MIXING AND SEGREGATION OF PARTICULATE SOLIDS
IN A MOTIONLESS MIXER

by

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CHAPTER I

INTRODUCTION

The mixing of particulate solids may be broadly defined as any pro-
cess that tends to eliminate existing inhomogeneities, or to reduce existing
gradients (Olecnizack, 1962). By this operation two or more solid materials
are scattered randomly in a mixer among each other by the random movement of
the particles. Although it is one of the oldest industrial operations, it
has remained essentially an art even to this day. Hence, it seems that
there is a clear necessity to clarify the presentation of fundamental con-
cepts such as mixing and degree of mixedness, and their proper application
(Weidenbaum, 1953). A better understanding of these terms will permit bet-
ter correlations between degree of mixedness and rate of mixing to be
obtained, and with these a solution to the most important problem, the
design of mixing equipment.

Solids mixing is referred to as (Fisher, 1963) blending when the par-
ticles being mixed have identical properties. In this situation, mixing
proceeds without separating effects until a random mixture is produced. In
this state, the probability of finding a particle of a given component is
the same at all points in the mixture and is equal to the fraction of that
component in the overall mixture. Generally, in the blending of dry solids,
individual particles are quite mobile. Under these conditions the diffusion
mechanism plays an important role in the process. In liquid and gases, dif-
fusion can be thought of as a "natural impulse," but in the case of solids

mixing, the solid particles will diffuse only as a result of an input of mechanical work. Thus, the mechanisms of mixing must be related to the flow properties of the materials and the method by which mechanical energy is applied (Hogg, 1971).

The second case is one where the particles have different characteristics. For this case, mixing and segregation occur simultaneously. The former predominates during early moments and only after a maximum degree of mixedness has been reached does segregation start to predominate. Eventually a limit is approached. At present this limit cannot be calculated, and must be determined experimentally.

Segregation is defined as that process which produces separation or classification of particles. The particles tend to segregate as a result of their having different characteristics. These differences will produce differential motions of individual particles, which makes them seek preferential regions in the mixer.

The various characteristics causing segregation in order of importance are: difference in size, difference in density, differences in shape, and electrical, magnetic and surface properties. Donald and Roseman (1962), Campbell and Bauer (1966), Williams and Shields (1967) and Williams (1969) have offered several mechanisms in an attempt to explain the causes of segregation and suggest some ways to prevent it.

Segregation is not only important in mixing operations, but it can be related to many problems in handling and storage. Some examples include: product variabilities in the ceramic and powder metallurgy industries, pelletizing and blending of detergent ingredients, blending of raw materials for glass making and preparation of furnace burden for elemental phosphorus

production.

The research described in this thesis has been conducted to investigate axial mixing and segregation of solid particles in a motionless mixer. The existing models of Rose (1959) and Weidanz (1960) were examined. In addition a new Semi-Empirical Kinetic Model has been proposed. The formal mathematical development associated with each of the models is presented. The experimental results are presented in several forms which illustrate the detailed and overall progress of mixing and segregation.

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CHAPTER 2

LITERATURE SURVEY

2.1 INTRODUCTION

The literature of solids mixing has been thoroughly reviewed by Weidenbaum (1953), Klothen (1969), Fan, et al. (1970), and Fan, et al. (1971). This chapter provides a brief discussion of the literature that is most pertinent to this work.

Although a considerable amount of work has been reported, little advance with respect to the design of solids mixing equipment has been achieved. The inter-relations between the mixer geometry, the solids characteristics and the operating conditions and how they affect the process of mixing are just in the process of development. There are three major points that require careful attention before the solution to problems encountered in the mixing of particulate solids can be obtained. These involve 1) a satisfactory definition of degree of mixedness, 2) analysis of the rate of mixing, and 3) the development of the design procedures for mixing equipment.

2.2 DEGREE OF MIXEDNESS

Several investigations in solids mixing have been conducted to find a means by which the process of mixing can be followed. Indices of mixing have been proposed, most of them being statistical in nature. Essentially the indices are different forms of the mean deviation, standard deviation or variance of a number of sample compositions. Kramer (1954) used the

ratio:

$$M = \frac{\sigma_o - \sigma}{\sigma_o - \sigma_r}$$

for a mixture of two materials distinguishable only by color

$$\sigma_o^2 = \bar{X}(1 - \bar{X})$$

$$\sigma_r^2 = \bar{X}(1 - \bar{X})/N$$

where

$\bar{X}$ = overall proportion of one component in the mixture,

N = total number of particles in a spot sample.

This definition involves the random sample standard deviation and the initial mixing standard deviation, and produces a mixing coefficient which varies from zero to unity.

Lacey (1954) has used the ratio

$$M = \frac{\sigma_o^2 - \sigma^2}{\sigma_o^2 - \sigma_r^2}$$

as an index of mixing. It has been shown that when the samples individually cover only a small composition range within the mixture, this definition is approximately independent of sample size.

Fan, et al. (1970), have reviewed over 30 definitions for degree of mixedness. In a recent work, Akao, et al. (1971), proposed a degree of mixedness for binary mixtures of uniform size particles in regular and random arrangements based on the coordination number. An imaginary or hypothetical particle model was proposed by Akao and Noda (1969) in evaluating the distribution of coordination numbers for fine-coarse particle mixtures.

A review of these papers is presented by Fan, et al. (1971).

2.3 MECHANISTIC MODELS

Deterministic approaches have been used to describe the motion of solid particles in solids mixers. A deterministic model is one in which each variable and parameter can be assigned a definite fixed number, or a series of fixed numbers for any given set of conditions.

In a probabilistic or stochastic model the principle of uncertainty is introduced. A stochastic process is a random phenomenon that is controlled by probabilistic laws. This approach to the solids mixing problem seems to be more fundamental and powerful for analyzing and understanding the complex mechanisms of solids mixing processes. With this type of model, mathematical intractability can be avoided.

Fan, et al. (1972), employed a Markov chain process (a stochastic process in which the probability that the state a physical system will be in a given time in the future can be obtained from the knowledge of its state at present) to describe the mixing of particles differing only by color in a motionless mixer of 12 helices. A fairly good agreement with the experimental data was obtained for up to seven steps of the Markov chain, or what is equivalent to seven consecutive passes of the mixture through the mixer.

Makarov and Gorbushin (1970) used the Markov process technique to describe the mechanisms of transition of particles in a circulation cell model. This model is proposed for the preliminary design of batch mixers for free flowing materials with closed loop internal circulation. It is assumed that the termination of convective mixing is the determining factor to obtain the optimum time of mixing because at some time $t_{conv} = t_{opt.}$, and the mixing process achieves an equilibrium with the segregation process. The main idea

in this model is to divide the internal operating volume of the mixer into a number of zones each of which has a characteristic particle flow pattern. Assuming that the laws governing the movement of particles through each zone is known, they determine the average residence time τ_i of particles in each zone and the standard deviation of the residence time distribution in any zone. If the system as a whole is linear, the total average residence time of a particle and the standard deviation for the entire mixer can be calculated. Experimental verification of the method was presented.

Fan, et al. (1973) used the axial dispersed plug flow model (ADPF) and the Stirred Tank-in-Series model to describe longitudinal mixing of solids particles in the mixer. With the ADPF model, mixing was characterized by a simple parameter, the modified Peclet number. Motionless mixers of four different lengths were tested for the axial dispersion of 1/8" and 5/32" lucite particles and wheat. The exit residence time distributions were determined. The cumulative distributions were between the apparent or modified Peclet numbers of 70 to 170 for 1/8" lucite particles and wheat, and between 90 to 220 for 5/32" lucite particles. The apparent or modified axial dispersion coefficients were obtained for each case. Lucite particles 1/8" and wheat exhibited higher apparent or modified axial dispersion coefficients than did 5/32" lucite particles.

Fan, et al. (1973), employed the Stirred Tanks-in-Series-Model and concluded that this model could be utilized to represent other continuous mixers, such as continuous cylinders with or without baffles, continuous ribbon blenders, and continuous screw mixers. Unlike the ADPF model, the Stirred Tanks-in-Series model may be modified to account for the end effects of the mixers by using tanks of different sizes. With this flexibility, the

model may find wide applications in continuous solids mixing.

Shinojara, et al. (1970), theoretically analyzed the mechanism of segregation and blending of a mixture of particles flowing out of the mass flow hopper. The Screen or the "hypothetical hoppers" model was proposed and flow tests were carried out to verify it. Satisfactory agreement between the experimental and calculated results verifies the model and the analysis. Shinojara, et al. (1972), on the basis of the Screening model, studied the pattern or degree of size segregation in filling a hopper. Experiments verified the derived equations and the following conclusions were obtained: 1) a zone where smaller particles are contained as a result of size segregation is observed; 2) the shape of the zone depends on the hopper geometry; and 3) the zone becomes larger with a smaller fraction of larger particles in the feed, longer distance of flow along the heap surface, and larger feed rate of particles.

Tanaka (1971) proposed a model describing the mechanisms of segregation of particles differing in size and density. This model is based on his experience with central filling hoppers. Particles form a conical surface as the bin is filled, and segregation occurs in the thinner surface layer moving on the heap. The heavy particles tend to remain near the central portion, while the lighter particles roll down the slope toward the bin wall. The model is thus called "Push-away" model.

Nienow, et al. (1972) examined the role of particle density and size, the minimum fluidization velocity and described the mechanisms of particle movement in two component fluidized beds. It was found that generally a fairly wide particle size difference can be tolerated without appreciable segregation because such beds exhibit a composite minimum fluidization velo-

city, very dependent on finer particles. However, even small density differences lead to ready settling of the denser particle, even when it has a lower minimum velocity of fluidization. Mixing profiles show that the bulk of the bed is of a constant composition which approaches the overall component proportion with increasing velocity.

2.4 SEMI-EMPIRICAL MODELS

In a Semi-Empirical model, a measure of the degree of mixedness is defined and generally a first order equation is proposed using this measure. The equation is integrated and the constants are estimated using the experimental data, by the least square method. In Table 1 a summary of rate equations presented by Fan, et al. (1970), is shown. Among these proposed models only two have considered the mixing and segregation effects operating together; they are Rose's model (1959) and Weidanz's model (1960).

Rose's model includes both mixing and demixing effects. The parameters in the mixing equation can be related to physical properties of the elements of the system; for example, the size, density and shape of the particles in a two component system and the dimensions of the mixer.

Weidanz considered a model in which two mixing operations are working simultaneously. The case of a rotating drum where the vertical mixing action is counteracted by a demixing due to gravity is cited. A more detailed review of these two models is considered in later sections.

Yano, et al. (1957, 1959, 1960), presented results of a rather detailed semi-empirical study including extensive experimental results. In a series of papers they covered the mixing of powders in several types of mixers such as V type mixers, double cone mixers and cubic type mixers. Experiments were conducted with two kinds of powders having the same particle size dis-

tribution, and with particles differing in size. They reported that all mixing curves, even for different shapes of mixers under different conditions, had similar features. Mixing curves were analyzed and discussed. They observed that when the particle size of the powders was comparatively large the values of the standard deviation became larger, and that as the particle size-ratio of the two components was increased it became more and more difficult to obtain an intimate mixture.

TABLE 1

SUMMARY OF RATE EQUATIONS

Investigators	Proposed model	Rate equation	Final equation given	Notations
Brothman, Wollan, and Feldman [B12]	The surface of separation of two constituents increases as mixing proceeds, following the first order mechanism.	$y_{t+1} = y_t + \phi(1-y_t)$	$P_t = 1 - e^{-k\,S_p(1-e^{-t})}$	y_t = proportion of the maximum theoretically possible surface of separation, S, which has been developed in t t = time k, c = constants S = interfacial area of surface per unit volume S_o = maximum surface per unit volume σ_r = standard deviation of complete random mixture σ = observed standard deviation z = size of agglomerate z_o = initial value of z z_f = equilibrium value of z
Coulson and Maitra [C17]	Same as above.	$\dfrac{dS}{dt} = k(S_o - S)$	$\ln\dfrac{100}{X} = kt$	
Weidenbaum and Bonilla [W2]	The ratio $\dfrac{\sigma_r}{\sigma}$ can be considered as the fractional distance toward equilibrium, which follows first order mechanism.	$\dfrac{d(\sigma_r/\sigma)}{dt}$ $= k[1 - (\sigma_r/\sigma)]$	$\ln\left[\dfrac{1}{1-(\sigma_r/\sigma)}\right]$ $= \ln\left[\dfrac{1}{1-(\sigma_r/\sigma)}\right]+k$	
Oyama and Ayaki [O10]	The observed variance follows first order mechanism.	$\dfrac{\partial \sigma^2}{\partial t} = -k(\sigma^2 - \sigma_r^2)$	$\ln\dfrac{\sigma^2-\sigma_r^2}{\sigma_o^2-\sigma_r^2} = -kt + c$	
Sakaio [S13]	The size of agglomerate diminishes as mixing proceeds, following first order mechanism.	$-\dfrac{dz}{dt} = K(z - z_f)$	$\ln\left(\dfrac{z - z_f}{z_o - z_f}\right) = -kt$	

TABLE 1--Continued

Investigators	Proposed model	Rate equation	Final equation given	Notations
Lacey [L2]	Axial diffusive mixing is assumed to occur in a horizontal cylinder.	$\dfrac{\partial c}{\partial t} = D_a \dfrac{\partial^2 c}{\partial x^2}$ I.C. and B.C. $c(x,0) = 1,\ 0 \le x < \bar{C}$ $= 0,\ \bar{C} < x < L$	$\sigma^2 = \dfrac{2}{\pi^2} \sum_{k=1}^{\infty} \left(\dfrac{\sin k\bar{C}\pi}{k}\right)^2 \cdot e^{-2k^2\pi^2 T}$	C=concentration of constituent $\bar{C}$=average concentration of key constituent k=integer $T = \dfrac{D_a t}{L^2}$ L=length of mixer D_a=axial diffusion coefficient N=number of revolutions of the mixer $m = \dfrac{D_\pi^2}{L^2}$ d=diameter of particles
Hogg, Cahn, Healy, and Fuerstenau [H14]	Same as above.	$\dfrac{\partial c}{\partial t} = D_a \dfrac{\partial^2 c}{\partial x^2}$ I.C. and B.C. i) $c(x,0)=0,\ \dfrac{-L}{2} \le x<0$ $=1,\ 0\le x<\dfrac{L}{2}$ ii) $\left.\dfrac{\partial c}{\partial x}\right|_{x=-\frac{L}{2}} = 0$ iii) $\left.\dfrac{\partial c}{\partial x}\right|_{x=\frac{L}{2}} = 0$	$\sigma^2 = \dfrac{2}{\pi^2} \sum_{k=1}^{\infty} \dfrac{1}{(2k-1)^2} \cdot \exp\left(-\dfrac{2(2k-1)^2\pi^2 D_a N}{L^2}\right)$	
Fisher [F2]	Same as above.	$\dfrac{\partial c}{\partial t} = D_a \dfrac{\partial^2 c}{\partial x^2}$ I.C. and B.C. i) $c(x,0)= 1,\ x=\dfrac{L}{2}$ $= 0,\ \text{otherwise}$ ii) $\left.\dfrac{\partial c}{\partial x}\right|_{x=0} = 0$ iii) $\left.\dfrac{\partial c}{\partial x}\right|_{x=L} = 0$	$\sigma^2 = 0.08333 + \dfrac{8}{\pi^3 d} \sum_{k=1}^{\infty} \dfrac{1}{k^3} e^{-mk^2 t} \cos k\pi s \sin\dfrac{k\pi d}{2L}$	

TABLE 1--Continued

Investigators	Proposed model	Rate equation	Final equation given	Notations
Otake, Kitaoka, and Tone [O3]	Radial diffusive mixing takes place in a cylindrical tank equipped with a paddle-type impeller.	$\frac{\partial C}{\partial t} = D_r \left(\frac{\partial^2 C}{\partial r^2} + \frac{1}{r}\frac{\partial C}{\partial r} \right)$ I.C. and B.C. i) $C(x,0)=1$, $0<r<R_i$ $=0$, $R_i<r<R_o$ ii) $\left.\frac{\partial C}{\partial r}\right|_{r=R_o} = 0$ iii) $\lim_{t\to\infty} C = \bar{C}$	$\sigma^2 = \frac{4R_i^2}{R_o^4} \sum_{k=1}^{\infty} \frac{J_1^2(m_k R_i)}{m_k^2 J_0^2(m_k R_o)} \cdot e^{-2D_r m_k^2 t}$	C=concentration of key constituent $\bar{C}$=average concentration of key constituent D_r=radial diffusion coefficient r=radial distance R_i=radius of key constituent charged in the tank R_o=radius of tank J=Bessel functions m_k=roots of J_1 $(m_k R_o) = 0$ L=length of mixer M=degree of mixedness N=number of revolutions of the mixer k=constant d=inside diameter of mixer Me=equilibrium value for the degree of mixedness A=coefficient defining rate of mixing B=coefficient
Gayle and Gary [G2]	Mixing proceeds by an initial second order mechanism and is followed by a slower first order mechanism.	$-\frac{dM}{dt} = k\frac{d^{1.45}M^2}{L^2}$	$\frac{1}{M} = 1 + kd^{1.45}\left(\frac{N}{L}\right)^2$	
Rose [R4]	A mixing and demixing potential are operative simultaneously.	$\frac{dM}{dt} = A(1-M) - B\psi$	$Me = \eta\left[1 - \left(\frac{B}{A}\right)^2\right]$	

TABLE 1--Continued

Investigators	Proposed model	Rate equation	Final equation given	Notations
Weidanz	In the vertical direction mixing and segregation occur, but in the horizontal direction only mixing is acting.	$$V_h \frac{dy_h}{dt} = [(Y_l - Y_h)b-c]$$ $$V_l \frac{dY_l}{dt} = [Y_r - Y_l]a$$	$$\frac{\sigma}{\sigma_o} \approx [e^{-2Qt} + \sigma_\infty^2(1-e^{-EQt})^2]^{1/2}$$	defining rate of demixing η=intrinsic efficiency of mixer t = time ψ=demixing potential Y = volumetric proportion of one of the constituents σ = standard deviation a=volume exchange per unit of time between left side and right side (horizontal) b=volume exchange per unit of time in vertical direction c=rate of demixing, a constant E = constant Q = constant

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CHAPTER 3

MODELS OF RATE OF MIXING

3.1 INTRODUCTION

The flow pattern of particles in a solids mixer is usually too complex to predict theoretically. It is, therefore, desirable to construct a model which simulates the actual situation to represent the mixing behavior of the particles. Several mixing mechanisms operate in a motionless mixer. They are (Fan, et al., 1972, 1973):

1) Multiple divisions and combinations of the flow of particles.

2) Mutual interactions of the particles and interaction of the particles with the helices and with the wall of the mixer.

3) Change of the direction of the flow of particles.

4) Difference in velocities of particles.

In general, mixing has been considered to depend on three mechanisms (Lacey, 1954). They are: (a) convective mixing--transfer of groups of adjacent particles from one location to another in the mixture, (b) diffusive mixing--distribution of particles over a freshly developed surface, and (c) shear mixing--setting up of slipping planes within the mixture. All three mechanisms function to some extent as mixing proceeds. For example, in the motionless mixer convection is produced by mechanisms 2) and 4); diffusion by mechanisms 1), 2), 3); and shear by mechanisms 3) and 4). The application of Newton's law of motion to description of particle motion in a mixer is extremely difficult, if not impossible.

The purpose of this chapter is to describe existing models accounting for mixing and segregation in binary systems. These include Rose's (1959), Weidanz's (1960), and the Semi-Empirical Kinetic Model developed in this work.

3.2 PREVIOUS WORK

3.2.1 Rose's Model

In this model the degree of mixedness is denoted by M (M = 0 corresponds to fully segregated material and M = 1 to a perfectly mixed batch). The degree of mixedness is defined as:

$$M = 1 - \frac{\sigma}{\sigma_o}$$

where

σ = standard deviation of samples drawn from the mixture,

σ_o = standard deviation corresponding to the unmixed material.

The model then postulates that the rate of the change of M is related to a mixing potential, 1-M (that gives rise to an interchange in particles) and a demixing potential, ψ (operative when a difference between the components exist) through the following equation:

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

where A and B are constants. The constant, A, depends on the geometrical and physical parameters of the mixer and particles. The constant, B, depends upon the geometry of the mixer, density differences, size differences and shape of the particles in a two component system.

According to Rose's analysis, the demixing potential, ψ, can be either negative or positive depending upon the initial state of the components in

the system. Rose cited a case where ψ is positive because the denser material is in the lower layer and the less dense material is in the upper layer, and thus segregation is maintained. However, if the denser material is above the less dense, it will sink through the less dense and produce mixing. If the operation is continued, the material rises to the top and therefore the demixing potential changes sign. This situation can be represented in terms of a simple equation as:

$$\psi^2 = 1 - M \qquad \text{or} \qquad \psi = \pm\sqrt{1 - M} \tag{2}$$

Differentiation of equation (2) with respect to t, followed by combination with equation (1) and integration subject to the initial condition, $M = 0$ at $t = 0$, gives the following results:

$$M = \eta\{1 - [(1 - B/A)e^{-At/2} + B/A]^2\} \tag{3}$$

$$M = \eta\{1 - [B/A - (1 + B/A)e^{-At/2}]^2\} \tag{4}$$

Here the intrinsic efficiency η has been introduced to account for imperfections in individual mixers. Equation (3) corresponds to the situation where the process is started with the denser component below, and equation (4) to the case where the denser component is above.

3.2.2 Weidanz's Model

Weidanz's (1960) derivations are based on the concept that there are two mixing operations at work simultaneously. For example, in a rotating drum the vertical mixing is counteracted by a demixing due to gravity, while axial mixing is undisturbed by this action. A schematic representation of this model is shown in Fig. 1. Here the components are initially separated by the vertical line. The volumetric proportion of the constituent on the
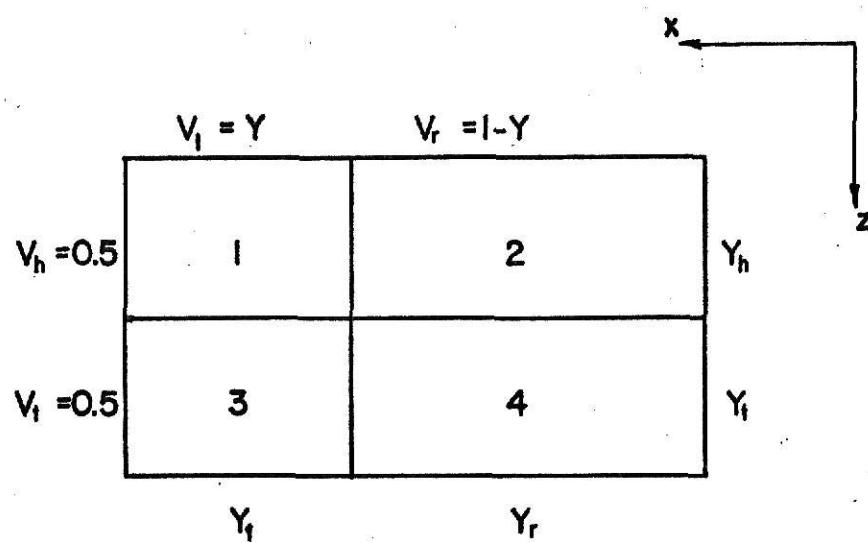
Fig. I. Schematical representation of Wedianz's
Model.

left hand side is equal to Y, and that of the constituent on the right hand side to 1-Y. The horizontal line divides the system in two sections of equal volume; the upper fraction of the total volume is denoted by V_h (=0.5) with a concentration Y_h, and the lower by V_t (=0.5) with concentration Y_t.

Weidanz considered that both mixing and segregation occur in the Z direction, but in X direction only mixing acts. In the analysis of the system he assumes that the exchange of small volumes between a region of high concentration and a region of low concentration produces mixing. For mixing in Z direction, it is assumed that the volume exchange per unit of time is constant (i.e., $\frac{dv}{dt} = b$). At the same time, demixing occurs in the vertical direction. The rate of segregation is assumed also to be a constant and it is noted as C. A net balance of the movement of volumes in Z direction gives:

$$V_h dY_h = [(Y_t - Y_h)b - c] \, dt \tag{5}$$

$$Y_h = Y_t = Y \quad \text{at} \quad t = 0$$

Solving for Y_h and performing the same analysis for Y_t, Weidanz obtained:

$$Y_h = Y - 0.5 \frac{c}{b} (1 - e^{-4bt}) \tag{6}$$

$$Y_t = Y + 0.5 \frac{c}{b} (1 - e^{-4bt}) \tag{7}$$

According to the definition of the standard deviation, and assuming that the volume of samples with concentration Y_h is proportional to V_h, and the volume of samples with concentration Y_t is proportional to V_t, Weidanz obtained:

$$\sigma^2 = \sum_{i=1}^{n} (Y_i - Y)^2 / n \tag{8}$$

$$\sigma_z{}^2 = V_h \, (Y_h - Y)^2 + V_t \, (Y_t - Y)^2 \tag{9}$$

$$\sigma_z = 0.5 \, \frac{c}{b} \, (1 - e^{-4bt}) \tag{10}$$

For the analysis of the X direction, it is considered that segregation is absent (c = 0) and therefore only mixing takes place. Following a derivation analogous to that given above:

$$V_1 dY_1 = a(Y_r - Y_1)^{dt} \tag{11}$$

Here a represents the net exchange of volume between the left and the right hand sides in a unit of time. Solving this equation subject to the initial condiction that $Y_1 = 1$ at t = 0 gives:

$$Y_1 = Y + (1 - Y) \, e^{-at/Y(1 - Y)} \tag{12}$$

By the same considerations employed in the analysis of the Z direction and with $Y_r = 0$ at t = 0, Weidanz obtained

$$\sigma_x{}^2 = V_1 (Y_1 - Y)^2 + V_r (Y_r - Y)^2 \tag{13}$$

$$\sigma_x = \sqrt{Y(1 - Y)} \, e^{-at/Y(1 - Y)} \tag{14}$$

The total effect can be obtained by taking advantage of the additive properties of the variance. Addition of equations (10) and (14) gives

$$\sigma^2 = \sigma_x{}^2 + \sigma_z{}^2 \tag{15}$$

or

$$\frac{\sigma}{\sigma_o} = [e^{-2Qt} + \sigma_\infty{}^2 (1 - e^{-EQt})^2]^{1/2} \tag{16}$$

where

$$a = Q\sigma_o^2 \qquad\qquad E = \text{constant}$$

$$C = 0.5\ \sigma_o\ EQ\ \sigma_\infty \qquad\qquad \sigma_\infty = \text{standard deviation at } t \to \infty$$

$$b = 0.25\ EQ \qquad\qquad Q = \text{constant}$$

In terms of Rose's definition for degree of mixedness, Weidanz's result takes the form

$$M = 1 - \frac{\sigma}{\sigma_o} = 1 - \{e^{-2Qt} + \sigma_\infty^2(1 - e^{-EQt})^2\}^{1/2} \tag{17}$$

Rose and Weidanz both applied their equations to Tanaka's data (presented by Rose) in which unmixed material was repeatedly passed through hoppers with different slopes. Both models showed a good agreement with the experimental data.

3.3 SEMI-EMPIRICAL KINETIC MODEL

The situation to be modeled is a binary system in which light or small particles are placed in the bottom of a mixing device and big or heavy particles are layered on top, as it is shown in Fig. 2. The situation where the denser or larger component is on the top at $t = 0$ (segregated state of type I) can be viewed as a metastable equilibrium state because, at least theoretically, the system can reach a more stable segregated state (the lighter component on the top, i.e., segregated state of type II). This observation is based on Tanaka's (1971) recent work on the stability of systems of solid particles which led to the development of the "Push-away" model. This model is based on the fact that heavy particles can push away both heavy and light particles underneath it, and light particles can push away only light particles located underneath it. This implies that a system in which light particles are on the top of heavy particles (segregated state of type II) is more stable than a system in a segregated state of type I, because light par-
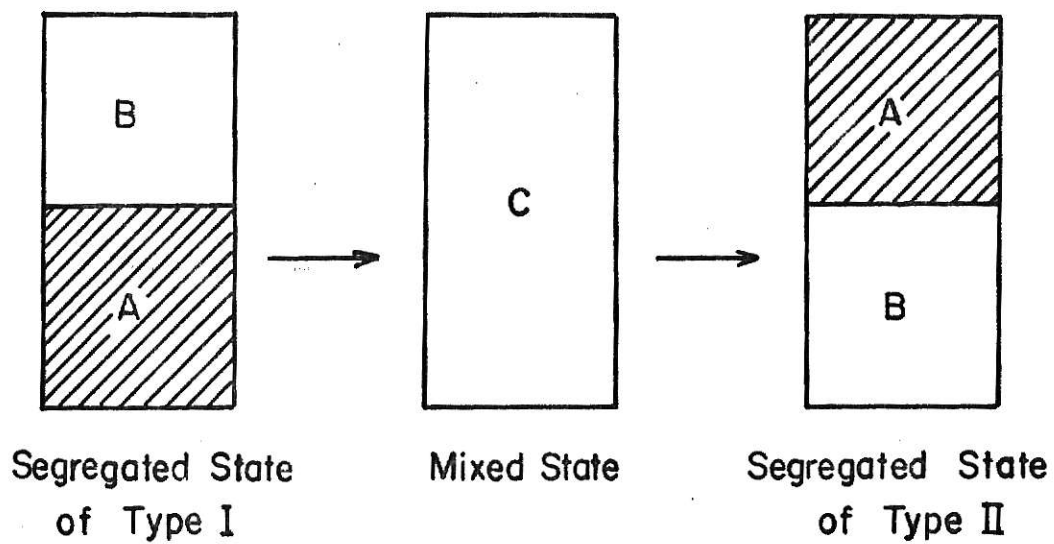
Fig. 2. Representation of the Semi-Empirical Kinetic Model.
A = Small or light particles
B = Big or heavy particles
C = Mixture of A and B

ticles cannot push away heavy particles. Various combinations of heavy-light particles and big-small particles are presented in Fig. 3.

When a system in the segregated state of type I is acted on by a device such as a motionless mixer, axial mixing is produced as a primary effect. This results principally from the difference in velocities of heavy and light particles (or big and small) streaming along the cross section of the mixer. This type of mixing is primarily of a convective nature. It is characterized by a rapid increase in the degree of mixing. The behavior of particles in this type of mixing has been observed to be only slightly dependent on the physical characteristics of the components (Makarov and Gorbushin, 1971).

At the same time that mixing is produced, various physical factors and properties of the particles (differences in size, density, shape, etc.) create a tendency to impede the formation of a stable random mixture. Initially these effects are not appreciable and it appears that only mixing is produced up to a point where a near random mixture is obtained. The mixed state is relatively unstable and thus further agitating action on the system results in a rapid decline in the degree of mixing and formation of a stable segregated state (i.e., the segregated state of type II).

The key assumption in the development of the model is that mixing and segregation can be described independently. This concept is very similar to that employed by Weidanz (1960). A kinetic approach, analogous to that employed in classical chemical kinetic analysis is used to model the mechanisms of mixing and segregation. The analogy is based on the fact that in classical chemical kinetic, under appropriate conditions materials may be transformed into new and different materials which constitute different
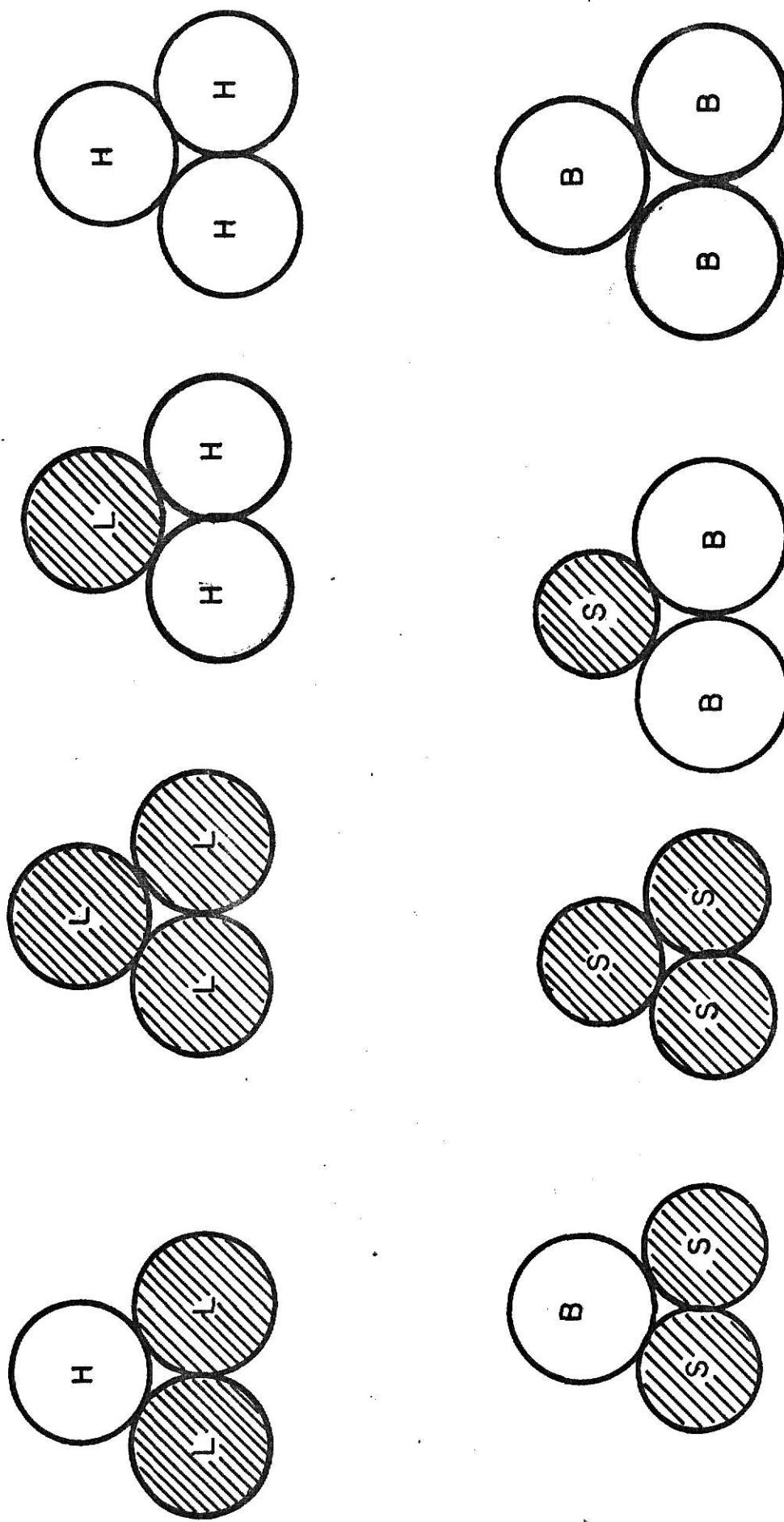
Fig. 3. Various combinations of heavy——light particles and big——small particles. (Tanaka, 1971).

chemical species (driving forces are provided by differences in concentration); in solids mixing, no new materials are created, but rather a new state in the system (driving forces are provided by differences in degrees of mixedness or segregation.

