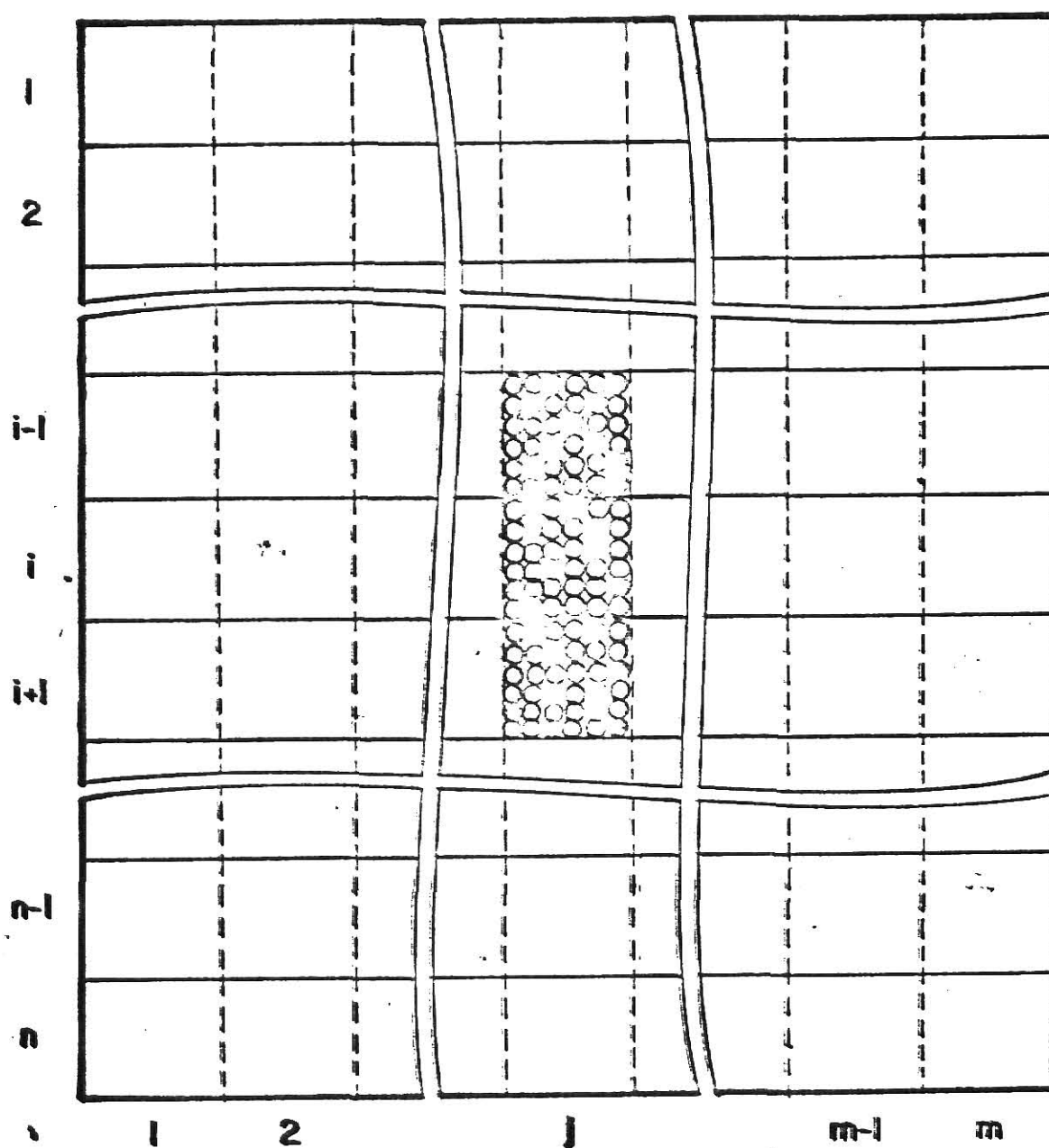


Fig.1. Schematic representation of sections, cells, and particles in the mixer.

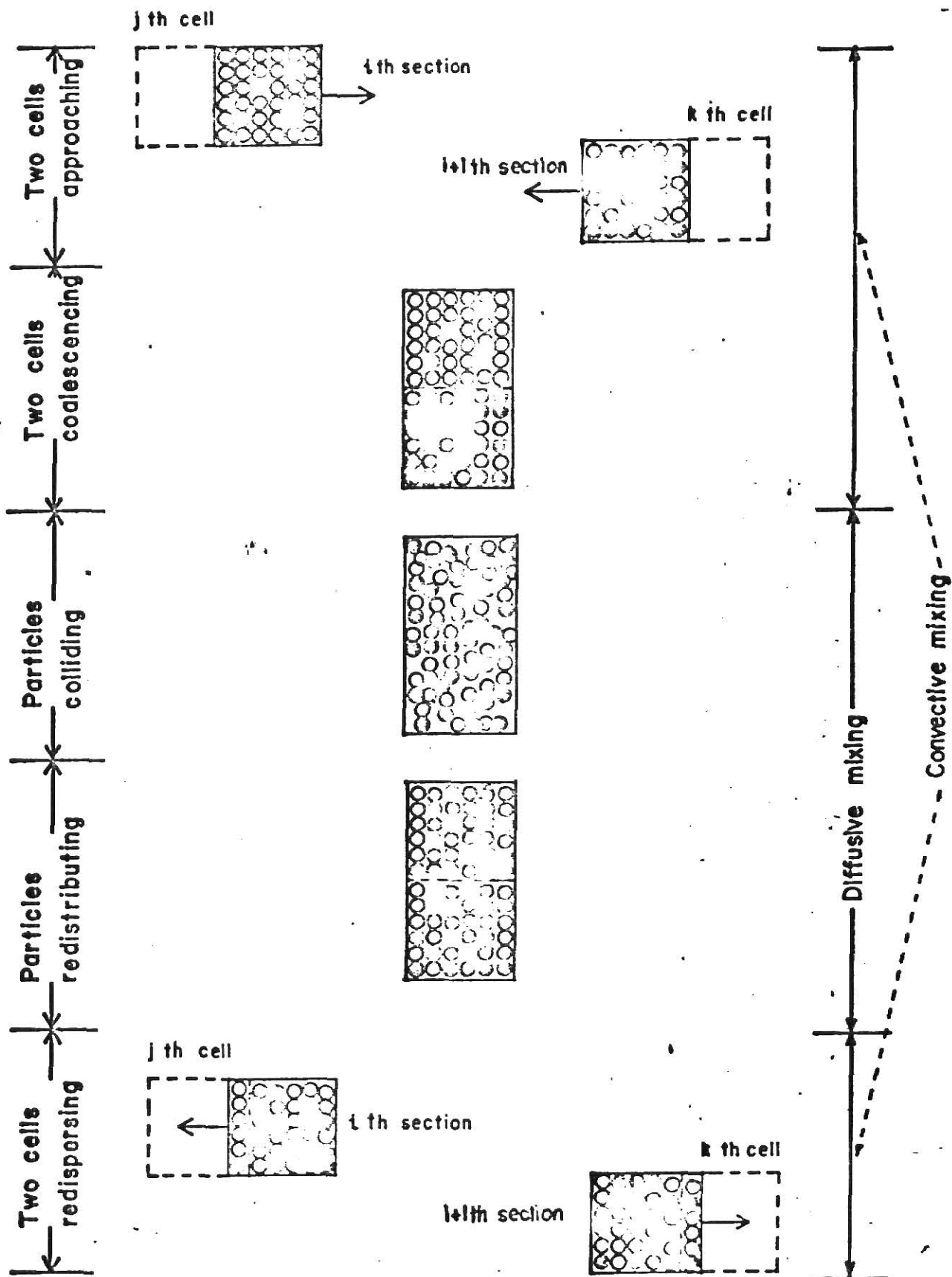


Fig.2. Representation of the visualized mechanism of mixing upon a coalescence.

Upon each coalescence, particles in one coalescing cell experience one-to-one random collisions with particles in the other coalescing cell and redistribute themselves instantaneously between the two coalescing cells. In the case of a key-nonkey particle collision, the key particle may find its new position in the coalescing cell either in the i -th section or in the $(i+1)$ -th section. The probability of the key particle to find its new position in the i -th section is denoted by α , and consequently that in the $(i+1)$ -th section is denoted by $(1-\alpha)$. The magnitude of the distribution ratio α is mainly dependent on physical characteristics of the particles. When the key and nonkey particles are identical in every aspect except color, α is equal to 0.5.

- (3) The two coalescing cells are redispersed back into their original positions in the respective sections.

Based on these assumptions, for an arbitrarily small interval Δt , we have (see Appendix 1)

The number concentration of key particles in cell j in the i -th section at $t + \Delta t$ provided that the coalescence is with cell k in the $(i+1)$ -th section

$$= [C_{i,j}(t) C_{i+1,k}(t) + \{[1 - C_{i,j}(t)] C_{i+1,k}(t) + C_{i,j}(t) [1 - C_{i+1,k}(t)]\} \times \alpha] U \Delta t + C_{i,j}(t) (1 - U \Delta t) \quad (1)$$

The number concentration of key particles in cell k in the $(i+1)$ -th section at $t + \Delta t$ provided that the coalescence is with cell j in the i -th section

$$= [C_{i,j}(t) C_{i+1,k}(t) + \{[1 - C_{i,j}(t)] C_{i+1,k}(t) + C_{i,j}(t) [1 - C_{i+1,k}(t)]\} \times (1 - \alpha)] U \Delta t + C_{i+1,k}(t) (1 - U \Delta t), \quad i = 1, 2, \dots, n-1 \quad (2)$$

$j, k = 1, 2, \dots, m$

where

$C_{i,j}(t)$ = number concentration of key particles in cell j in the i -th section at t ,

$C_{i+1,k}(t)$ = number concentration of key particles in cell k in the $(i+1)$ -th section at t ,

U = number of coalescences per unit time.

Mixing particles of different sizes is complicated by the fact that the total number of particles in a cell is a function of the composition and the structure (packing) of the mixture in that cell. The great difficulty involved in such systems can be overcome by assuming that replacement of a key particle lost from the cell can be accomplished either by another key particle or a number of nonkey particles provided that the key particles are larger than the nonkey particles. Under this circumstance, the assumption of one-to-one particle collision is no longer valid unless a certain number of the nonkey particles is treated as an aggregate, which behaves as if it is a single particle. For this purpose, the apparent number concentration of key particles in cell j in the i -th section is defined as

$$C_{i,j}(t)_a = \frac{X_{i,j}(t)}{X_{i,j}(t) + \frac{Y_{i,j}(t)}{r}}, \quad \begin{array}{l} i = 1, 2, \dots, n \\ j = 1, 2, \dots, m \end{array} \quad (3)$$

in which

$X_{i,j}(t)$ = number of key particles in cell j in the i -th section,

$Y_{i,j}(t)$ = number of nonkey particles in cell j in the i -th section,

r = bulk volume ratio of a key particle to a nonkey particle.

With this definition, Equations (1) and (2) are applicable to the mixing of different size particles.

3.3 SYSTEM EQUATIONS

The probability that cell j in the first (top) section and cell k in the second section are selected as the two cells of a coalescing pair (see Appendix 2)

$$= \frac{3}{2 n m^2}, \quad j, k = 1, 2, \dots, m \quad (4)$$

The probability that cell j in the i -th section and cell k in the $(i+1)$ -th section are selected as the two cells of the coalescing pair (see Appendix 2)

$$= \frac{1}{n m^2}, \quad i = 2, 3, \dots, n-2 \quad (5)$$

$$j, k = 1, 2, \dots, m$$

The probability that cell j in the $(n-1)$ -th section and cell k in the n -th (bottom) section are selected as the two cells of the coalescing pair

= the probability that cell j in the first section and cell k in the second section are selected as the two cells of the coalescing pair

$$= \frac{3}{2 n m^2}, \quad j, k = 1, 2, \dots, m \quad (6)$$

The change in the number concentration of key particles in cell j in the i -th section during Δt due to the coalescence provided that the coalescence is with cell k in the $(i+1)$ -th section

$$= [C_{i,j}(t) C_{i+1,k}(t) + \{[1 - C_{i,j}(t)] C_{i+1,k}(t) + C_{i,j}(t) [1 - C_{i+1,k}(t)]\} \\ \times \alpha] U \Delta t + C_{i,j}(t) (1 - U \Delta t) - C_{i,j}(t), \quad (7)$$

$$i = 1, 2, \dots, n-1$$

$$j, k = 1, 2, \dots, m$$

The change in the number concentration of key particles in cell j in the i -th section during Δt due to the coalescence provided that the coalescence is with cell k in the $(i-1)$ -th section

$$= [C_{i,j}(t) C_{i+1,k}(t) + \{[1 - C_{i,j}(t)] C_{i+1,k}(t) + C_{i,j}(t) [1 - C_{i+1,k}(t)]\} \times (1 - \alpha)] U \Delta t + C_{i,j}(t) (1 - U \Delta t) - C_{i,j}(t), \quad (8)$$

$$i = 2, 3, \dots, n$$

$$j, k = 1, 2, \dots, m$$

The expected change in the number concentration of key particles in cell j in the first section during Δt

$$\begin{aligned} &= E \{C_{1,j}(t+\Delta t) - C_{1,j}(t)\} \\ &= \frac{3}{2 n m^2} \sum_{k=1}^m \left\{ [C_{1,j}(t) C_{2,k}(t) + \{[1 - C_{1,j}(t)] C_{2,k}(t) + C_{1,j}(t) \times [1 - C_{2,k}(t)]\} \alpha] U \Delta t + C_{1,j}(t) (1 - U \Delta t) - C_{1,j}(t) \right\} \\ &= \frac{3 U \Delta t}{2 n m^2} \sum_{k=1}^m [C_{1,j}(t) C_{2,k}(t) + \{[1 - C_{1,j}(t)] C_{2,k}(t) + C_{1,j}(t) \times [1 - C_{2,k}(t)]\} \alpha - C_{1,j}(t)], \quad j = 1, 2, \dots, m \end{aligned} \quad (9)$$

The expected change in the number concentration of key particles in cell j in the second section during Δt

$$\begin{aligned} &= E \{C_{2,j}(t+\Delta t) - C_{2,j}(t)\} \\ &= \frac{3 U \Delta t}{2 n m^2} \sum_{k=1}^m [C_{2,j}(t) C_{1,k}(t) + \{[1 - C_{2,j}(t)] C_{1,k}(t) + C_{2,j}(t) \times [1 - C_{1,k}(t)]\} (1 - \alpha) - C_{2,j}(t)] + \frac{U \Delta t}{n m^2} \sum_{k=1}^m [C_{2,j}(t) C_{3,k}(t) + \{[1 - C_{2,j}(t)] C_{3,k}(t) + C_{2,j}(t) [1 - C_{3,k}(t)]\} \alpha - C_{2,j}(t)], \quad (10) \\ & \quad j = 1, 2, \dots, m \end{aligned}$$

The expected change in the number concentration of key particles in cell j

in the i -th section during Δt

$$\begin{aligned}
 &= E \{C_{i,j}(t+\Delta t) - C_{i,j}(t)\} \\
 &= \frac{U \Delta t}{n m^2} \sum_{k=1}^m \left[C_{i,j}(t) C_{i-1,k}(t) + \{[1 - C_{i,j}(t)] C_{i-1,k}(t) + C_{i,j}(t) \right. \\
 &\quad \times [1 - C_{i-1,k}(t)]\} (1 - \alpha) - C_{i,j}(t) + C_{i,j}(t) C_{i+1,k}(t) + \{[1 - C_{i,j}(t)] \\
 &\quad \times C_{i+1,k}(t) + C_{i,j}(t) [1 - C_{i+1,k}(t)]\} \alpha - C_{i,j}(t) \Big] , \quad (11) \\
 &\quad i = 3, 4, \dots, n-2 \\
 &\quad j = 1, 2, \dots, m
 \end{aligned}$$

The expected change in the number concentration of key particles in cell j in the $(n-1)$ -th section during Δt

$$\begin{aligned}
 &= E \{C_{n-1,j}(t+\Delta t) - C_{n-1,j}(t)\} \\
 &= \frac{U \Delta t}{n m^2} \sum_{k=1}^m \left[C_{n-1,j}(t) C_{n-2,k}(t) + \{[1 - C_{n-1,j}(t)] C_{n-2,k}(t) + C_{n-1,j}(t) \right. \\
 &\quad \times [1 - C_{n-2,k}(t)]\} (1 - \alpha) - C_{n-1,j}(t) \Big] + \frac{3 U \Delta t}{2 n m^2} \sum_{k=1}^m \left[C_{n-1,j}(t) C_{n,k}(t) \right. \\
 &\quad \left. + \{[1 - C_{n-1,j}(t)] C_{n,k}(t) + C_{n-1,j}(t) [1 - C_{n,k}(t)]\} \alpha - C_{n-1,j}(t) \right] , \quad (12) \\
 &\quad j = 1, 2, \dots, m
 \end{aligned}$$

The expected change in the number concentration of key particles in cell j in the n -th section during Δt

$$\begin{aligned}
 &= E \{C_{n,j}(t+\Delta t) - C_{n,j}(t)\} \\
 &= \frac{3 U \Delta t}{2 n m^2} \sum_{k=1}^m \left[C_{n,j}(t) C_{n-1,k}(t) + \{[1 - C_{n,j}(t)] C_{n-1,k}(t) + C_{n,j}(t) \right. \\
 &\quad \times [1 - C_{n-1,k}(t)]\} (1 - \alpha) - C_{n,j}(t) \Big] , \quad j = 1, 2, \dots, m \quad (13)
 \end{aligned}$$

Dividing Equation (9) by Δt , and setting $\Delta t \rightarrow 0$, we obtain

$$E \left\{ \frac{dC_{1,j}(t)}{dt} \right\}$$

$$= \frac{3U}{2nm^2} \sum_{k=1}^m \left[c_{1,j}(t) c_{2,k}(t) + \{[1 - c_{1,j}(t)] c_{2,k}(t) + c_{1,j}(t) \times [1 - c_{2,k}(t)]\} \alpha - c_{1,j}(t) \right], \quad j = 1, 2, \dots, m \quad (14)$$

Similarly,

$$\begin{aligned} & E \left\{ \frac{dc_{2,j}(t)}{dt} \right\} \\ &= \frac{3U}{2nm^2} \sum_{k=1}^m \left[c_{2,j}(t) c_{1,k}(t) + \{[1 - c_{2,j}(t)] c_{1,k}(t) + c_{2,j}(t) \times [1 - c_{1,k}(t)]\} (1 - \alpha) - c_{2,j}(t) \right] + \frac{U}{nm^2} \sum_{k=1}^m \left[c_{2,j}(t) c_{3,k}(t) \right. \\ & \quad \left. + \{[1 - c_{2,j}(t)] c_{3,k}(t) + c_{2,j}(t) [1 - c_{3,k}(t)]\} \alpha - c_{2,j}(t) \right], \quad (15) \\ & \quad j = 1, 2, \dots, m \end{aligned}$$

$$\begin{aligned} & E \left\{ \frac{dc_{i,j}(t)}{dt} \right\} \\ &= \frac{U}{nm^2} \sum_{k=1}^m \left[c_{i,j}(t) c_{i-1,k}(t) + \{[1 - c_{i,j}(t)] c_{i-1,k}(t) + c_{i,j}(t) \times [1 - c_{i-1,k}(t)]\} (1 - \alpha) - c_{i,j}(t) + c_{i,j}(t) c_{i+1,k}(t) \right. \\ & \quad \left. + \{[1 - c_{i,j}(t)] c_{i+1,k}(t) + c_{i,j}(t) [1 - c_{i+1,k}(t)]\} \alpha - c_{i,j}(t) \right], \quad (16) \end{aligned}$$

$$i = 3, 4, \dots, n-2$$

$$j = 1, 2, \dots, m$$

$$\begin{aligned} & E \left\{ \frac{dc_{n-1,j}(t)}{dt} \right\} \\ &= \frac{U}{nm^2} \sum_{k=1}^m \left[c_{n-1,j}(t) c_{n-2,k}(t) + \{[1 - c_{n-1,j}(t)] c_{n-2,k}(t) + c_{n-1,j}(t) \right. \end{aligned}$$

$$\begin{aligned}
& \times [1 - C_{n-2,k}(t)] \} (1 - \alpha) - C_{n-1,j}(t) \Big] + \frac{3U}{2nm^2} \sum_{k=1}^m \left[C_{n-1,j}(t) C_{n,k}(t) \right. \\
& + \{ [1 - C_{n-1,j}(t)] C_{n,k}(t) + C_{n-1,j}(t) [1 - C_{n,k}(t)] \} \alpha \\
& \left. - C_{n-1,j}(t) \right], \quad j = 1, 2, \dots, m \quad (17)
\end{aligned}$$

$$\begin{aligned}
& E \left\{ \frac{dC_{n,j}(t)}{dt} \right\} \\
& = \frac{3U}{2nm^2} \sum_{k=1}^m \left[C_{n,j}(t) C_{n-1,k}(t) + \{ [1 - C_{n,j}(t)] C_{n-1,k}(t) + C_{n,j}(t) \right. \\
& \left. \times [1 - C_{n-1,k}(t)] \} (1 - \alpha) - C_{n,j}(t) \right], \quad j = 1, 2, \dots, m \quad (18)
\end{aligned}$$

Equations (14) through (18) can be solved numerically to give the number concentration of key particles in each cell. The number concentration of key particles in each section can be calculated by averaging all the cells in the section. When the model Equations (14) through (18) is applied to a mixer operated in a semi-batch mode, e.g., repeatedly passing a mixture through a motionless mixer, the time of mixing can be considered to be proportional to an integer number of passes when end effects are negligibly small.

A more convenient discrete time approach is described in the next section.

3.4 MONTE CARLO SIMULATION

When Δt equals to $1/U$ (i.e., period of a coalescence), Equations (1) and (2) reduce to, respectively,

$$\begin{aligned} & C_{i,j}(t + \frac{1}{U}) \\ &= C_{i,j}(t) C_{i+1,k}(t) + \{[1 - C_{i,j}(t)] C_{i+1,k}(t) + C_{i,j}(t) \\ & \quad \times [1 - C_{i+1,k}(t)]\} \alpha, \end{aligned} \quad (19)$$

and

$$\begin{aligned} & C_{i+1,k}(t + \frac{1}{U}) \\ &= C_{i,j}(t) C_{i+1,k}(t) + \{[1 - C_{i,j}(t)] C_{i+1,k}(t) + C_{i,j}(t) \\ & \quad \times [1 - C_{i+1,k}(t)]\} (1 - \alpha), \end{aligned} \quad \begin{aligned} i &= 1, 2, \dots, n-1 \\ j, k &= 1, 2, \dots, m \end{aligned} \quad (20)$$

where $C_{i,j}(t + \frac{1}{U})$ and $C_{i+1,k}(t + \frac{1}{U})$, by their definitions, are respectively the number concentration in cell j of the i -th section and that in cell k of the $(i+1)$ -th section after the coalescence.

According to the Monte Carlo simulation technique (see e.g., Meyer, 1956; Shreider, 1966) the mixing process described by Equations (14) through (18) can be simulated by using only Equations (19) and (20) in the following manner (see Fig. 3):

- (1) A section is selected at random by generating a random number i_1 where $1 \leq i_1 \leq n$.
- (2) A cell is selected at random from this section by generating another random number j_1 where $1 \leq j_1 \leq m$.
- (3) If the section selected in (1) is neither the top section nor

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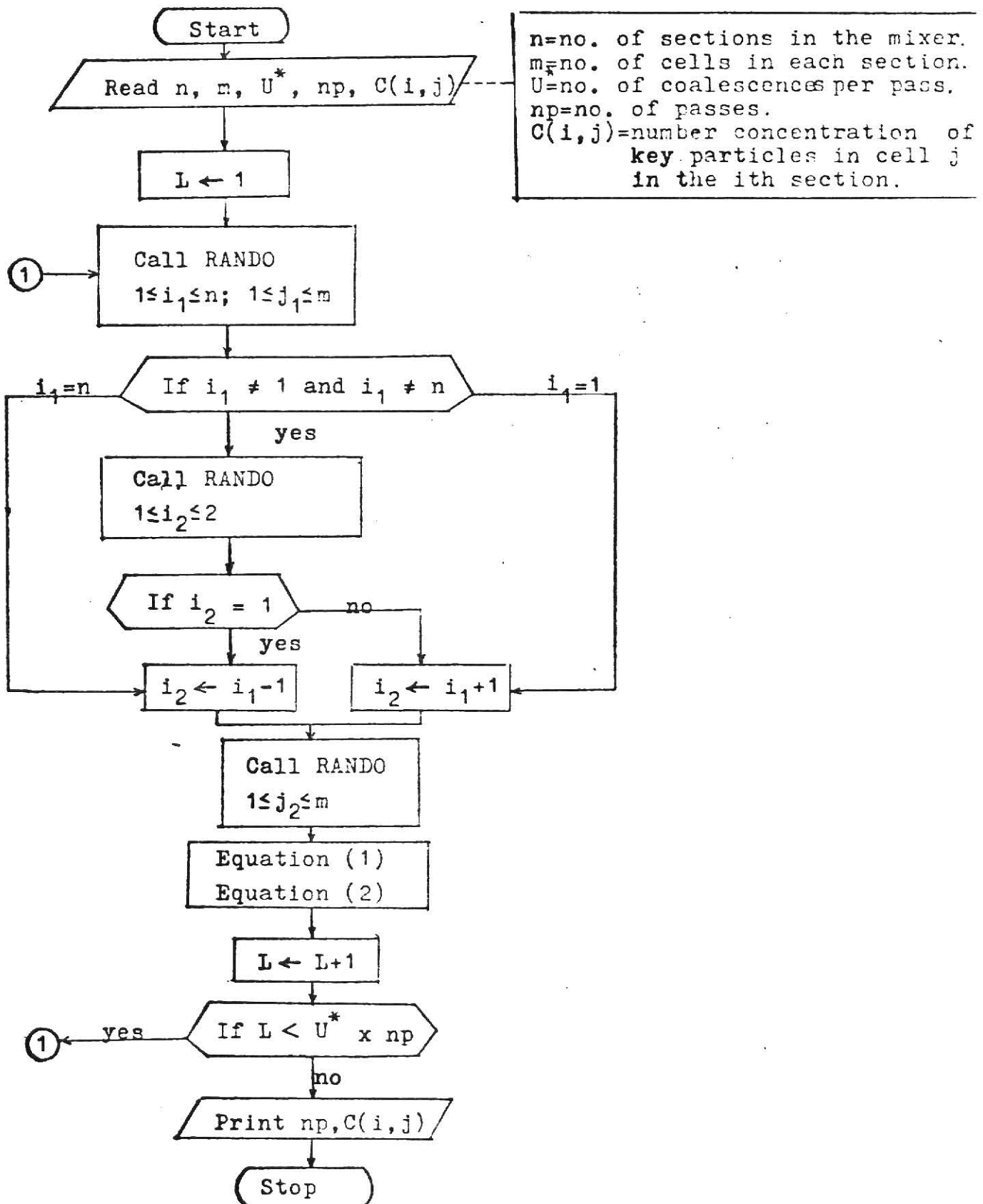


Fig. 3 Flowchart for the Monte Carlo simulation.

the bottom section, one of the two adjacent sections is selected at random by generating a third random number i_2 where $1 \leq i_2 \leq 2$. If the section selected in (1) is the top section, the second section from the top is always selected, and if the section selected in (1) is the bottom section, the second section from the bottom is always selected.

- (4) A cell is selected at random from the section selected in (3) by generating a random number j_2 where $1 \leq j_2 \leq m$.
- (5) The two cells selected in (2) and (4) constitute the coalescing pair. Redistribution of particles in these cells after the coalescence is computed according to Equations (19) and (20).
- (6) Repeat steps (1) through (5) until a desired number of coalescences is obtained.

3.5 EXPERIMENTAL DATA

The procedures employed to obtain the data (Gelves-Avocha, 1973) in this work are outlined below.

The mixing device employed to obtain the data (Kenics mixer) consisted of a 1.5-in. ID pyrex tube with a number of helical elements. Experiments were carried out by three basic operations: loading, passing, and sampling. In the loading process, key particles (see Table 1) were poured into the feeder first; particles of the second component (nonkey particles) were carefully layered on top. After loading, the feeder was placed above the mixer and the gate valve was withdrawn rapidly, permitting the slug of particles to fall freely through the mixer. The particles were caught in the collector; then the feeder and the collector were interchanged to carry out the second pass. This process was repeated to give the desired number of passes. To determine the fluctuation of the concentration of the key particles in the mixer, the variance was calculated by using the equation

$$\sigma^2 = \frac{\sum_{i=1}^m (c_i - \bar{c})^2}{n - 1} \quad (21)$$

where \bar{c} denotes the mean concentration of the key particles in the mixer and n represents the number of samples taken from the mixture. For the present case the entire mixture was divided into samples. The degree of mixedness was calculated for each experiment according to the definition

$$M = 1 - \frac{\sigma^2}{\sigma_0^2} \quad (22)$$

where σ_0^2 is the variance of the completely segregated state of the mixture, which can be calculated by

$$\sigma_0^2 = \bar{c} (1 - \bar{c}) \quad (23)$$

3.6 RESULTS AND DISCUSSION

Computations for solving Equations (14) through (18) by means of fourth-order Runge-Kutta method have been performed on an IBM 370 computer by dividing the mixer into 9 fictitious sections in series in the vertical direction with 10 cells arranged horizontally within each section. Simulations by means of the Monte Carlo method have also been carried out by using the same scheme and computer. Typical results are shown in Figs. 4a through 4e, which give concentration profiles for the axial mixing of 3/16" glass beads with the same size steel balls. Good agreements between the experimental data and the two simulations can be observed in general.

The overall progress of the mixing and demixing process can also be illustrated by a plot of the degree of mixedness (M) versus the number of passes as shown in Fig. 5. As expected, the simulated result based on Equations (14) through (18) is deterministic in nature while that based on the Monte Carlo simulation is stochastic in nature. In this aspect, the Monte Carlo simulation appears to be a better representation of the process. The difference in the two simulated results is not significant at the 5% confidence level. However, the computing time needed for the simulation based on Equations (14) through (18) is about 10 times as much as that needed for the Monte Carlo simulation. Simulation for all other particle systems have been performed by using the Monte Carlo simulation. The degree of fluctuation in the Monte Carlo simulations is dependent upon the number of cells identified in the mixer. It can be reduced by increasing the number of cells at the expense of more computing time. Nevertheless, 90 cells in the mixer have been found to be sufficient in this study.

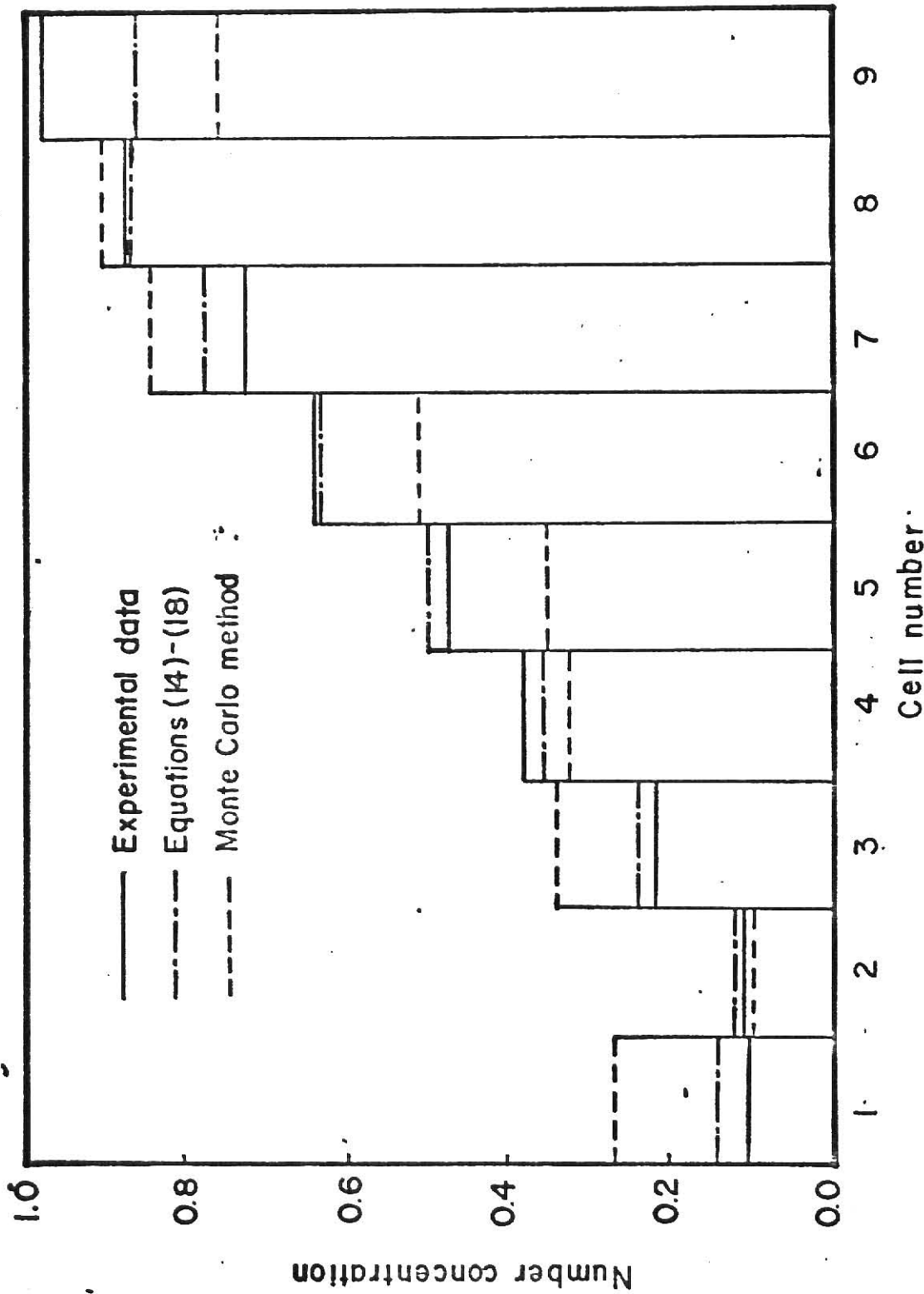


Fig. 4a. Concentration profile for mixing 3/16" glass particles with 3/16" steel particles after 2 passes in the 12 helices mixer.