3.3.1 Formulation of the Model Equations

The following notations are employed:

1) Big or heavy particles will be referred to as species B, and small or light as species A.

2) [A+B] represent particles in the segregated state of type I (big or heavy particles on the top and small or light particles in the bottom).

3) C represents a mixed state of A and B with any degree of mixedness.

4) [B+A] represent particles in the segregated state of type II (light or small particles on the top and big or heavy particles in the bottom).

5) Mixing will be referred to as a decrease of particles in the segregated state of type I, and segregation as an increase of particles in the segregated state of type II.

The following concepts are employed in developing the present model by analogy with chemical kinetics:

1) In chemical kinetics the rate at which a material appears or disappears is studied in terms of concentrations or conversions as a function of time. By analogy, in solids mixing the rate can be studied in terms of a degree of mixing or a degree of segregation. The degree of mixedness can be defined as:

$$M = 1 - f_{total} \qquad (18)$$

where

$$f_{total} = \frac{\sigma^2}{\sigma_o^2} \qquad (19)$$

Here, f_{total} is the ratio of the variance among samples (of key particles) at time t, to the variance at t = 0. This ratio indicates the overall degree of segregation existing in the system.

The degree of segregation contributed by the particles in the segregated state of type I, f_I, is defined by:

$$f_I = \frac{\sigma_I^2}{\sigma_o^2} \qquad (20)$$

where σ_I^2 is the variance among samples at time t (assuming that only mixing takes place, that is only a decrease in the degree of segregation of type I takes place), and σ_o^2 is the variance, i.e., at t = 0.

The degree of segregation contributed by the particles in the segregated state of type II, f_{II}, is defined as:

$$f_{II} = \frac{\sigma_{II}^2}{\sigma_o^2} \qquad (21)$$

where σ_{II}^2 is the variance among samples at any time t (assuming that only segregation takes place, that is, only an increase in the degree of segregation of type II occurs), and σ_o^2 is the initial variance. The linear combination of f_I and f_{II} gives the total degree of segregation in the system, f_{total}.

2) In chemical kinetics, concentration gradients provide driving forces for reaction. By analogy, the driving forces for the decrease of particles in a segregated state of type I and the increase of particles in a segregated state of type II are functions of f_I and f_{II}, respectively.

3.3.2 Rate at Which Particles Mix

The mechanism of mixing can be symbolically represented as:

$$[A + B] \xrightarrow{\text{K}_1} C \tag{22}$$

where

$$K_1 = \text{mixing constant}$$

The rate at which the number of particles in a segregated state of type I decreases is considered to be proportional to f_I.

$$\begin{array}{l}\text{Rate at which particles} \\ \text{in the segregated state} \\ \text{of type I decrease}\end{array} = -\frac{df_I}{dt} = K_1 \, f_I \tag{23}$$

Integration of equation (23) with the initial condition $f_I = 1$ at $t = 0$ leads to

$$f_I = e^{-K_1 \, t} \tag{24}$$

Equation (24) represents the degree of segregation contributed by the particles in the segregated state of type I, at any moment. A schematical representation of the mechanism of mixing is shown in Fig. 2.

3.3.3 Rate at Which Particles Segregate

It is assumed here that the segregation process can be represented in terms of an autocatalytic reaction mechanism (Boudart, 1968):

$$C \xrightarrow{\text{K}_2} [B + A] \tag{25}$$

$$C + [B + A] \xrightarrow{\text{K}_3} [B + A] \tag{26}$$

The above mechanism represents a situation where the product acts as a catalyst to greatly accelerate the breakdown of C. In this case $K_3 >>> K_2$.

It seems reasonable to assume at this point that the driving force for

segregation is proportional to the distance away from equilibrium. This force is defined as the segregation potential ψ by:

$$\psi = \frac{\sigma_\infty^2}{\sigma_o^2} - f_{11} \tag{27}$$

where f_{11} is defined by equation (21) and the ratio $\frac{\sigma_\infty^2}{\sigma_o^2}$ indicates the most likely degree of segregation after particles have been agitated for an infinite time (σ_∞^2 and σ_o^2 are the variances at $t \sim \infty$ and at $t = 0$, respectively). According to this definition for the demixing potential, at $t \sim \infty$

$$(f_{11})_\infty = \frac{\sigma_\infty^2}{\sigma_o^2} \tag{28}$$

and the driving force to produce a segregated state of type II (segregation potential, ψ) will vanish. In other words, an equilibrium will be obtained. In the ideal case in which a complete separation of the components is achieved, σ_∞^2 will be equal to σ_o^2, and the segregation potential becomes $(1 - f_{11})$. A schematical representation of the mechanisms is shown in Fig. 2. The rate, r_1, at which particles form a segregated state of type II according to equation (25) is given by:

$$r_1 = \frac{df_{11}}{dt} = K_2 \psi = K_2 \left(\frac{\sigma_\infty^2}{\sigma_o^2} - f_{11}\right) \tag{29}$$

In here it is assumed that r_1 is first order with respect to ψ. The rate, r_2, at which particles form the segregated state of type II according to equation (26) is given by:

$$r_2 = \frac{df_{11}}{dt} = K_3 \psi f_{11} = K_3 \left(\frac{\sigma_\infty^2}{\sigma_o^2} - f_{11}\right) f_{11} \tag{30}$$

Here it is assumed that r_2 is first order with respect to ψ and first order with respect to f_{II}. The overall rate of formation of the segregated state of type II is then:

$$r_0 = r_1 + r_2 = K_2\left(\frac{\sigma_\infty^2}{\sigma_o^2} - f_{II}\right) + K_3\left(\frac{\sigma_\infty^2}{\sigma_o^2} - f_{II}\right) f_{II} \tag{31}$$

Thus:

$$\frac{df_{II}}{dt} = (K_2 + K_3 f_{II})\left(\frac{\sigma_\infty^2}{\sigma_o^2} - f_{II}\right) \tag{32}$$

Setting K_2/K_3 equal to ρ, separating variables and integrating leads (see Appendix) to the following result:

$$K_3 t = \frac{1}{\rho + \frac{\sigma_\infty^2}{\sigma_o^2}}\left[\ln\left\{\frac{f_{II} + \rho}{\frac{\sigma_\infty^2}{\sigma_o^2} - f_{II}}\right\} - \ln \zeta\right] \tag{33}$$

where ζ is an integration constant. Equation (33) can be transformed to

$$\frac{f_{II} + \rho}{\frac{\sigma_\infty^2}{\sigma_o^2} - f_{II}} = \zeta \exp\left[K_3\left(\rho + \frac{\sigma_\infty^2}{\sigma_o^2}\right)t\right]$$

thus:

$$f_{II} = \frac{\zeta \frac{\sigma_\infty^2}{\sigma_o^2} \exp\left[K_3\left(\rho + \frac{\sigma_\infty^2}{\sigma_o^2}\right)t\right] - \rho}{1 + \zeta \exp\left[K_3\left(\rho + \frac{\sigma_\infty^2}{\sigma_o^2}\right)t\right]} \tag{34}$$

By considering the initial condition $f_{II} = 0$ at $t = 0$

$$\zeta = \rho \frac{\sigma_o^2}{\sigma_\infty^2}$$

and

$$f_{II} = \frac{\rho\{\exp[K_3(\rho + \frac{\sigma_\infty^2}{\sigma_o^2})t] - 1\}}{1 + \rho \frac{\sigma_o^2}{\sigma_\infty^2} \exp[K_3(\rho + \frac{\sigma_\infty^2}{\sigma_o^2})t]} \tag{35}$$

Equation (35) represents the degree of segregation contributed by particles in the segregated state of type II at any moment.

On the basis that mixing and segregation can be described independently, the overall degree of segregation of the system is equal to the linear combination of f_I and f_{II} (additive properties of the variance). According to this the following expression results:

$$f_{total} = f_I + f_{II} \tag{36}$$

And by substitution of equations (24) and (35) into equation (36):

$$f_{total} = e^{-K_1 t} + \frac{\rho\{\exp[K_3(\rho + \frac{\sigma_\infty^2}{\sigma_o^2})t] - 1\}}{1 + \rho \frac{\sigma_o^2}{\sigma_\infty^2} \exp[K_3(\rho + \frac{\sigma_\infty^2}{\sigma_o^2})t]} \tag{37}$$

According to the definition for degree of mixedness given by equation (18),

$$M = 1 - e^{-K_1 t} - \frac{\rho\{\exp[K_3(\rho + \frac{\sigma_\infty^2}{\sigma_o^2})t] - 1\}}{1 + \rho \frac{\sigma_o^2}{\sigma_\infty^2} \exp[K_3(\rho + \frac{\sigma_\infty^2}{\sigma_o^2})t]} \tag{38}$$

when

$$t = 0 \qquad M = 0$$

$$t \sim \infty \qquad M_e = 1 - \frac{\sigma_\infty^2}{\sigma_o^2}$$

Inspection of equation (38) indicates that mixing predominates initially, and that after some period of time the mixing term becomes negligible, leaving only the term that accounts for segregation. For particles with identical properties $(K_2 = 0)$, equation (38) becomes:

$$M = 1 - e^{-K_1 t} \tag{39}$$

An equation similar in form to (39) can be obtained by making an algebraic transformation in Weidanz's model (with $C = 0$) or by assuming an efficiency equal to 1 in Rose's model (with $B = 0$). However, for particles with differences in physical properties, the models are completely different.

When the models [equations (4), (16), and (38)] are applied to mixers operated in a semibatch process like the case studied (in which the particles are the system under consideration), the time of mixing can be considered to be proportional to an integer number of passes in the case of small end effects. For example,

$$t \simeq \alpha \left(\frac{time}{pass} \right) N.$$

NOTATION

A   Constant of mixing in Rose's model

B   Constant of demixing in Rose's model

E   Constant

f   Ratio between the variance among samples at time t and that at t = 0

K_1 Constant of mixing in the Semi-Empirical Kinetic model

K_2 Constant

K_3 Constant

M   Degree of mixedness

M_e Degree of mixedness at equilibrium

n   Number of samples

N   Number of passes

Q   Constant

V_h Upper fraction of the total volume

Y   Volumetric proportions of one of the constituents

GREEK LETTERS

σ_o Standard deviation at t = 0

σ_∞ Standard deviation at t $\sim \infty$

σ Standard deviation

ψ Segregation potential

η Efficiency

ρ Constant

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APPENDIX

Derivation of equation (34) from (32)

$$\frac{df_{11}}{dt} = (K_2 + K_3 f_{11})(\frac{\sigma_\infty^2}{\sigma_o^2} - f_{11}) \qquad (A-1)$$

factorizing K_3 in (A-1):

$$\frac{df_{11}}{dt} = K_3 (\frac{K_2}{K_3} + f_{11})(\frac{\sigma_\infty^2}{\sigma_o^2} - f_{11}) \qquad (A-2)$$

setting $\frac{K_2}{K_3}$ equal to ρ leads to:

$$\frac{df_{11}}{dt} = K_3 (\rho + f_{11})(\frac{\sigma_\infty^2}{\sigma_o^2} - f_{11}) \qquad (A-3)$$

separating variables:

$$\int \frac{df_{11}}{[\rho + f_{11}][\frac{\sigma_\infty^2}{\sigma_o^2} - f_{11}]} = K_3 \int dt \qquad (A-4)$$

According to Mathematical Handbook (by Spiegel,[*] 1968), (A-4) can be integrated as:

$$\int \frac{dx}{(ax + b)(q + px)} = \frac{1}{bp - aq} [\ln(\frac{px + q}{ax + b}) + \ln \zeta] \qquad (A-5)$$

making analogy to equation (A-5), equation (A-4) becomes:

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[*] Spiegel, R. M., "Mathematical handbook of formulas and tables," Schaum's outline series, McGraw-Hill Book Co., 62 (1968).

$$a = 1 \qquad\qquad q = \frac{\sigma_\infty^2}{\sigma_o^2}$$

$$b = \rho$$

$$p = -1$$

$$K_3 t = \int \frac{df_{||}}{(f_{||} + \rho)(\frac{\sigma_\infty^2}{\sigma_o^2} - f_{||})} = \frac{1}{-\rho - \frac{\sigma_\infty^2}{\sigma_o^2}}[\ln(\frac{\frac{\sigma_\infty^2}{\sigma_o^2} - f_{||}}{\rho + f_{||}}) + \ln \zeta] \qquad (A-6)$$

changing the sign in righthand side of (A-6) gives:

$$K_3 t = \frac{1}{\rho + \frac{\sigma_\infty^2}{\sigma_o^2}}[\ln(\frac{\rho + f_{||}}{\frac{\sigma_\infty^2}{\sigma_o^2} - f_{||}}) - \ln \zeta] \qquad (A-7)$$

(A-7) is equation (33) in the text. Multiplying (A-7) by $(\rho + \frac{\sigma_\infty^2}{\sigma_o^2})$ gives:

$$K_3 (\rho + \frac{\sigma_\infty^2}{\sigma_o^2}) t = \ln\{\frac{\rho + f_{||}}{\zeta (\frac{\sigma_\infty^2}{\sigma_o^2} - f_{||})}\} \qquad (A-8)$$

or:

$$\exp[K_3 (\rho + \frac{\sigma_\infty^2}{\sigma_o^2}) t] = \frac{f_{11} + \rho}{\zeta (\frac{\sigma_\infty^2}{\sigma_o^2} - f_{11})} \qquad (A-9)$$

Multiplying equation (A-9) by $\zeta(\frac{\sigma_\infty^2}{\sigma_o^2} - f_{11})$ gives:

$$\zeta \frac{\sigma_\infty^2}{\sigma_o^2} \exp[K_3 (\rho + \frac{\sigma_\infty^2}{\sigma_o^2}) t] - \zeta f_{11} \exp[K_3 (\rho + \frac{\sigma_\infty^2}{\sigma_o^2}) t] = \rho + f_{11} \qquad (A-10)$$

Equation (A-10) can be rewritten as:

$$\zeta \frac{\sigma_\infty^2}{\sigma_o^2} \exp[K_3 (\rho + \frac{\sigma_\infty^2}{\sigma_o^2}) t] - \rho = f_{11} \{1 + \zeta \exp[K_3 (\rho + \frac{\sigma_\infty^2}{\sigma_o^2}) t]\} \qquad (A-11)$$

or

$$f_{11} = \frac{\zeta \frac{\sigma_\infty^2}{\sigma_o^2} \exp[K_3 (\rho + \frac{\sigma_\infty^2}{\sigma_o^2}) t] - \rho}{1 + \zeta \exp[K_3 (\rho + \frac{\sigma_\infty^2}{\sigma_o^2}) t]} \qquad (A-12)$$

This is equation (34) in the text.

CHAPTER 4

EXPERIMENTAL WORK

4.1 INTRODUCTION

This section presents a description of the apparatus, particle systems and the experimental procedures employed in this work.

4.2 APPARATUS

The experimental apparatus is shown schematically in Fig. 1. It consisted of a feeder, a motionless mixer, a collector, a simple gate valve, and a sampling device.

The motionless mixer used in this work was constructed from yellow sheet brass (0.025 inch thickness). The sheet brass was sheared into strips 1.5 inches wide and the strips were twisted on a lathe to form right handed and left handed helical ribbons of the same number turns per unit length. The ribbons were cut every 180° to give helical elements (helices).

The helices were joined with solder in an alternating pattern of clockwise and counterclockwise elements meeting at right angles to one another to form a chain. The chain of helices was then inserted in a 1.5 inch ID pyrex tube of the same length as the chain of helices. Friction between the tube wall and the edges of the helices prevented slipping.

Figure 2 presents a photograph of a two-element unit which illustrates the geometric configuration. This particular geometric arrangement results in multiple motions being imposed on the flowing stream. On contacting the first element the stream is split into two parts and each part is twisted in
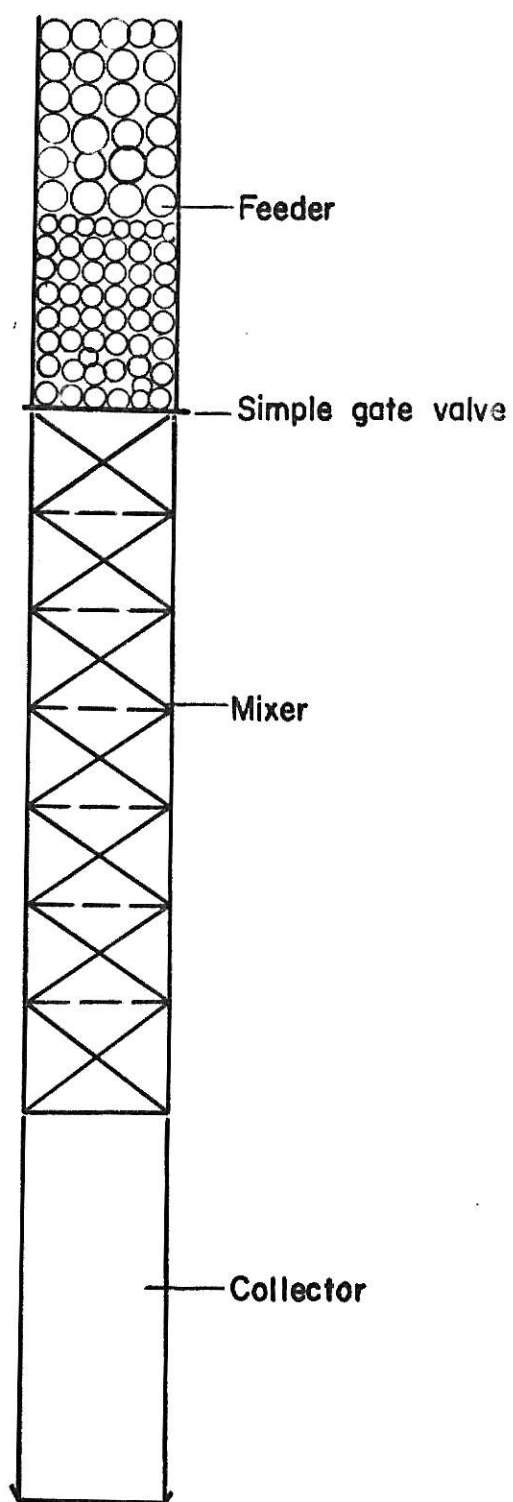
Fig. 1. Schematical representation of an experimental set up.

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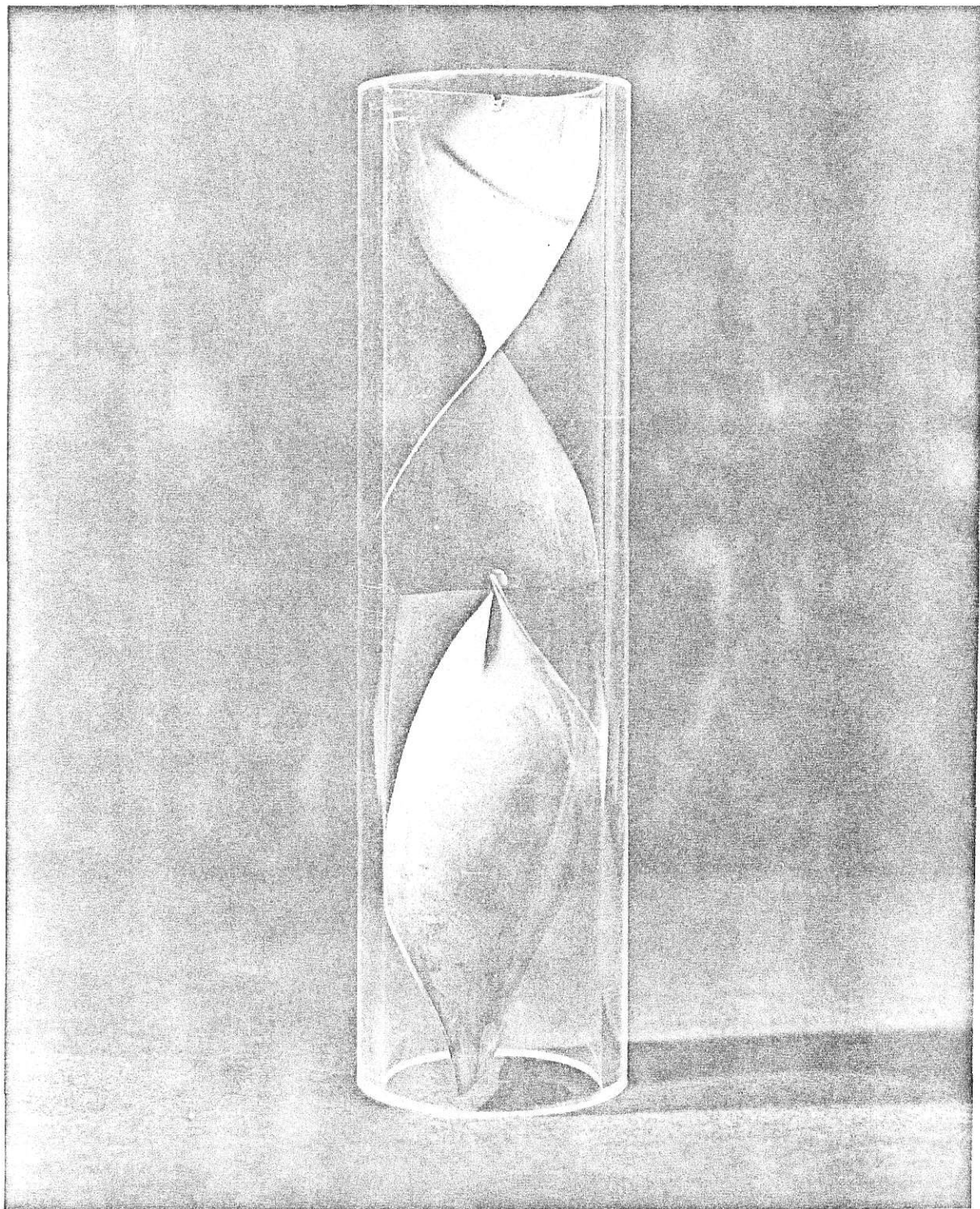THIS IS AS RECEIVED FROM CUSTOMER.

Fig. 2. A motionless mixer similar to the one described in the text.

either a clockwise or a counterclockwise direction depending on the configu-
ration of the elements. On contacting the second element the two streams
are split into four parts and twisted in a direction opposite to that one of
the first element. This process is repeated; twisting in opposite directions
with alternating elements and dividing the flowing stream into an increasing
number of strata with each element. Interaction of the particles with the
helices, with the wall of the mixer and with each other gives additional
mixing action. Although the mechanism of shear mixing may not be as promi-
nent as it is in mixing of fluids forced through the mixer, mixing does occur
as a result of the differences in velocities of particles flowing along the
surfaces of the helices and wall.

Mixers of 2, 5, 7, 8, 9, and 12 helices were employed in this study.
All of these had the same geometric characteristics with the exception of the
length of the chain of helices. The feeder was simply a 1 foot length of
pyrex glass tube 1.5" ID. The collector is identical in dimensions to the
feeder and was graduated at one-half inch intervals along the length of the
tube to serve as a guide for sampling purposes. The gate valve was simply a
piece of filter paper fixed to the bottom of the feeder with a rubber band.
When the rubber band was removed the paper could be pulled out rapidly.

Samples of approximately equal volume were removed from the collector
at the end of each experiment. In the case of all but the extremely dense
particles, the particles were removed from the collector by gentle suction.
A glass tube with a diameter only slightly larger than the particles permit-
ted the withdrawing of the particles in the sample one by one. This device
made it possible to obtain a near uniform interface between samples and did
not produce any disturbance of the mixture in the sampling process. In the

case of dense particles an apparatus similar to that designed by Fusternau (1967) was employed. This device was simply a 1.5 inch ID plexiglass tube which was divided into sections of equal size (1 inch), so that they could be removed section by section. Figure 3 illustrates schematically this sampler together with one cylindrical section.

4.3 PARTICLE SYSTEMS

Four basic systems were employed. These consisted of (1) particles with identical physical properties (size, shape, density and material) distinguishable only by difference in color; (2) particles differing only in size; (3) particles differing in density (material differs here also; and (4) particles differing in both size and density (material differs here also).

Spherical particles made of lucite, glass, and steel were employed. Their physical properties are listed in Table 1. For the case of particles with identical properties, two size levels were considered. Particles were not available to permit experiments at different density levels. For the case of size differences, the size of the smaller particles was held constant while the diameter of the larger particles was varied. A similar situation was possible for some of the experiments involving density differences. However, one experiment with steel and glass particles did not fit this criteria. Only one combination was examined for both size and density differences. The particles systems employed are summarized in Table 2.

4.4 EXPERIMENTAL PROCEDURE

Experiments were carried out by following three basic operations: loading, passing and sampling. Mixing experiments were divided as: mixing of particles differing only by color, mixing of particles with differences in
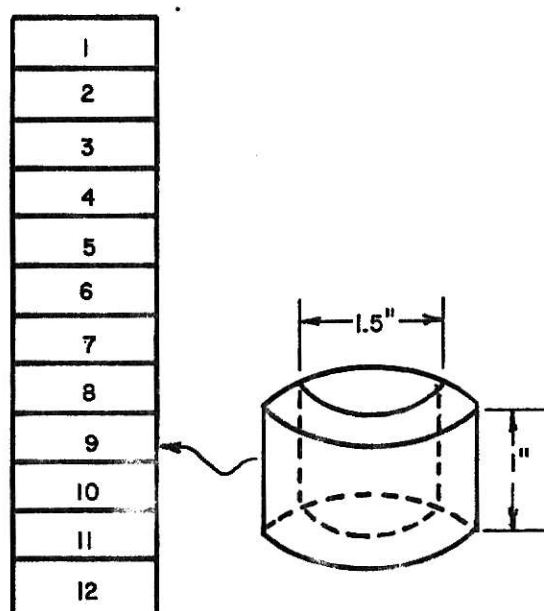
Fig. 3. Schematic diagram of the sampler with one of
the removable sections.

TABLE 1

PROPERTIES OF THE SOLID PARTICLES

Material	Particle diameter (inch)	Average weight (g)	Standard deviation (g)	Density (g/cc)
Lucite	1/8	0.0184	0.00113	1.18
Lucite	5/32	0.0384	0.00068	1.18
Lucite	3/16	0.0684	0.0032	1.18
Glass	3/16	0.145	0.0026	2.42
Steel	3/16	0.429	0.000	7.6

TABLE 2

DESCRIPTION OF THE EXPERIMENTS

System	Particles diameter (inch)	Overall composition of key particles	Number of helices	Sampling interval (inch)	Sampling technique
White and red lucite	1/8	0.5	2, 5, 12	0.5	Suction
White and red lucite	5/32	0.5	2, 5, 12	1.0	Suction
White and red lucite	1/8-5/32	0.6	2, 5, 7, 9, 12	0.5	Suction
White and red lucite	1/8-3/16	0.68	2, 5, 8, 12	0.5	Suction
Lucite-glass	3/16	0.52	2, 5, 8, 12	1.0	Suction
Lucite	3/16	0.515	12	1.0	Sampler in Fig. 3.
Glass-steel	3/16	0.5	12	1.0	Sampler in Fig. 3.
Lucite-steel	3/16	0.52	12	1.0	Sampler in Fig. 3.
Lucite-glass	5/32-3/16	0.58	2, 5, 8, 12	1.0	Sampler in Fig. 3.

size, mixing of particles with differences in density and mixing of particles with both size and density differences.

The loading process consisted of closing the bottom of the collector and feeder with filter paper and pouring the particles into the feeder. The key particles (small, light or colored) were poured in first and the second component was carefully layered on top to avoid disturbing the interface. The total number of particles was constant for each individual system studied, but varied between individual systems.

After loading, the feeder was placed above the mixer as indicated in Fig. 1. The filter paper gate valve was withdrawn rapidly, permitting the slug of particles to fall through the mixer. The particles were caught in the collector which was closed at the bottom with filter paper. The feeder was removed and the bottom again closed. The feeder and collector were then interchanged to permit a second pass   thout disturbing the bed of particles in the process. This process was repeated an even number of times to give the desired number of passes.

After the desired number of passes was reached samples of approximately equal volume were withdrawn from the collector. A sample interval of either 1/2 inch or 1 inch was employed, depending on particle size, the former being used for the smaller particles.

Particles were withdrawn from the collector starting at the top. In most cases these were removed with the suction device with care being taken to obtain a uniform interface between samples. The entire mixture was divided into samples by this method. In the case of extremely dense particles, a sectioned collector was used which permitted the samples (1" intervals) to be obtained by dismantling the collector section by section from top to bot-

tom. The samples thus obtained were separated manually (with aid of U. S. standard sieves when the particles differed in size) to obtain a count on the number of key particles and total particles in each sample.

Experiments were conducted with each system of particles over a range of number of passes ranging from 2 to about 30. These experiments were repeated with several mixers containing from 2 to 12 helices. In all cases the fraction of key particles ranged from 0.5 to 0.7. The total number of particles in the slug ranged from 5000-7000. Table 2 summarizes the various experimental combinations examined.

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CHAPTER 5

TREATMENT OF DATA

5.1 INTRODUCTION

This section presents a summary of the fundamental definitions and the methods employed to analyze the data. After passing the slug of particles through the mixer the desired number of times, the slug was divided into axial samples of approximately equal volume. The average concentration or fraction of key particles (small or light) was determined for each sample and recorded as a function of axial position. These data were utilized directly to evaluate the variance, standard deviation, and degree of mixedness.

5.2 DEFINITIONS

5.2.1 Average Concentration

The average concentration for a sample is defined as the number of key particles in the sample, divided by the total number of particles in the sample, i.e., if a sample has 270 small particles and the total number of particles is 450, then the average concentration is 0.6. The overall concentration is defined as the total number of key particles divided by the total number of the particles in the slug.

5.2.2 Variance and Standard Deviation

Because of the random nature of mixing operations, statistical analysis becomes the approach most frequently utilized for examining data. The stan-

dard deviation and the variance of samples taken from a mixture are the most frequently employed for the purpose of analysis.

The variance is denoted by σ^2 and is defined as:

$$\sigma^2 = \sum_{i=1}^{n} \frac{(X_i - \bar{X})^2}{n - 1} \tag{1}$$

where

X_i = average concentration of the key particles (small or light) in a selected axial sample,

$\bar{X}$ = overall concentration of the key particles in the total mixture,

n = the number of samples taken from the mixture. For the present case, the entire mixture was divided into samples.

The positive square root of the variance is called the standard deviation and is denoted by σ, i.e.,

$$\sigma = + \sqrt{\sigma^2} \tag{2}$$

Roughly speaking, the variance is a measure of the spread or dispersion of the values which the variable X_i can assume.

5.2.3 Degree of Mixedness

Definitions for a degree of mixedness in terms of a minimum number of parameters usually employ the variance or standard deviation to describe the state of a mixture. In the present work two definitions for degree of mixedness are considered.

The first is defined in terms of the standard deviation:

$$M = 1 - \frac{\sigma}{\sigma_o} \tag{3}$$

$$1 \geq M \geq 0$$

where σ is the standard deviation of the total number of samples taken from the mixture as defined by equation (2), and σ_o is the initial standard deviation, i.e., before mixing. For a mixture of two components σ_o is given by Lacey (1954) as:

$$\sigma_o{}^2 = \bar{X}(1 - \bar{X}) \tag{4}$$

Here $\bar{X}$ is the overall composition of the key particles in the mixture. The above definition of the degree of mixedness was employed in the model developed by Rose (1959) which was discussed in section 3.2.1.

The second index of mixedness is defined in terms of the variance:

$$M = 1 - \frac{\sigma^2}{\sigma_o{}^2} \tag{5}$$

where σ^2 and $\sigma_o{}^2$ are given by equations (1) and (4) respectively. This definition is employed in the Semi-Empirical Kinetic Model developed in this study. This equation was obtained by simplification of Lacey's (1954) definition for degree of mixedness which is stated in section 2.2. It was assumed as an approximation that $\sigma_o{}^2 \gg \sigma_r{}^2$ and that no sampling error exists; for this case equation (5) results. This definition is employed to keep the index positive; it approaches unity for the random mixture. The equation based on variance approaches unity more rapidly than the expression based on the standard deviation. According to Donald and Roseman (1962), only an index based on the variance can be adapted to be theoretically independent of sample size, and thus only this type of index gives a consistently accurate value over the whole course of mixing. Further, such an index is more satisfactory statistically than indices based on other statis-

tical terms, since the variance has additive properties.

5.2.4 Mixing Action Number (M.A.N.)

The Mixing Action Number (M.A.N.) is defined as the product of the number of helices in the mixer and the number of passes. In this case it is assumed that all the geometric characteristics (angle of twisting, length and width of the elements, etc.) except the number of elements must be the same. As an example for a mixer with five helices and the case of four passes, the M.A.N. is equal to 20 (5 helices x 4 passes = 20).

5.2.5 Expansion and Compaction

Expansion or compaction effects were measured after each pass of particles through the mixer. Percentage of expansion is defined as:

$$\% \text{ of expansion} = \frac{V - V_o}{V_o} \times 100 \tag{6}$$

where V_o is the initial volume of the slug of particles and V is the volume of the slug of particles after N passes. If the difference between V and V_o is negative, it is referred to as a percentage of compaction.

The void volume ε is defined as the ratio of the volume of the voids to the volume of the particles as indicated by the following equation:

$$\varepsilon = \frac{V - V_p}{V_p} \tag{7}$$

where

V = volume of the slug of particles

V_p = volume of the particles ($\frac{4\pi R^3}{3}$ x number of particles)

5.3 METHODS OF ANALYSIS

5.3.1 Expansion and Compaction

Expansion or compaction and void volumes were calculated and studied as a function of the number of passes for particles differing only by color. The purpose was to examine the change of the bulk density with respect to the degree of mixedness reached by the particles after a defined number of passes. Plots of void volumes or bulk densities versus M were made to examine the changes graphically.

5.3.2 Concentration Profiles

A plot of the average concentration versus the section number (or relative position) provides an axial concentration profile. In the data to be presented section numbers increase in proceeding from top to the bottom of the collector (i.e., section 1 refers to the sample from the top of the collector). Concentration profiles change with the number of helical elements used, the total number of passes, and the physical properties of the particles. Profiles provide a detailed description of the progress of mixing and segregation as well as means for graphically comparing the effects of varying the physical properties of the particles.

5.3.3 Standard Deviation vs. Number of Passes

A plot of the standard deviation versus the number of passes provides one means for describing the overall progress of mixing. In the present work a semilog plot is employed (log σ vs N). This type of plot results in a very short curvilinear transient period at the start of a run, a linear period, and a steady state period that indicates a diffusional mixing when particles have identical physical properties. Or, a transient, a linear,

and a region in which the standard deviation increases when particles present differences in physical properties.

The linear region indicates that mixing is the mechanism that predominates. In this region the particles are mixed principally by the mutual interaction of the particles, and by the differences in velocities of the stream of particles along the cross section of the mixer. The process of mixing in this region is called convective. Straight lines obtained for this portion were correlated as a function of the number of passes. Slopes of the straight lines (ϕ), identified as coefficients of mixing velocities (Yano, et al., 1960) were calculated by a linear least square regression analysis.

These plots provided a means to extrapolate the values of the standard deviation and estimate the value of σ_∞ (value reached by the standard deviation after a large number of passes).

Plots of σ vs N were employed to represent experimental data for comparison with Weidanz's model.

5.3.4 Degree of Mixedness vs. Number of Passes

A plot of the degree of mixedness vs number of passes provided another means for describing the overall progress of mixing. Two definitions of the degree of mixedness were employed in this study as given by equations (3) and (5). The curves provided a means for graphically comparing the effects of variables such as the number of helices and the physical properties of the particles. For this purpose only the definition for the degree of mixedness based on the variance was employed. In addition the curves were also used to compare the theoretical predictions of the models with the experimental data.

5.3.5 Degree of Mixedness vs. M.A.N.

A plot of the degree of mixedness vs the Mixing Action Number (M.A.N.) provides a test of the assumption of negligible end effects (it appears that the major factor influencing the ability of the plot to correlate data will be the significance of end effects). If these are small a good correlation will be expected. Data in this form also provide a means for graphically comparing the effects of changes in the physical properties of the mixer.

5.3.6 Comparison of Models with Experimental Data

The experimental data were utilized to determine the parameters which characterize each of the three models considered in the present work. Bard's method for nonlinear parameter estimation was employed for this purpose. The parameters of η, A and B were estimated for Rose's model with data expressed in terms of the degree of mixedness as a function of the number of passes (assuming that the time of mixing is proportional to an integer number of passes). Here the degree of mixedness was based on the standard deviation as defined by equation (3). The best fit parameters were then used to calculate points for the M vs N curve with the aid of equation (4) in Chapter 3. Both graphical and numerical comparisons were made between the predicted and experimental data. Data were examined for different number of helices in the mixer as well as for a variety of combinations of particle physical properties.

The parameters Q and E of Weidanz's model were estimated with data expressed in terms of the standard deviation as a function of the number of passes (assuming as an approximation that the time of mixing is proportional to an integer number of passes). The best fit parameters were then used to calculate points for the σ vs N curve with the aid of equation (16) in Chap-

ter 3. Both graphical and numerical comparisons were made between the pre-
dicted and experimental data. Data were examined for different number of
helices in the mixer as well as for a variety of combinations of particle
physical properties.

The parameters K_1, K_2 and K_3 of the proposed kinetic model were
estimated from data expressed in terms of the degree of mixedness as a
function of the number of passes (assuming that the time of mixing is pro-
portional to an integer number of passes). Here the degree of mixedness was
based on the variance as defined by equation (5). The best fit parameters
were utilized to calculate points for the M vs N curve with the aid of equa-
tion (38) in Chapter 3, and the estimated value of σ_∞. The method employed
to estimate σ_∞ was outlined in section 5.3.3. Both graphical and numerical
comparisons were made between the predicted and experimental data. Data
were examined for different number of helices in the mixer as well as for a
variety of combinations of particle physical properties. Plots of the para-
meters K_1, K_2 and K_3 as functions of the number of helices in the mixer were
also made.

5.3.7 Curve Fitting Methods

Bard's program for nonlinear parameter estimation was used to fit
experimental data to the proposed models. This program can handle up to 20
parameters, 10 variables per experiment, and 200 experiments. The central
function of the program is to find the values of the parameters (θ) that
minimize the objective function $G(\theta)$ (difference between the proposed model
and the experimental data). The Gauss-Newton method, with modifications by
Greenstadt-Eisespress (1967), Bard (1967), and Carroll (1961) is used for
effecting the minimization. The minimization of the function proceeds in an

iterative fashion. Each iteration is started with an initial guess for the parameters and proceeds to find a new guess such that $G(\theta') < G(\theta°)$. This now becomes the initial guess for the next iteration. In the course of each iteration two factors must be determined: a direction $\Delta\theta$ to proceed in, and length λ of the step to be taken along this direction, so that $\theta' = \theta° + \lambda\theta$. To be admissible, the direction must be such that as one proceeds along it from $\theta°$ the value of G decreases at least initially, i.e., for sufficiently small λ. The value of λ is ideally such as to take us near the peak of G along the chosen direction. Also, λ must be small enough to insure that θ' satisfies all bounds and constraints on the parameters. To use this program, only one subroutine must be written by the user to define the function to be fitted and its derivatives. Upper and lower bounds as well as initial guesses for the parameters have to be specified by the user. The linear least square method was used to estimate the coefficients of mixing velocity.

NOTATION

A	Constant of mixing in Rose's model
B	Constant of demixing in Rose's model
D	Diameter, inch
E	Constant
K_1	Constant of mixing in the Semi-Empirical Kinetic model
K_2	Constant
K_3	Constant
M	Degree of mixedness
N	Number of passes
Q	Constant
V	Volume of the slug of particles after N passes
V_o	Initial volume of the slug of particles
V_p	Volume of the particles
$\bar{X}$	Overall proportion of one of the constituents

GREEK LETTERS

σ	Standard deviation
σ_o	Standard deviation at t = 0
σ_∞	Standard deviation at t
ϵ	Ratio of the volume of the voids to the volume of the particles
θ	Any parameter
ρ	Density, g/cc
ϕ	Coefficient of mixing velocity

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CHAPTER 6

MIXING OF PARTICLES WITH THE SAME SIZE AND DENSITY

6.1 INTRODUCTION

The purpose of this part of the study was to investigate axial mixing in motionless mixers with particles having identical physical properties. The study was concerned with the influence of the diameter of the particles, number of helices in the mixer, and the number of passes on the rate of mixing. Data on the extent of compaction or expansion as a function of the number of passes through the mixer was also obtained. The adequacy of existing models for describing axial mixing in a motionless mixer was examined as well as the new model proposed in this work.

6.2 CONCENTRATION PROFILES

The experimental data were obtained in the form of particle concentration versus axial position in the collector, from which concentration profiles could be recovered easily. As mixing progressed the distribution of sample compositions changed in the direction of a uniform concentration profile. The change in distribution with increasing number of passes is illustrated in Fig. 1.

The characteristics of the concentration profiles obtained with different number of helices in the mixer were examined. For a small number of helices (less than 5) the mixing action took place very slowly; that is, a large number of passes was required to obtain a random mix. For a large number of helices (12) the mixing action was rapid and a random mix was
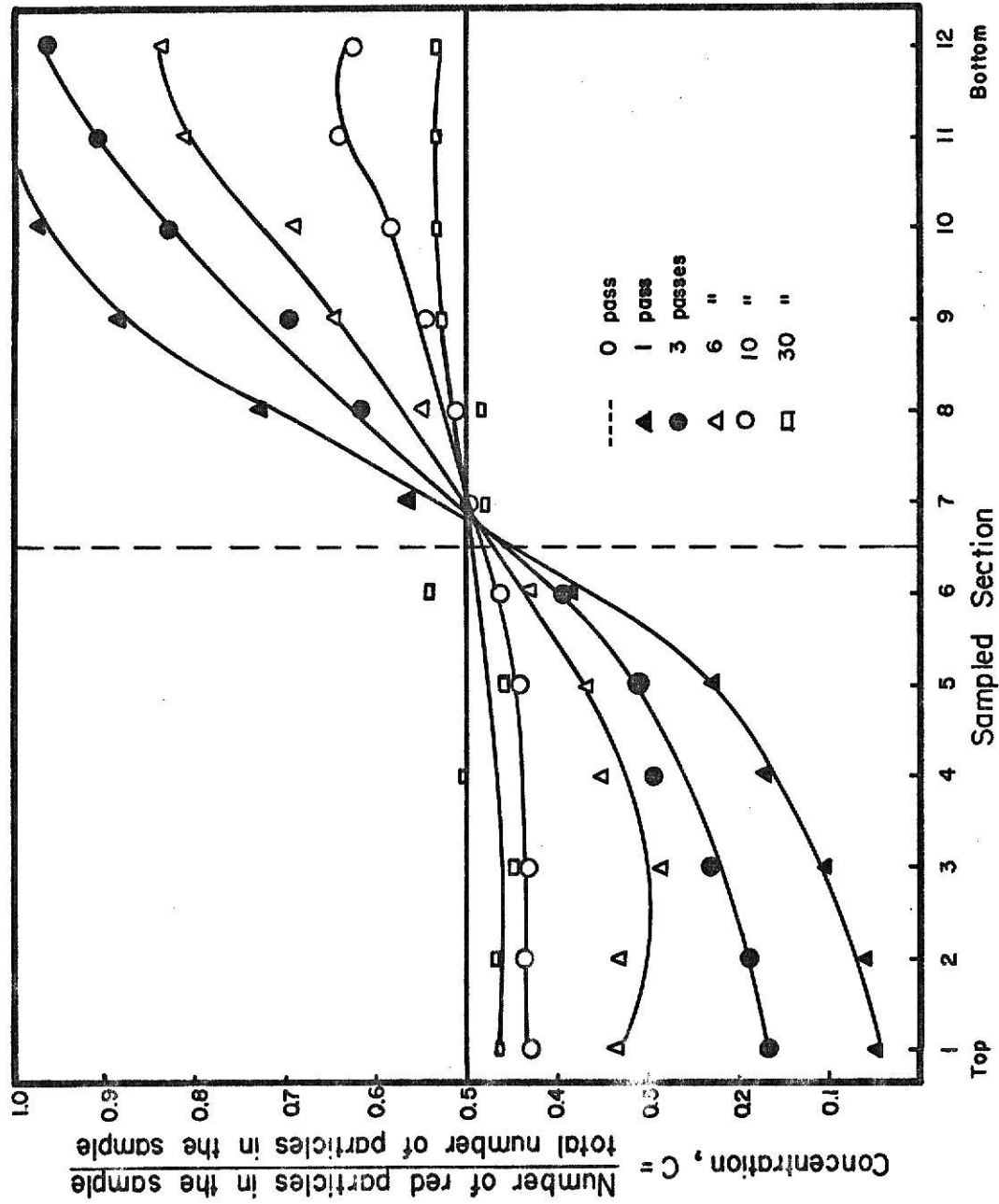
Fig. I. Concentration profiles for a 12 helices mixer with Lucite particles 1/8" in diameter.

obtained with relatively few passes. As the profile approached the overall concentration line (as illustrated by the profile for 30 passes in Fig. 1), it was observed that the experimental points oscillate. The reason for these oscillations probably is that as the mixture approaches the random state any small error in sampling can produce a difference in the average concentrations of the spot samples. In an ideal situation such fluctuations should not appear and the concentration will be the same along the test section when the perfect random mixture is reached.

It was also observed that the concentration profiles cross at a common point. Ideally the point should represent the initial position of the interfacial surface. As expected, the crossing point was between sections 6 and 7; however, it was displaced a little to the right. This displacement could occur as a result of one or more of the following reasons: (1) uneven layering of the particles at the interface initially; (2) bouncing of the particles when they hit the filter paper in the collection procedure; and (3) although the physical properties of the particles were approximately the same, the small deviations present might be sufficient to produce the observed phenomena.

6.3 STANDARD DEVIATION

The composition data can be expressed in terms of standard deviation. A semilog plot of σ vs the number of passes (N) also serves to illustrate the progress of mixing. Typically a plot of log vs N shows a curvilinear transient section at the early stage, a linear section and possibly an equilibrium section (if the product of the number of passes and the number of helices is sufficiently large). For motionless mixers of 2, 5, and 12 helices with 5/32" particles, only a very small transient period followed by

a linear period were noted as shown in Fig. 2. For the same mixers and 1/8"
particles, a steady state section was observed after 10 passes through a 12-
helices mixer.

The linear region of the plot of ln σ vs N can be represented by a
relationship of the form:

$$\sigma = \sigma_o' \, e^{-\phi N} \qquad\qquad\qquad (1)$$

where ϕ can be obtained from the slope of the semilogarithmic plot. Yano,
et al. (1960), identified ϕ as a "coefficient of mixing velocity." Lineari-
zation of equation (1) followed by a linear least square regression analysis
for the experimental data permitted the calculation of σ_o' and the coeffi-
cients of mixing velocity (ϕ).

Table 1 summarizes the calculated value. Examination of the table
shows that ϕ increases as the number of helices in the mixer increases.
This is logically so because as the number of helices in the mixer is aug-
mented, there will be more agitation and this will tend to increase the
homogeneity of the mixture. If results are compared in terms of particle
size, a marked decrease in ϕ is noted as the particle size increases. These
results indicate that the rate of axial mixing is strongly dependent on par-
ticle size, and that a random mix is achieved most rapidly when the particle
diameter is small and the number of helices in the mixer is large.

6.4 DEGREE OF MIXEDNESS VS. NUMBER OF PASSES

The degree of mixedness can be calculated from the variance with the
aid of equation (5) in Chapter 5. A plot of M vs N provides still another
means for illustrating the progress of mixing. Plots of degree of mixedness
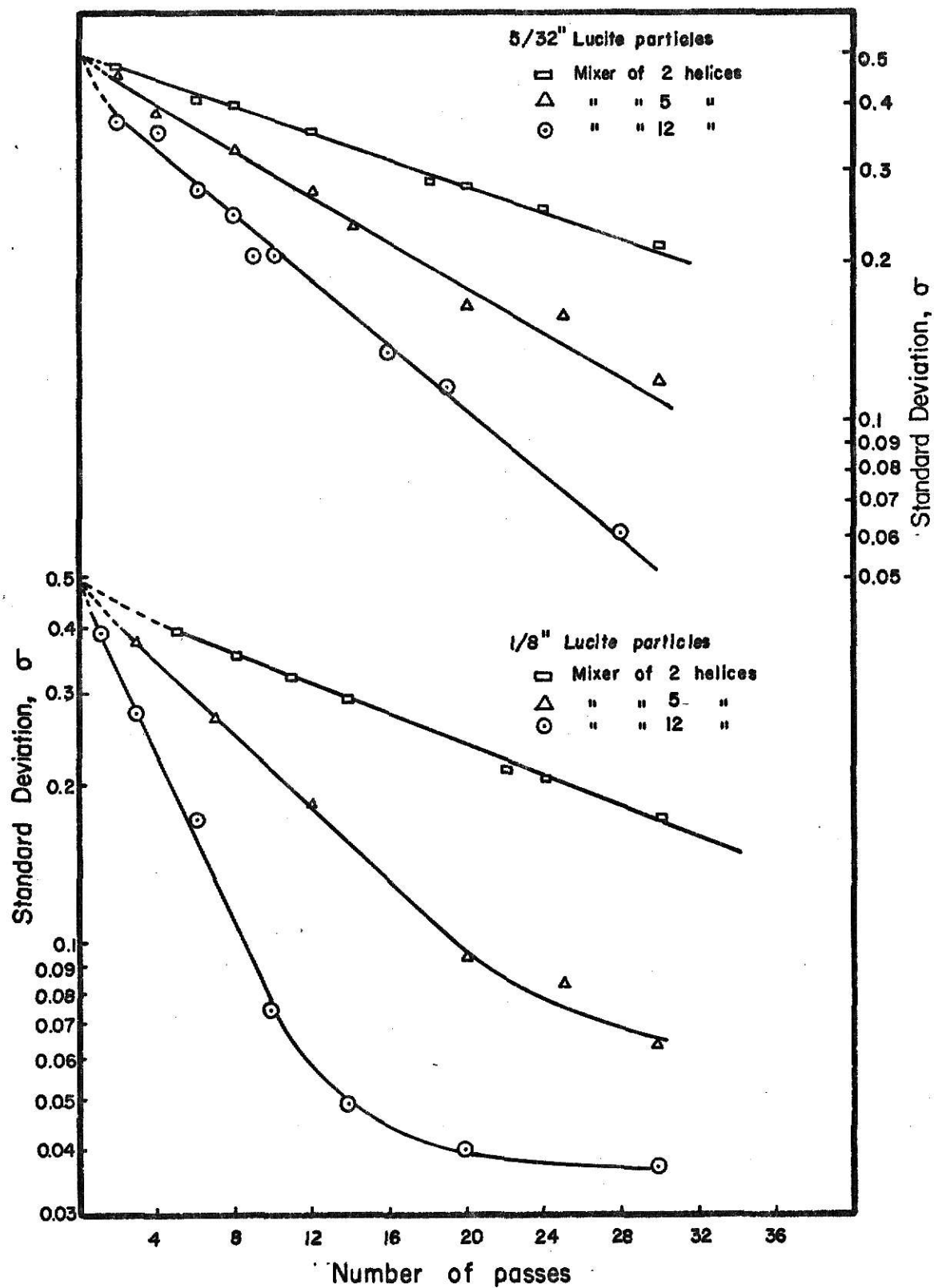vs number of passes are shown in Figs. 3 and 4. Here each point is the

Fig. 2. Correlation of σ vs N° of passes for mixers with particles of identical properties.

TABLE I

CALCULATED VALUES FOR THE COEFFICIENT OF MIXING VELOCITY

No. of helices	Particle diameter (inch)	$\sigma_o{}'$	ϕ (1/pass)
2	1/8	0.47	0.033
5	1/8	0.48	0.081
12	1/8	0.48	0.184
2	5/32	0.49	0.028
5	5/32	0.46	0.042
12	5/32	0.42	0.074
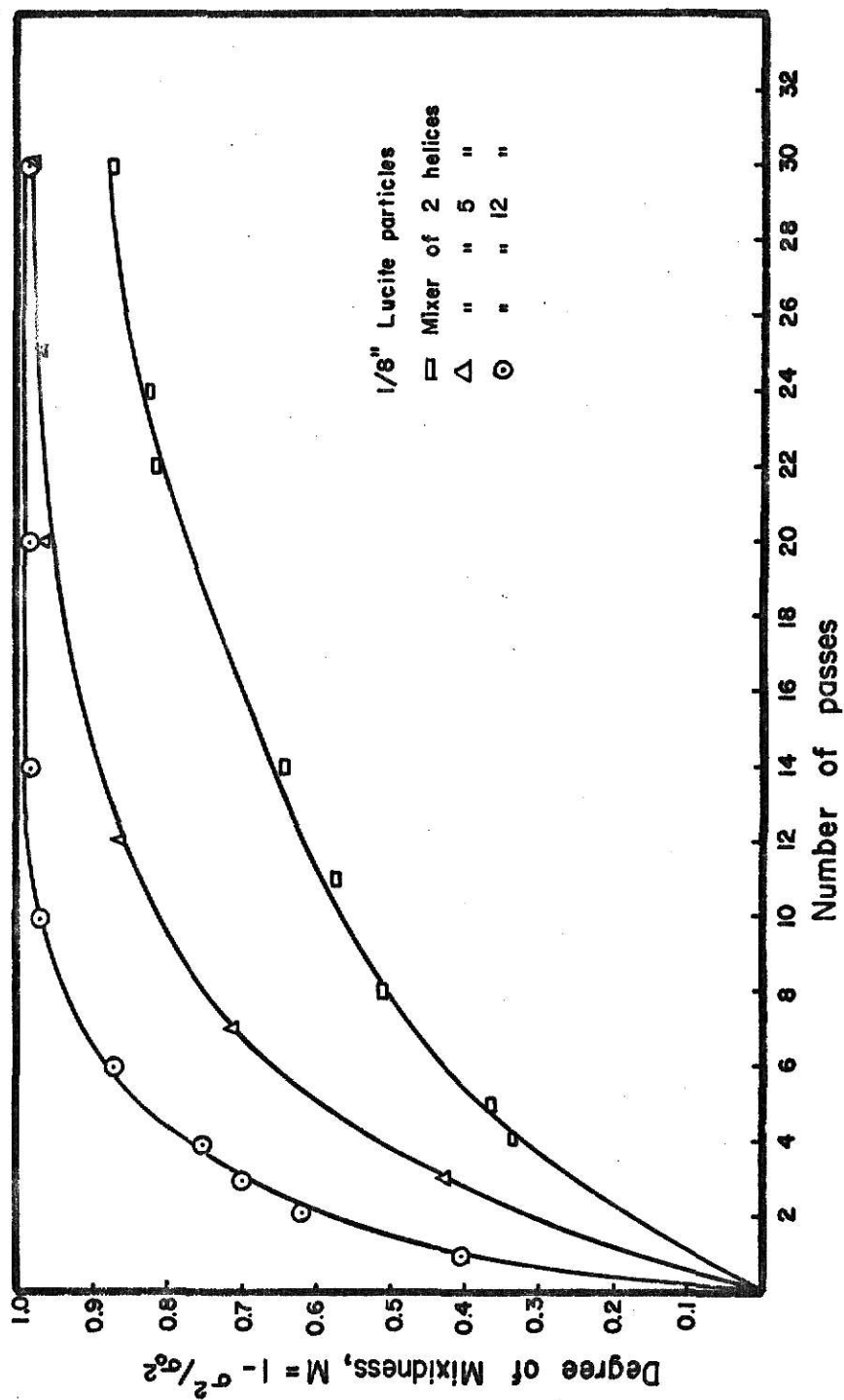
Fig. 3. Rate of axial mixing for particles with same size and density in a motionless mixer.

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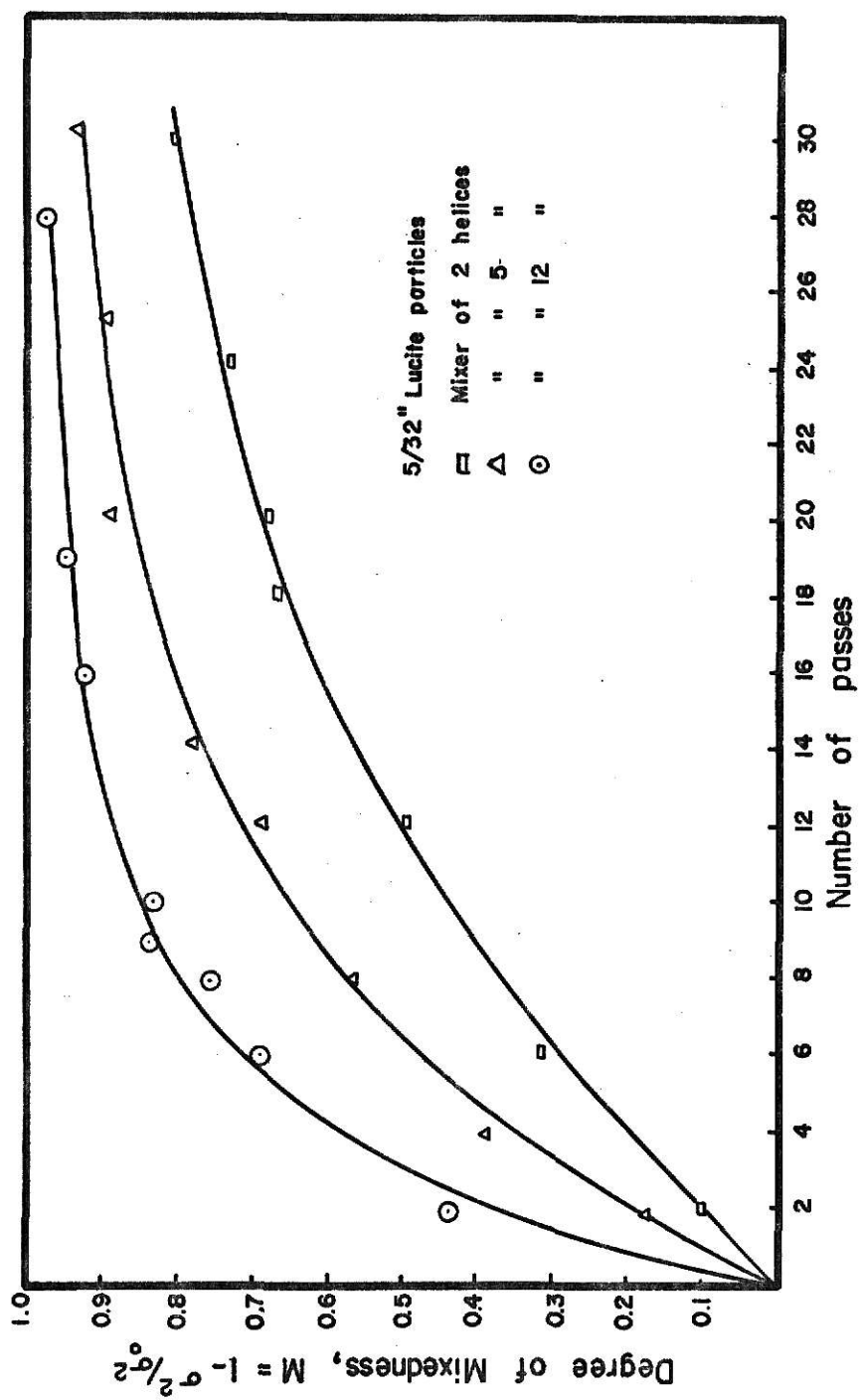
Fig. 4. Rate of axial mixing for particles with same size and density in a motionless mixer.

average of at least two experiments. The plots show that the rate of mixing is very rapid initially and decreases as the number of passes is increased. If the number of passes is sufficiently large an equilibrium will be obtained which is very close to a random mix. As noted previously, mixing progresses most rapidly when the particle size is small and the number of helices is large.

6.5 MIXING ACTION NUMBER (M.A.N.)

M.A.N. were calculated for all the data for each particle size according to the definition given previously. Figures 5 and 6 illustrate the results for 1/8" and 5/32" particles respectively. The plot for 1/8" particles shows a good correlation among the experimental points. For the 5/32" particles some scattering is noted in the experimental data in the region bounded by M.A.N. of 40 to 110. The scatter could be a result of sampling errors, electrostatic effects, or small end effects. Although the correlation is not as good as the one observed for 1/8" particles, it is still satisfactory. On the basis of the results, it can be concluded that the end effects can be considered negligible. Thus, the same degree of mixedness can be reached with a mixer of 2, 5, and 12 helices as long as the Mixing Action Number is the same.