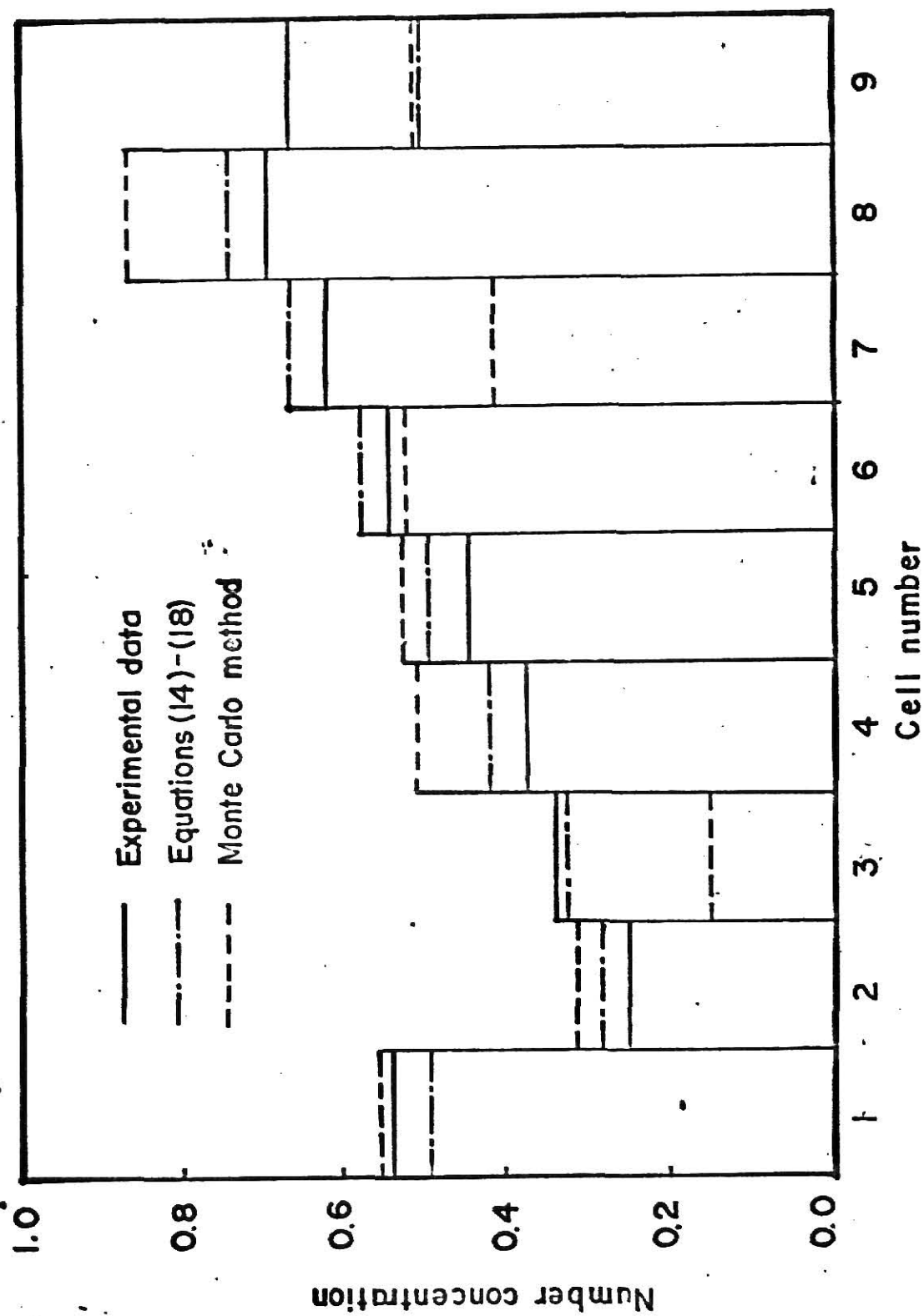


Fig.4b. Concentration profile for mixing 3/16" glass particles with 3/16" steel particles after 4 passes in the 12 helices mixer.

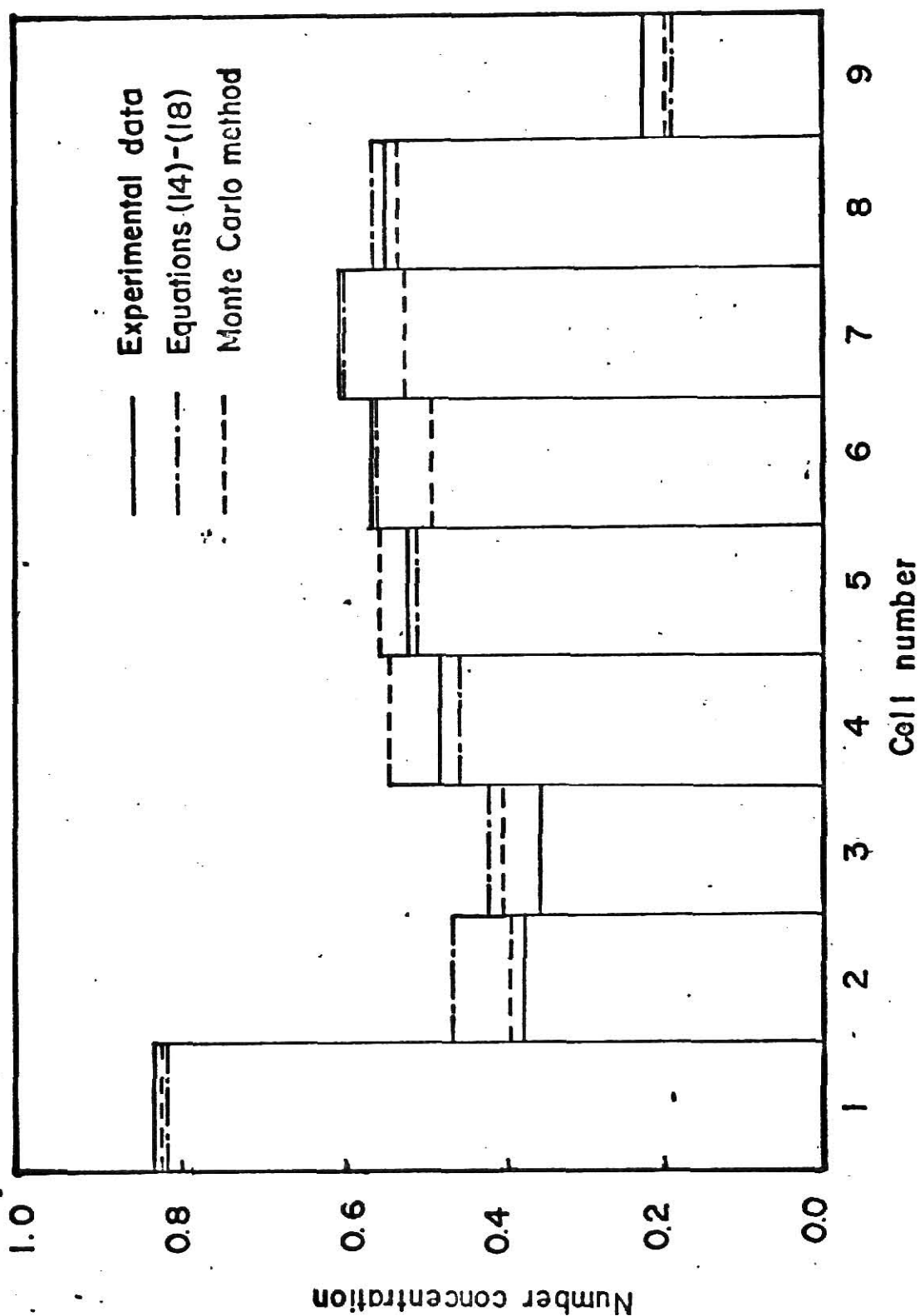


Fig.4c. Concentration profile for mixing 3/16" glass particles with 3/16" steel particles after 6 passes in the 12 helices mixer.

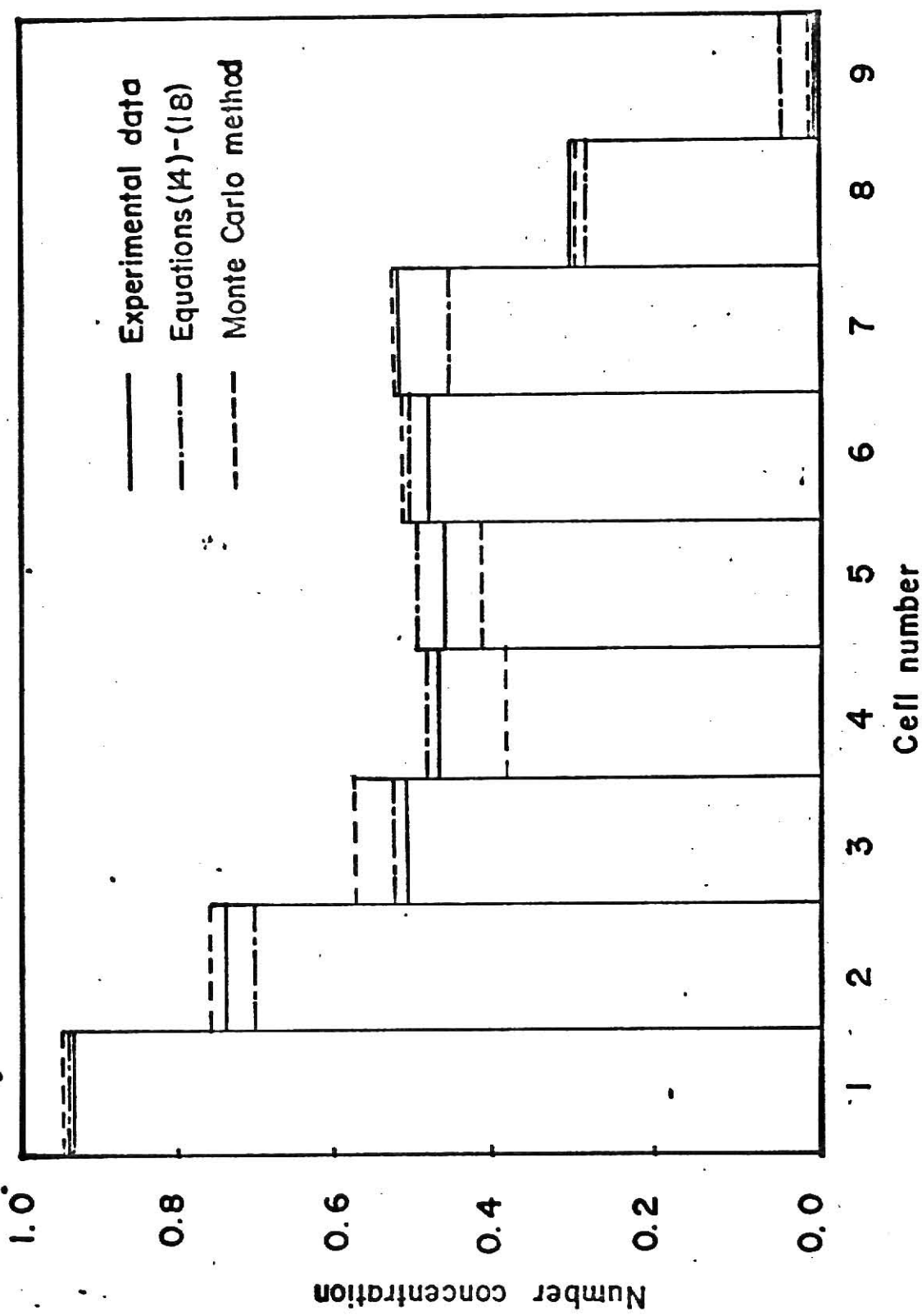


Fig. 4d. Concentration profile for mixing 3/16" glass particles with 3/16" steel particles after 8 passes in the 12 helices mixer.

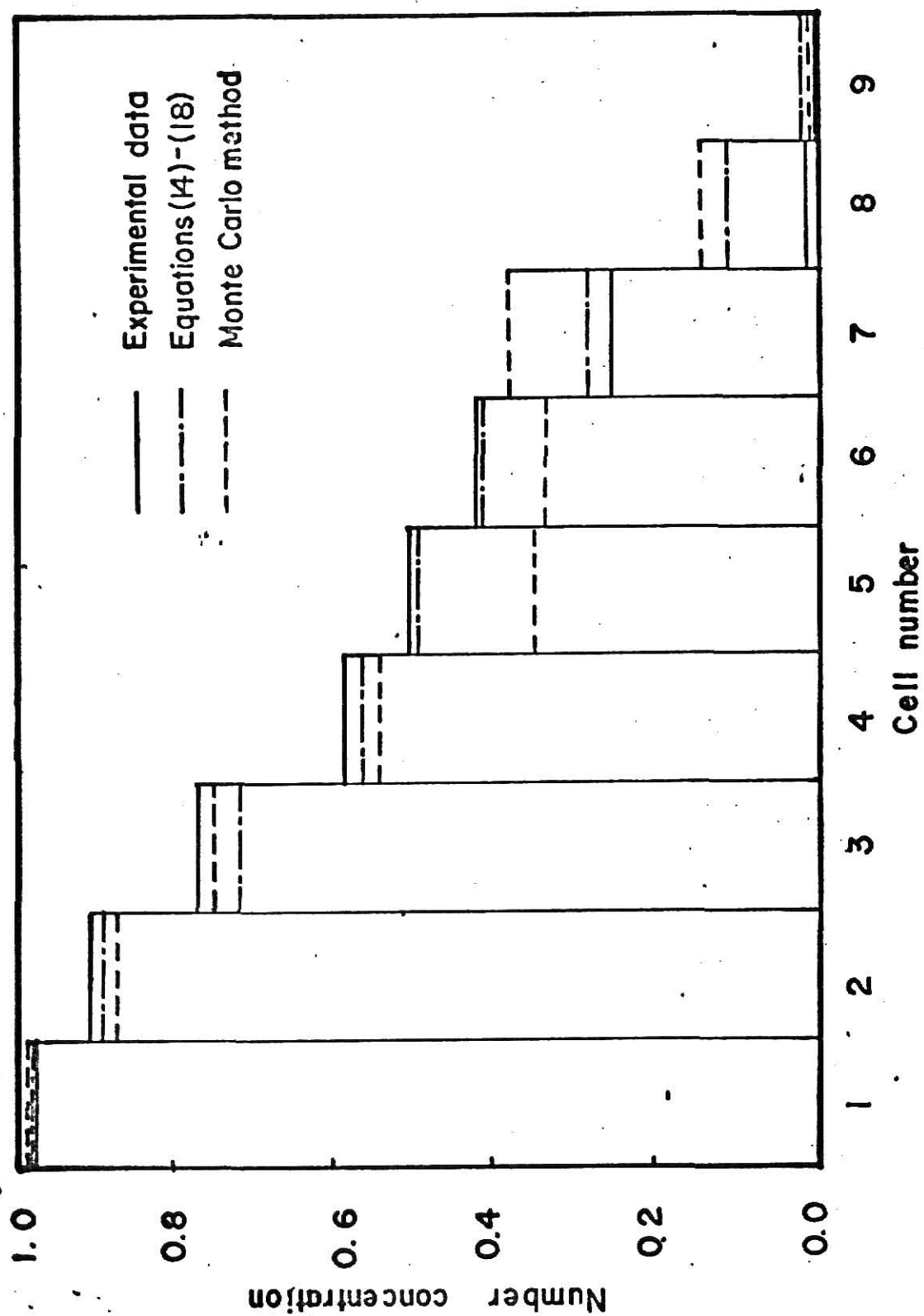


Fig.4e. Concentration profile for mixing 3/16" glass particles with 3/16" steel particles after 10 passes in the 12 helices mixer.

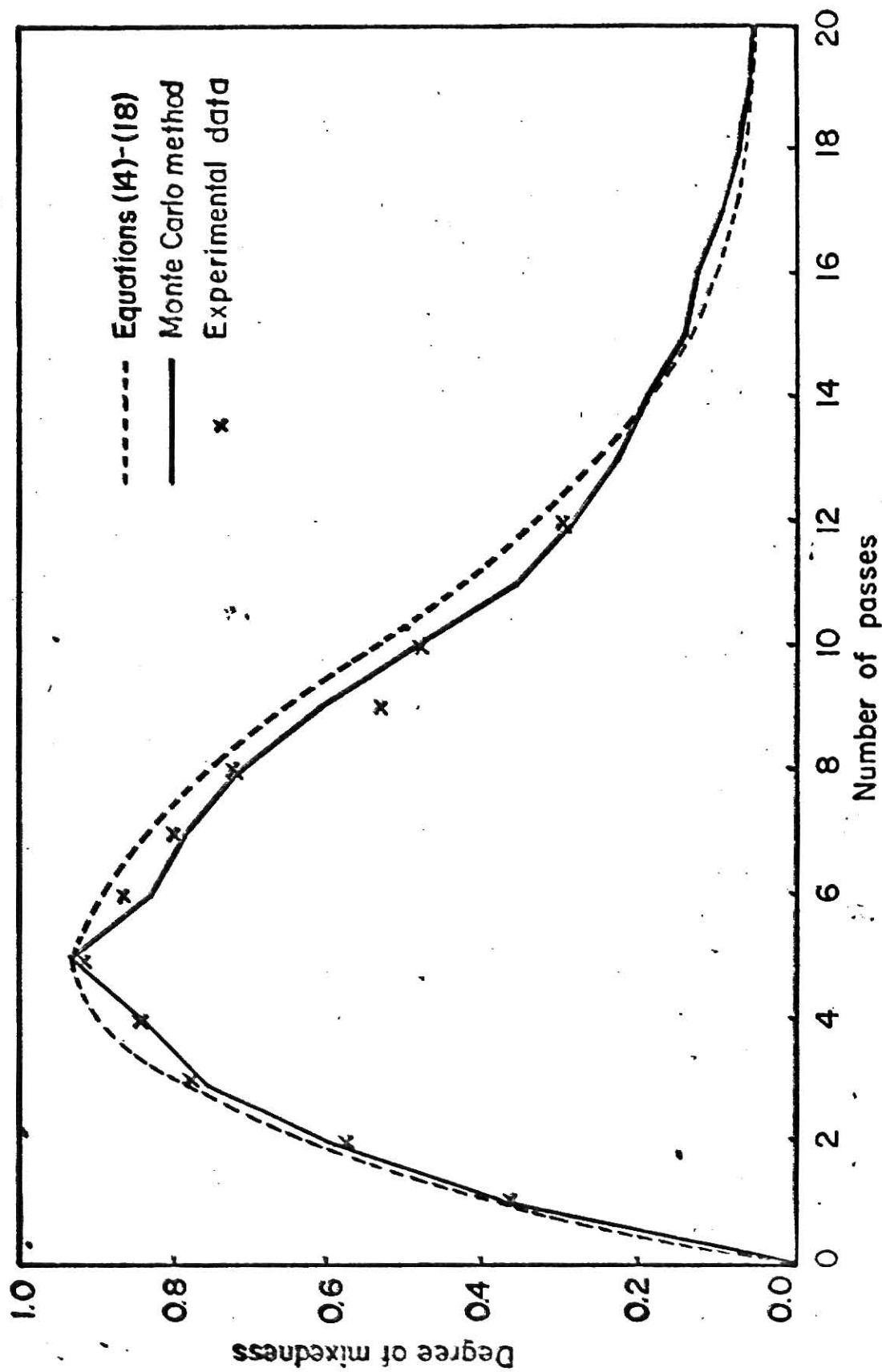


Fig.5. Degree of mixedness versus number of passes for mixing 3/16" glass particles with 3/16" steel particles in the 12 helices mixer.

3.6.1 Nonsegregating Particle Systems

Experimental data (Gelves-Arocha, 1973) in Fig. 6 show that the rate of mixing was relatively fast initially and decreased as the number of passes increased. After a sufficient number of passes, a dynamic equilibrium state was reached and no further improvement in the homogeneity may be obtained. The rate of mixing was strongly dependent on the particle size. Mixing progressed most rapidly when the particle size was small. The progress of mixing for this case can be described by the proposed model with $\alpha = 0.5$ as shown in Fig. 6. The values of the coalescence rate (number of coalescences per pass), U^* , estimated from Fig. 6 are represented in Table 1 and plotted against $(1/\text{Particle diameter})^3$ as shown in Fig. 7. A near straight line can be obtained from the plot indicating that an important factor for mixing is the volume of particles. The line passes through the origin when extrapolated. This implies that as particles become excessively large and therefore $(1/\text{Particle diameter})^3$ excessively small, no mixing can be achieved in the mixer. The result is intuitively expected as mixing is limited by the dimension of a mixer when the particles mixed are extremely large.

The coalescence rate is plotted against the number of helices in the mixer as shown in Fig. 8, which indicates the following correlations

$$U^* = 11 + 20 \times (\text{number of helices}) \quad (24)$$

for 1/8" lucite particles,

and

$$U^* = 14 + 9 \times (\text{number of helices}) \quad (25)$$

for 5/32" lucite particles.

The nonzero intercepts imply that even without mixing elements in the mixer, slight mixing can still be achieved.

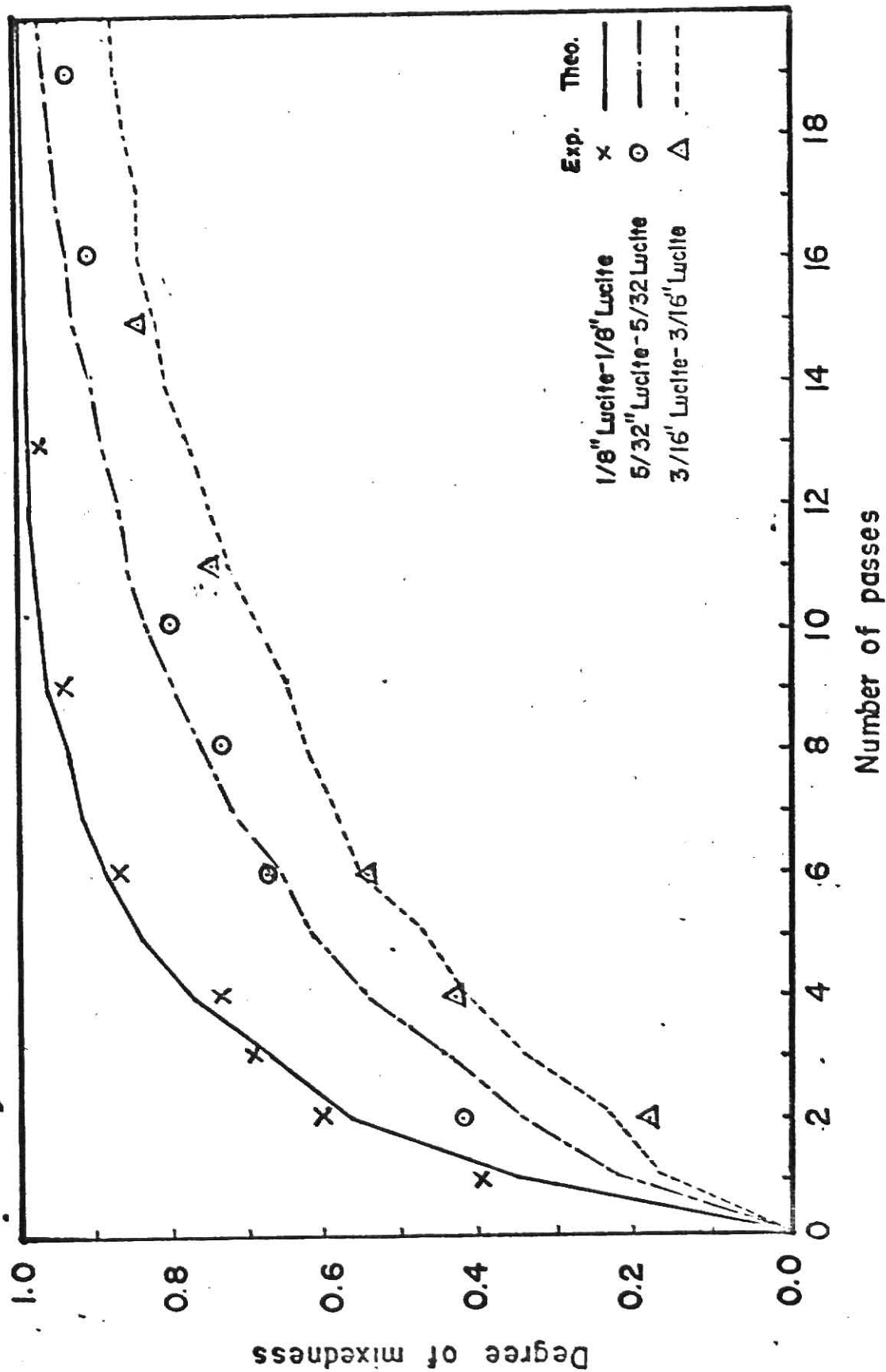


Fig.6. Effect of particle size on the rate of mixing for nonsegregating particle systems in the 12 helices mixer.

Table 1. Estimated Values of Coalescence and Distribution Parameters
Based on Experimental Data by Gelves-Arocha (1973).

No. of helices in the mixer	Key Component (Charged in the bottom)			Second Component			No. Fraction of Key Component	Coalescence rate, U*	Distribution ratio, α
	Material	Particle size (in.)	Density (g/cm ³)	Material	Particle size (in.)	Density (g/cm ³)			
2	Lucite	1/8	1.18	Lucite	1/8	1.18	0.500	48	0.500
5	"	"	"	"	"	"	"	113	"
12	"	"	"	"	"	"	"	253	"
2	"	5/32	"	"	5/32	"	"	35	"
5	"	"	"	"	"	"	"	64	"
12	"	"	"	"	"	"	"	121	"
12	"	3/16	"	"	3/16	"	0.515	78	"
5	"	1/8	"	"	"	"	0.680	55	0.726
8	"	"	"	"	"	"	"	77	0.773
12	"	"	"	"	"	"	"	115	0.707
5	"	"	"	"	"	"	0.600	69	0.715
7	"	"	"	"	5/32	"	"	85	0.726
9	"	"	"	"	"	"	"	104	0.747
12	"	"	"	"	"	"	"	128	0.762
2	"	3/16	"	Glass	3/16	2.42	0.515	22	0.773
5	"	"	"	"	"	"	"	63	0.777
8	"	"	"	"	"	"	"	66	0.875
12	"	"	"	"	"	"	"	100	0.927
12	"	"	"	Steel	"	7.60	0.52	184	0.987
12	"	"	"	"	"	"	0.50	105	0.958
5	Glass	5/32	2.42	Glass	"	2.42	0.58	51	0.985
8	Lucite	"	1.18	"	"	"	"	84	0.991
12	"	"	"	"	"	"	"	115	0.951

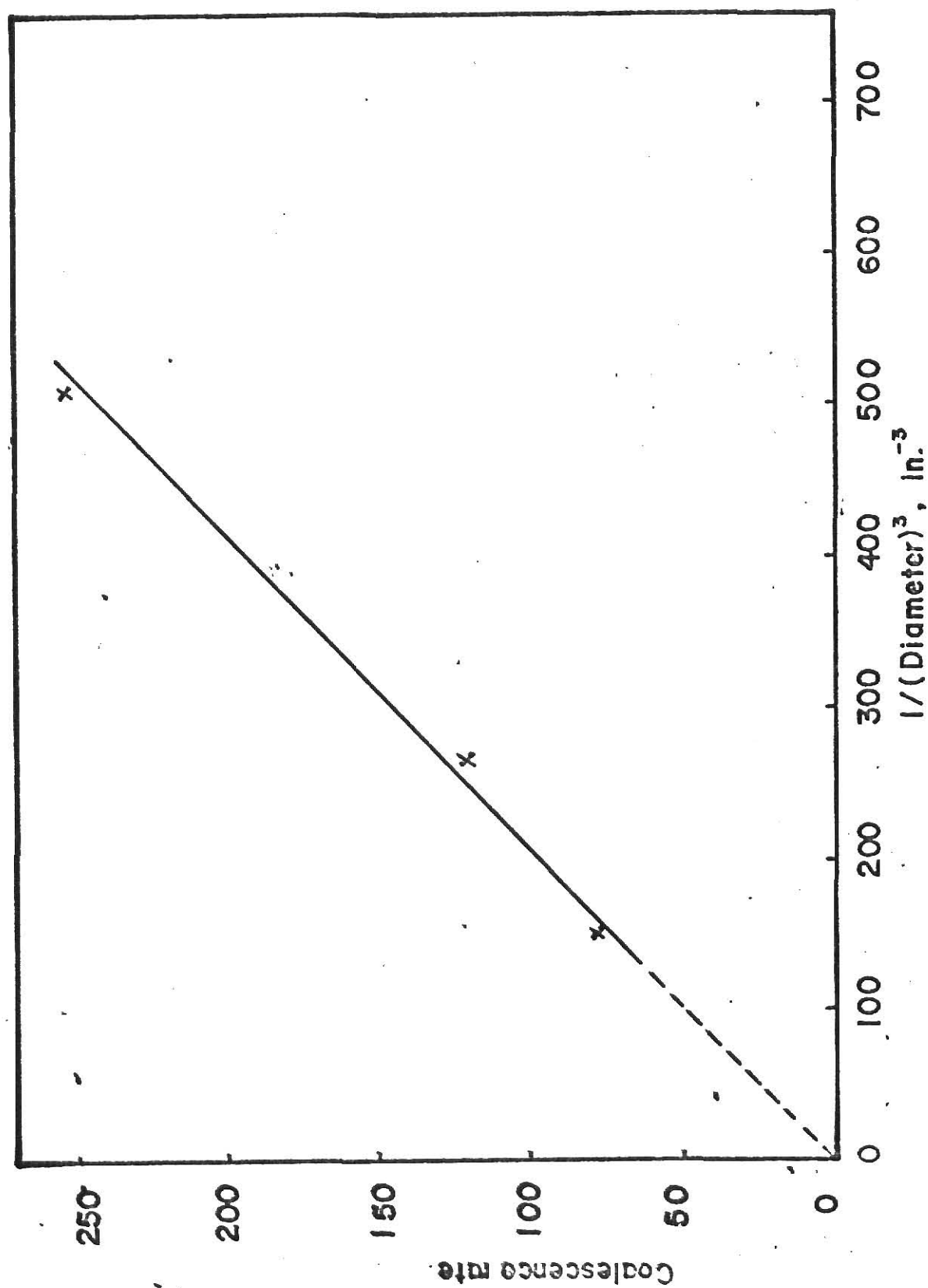


Fig.7 . Coalescence rate as a function of reciprocal of particle diameter to the third power for mixing nosegregating particle systems in the 12 helices mixer.