6.6 EXPANSION AND COMPACTION

In the course of the experiments the total volume was also measured as a function of the number of passes. A plot of the percentage of expansion vs void fraction gave straight lines with the same slopes for mixers of 2, 5, and 12 helices. This implies that the percentage of expansion depends only on the initial void fraction (Fan, et al., 1971). Table 2 summarizes
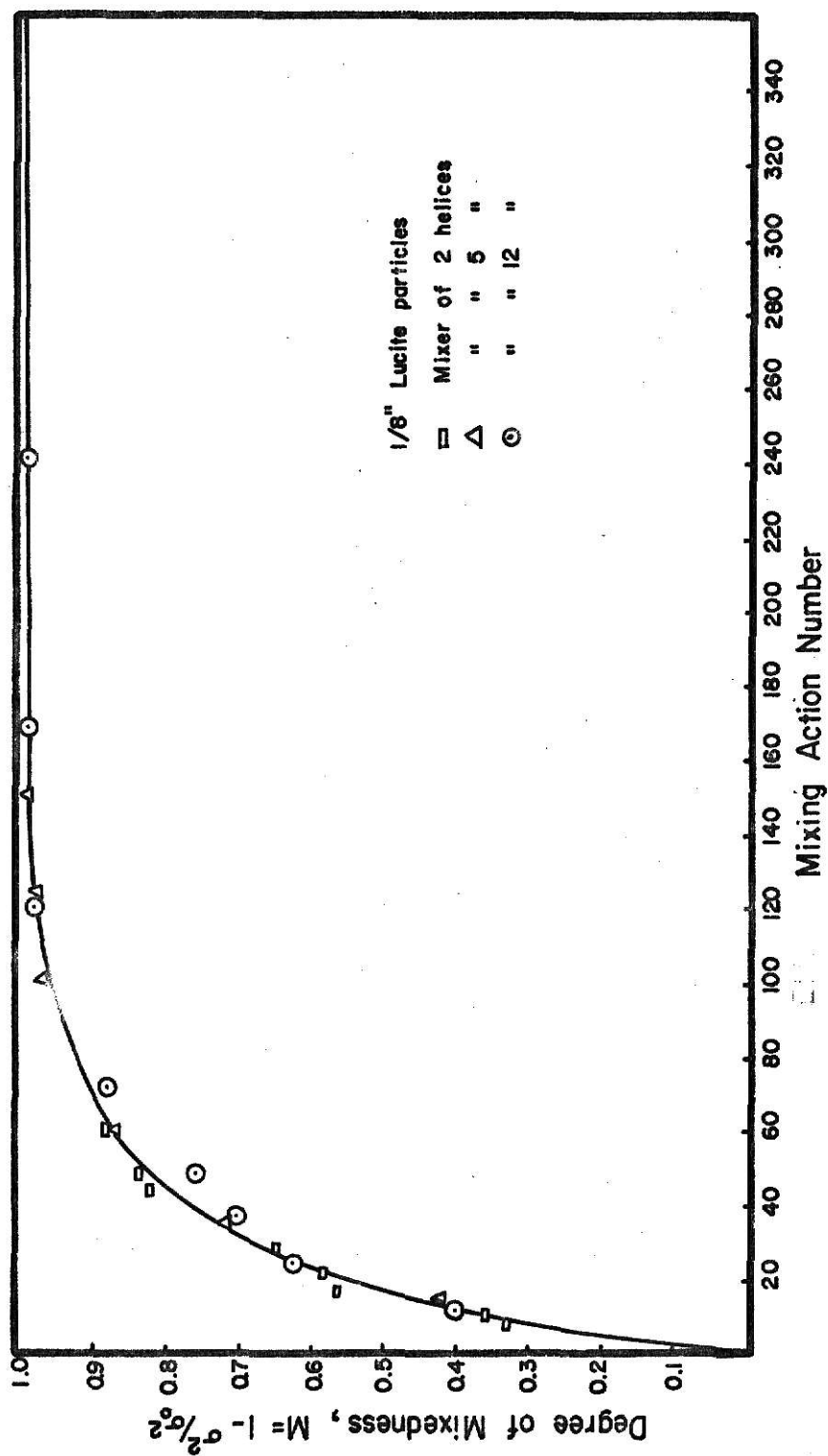
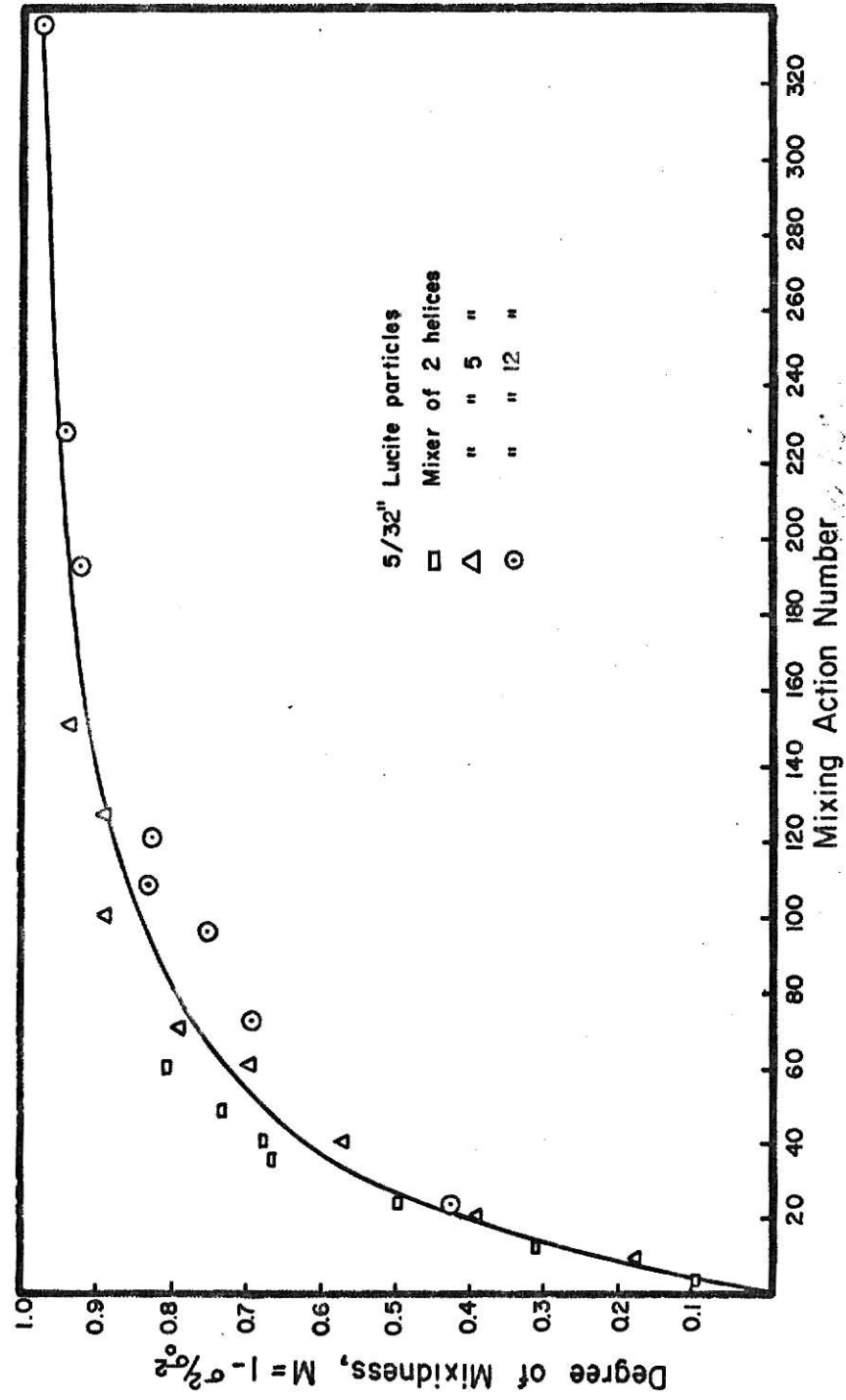Fig. 5. Correlation between degree of mixedness and M.A.N.

Fig. 6. Correlation between degree of mixedness and M.A.N.

TABLE 2

INFLUENCE OF NUMBER OF PASSES IN VOID VOLUME

No. of helices	No. of passes	Particles diameter (inch)	Expansion after N passes $(V - V_0)/V_0$	Void volume $\varepsilon = \dfrac{V - V_p}{V_p}$
5	12	1/8	0.041	Initial: 0.567 After N passes: 0.63
5	20	1/8	0.042	0.54 0.605
5	25	1/8	0.052	0.558 0.639
5	30	1/8	0.031	0.557 0.605
12	2	1/8	0.031	0.553 0.601
12	14	1/8	0.02	0.569 0.601
12	20	1/8	0.031	0.537 0.585
2	6	5/32	0.041	0.649 0.72
2	12	5/32	0.041	0.639 0.7
2	10	5/32	0.035	0.668 0.716
2	24	5/32	0.04	0.658 0.726
12	16	5/32	0.041	0.639 0.707

the pertinent data.

Bulk densities were calculated and are presented in Table 3. A plot of the bulk density vs the degree of mixedness shows that the change in bulk density with the number of passes is very small. Because of the insignificant change (in the bulk density) it was concluded that the expansion has a small effect on the total number of particles (580 ± 40 for 1/8" particles) contained in a spot sample of fixed volume. Therefore it appears reasonable to neglect expansion effects in the present work. In other words, the total number of particles in a given sample is independent of the number of passes.

6.7 ROSE'S MODEL

For the mixing of particles with identical physical properties in a motionless mixer Rose's model becomes:

$$M = \eta \ (1 - e^{-AN}) \tag{2}$$

where

$$M = 1 - \sigma/\sigma_o.$$

The formulation of this equation was considered in section 3.2.1. The parameters η and A of the model were estimated utilizing Bard's technique for nonlinear parameter estimation (1967) for each set of experimental M vs N data. The results are presented in Table 4. A comparison between the experimental data and Rose's model is shown in Fig. 7. The agreement is fairly good, giving rise to sums of squares of residuals of approximately 2×10^{-3} for 2 helices, 6.2×10^{-4} for 5 helices, and 3.7×10^{-3} for 12 helices.

Examination of the calculated parameters (Table 4) shows that the efficiency of the mixer, η, is a function of the number of helices in the mixer,

TABLE 3

INFLUENCE OF DEGREE OF MIXEDNESS IN BULK DENSITY

No. of helices	No. of passes	Particles diameter (inch)	Bulk density (g/cc)	Degree of mixedness $M = 1 - \sigma^2/\sigma_o^2$
12	2	1/8	0.691	0.6
12	14	1	0.691	0.99
12	20	3	0.8	0.994
12	30	1/8	0.705	0.994
2	2	5/32	0.687	0.105
2	6	5/32	0.687	0.319
2	12	5/32	0.69	0.5
2	18	5/32	0.683	0.67
2	24	5/32	0.683	0.737
2	30	5/32	0.683	0.82
12	2	5/32	0.69	0.439
12	6	5/32	0.686	0.695
12	19	5/32	0.69	0.946
12	20	5/32	0.690	0.985

TABLE 4

ESTIMATED PARAMETERS FOR ROSE'S MODEL

No. of helices	Particles diameter (inch)	Efficiency	A (1/pass)
2	1/8	0.809	0.054
5	1/8	0.919	0.1
12	1/8	0.932	0.223
2	5/32	0.831	0.038
5	5/32	0.845	0.065
12	5/32	0.893	0.113
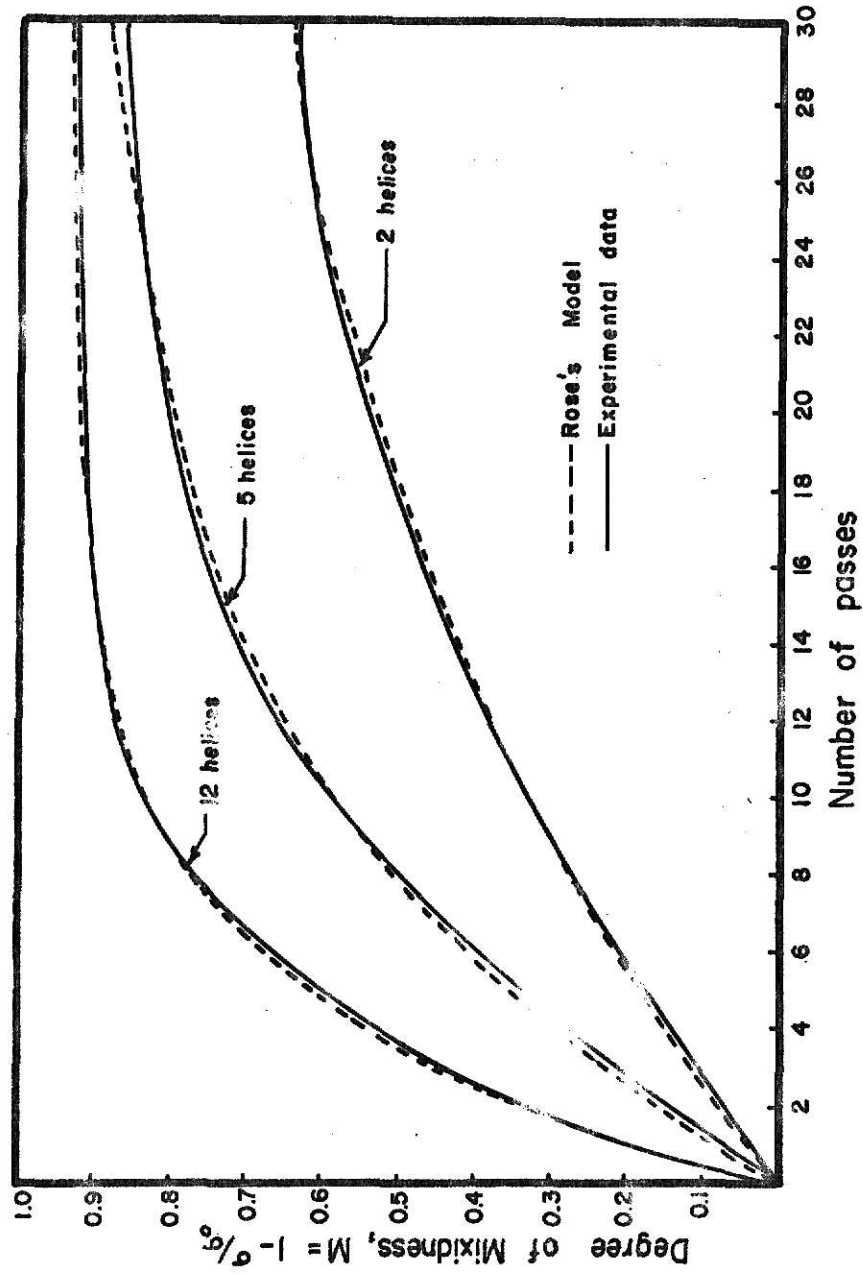
Fig. 7 Comparison between experimental data and Rose's Model Mixers with 1/8" Lucite particles.

and the particle size. However, it appears that the particle diameter has a stronger influence on the efficiency than the number of helices.

Inspection of equation (2) for the case of N approaching infinity reveals that the limit of the degree of mixedness must be equal to η. Rose has claimed that the efficiency depends only on the mixer and not on the characteristics of the mixture, but according to the observations of the present study it depends on the geometry of the mixer, the diameter of the particles, and the definition used for the degree of mixedness.

6.8 SEMI-EMPIRICAL KINETIC MODEL

Weidanz's model (1960) was not considered because it reduces to the Semi-Empirical Kinetic model in the case of identical particles. For particles having identical physical properties, the Semi-Empirical model of this work becomes:

$$M = 1 - e^{-K_1 N} \qquad (3)$$

where M is given by the definition based on the variance [equation (5) in Chapter 5] and K_1 is a constant which depends upon the physical characteristics of the materials being mixed and the geometry of the mixer.

Using experimental data for 1/8" and 5/32" particles, values for K_1 were determined with the aid of Bard's technique (1967) for nonlinear parameter estimation. The results are presented in Table 5. A comparison between the experimental data and the model is presented in Fig. 8. The agreement is fairly good over the range of experimental data. Sums of square of residuals were respectively, 9.3×10^{-3}, 7.0×10^{-4}, and 8.8×10^{-3} for the mixers of 2, 5, and 12 helices with 1/8" particles. Similar results were obtained for 5/32" particles and the same mixers.

TABLE 5

ESTIMATED VALUES FOR K_1

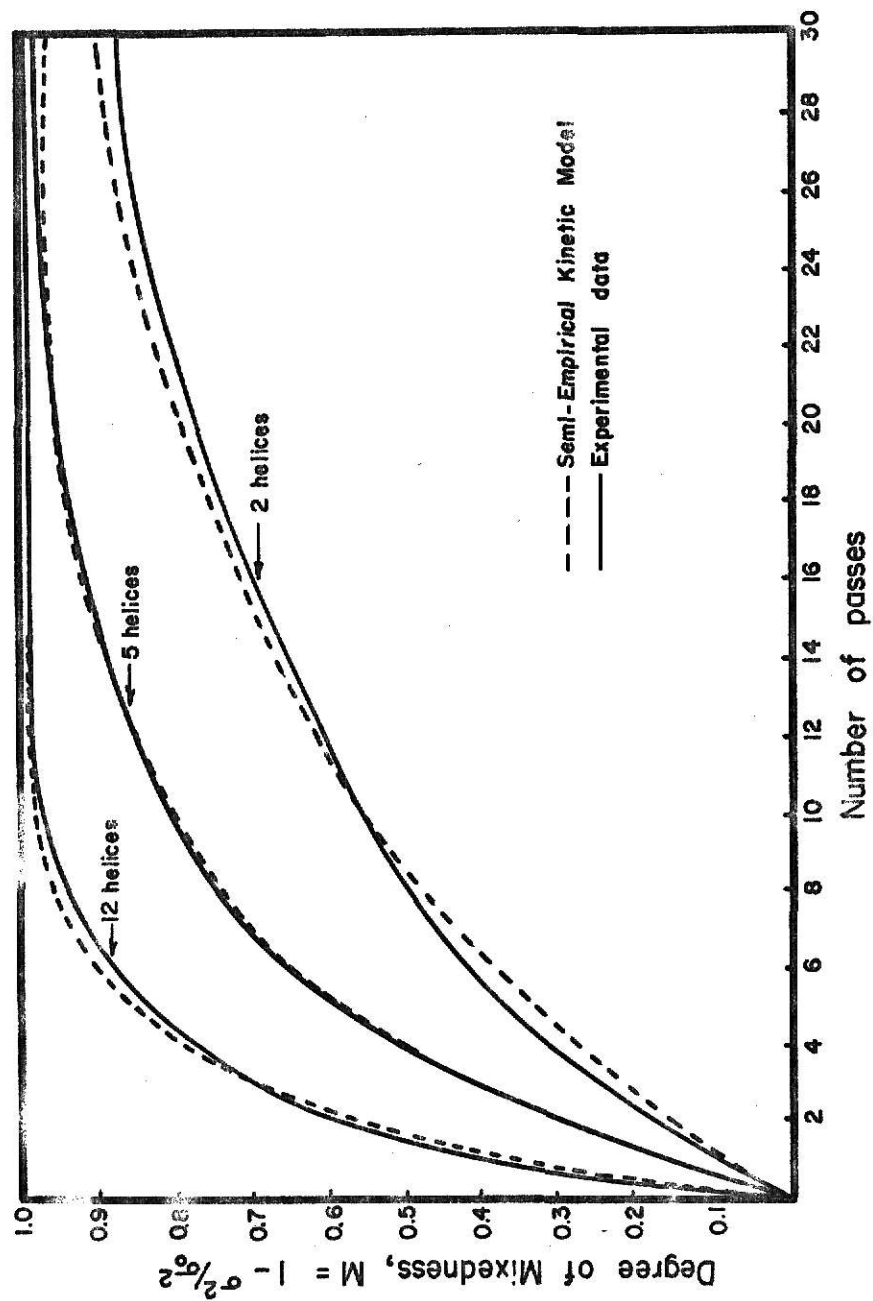| No. of helices	Particles diameter (inch)	K_1 (1/pass)
2	1/8	0.083
5	1/8	0.176
12	1/8	0.4
2	5/32	0.0575
5	5/32	0.106
12	5/32	0.19°

Fig. 8. Comparison between experimental data and the Semi-Empirical Kinetic Model for mixers with 1/8" Lucite particles.

Plots of K_1 vs the number of helices resulted in straight lines over the range of experimental data for each particle size. The slope of the line increased as the particle diameter was decreased; this resulted from the fact (Table 5) that the rate of mixing decreased with an increase in the diameter of the particles. Slopes were calculated and the following equations for K_1 as functions of the number of helices were obtained:

1/8" Lucite particles $\qquad K_1 = 3.35 \times 10^{-2} \times$ (number of helices)

5/32" Lucite particles $\qquad K_1 = 1/8 \times 10^{-2} \times$ (number of helices)

For the case of experimental data as functions of the Mixing Action Number, the Semi-Empirical Kinetic model becomes:

$$M = 1 - e^{-K_1 (M.A.N.)} \tag{4}$$

For this case, the experimental data were also well described by the Semi-Empirical model. Values of K_i' were determined and the following results obtained:

for 1/8" $\qquad K_1' = 0.0316$ 1/pass-helice

for 5/32" $\qquad K_1' = 0.0232$ 1/pass-helice

These results illustrate again that the mixing rate increases as the particle size decreases.

6.9 CONCLUSIONS

1) Comparison of Rose's model and the new model of this study (on the basis of the sums of squares of residuals) indicates that both are equally applicable to the case of mixing particles with identical properties. The major difference in the models is that the new model contains one adjustable parameter, whereas Rose's model contains two.

2) Examination of the data by several methods shows that the rate of mixing increases as the number of helices in the mixer is increased. This result is intuitively expected.

3) Examination of the data by several methods shows that the rate of mixing decreases as the particle size increases.

4) The mixing parameter K_1 of the Semi-Empirical model is a linear function of the number of helices in the mixer for a given particle system.

5) A plot of M vs M.A.N. provides a correlation of data (for a given particle system) obtained with mixers containing different numbers of helices. This result indicates that end effects in the mixer are small.

6) A semilog plot of the standard deviation versus the number of passes results in a linear region from which a coefficient of mixing velocity can be extracted.

7) Percentage of expansion vs void fraction gave straight lines with the same slope, implying that the percentage of expansion depends only on the initial void fraction. Bulk density changes with the degree of mixedness were found to be negligible.

NOTATION

A   Constant of mixing in Rose's model

B   Constant of demixing in Rose's model

D   Diameter, inch

E   Constant

K_1 Constant of mixing in the Semi-Empirical Kinetic model

K_2 Constant

K_3 Constant

M   Degree of mixedness

N   Number of passes

Q   Constant

V   Volume of the slug of particles after N passes

V_o Initial volume of the slug of particles

V_p Volume of the particles

GREEK LETTERS

σ Standard deviation

σ_o Standard deviation at $t = 0$

σ_∞ Standard deviation at $t \sim \infty$

ϵ Ratio of the volume of the voids to the volume of the particles

ρ Density, g/cc

ϕ Coefficient of mixing velocity

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CHAPTER 7

MIXING OF PARTICLES OF DIFFERENT SIZE AND IDENTICAL DENSITY

7.1 INTRODUCTION

The purpose of this chapter is to present results of the investigation of axial mixing in motionless mixers for particles having identical physical properties, with the exception of size. The study was concerned with the influence of the size differences, number of helices, and number of passes on the rate of mixing. The adequacy of existing models for describing axial mixing and segregation in a motionless mixer was examined, as well as the new model proposed in this work.

7.2 CONCENTRATION PROFILES

Typical concentration profiles for a system in which the particles differ only in size are shown in Fig. 1. They illustrate how the axial concentration of the key particles (small particles in this case) changed as a function of the number of passes. Initially, only mixing was noted, but as the number of passes was increased, segregation pockets started to appear at each end of the slug of particles (as a result of size differences). However, the net effect (initially) was one of increasing the overall degree of mixedness. Further increase in the number of passes produced a continued movement of small particles to the top of the slug of particles, indicating that segregation predominated.

It was also observed that the concentration profiles changed more rapidly as the number of helices in the mixer was increased. In the case of
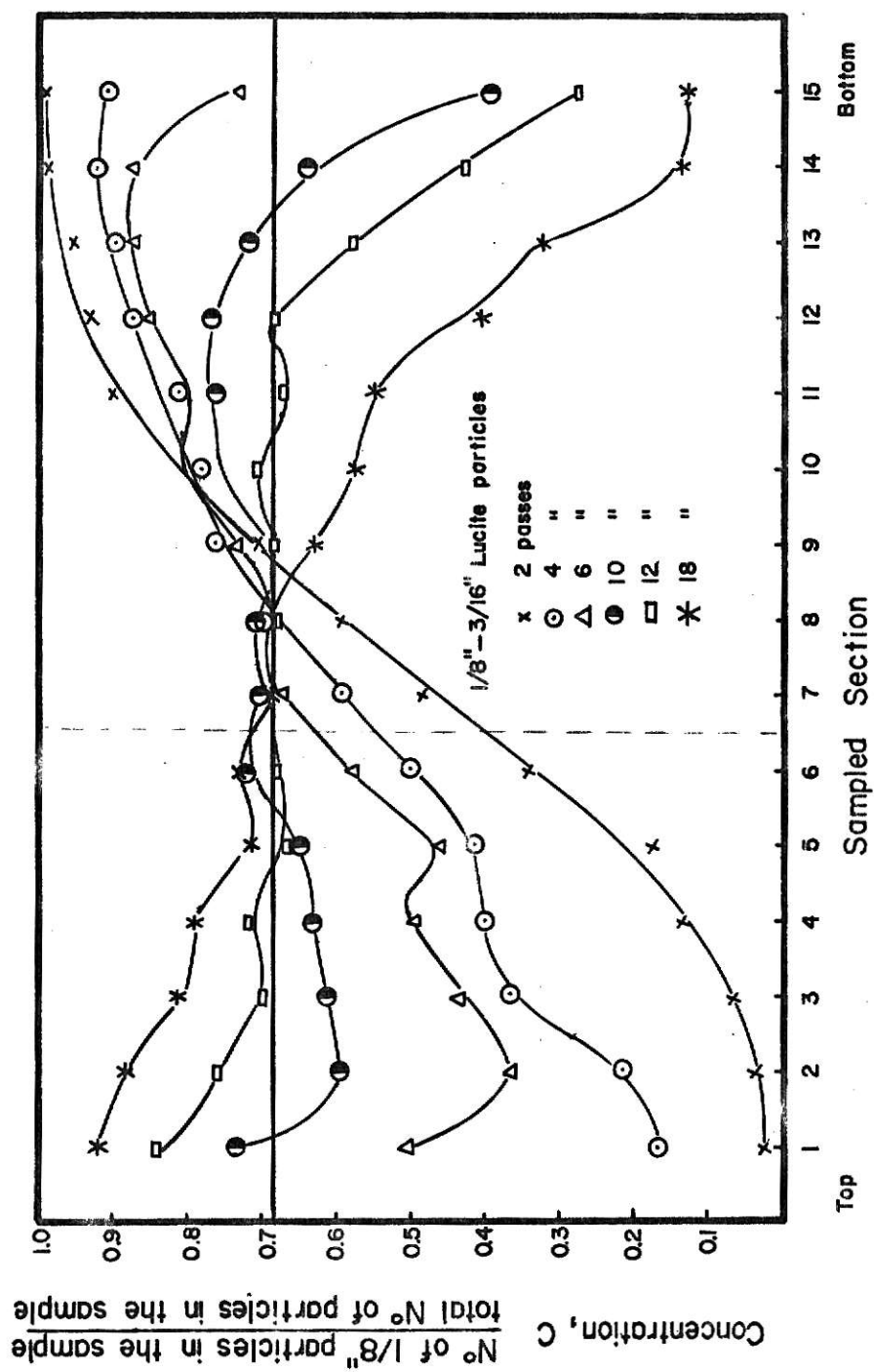
Fig. I. Concentration profiles for an 8 helices mixer with Lucite particles 1/8 and 3/16 inches in diameter.

identical particles, a random mixture was obtained after the particles had been agitated for a long time. For the system of particles with differences in size, a mixture which was not far removed from being random was obtained, after which further mixing action produced a separation of the particles. If the mixing process was carried out long enough, a near complete separation of the particles would occur with the small particles on the top and the large ones on the bottom.

In the ideal case in which the particles have identical physical properties, the concentration profiles intersect at the point where the initial interfacial surface was located. When the particles present differences in size the crossing point is markedly displaced to the right, as can be observed in Fig. 1. This point is expected (according to the ideal situation) between sections 6 and 7, and it is approximately in section 8.

7.3 STANDARD DEVIATION

The behavior of the standard deviation as a function of the number of passes, number of helices in the mixer, and size differences was also examined. Typically, a plot of $\log_{10}\sigma$ vs N showed a transient period and a linear period. But instead of the steady state period observed with particles of identical physical properties, a region in which the standard deviation increases nonlinearly was noted. The linear portion represents the region where mixing is the dominant effect. The minimum value of σ corresponds to a maximum degree of homogeneity for the system. The nonlinear portion represents the region where segregation is the dominant effect. The experimental observations suggest that the nonlinear portion tends to a limit as the number of passes becomes large.

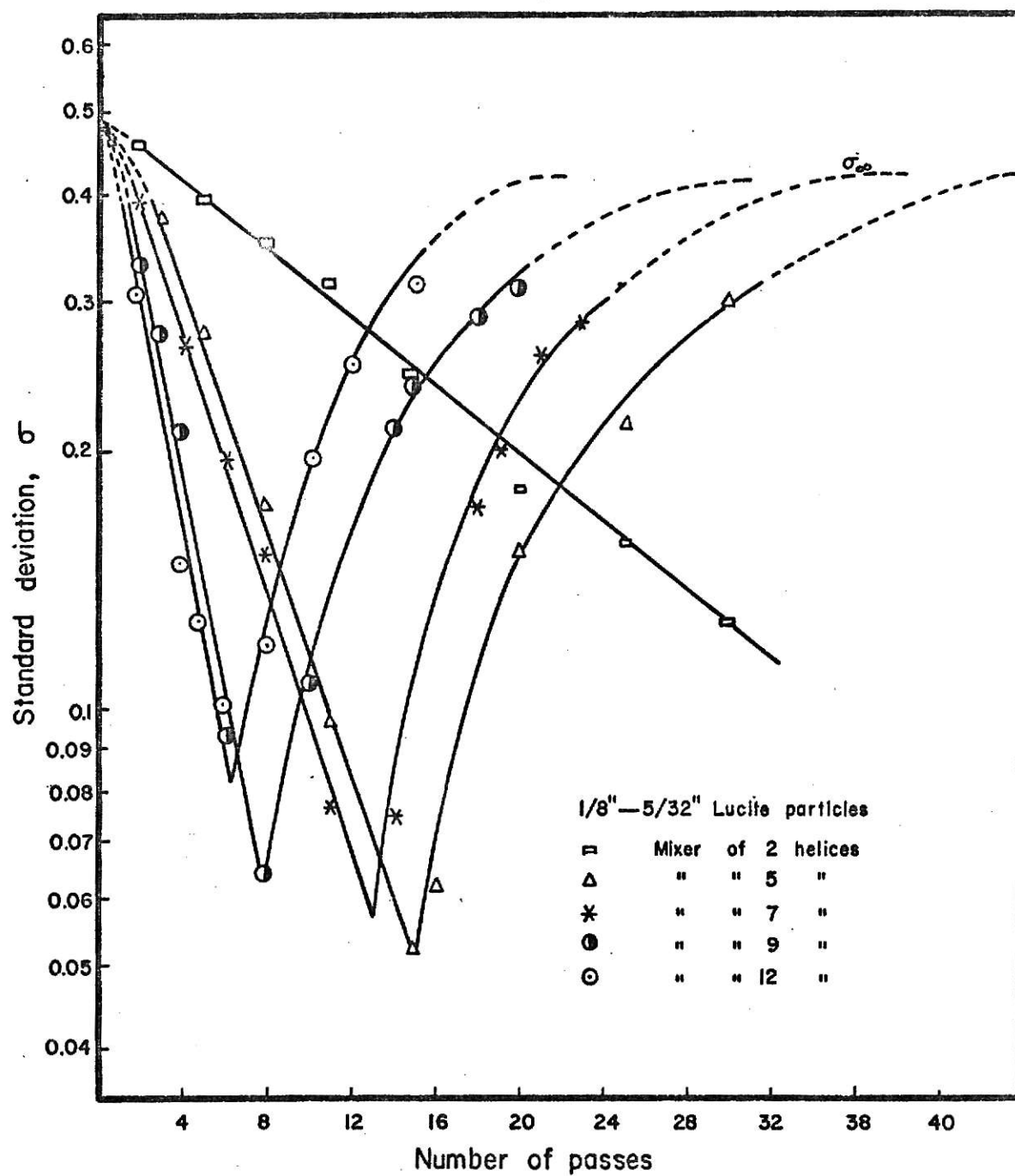Experimental results in the motionless mixer are presented in Figs. 2

Fig. 2. Correlation for σ and number of passes for motionless
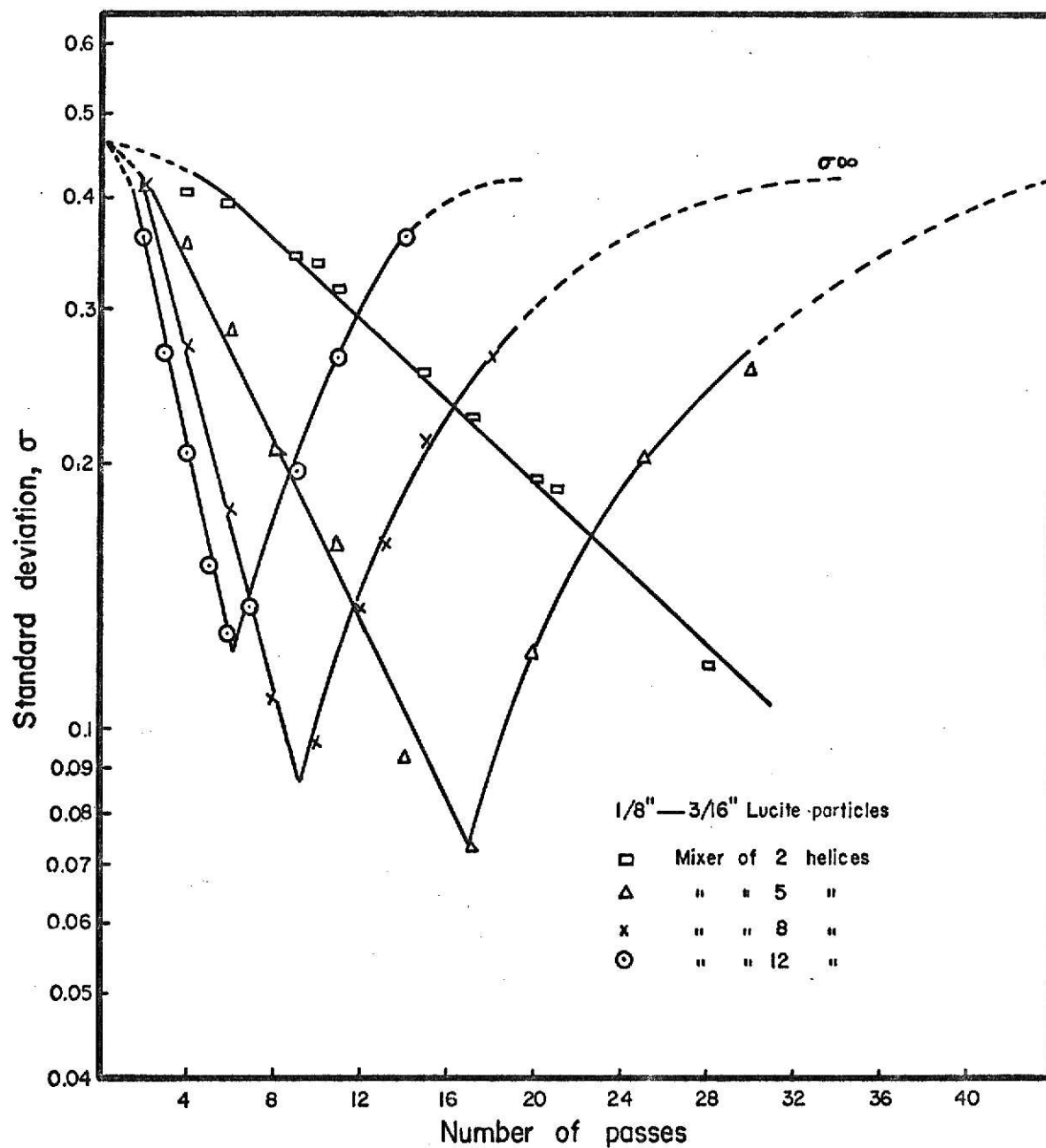mixers with particles of different size and same density.