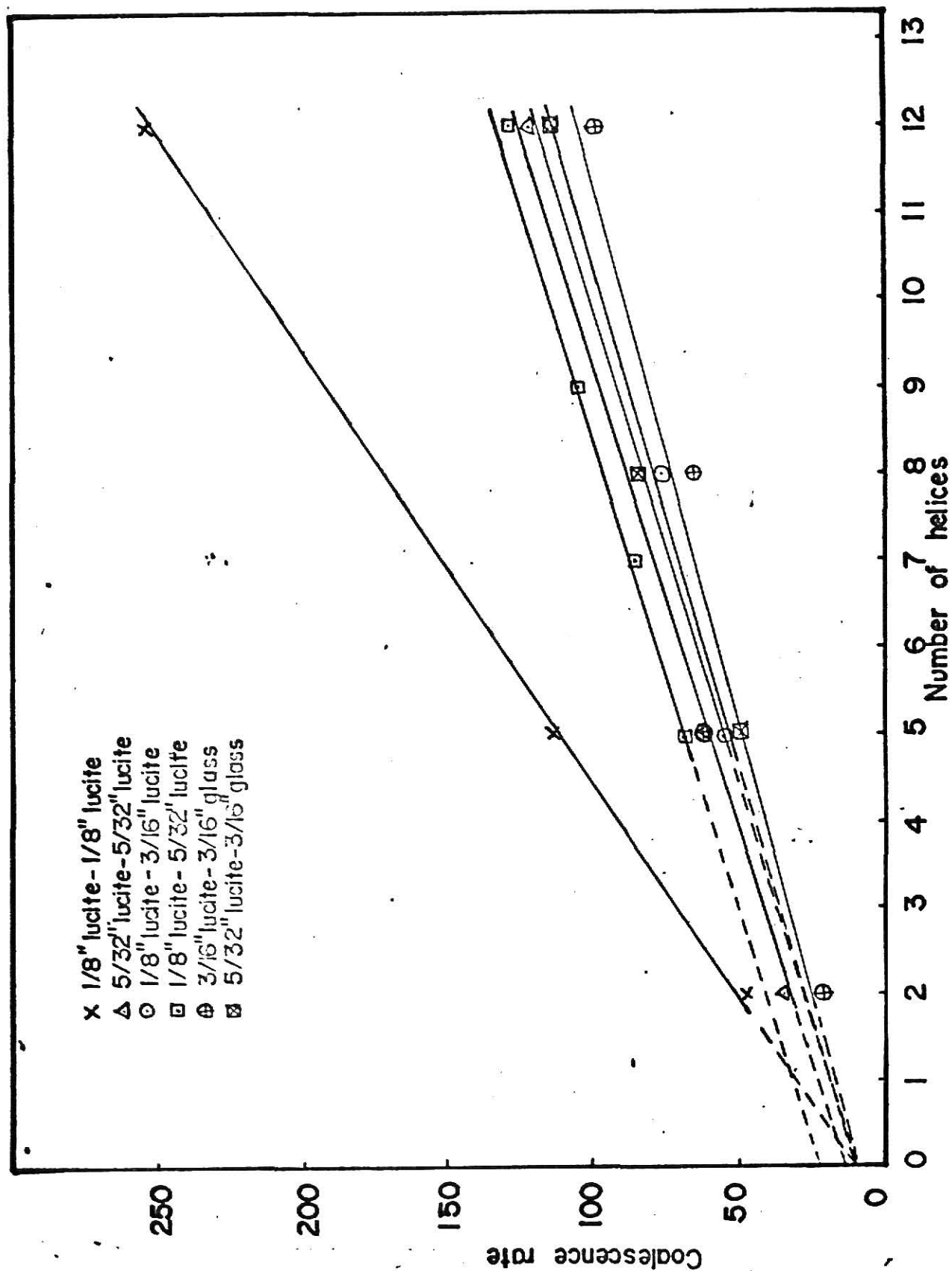


Fig.8 . Estimated coalescence rates as functions of the number of helices in the mixer.

3.6.2 Segregating Particle Systems

For the system with size difference, the small particles were initially placed at the bottom and the large particles were layered on the top (Gelves-Arocha, 1973). A typical degree of mixedness versus time plot (see Fig. 9) shows a rapid initial rise in M to a maximum, followed by a decline to a terminal value corresponding to a dynamic equilibrium state. Estimated coalescence rates presented in Table 1 can be correlated by

$$U^* = 22 + 9 \times (\text{number of helices}) \quad (26)$$

for $1/8''$ - $5/32''$ lucite particles,

and

$$U^* = 10 + 9 \times (\text{number of helices}) \quad (27)$$

for $1/8''$ - $3/16''$ lucite particles.

The coalescence rate for the $1/8''$ - $3/16''$ lucite particle system was smaller than that for the $1/8''$ - $5/32''$ lucite particle system while the size difference for the former was greater than that for the latter. It is possible that in addition to size difference, the coalescence rate is also a function of individual particle sizes. The size and the size difference may couple together in a complex fashion that is beyond our description at this stage.

The distribution ration, α , which appeared in the model, indicates the tendency of a particle system to segregate. While it is impossible to determine explicitly the dependence of the distribution ratio on the size difference from the limited number of the available data, it appears that an increase in the size difference results in an increase in α .

Figure 8 shows that the linear relationship between the coalescence rate and the number of helices is less obvious for the system with density difference than that for the system with size difference. It

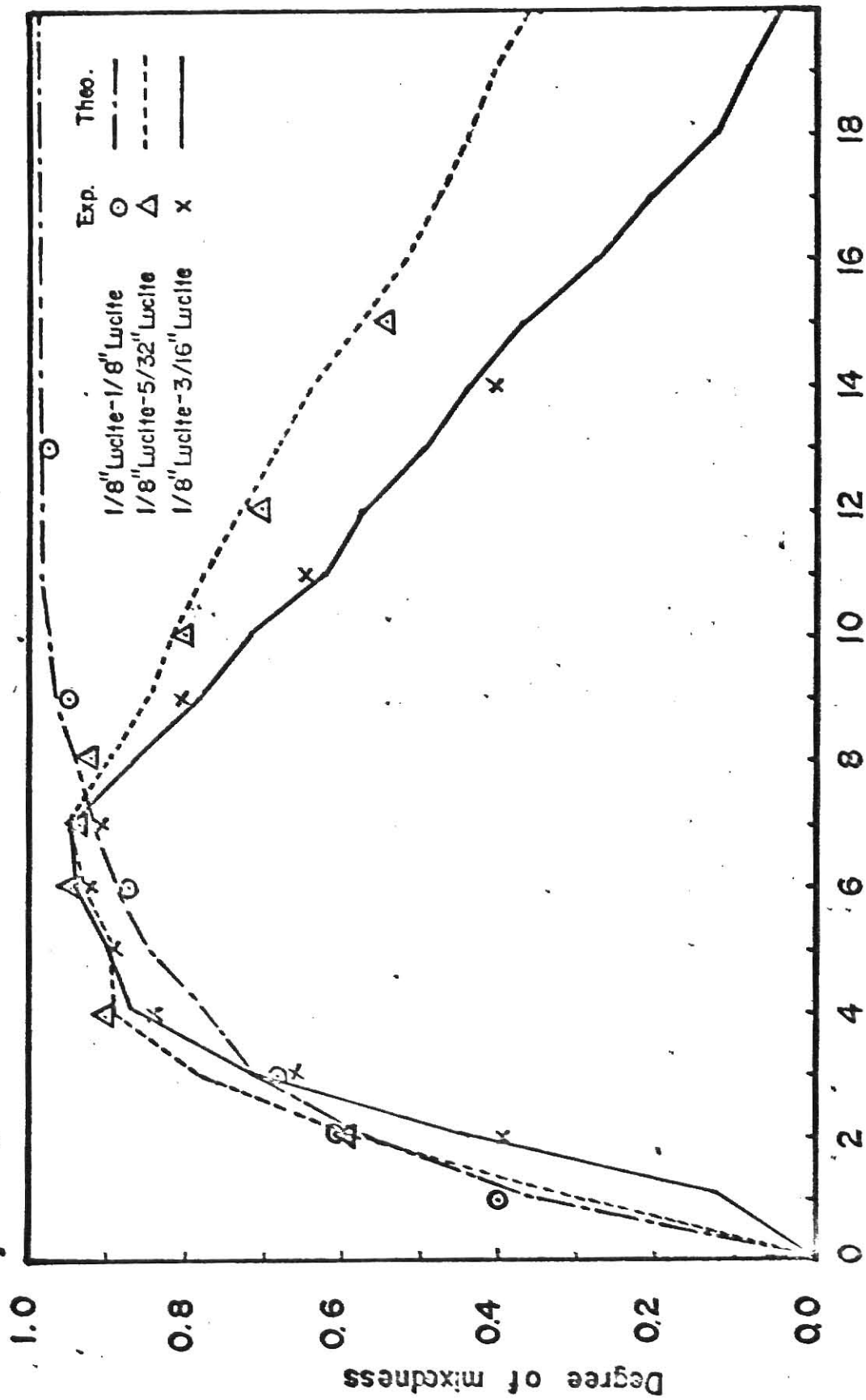


Fig.9. Effect of particle size difference on the rate of mixing and segregation in the 12 helices mixer.

may be that the end effects are more pronounced for the former than for the latter. The rate of mixing for various combinations of particles differing primarily in density is shown in Fig. 10, which reveals the fact that the greater the density difference the faster the mixing and demixing. The estimated coalescence rate is plotted against $\Delta\rho/\rho_m$ in Fig. 11. It appears that the rate of increase in the coalescence rate with $\Delta\rho/\rho_m$ is reduced as the density difference becomes smaller. After the density difference reaches a certain value, the increase in the coalescence rate becomes so drastic that eventually the particle system mix and demix within a single pass. $\alpha/(1 - \alpha)$ is plotted against $\Delta\rho/\rho_m$ in the same figure, which indicates the correlation,

$$\frac{\alpha}{(1 - \alpha)} = 1.1 + 20.6 \times \Delta\rho/\rho_m \quad (28)$$

For the system with both density and size differences, the small particles were initially placed at the bottom and the coarse were layered on the top. The degree of mixedness for this case is plotted as a function of the number of passes in Fig. 12. As can be seen from the figure, the rates of mixing and demixing are much greater than the two previous systems. The combined effect of size and density differences is greater than the single effect of either the size or density difference.

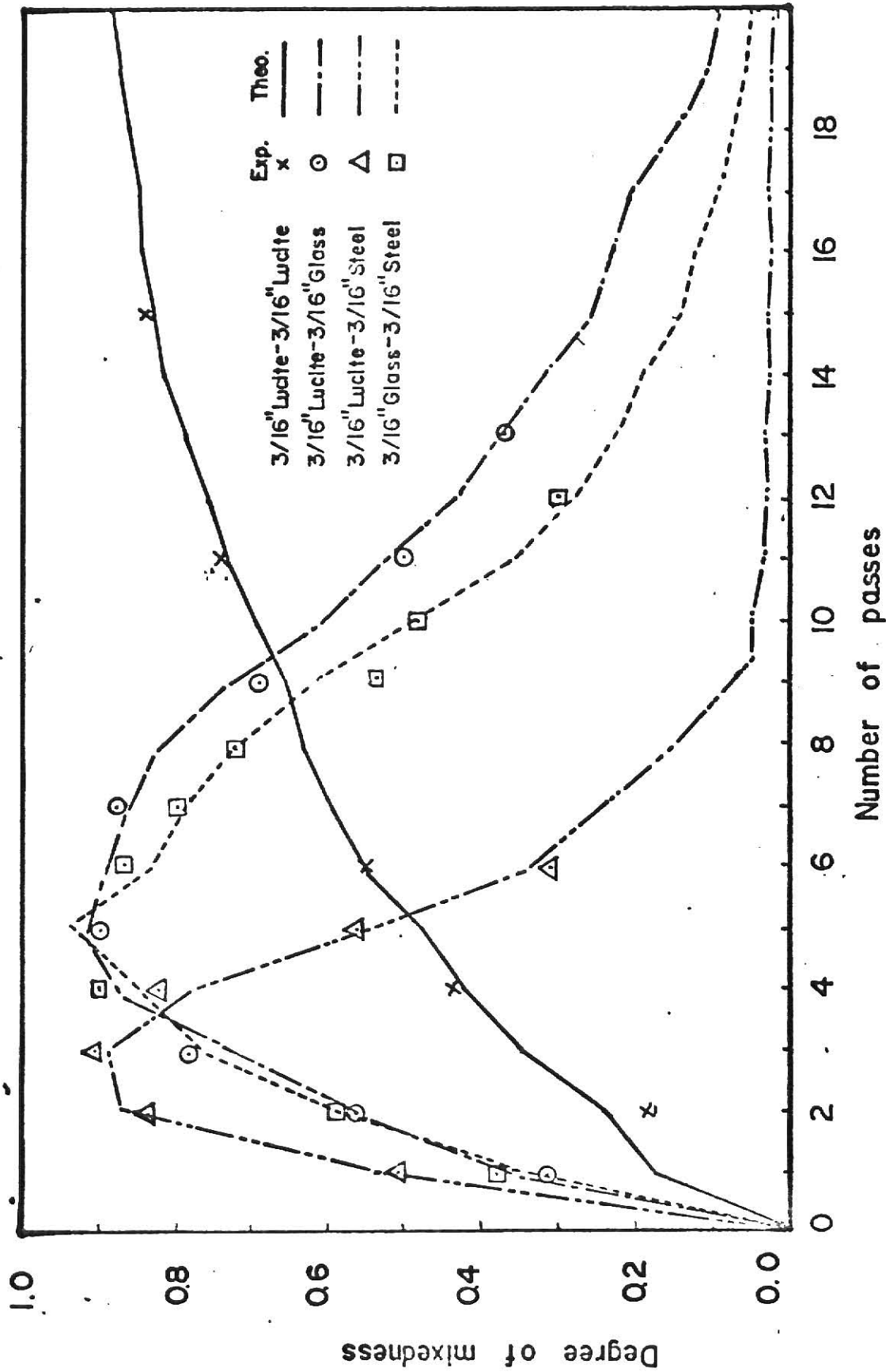


Fig.10. Effect of particle density difference on the rate of mixing and segregation in the 12 helices mixer.

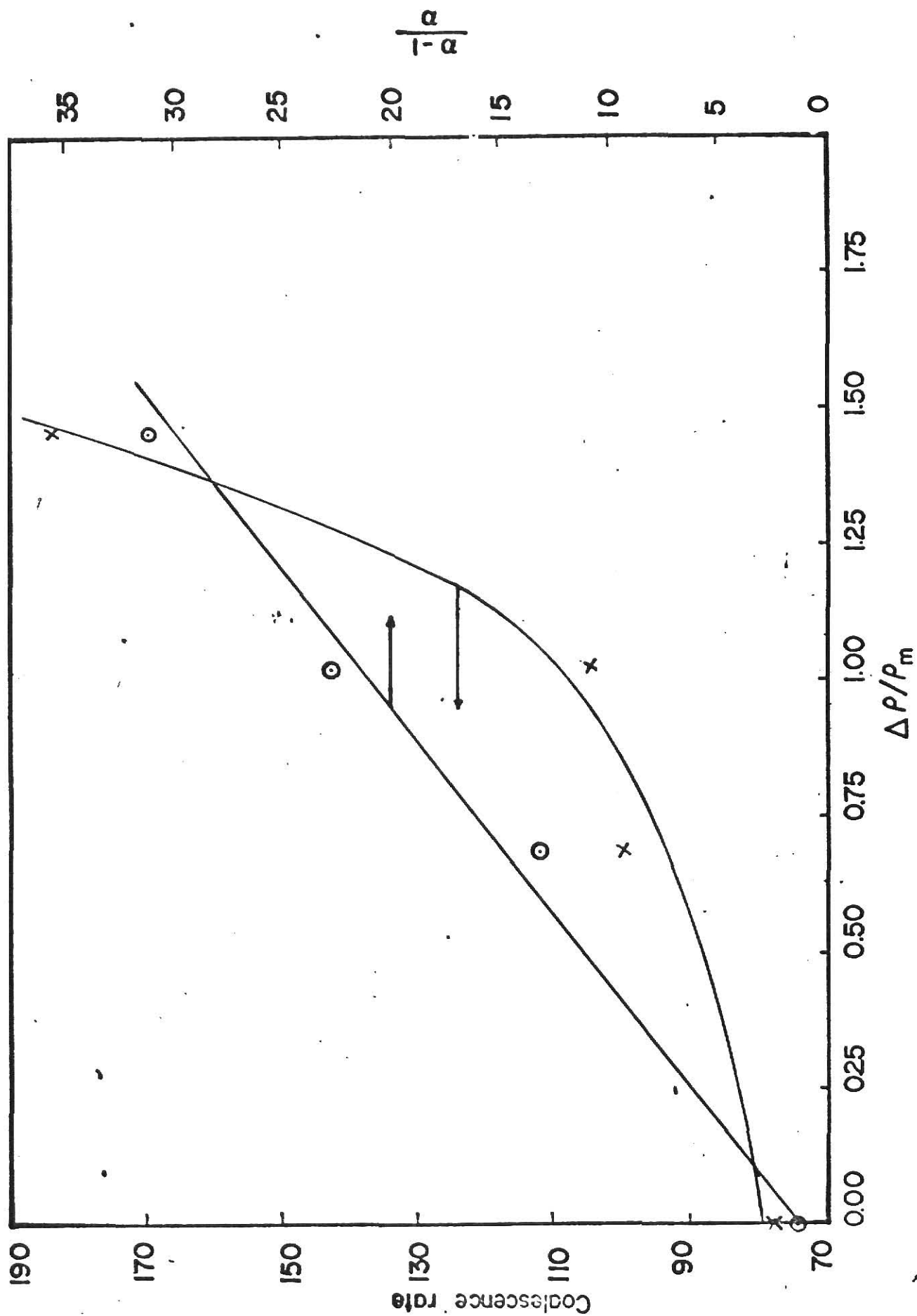


Fig.11. Estimated parameters as functions of $\Delta P/P_m$ for mixing particles of different densities in the 12 helices mixer.

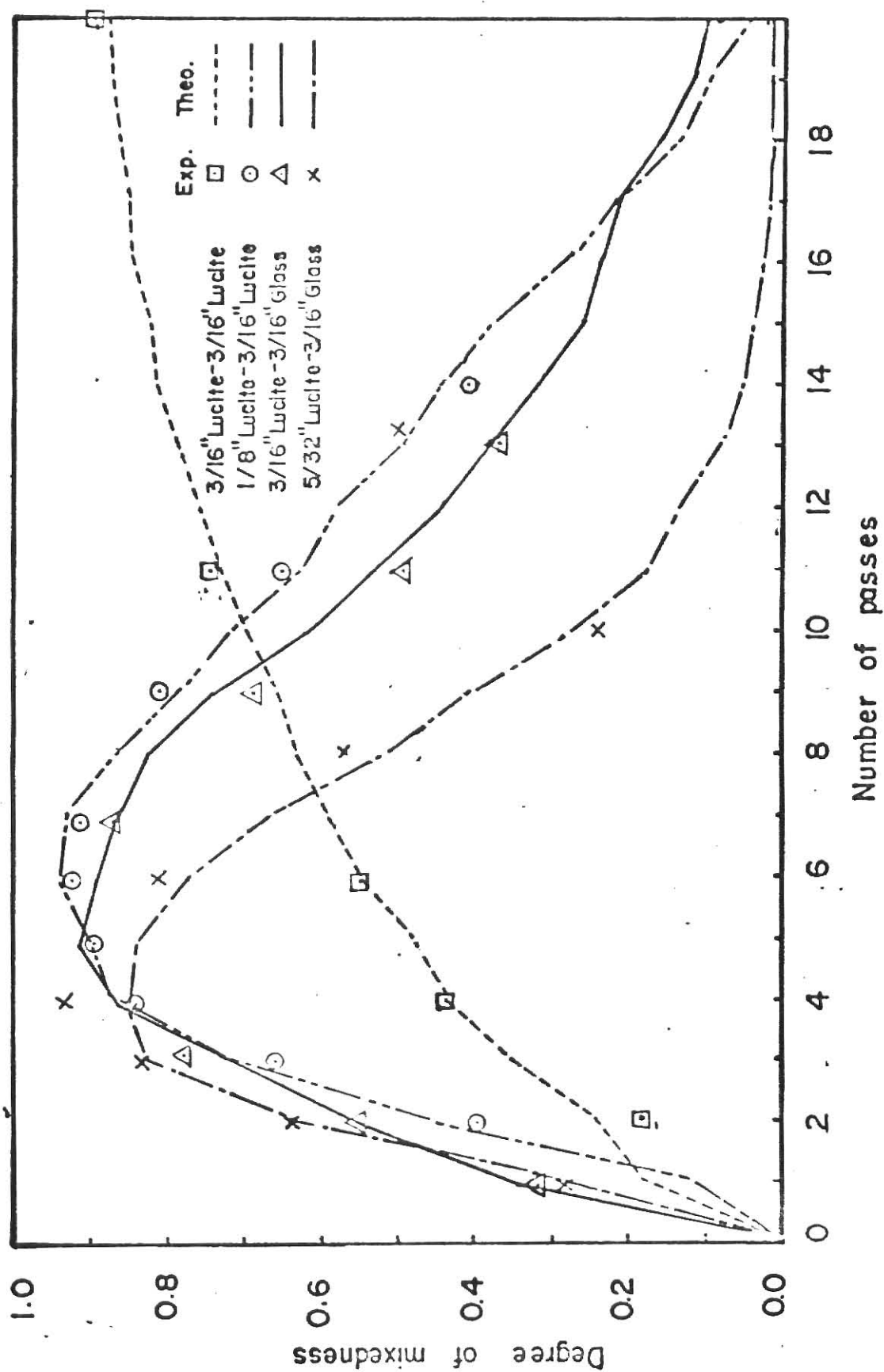


Fig.12. Effect of particle size and/or density differences on the rate of mixing for various particle systems in the 12 helices mixer.

3.7 CONCLUSIONS

A modified coalescence-dispersion model has been developed and successfully applied to correlation of the available experimental data (Gelves-Arocha, 1973). For the nonsegregating systems and the segregating systems with size difference, the coalescence rate is a linear function of the number of helices in the mixer. The linear relationship between the coalescence rate and the number of helices in the mixer is less obvious for the system with density difference than for the system with size difference. The dependence of the coalescence rate on the size of particles indicates that an important factor for mixing is the volume of individual particles for the nonsegregating system. The distribution ratio appeared to be dependent upon the size, the size difference, the density, and the density difference.

While previous proposed deterministic model (Fan et al., 1975) for mixing of segregating particle systems in motionless mixers contains four parameters, the present stochastic model contains only two parameters. Unlike the deterministic model, the present model is capable of generating concentration distribution as a function of time and its applicability is not constrained by the initial concentration distribution in the mixer. Therefore, the present model may be valuable in understanding the mechanisms of mixing in the motionless mixers as well as for the improvement of mixer design.

To derive a model which can take into account additional details of mixing mechanisms such as the cell size distribution and the coalescence rate distribution in the mixer than the present model can, additional analysis and experimentation are required.

NOTATION

\bar{C} = average number concentration of key particles in the mixer

C_i = average number concentration of key particles in section i

$C_{i,j}$ = number concentration of key particles in cell j of the i -th section

$(C_{i,j})_a$ = apparent number concentration of key particles in cell j of the i -th section

m = number of divided cells in each section

n = number of divided sections in the mixer

np = number of passes through the mixer

r = bulk volume ratio of a key particle to a nonkey particle

U = number of coalescences per unit time

U^* = number of coalescences per pass through the mixer

$X_{i,j}$ = number of key particles in cell j in the i -th section

$Y_{i,j}$ = number of nonkey particles in cell j in the i -th section

α = distribution ratio

ℓ = total number of particles in each cell

σ^2 = variance

σ_o^2 = variance of the completely segregated state

ρ = density

ρ_m = average density

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APPENDIX I. DERIVATION OF EQUATIONS (1) AND (2).

According to assumption (1), the number of particles in each cell is a constant which is to be denoted by l . Based on assumption (2), upon coalescence, each particle in one of the two coalescing cells experiences a one-to-one collision with another particle in the remaining cell in a completely random manner. Thus, l collisions occur for each coalescence.

$$\begin{aligned} &\text{The number of collisions per unit time} \\ &= lU \end{aligned} \tag{A1.1}$$

where U denotes the number of coalescence per unit time.

$$\begin{aligned} &\text{The number of collisions during an arbitrarily small interval } \Delta t \\ &= lU\Delta t \end{aligned} \tag{A1.2}$$

$$\begin{aligned} &\text{The probability of key-key particle collisions provided that cell } j \\ &\text{in the } i\text{-th section coalescences with cell } k \text{ in the } (i+1)\text{-th section} \\ &= C_{i,j}(t) C_{i+1,k}(t), \quad \begin{aligned} i &= 1, 2, \dots, n-1 \\ j, k &= 1, 2, \dots, m \end{aligned} \end{aligned} \tag{A1.3}$$

$$\begin{aligned} &\text{The expected number of key-key particle collisions during } \Delta t \\ &\text{provided that cell } j \text{ in the } i\text{-th section coalescences with cell } k \text{ in} \\ &\text{the } (i+1)\text{-th section} \\ &= [C_{i,j}(t) C_{i+1,k}(t)] (lU\Delta t), \quad \begin{aligned} i &= 1, 2, \dots, n-1 \\ j, k &= 1, 2, \dots, m \end{aligned} \end{aligned} \tag{A1.4}$$

Based on assumption (2), after each key-key particle collisions, the two particles in the colliding pair split instantaneously and distribute randomly between the coalescing cells. Thus,

The expected number of key particles in cell j in the i -th section resulting from key-key particle collisions during Δt provided that

the coalescence is with cell k in the (i+1)-th section

$$= [C_{i,j}(t) C_{i+1,k}(t)] (\ell U \Delta t) \times 1, \quad i = 1, 2, \dots, n-1 \quad (A1.5)$$

$$j, k = 1, 2, \dots, m$$

The expected number of key particles in cell k in the (i+1)-th section resulting from key-key particle collisions during Δt provided that the coalescence is with cell k in the (i+1)-th section

$$= [C_{i,j}(t) C_{i+1,k}(t)] (\ell U \Delta t) \times 1, \quad i = 1, 2, \dots, n-1 \quad (A1.6)$$

$$j, k = 1, 2, \dots, m$$

The probability of key-nonkey particle collisions provided that cell j in the i-th section coalescences with cell k in the (i+1)-th section

$$= [1 - C_{i,j}(t)] C_{i+1,k}(t) + C_{i,j}(t) [1 - C_{i+1,k}(t)], \quad (A1.7)$$

$$i = 1, 2, \dots, n-1$$

$$j, k = 1, 2, \dots, m$$

The expected number of key-nonkey particle collisions during Δt provided that cell j in the i-th section coalescences with cell k in the (i+1)-th section

$$= \{[1 - C_{i,j}(t)] C_{i+1,k}(t) + C_{i,j}(t) [1 - C_{i+1,k}(t)]\} (\ell U \Delta t), \quad (A1.8)$$

$$i = 1, 2, \dots, n-1$$

$$j, k = 1, 2, \dots, m$$

According to assumption (2), upon collision, a key particle will have a probability of α to find its new position in cell j in the i-th section and a probability of $(1-\alpha)$ to find its new position in cell k in the (i+1)-th section. Thus,

The expected number of key particles in cell j in the i-th section resulting from key-nonkey particle collisions during Δt provided that the coalescence is with cell k in the (i+1)-th section

$$= \{[1 - C_{i,j}(t)] C_{i+1,k}(t) + C_{i,j}(t) [1 - C_{i+1,k}(t)]\} (\ell U \Delta t), \quad (A1.9)$$

$$i = 1, 2, \dots, n-1$$

$$j, k = 1, 2, \dots, m$$

The expected number of key particles in cell k in the (i+1)-th section resulting from key-nonkey particle collisions during Δt provided that the coalescence is with cell k in the (i+1)-th section

$$= \{[1 - C_{i,j}(t)] C_{i+1,k}(t) + C_{i,j}(t) [1 - C_{i+1,k}(t)]\} (\ell U \Delta t) (1-\alpha), \quad (A1.10)$$

$$i = 1, 2, \dots, n-1$$

$$j, k = 1, 2, \dots, m$$

Collisions between nonkey particles contribute nothing to the concentration of key particles.

The number fraction of particles in a cell which do not collide during Δt provided that the cell is selected as one cell of a coalescing pair

$$= [\ell - (\ell U \Delta t)] / \ell$$

$$= 1 - U \Delta t \quad (A1.11)$$

The expected number of key particles in cell j in the i-th section which do not collide during Δt provided that the coalescence is with cell k in the (i+1)-th section

$$= C_{i,j}(t) \ell (1 - U \Delta t), \quad i = 1, 2, \dots, n-1 \quad (A1.12)$$

$$j = 1, 2, \dots, m$$

The expected number of key particles in cell k in the (i+1)-th section which do not collide during Δt provided that the coalescence is with cell j in the i-th section

$$= C_{i+1,k}(t) \ell (1 - U \Delta t), \quad i = 1, 2, \dots, n-1 \quad (A1.13)$$

$$j = 1, 2, \dots, m$$

The total expected number of key particles in cell j in the i -th section at $t+\Delta t$ provided that the coalescence is with cell k in the $(i+1)$ -th section

$$= [C_{i,j}(t) C_{i+1,k}(t) (\ell U \Delta t) \times 1] + \left\{ \{ [1 - C_{i,j}(t)] C_{i+1,k}(t) + C_{i,j}(t) \times [1 - C_{i+1,k}(t)] \} (\ell U \Delta t) \alpha \right\} + [C_{i,j}(t) \ell (1 - U \Delta t)], \quad (A1.14)$$

$$i = 1, 2, \dots, n-1$$

$$j, k = 1, 2, \dots, m$$

The total expected number of key particles in cell k in the $(i+1)$ -th section at $t+\Delta t$ provided that the coalescence is with cell j in the i -th section

$$= [C_{i,j}(t) C_{i+1,k}(t) (\ell U \Delta t) \times 1] + \left\{ \{ [1 - C_{i,j}(t)] C_{i+1,k}(t) + C_{i,j}(t) \times [1 - C_{i+1,k}(t)] \} (\ell U \Delta t) (1 - \alpha) \right\} + [C_{i+1,k}(t) \ell (1 - U \Delta t)], \quad (A1.15)$$

$$i = 1, 2, \dots, n-1$$

$$j, k = 1, 2, \dots, m$$

The expected concentration of key particles in cell j in the i -th section at $t+\Delta t$ provided that the coalescence is with cell k in the $(i+1)$ -th section

$$= \left[C_{i,j}(t) C_{i+1,k}(t) + \{ [1 - C_{i,j}(t)] C_{i+1,k}(t) + C_{i,j}(t) [1 - C_{i+1,k}(t)] \}^\alpha \right] \times U \Delta t + C_{i,j}(t) (1 - U \Delta t), \quad i = 1, 2, \dots, n-1 \quad (A1.16)$$

$$j, k = 1, 2, \dots, m$$

which is equation (1).

The expected concentration of key particles in cell k in the $(i+1)$ -th section at $t+\Delta t$ provided that the coalescence is with cell j in the i -th section

$$= \left[C_{i,j}(t) C_{i+1,k}(t) + \{ [1 - C_{i,j}(t)] C_{i+1,k}(t) + C_{i,j}(t) [1 - C_{i+1,k}(t)] \}^\alpha \right] \times (1 - \alpha) U \Delta t + C_{i+1,k}(t) (1 - U \Delta t), \quad i = 1, 2, \dots, n-1 \quad (A1.17)$$

$$j, k = 1, 2, \dots, m$$

which is equation (2).

$$\begin{aligned}
&= \frac{1}{n \times m} \times \frac{1}{m} + \frac{1}{n \times m} \times \frac{1}{2 \times m} \\
&= \frac{3}{2 \times n \times m^2}
\end{aligned} \tag{A2.7}$$

which is Equation (4).

The probability that cell j in the i -th section is selected as the first cell of a coalescing pair

$$= \frac{1}{n \times m} \tag{A2.8}$$

The conditional probability that cell k in the $(i+1)$ -th section is selected as the second cell of the coalescing pair provided that cell j in the i -th section is selected as the first cell of the coalescing pair

$$= \frac{1}{2 \times m} \tag{A2.9}$$

The probability that cell j in the i -th section is selected as the first cell of the coalescing pair and cell k in the $(i+1)$ -th section is selected as the second cell of the coalescing pair

$$= \frac{1}{n \times m} \times \frac{1}{2 \times m} \tag{A2.10}$$

There is equal probability that cell k in the $(i+1)$ -th section is selected as the second cell of a coalescing pair and cell j in the i -th section is selected as the second cell of the coalescing cell. Therefore, the probability that cell j in the i -th section and cell k in the $(i+1)$ -th section are selected as the two cells of the coalescing pair

$$\begin{aligned}
&= \frac{1}{n \times m} \times \frac{1}{2 \times m} \times 2 \\
&= \frac{1}{n \times m^2}
\end{aligned} \tag{A2.11}$$

which is Equation (5).

APPENDIX 2. DERIVATION OF EQUATIONS (4) AND (5).

The probability that cell j in the first section is selected as the first cell of a coalescencing pair

$$= \frac{1}{n \times m} \quad (\text{A2.1})$$

The conditional probability that cell k in the second section is selected as the second cell of the coalescencing pair provided that cell j in the first section is selected as the first cell of the coalescencing pair

$$= \frac{1}{m} \quad (\text{A2.2})$$

The probability that cell j in the i -th section is selected as the first cell of the coalescencing pair

$$= \frac{1}{n \times m} \times \frac{1}{m} \quad (\text{A2.3})$$

The probability that cell k in the second section is selected as the first cell of a coalescencing pair

$$= \frac{1}{n \times m} \quad (\text{A2.4})$$

The conditional probability that cell j in the first section is selected as the second cell of the coalescencing pair provided that cell k in the second section is selected as the first cell of the coalescencing pair

$$= \frac{1}{2 \times m} \quad (\text{A2.5})$$

The probability that cell k in the second section is selected as the first cell of the coalescencing pair and cell j in the first section is selected as the second cell of the coalescencing pair

$$= \frac{1}{n \times m} \times \frac{1}{2 \times m} \quad (\text{A2.6})$$

Therefore, the probability that cell j in the first section and cell k in the second section are selected as the two cells of the coalescencing cell

CHAPTER 4
MIXING OF LARGE PARTICLES IN A TWO-DIMENSIONAL GAS
FLUIDIZED BED - Nonsegregating System

4.1 INTRODUCTION

Solids mixing in a fluidized bed has been the subject of many experimental and theoretical studies. The simplicity of design, the ease of application in continuous systems, and the turbulent motion of particles are attractive characteristics of a fluidized bed which make it a potential device for solids mixing.