Fig. 3. Correlation for σ and number of passes for motionless mixers with particles of different size and same density.

and 3 for particle combinations of 1/8"-5/32" and 1/8"-3/16", respectively.
The linear portion was correlated as function of the number of passes by
linearizing equation (1) in Chapter 6 followed by a linear least square
regression with the experimental data. Results for σ_o' and ϕ are shown in
Table 1. The coefficients of mixing velocity show a definite tendency to
increase as the number of helices in the mixer is increased, but decrease on
increasing the size difference between the particles.

Comparing ϕ values for identical particles (presented in Table 1 of
Chapter 6) with those for particles differing in size (Table 1), it is noted
that: 1) when the particles are identical, ϕ increases with decreasing par-
ticle diameter. 2) When the particle sizes are different, it appears that ϕ
increases with an increase in the size difference from zero to some critical
value (ΔD_c), then decreases with further increase in size difference. 3) It
appears that ϕ values (within those examined by experiment) are always
greater for mixing particles with differences in size than for particles with
identical physical properties. In other words, the presence of a size dif-
ference results in a faster rate of mixing.

7.4 DEGREE OF MIXEDNESS

The behavior of plots of the degree of mixedness (defined in terms of
the variance) vs the number of passes was also examined in a motionless mixer.
Experimental results are presented in Figs. 4 and 5. Typically, the plot of
M vs N shows a rapid initial rise in M up to a maximum (which is not too far
removed from a random mixture), followed by a decline in M which levels off
to a non-zero value.

In all cases, the smaller particles (1/8") were initially placed at the
bottom and the coarser (5/32" or 3/16") were layered on top. The disturbance

TABLE 1

CALCULATED VALUES OF THE COEFFICIENTS OF MIXING
VELOCITY FOR PARTICLES DIFFERING IN SIZE

No. of helices	Particles diameter (inch)	$\sigma_o{}'$	ϕ (1/pass)
2	1/8-5/32	0.536	0.054
5	1/8-5/32	0.595	0.163
7	1/8-5/32	0.560	0.181
9	1/8-5/32	0.59	0.277
12	1/8-5/32	0.54	0.290
2	1/8-3/16	0.546	0.053
5	1/8-3/16	0.567	0.123
8	1/8-3/16	0.65	0.223
12	1/8-3/16	0.568	0.26
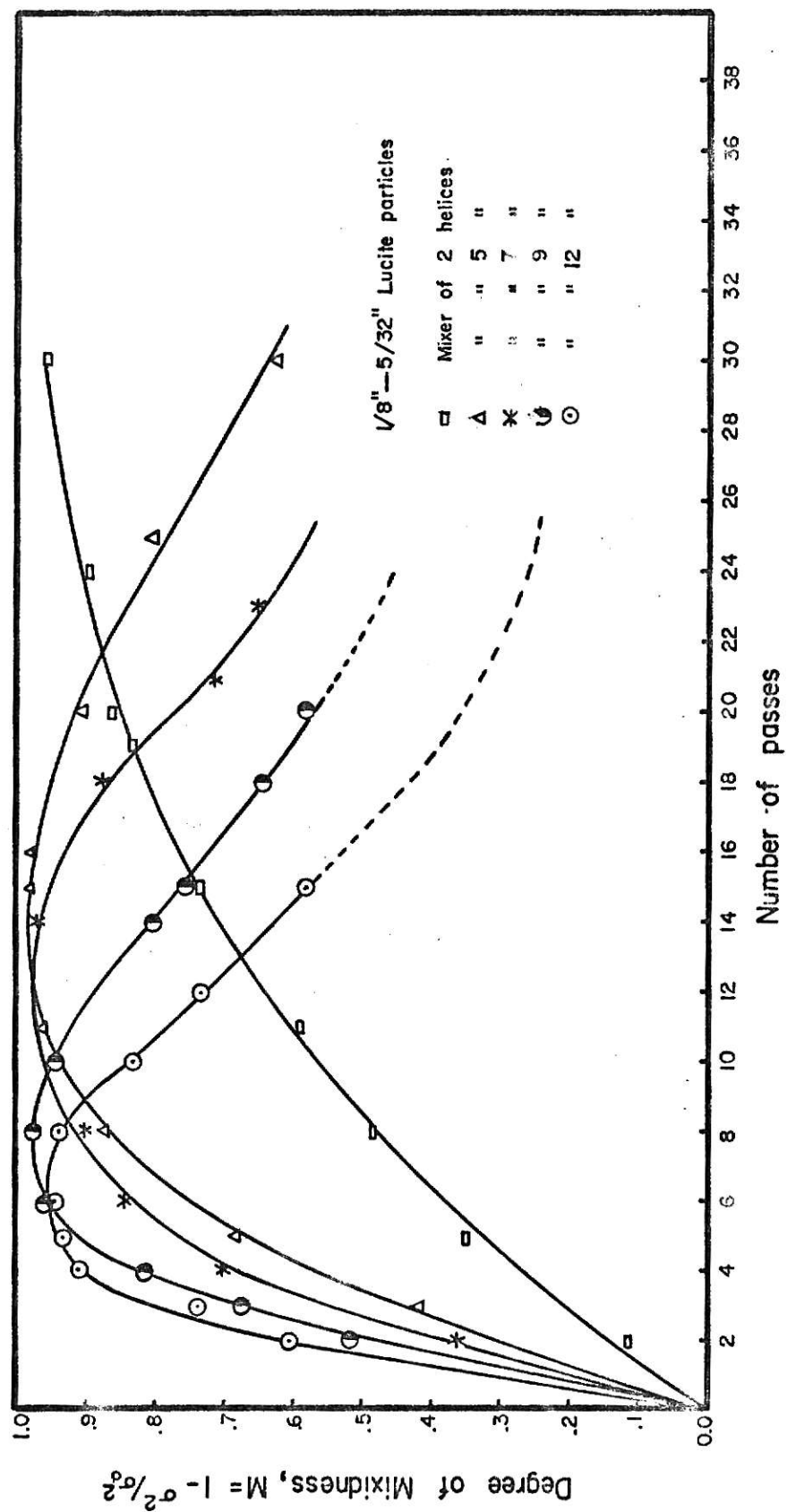
Fig. 4. Rate of Axial mixing for Different Size particles and Same density.
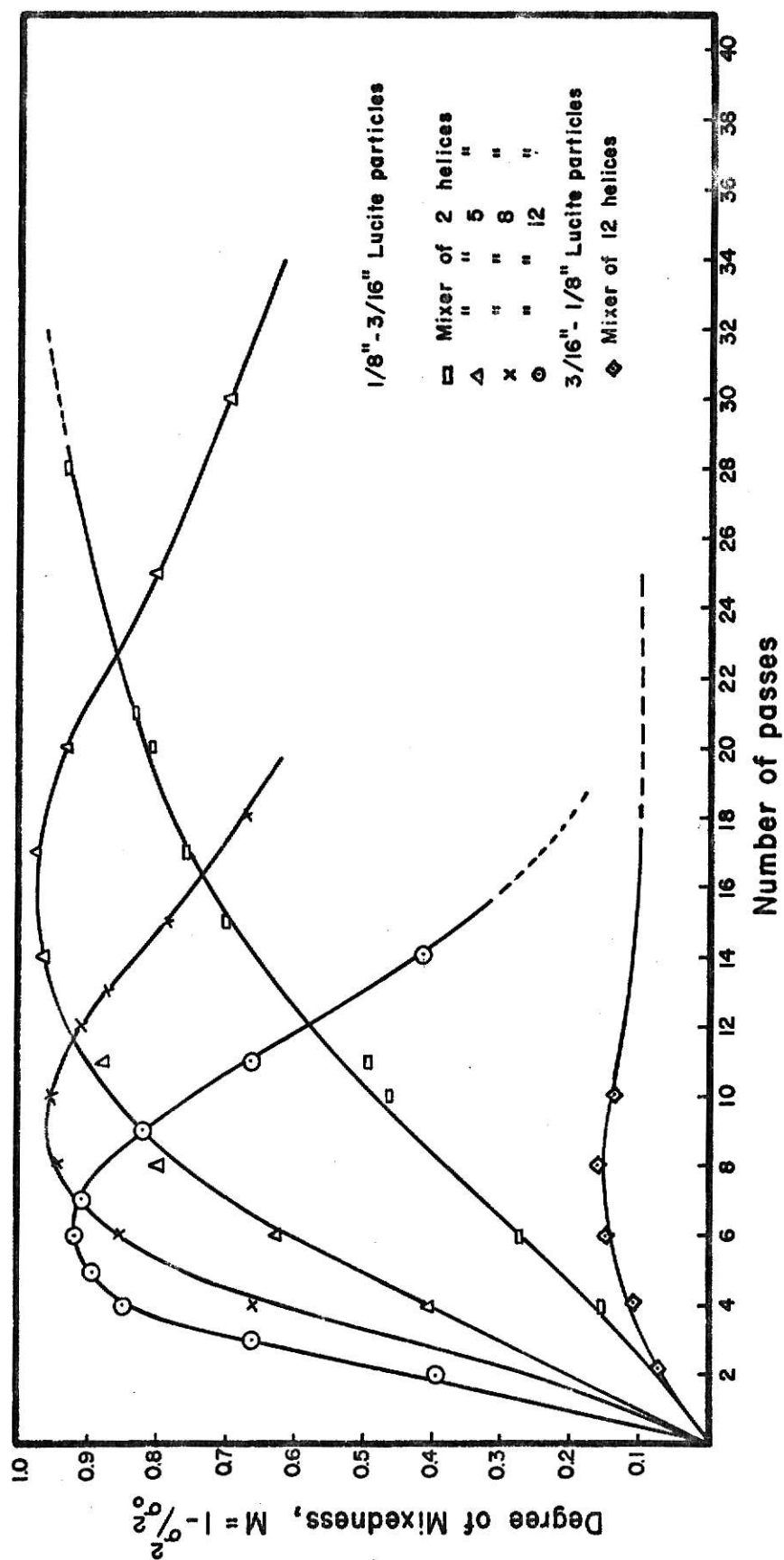
Fig. 5. Rate of axial mixing for different size particles and same density.

produced by passing through the mixer resulted in the smaller particles moving from the bottom to higher positions. As a result of this action, homogeneity improved rapidly during early moments of mixing and only after a maximum degree of mixedness was reached did the segregation effect start to appear. This behavior was accelerated as the number of helices in the mixer increased.

For a small number of helices (1 or 2) the mixing effect was predominant, and only after a large number of passes was the segregation effect observed. Increasing the size difference resulted in lower degrees of maximum mixedness.

When the process was started in the state of type II segregation (larger particles in the bottom and smaller on the top), very small rates of mixing were observed. One experiment starting with this situation was performed for a 12 helices mixer with 1/8" and 3/16" Lucite particles. Maximum degrees of mixedness of the order of 0.1 were obtained. It appears that the equilibrium obtained for this case was the same as that one reached when the process was started in the segregated state of type I (light in bottom, heavy top) and mixing was carried out for a long time, as illustrated in Fig. 5.

7.5 MIXING ACTION NUMBER (M.A.N.)

The data were expressed in terms of M.A.N. as defined previously. Figure 6 illustrates the results for 1/8"-5/32" Lucite particles. A reasonable correlation is observed for the experimental data. For the 1/8"-3/16" system, the correlation is not as good, but it is still satisfactory. In comparison with same size particles, similar correlations are obtained.
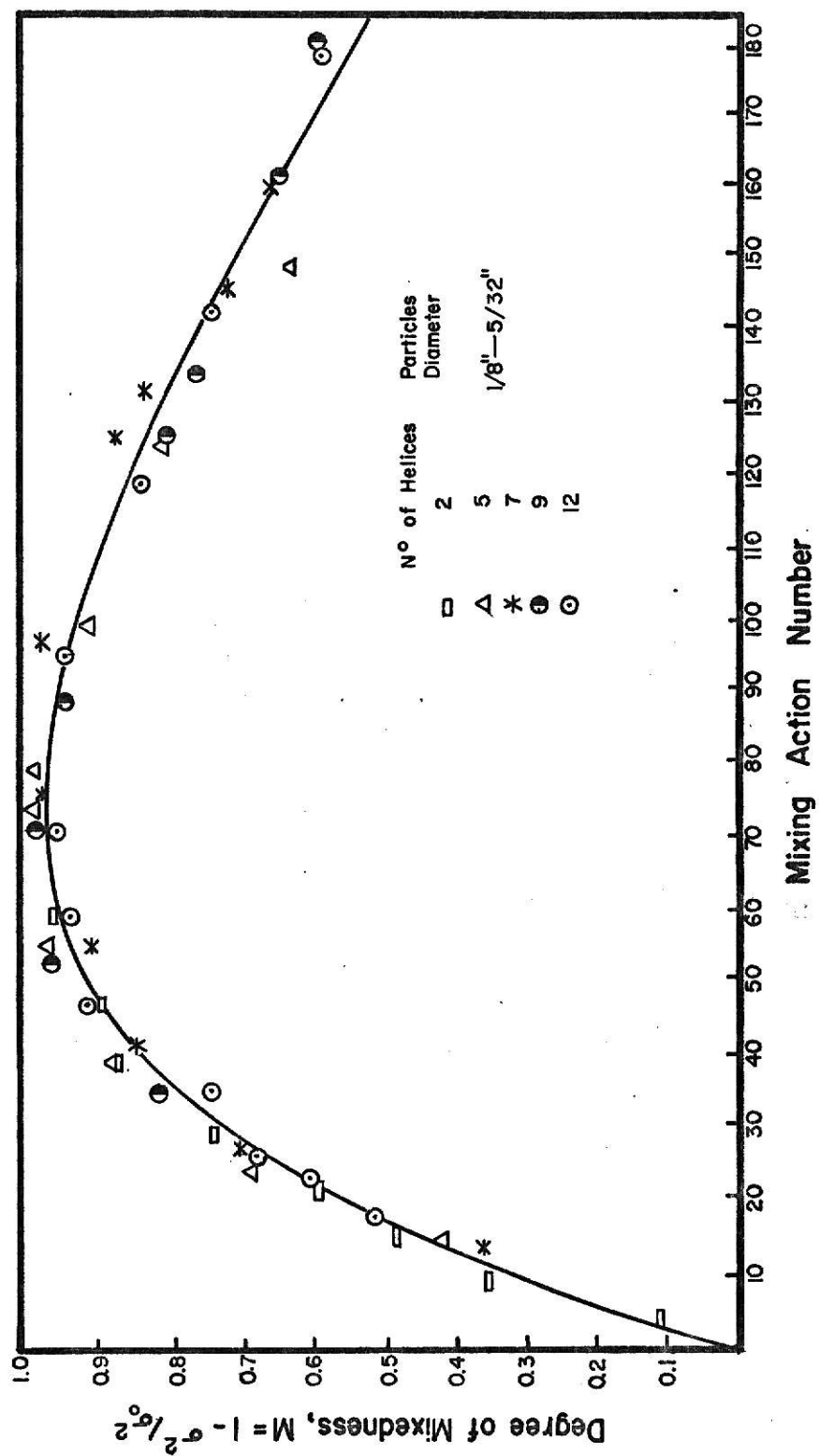
Fig. 6.   Correlation between the degree of mixedness M and M.A.N. for mixers with particles of different size.

7.6 ROSE'S MODEL

For the mixing of particles with differences in physical properties in a motionless mixer Rose's model becomes:

$$M = \eta\{1 - [B/A - (1 + B/A)e^{-AN/2}]^2\} \tag{1}$$

where

$$M = 1 - \sigma/\sigma_o$$

The formulation of this equation was considered in section 3.2.1. The parameters η, B, and A of Rose's model were estimated with the aid of Bard's technique for nonlinear parameter estimation (1967), for each set of experimental M vs N data. Fairly good agreement is obtained between experimental data and the model as is shown in Fig. 7.

However, there is an inconsistency between the estimated parameters and the theory behind the model. It was pointed out (Danckwerts, 1959) that the parameter A should always be greater than the demixing parameter B. The reasoning is as follows: if we consider equation (1) in Chapter 3, and imagine a situation where all the heavy material is at the bottom, then the degree of mixedness M is equal to zero, and according to the definition for the demixing potential, ψ is equal to 1. Under the limiting conditions,

$$\frac{dM}{dt} = A - B \tag{2}$$

and if B > A, the rate of mixing will be negative. In this case the system is already as demixed as possible, but according to equation (2) it would continue to demix.

An alternative explanation can be obtained through a consideration of equation (1) as $t \rightarrow \infty$
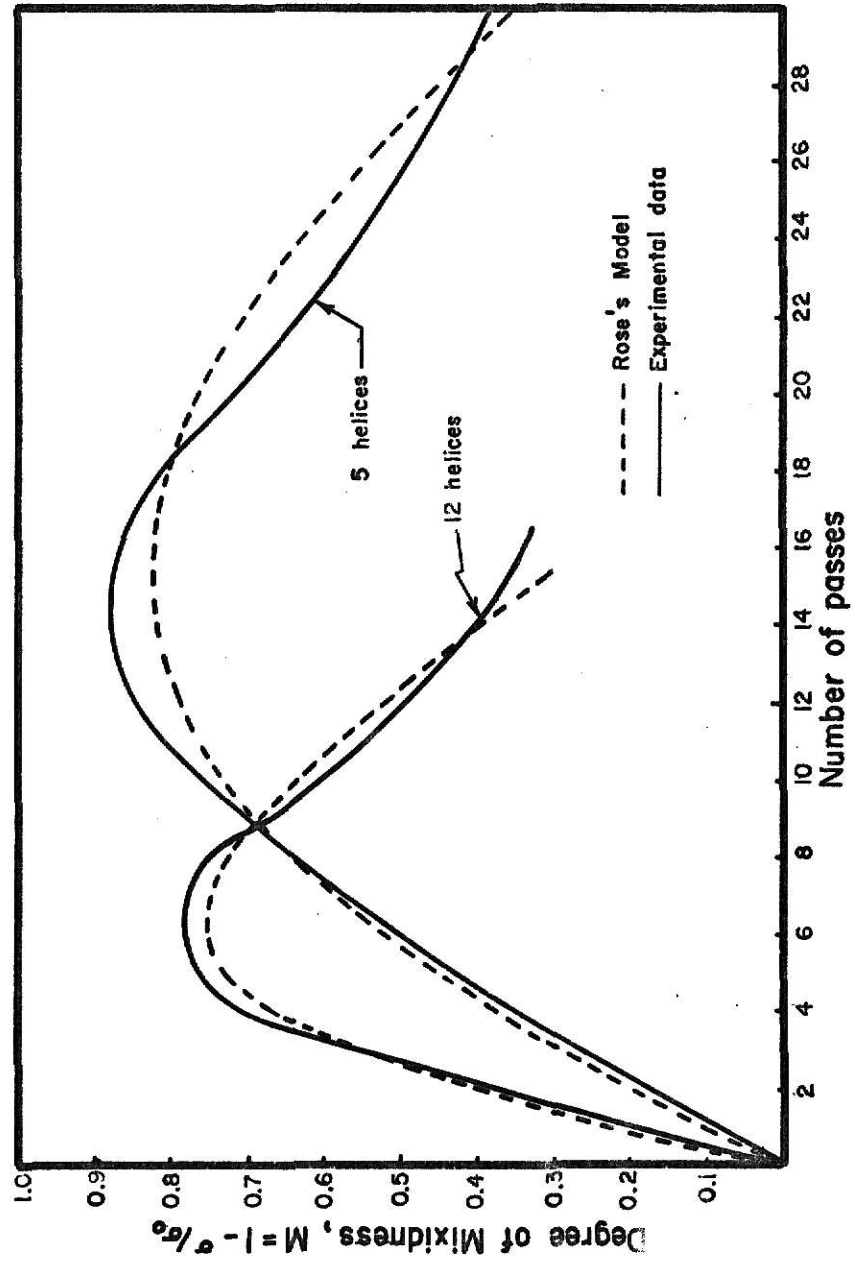
$$M = (1 - B^2/A^2) \tag{3}$$

Fig. 7. Comparison between experimental data and Rose's model.
for mixes with 1/8″—5/32″ Lucite particles.

If B > A the degree of mixedness will be negative, which is inconsistent physically. In all cases for which this model was tested, the values estimated for the parameters A and B showed this inconsistency; in other words, B was always greater than A. When the definition for degree of mixedness in terms of the variance was employed with this model the same inconsistency was obtained. Thus, this model cannot represent the experimental data with the motionless mixer on the basis of physical incompatibility.

7.7 WEIDANZ'S MODEL

For the mixing of particles with differences in physical properties in a motionless mixer, Weidanz's model (1960) becomes:

$$\frac{\sigma}{\sigma_o} = \{e^{-2QN} + \sigma_\infty^2 (1 - e^{-EQN})^2\}^{1/2} \tag{4}$$

The formulation of this equation was considered in section 3.2.2. The parameters E and Q of Weidanz's model were estimated by Bard's technique (1967) for each set of σ vs N data. The value of σ_∞ required in equation (4) was estimated from the plot of $\log \sigma$ vs N as in Fig. 2. A comparison between experimental data and the model is shown in Fig. 8. As can be seen from this figure, the agreement is very poor.

The lack of agreement probably results because the key assumption (i.e., that small volumes interchanged between upper and lower portions of the mixer are constant with time) is not true in this case. In the present case, the constant b (b = 0.25 EQ, a measure of the mixing in the vertical direction) and the constant C (C = 0.5 $\sigma_o\sigma_\infty$ EQ, a measurement of segregation) are not likely to be simple constants as was assumed in the model.
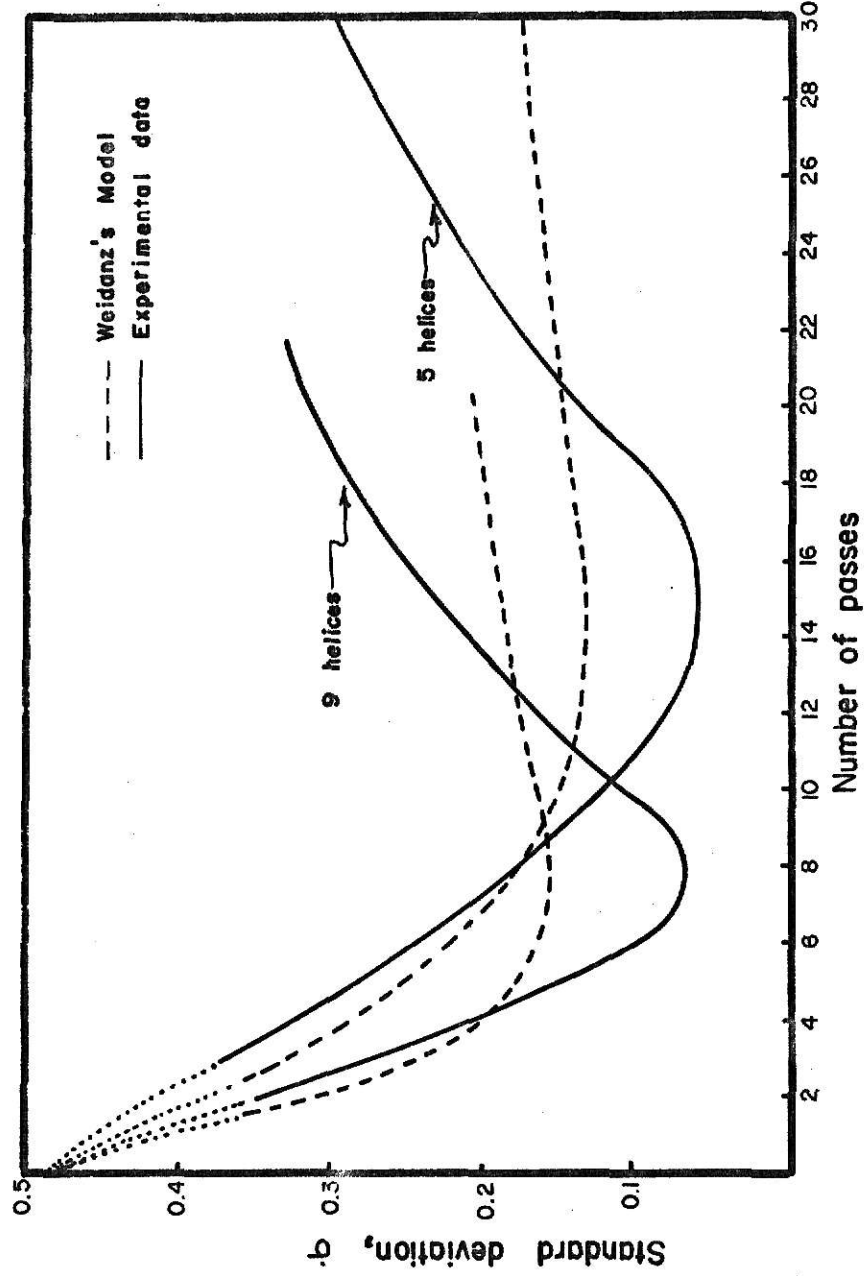
Fig. 8. Comparison between experimental data and Weidanz's Model for the mixing of 1/8"–5/32" Lucite particles.

7.8 SEMI-EMPIRICAL KINETIC MODEL

For the mixing of particles with differences in physical properties in a motionless mixer the Semi-Empirical Kinetic Model becomes:

$$M = 1 - e^{-K_1 N} + \frac{K_2/K_3 \{\exp(K_2 + K_3 \frac{\sigma_\infty^2}{\sigma_o^2}) N - 1\}}{1 + \frac{K_2}{K_3} \frac{\sigma_o^2}{\sigma_\infty^2} \exp(K_2 + K_3 \frac{\sigma_\infty^2}{\sigma_o^2}) N} \qquad (5)$$

where

$$M = 1 - \frac{\sigma^2}{\sigma_o^2}$$

The formulation of this equation was considered in section 3.3.3. The values of σ_∞ were determined by extrapolation of the log σ vs N curves as indicated in Figs. 2 and 3. The constant K_1 can be estimated by plotting $\log_{10} (1 - M)$ vs the number of passes for the initial experimental points up to the maximum degree of mixedness, and calculating the slope of the best straight line through the points. Alternatively K_1 can be estimated by Bard's method (along with the values of K_2 and K_3). Values of K_1 estimated by the two methods agree to within 3 to 10%.

An illustration of the approximation obtained with the model is presented in Fig. 9 for mixers of 7 and 12 helices with Lucite particles 1/8''-5/32'' in diameter. The general result for mixing of 1/8''-5/32'' gave a sum of squares of residuals of the order of 0.011 (0 to 5% difference) between the experimental data and the values predicted by the model. The fitting for 1/8''-3/16'' was not as good; however, it was acceptable. The sum of squares of residuals was of the order of 0.03 (0 to 8% difference).
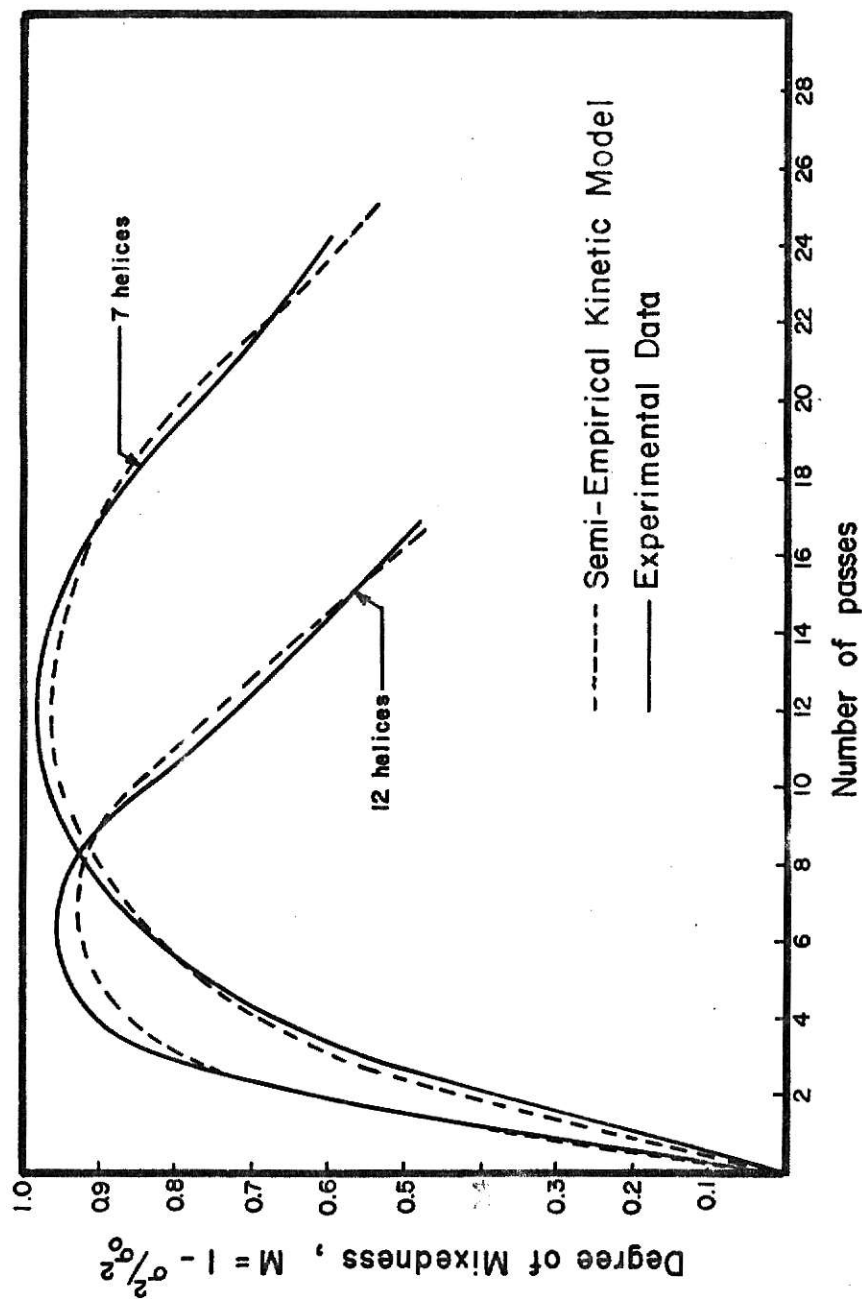
Fig. 9. Approximation given by the Semi-Empirical Kinetic Model for the mixing of 1/8" – 5/32" Lucite particles.

The results of the parameter estimation (Bard's method) for the 1/8"-5/32" and 1/8"-3/16" systems are plotted in Fig. 10 as functions of the number of helices. The values of the mixing constant K_1 show a linear relationship with the number of helices in the mixer and are larger for 1/8"-5/32" than for 1/8"-3/16". The values were correlated by the following equations:

for 1/8"-5/32" $K_1 = 4.34 \times 10^{-2} \times$ (number of helices)

for 1/8"-3/16" $K_1 = 3.25 \times 10^{-2} \times$ (number of helices)

With reference to K_1 it is also noted from Figure 10 that:

1) For identical particles, K_1 increases with decreasing particle diameter.

2) K_1 increases as the size difference is increased from zero to some critical value $(\Delta D)_c$ (for the experimental systems covered, this $(\Delta D)_c$ seems to be the order of 1/32"), but beyond this point K_1 decreases with further increase in size difference.

3) K_1 values (over the experimental range covered) are not always greater for mixing of particles with differences in size than for particles with identical properties. This last point appears to be in contradiction with the conclusion obtained under the coefficient of mixing velocity criteria that the rate of mixing particles with differences in size is always greater than the rate of mixing for identical particles. The reason for this contradiction seems to be a result of the number of parameters estimated for the linear region. In the case of the Semi-Empirical model, only one parameter (the slope K_1) is calculated. In the case of the coefficient of mixing velocity, $\sigma = \sigma_o' e^{-\phi N}$, two parameters ($\sigma_o'$ and ϕ) are estimated. Therefore, variations in the slope, according to the model used, could give this kind of result.