The phenomenon of solids mixing in fluidized beds has been described by many investigators (see e.g., Rowe and Partridge, 1962; Kunii and Levenspiel, 1969; Schugerl, 1969), and wake mixing, bubble induced drift and gross circulation have been considered as the three principal mechanisms of solids mixing.

Two models have been used to describe the axial solids mixing in fluidized beds. Brötz (1952), May (1959), Levey et al. (1960), and Massimilla and Westwater (1960) applied essentially linear diffusion models in investigating the effect of system variables on the rate of mixing. Katz and Zenz (1954) and Talmor and Benenati (1963) proposed a circulation model by considering material balances about two completely mixed regions in a fluidized bed and correlated the model with published information.

Few reports are available on the radial solids mixing in fluidized beds despite its practical importance. Brötz (1956) measured the rate of radial mixing in a two-dimensional fluidized bed and assumed a diffusion model to describe the mixing data. Gabor (1964) studied the radial mixing in fluidized and fluidized-packed beds. Mori and Nakamura (1965) reported experimental results of radial mixing and its relation to the bubbling

phenomenon. Wen (1974) concluded that in general, correlations for estimation of the radial solids mixing are lacking.

Research has been conducted mainly in systems where particle sizes are smaller than 500 μm . However, there are some applications of fluidization where the use of large particles can be advantageous (see e.g., Cox, 1958; Anon., 1961; Brown et al., 1972; Vogel et al., 1975; Wilson and Gillmore, 1975). Geldart and Cranfield (1972) carried out some hydrodynamic studies on the gas fluidization of particles varying in size from 0.8 to 2.4 mm. Cranfield and Geldart (1974) reported a bubble formation mechanism and several unusual features for a large particle bed. Correlations for estimation of the solids mixing rate for large particle beds are scarce.

In this study, extremely large particles ($1 \frac{7}{16}$ -in. table tennis or ping-pong balls) were used as fluidizing particles. It should be of interest to note that table tennis balls or similar particles are actually employed in turbulent contactors which are essential three phase fluidized beds (see e.g, Anon., 1959; Kielbach, 1961; Douglas et al., 1963; Douglas, 1964). Both axial mixing and radial mixing were carried out in a two-dimensional gas fluidized bed. The bed employed may be regarded as truly two-dimensional in character, since the dimension of the bed (130-in. x 15-in. x $1 \frac{3}{4}$ -in.) was such that it gave rise to essentially only two-dimensional motion of the particles in the bed. A discrete time stationary random walk model presented by Wang and Fan (1975) has been extended to a continuous time stationary random walk model for the axial and radial solids mixing. The model is a special format of the Markov process which has been applied by many investigators (Oyama and Ayaki, 1956; Oleniczak, 1962; Inoue and Yamaguchi, 1969; Chen et al., 1972; Lai and Fan, 1975) to study the performance of conventional solids mixers.

4.2 THEORETICAL

The motion of particles which leads to axial mixing is described as a one-dimensional process by equally dividing the bed into m fictitious sections in series in the vertical direction (Fig. 1). The motion of particles which leads to radial mixing is described as a one-dimensional process by dividing the bed into m fictitious sections in series in the horizontal direction (Fig. 2). To obtain rate expressions for both mixing process, the following two assumptions are made:

- (A1) The concentration distribution inside a section is spatially uniform and each section is a discrete state.
- (A2) The motion of particles can be described by a random walk process.

The random walk is a Markov chain $\{X_i, i = 1, 2, \dots, m\}$ on a state space consisting of integers with the property that if the system is in a given state i , the system in a single transition either remains at state i or moves to one of the states adjacent to state i ; in other words, the particle can only move to the nearest neighbors (see e.g., Parzen, 1967; Barber et al., 1970). The transition probabilities associated with such a random walk process in an arbitrarily small time interval Δt can be stated in terms of:

- (1) The probability of a key particle transitioning from state i to state $(i-1)$, which is $A_{i,i-1}(t)\Delta t$.
- (2) The probability of a key particle transitioning from state i to state $(i+1)$, which is $A_{i,i+1}(t)\Delta t$.
- (3) The probability of a key particle remaining in state i , which is equal to $\{1 - [A_{i,i-1}(t) + A_{i,i+1}(t)]\}\Delta t$.

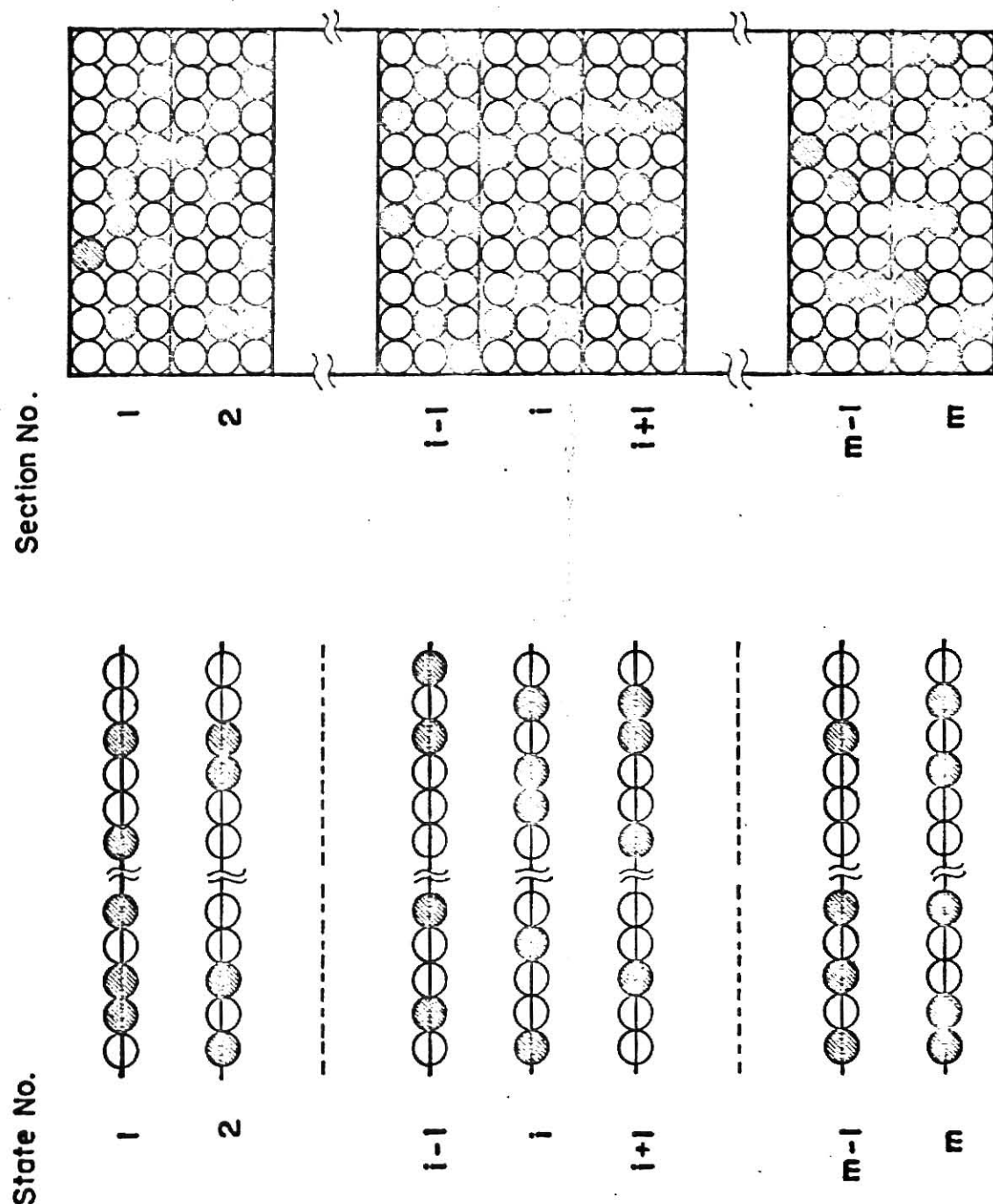


Fig.1. Schematic representation of sections and states for the axial mixing.

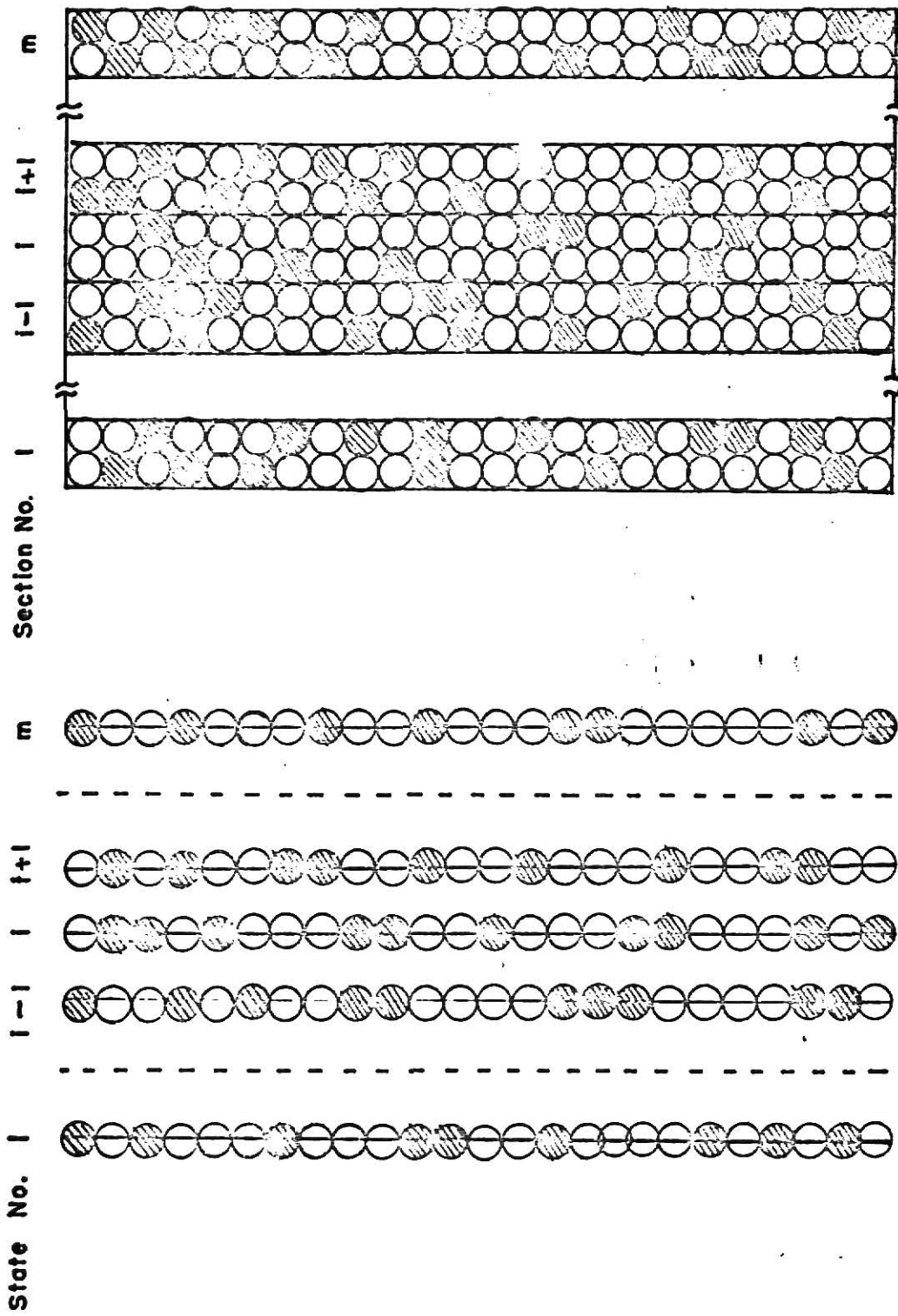


Fig.2. Schematic representation of sections and states for the radial mixing.

In these expressions, $A_{i,i-1}(t)$ and $A_{i,i+1}(t)$ represent the intensities of transition (transition rates) from state i to state $(i-1)$ and state $(i+1)$, respectively. The corresponding transition probability matrix in Δt is of the form:

(see the following pages)

For simplification, two additional assumptions are made:

- (A3) The random walk process can be described as a stationary process, i.e., a process with the intensity of transition remains invariant with respect to time.
- (A4) The intensity of transition from a state to an adjacent state remains spatially invariant.

Under these assumptions,

$$A_{1,2} = \alpha \tag{2}$$

$$A_{i,i-1} = A_{i,i+1} = \alpha, \quad i = 2, 3, \dots, m-1 \tag{3}$$

$$A_{m,m-1} = \alpha \tag{4}$$

where α is a positive real constant. Thus Equation (1) becomes

(see the following pages)

(1)

0	0	0	0
0	0	0	0
0	0	0	0
\cdot	\cdot	\cdot	\cdot
\cdot	\cdot	\cdot	\cdot
\cdot	0	0	0
$1 - [A_{m-2,m-3}(t) + A_{m-2,m-1}(t)]\Delta t$	$A_{m-2,m-1}(t)\Delta t$		0
$A_{m-1,m-2}(t)\Delta t$	$1 - [A_{m-1,m-2}(t) + A_{m-1,m}(t)]\Delta t$	$A_{m-1,m}(t)\Delta t$	
0	$A_{m,m-1}(t)\Delta t$	$1 - A_{m,m-1}(t)\Delta t$	

$$\underline{P} = [P_{i,j}] =$$

$$\begin{bmatrix} 1-A_{1,2}(t)\Delta t & A_{1,2}(t)\Delta t & 0 & 0 & 0 & 0 \\ A_{2,1}(t)\Delta t & 1-[A_{2,1}(t) + A_{2,3}(t)]\Delta t & A_{2,3}(t)\Delta t & 0 & 0 & 0 \\ 0 & A_{3,2}(t)\Delta t & 1-[A_{3,2}(t) + A_{3,4}(t)]\Delta t & A_{3,4}(t)\Delta t & 0 & 0 \\ 0 & 0 & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ 0 & 0 & 0 & 0 & \cdot & A_{m-2,m-3}(t)\Delta t \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{bmatrix}$$

$$\begin{bmatrix}
 1-\alpha\Delta t & \alpha\Delta t & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
 \alpha\Delta t & 1-2\alpha\Delta t & \alpha\Delta t & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
 0 & \alpha\Delta t & 1-2\alpha\Delta t & \alpha\Delta t & 0 & 0 & 0 & 0 & 0 & 0 \\
 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
 \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\
 \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\
 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
 \end{bmatrix}$$

$$P = [P_{i,j}] =$$

Mathematically, the concentration of key particles at $(t+\Delta t)$ can be related to the concentration of key particles at t and the corresponding transition probability matrix as (Parzen, 1967)

$$E\{C(t+\Delta t)\} = \underline{C}(t) \underline{P} \text{ or } C_j = \sum_{i=1}^m C_i P_{i,j}, \quad j = 1, 2, \dots, m \quad (6)$$

where

$$\underline{C}(t) = [C_1(t) \ C_2(t) \ \dots \ C_i(t) \ \dots \ C_{m-1}(t) \ C_m(t)] \quad (7)$$

Accordingly,

$$E\{C(t+\Delta t)\} = C_1(t) (1 - \alpha \Delta t) + C_2(t) \alpha \Delta t \quad (8)$$

$$E\{C(t+\Delta t)\} = C_{i-1}(t) \alpha \Delta t + C_i(t) (1 - 2\alpha \Delta t) + C_{i+1}(t) \alpha \Delta t, \quad (9)$$

$$i = 2, 3, \dots, m-1$$

$$E\{C(t+\Delta t)\} = C_m(t) (1 - \alpha \Delta t) + C_{m-1}(t) \alpha \Delta t \quad (10)$$

Equations (8), (9), and (10) can be simplified by carrying out multiplication, collecting common terms, dividing through by Δt , and taking the limit as $\Delta t \rightarrow 0$ to obtain

$$E\left\{\frac{dC_1(t)}{dt}\right\} = \alpha C_2(t) - \alpha C_1(t) \quad (11)$$

$$E\left\{\frac{dC_i(t)}{dt}\right\} = \alpha C_{i-1}(t) - 2\alpha C_i(t) + \alpha C_{i+1}(t), \quad (12)$$

$$i = 2, 3, \dots, m-1$$

$$E\left\{\frac{dC_m(t)}{dt}\right\} = \alpha C_{m-1}(t) - \alpha C_m(t) \quad (13)$$

To determine fluctuation of the concentration of the key particles in the bed, the variance can be calculated as

$$\sigma^2(t) = \frac{1}{m-1} \sum_{i=1}^m [C_i(t) - \bar{C}]^2 \quad (14)$$

where \bar{C} denotes the mean number concentration of the key particles in the bed. The degree of mixedness, $M(t)$, is calculated according to the definition

$$M(t) = 1 - \frac{\sigma^2(t)}{\sigma_o^2} \quad (15)$$

where σ_o^2 is the variance of the completely segregated state of the mixture.

4.3 EXPERIMENTAL

The experimental set-up, particle system, and procedure are described below.

4.3.1 Set-Up

The schematic diagram of the experimental apparatus and equipment is shown in Fig. 3. The main section of the bed was 130-in. high, 15-in. wide and $1\frac{3}{4}$ -in. thick. The height of the particle bed was indicated by a scale attached to the bed. The bed was supported by a drilled standard steel distributor which contained 156 $\frac{3}{64}$ -in. holes arranged in square pitch. A $15\frac{1}{2}$ -in. calming section beneath the distributor consisted of two packed sections, one at the bottom with $\frac{7}{8}$ -in. glass beads, the other at the top with $1\frac{7}{16}$ -in. sand filled ping-pong balls. A manometer attached to the bed was used to measure the pressure drops across the distributor and the bed. Air flow from a compressor was controlled with a needle valve and measured with a rotameter. The air was directed through the bed or to the vent line by a 3-way valve to minimize the transient period during the start-up.

1. Rotary Vane Air compressor

2. Control Valves

3. Rotameters

4. Thermometer

5. Pressure Gauge

6. Control Valve

7. 3-Way Valve

8. Sphere Packing

9. Distributer

10. 2-D Fluidized Bed

11. Manometers

12. Thermometer

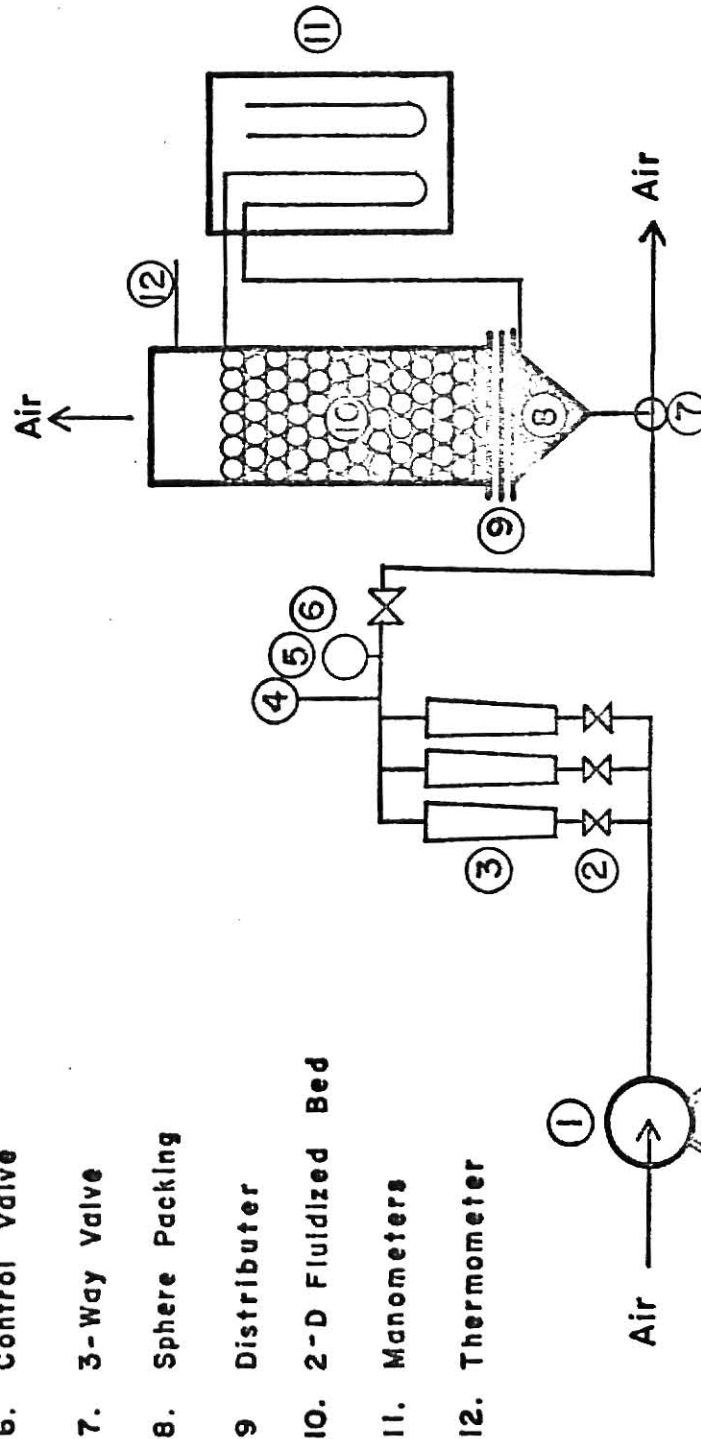


Fig.3. Schematic diagram of experimental apparatus.

4.3.2 Particle System

Halex 3-star table tennis balls, $1\frac{7}{16}$ -in. in diameter and 2.42 g in weight were used as fluidizing particles. The balls in white were denoted as key particles and the balls in yellow as nonkey particles throughout the experiment. The particle diameter was measured by the displacement volume in water. No significant difference in size and weight was observed between the white and yellow particles. The minimum fluidization rate of the particle at 76-in. static bed height was found to be 6.78 ft./sec. by measuring pressure drops across the particle bed at several air rates. A full-log plot of the pressure drop vs. the air rate was used to determine the minimum fluidization rate.

4.3.3 Procedure

For determination of axial mixing, the key particles were poured into the bed first and the nonkey particles were carefully layered on top. The air from the compressor was metered and allowed to vent-off initially through the by-pass line fitted with the 3-way valve. The 3-way valve was then quickly switched, forcing the air to flow through the bed, and timing started. After a predetermined period of mixing, the by-pass line was opened to terminate the fluidization. The number of key particles and that of nonkey particles in each section were recorded.

Two sets of runs were devised to test the intermediate termination and reinitiation of the fluidization on mixing. In the first, each experiment was started with a fresh charge and the experiment was terminated when the fluidization was terminated. In the second, the operating conditions were identical with those of the first except that the bed was collapsed and refluidized between successive mixing interval. The

difference in the results of the two sets was not significant at the 5% confidence level. All other experiments for the axial mixing were run in the second operating mode.

For determination of radial mixing, the key particles were charged to one side of a centrally located vertical partition and the nonkey particles were charged to the other side. After the particles were fluidized, the partition was quickly removed, allowing the particles to mix. Each experiment was started with a fresh charge and the experiment was terminated when the fluidization was terminated.

Throughout each experiment, the air temperature was kept at $25.0^{\circ}\text{C} \pm 0.5$ and the pressure on the top of the bed was 1 atmosphere.

4.4 RESULTS AND DISCUSSION

4.4.1 Fluidization Characteristics

Each particle oscillated slightly around a fixed position in the bed prior to the onset of bubbling. Transition from the bubbling-bed condition to the slugging-bed condition was almost immediate. This may be typical for fluidizing particles of large size and low density (Brown, 1972). Lenticular slugs were generated at 10-20 in. above the distributor and rose slowly through the bed because of its shape. Occasionally, a rising slug was broken into smaller irregular bubbles by the down-flowing particles, which rose through the bed more rapidly than the original slug probably due to their more streamlined shapes.

The minimum bubbling rate, U_b , was observed to be approximately $1.3 \times U_{mf}$ as compared to $1.2 \times U_{mf}$ reported by Rowe and Sutherland (1964) for their small particle bed (0.4-0.5 mm). No appreciable solids mixing

occurred below the minimum bubbling rate. Bubble induced drift and gross solids circulation appeared to be predominant solids mixing mechanisms in the large particle bed of the present study. The contribution from wake mixing appeared to be minor.

4.4.2 Axial Solids Mixing

Figures 4a through 4d give axial concentration distributions of the key particles respectively at the mixing times of 30 seconds, 60 seconds, 90 seconds, and 150 seconds at an air rate of 9.77 ft./sec. These figures show the overall progress of the axial mixing process. Appreciable scattering of the data was expected since the system was essentially a stochastic system with a small population (see e.g., Asaki and Kondo, 1965; King, 1968) containing only about 30 balls in each section.

Concentration distributions predicted by the proposed model are also presented in Figs. 4a through 4d. Concentration predicted by the model is generally higher than the experimental data in the upper part of the bed and the opposite is true in the lower part of the bed. These discrepancies have probably arisen from the fact that the distribution of the axial mixing intensity along the bed is not taken into account in the model. Obviously, the motion of particles directly above the gas distributor was different from that in the main body. The particles may be completely stagnant, semi-stagnant, or fully mobile. Combination of these various types of behavior was found across the distributor in the present study. The mixing pattern in the top layer of the bed also was different from that in the main body of the bed because the slugs continually broke up the surface and the particles were scattered over the surface. In order to derive a model which can take into account these various types of complex

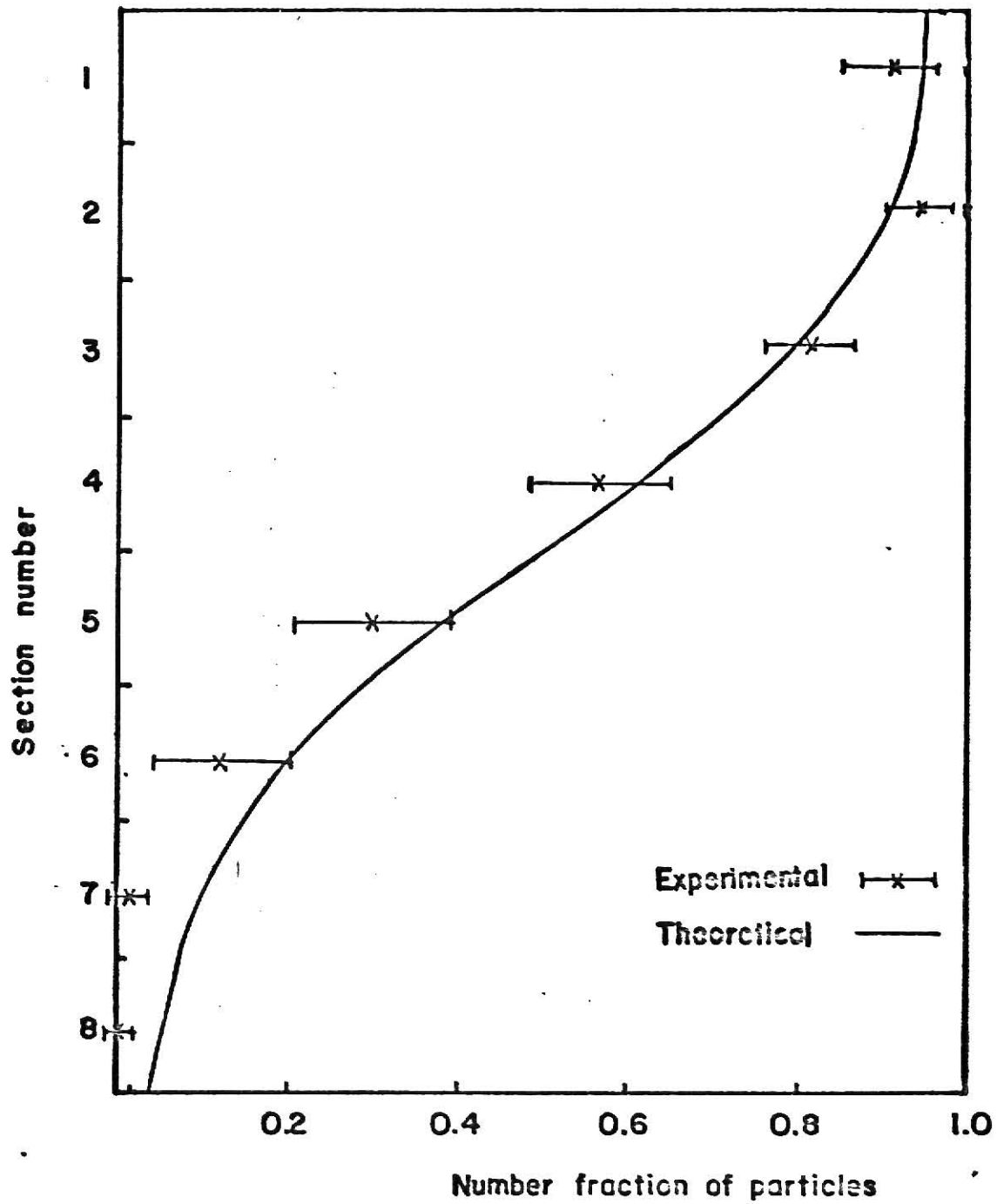


Fig. 4 a. Axial concentration distribution of particles after 30 seconds of mixing at an air rate of 9.77 ft./sec..

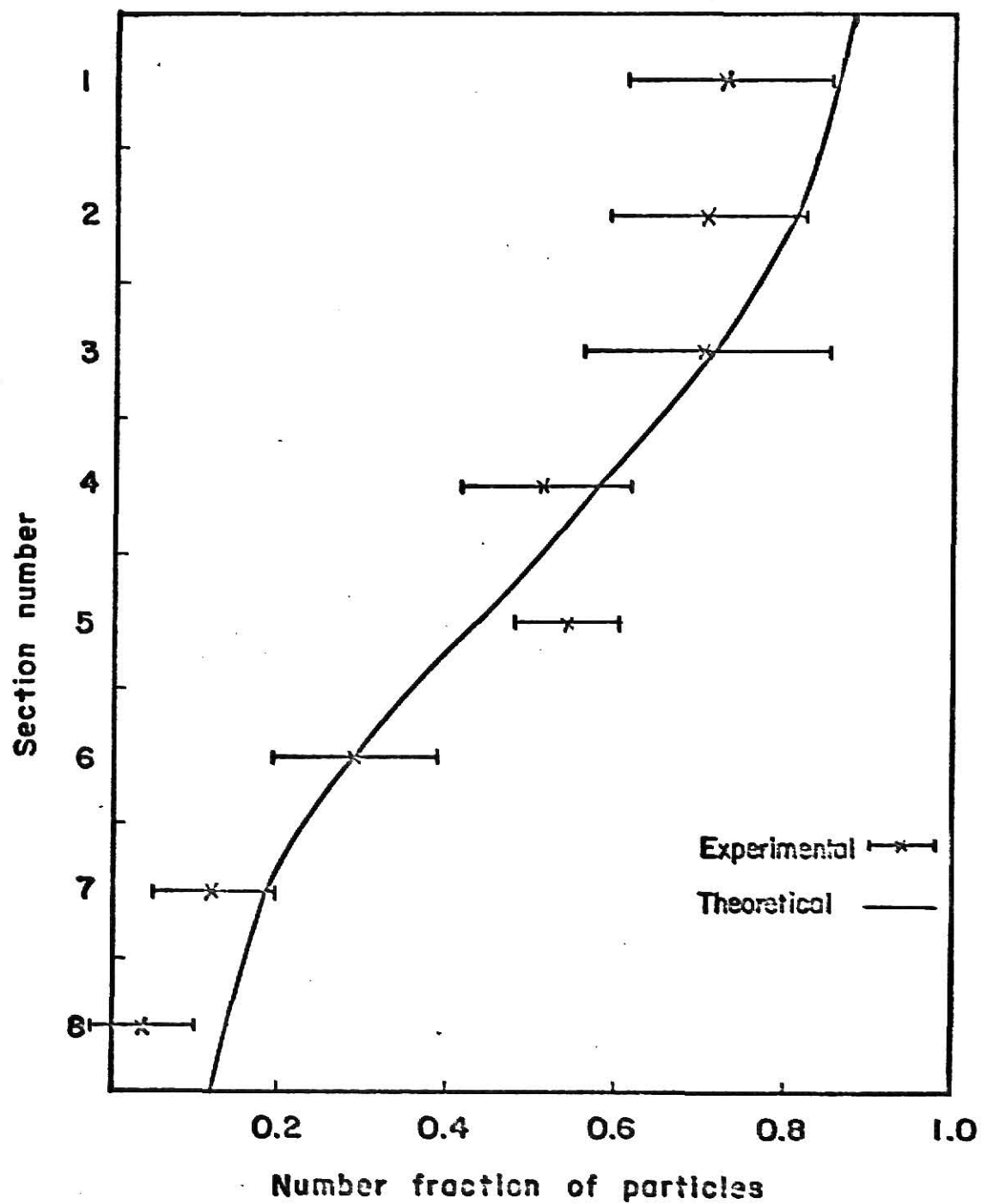


Fig.4 b. Axial concentration distribution of particles after 60 seconds of mixing at air rate 9.77 ft/sec .

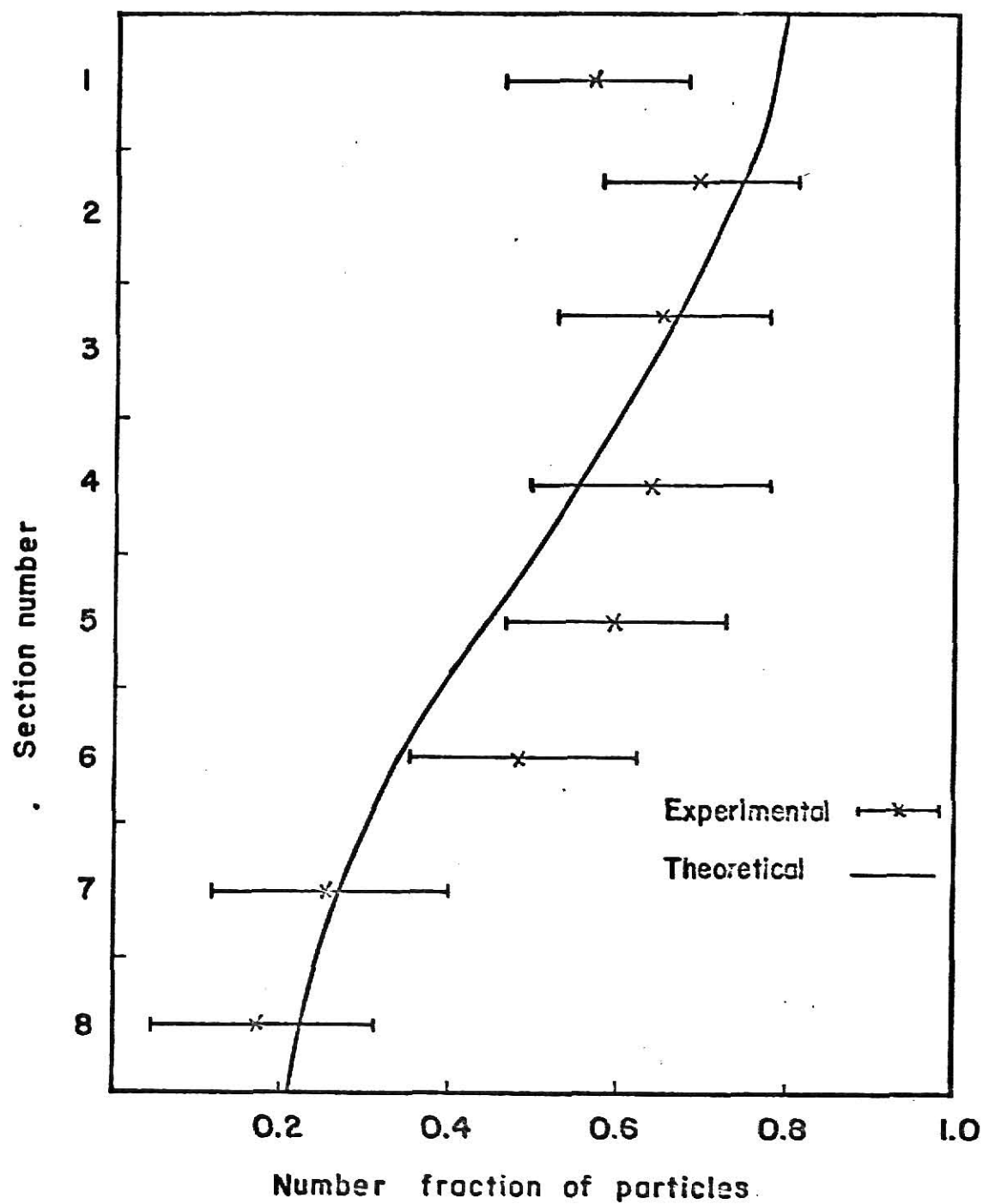


Fig.4c. Axial concentration distribution of particles after 90 seconds of mixing at air rate 9.77 ft/sec .

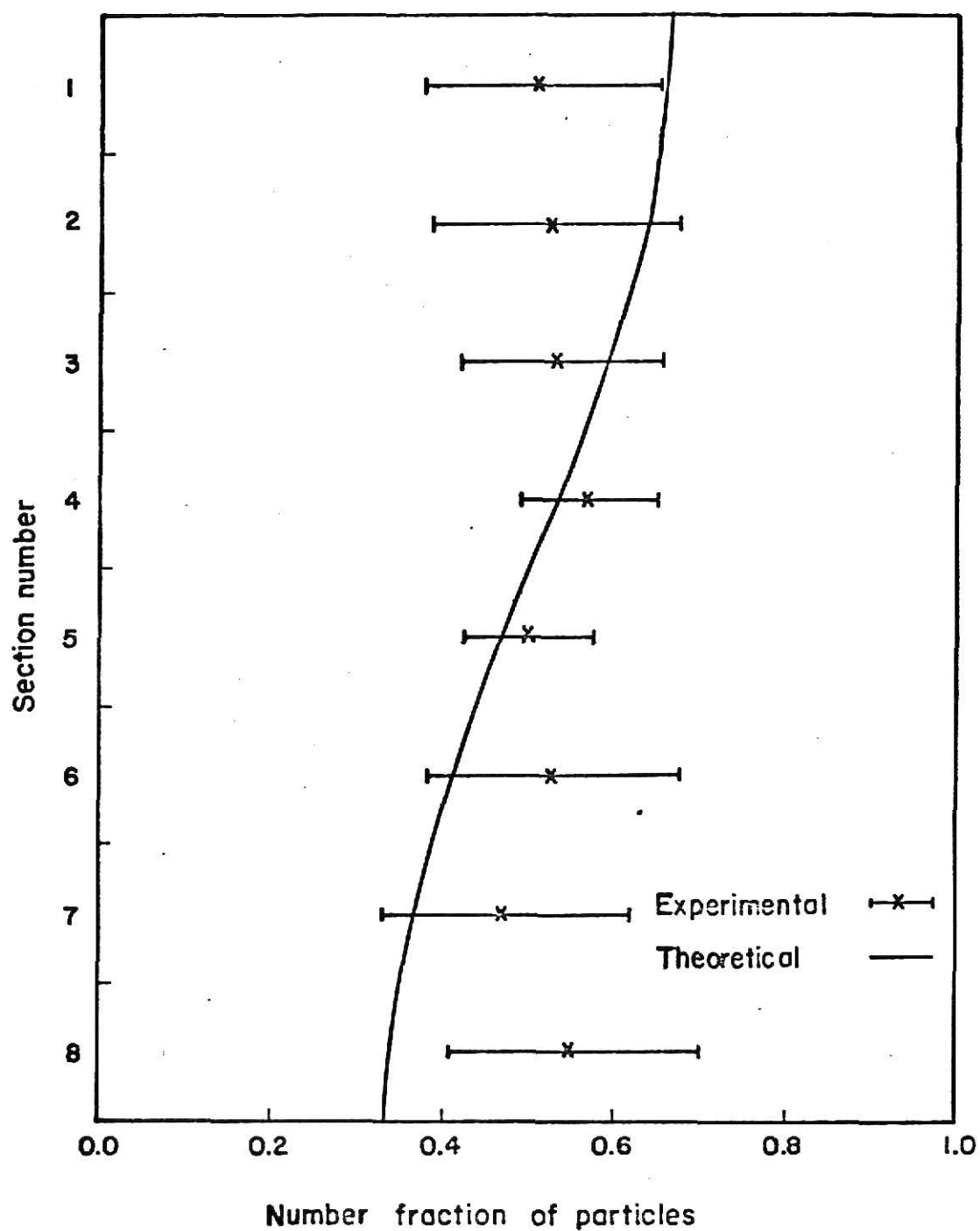


Fig. 4d. Axial concentration distribution of particles after 150 seconds of mixing at air rate 9.77 ft/sec.

behavior, additional analysis and experimentation are required.

The degree of mixedness of a mixture can be determined from Equation (15). A plot of the degree of mixedness against the mixing time provides an indication of the overall progress of mixing. As shown in Fig. 5, the rate of axial mixing is rapid initially and decreases with the mixing time. When the mixing time was prolonged, the degree of mixedness was reduced, indicating a segregation tendency in the system. This segregation tendency might have resulted from the surface property differences between the particles.

The effect of air rate on the rate of axial mixing is shown in Fig. 5. The increases in the rate of axial mixing with the increase in the air rate can be readily understood because the mixing in a gas fluidized bed is predominantly induced by bubbles which increase both in number and size when the air rate is increased.

Theoretical prediction of the degree of mixedness as a function of the mixing time is also shown in Fig. 5. Values of the parameter (see Table 1), α , is plotted against $(U - U_b)^2$ as shown in Fig. 6, which indicates the following correlation:

$$\alpha = 0.038 \times (U - U_b)^2 \quad (16)$$

This implies that no mixing occurs when the air rate is less than the minimum bubbling rate. The expression of α given by Equation (16) is similar to the expression of the first-order rate constant proposed by Nicholson and Smith (1966) for small particles. In other words, the excess air rate which is responsible for formation of bubbles should be $(U - U_b)$ for a large particle bed instead of $(U - U_{mf})$ which has been conventionally used for a small particle bed.

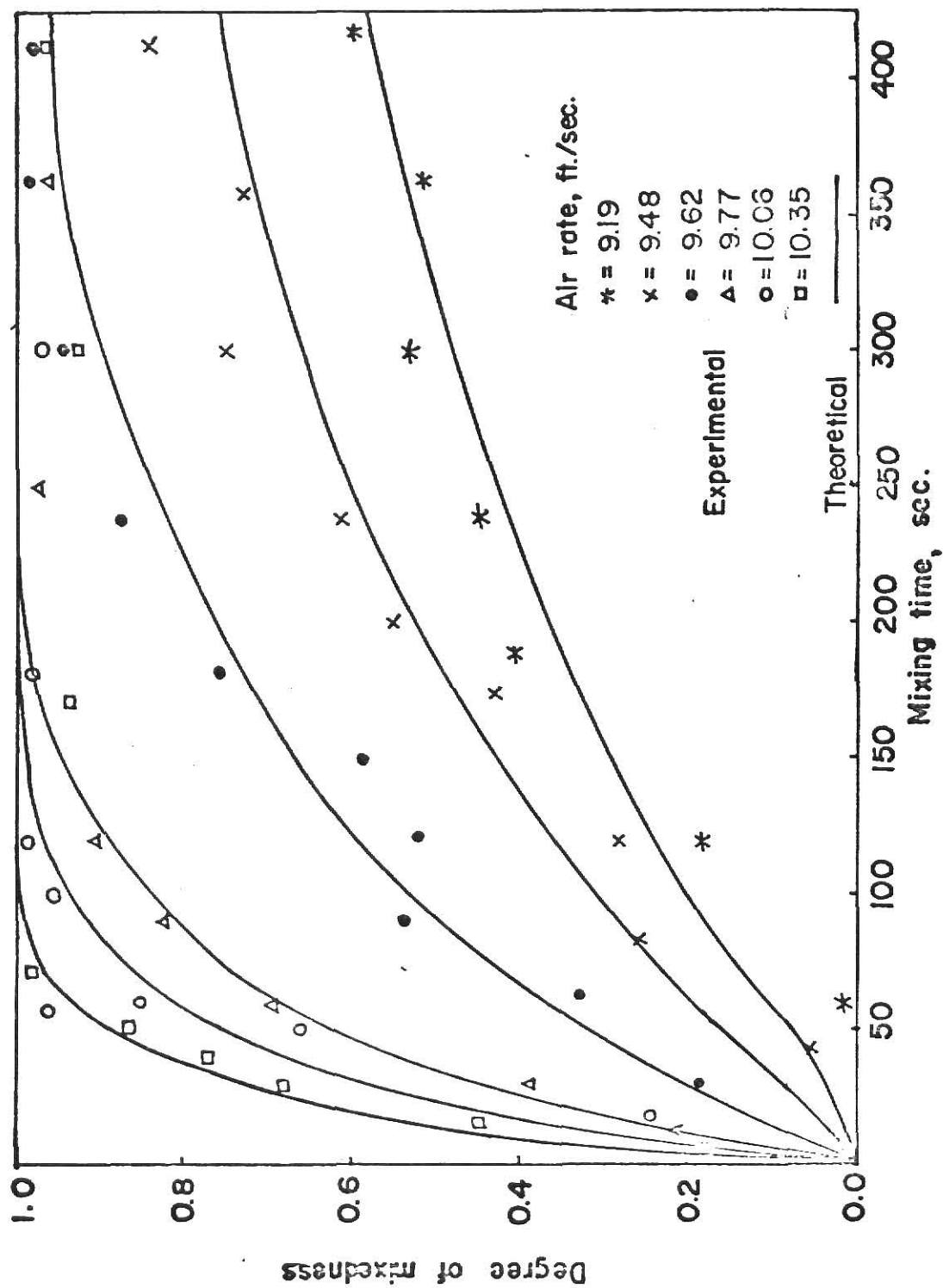


Fig.5 Degree of mixedness vs. mixing time for the axial mixing.

Table 1

Parameter of the Random Walk Model
for the Axial Solids Mixing.

<u>Air Rate, ft./sec.</u>	<u>α, sec.⁻¹</u>	<u>Standard Deviation of fitting</u>
9.19	3.9×10^{-3}	0.057
9.48	6.5×10^{-3}	0.068
9.62	1.4×10^{-2}	0.063
9.77	3.5×10^{-2}	0.029
10.06	5.4×10^{-2}	0.034
10.35	8.8×10^{-2}	0.045

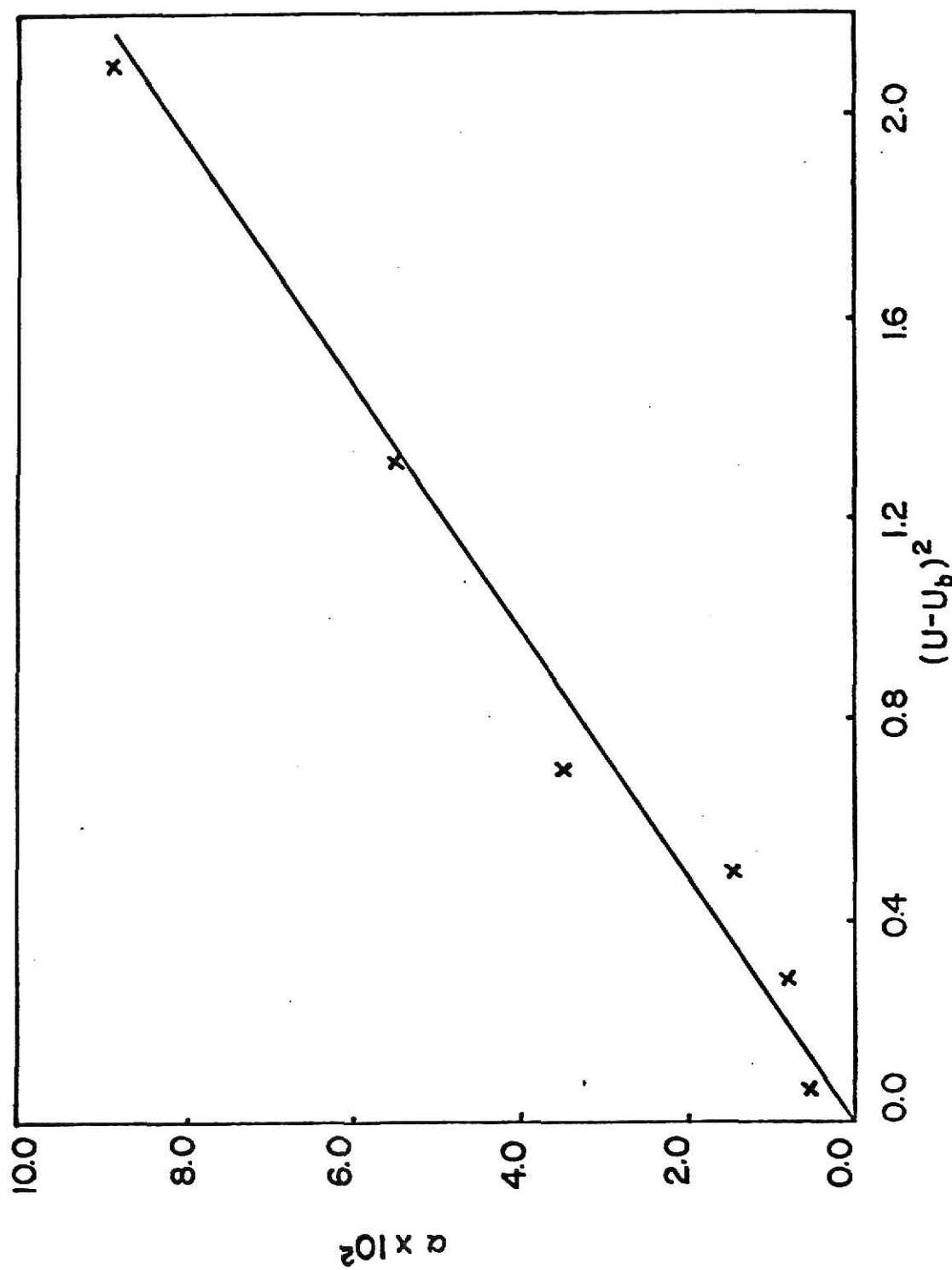


Fig.6 The effect of air rate on α' for the axial mixing.

4.4.3 Radial Solids Mixing

The overall progress of radial solids mixing in terms of the degree of mixedness is presented in Fig. 7. No appreciable segregation occurred in the radial direction. The degree of mixedness predicted by the model is also shown in the same figure. Values of the parameter, α , estimated from the data shown in Fig. 7 are presented in Table 2 and plotted against $(U - U_b)^2$ as shown in Fig. 8, indicating that α varies with the excess air rate as

$$\alpha = 0.11 \times (U - U_b)^2 \quad (17)$$

According to Gabor (1964), the rate of radial solids mixing is proportional to $(U - U_{mf})^{1.25}$ for a small particle bed (0.196 mm). Stronger dependence of the rate of radial solids mixing on the excess air rate was reported by Brötz (1956), and Mori and Nakamura (1965) for large particle beds (0.4-0.5 mm and 0.6 mm, respectively). It appears that the dependence of the rate of radial solids mixing on the excess air rate is increased with the size of particles.

Equations (16) and (17) suggest that radial solids mixing is more rapid than axial solids mixing. However, such a conclusion may not be true for other fluidized beds of different dimensions and particles because of the probable change in the predominant mixing mechanisms.

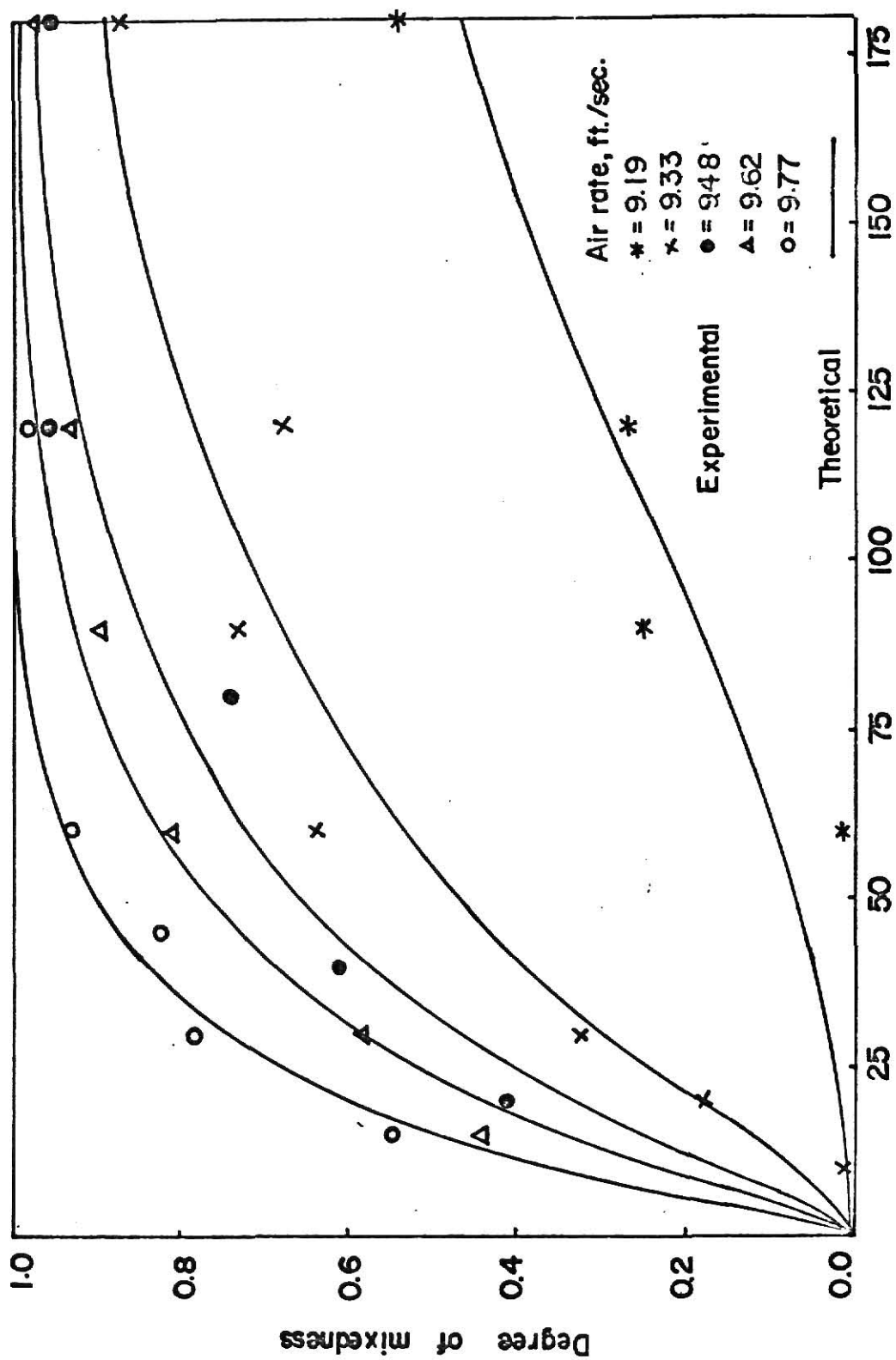


Fig.7 Degree of mixedness vs. mixing time for the radial mixing.

Table 2

Parameter of the Random Walk Model
for the Radial Solids Mixing.

<u>Air Rate, ft./sec.</u>	<u>α , sec.⁻¹</u>	<u>Standard Deviation of fitting</u>
9.19	5.4×10^{-3}	0.071
9.33	2.4×10^{-2}	0.068
9.48	3.9×10^{-2}	0.057
9.62	5.5×10^{-2}	0.030
9.77	8.6×10^{-2}	0.042

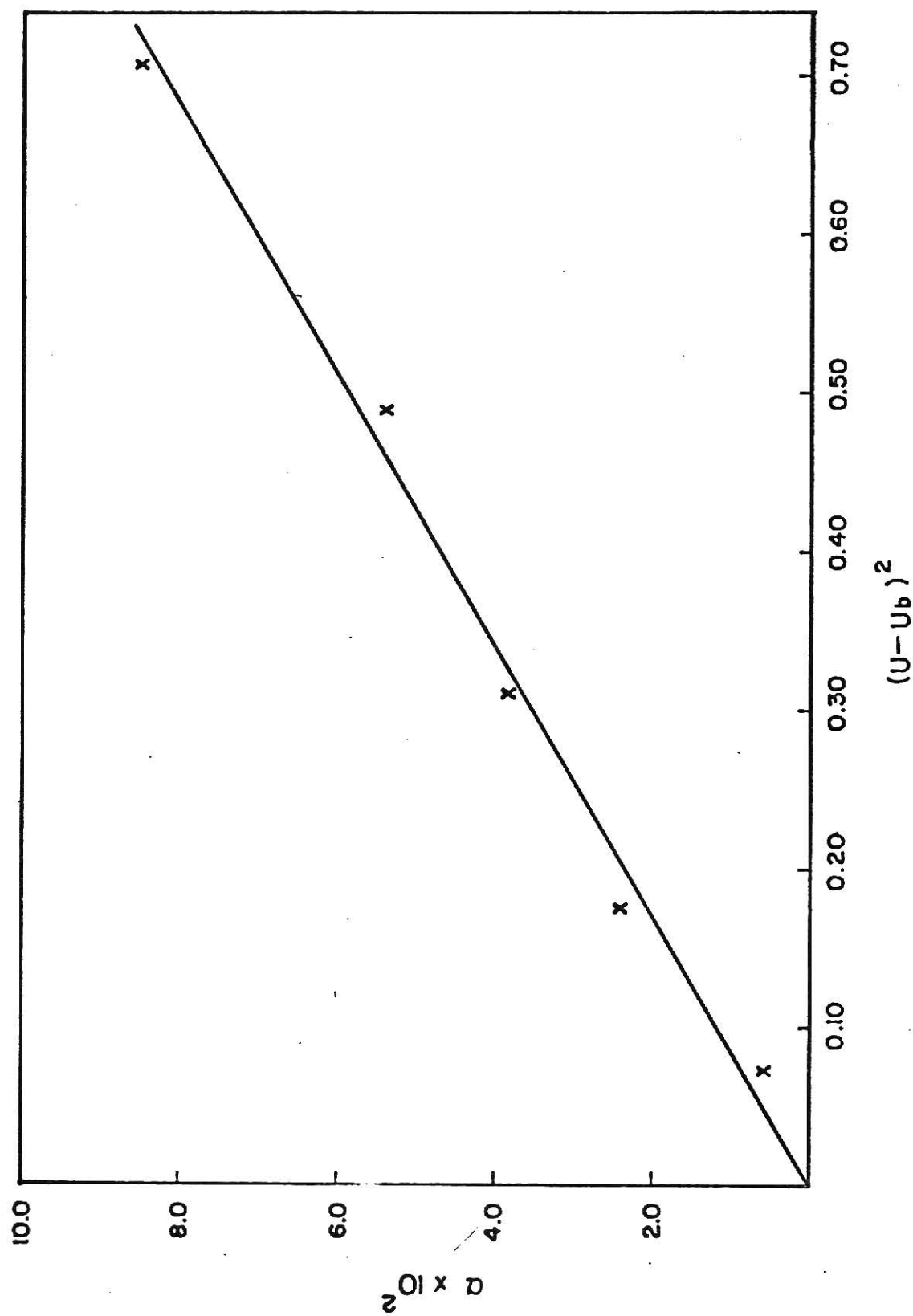


Fig.8. The effect of air rate on α for the radial mixing.

4.5 CONCLUSIONS

Several unusual features were observed. Each particle oscillated slightly around a fixed-position in the bed prior to the onset of bubbling. The transition from the bubbling-bed condition to the slugging-bed condition was almost immediate. The minimum bubbling rate, U_b , was observed to be approximately $1.3 \times U_{mf}$ as compared to $1.2 \times U_{mf}$ reported by Rowe and Sutherland (1964) for their small particle bed (0.3-0.5 mm). No appreciable solids mixing occurred below the minimum bubbling rate. Bubble induced drift and gross solids circulation appeared to be predominant solids mixing mechanisms in this large particle bed. The contribution from wake mixing appeared to be negligible.

A stationary random walk model which is equally applicable to the axial and radial solids mixings, was successfully employed to represent the experimental data. It was found that the rate of solids mixing can be correlated as a function of the excess air rate.

NOTATION

$A_{i,i-1}(t)$ = intensity of transition from state i to state $(i-1)$ at time t

$A_{i,i+1}(t)$ = intensity of transition from state i to state $(i+1)$ at time t

$C_i(t)$ = concentration of key particles in section i at time t

$C_j(t+\Delta t)$ = concentration of key particles in section j at time $t+\Delta t$.

m = number of sections

$M(t)$ = degree of mixedness at time t

$\underline{P}(t)$ = transition matrix at time t

U = air rate

U_b = minimum bubbling rate

U_{mf} = minimum fluidization rate

\bar{C} = mean number concentration of key particles

Greek Letters

α = parameter in the model

$\sigma^2(t)$ = variance at time t

$\sigma_o^2(t)$ = variance of the completely segregated state

Table A1. Fluidization data

Static bed height in.	Air rate ft./sec.	Pressure drop across the bed in. H ₂ O	Pressure drop across the distributor in. H ₂ O	Bed expansion %
76.2	2.92	0.10	negligible	0
76.2	3.65	0.20	negligible	0
76.2	4.37	0.30	0.05	0
76.2	7.29	0.65	0.15	negligible
76.2	8.02	0.70	0.20	negligible
76.2	8.75	0.75	0.20	1
76.2	9.48	0.80	0.25	4
76.2	10.21	0.85	0.30	13
76.2	10.94	0.90	0.35	35

Table A2a. Axial concentration distribution of the key (white) particle after 30 seconds of mixing at an air rate of 9.77 ft./sec.

<u>Run no.</u>	<u>Section 1</u>	<u>Section 2</u>	<u>Section 3</u>	<u>Section 4</u>	<u>Section 5</u>	<u>Section 6</u>	<u>Section 7</u>	<u>Section 8</u>
1	0.000	0.000	0.111	0.556	0.769	0.899	0.958	1.000
2	0.086	0.033	0.038	0.407	0.571	0.741	1.000	1.000
3	0.111	0.000	0.036	0.385	0.621	0.950	1.000	1.000
4	0.000	0.000	0.071	0.385	0.786	0.931	1.000	1.000
5	0.041	0.057	0.132	0.445	0.745	0.876	0.958	1.000

Table A2b. Axial concentration distribution of the key (white) particles after 60 seconds of mixing at an air rate of 9.77 ft./sec.

<u>Run no.</u>	<u>Section 1</u>	<u>Section 2</u>	<u>Section 3</u>	<u>Section 4</u>	<u>Section 5</u>	<u>Section 6</u>	<u>Section 7</u>	<u>Section 8</u>
1	0.175	0.172	0.172	0.414	0.444	0.862	0.920	1.000
2	0.372	0.286	0.111	0.267	0.407	0.607	0.840	1.000
3	0.183	0.179	0.290	0.533	0.408	0.750	0.846	0.946
4	0.274	0.459	0.483	0.479	0.476	0.646	0.746	0.855
5	0.242	0.361	0.378	0.444	0.561	0.711	0.872	1.000

Table A2c. Axial concentration distribution of the key (white) particles after 90 seconds of mixing at an air rate of 9.77 ft./sec.

<u>Run no.</u>	<u>Section 1</u>	<u>Section 2</u>	<u>Section 3</u>	<u>Section 4</u>	<u>Section 5</u>	<u>Section 6</u>	<u>Section 7</u>	<u>Section 8</u>
1	0.528	0.586	0.520	0.423	0.444	0.370	0.458	0.571
2	0.337	0.300	0.345	0.500	0.519	0.524	0.769	0.852
3	0.436	0.192	0.172	0.423	0.400	0.621	0.875	0.852
4	0.610	0.536	0.429	0.115	0.172	0.393	0.761	0.889
5	0.353	0.192	0.300	0.357	0.519	0.667	0.800	0.926

Table A2d. Axial concentration distribution of the key (white) particles after 150 seconds of mixing at an air rate of 9.77 ft./sec.

<u>Run no.</u>	<u>Section 1</u>	<u>Section 2</u>	<u>Section 3</u>	<u>Section 4</u>	<u>Section 5</u>	<u>Section 6</u>	<u>Section 7</u>	<u>Section 8</u>
1	0.453	0.393	0.556	0.464	0.519	0.607	0.630	0.556
2	0.657	0.552	0.360	0.333	0.448	0.444	0.542	0.529
3	0.538	0.679	0.621	0.536	0.586	0.276	0.296	0.333
4	0.526	0.429	0.333	0.379	0.407	0.393	0.520	0.186
5	0.516	0.593	0.621	0.586	0.393	0.259	0.481	0.522

Table A3. Degree of mixedness for the axial mixing

Air rate ft./sec.	Mixing time sec.		20	30	40	60	80	120	180	240	300	360	420
9.19			--	--	--	0.018	--	0.183	0.411	0.442	0.508	0.489	0.573
9.48			--	--	0.052	--	0.258	0.271	0.407	0.618	0.759	0.715	0.841
9.62			--	0.171	--	0.323	--	0.512	0.742	0.898	0.937	0.981	0.979
9.77			--	0.397	--	0.639	--	0.861	0.948	0.966	0.921	0.976	--
10.06			0.234	0.507	--	0.823	--	0.982	0.977	--	0.958	--	--
10.35			0.435	0.683	0.769	0.962	0.947	0.944	0.912	--	0.934	--	--

Table A4. Degree of mixedness for the radial mixing

Air rate ft./sec.	Mixing time sec.		10		15		20		30		40		60		90		120		180	
9.19			--		--		--		--		--		0.018		0.263		0.276		0.510	
9.33			0.017		--		0.183		0.328		--		0.624		0.725		0.663		0.831	
9.48			--		--		0.428		--		0.605		--		--		0.971		0.969	
9.62			--		0.436		--		0.588		--		0.810		0.903		0.942		0.979	
9.77			--		0.543		--		0.789		--		0.927		--		0.984		--	

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CHAPTER 5
MIXING OF LARGE PARTICLES IN A TWO-DIMENSIONAL
GAS FLUIDIZED BED - Segregating System

5.1 INTRODUCTION

Solids mixing in a fluidized bed has been the subject of many experimental and theoretical studies. The simplicity of design, the ease of application in continuous systems, and the turbulent motion of particles have been most frequently mentioned as the attractive characteristics associated with a fluidized bed, which make it a potential device for solids mixing (see e.g., Kunii and Levenspiel, 1969).