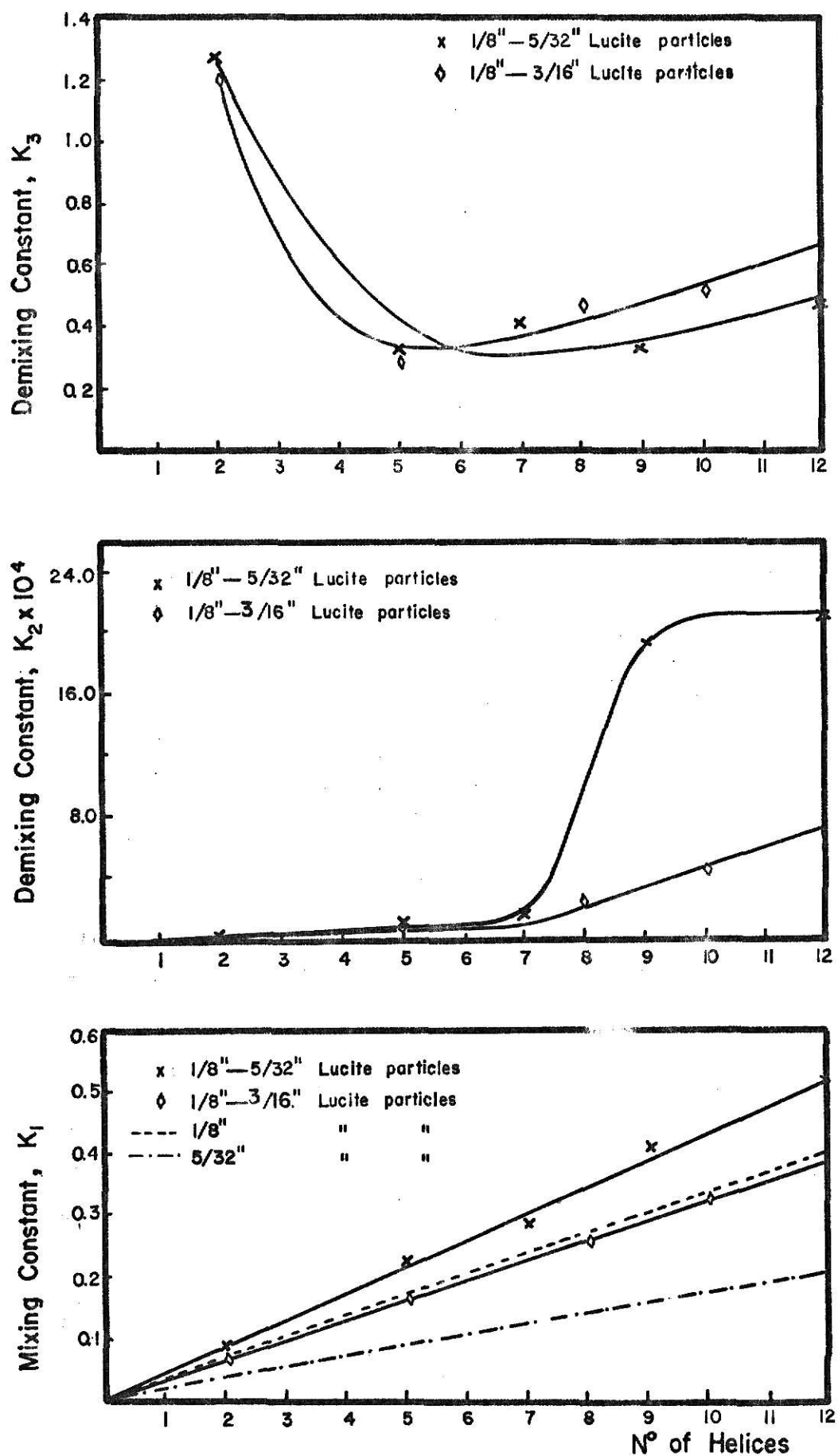Inspection of the plot of K_2 vs number of helices shows that values for

Fig. 10. Estimated parameters as a function of the N° of Helices.

the demixing constant K_2 are smaller for 1/8"-3/16" system than for 1/8"-5/32" system. This indicates that K_2 does not increase in a continuous form with the increase of size differences. The parameter K_2 will go through a maximum value when a critical difference in the diameter of the particles has been reached [for the systems covered, this $(\Delta D)_c$ seems to be of the order of 1/32"] after which it will start decreasing with further increase of the size difference. This seems reasonable if a situation where the differences in size are very large is considered. In this case the segregated state of type II is not likely to occur because the existent voids always allow the finer particles to fall down into the lower layers. It is also observed that when the number of helices in the mixer is less than 7, K_2 shows very small values. After 7 helices a sharp change in K_2 with small increase in the number of helices is observed.

Figure 10 also includes a plot of K_3 versus the number of helices. The K_3 values are observed to be, in some cases, greater for 1/8"-3/16" than for 1/8"-5/32". It is also noted that K_3 starts with a high value for mixers of small number of helices and decreases with the number of helices to a minimum value. After this point it increases (with a very small slope) as the number of helices in the mixer is increased.

The results obtained for K_2 and K_3 seem to be in very good agreement with the mechanisms of segregation proposed in the Semi-Empirical model. According to the mechanism, the mixed state is relatively unstable and tends to break down with a very small rate constant K_2 ($K_1 \gg K_2$ to a more stable state. However, when any small number of particles in the segregated state of type II appear, they act as a catalyst for the breakdown and a second mechanism with a much larger rate constant K_3 takes place, to produce addi-

tional particles in the segregated state of type II. The order of magnitude of the K_2 values suggest that the initial breakdown is the rate controlling step in passing from a mixed state to a segregated state of type II.

7.9 CONCLUSIONS

1) Graphical comparison of Rose's model, Weidanz's model, and the new model of this study with the experimental data indicates that only Rose's and the new model are capable of providing a sufficiently good description of the data. The best fit parameters of Rose's model are, however, physically incompatible. Thus the new model is the only one capable of reasonably describing both mixing and segregation in a motionless mixer. The best fit parameters of the new model appear to be compatible with the postulated mechanism on which it is based.

2) Examination of the data by several methods indicates that the presence of a modest size difference will tend to accelerate the rate of mixing in comparison with the case of identical particles. The effect of increasing size difference on rate appears to follow the path of rising very rapidly initially, passing through a maximum and diminishing to a point of equilibrium.

3) A plot of M vs M.A.N. provides a reasonable correlation of data (for a given particle system) obtained with mixers containing different numbers of helices. This result indicates that end effects in the mixer are small.

4) The mixing parameter K_1 of the new model is a linear function of the number of helices in the mixer for given particle systems.

5) The demixing parameter which determines the rate of segregation is K_2. According to the mechanism proposed, K_2 must have small values. It

represents the formation of a small amount of particles in the segregated state of type II. These particles act as a catalyst for accelerating the formation of the segregated state of type II.

NOTATION

A	Constant of mixing in Rose's model
B	Constant of demixing in Rose's model
D	Diameter, inch
E	Constant
K_1	Constant of mixing in the Semi-Empirical Kinetic model
K_2	Constant
K_3	Constant
M	Degree of mixedness
N	Number of passes
Q	Constant

GREEK LETTERS

σ	Standard deviation
σ_0	Standard deviation at $t = 0$
σ_∞	Standard deviation at $t = \infty$
ϕ	Coefficient of mixing velocity
η	Efficiency
ψ	Demixing potential

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CHAPTER 8

MIXING OF PARTICLES OF DIFFERENT DENSITY AND IDENTICAL SIZE

8.1   INTRODUCTION

The purpose of this chapter is to present the results of the investigation of axial mixing in motionless mixers with particles having identical physical properties with the exception of density. The study was concerned with the influence of the density differences, number of helices and number of passes on the rate and extent of mixing. The adequacy of existing models for describing axial mixing and segregation in a motionless mixer was examined, as well as the new model proposed in this work.

8.2   CONCENTRATION PROFILES

Typical concentration profiles for a system in which the particles differ only in density are shown in Fig. 1. They illustrate how the axial concentration of the key particles (light particles in this case) change as a function of the total number of passes. Initially only mixing was noted, but as the number of passes was increased segregation pockets started to appear at each end of the slug of particles (as a result of the differences in density). However, the net effect (initially) was one of increasing the overall degree of mixedness. Further increase in the number of passes produced a continued movement of light particles to the top of the slug of particles, which indicates that segregation starts to predominate.

It was also observed that the concentration profiles changed more rapidly as the number of helices in the mixer was increased. This is logi-
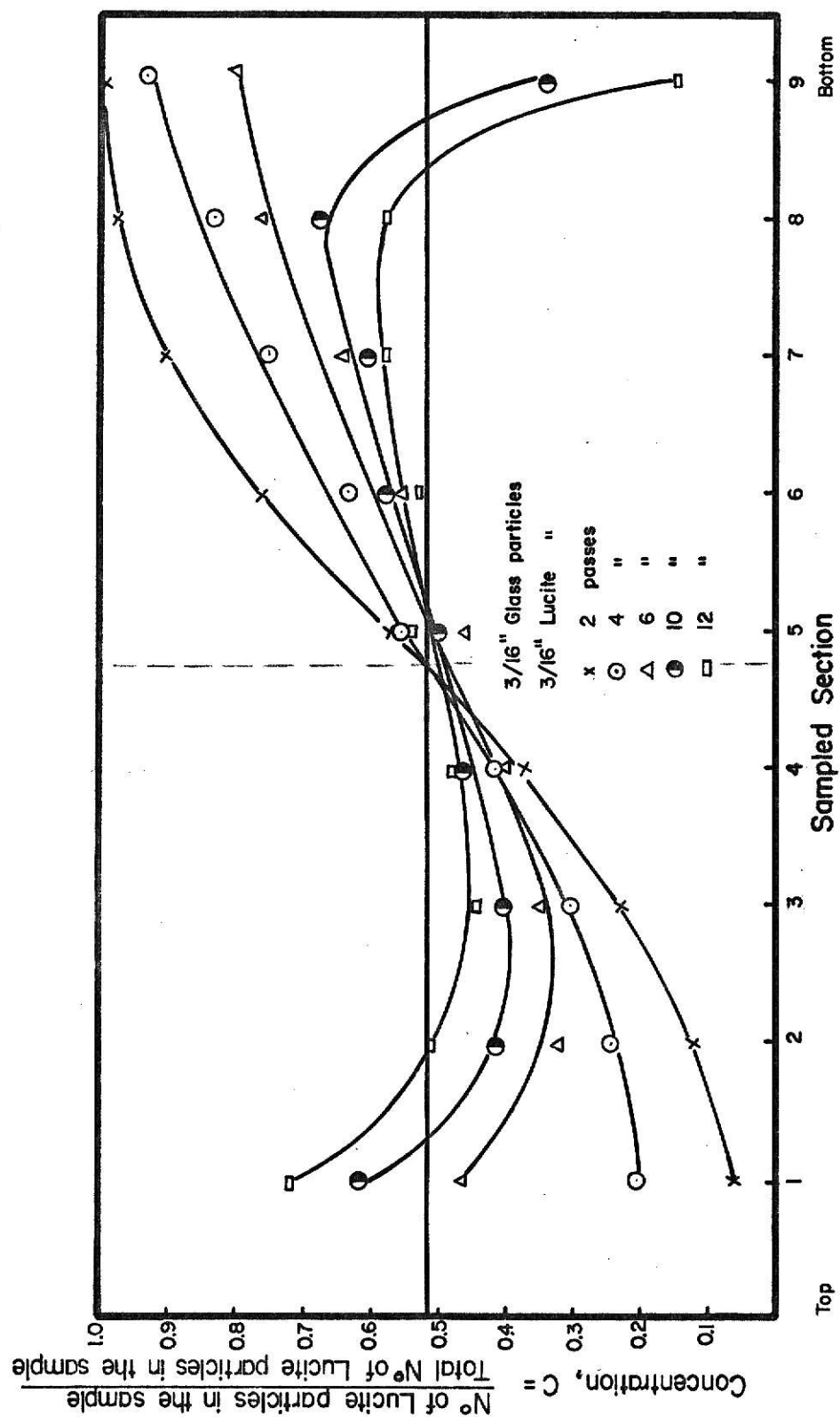
Fig. I. Variation of the concentration profiles with the number of passes for an 8 helices mixers.

cally so, because as the number of helices increased the agitation of the
particles was greater and the homogeneity among samples should change fas-
ter. In the case of identical particles, a random mixture was obtained after
the particles were agitated for a long time. For the system of particles
with differences in density a mixture which was not far removed from being
random was obtained, after which further mixing action produced a separation
of the particles. If the mixing process was carried out long enough a near
complete separation of the particles would occur, with the light particles on
the top and the heavy ones in the bottom.

It was also observed that as the differences in density were increased:
1) more rapid changes in concentration are obtained, and 2) the onset of
segregation was observed at an earlier point (fewer passes).

8.3 STANDARD DEVIATION

The behavior of the standard deviation as a function of the number of
passes, number of helices in the mixer, and density differences was also
examined. A typical plot of $\log_{10} \sigma$ vs N showed a transient period, a linear
period and a region where σ increased with increasing the number of passes.
The linear portion represents the region where mixing is the dominant effect.
The minimum value of σ corresponds to a maximum degree of homogeneity for the
system. The nonlinear portion represents the region where segregation is the
dominant effect. The experimental observations suggest that the nonlinear
portion tends to a limit as the number of passes becomes large.

Experimental results with the motionless mixer are presented in Figs. 2
and 3. From the figures it can be seen that the shapes of the curves are
very similar to those obtained for size differences. The linear portion of
the data was described by equation (1) in Chapter 6. Values for σ_o' and ϕ
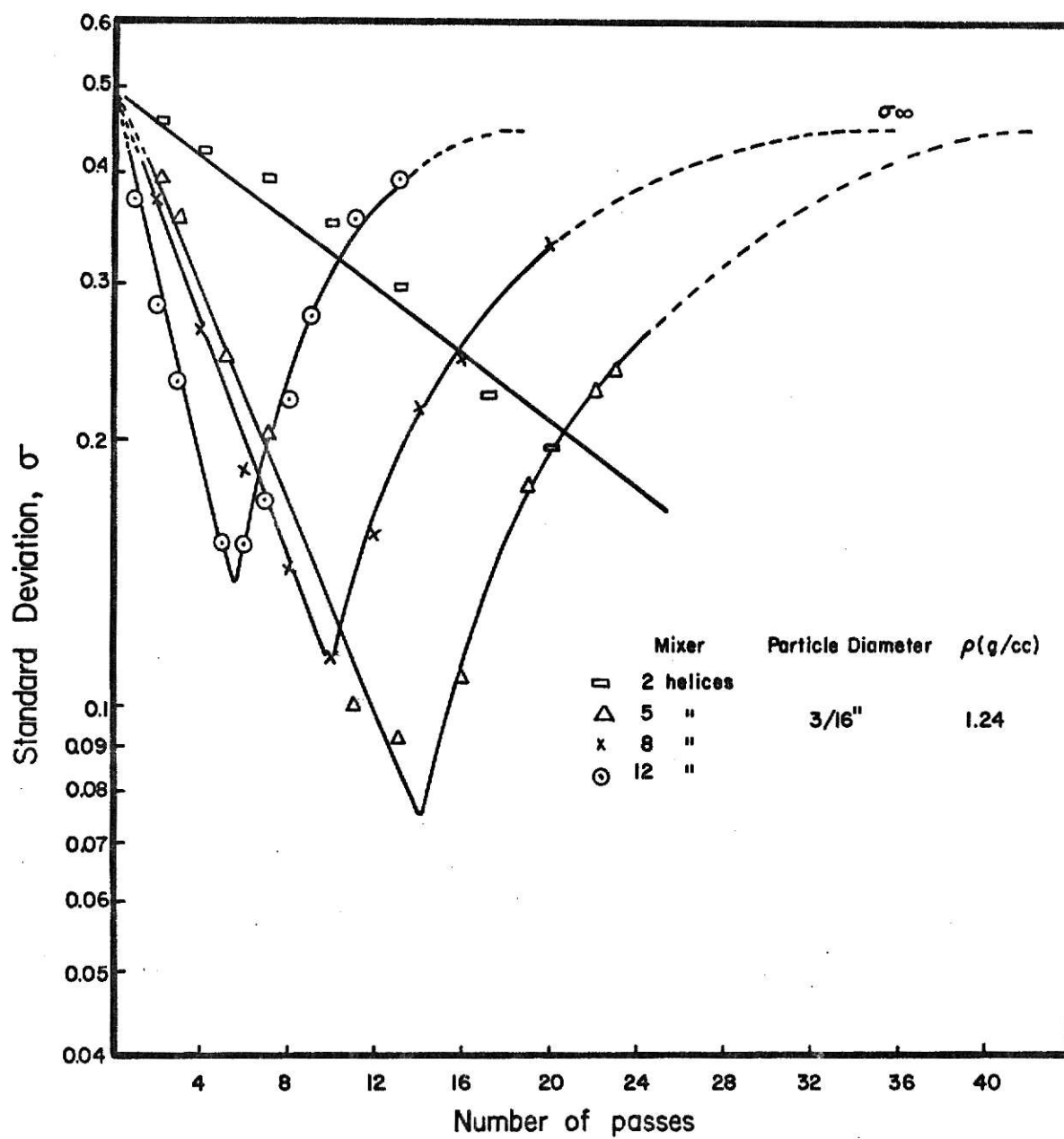
Fig. 2 . Correlation between σ and number of passes for mixers with particles of same size but different density.
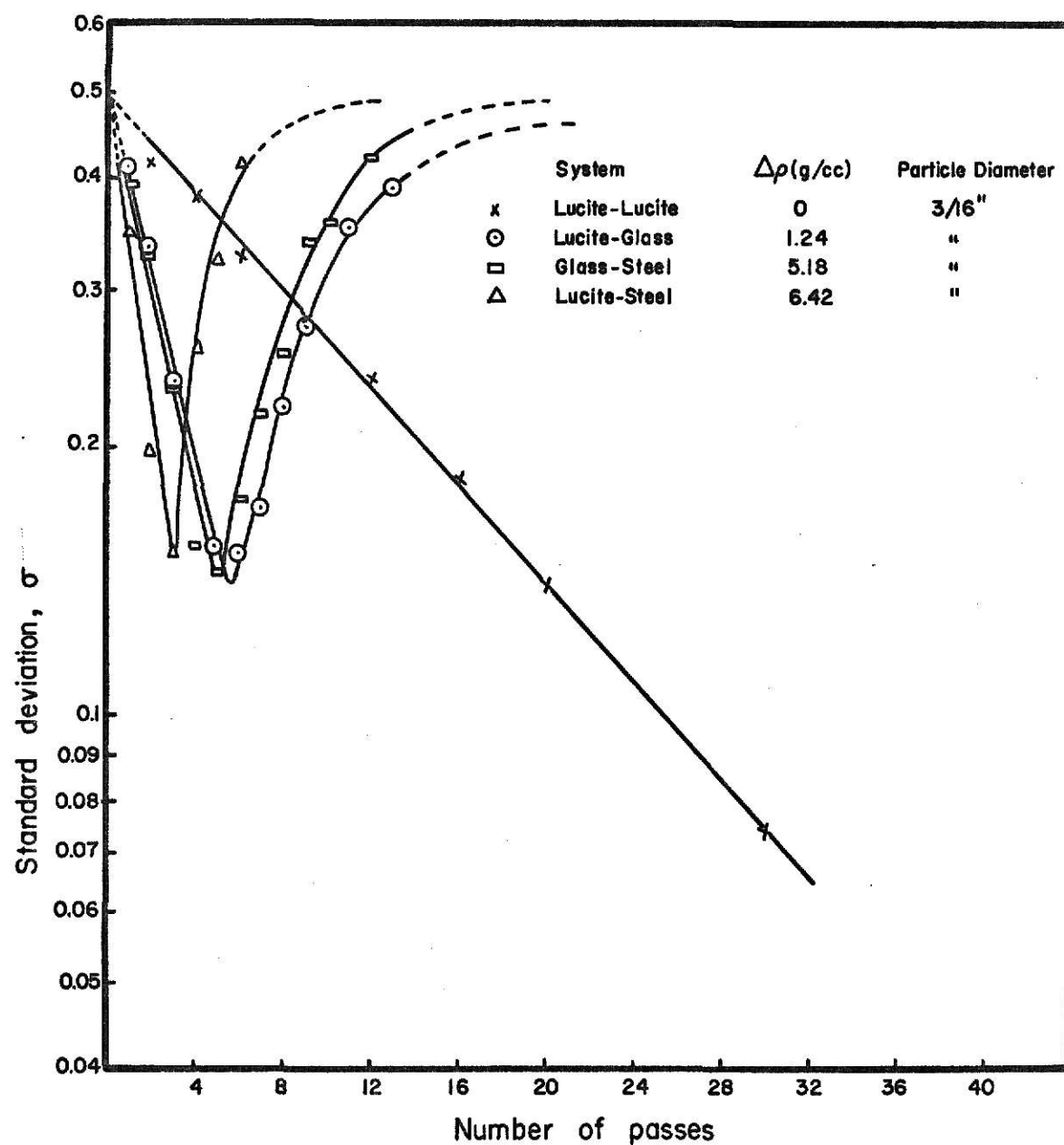
Fig. 3.   Correlation for standard deviation vs number of passes
when mixing particles of same size but different density.

were calculated and are presented in Table 1. For a fixed density difference
($\Delta\rho$ = 1.24 g/cc) the coefficient of mixing velocity increases with the number
of helices, as expected.

As the density difference was increased it was observed that the coef-
ficient of mixing velocity increased. This result indicates that a substan-
tial increase in the mixing action can be obtained over that for no differ-
ence in density. Numerical values are provided in Table 1.

In comparison with size differences, the coefficients of mixing velo-
city did not appear to go through a maximum value beyond which further
increase in density difference would produce a decrease on the rate of mix-
ing. It seems, in this case, the coefficient of mixing velocity will keep
increasing with the increase in density differences. Eventually a point will
be reached at which the pass of heavy particles from top to bottom will be so
fast that the particles will mix and demix in a single pass through the
mixer.

8.4 DEGREE OF MIXEDNESS

The behavior of plots of the degree of mixedness (defined in terms of
the variance) vs the number of passes was also examined in a motionless
mixer. Experimental results are presented in Figs. 4 and 5. Typically, the
plot of M vs N shows a rapid initial rise in M up to a maximum (which is not
too far removed from a random mixture), followed by a decline in M which
levels off to a non-zero value.

In all cases the light particles were initially placed at the bottom
and the heavy were layered on top. The disturbance produced by passing
through the mixer resulted in the movement of light particles to higher posi-
tions. As a result, it was observed that the degree of mixedness increased

TABLE 1

CALCULATED VALUES OF THE COEFFICIENTS OF MIXING
VELOCITY FOR (3/16") PARTICLES DIFFERING IN DENSITY

No. of helices	Density difference (g/cc)	σ_o'	ϕ (1/pass)
2	1.24	0.529	0.048
5	1.24	0.52	0.14
8	1.24	0.483	0.149
12	1.24	0.486	0.21
12	0	0.483	0.062
12	5.18	0.53	0.276
12	6.42	0.50	0.417
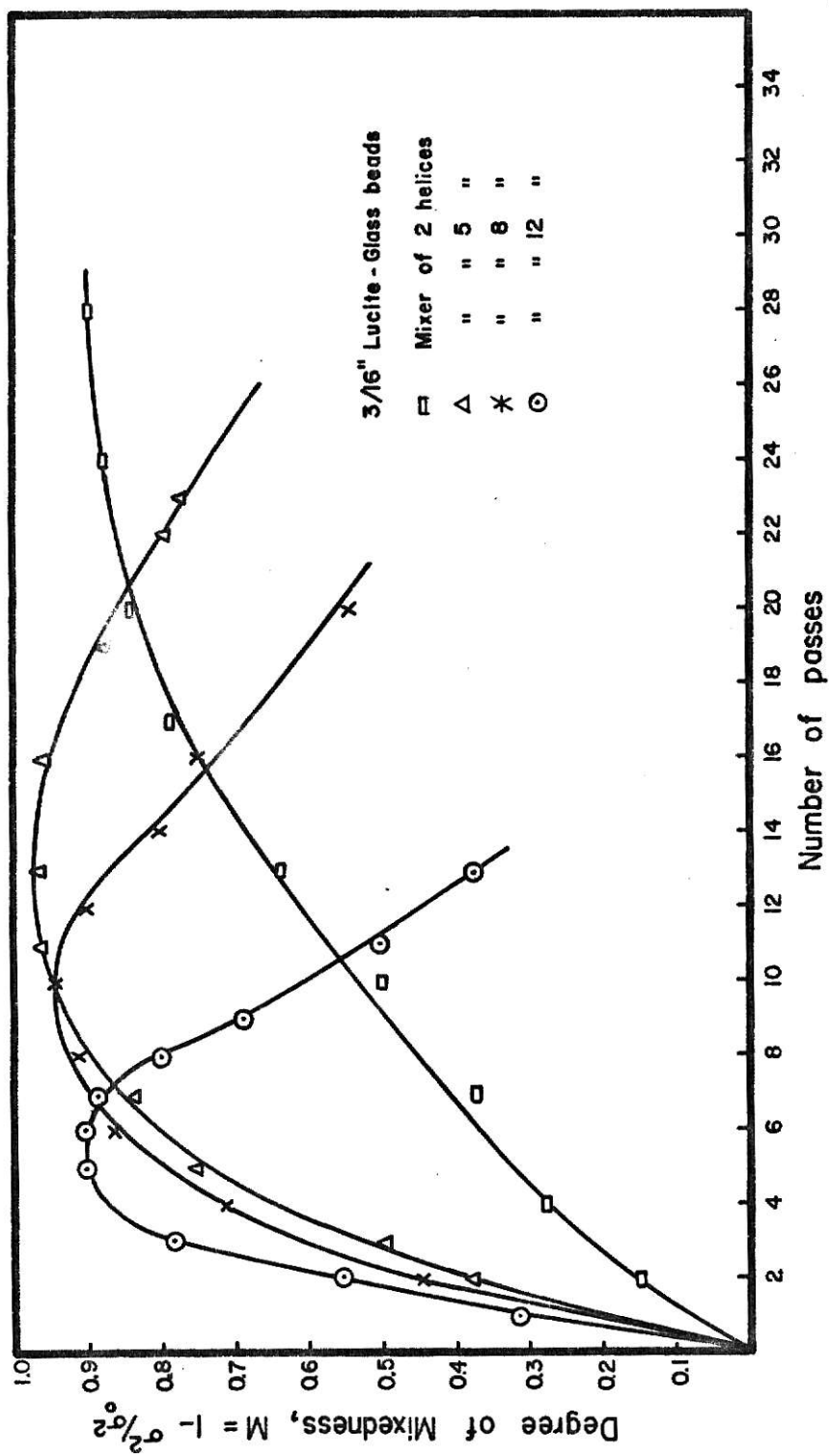
Fig. 4. Rate of axial mixing for particles with the same size but with different density.
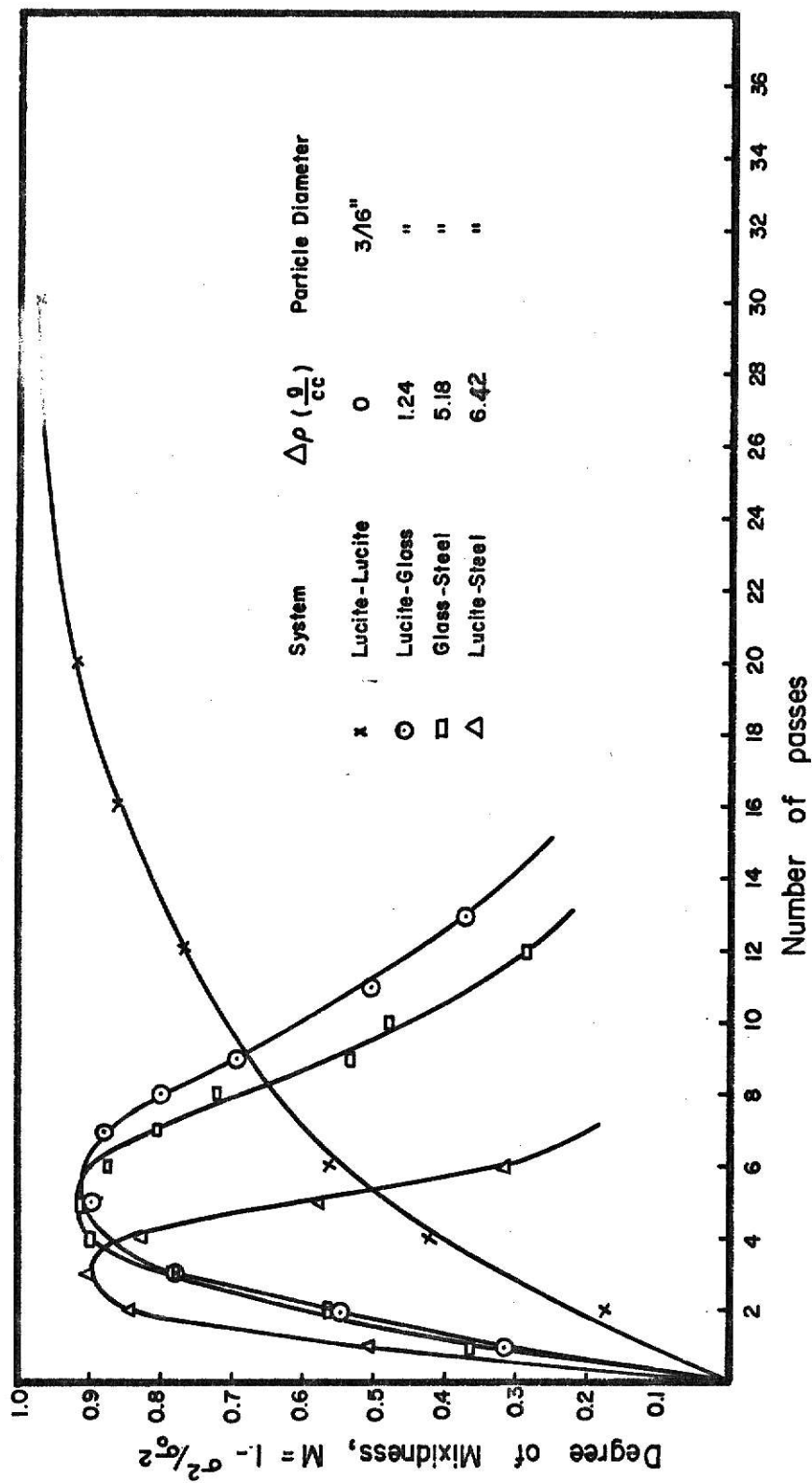
Fig. 5.   Effect of the difference in density in the rate of axial mixing for a 12 helices mixer with particles of the same size.

with the number of passes and only after a maximum degree of mixedness was reached did the segregation effect start to appear. The rate of mixing and segregation increased with the number of helices. The number of passes needed to reach a maximum degree of mixedness was reduced as the number of helices was increased.

For a small number of helices (1 or 2) the mixing effect was predominant, and only after a large number of passes was the segregation effect observed. Increasing the density difference resulted in lower degrees of maximum mixedness. It was also observed that when the density differences were greater than 1.24 g/cc, a complete separation of the material could be obtained with relatively few passes through the mixer.

When the process was started in reverse (i.e., heavy particles in the bottom and light particles on the top) and the density difference was 1.24 g/cc a very small amount of mixing was observed. For $\Delta\rho$ greater than 1.24 g/cc no mixing was achieved at all.

8.5 MIXING ACTION NUMBER (M.A.N.)

The data were expressed in terms of M.A.N. as defined previously. Figure 6 illustrates the results for mixing particles with $\Delta\rho$ = 1.24 g/cc. A reasonable correlation is observed for the experimental points up to the maximum degree of mixedness; however, beyond this the correlation is not as good. Some spread is noted between experimental data for mixers with 8 and 12 helices. In spite of this the general correlation can be considered acceptable. In comparison with size difference, much better correlations were obtained in that case.
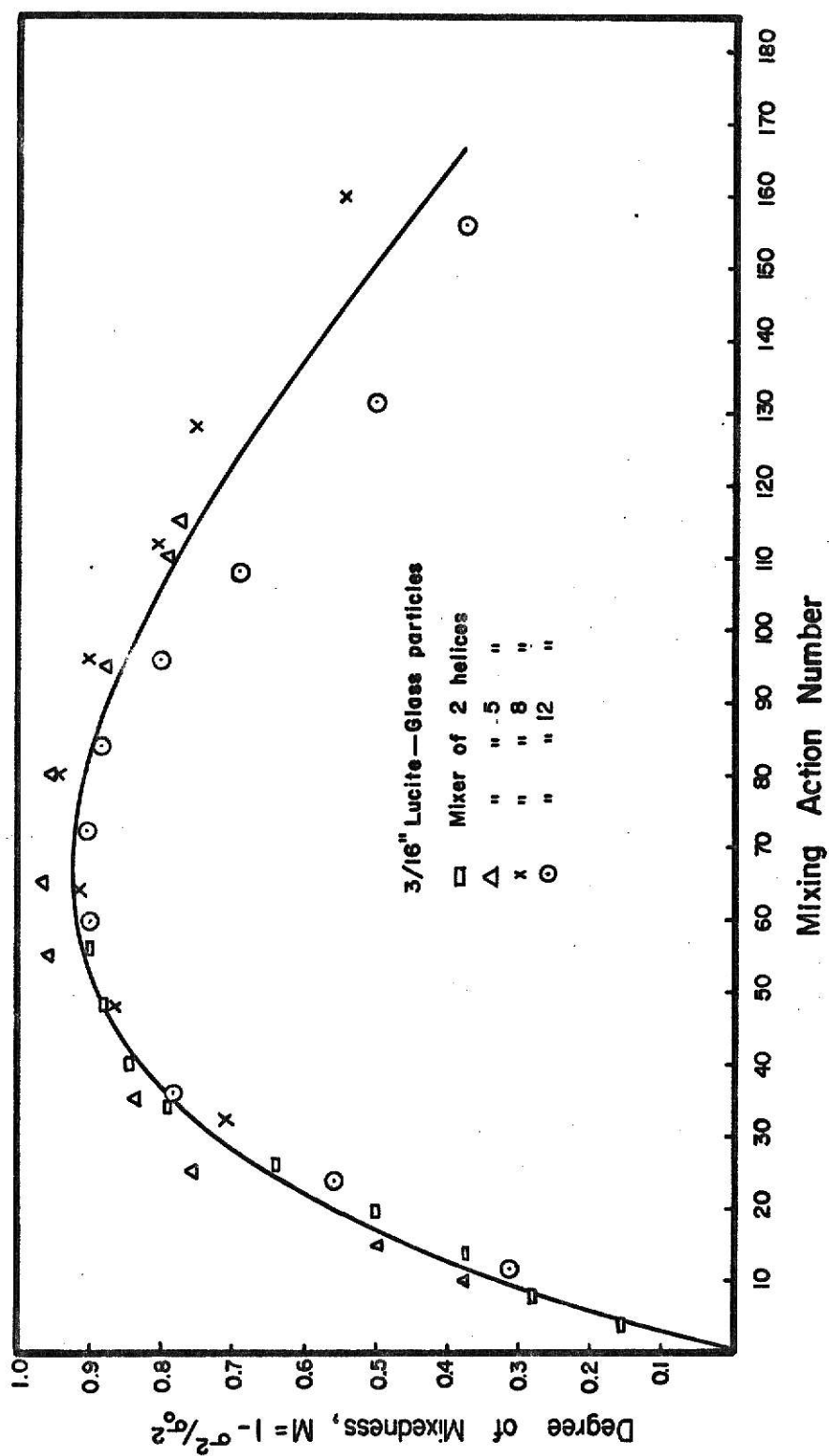
Fig. 6 . Correlation between degree of mixedness M and M.A.N. for mixers with particles of the same size but different density.