It has been known that gas fluidized beds have excellent and rapid mixing characteristics for nonsegregating particle systems. Much effort, both experimental and theoretical, has been spent in explaining this fact (see e.g., Brotz, 1952; May, 1960; Levey et al., 1960; Kunii and Levenspiel, 1969). However, in industrial solids mixing, it is often required to mix particles which differ widely in physical properties. In fluidized beds containing particles of different physical properties, segregation which prevents the attainment of a random state of mixing is usually encountered.

The forces which promote particle segregation in a fluidized bed have been discussed by Sutherland and Wong (1964), and Pruden and Epstein (1964). Though there has been moderate interest in studying the segregation behavior of a mechanically homogenized mixture during fluidization (see e.g., Wen and Yu, 1966; Gelperin et al., 1967; Pena et al., 1968; Goossens et al., 1971), little has been done in investigating

the mixing of segregating particles in a fluidized bed. Nicholson and Smith (1966) studied the axial mixing of particles differing in density in a fluidized bed and proposed a first-order rate equation to describe the progress of mixing in the short mixing time. Nienow et al. (1972) reported the role of particle size, particle density, and air rate on the segregation behavior in a gas fluidized bed. They concluded that a fairly wide particle size difference can be tolerated without appreciable segregation while even a small density difference leads to readily settling of the denser particles. It should be of interest to note that this is in contrast to the situation in conventional mixers, where the size difference is the most important segregation factor (see e.g., Williams, 1975). Gibilaro and Rowe (1974) formulated a model based on four physical mechanisms: overall particle circulation, interchange between wake and bulk phases, axial dispersion, and segregation. Experimentally observed equilibrium concentration distributions can be satisfactorily predicted from this model.

In this study, extremely large particles ($1 \frac{7}{16}$ -in.) with density difference were used as fluidizing particles. Both axial and radial mixings were carried out in a two-dimensional gas fluidized bed. The bed employed may be regarded as truly two-dimensional in character, since the dimension of the bed (130-in. x 15-in. x $1 \frac{3}{4}$ -in.) was such that it gave rise to essentially only two-dimensional motion of the particles in the bed. A nonstationary random walk model has been developed to represent the axial concentration distribution in a segregating fluidized particle system as a function of the operating time.

5.2 THEORETICAL

The motion of particles which leads to axial mixing is described as a one-dimensional process by equally dividing the entire length of the bed into m fictitious sections in series in the vertical direction, with the last section being that adjacent to the gas distributor (Fig. 1).

To obtain the rate expression for the mixing process, the following assumptions are made:

- (A1) The concentration distribution inside a section is spatially uniform and each section is a discrete state.
- (A2) The motion of particles can be described by a random walk process. The random walk is a Markov chain $\{X_i, i = 1, 2, \dots, m\}$ on a state space consisting of integers with the property that if the system is in a given state i , then in a single transition the system either remains at state i or moves to one of the states immediately adjacent to state i ; in other words, the particle can only move to the nearest neighbors (see e.g., Parzen, 1967; Barber et al., 1970).
- (A3) The bed consists of two mixing regions, the segregated region at the bottom or lower part of the bed and the completely mixed region at the top or the upper part of the bed. The proportion of the two regions is dependent upon the characteristics of particles, the geometry of the bed and the operating conditions.
- (A4) The random walk process in the completely mixed region is a stationary process, i.e., a process with the intensity of transition (transition rate) invariant with respect to time.
- (A5) The random walk process in the segregated region is a nonstationary

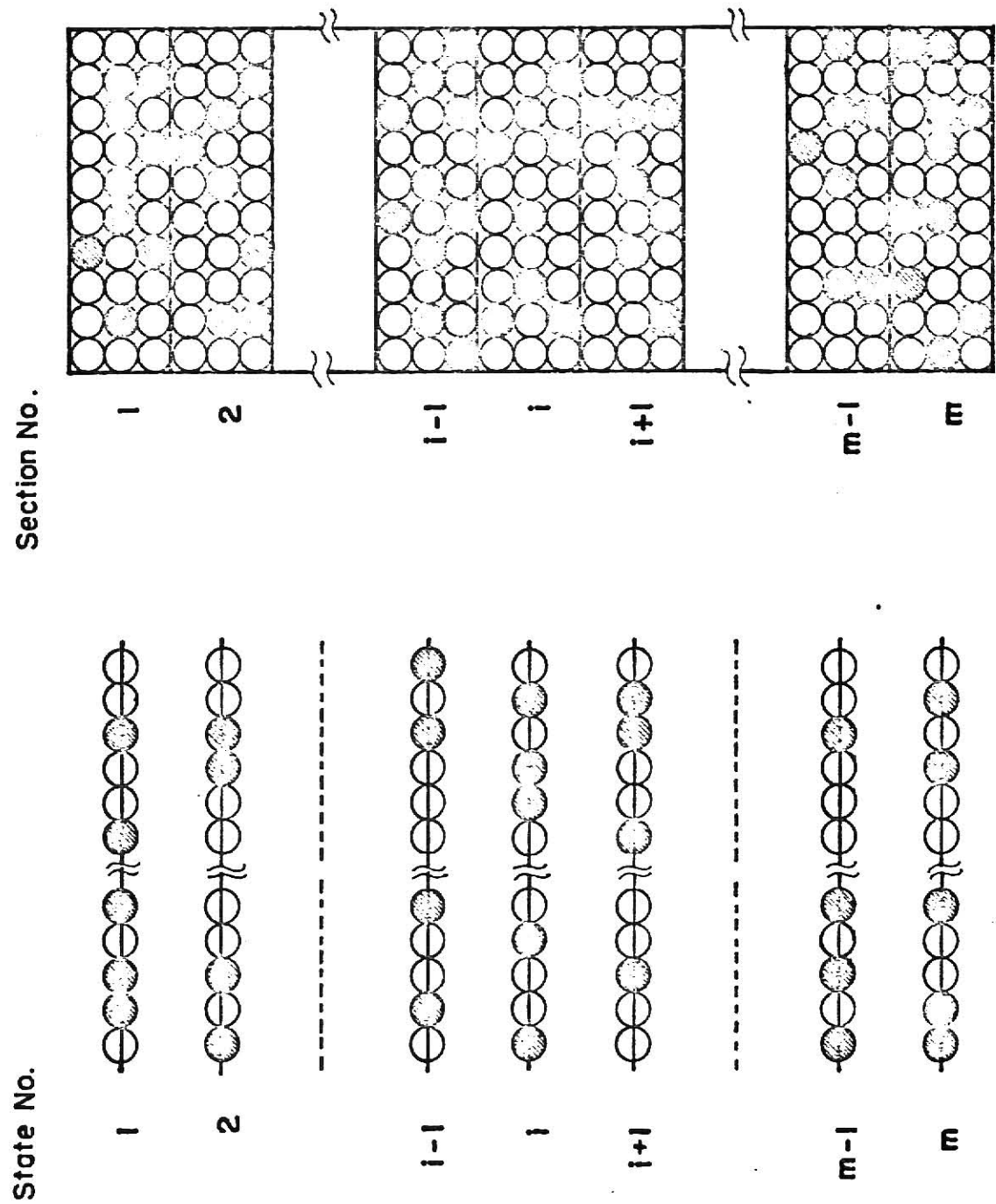


Fig.1. Schematic representation of sections and states for the axial mixing.

process, i.e., a process with the intensity of transition as a function of time.

Under these assumptions, the transition probability matrix associated with the mixing process in an arbitrarily small time interval Δt can be written as

$$\text{(see the following pages)} \quad (1)$$

where

$A_{i,i-1}(t)$ = intensity of transition of a key particle from state i to state $(i-1)$,

$A_{i,i+1}(t)$ = intensity of transition of a key particle from state i to state $(i+1)$,

k = section number above which complete mixing occurs, i.e., the border line between state $(k-1)$ and state k divided the completely mixed region and the segregated region.

The expressions, $A_{i,i-1}(t) \Delta t$ and $A_{i,i+1}(t) \Delta t$, which appear in the matrix, represent respectively the probability of the key particle transition from state i to state $(i-1)$ and state $(i+1)$ in Δt , consequently, $\{1 - [A_{i,i-1}(t) + A_{i,i+1}(t)] \Delta t\}$ represents the probability of the key particle remaining in state i . For further simplification, the following additional assumptions are made:

(A6) In the completely mixed region, the intensity of transition from a state to an adjacent state remains spatially invariant. Thus,

$$A_{1,2} = \alpha \quad (2)$$

$$A_{i,i-1} = A_{i,i+1} = \alpha, \quad i = 2, 3, \dots, k-1 \quad (3)$$

where α is a positive real constant.

(A7) Only interactions between particles in the adjacent states are

(1) - 1

$$\begin{aligned}
 & \left[\begin{array}{cccccccccccccccc}
 1-A_{1,2}\Delta t & A_{1,2}\Delta t & 0 & 0 & \dots & 0 & 0 & 0 & 0 \\
 A_{2,1}\Delta t & 1-(A_{2,1}+A_{2,3})\Delta t & A_{2,3}\Delta t & 0 & \dots & 0 & 0 & 0 & 0 \\
 0 & A_{3,2}\Delta t & 1-(A_{3,2}+A_{3,4})\Delta t & A_{3,4}\Delta t & \dots & 0 & 0 & 0 & 0 \\
 0 & 0 & \cdot & \cdot & \dots & \cdot & \cdot & \cdot & \cdot \\
 \cdot & \cdot & \cdot & \cdot & \dots & \cdot & \cdot & \cdot & \cdot \\
 \cdot & \cdot & \cdot & \cdot & \dots & \cdot & \cdot & \cdot & \cdot \\
 0 & 0 & 0 & 0 & \dots & A_{k-1,k-2}\Delta t & 1-(A_{k-1,k-2}+A_{k-1,k})\Delta t & A_{k-1,k}\Delta t \\
 0 & 0 & 0 & 0 & \dots & 0 & A_{k,k-1}(\tau)\Delta t & 1-(A_{k,k-1}(\tau)+A_{k,k+1}(\tau))\Delta t \\
 \cdot & \cdot & \cdot & \cdot & \dots & \cdot & \cdot & \cdot \\
 \cdot & \cdot & \cdot & \cdot & \dots & \cdot & \cdot & \cdot \\
 \cdot & \cdot & \cdot & \cdot & \dots & \cdot & \cdot & \cdot \\
 0 & 0 & 0 & 0 & \dots & 0 & 0 & 0 \\
 0 & 0 & 0 & 0 & \dots & 0 & 0 & 0 \\
 0 & 0 & 0 & 0 & \dots & 0 & 0 & 0
 \end{array} \right]
 \end{aligned}$$

$$\underline{P}(\tau) = [P_{1,j}(\tau)] =$$

(1)

(1) - 2

0	...	0	0	0	0	0	0	0	0
0	...	0	0	0	0	0	0	0	0
0	...	0	0	0	0	0	0	0	0
.
.
.
0	...	0	0	0	0	0	0	0	0
$A_{k,k+1}(t)\Delta t$...	0	0	0	0	0	0	0	0
.
.
.	0
0	...	$A_{m-2,m-3}(t)\Delta t$	$1-[A_{m-2,m-3}(t)+A_{m-2,m-1}(t)]\Delta t$	$A_{m-2,m-1}(t)\Delta t$	0	0	0	0	0
0	...	0	$A_{m-1,m-2}(t)\Delta t$	$1-[A_{m-1,m-2}(t)+A_{m-1,m}(t)]\Delta t$	$A_{m-1,m}(t)\Delta t$	0	0	0	0
0	...	0	0	0	$A_{m,m-1}(t)\Delta t$	$1-A_{m,m-1}(t)\Delta t$	0	0	0

significant in the segregated region. Those between particles in the same state and those between particles beyond the adjacent states are ignored. The transition of a particle in the segregated region is the result of an interchange of states between a pair of particles in the adjacent states. For a key particle in state i , four possible combinations of the key particles in state $(i-1)$ and state $(i+1)$, are possible as shown in Fig. 2. Associated with each combination, two transitions, each with a specific intensity, may occur. The intensity of each transition is primarily dependent upon the stability of the original state and that of the final state of that transition. Assuming that the key particle is lighter than the nonkey particle, the key particle will be more stable in the upper state [state $(i-1)$] than in the lower state [state i] and vice versa. Any transition from a less stable state to a more stable one is classified as a favored transition; that from a more stable state to a less stable one is classified as an unfavored transition. If the stability of a system remains unchanged, the transition is classified as an equivalent or neutral transition.

The intensities of the three transitions are assumed to be:

$$(8) \quad \text{The intensity of an equivalent transition} = \alpha ,$$

$$(9) \quad \text{The intensity of a favored transition} = \alpha + \beta ,$$

$$(10) \quad \text{The intensity of an unfavored transition} = \alpha - \beta ,$$

In these expressions, β is a positive real constant which is less than or equal to α . Thus, magnitudes of the intensity of transition can be assigned with reference to Fig. 2 and are summarized in Table 1. Accordingly, the intensity of a key particle transitioning from state i to state $(i-1)$ is

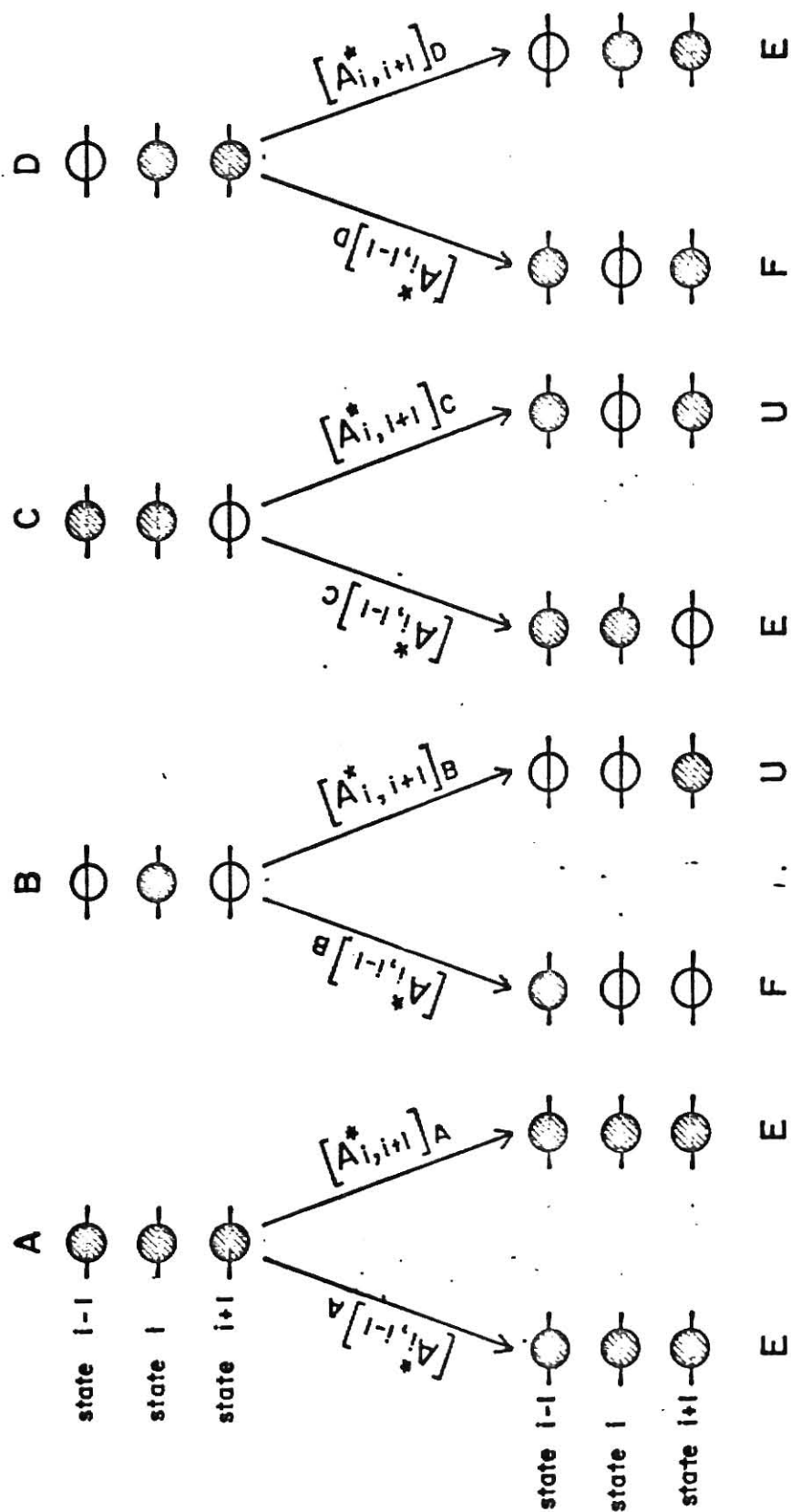


Fig.2. Possible combinations of a key particle in state i with particles in the adjacent states and its possible transitions (● = key particle; ○ = non key particle; E = equivalent transition; F = favored transition; U = unfavored transition).

Table 1

Occurrence probabilities and transition probabilities
for combinations A, B, C, and D in Fig. 2.

<u>Combination</u>	<u>Probability of occurrence</u>	<u>$A_{i,i-1}^*$</u>	<u>$A_{i,i+1}^*$</u>
A	$C_{i-1}(t) \times C_{i+1}(t)$	α	α
B	$[1-C_{i-1}(t)] \times [1-C_{i+1}(t)]$	$\alpha+\beta$	$\alpha-\beta$
C	$C_{i-1}(t) \times [1-C_{i+1}(t)]$	α	$\alpha-\beta$
D	$[1-C_{i-1}(t)] \times C_{i+1}(t)$	$\alpha+\beta$	α

$$\begin{aligned}
& A_{i,i-1}(t) \\
&= C_{i-1}(t) C_{i+1}(t) [A_{i,i-1}^*]_A + [1 - C_{i-1}(t)] [1 - C_{i+1}(t)] [A_{i,i-1}^*]_B \\
&\quad + C_{i-1}(t) [1 - C_{i+1}(t)] [A_{i,i-1}^*]_C + [1 - C_{i-1}(t)] C_{i+1}(t) [A_{i,i-1}^*]_D \\
&= \alpha + \beta - \beta C_{i-1}(t), \quad i = k, k+1, \dots, m \quad (4)
\end{aligned}$$

Similarly, the intensity of a key particle transitioning from state i to state $(i+1)$ is

$$\begin{aligned}
& A_{i,i+1}(t) \\
&= C_{i-1}(t) C_{i+1}(t) [A_{i,i+1}^*]_A + [1 - C_{i-1}(t)] [1 - C_{i+1}(t)] [A_{i,i+1}^*]_B \\
&\quad + C_{i-1}(t) [1 - C_{i+1}(t)] [A_{i,i+1}^*]_C + [1 - C_{i-1}(t)] C_{i+1}(t) [A_{i,i+1}^*]_D \\
&= \alpha - \beta + \beta C_{i+1}(t), \quad i = k, k+1, \dots, m-1 \quad (5)
\end{aligned}$$

Substituting Equations (2), (3), (4) and (5) into Equation (1), the transition probability matrix can be rewritten as

$$(\text{see following pages}) \quad (6)$$

Mathematically, the concentration of key particles at $(t+\Delta t)$ can be related to the concentration of the key particles at t and the corresponding transition probability matrix as

$$E\{\underline{C}(t+\Delta t)\} = \underline{C}(t)\underline{P}(t) \quad \text{or} \quad C_j = \sum_{i=1}^m C_i P_{i,j}, \quad j = 1, 2, \dots, m \quad (7)$$

where

$$\underline{C}(t) = [C_1(t) \ C_2(t) \ \dots \ C_i(t) \ \dots \ C_{m-1}(t) \ C_m(t)] \quad (8)$$

Accordingly,

$$E\{C_1(t+\Delta t)\} = C_1(t)(1-\alpha\Delta t) + C_2(t)\alpha\Delta t \quad (9)$$

$$E\{C_i(t+\Delta t)\} = C_{i-1}(t)\alpha\Delta t + C_i(t)(1 - 2\alpha\Delta t) + C_{i+1}(t)\alpha\Delta t, \quad (10)$$

$$i = 2, 3, \dots, k-2$$

(6) - 2

...	0	0	0	0	0	0
...	0	0	0	0	0	0
...	0	0	0	0	0	0
...
...
...
...	0	0	0	0	0	0
...	0	0	0	0	0	0
...
...
...
...	[$\alpha + \beta - \beta C_{m-3}(t)$] Δt	$1 - [2\alpha - \beta C_{m-3}(t) + \beta C_{m-1}(t)]\Delta t$	[$\alpha + \beta - \beta C_{m-1}(t)$] Δt	0	0	0
...	0	[$\alpha + \beta - \beta C_{m-2}(t)$] Δt	$1 - [2\alpha - \beta C_{m-2}(t) + \beta C_{m-1}(t)]\Delta t$	[$\alpha + \beta - \beta C_m(t)$] Δt		
...	0	0	[$\alpha + \beta - \beta C_{m-1}(t)$] Δt	$1 - [\alpha + \beta - \beta C_{m-1}(t)]\Delta t$		

(6)

Continue to Page 123.

$$E\{C_{k-1}(t+\Delta t)\} = C_{k-2}(t)\alpha\Delta t + C_{k-1}(t)(1 - 2\alpha\Delta t) + C_k(t)[\alpha + \beta - \beta C_{k-1}(t)]\Delta t \quad (11)$$

$$E\{C_k(t+\Delta t)\} = C_{k-1}(t)\alpha\Delta t + C_k(t)\{1 - [2\alpha - \beta C_{k-1}(t) + \beta C_{k+1}(t)]\Delta t\} + C_{k+1}(t)[\alpha + \beta - \beta C_k(t)]\Delta t \quad (12)$$

$$E\{C_i(t+\Delta t)\} = C_{i-1}(t)[\alpha - \beta + \beta C_i(t)]\Delta t + C_i(t)\{1 - [2\alpha - \beta C_{i-1}(t) + \beta C_{i+1}(t)]\Delta t\} + C_{i+1}(t)[\alpha + \beta - \beta C_i(t)]\Delta t, \quad i = k+1, k+2, \dots, m-1 \quad (13)$$

$$E\{C_m(t+\Delta t)\} = C_{m-1}(t)[\alpha - \beta + \beta C_m(t)]\Delta t + C_m(t)\{1 - [\alpha + \beta - \beta C_{m-1}(t)]\Delta t\} \quad (14)$$

Equations (9) through (14) can be simplified by carrying out multiplication, collecting common terms, dividing through Δt , and taking the limit as $t \rightarrow 0$ to obtain

$$E\left\{\frac{dC_1(t)}{dt}\right\} = -\alpha C_1(t) + \alpha C_2(t) \quad (15)$$

$$E\left\{\frac{dC_i(t)}{dt}\right\} = \alpha C_{i-1}(t) - 2\alpha C_i(t) + \alpha C_{i+1}(t), \quad i = 2, 3, \dots, k-2 \quad (16)$$

$$E\left\{\frac{dC_{k-1}(t)}{dt}\right\} = \alpha C_{k-2}(t) - 2\alpha C_{k-1}(t) + [\alpha + \beta - \beta C_{k-1}(t)]C_k(t) \quad (17)$$

$$E\left\{\frac{dC_k(t)}{dt}\right\} = \alpha C_{k-1}(t) - [2\alpha - \beta C_{k-1}(t) + \beta C_{k+1}(t)]C_k(t) + [\alpha + \beta - \beta C_k(t)]C_{k+1}(t) \quad (18)$$

$$E\left\{\frac{dC_i(t)}{dt}\right\} = [\alpha - \beta + \beta C_i(t)]C_{i-1}(t) - [2\alpha - \beta C_{i-1}(t) + \beta C_{i+1}(t)]C_i(t) + [\alpha + \beta - \beta C_i(t)]C_{i+1}(t), \quad i = k+1, k+2, \dots, m-1 \quad (19)$$

$$E\left\{\frac{dC_m(t)}{dt}\right\} = [\alpha - \beta + \beta C_m(t)]C_{m-1}(t) - [\alpha + \beta - \beta C_{m-1}(t)]C_m(t) \quad (20)$$

When $\beta=0$ and/or $k=0$, the nonstationary random walk model is reduced to the stationary random walk model (Chapter 4).

$$E\left\{\frac{dC_1(t)}{dt}\right\} = \alpha C_2(t) - \alpha C_1(t) \quad (21)$$

$$E\left\{\frac{dC_i(t)}{dt}\right\} = \alpha C_{i-1}(t) - 2\alpha C_i(t) + \alpha C_{i+1}(t) , i = 2, 3, \dots, m-1 \quad (22)$$

$$E\left\{\frac{dC_m(t)}{dt}\right\} = \alpha C_{m-1}(t) - \alpha C_m(t) \quad (23)$$

The stationary random walk model can be applied to systems where segregation does not occur.

The motion of particles which leads to radial mixing is described as a one-dimensional process by equally dividing the bed into m fictitious sections in series in the horizontal direction (Fig. 3). It is postulated that the density difference between the particles does not give rise to the radial segregation. Provided that this is true, the stationary random walk model, expressed as Equations (21) through (23), can also be applied to representation of the radial mixing process.

To determine fluctuation of the concentration of the key particles in the bed, the variance can be calculated as

$$\sigma^2(t) = \frac{1}{m-1} \sum_{i=1}^m [C_i(t) - \bar{C}]^2 \quad (24)$$

where \bar{C} denotes the mean number concentration of the key particles in the bed. The degree of mixedness, $M(t)$, is calculated according to the definition

$$M(t) = 1 - \frac{\sigma^2(t)}{\sigma_o^2} \quad (25)$$

where σ_o^2 is the variance of the completely segregated state of the mixture.

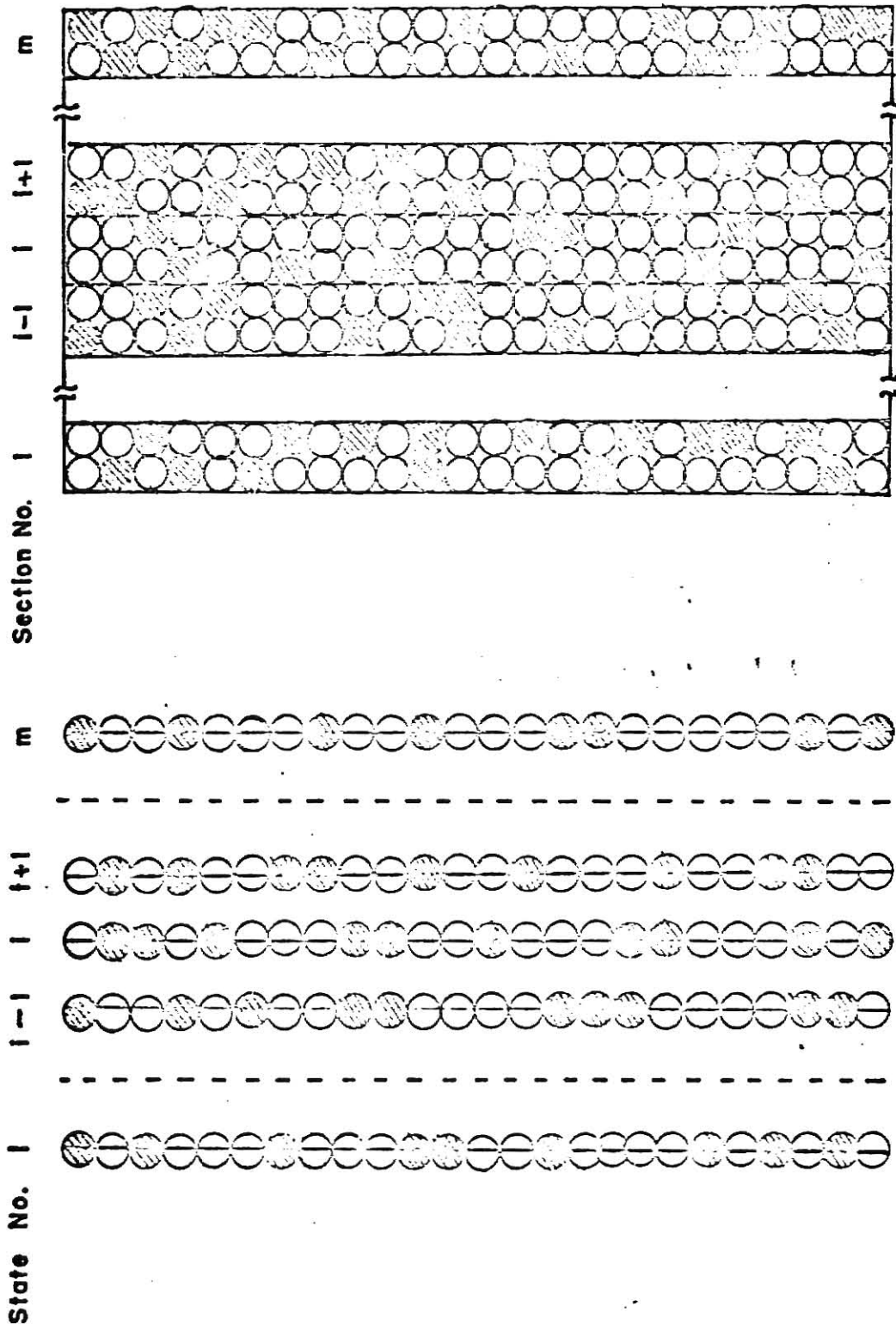


Fig.3. Schematic representation of sections and states for the radial mixing.

5.3 EXPERIMENTAL

The experimental set-up, particle system, and procedures are described below.

5.3.1 Set-Up

The schematic diagram of the experimental apparatus and equipment is shown in Fig. 4. The main section of the bed was 130-in. high, 15-in. wide and $1\frac{3}{4}$ -in. thick. The height of the particle bed was indicated by a scale attached to the bed. The bed was supported by a drilled standard steel distributor which contained 156 $\frac{3}{64}$ -in. holes arranged in square pitch. A $15\frac{1}{2}$ -in. calming section beneath the distributor consisted of two packed sections, one at the bottom with $\frac{7}{8}$ -in. glass beads, the other at the top with $1\frac{7}{16}$ -in. sand filled ping-pong balls. A manometer attached to the bed was used to measure the pressure drops across the distributor and the bed. Air flow from a compressor was controlled with a needle valve and measured with a rotameter. The air was directed through the bed or to the vent line by a 3-way valve to minimize the transient period during the start-up.

5.3.2 Particle System

Halex 3-star table tennis balls, $1\frac{7}{16}$ -in. in diameter and 2.42 g in weight, were used throughout the experiment as key particles. Nonkey particles were the same sized table tennis balls, of which the weight was changed to be 2.50 g by sealing dried sand inside. The minimum fluidization rate of the balls was found by measuring pressure drops across the particle bed at several air rates. A full-log plot of the pressure drop vs. the air rate was used to determine the minimum fluidization rate. This rate was determined to be 6.72 ft./sec. for 2.42 g table tennis balls and 6.80 ft./sec. for the 2.50 g table tennis balls at 57-in. static bed height.

1. Rotary Vane Air compressor

2. Control Valves

3. Rotameters

4. Thermometer

5. Pressure Gauge

6. Control Valve

7. 3-Way Valve

8. Sphere Packing

9. Distributer

10. 2-D Fluidized Bed

11. Manometers

12. Thermometer

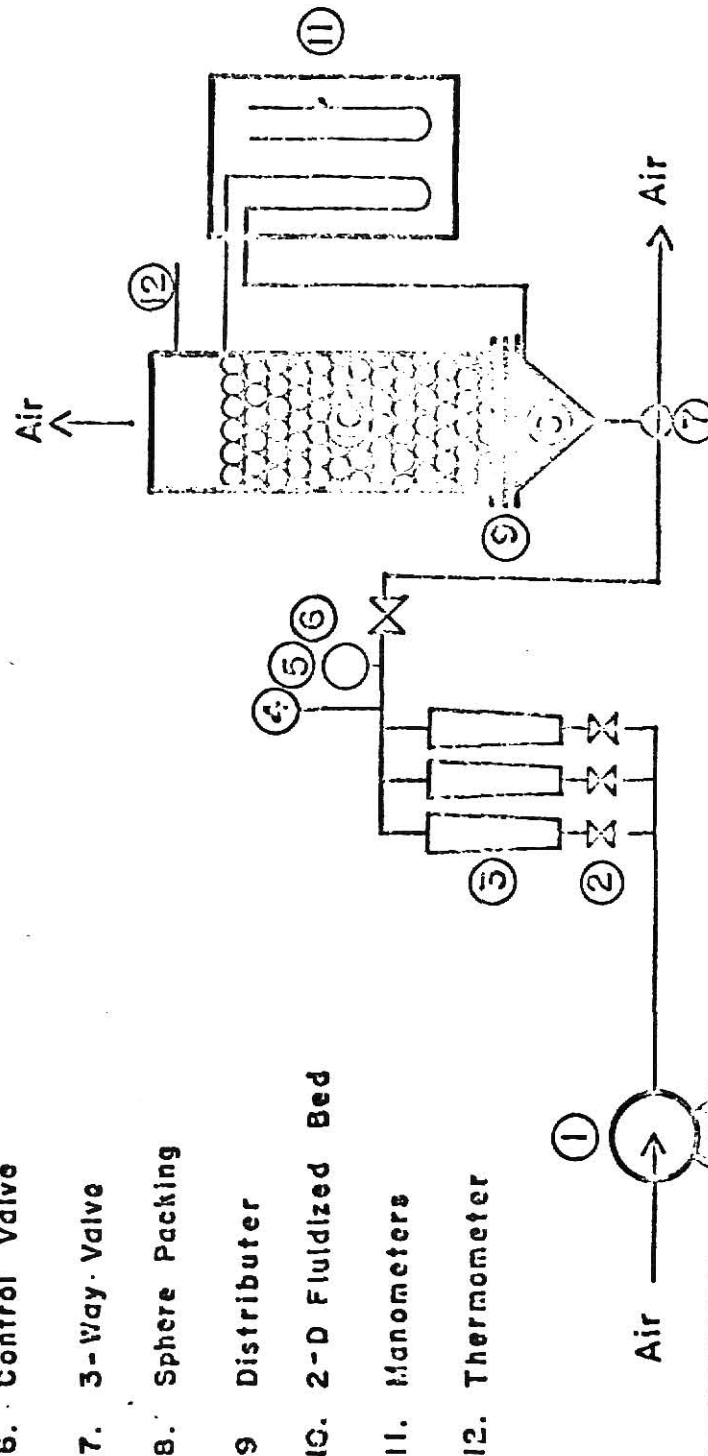


Fig.4. Schematic diagram of experimental apparatus.

5.3.3 Procedures

For determination of axial mixing, one type of particle was poured into the bed first and then the second component was carefully layered on top. The air from the compressor was metered and allowed to vent-off initially through the by-pass line fitted with the 3-way valve. This valve was then quickly switched, forcing the air to flow through the bed with a timer started. After a predetermined period of mixing, the by-pass line was opened to terminate the fluidization. The number of key particles and that of nonkey particles in each section were recorded.

Two sets of runs were devised to test the intermediate termination and reinitiation of the fluidization on mixing. In the first, each experiment was started with a fresh charge and the experiment was terminated when the fluidization was terminated.

In the second, the operating conditions were identical with those of the first except that the bed was collapsed and refluidized between successive mixing intervals. The difference in the results of the two sets was not significant at the 2.5% confidence level. All other experiments for the axial mixing were run in the second operating mode.

For determination of radial mixing, the key particles were charged to one side of a centrally located vertical partition and the nonkey particles were charged to the other side. After the particles were fluidized, the partition was quickly removed, allowing the particles to mix. Each experiment was started with a fresh charge and the experiment was terminated.

Throughout each experiment, the air temperature was kept at $25.0^{\circ}\text{C} \pm 0.5$ and the pressure on the top of the bed was 1 atmosphere.

5.4 RESULTS AND DISCUSSION

5.4.1 Fluidization Characteristics

Each particle oscillated slightly around a fixed position in the bed prior to the onset of bubbling. Transition from the bubbling-bed condition to the slugging-bed condition was almost immediate. This may be typical for fluidizing particles of large size and low density (Brown, 1972).

Lenticular slugs were generated at 10-20 in. above the distributor and rose slowly through the bed because of their shapes. Occasionally, a rising slug was broken into smaller irregular bubbles by the down-flowing particles, which rose through the bed more rapidly than the original slug probably due to their more streamlined shapes. The minimum bubbling rate, U_b , was observed to be approximately $1.3 \times U_{mf}$ of the denser particles. No appreciable solids mixing occurred below the minimum bubbling rate. Bubble induced drift and gross solids circulation appeared to be predominant solids mixing mechanisms in the large particle bed of the present study. The contribution from wake mixing appeared to be negligible.

Apparently segregation resulted from preferentially transporting the lighter particles upwards with rising bubbles and interparticle competition for the voidage left temporarily behind the rising bubbles.

5.4.2 Axial Solids Mixing

Figures 5a through 5d give, respectively, axial concentration distributions of the key particles (lighter particles) respectively at the mixing times of 10 seconds, 25 seconds, 60 seconds, and 120 seconds of mixing at an air rate of 9.77 ft./sec. These figures show the overall progress of the axial mixing process. The data in these figures were

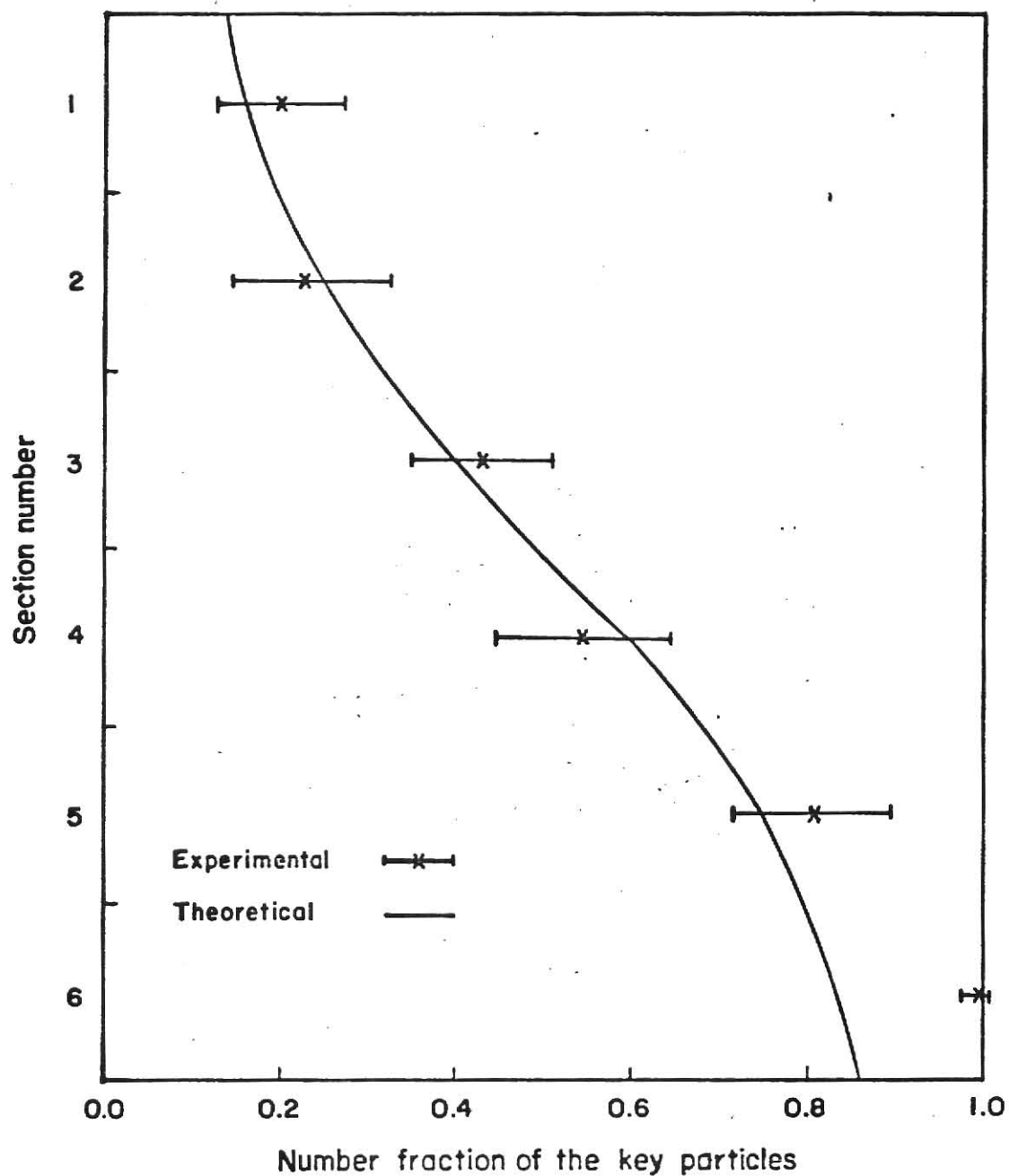


Fig. 5a. Axial cocentration distribution of the key particles after 10 seconds of mixing at an air rate of 9.77 ft./sec..

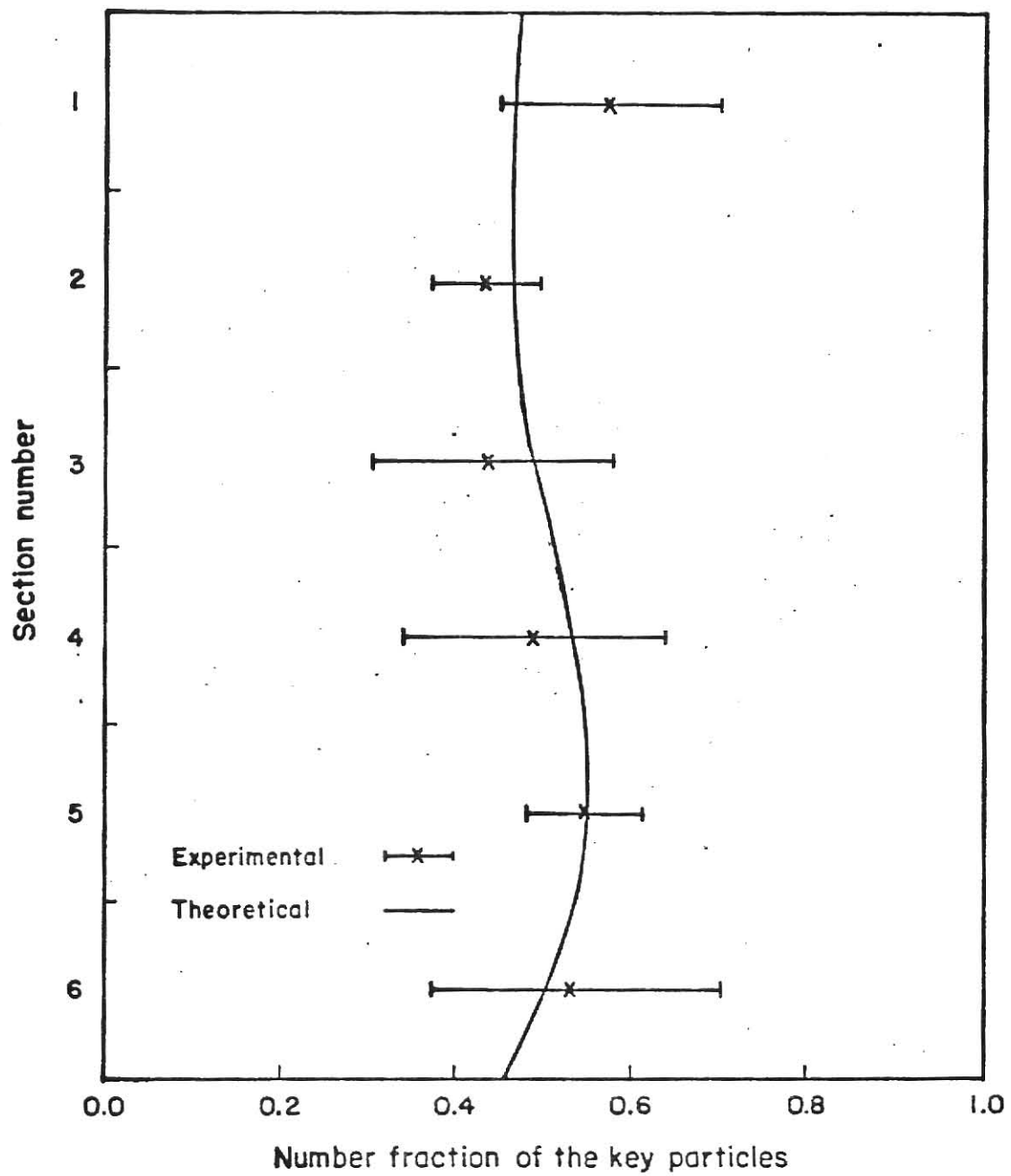


Fig.5b. Axial concentration distribution of the key particles after 25 seconds of mixing at an air rate of 9.77 ft./sec..

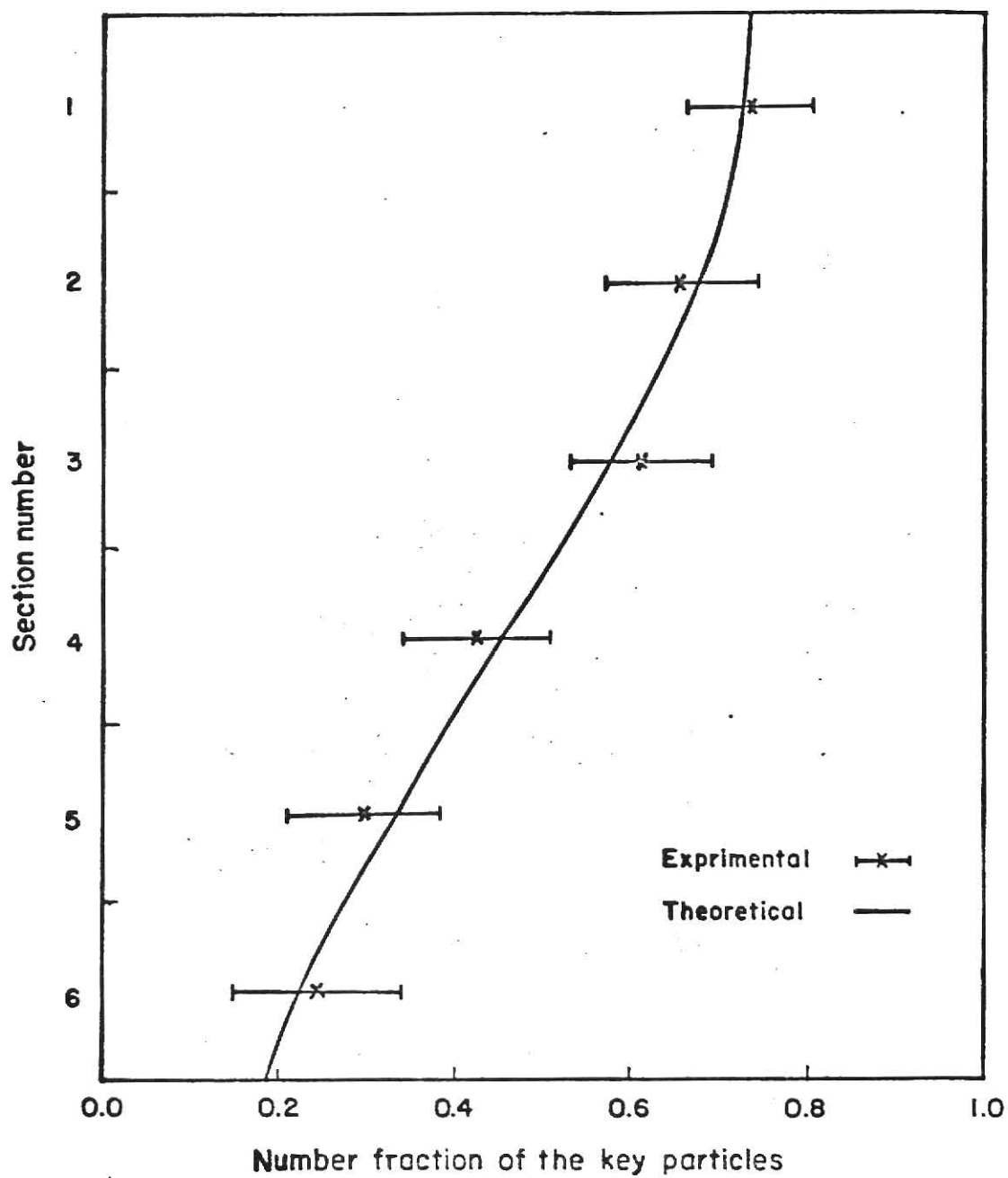


Fig.5c. Axial concentration distribution of the key particles after 60 seconds of mixing at an air rate of 9.77 ft./sec..

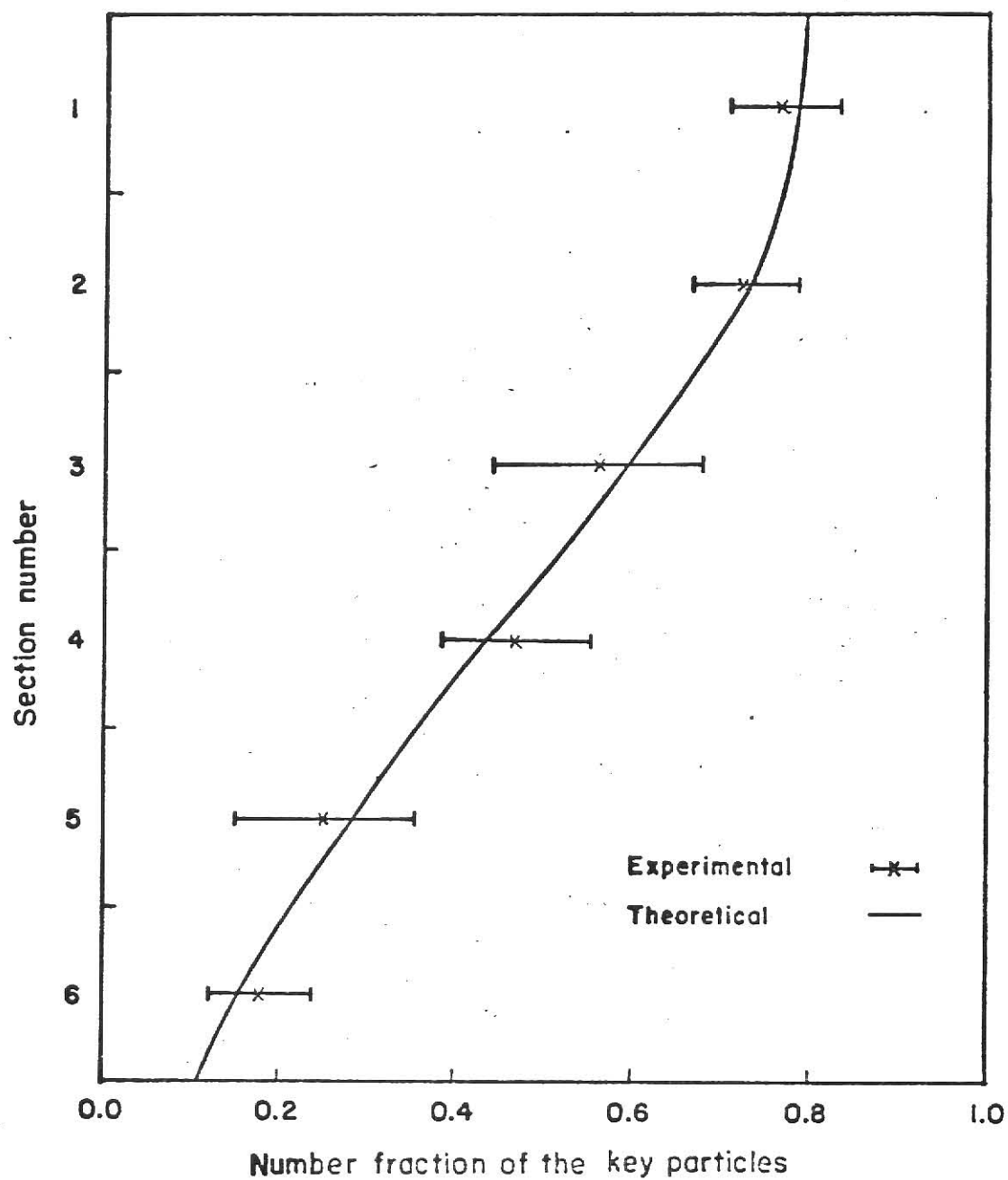


Fig. 5d. Axial cocentration distribution of the key particles after 120 seconds of mixing at an air rate of 9.77 ft./sec..

obtained by initially placing the key particles at the bottom and the nonkey particles at the top. As mentioned previously, appreciable scattering of the experimental data is expected as the system consisted of about 30 balls in each section, i.e., a stochastic system with a rather small population (Asaki and Kondo, 1965; King, 1968).

As can be seen from Figs. 5a through 5d, the overall homogeneity of the system was initially increased with the operating time. When the operating time was prolonged, a continued movement of the key particles to the top of the bed was observed until a dynamic equilibrium was established between the mixing and segregation tendencies. Only slight fluctuation in the concentration distribution could be observed thereafter.

Typical equilibrium concentration distributions of the key particles are shown in Fig. 6. At a moderate air rate (9.77 ft./sec.), the equilibrium concentration distribution was characterized by the fact that the concentration gradient existed only at the bottom of the bed, and the upper part of the bed remained spatially uniform. The concentration of the key particles in the upper layer of uniform concentration was decreased while the proportion of the upper layer was increased with the increase in the air rate. Uniform concentration over the entire bed was not obtained even at an air rate of 11.17 ft./sec., above which the particles elutriated from the bed. At a low air rate (9.48 ft./sec.), the concentration gradient developed throughout the bed. In general, the homogeneity increased rapidly in the low air rate range and slowly in the high air range as illustrated in Fig. 7.

Concentration distributions represented by the proposed model, expressed as Equations (15) through (20), are also presented in Figs. 4a through 4d. Agreement between the model and the experimental data is

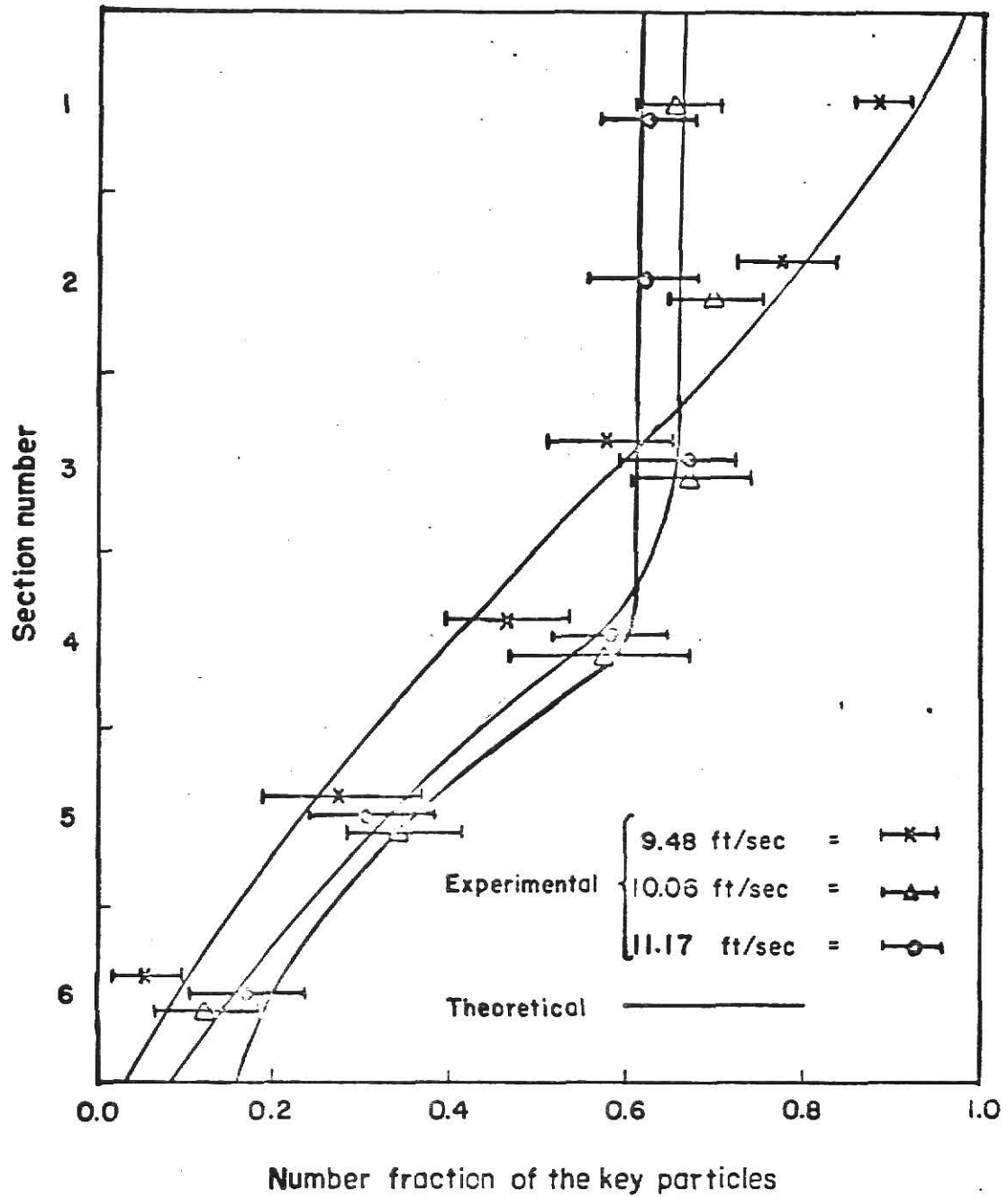


Fig. 6 Equilibrium axial concentration distribution of the key particles.

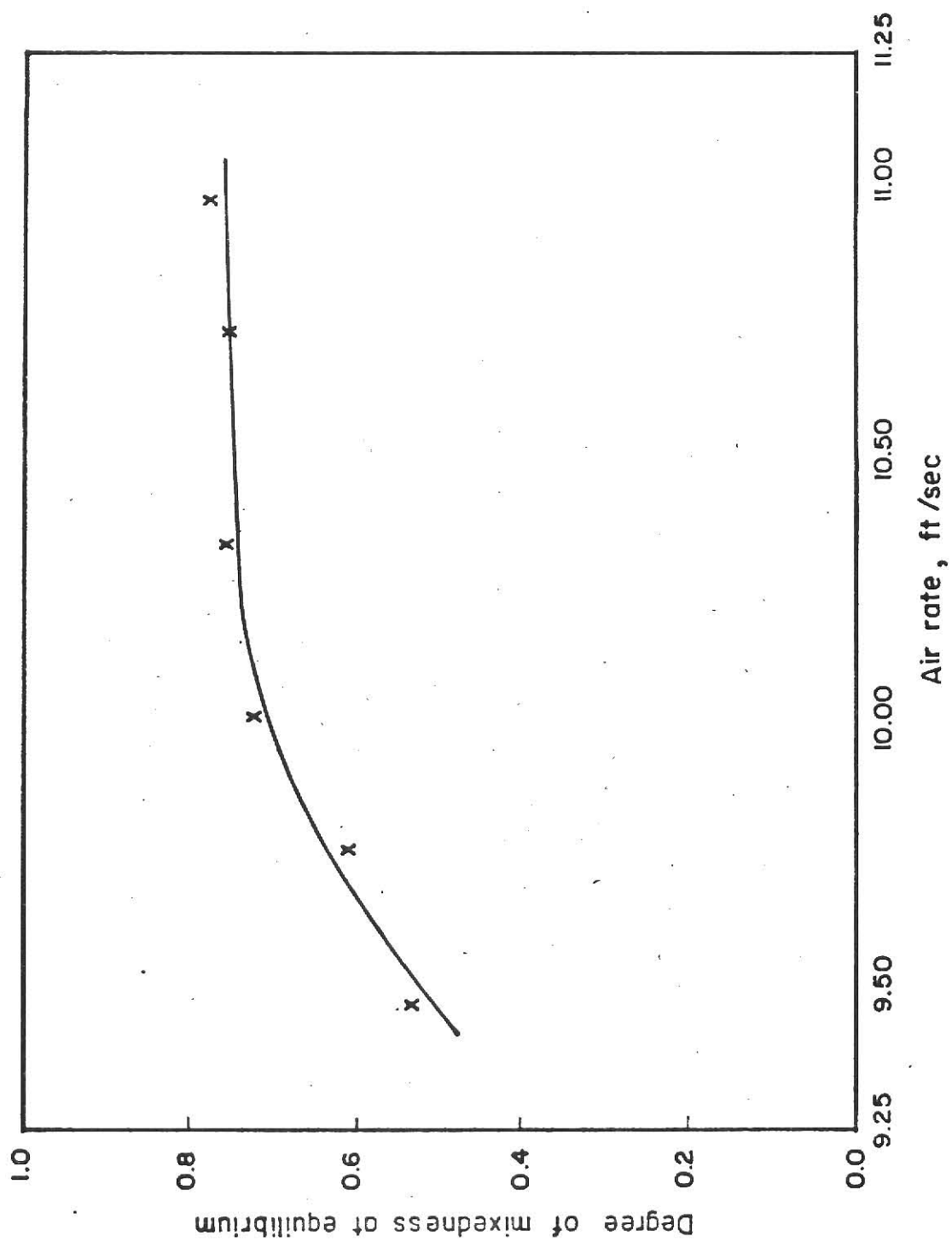


Fig. 7. Degree of mixedness at equilibrium vs. air rate.

sufficiently good. Obviously, the motion of particles directly above the gas distributor was different from that in the main body. The particles may be completely stagnant, semi-stagnant, or fully mobile. Combinations of these various types of behavior were found across the distributor in the present study. Mixing pattern in the top layer of the bed was also different from that of the main body of the bed because the slugs continuously broke up the surface and the particles were scattered over the surface. In order to derive a model which can take into account these various types of complex behavior, additional analysis and experimentation are required.

The degree of mixedness of a mixture can be determined from Equation (25). A plot of the degree of mixedness against the operating time provides an indication of the overall progress of mixing. As shown in Fig. 8, where the data were obtained by initially charging the key particles at the bottom, the degree of mixedness increased rapidly up to a maximum and then declined to a terminal value corresponding to the equilibrium state. Values of the parameter, α , β , and k , of the model estimated from Fig. 8, are presented in Table 2. The parameter, α , which serves as an index for the rate of mixing, can be correlated with air rate as (see Fig. 9)

$$\alpha = 0.74 \times (U - U_b)^{2.94} \quad (26)$$

This result was expected, since the air rate was responsible for the formation of bubbles which induced the mixing. The parameter, β , which indicates the segregation tendency of a particle system (here $\beta = 0$ represents a nonsegregating particle system), varies with the air rate according to the equation (see Fig. 10)

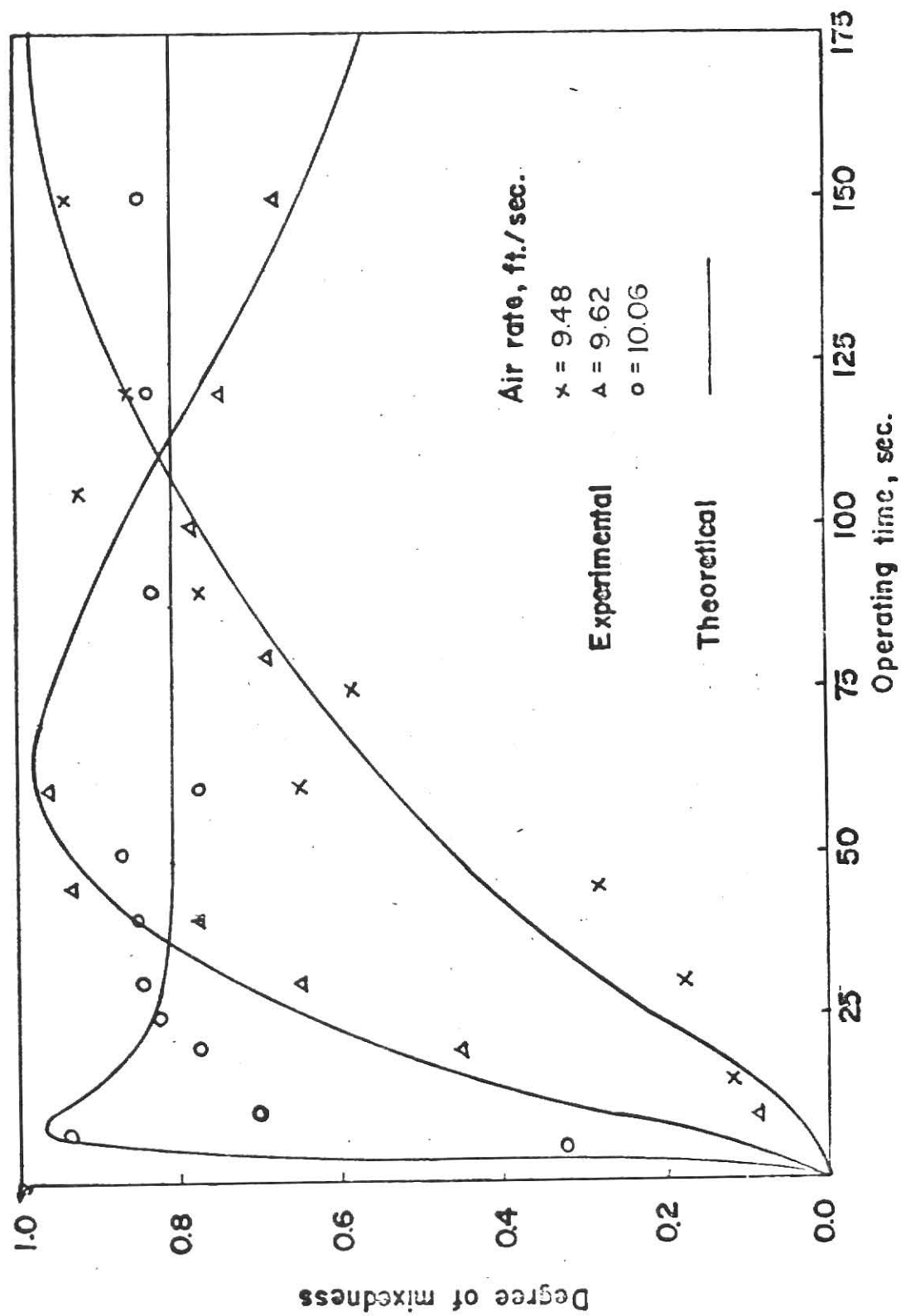


Fig. 8. Degree of mixedness vs. operating time for the axial mixing.