8.6 ROSE'S MODEL

For the mixing of particles with differences in physical properties in a motionless mixer Rose's (1959) model becomes:

$$M = \eta\{1 - [B/A - (1 + B/A)e^{-AN/2}]^2\} \tag{1}$$

where

$$M = 1 - \frac{\sigma}{\sigma_o}$$

The formulation of this equation was considered in section 3.2.1. The parameters η, B, and A of Rose's model were estimated with the aid of Bard's technique (1967) for nonlinear parameter estimation, for each set of experimental M vs N data. Fairly good agreement is obtained between the data and the model as can be seen from Fig. 7. However, the physical inconsistency of B > A was observed. This inconsistency was also observed with the system in which particles differed only in size, and is discussed in detail in section 7.6.

8.7 WEIDANZ'S MODEL

For the mixing of particles with differences in physical properties in a motionless mixer Weidanz's (1960) model becomes:

$$\frac{\sigma}{\sigma_o} = \{e^{-2QN} + \sigma_\infty^2 (1 - e^{-EQN})^2\}^{1/2} \tag{2}$$

The formulation of this equation was considered in section 3.2.2. The parameters E and Q of Weidanz's model were estimated by Bard's technique for each set of σ vs N data. The value of σ_∞ required in equation (2) was estimated from the plot of log σ vs N as in Fig. 2. A comparison between experi-
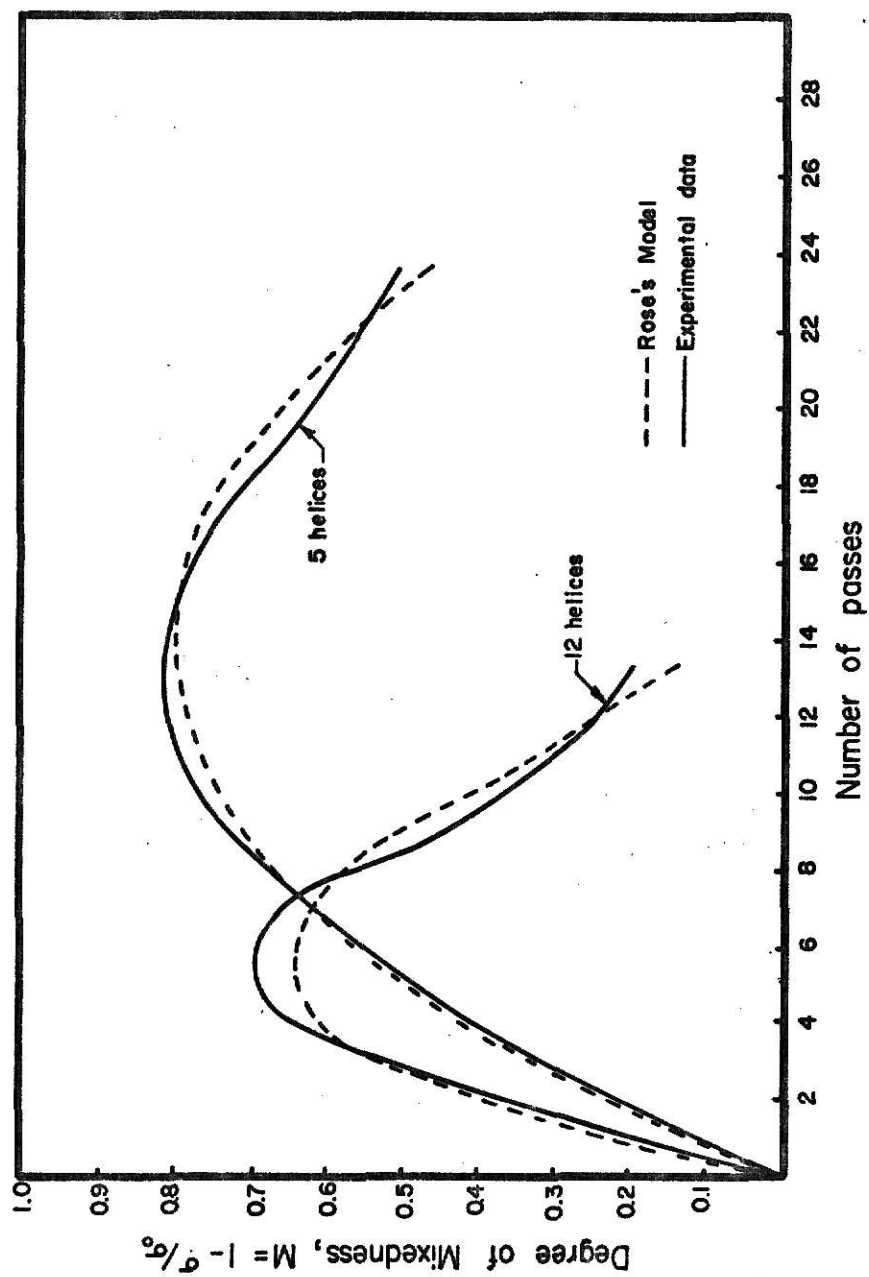
Fig. 7. Comparison between Rose's Model and experimental data for the mixing of 3/16" Lucite-Glass particles.

mental data and the model is shown in Fig. 8. Although the fitting is a
little better than with different size particles, it is still very poor.
The reason for the poor fitting seems to be that the constant b (b = 0.25
EQ, a measure of the mixing in the vertical direction) and the constant C
(C = 0.5 $\sigma_0\sigma_\infty$ EQ, a measure of segregation) are not likely to be simple con-
stants, as was assumed in the model.

8.8 SEMI-EMPIRICAL KINETIC MODEL

For the mixing of particles with differences in physical properties in
a motionless mixer the Semi-Empirical Kinetic model becomes:

$$M = 1 - e^{-K_1 N} + \frac{K_2/K_3 \{\exp(K_2 + K_3 \frac{\sigma_\infty^2}{\sigma_0^2}) N - 1\}}{1 + \frac{K_2}{K_3} \frac{\sigma_0^2}{\sigma_\infty^2} \exp(K_2 + K_3 \frac{\sigma_\infty^2}{\sigma_0^2}) N} \tag{3}$$

where

$$M = 1 - \frac{\sigma^2}{\sigma_0^2}$$

The formulation of this equation was considered in section 3.3.3.
Values of σ_∞ were determined by extrapolation of the log σ vs N curves as
shown in Figs. 2 and 3. With the aid of Bard's method (1967) for nonlinear
parameter estimation, the parameters K_1, K_2, and K_3 of the model were ob-
tained. An illustration of the approximation obtained with the model is
presented in Fig. 9 for the system of Lucite and glass particles. The
general result for this system of particles gave a maximum error of 5% between
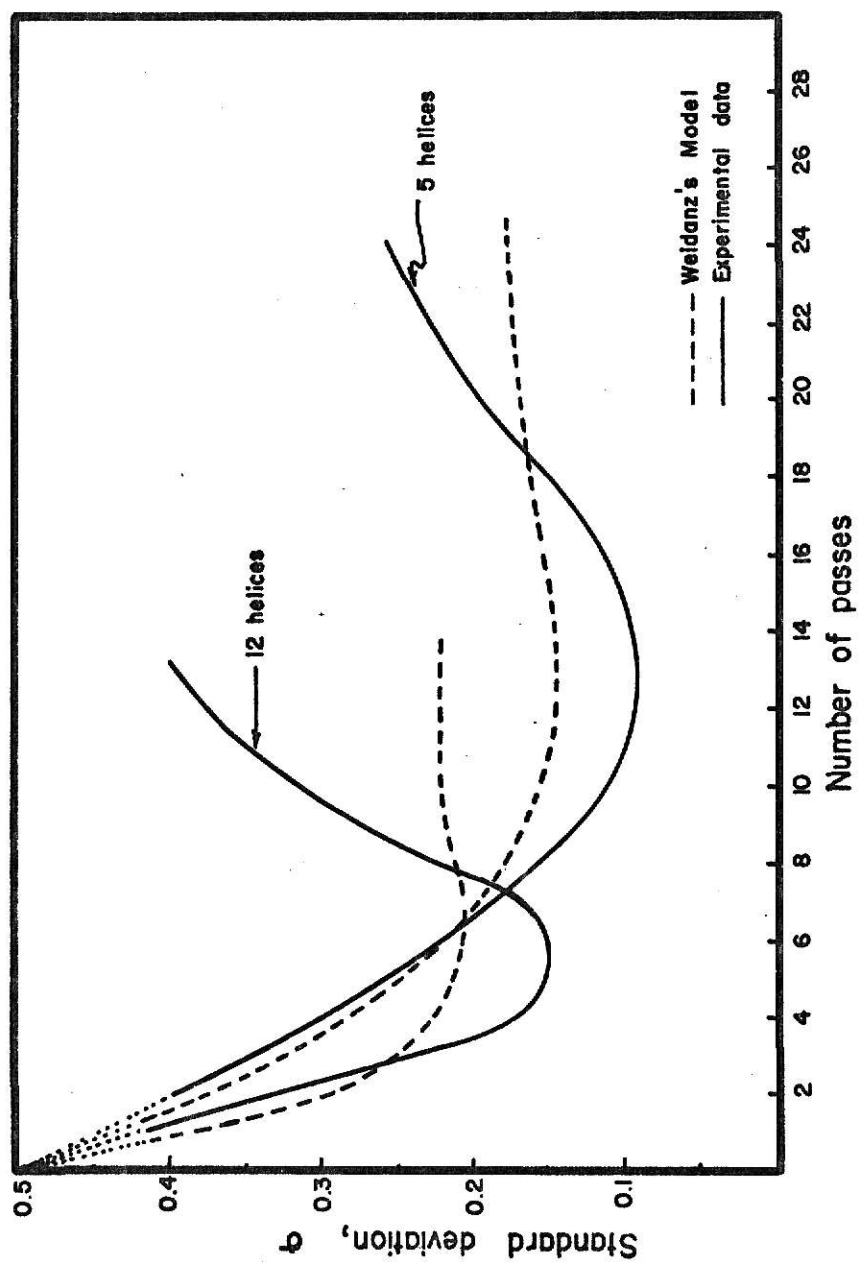experimental data and the model predictions (sums of squares of residuals

Fig. 8. Comparison between Weidanz's Model and experimental data for the mixing of 3/16" Lucite-Glass particles.
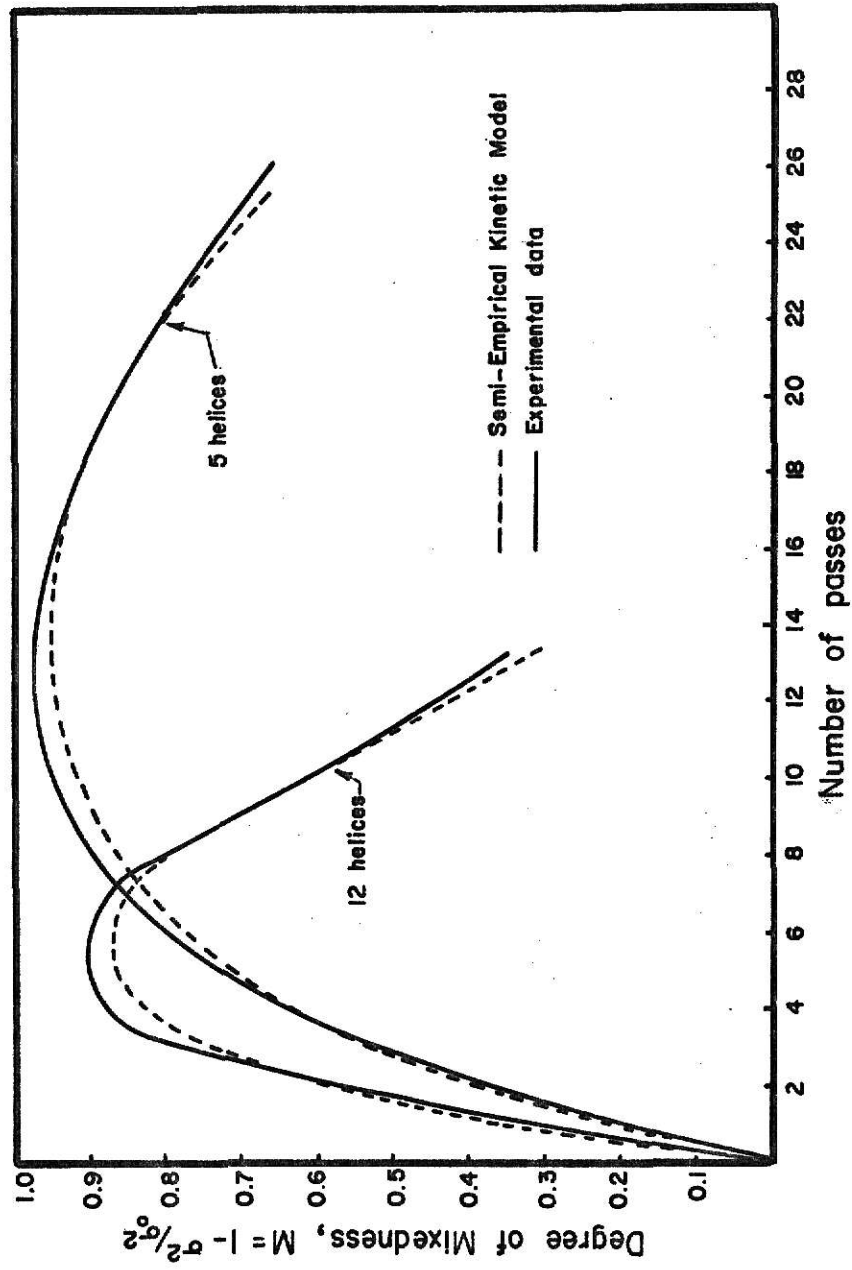
Fig. 9. Approximation given by the Semi-Empirical Kinetic Model for the mixing of 3/16" Lucite-Glass particles.

were of the order of 8.0×10^{-3}). Similar agreement was obtained even with extreme density differences.

The results of the parameter estimation for the system with $\Delta\rho = 1.24$ g/cc are plotted in Fig. 10 as a function of the number of helices. The values for the mixing constant K_1 show a linear relationship with the number of helices in the mixer. The K_1 values were correlated by the following equation:

$$\Delta\rho = 1.24 \text{ g/cc} \qquad K_1 = 3.75 \times 10^{-2} \times \text{(number of helices)}$$

The shape of the curves for K_2 and K_3 are similar to those obtained with particles differing only in size. The parameter K_2 has low values for mixers with small number of helices. After about 7 helices there is a pronounced change in K_2 values as the number of helices in the mixer is increased. The plot of K_3 vs the number of helices indicates that for mixers of small number of helices K_3 starts with a high value and decreases with the number of helices to a minimum value. After this point it increases as the number of helices in the mixer is increased.

Table 2 shows the effect of density differences on the parameters of the Semi-Empirical model for a 12-helices mixer. A plot of K_1 vs $\Delta\rho/\rho_m$ shows a linear relationship, thus indicating that increasing the density differences results in more rapid mixing. The relative influence of density differences on K_2 can be also implied by inspection of Table 2. According to the data it seems that K_2 increases as the density difference increases from zero to some optimal value (value at which the density difference becomes very important). This optimal value appears to be very close to 1.24 g/cc. According to Donald and Roseman (1962) the optimal value is 1.2. Beyond this point K_2 decreases with further increase in density differences. The values for the
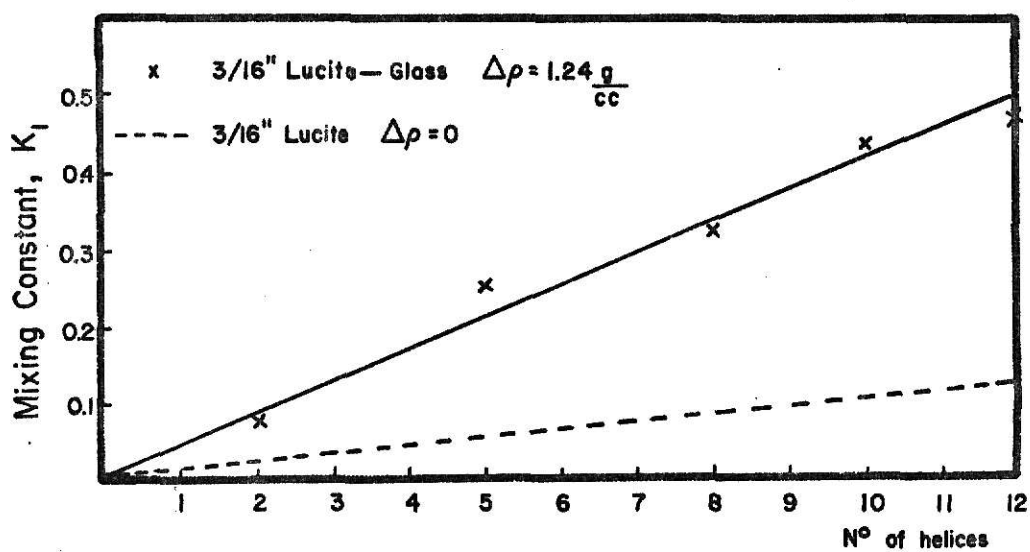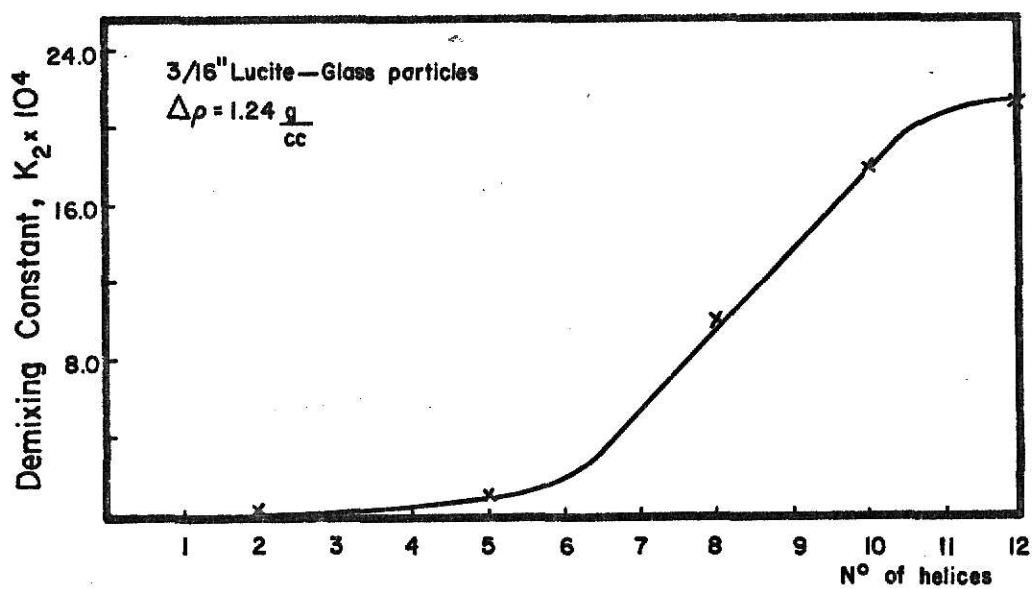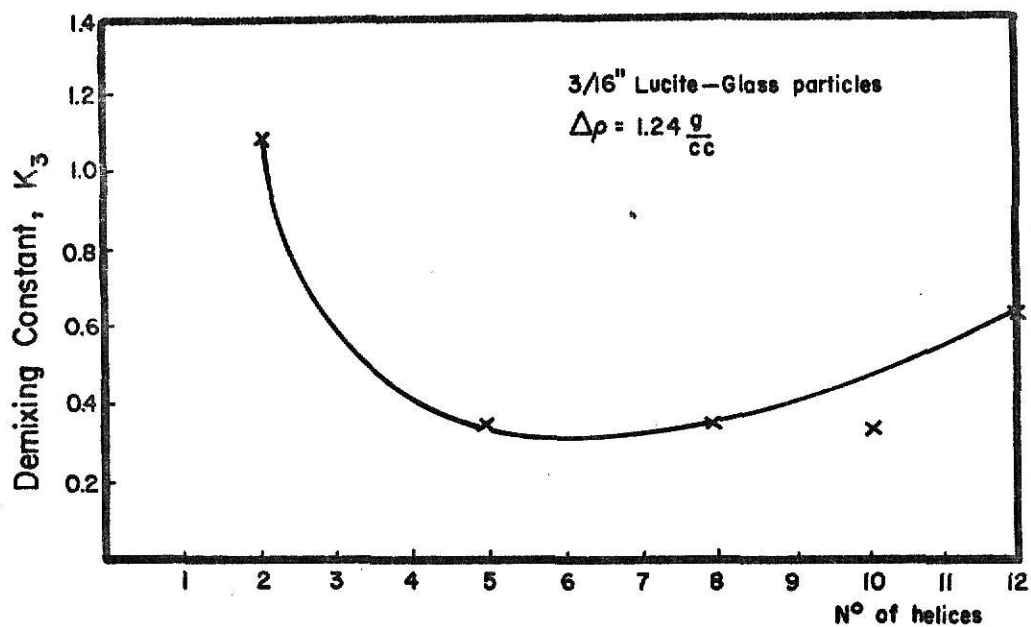
Fig. 10. Estimated parameters as a function of N° of helices.

TABLE 2

EFFECTS OF THE DENSITY DIFFERENCES ON THE
PARAMETERS OF THE SEMI-EMPIRICAL MODEL
FOR A 12 HELICES MIXER

System	$\Delta\rho$ (g/cc)	Particle diameter (inch)	K_1	$K_2 \times 10^3$	K_3
Lucite-Lucite	0	3/16	0.12	0	0
Lucite-glass	1.24	3/16	0.46	1.85	0.638
Lucite-steel	6.42	3/16	0.826	1.17	1.31
Glass-steel	5.18	3/16	0.51	2.46	0.54

parameter K_3 tend to increase with increasing the density differences.

Results obtained for K_2 and K_3 appear to be in a good agreement with the mechanisms of segregation proposed in the Semi-Empirical Kinetic model. According to the mechanism, the mixed state is relatively unstable and tends to break down with a very small rate constant K_2 ($K_1 \gg K_2$) to a more stable state. However, when any small number of particles in the segregated state of type II appear, they act as a catalyst for the breakdown and a second mechanism with a much larger rate constant K_3 takes place to produce additional particles in the segregated state of type II. The order of magnitude of the K_2 values suggests that the initial breakdown is the rate controlling step in the formation of a segregated state of type II. A strong tendency to form the segregated state of type II will exist when K_2 and K_3 both have high values. This situation is observed in the case of density differences higher than 1.24 g/cc (for example, as in the Lucite-steel system). The case in which both K_2 and K_3 are small will tend to produce negligible segregation. This situation occurs when, for example, the density differences are lower than the optimum value of 1.24 g/cc.

A relative comparison between the effect of size and density differences on rate of mixing is very difficult to make at this point because information is lacking on the effects of density levels. Although there is limited information for a comparison, other authors (Fisher, 1963; Donald and Roseman, 1962; Brown and Richards, 1970; Van Demburg and Bauer, 1964; Fan, et al., 1972) and some of the present experiments seem to indicate that very small differences in size can produce higher rates of mixing and segregation than a moderate density difference. In the case of large size differences, however, the present results appear to indicate that large density differences will

have a more pronounced effect than large or modest size differences.

8.9 CONCLUSIONS

1) Graphical comparison of Rose's model, Weidanz's model, and the new model of this study with the experimental data indicates that only Rose's and the Semi-Empirical Kinetic model are capable of providing a good description of the data. The best fit parameters of Rose's model are, however, physically incompatible. Thus, the new model is the only one capable of describing the mixing and segregation of particles differing only in density. The best fit parameters of the new model appear to be compatible with the proposed mechanism on which it is based.

2) Examination of the data by several methods indicates that the presence of density differences will tend to accelerate the rate of mixing in comparison with the case of identical particles. The rate of mixing increases with increasing density differences.

3) A plot of M vs M.A.N. provides a reasonable correlation of data (for a given particle system) obtained with mixers containing different numbers of helices. This result indicates that the end effects in the mixer are small.

4) The mixing parameter of the new model (K_1) is a linear function of the number of helices in the mixer, and of the ratio between the density difference and the average density of the two components $(\Delta\rho/\rho_m)$.

5) The demixing parameter which determines the rate of segregation is K_2. According to the mechanism proposed K_2 must have small values. It represents the formation of a small amount of particles in the segregated state of type II. These particles act as a catalyst for accelerating the formation of the segregated state of type II.

6) Density differences ($\geq$ 1.24 g/cc) can substantially increase the rate of mixing and segregation. As the density differences are increased, the rate at which particles mix and demix increases, but the point where segregation starts is reached earlier.

NOTATION

A   Constant of mixing in Rose's model

B   Constant of demixing in Rose's model

D   Diameter, inch

E   Constant

K_1 Constant of mixing in the Semi-Empirical Kinetic model

K_2 Constant

K_3 Constant

M   Degree of mixedness

N   Number of passes

Q   Constant

GREEK LETTERS

σ Standard deviation

σ_o Standard deviation at $t = 0$

σ_∞ Standard deviation at t

ϕ Coefficient of mixing velocity

ρ_m Average density of the two components

$\Delta\rho$ Density difference

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CHAPTER 9

MIXING OF PARTICLES WITH DIFFERENCES IN SIZE AND DENSITY

9.1 INTRODUCTION

The purpose of this chapter is to present the results of the investigation of axial mixing in motionless mixers for particles differing in both size and density. The study was concerned with the influence of size and density differences, number of helices, and number of passes on the rate of mixing. The Semi-Empirical Kinetic model was the only one employed to represent the experimental data. Rose's and Weidanz's models were not used because of the physical incompatibility shown by the former and the poor fitting obtained with the latter in the previous studies (Chapters 7 and 8.)

It has already been indicated that in a dense-light system the denser particles tend to go to the bottom of the slug of particles, and when they are of the same density but different in size the larger particles tend to go to the bottom. Therefore, it might be expected that a combination of small-light particles and heavy-big particles would exhibit increasing segregating tendencies. This phenomenon was studied for a system of Lucite-glass particles 5/32" and 3/16" in diameter.

9.2 CONCENTRATION PROFILES

Typical concentration profiles for the case of differences in both size and density are shown in Fig. 1. The data in the figure are for an 8-helices mixer with 5/32" Lucite particles and 3/16" glass particles. They illustrate how the axial concentration of the key particles (5/32" Lucite) change as a
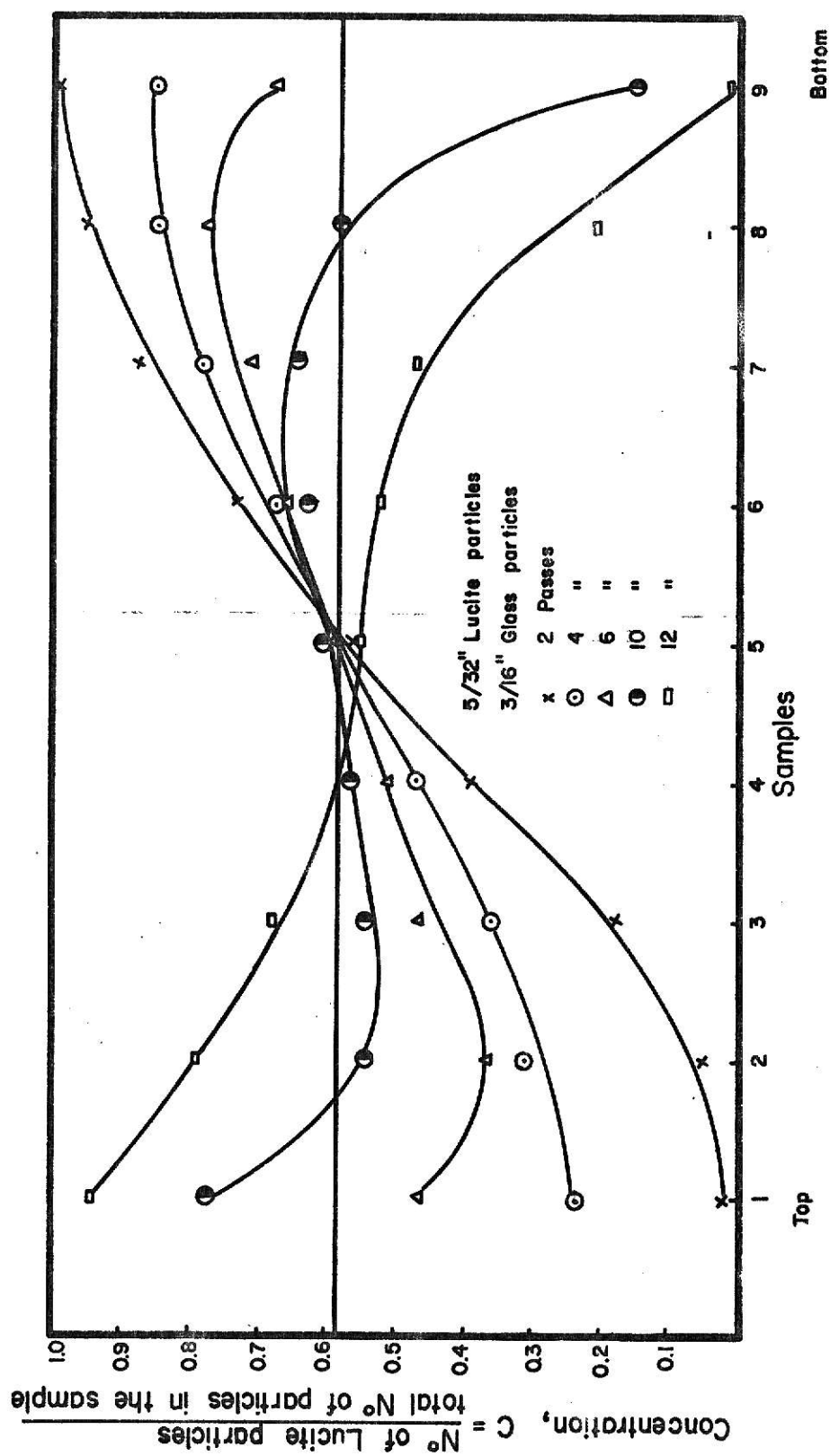
Fig. I. Variation of the concentration profiles with the number of passes for an 8 helices mixer.

function of the total number of passes. Initially only mixing is observed, but as the number of passes is increased segregation pockets start appearing at each end of the slug of particles (as a result of size and density differences). However, the net effect (initially) is one of increasing the overall degree of mixedness. Further increase in the number of passes produces a continued movement of the Lucite particles (smaller and lighter particles) to the top of the slug of particles, indicating a predominance of the segregation effect. The concentration profiles for a 12-helices mixer when compared to those for fewer helices show that the homogeneity of the mixture increases faster as the number of helices is increased. In the case of 12 helices, only 4 passes can produce a good mixture. After a maximum homogeneity has been reached, separation of the particles is noted in top and bottom sections, although mixed material remains in the middle sections. Segregation is observed to take place to a lesser extent (after an equal number of passes) in the case of only size or density differences than in the case of differences in both properties.

9.3 STANDARD DEVIATION

The behavior of the standard deviation as a function of the number of passes and number of helices in the mixer for particles with differences in both size and density was also examined. A typical plot of $\log_{10} \sigma$ vs N showed a transient period, a linear period, and a region where σ increased (nonlinearly) with increasing the number of passes. The linear portion represents the region where mixing is the predominant effect. The minimum value of σ corresponds to a maximum degree of homogeneity for the system. The nonlinear portion represents the region where segregation is the dominant effect. The experimental observations suggest that the nonlinear portion tends to a

limit as the number of passes becomes large.

Experimental results in the motionless mixer with Lucite (5/32") particles and glass (3/16") are presented in Fig. 2. The linear portion was correlated by equation (1) in Chapter 6. Values for σ_o' and ϕ were calculated and are presented in Table 1. As expected, ϕ increases with the number of helices in the mixer. Comparison of ϕ values with those of the previous cases (size differences, or density differences) shows that the coefficients of mixing velocity are larger for the combined effect. The results indicate that the rate of mixing proceeds faster, but the mixing period is shorter and segregation will start earlier.

9.4 DEGREE OF MIXEDNESS

The behavior of plots of the degree of mixedness (defined in terms of the variance) vs the number of passes was also examined in a motionless mixer. Experimental results are presented in Fig. 3. Typically, the plot of M vs N shows a rapid initial rise in M up to a maximum (which is not far removed from a random mixture), followed by a decline in M which levels off to a non-zero value.

In all cases the small particles (5/32" Lucite) were initially placed at the bottom and the coarser (3/16" glass) were layered on top. The disturbance produced by passing through the mixer resulted in the Lucite particles moving from the bottom to higher positions. As a result of the action the homogeneity improved rapidly during the early moments of mixing, and only after a maximum degree of mixedness was achieved did the segregation effect start to appear. This behavior was accelerated as the number of helices in the mixer increased.
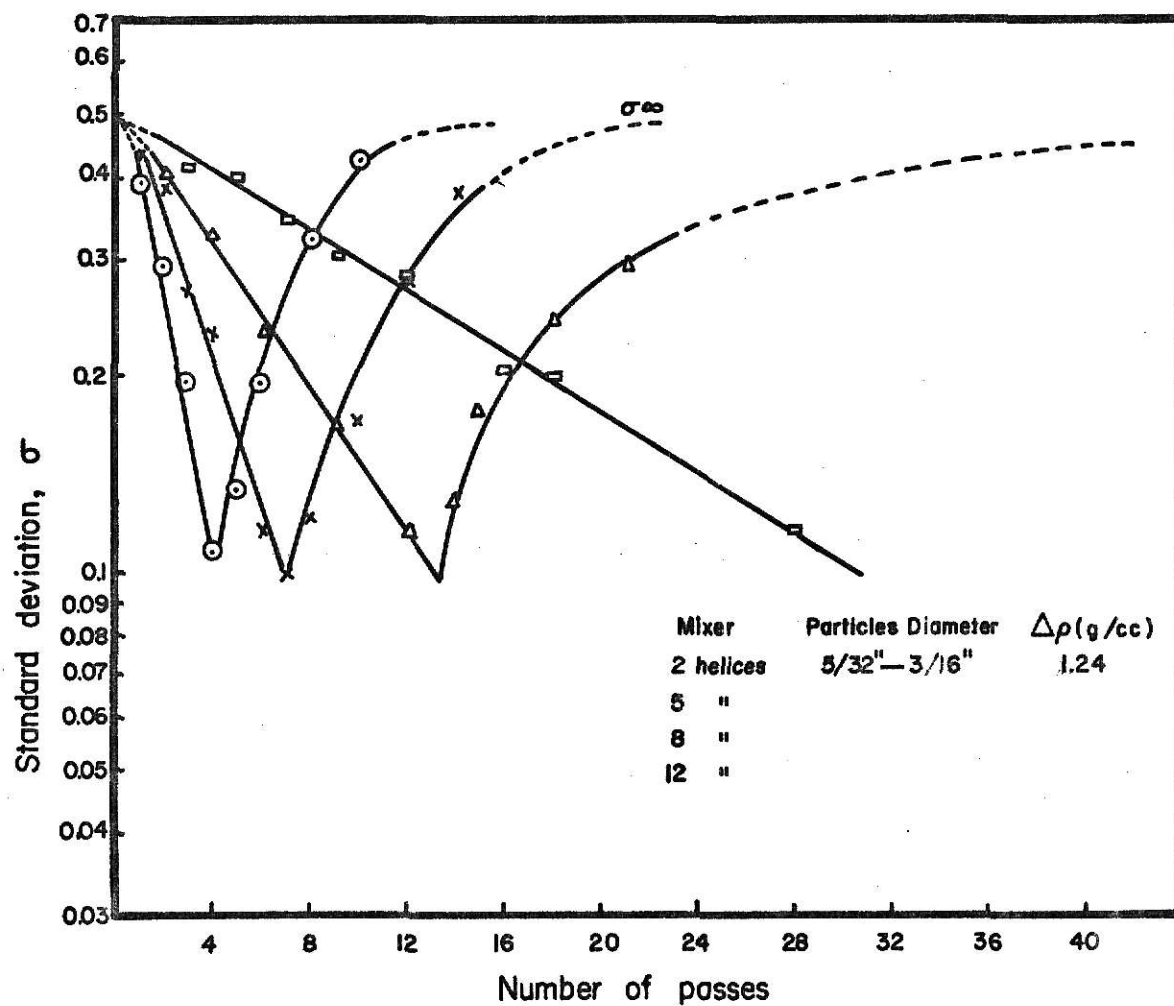
Fig. 2. Correlation for σ and number of passes for motionless mixers with particles differing in size and density.

TABLE 1

INFLUENCE OF SIZE AND DENSITY DIFFERENCES
IN THE STANDARD DEVIATION

No. of helices	Particles diameter (inch)	$\Delta\rho$ (g/cc)	$\sigma_o{}'$	ϕ (1/pass)
2	5/32-3/16	1.24	0.513	0.055
5	5/32-3/16	1.24	0.521	0.126
8	5/32-3/16	1.24	0.601	0.263
12	5/32-3/16	1.24	0.658	0.429
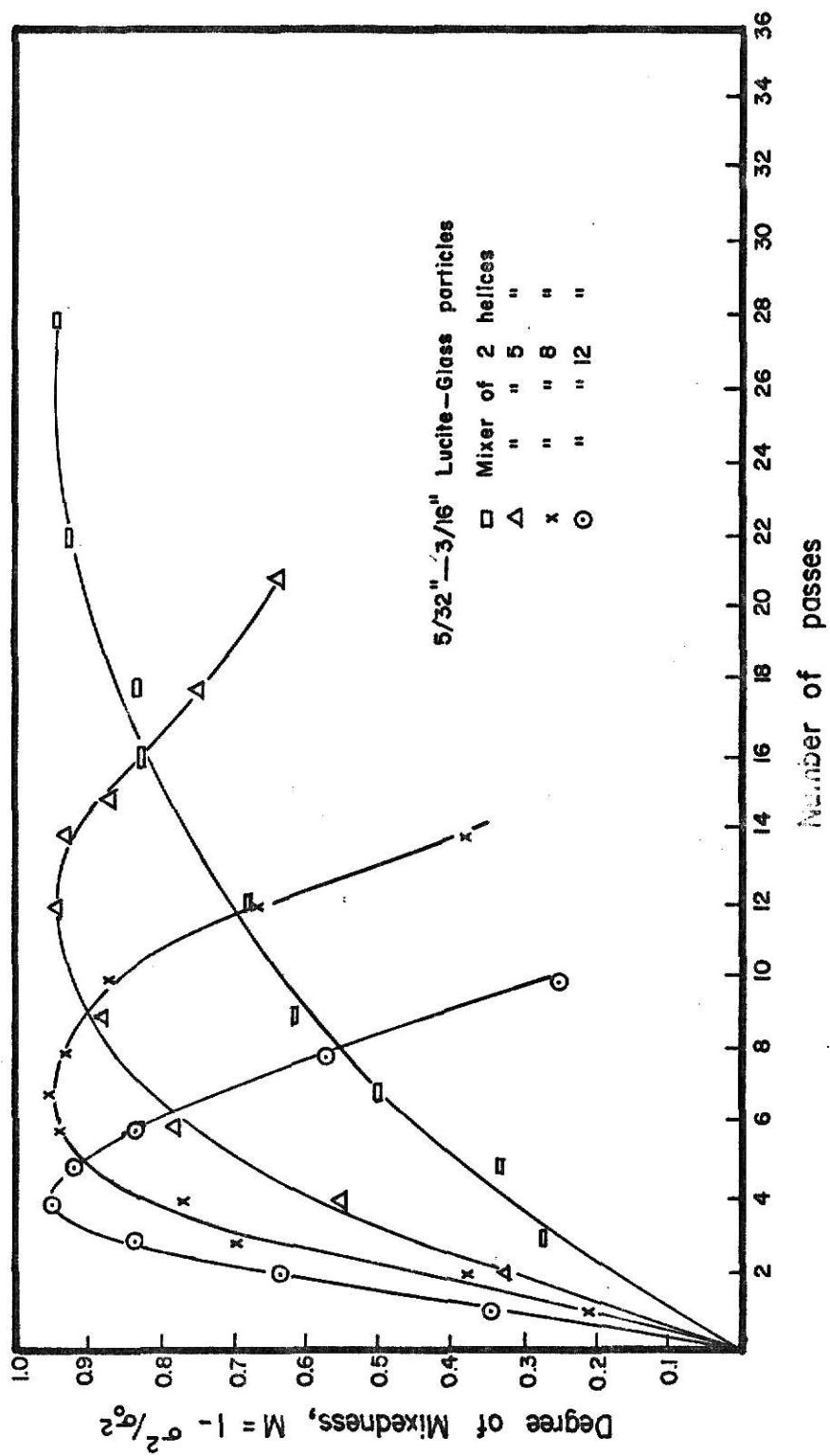
Fig. 3. Rate of axial mixing for particles differing in size and density.

9.5 MIXING ACTION NUMBER (M.A.N.)

The data were expressed in terms of M.A.N. as defined previously. Figure 4 presents the results for the 5/32"-3/16" Lucite-glass system. The plot shows a good correlation among the experimental data. In comparison with size differences similar correlations were obtained. With respect to the system where only density differences exist the correlation is considerably better.

9.6 SEMI-EMPIRICAL KINETIC MODEL

For the mixing of particles with differences in physical properties in a motionless mixer the Semi-Empirical Kinetic model becomes:

$$M = 1 - e^{-K_1 N} + \frac{K_2/ K_3 \left[\exp\left(K_2 + K_3 \frac{\sigma_\infty^2}{\sigma_o^2}\right) N - 1\right]}{1 + \frac{K_2}{K_3} \frac{\sigma_o^2}{\sigma_\infty^2} \exp\left(K_2 + K_3 \frac{\sigma_\infty^2}{\sigma_o^2}\right) N} \tag{1}$$

where

$$M = 1 - \frac{\sigma^2}{\sigma_o^2}$$

The formulation of this equation was considered in section 3.3.3. The values of σ_∞ were determined by extrapolation of the log σ vs N curves as is shown in Fig. 2. Using Bard's technique (1967) for nonlinear parameter estimation, the parameters of the model (K_1, K_2, and K_3) were obtained. An illustration of the approximation obtained with the model is presented in Fig. 5 for mixers of 5 and 12 helices. The general result for the system of particles gave a maximum error of 5% (sums of square of residuals of the
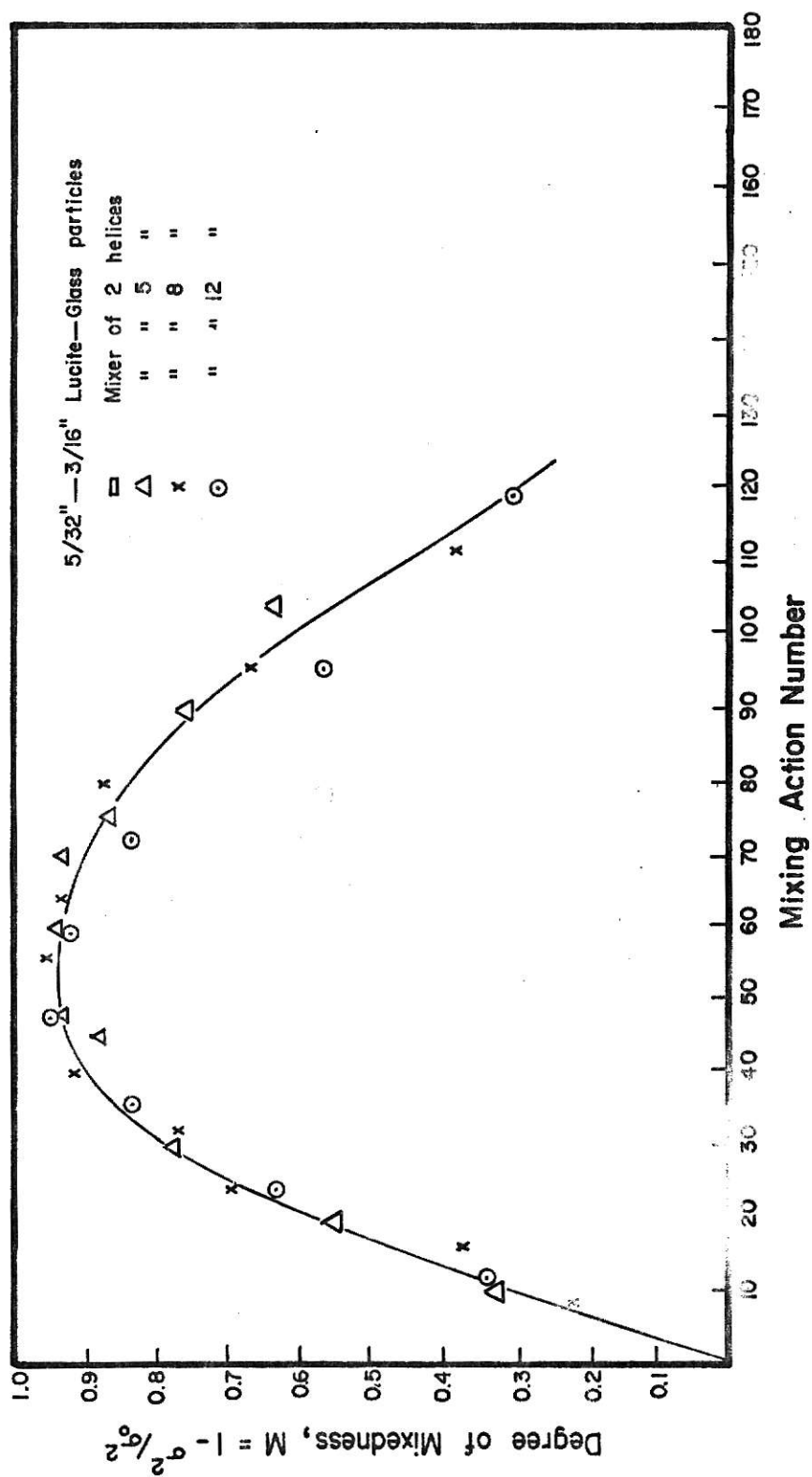
Fig. 4. Correlation between degree of mixedness and M. A. N.
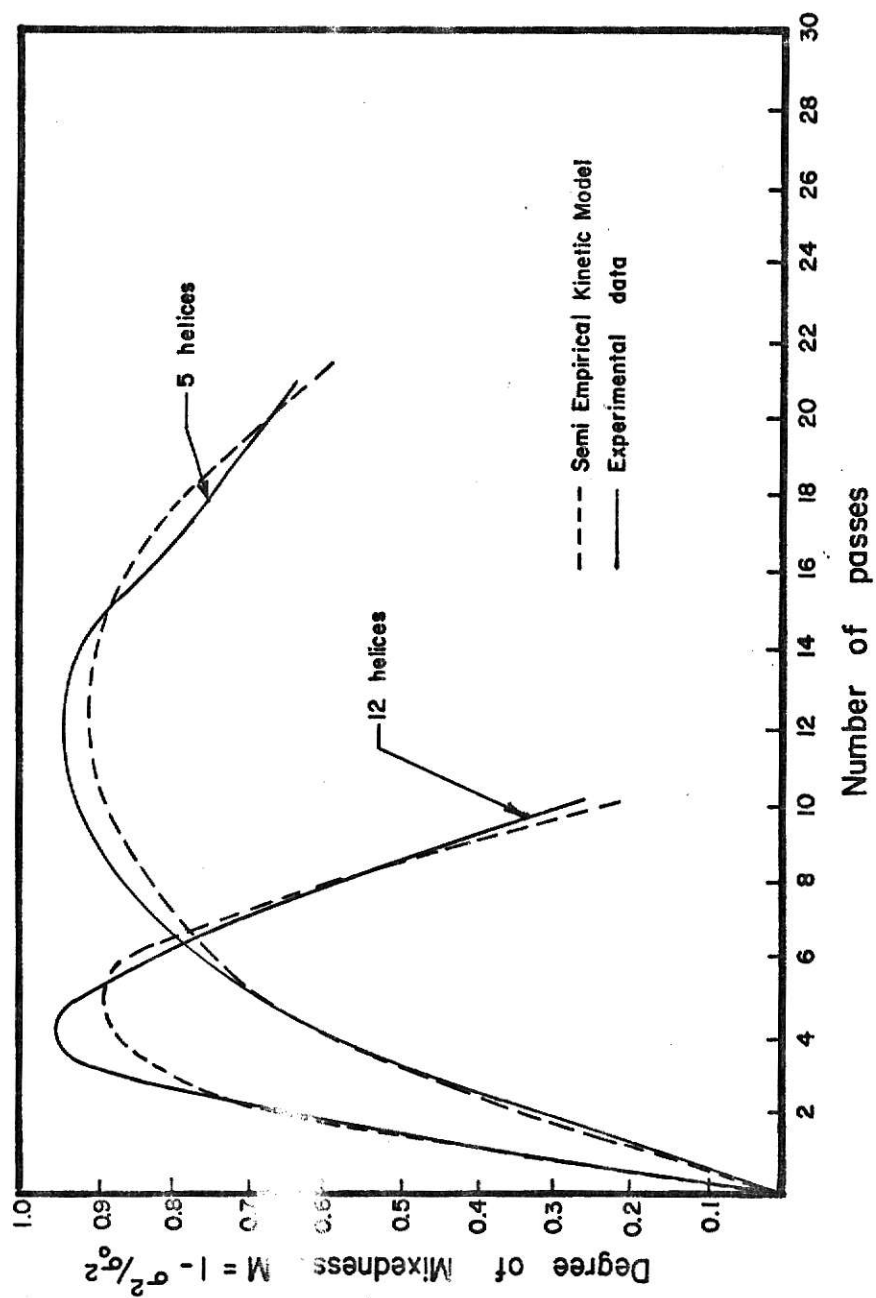
142

Fig. 5. Approximation given by the Semi-Empirical Kinetic Model for the mixing of particles differing in size and density.

order of 0.01) between the experimental data and the model predictions. However, for an 8-helices mixer a maximum error of 10% (sum of square of residuals of the order of 0.04) was observed.

The results of the parameter estimation for the system are plotted in Fig. 6 as a function of the number of helices. The values for the mixing constant K_1 show a linear relationship with the number of helices in the mixer. The magnitudes of K_1 were larger than for identical particles, particles with size differences, and particles with density differences, indicating a faster rate of mixing over the previous cases. The mixing constant K_1 was correlated as a function of the number of passes by the following equation:

5/32"-3/16"

$\Delta\rho = 1.24$ g/cc $K_1 = 4.5 \times 10^{-2} \times$ (number of helices)   (2)

The shape of the curves for K_2 and K_3 are similar to those obtained with particles differing only in size or density. The parameter K_2 has low values for mixers of small number of helices. After about 7 helices there is an increase in K_2 values as the number of helices in the mixer is increased. In general, K_2 values were of the same order as K_2 for particles with a size difference (1/8"-3/16"). The parameter K_3 starts with high values for mixers of small number of helices, and decreases with the number of helices to a minimum value. After this point it increases as the number of helices in the mixer is increased. Values of K_3 with mixers of more than 5 helices were greater than K_3 values observed when only size or density differences were present.

Results obtained for K_2 and K_3 appear to be in good agreement with the machanisms of segregation proposed in the Semi-Empirical Kinetic model.
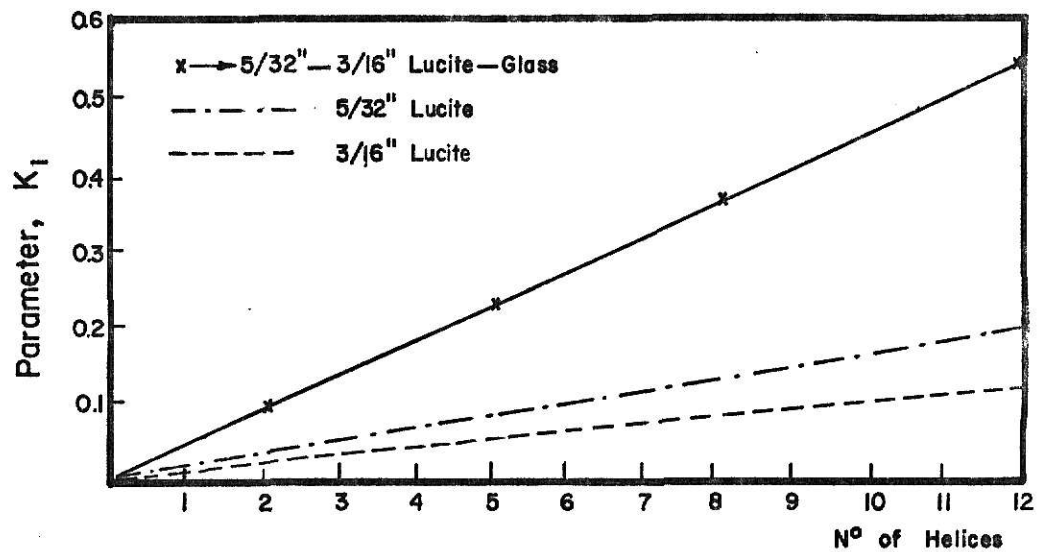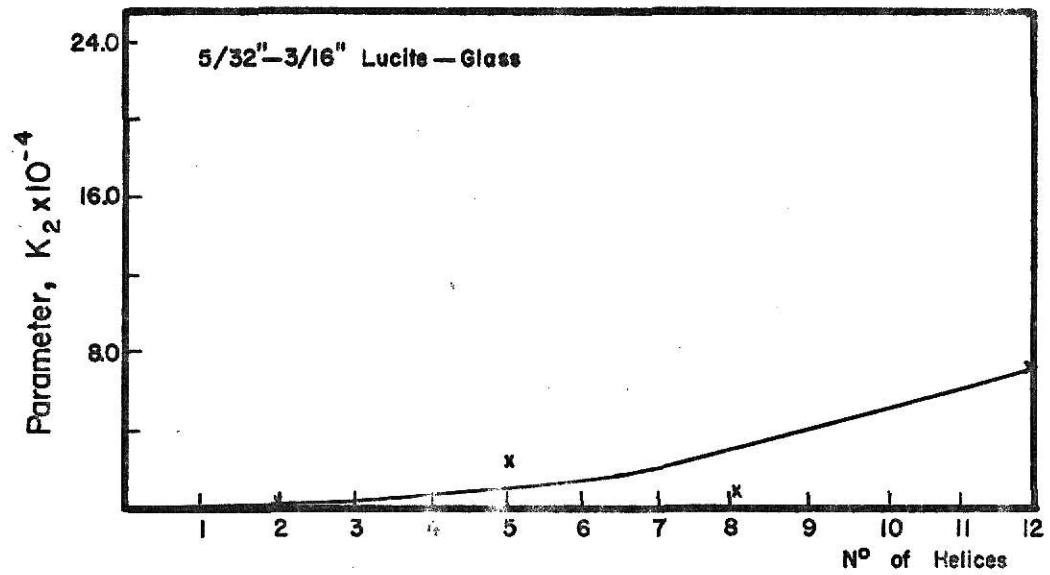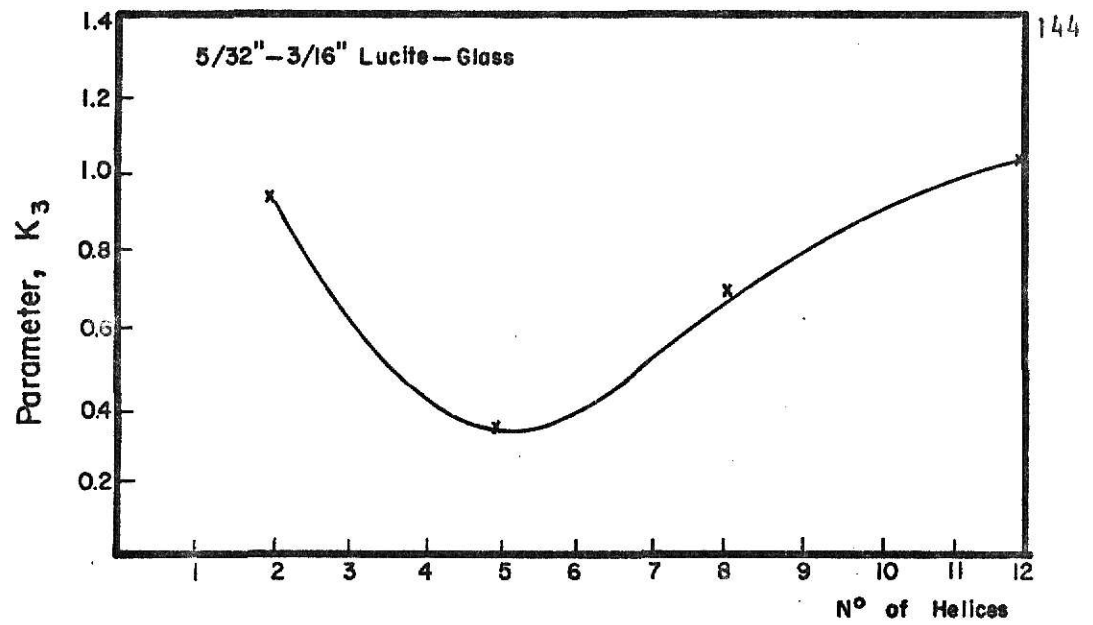
Fig. 6. Estimated parameters as a function of the N° of Helices.

According to the mechanisms, the mixed state is relatively unstable and tends to break down with a very small rate constant K_2 ($K_1 \gg K_2$) to a more stable state. However, when any small number of particles in the segregated state of type II appear they act as catalyst for the breakdown, and a second mechanism with a much larger rate constant K_3 takes place to produce additional particles in the segregated state of type II. The order of magnitude of the K_2 values suggest that the initial breakdown is the rate controlling step in the formation of a segregated state of type II.

9.7 CONCLUSIONS

1) The Semi-Empirical Kinetic model of this work has been successfully applied to describe the axial mixing of particles with differences in size and density. The best fit parameters appear to be compatible with the proposed mechanism on which the model is based.

2) Small values were observed for K_2, indicating that the initial breakdown of the mixed state is the controlling step in going to a segregated state of type II.

3) Good correlations among the experimental data were observed under the M.A.N. criteria, indicating that end effects can be neglected.

4) Differences in both size and density (for small-light with big-heavy system) considerably increase the rate of mixing and segregation. The rates are notably different from those for size or density difference alone.

NOTATION

D	Diameter, inch
K_1	Consta of mixing in the Semi-Empirical Kinetic model
K_2	Consta
K_3	Constant
M	Degree of mixedness
N	Number of passes

GREEK LETTERS

σ	Standard deviation
σ_o	Standard deviation at $t = 0$
σ_∞	Standard deviation at $t \sim \infty$
ϕ	Coefficient of mixing velocity
ρ	Density, g/cc

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CHAPTER 10

CONCLUSIONS AND RECOMMENDATIONS

The major conclusions of the study are summarized below.

1) Comparison of Rose's model and the new model of this study (on the basis of the sum of squares of residuals) indicates that both are equally applicable to the case of mixing particles with identical properties. The major difference in the models is that the new model contains one parameter, whereas Rose's model contains two.

2) Graphical comparison of Rose's model, Weidanz's model, and the Semi-Empirical Kinetic model with experimental data for particles with differences in size, density, or both, indicates that only Rose's and the new model are capable of providing a good description of the data. The best fit parameters of Rose's model are, however, physically incompatible. Thus, the new model is the only one capable of describing the mixing and segregation of the experimental data. The best fit parameters of the new model appear to be compatible with the proposed mechanism on which it is based.

3) Plots of M vs M.A.N. provided a reasonable correlation of data (for a given particle system) obtained with mixers containing different numbers of helices. This result indicates that the end effects in the mixer are small.

4) For particles with identical physical properties only mixing was obtained. If the particles presented a difference in size or density, mixing was

observed during early moments, and only after a maximum degree of mixedness was reached did the segregation effect start to appear.

5) For particles with identical physical properties, the parameter K_1 increased with decreasing particle diameter.

6) The parameter K_1 increased as the size difference was increased from zero to some critical value $(\Delta D)_c$ (this critical difference was $1/32''$ for the cases studied); beyond this point, K_1 decreased with further increase in size difference.

7) In general, higher rates of mixing can be observed for the mixing of particles with differences in size, density, or both than for identical particles.

8) Small size differences can produce higher rates of mixing and segregation than moderate density differences. In the case of large size differences however, the present results appear to indicate that large density differences may have a more pronounced effect.

9) For all the cases studied, the parameter K_1 was a linear function of the number of helices in the mixer, and of the ratio between the density difference and the average density of the two components.

10) The termination of convective mixing seems to determine the optimum time of mixing.

11) When particles present differences in size, density, or both, the use of small number of helices will be useful to control segregation. Another means of controlling segregation is the use of particles with identical physical properties.

12) The demixing parameter which determines the rate of segregation is K_2. According to the mechanisms proposed, the mixed state is relatively

unstable and tends to break down with a very small rate constant K_2 $(K_1 \gg K_2)$ to a more stable state. However, when any small number of particles in the segregated state of type II appear, they act as a catalyst for the breakdown and a second mechanism with a much larger rate constant K_3 takes place to produce additional particles in the segregated state of type II.

The motionless mixer was found to be a good segregator and very easy to operate. The preliminary investigation on the mixing of particles differing in size, density or both, showed encouraging results. Some recommendations for further studies are:

1) Study of the importance of density levels in mixing and segregation. Relative importance of density differences of lower values than 1.24 g/cc. Effect of mixing small-heavy with big-light particles on segregation. Another study involving the mixing of particles of different shapes would be valuable, too.

2) Research in solids mixing should have design as its ultimate goal; hence, correlations for the parameters K_2 and K_3 in terms of number of helices in the mixer and correlations for all the parameters $(K_1, K_2, \text{ and } K_3)$ in terms of dimensionless numbers need to be developed. The influence of other variables such as diameter of the mixer, angle of twisting of helices, order of helices, etc., on the parameters of the Semi-Empirical model need some study, too. A considerable extension of the experimental work would be required.

3) A method to estimate σ_∞ values in terms of physical properties of the particles.

4) The model and results of this study might be useful for optimization of

the rate of axial mixing by computer simulation of the process.

5) Application of the motionless mixer to the solution of separation problems.

6) The importance of electrostatic effects on mixing and segregation, because electrostatic effects can produce demixing by creating agglomeration.

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MIXING AND SEGREGATION OF PARTICULATE SOLIDS
IN A MOTIONLESS MIXER

by

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AN ABSTRACT OF A MASTER'S THESIS

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Motionless mixers were used to investigate the rate of mixing and segregation of particulate solids. Spherical Lucite, Glass and Steel particles were utilized. The influence of size and density differences on mixing and segregation were considered.

Experimental data were utilized to evaluate average concentrations, variance and standard deviation. A degree of mixedness was defined and experimental data were transformed by employing the definition. Plots of degree of mixedness vs number of passes were constructed. A definition of the mixing action number (M.A.N.) was presented, in which it was assumed that end effects were negligible. Correlations of the experimental data on the basis of the M.A.N. criteria were obtained.

Existing mathematical models (Rose's and Weidanz's) as well as a new Semi-Empirical Kinetic Model were proposed to describe the complex mixing and segregation mechanisms. The Semi-Empirical Kinetic Model described the experimental data reasonably well for particles with identical physical properties, and particles differing in size, density, or both. Rose's and Weidanz's models were found unsuitable for describing mixing and segregation in the motionless mixer.

The parameters of the models were estimated and plotted as a function of the number of helices in the mixer. The parameter K_1 of the Semi-Empirical Model showed a linear relationship with the number of helices in the mixer. For mixers of small number of helices, the parameter K_2 was small; after a 7 helices mixer was used, a pronounced change in the parameter K_2 was observed and it increased to higher values. On the basis of this model, it seemed reasonable to conclude that K_2 is the controlling parameter in passing from a segregated state of type I to a segregated

state of type II.

It was also observed in general, that higher rates of mixing could be obtained with particles differing in physical properties than with particles of identical properties.