Table 2

Best fit parameters α , β , and k of the nonstationary random walk model
for axial mixing of the particles at 25°C.

<u>Air rate, ft./sec.</u>	<u>α, sec.⁻¹</u>	<u>β, sec.⁻¹</u>	<u>k</u>	<u>Standard deviation of fitting</u>
9.48	1.5×10^{-2}	8.0×10^{-3}	1	0.108
9.62	3.9×10^{-2}	2.3×10^{-2}	1	0.119
9.77	1.4×10^{-1}	4.7×10^{-2}	2	0.123
10.06	4.8×10^{-1}	2.3×10^{-1}	3	0.052
*9.77	2.3×10^{-2}	7.7×10^{-3}	2	0.045

* Lighter particles charged at the top.

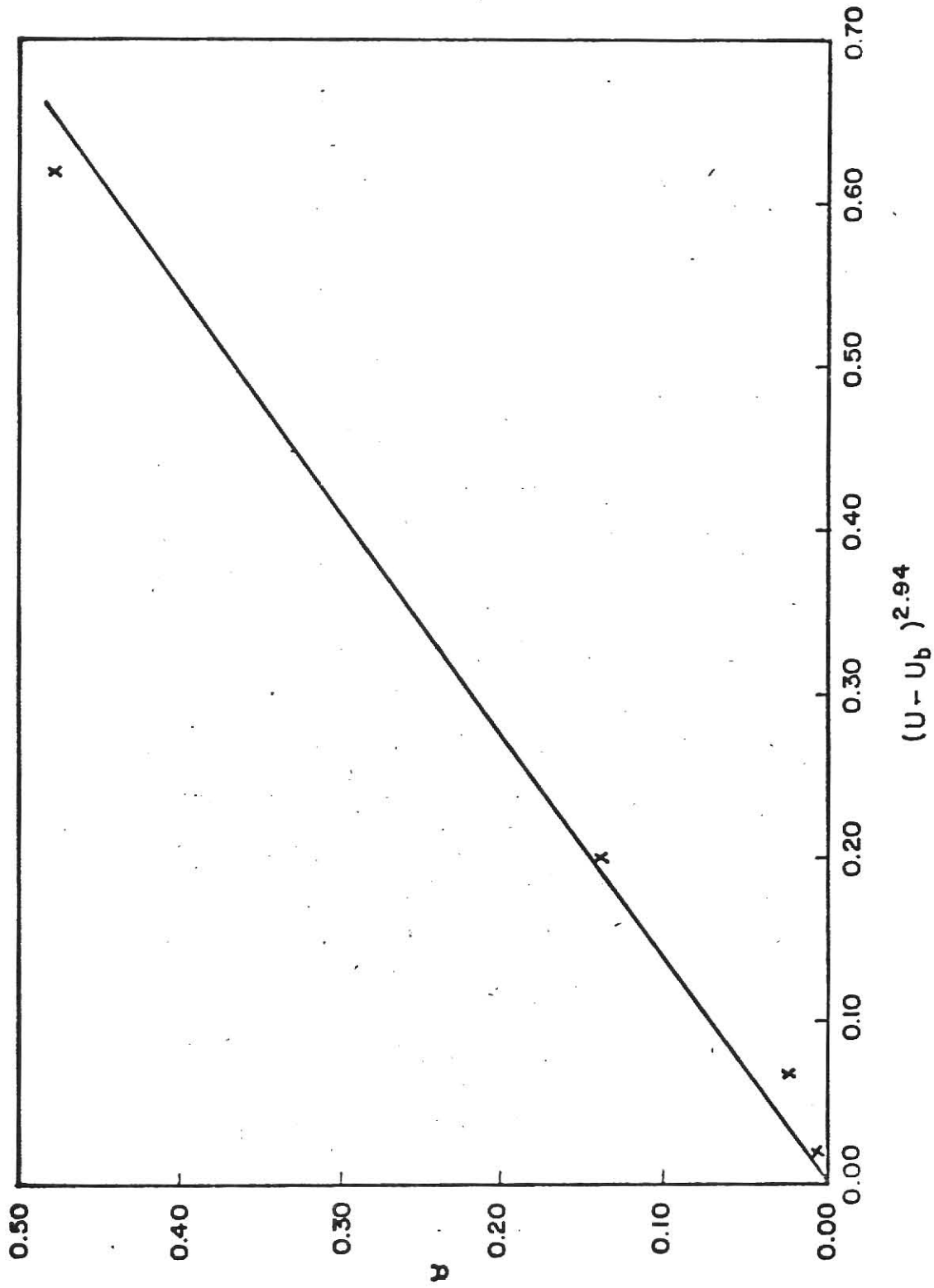


Fig.9 . The effect of air rate on α for the axial mixing .

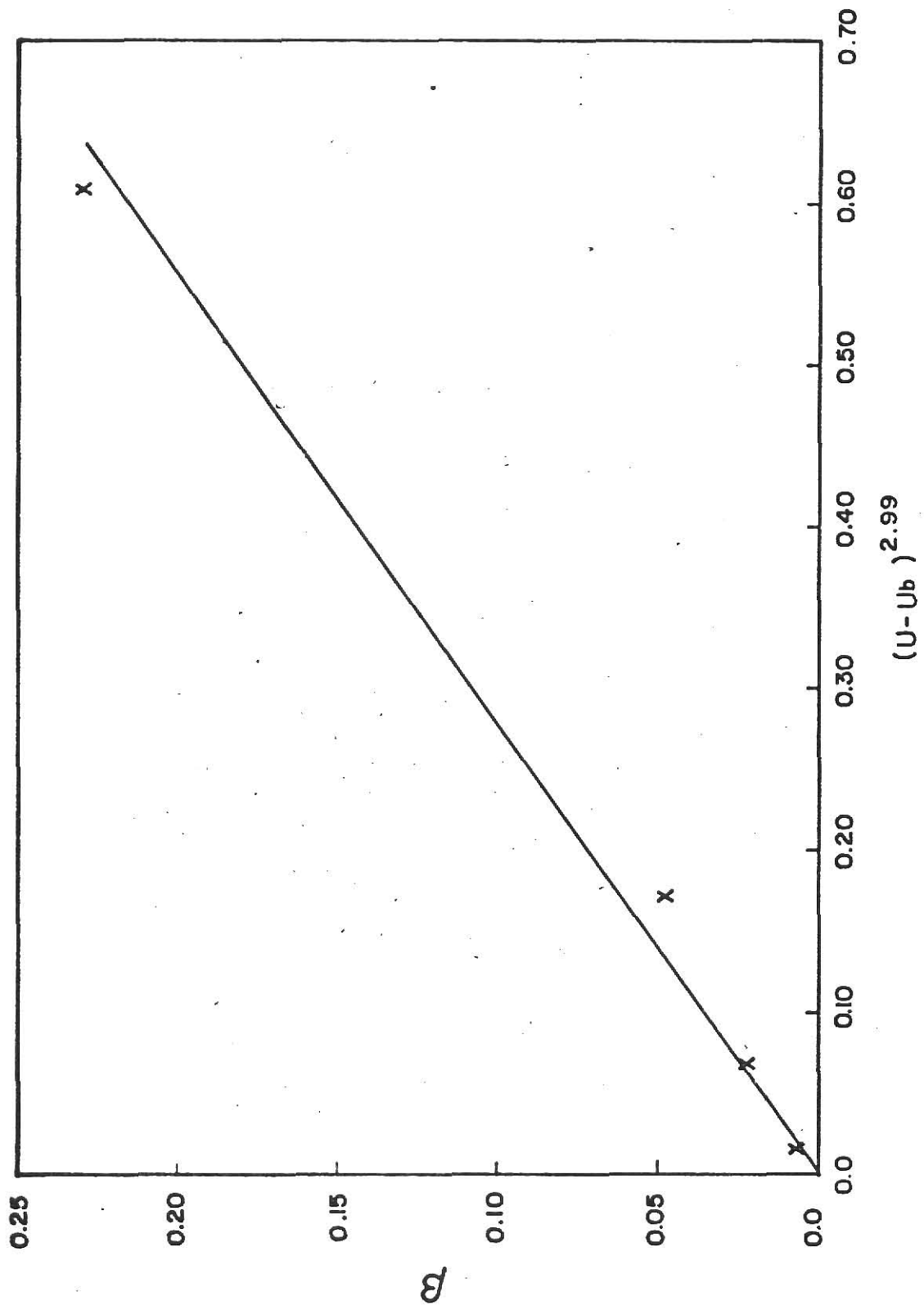


Fig. 10. The effect of air rate on β for the axial mixing.

$$\beta = 0.36 \times (U - U_b)^{2.99} \quad (27)$$

Since additional voidage was created by increase of the air rate, this correlation appears to be consistent with the hypothesis by Pramoda (1969): segregation is the result of an interparticle competition for free void space. The parameter, k , specifies the proportion of the constant concentration layer over the entire bed. The value of k was increased with the increase in the air rate. k may or may not be a time invariant. Further work is required to determine this dependency.

When the process was started in reverse (denser particles at the bottom and lighter particles at the top), only a small extent of mixing was observed. One such experiment was performed at an air rate of 9.77 ft./sec. Apparently the equilibrium state reached in this case was the same as that reached when the process was started in the normal order (denser particles at the bottom, and lighter particles at the top), and mixing was carried out for a long time, as illustrated in Fig. 11.

5.4.3 Radial Solids Mixing

The overall progress of radial mixing in terms of the degree of mixedness is presented in Fig. 12. No appreciable segregation occurred in the radial direction. As postulated, the progress of mixing in this direction can be described by the nonstationary random walk model expressed as Equations (21) through (23). Values of the parameter, α , estimated from the data shown in Fig. 12 are presented in Table 3. The parameter can be correlated with the air rate as (see Fig. 13)

$$\alpha = 0.23 \times (U - U_b)^{1.49} \quad (28)$$

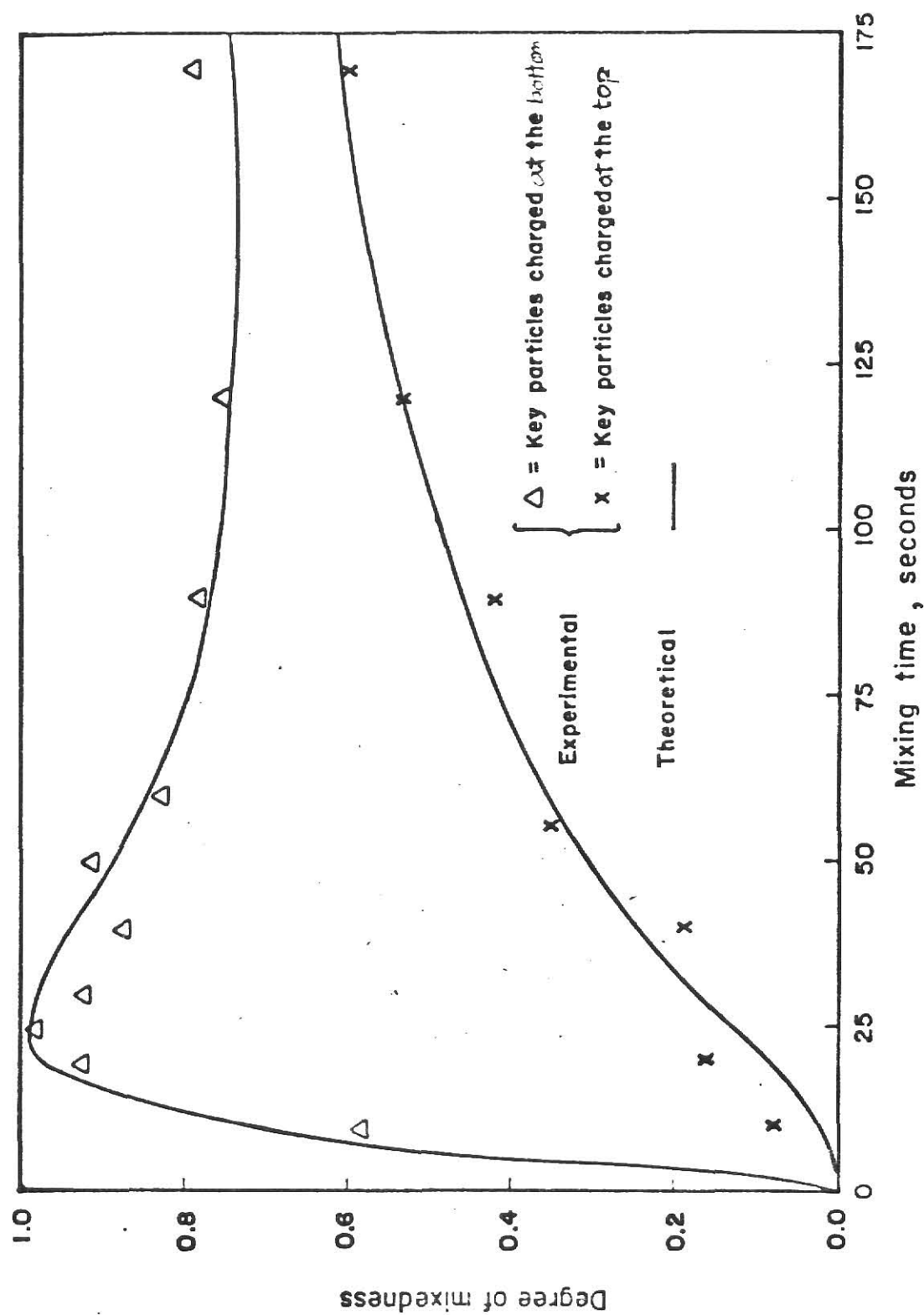


Fig. 11. Degree of mixedness vs. mixing time at air rate 9.77 ft/sec.

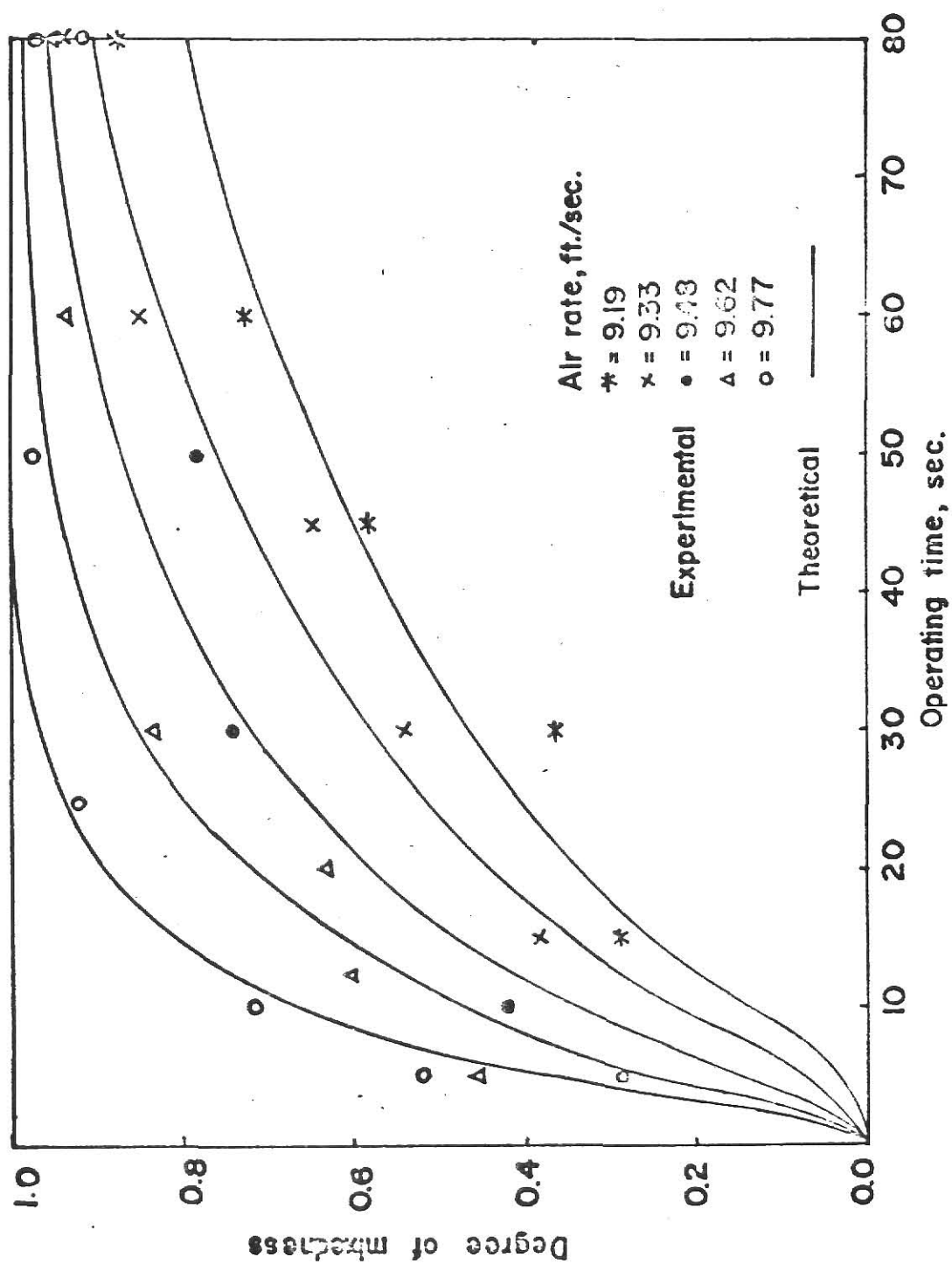


Fig. 12 Degree of mixedness vs. operating time for the radial mixing.

Table 3

Best fit parameter α of the random walk model
for radial mixing of the particles at 25°C.

<u>Air rate, ft./sec.</u>	<u>α, sec.⁻¹</u>	<u>Standard deviation of fitting</u>
9.19	3.8×10^{-2}	0.057
9.33	5.3×10^{-2}	0.041
9.48	7.9×10^{-2}	0.068
9.62	1.2×10^{-1}	0.081
9.77	2.1×10^{-1}	0.062

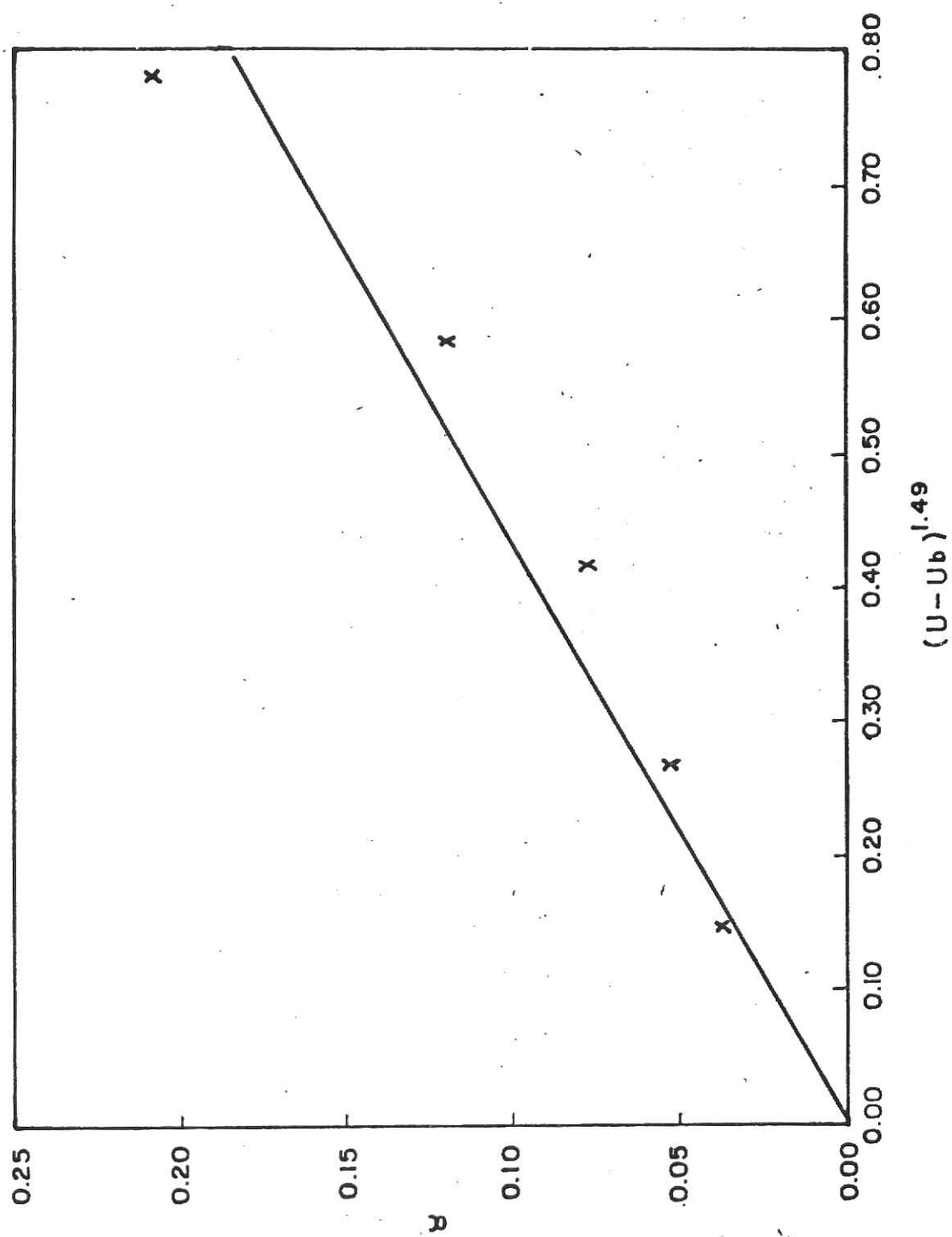


Fig.13 The effect of air rate on α for the radial mixing.

5.5 CONCLUSIONS

Fluidization characteristics were essentially identical to those of nonsegregating system of the previous chapter (Chapter 4). The transition from the bubbling-bed condition to the slugging-bed condition was almost immediate. The minimum bubbling rate, U_b , was observed to be approximately $1.3 \times U_{mf}$ of the denser particles. No appreciable solids mixing occurred below the minimum bubbling rate.

Axial mixing was more rapid for the process where the lighter particles were initially placed at the bottom and the denser particles were placed at the top than for the process which was started in reverse. It appears that a common equilibrium concentration distribution can be obtained for both the processes. At a moderate air rate, the equilibrium concentration distribution was characterized by the fact that the concentration gradient existed only at the bottom of the bed and remained spatially uniform in the upper part of the bed. Radial mixing was more rapid than the axial mixing. No appreciable segregation occurred in the radial direction.

The principal significance of this work is that it provides a nonstationary random walk model which can represent the axial concentration distribution of a segregating fluidized large particle system as a function of the operating time. The model is equally applicable to representation of the radial concentration distribution when it is reduced to a stationary form.

NOTATION

$A_{i,i-1}(t)$ = intensity of transition from state i to state $i-1$ at time t

$A_{i,i+1}(t)$ = intensity of transition from state i to state $i+1$ at time t

$C_i(t)$ = concentration of key particles in section i at time t

$C_j(t+\Delta t)$ = concentration of key particles in section j at time $t+\Delta t$

m = number of sections

$M(t)$ = degree of mixedness at time t

k = a parameter in the model

$\underline{P}(t)$ = transition matrix at time t

U = air rate

U_b = minimum bubbling rate

U_{mf} = minimum fluidization rate

\bar{C} = mean number concentration of key particles

Greek Letters

α, β = parameters in the model

$\sigma^2(t)$ = variance at time t

σ_o^2 = variance of the completely segregated state

Table A1a. Axial concentration distribution of the key particles after 10 second of mixing at an air rate of 9.77 ft./sec.

<u>Run No.</u>	<u>Section 1</u>	<u>Section 2</u>	<u>Section 3</u>	<u>Section 4</u>	<u>Section 5</u>	<u>Section 6</u>
1	0.067	0.129	0.333	0.607	0.963	1.000
2	0.103	0.152	0.444	0.720	0.852	1.000
3	0.143	0.148	0.481	0.692	0.923	0.963
4	0.030	0.069	0.480	0.778	0.815	1.000
5	0.000	0.107	0.308	0.800	1.000	1.000

Table A1b. Axial concentration distribution of the key particles after 25 seconds of mixing at an air rate of 9.77 ft./sec.

<u>Run no.</u>	<u>Section 1</u>	<u>Section 2</u>	<u>Section 3</u>	<u>Section 4</u>	<u>Section 5</u>	<u>Section 6</u>
1	0.750	0.429	0.375	0.357	0.556	0.538
2	0.424	0.519	0.429	0.679	0.556	0.481
3	0.444	0.429	0.667	0.630	0.586	0.308
4	0.778	0.393	0.333	0.321	0.597	0.577
5	0.667	0.393	0.259	0.448	0.435	0.786

Table A1c. Axial concentration distribution of the key particles after 60 seconds of mixing at an air rate of 9.77 ft./sec.

<u>Run no.</u>	<u>Section 1</u>	<u>Section 2</u>	<u>Section 3</u>	<u>Section 4</u>	<u>Section 5</u>	<u>Section 6</u>
1	0.788	0.500	0.546	0.480	0.372	0.259
2	0.706	0.750	0.750	0.500	0.148	0.148
3	0.625	0.690	0.560	0.516	0.444	0.192
4	0.800	0.690	0.643	0.400	0.182	0.179
5	0.758	0.655	0.536	0.321	0.360	0.397

Table A1d. Axial concentration distribution of the key particles after 120 seconds of mixing at an air rate of 9.77 ft./sec.

<u>Run no.</u>	<u>Section 1</u>	<u>Section 2</u>	<u>Section 3</u>	<u>Section 4</u>	<u>Section 5</u>	<u>Section 6</u>
1	0.697	0.760	0.607	0.444	0.385	0.150
2	0.906	0.769	0.690	0.333	0.115	0.192
3	0.703	0.630	0.571	0.577	0.280	0.214
4	0.833	0.769	0.571	0.500	0.182	0.107
5	0.800	0.700	0.360	0.500	0.280	0.259

Table A2a. Equilibrium axial concentration distribution of the key particles at an air rate of 9.48 ft./sec.

<u>Run no.</u>	<u>Section 1</u>	<u>Section 2</u>	<u>Section 3</u>	<u>Section 4</u>	<u>Section 5</u>	<u>Section 6</u>
1	0.844	0.769	0.517	0.600	0.214	0.037
2	0.865	0.769	0.577	0.500	0.219	0.000
3	0.861	0.814	0.464	0.500	0.296	0.000
4	0.861	0.741	0.536	0.539	0.250	0.000
5	0.865	0.750	0.519	0.621	0.120	0.037

Table A2b. Equilibrium axial concentration distribution of the key particles at an air rate of 10.06 ft./sec.

<u>Run no.</u>	<u>Section 1</u>	<u>Section 2</u>	<u>Section 3</u>	<u>Section 4</u>	<u>Section 5</u>	<u>Section 6</u>
1	0.630	0.571	0.630	0.643	0.320	0.111
2	0.784	0.607	0.520	0.571	0.320	0.115
3	0.806	0.607	0.500	0.500	0.400	0.069
4	0.703	0.759	0.577	0.536	0.333	0.084
5	0.630	0.750	0.643	0.542	0.320	0.069

Table A2c. Equilibrium axial concentration distribution of the key particles at an air rate of 11.17 ft./sec.

<u>Run no.</u>	<u>Section 1</u>	<u>Section 2</u>	<u>Section 3</u>	<u>Section 4</u>	<u>Section 5</u>	<u>Section 6</u>
1	0.541	0.679	0.640	0.690	0.308	0.182
2	0.639	0.621	0.600	0.690	0.360	0.111
3	0.632	0.615	0.667	0.536	0.400	0.148
4	0.605	0.579	0.675	0.472	0.375	0.154
5	0.605	0.610	0.756	0.556	0.360	0.179

Table A4. Degree of mixedness for the radial mixing

<u>Air rate</u> <u>ft./sec.</u>	<u>5 sec.</u>	<u>10 sec.</u>	<u>15 sec.</u>	<u>20 sec.</u>	<u>25 sec.</u>	<u>30 sec.</u>	<u>45 sec.</u>	<u>50 sec.</u>	<u>60 sec.</u>	<u>80 sec.</u>
9.19	--	--	0.280	--	--	0.362	0.585	--	0.642	0.864
9.33	--	--	0.385	--	--	0.535	0.623	--	0.836	0.933
9.48	0.280	0.429	--	--	--	0.733	--	0.794	--	0.915
9.62	0.471	0.603	--	0.633	--	0.866	--	--	0.953	0.941
9.77	0.526	0.716	--	--	0.938	--	--	0.970	--	0.963

Table A3. Degree of mixedness for the axial mixing

Air rate ft./sec.	<u>5 sec.</u>	<u>10 sec.</u>	<u>15 sec.</u>	<u>20 sec.</u>	<u>30 sec.</u>	<u>45 sec.</u>	<u>60 sec.</u>	<u>90 sec.</u>	<u>120 sec.</u>	<u>180 sec.</u>
9.48	--	--	0.121	---	0.175	0.238	0.608	0.780	0.863	0.940
9.62	--	0.086	--	0.459	0.647	0.945	0.735	--	0.755	0.685
9.77	--	0.583	--	0.874	0.925	--	0.830	0.775	0.740	--
9.92	0.136	0.433	--	0.947	0.810	--	0.842	0.777	0.816	0.803
10.06	0.316	0.945	0.701	0.771	0.843	--	0.773	0.831	0.833	0.840
*9.77	--	0.077	--	0.168	--	--	0.340	0.422	0.519	--

* key particles charged at the top

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CHAPTER 6

CONCLUSION

This chapter summarizes significant conclusions of this work and several recommendations for further studies.

6.1 CONCLUSIONS

- (1) A modified coalescence-dispersion model has been developed and successfully applied to correlation of the available solids mixing data in a motionless (Kenics) mixer. For the nonsegregating systems and segregating systems with size difference, the coalescence rate is a linear function of the number of helices in the mixer. The distribution ratio appears to be dependent upon the size, size difference, density, and density difference.
- (2) The fluidization of large particle systems in a two-dimensional gas fluidized bed is characterized by the immediate transition from the bubbling-bed condition to the slugging-bed condition. The minimum bubbling rate is approximately $1.3 \times U_{mf}$. No appreciable solids mixing occurs below the minimum bubbling rate.
- (3) At a moderate air rate, the axial equilibrium concentration distribution of the segregating large particle system in the two-dimensional gas fluidized bed is characterized by the fact that the concentration gradient exists only at the bottom of the bed and remains spatially uniform in the upper part of the bed. No concentration gradient exists at equilibrium in the radial direction.
- (4) A nonstationary random walk model has been developed to represent the axial concentration distribution of the segregating fluidized

large particle system as a function of the operating time. The model reduces to a stationary random walk model which is applicable to axial mixing of the nonsegregating particle system. The stationary random walk model is also applicable to the representation of radial mixing data.

6.2 RECOMMENDATIONS

- (1) The modified coalescence-dispersion model can be extended to take into account additional details of mixing mechanisms such as the cell size distribution and the coalescence rate distribution in the mixer. To derive such a model, additional analysis and experimentation are required.
- (2) The slugs which prevail in a two-dimensional large particle bed can be disadvantageous, e.g., they reduce the gas-solid contact time and cause local inhomogeneity in the bed. The possibility of introducing suitable internals into the system to break up the slugs should be considered.
- (3) More realistic mixtures, e.g., powdered mixtures, should be selected for study. Electrostatic charges on the surface of particles constitute perhaps the least understood causes that tend to prevent particles to form a random mixture. In some cases, this may result in the formation of an ordered mixture, and may provide a solution to the segregation.

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MIXING OF SEGREGATION PARTICLES

by

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B.S., National Taiwan University, 1971

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1976

ABSTRACT

A modified coalescence-dispersion model has been developed for the axial mixing of segregating particle systems in a motionless mixer (Kenics mixer). The validity of the present model has been tested against the available experimental data (Gelves-Arocha, 1973). Relationships between parameters of the proposed model and physical properties of the system have been analyzed.

Table tennis or ping-pong balls, $1 \frac{7}{16}$ -in. in diameter, which were identical in every aspect except color, were employed as a nonsegregating particle system, and the same size table tennis balls with density difference were employed as a segregating particle system to study the fluidization and solids mixing characteristics of large particle systems in a two-dimensional gas fluidized bed. The bed employed may be regarded as two-dimensional in character, since the dimension of the bed (130-in. x 15-in. x $1 \frac{3}{4}$ -in.) was such that it gave rise to essentially only two-dimensional motion of the particles.

A nonstationary random walk model has been developed to represent the axial concentration distribution of the segregating fluidized large particle system as a function of the operating time. The model reduces to a stationary random walk model which is applicable to the axial mixing of the nonsegregating particle system. The stationary random walk model is also applicable to the representation of radial mixing data. It was found that the rates of both axial and radial solids mixing can be generally correlated as a function of the excess air rate.