

Application of a Discrete Mixing Model to the Study of Mixing of Multicomponent Solid Particles

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A discrete steady-state Markov chain model of the solids mixing process in a multicomponent homogeneous particle system is proposed. Particles in a homogeneous particle system have identical physical properties and are distinguishable only through the characteristics of tracers. The variances of sample compositions at the completely randomized and the completely segregated states are derived from the model. The degree of mixedness based on variances is defined from these two reference limits. The model can predict the concentration distribution of the degree of mixedness of a multicomponent homogeneous particle system blended by passing through a motionless mixer, from prior knowledge of one-step transition probabilities. These transition probabilities can be obtained from mixing experiments with binary homogeneous particle systems of the same size, density, and shape as those in the multicomponent systems. The experimental results of mixing a three-component homogeneous particle system in motionless mixers are in good agreement with those predicted from the model.

I. Introduction

This study is concerned with a stochastic approach to the mixing of multicomponent solid particles. In the past such an approach has been applied only to the mixing of two-component particle systems. In mixing solid particles, a mixed state is usually attained through the motion of particles induced by the contact between particles and mixer. Mixers may be in motion (e.g., rotating drum mixer) or motionless (e.g., reversed helices mixer).

It is well known that several predominating mechanisms, shear, and bulk motion of particles (convective mechanism), act on particles and eventually lead to a random state of the particles in a mixer (Lacey, 1954). In his study of a two-component homogeneous particle system in a drum mixer, Lacey (1943) described the phenomenon of mixing by simulating it mathematically as a diffusional process. Such a simple approach is not applicable to a mixer in which several mechanisms predominate. The deterministic governing equations that describe the mixing process in such a mixer are usually too difficult to solve. Inoue and Yamaguchi (1969) used a stochastic approach to the study of solids mixing in a V-type mixer. They considered the mixing process as a steady-state unrestricted simple Markov process (first-order Markov chain). This stochastic approach was also applied by Chen et al. (1972) to their study of the mixing of a binary homogeneous mixture by passing through a motionless mixer. A binary homogeneous particle mixture contains two groups of particles with the same size and density but with different colors.

Due to their simplicity, binary systems have been studied most often by researchers in the field of solids mixing. Relatively little has appeared in the literature on the mixing of polynary (multicomponent) systems (see definition in section II). Development of a stochastic model for the mixing of a polynary homogeneous (or nonsegregating) particle system is the subject of this paper. A tertiary or three-component system is experimentally studied to verify the model. One of the advantages of the present stochastic approach is that the Markov process can be applied to almost any class of mixers and the design complication of a mixer

does not create much difficulty in the use of this approach. The approach can be extended easily to segregating materials (different size, different density, etc.).

Through high-speed photography Chen et al. (1972) were able to observe the operation of several mixing principles in a motionless mixer. They are (1) multiple division and combinations of the flow of particles, (2) interaction of the particles with other particles, the helices, and the wall of the mixers, (3) change in the direction of the flow of particles, and (4) difference in the velocity profile of the particle. It can be expected that all these mechanisms (diffusion, convection, and shear) of the solids mixing are present to some extent, in the motionless mixer employed in this work.

II. Classification of Solid Particle Systems

A convenient and consistent scheme for classifying particle systems should be established to facilitate the investigation of the mixing of solid particles. Particle systems may be classified into three general classes, unitary, binary, and polynary. However, a unitary class which consists of only identical particles does not come into the picture in considering the mixing process. A binary particle system contains two different types of particles while a polynary particle system contains more than two different types. Each type of particle system can be further divided into two subclasses, homogeneous and heterogeneous. A homogeneous subclass has the same physical properties and is distinguishable through the characteristics of tracers, for example, color, radioactivity, etc. Here materials with physical properties which do not interact with the operation of a mixer are considered tracers. On the other hand, a heterogeneous subclass has different physical properties, such as size, density, and shape. For example, a particle system containing four distinctly different combinations of size and density is called a four-component or polynary heterogeneous particle system. The particle system employed by Lacey (1954) which contained identical particles distinguishable only by color (black and white) was a two-component or binary homogeneous particle system. The general classification is summarized in Table I. Examples of different classes of particle systems are also given in Table I.

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Table I. Classification of Particle Systems

Particle system classification Definition	Binary or two-component system		Polynary or multicomponent system	
	Containing two different classes of particles		Containing more than two different classes of particles	
Sub-classification	Homogeneous	Heterogeneous	Homogeneous	Heterogeneous
Definition	Same physical properties distinguishable only by tracers (color, radioactivity, etc.)	Different physical properties such as size, shape, etc.	Same physical properties	Different physical properties
Examples	Black and white particles of the same material with uniform size distribution	Particles of the same material but with two different sizes	Particles of more than two kinds of color of the same material with uniform size distribution	Particles of different densities and different sizes

III. Development of a Discrete Mixing Model

The stochastic process is a description of random phenomena changing with respect to time. A stochastic process may be thought of as a set of random variables X_t depending on a parameter $t \in T$. Here the parameter set T may be interpreted as the time. For example, X_t may be the concentration of a certain component in a mixture, the number of particles in a sample, or the position of a particle in space. A Markov process can be briefly defined as a process with the property that, given the value of X_t , the value of X_a ($a > t$) does not depend on the value of X_b ($b < t$). In other words, the probability of any particular future behavior of the process, when its present state is known exactly, is not changed by additional knowledge of its past behavior. However, if our knowledge of the present state of the process is imprecise, then the probability of some future behavior will, in general, be altered by additional information relating to the past behavior of the system. This work was concerned with the application of a specific Markov process, namely the discrete time Markov chain, for which both the state space and index parameter are discrete, to the blending of a multicomponent or polynary homogeneous particle system.

The model is experimentally verified on a tertiary homogeneous particle system. The discrete time Markov chain can be formally defined in the analysis of a mixing process as follows (Parzen, 1962).

Suppose that we are given a sequence of experiments and as a result of each experiment there can be one and only one event from a finite or countable set of pairwise mutually exclusive events S_1, S_2, S_3, \dots , where

$$S_j: X_{t-N\tau} = j$$

or simply

$$S_j: X_N = j \quad (j = 1, 2, \dots)$$

The set $S = [S_1, S_2, \dots]$ is called the state space. When the event S_j occurs, the system is said to have passed into the state of S_j .

A sequence of random variables $[X_N, N = 0, 1, 2, \dots]$ can be defined as a Markov chain if, for every finite collection of integers $N_1 < \dots < N_n < N$, we have

$$Pr [X_N | X_{N_1}, X_{N_2}, \dots, X_{N_n}] = Pr [X_N | X_{N_n}] \quad (1)$$

If the possible values of X_N are $1, 2, \dots$, the Markov chain is denumerable. In this case

$$Pr [X_{N+1} = i | X_N = j] = p_{ij}^{(N)} \quad (i, j = 1, 2, \dots) \quad (2)$$

If $p_{ij}^{(N)}$ is independent of N for all i, j , the Markov chain is homogeneous. For a homogeneous Markov chain, the transition probabilities are constant, i.e.

$$Pr [X_{N+1} = i | X_N = j] = p_{ij} = \text{constant} \quad (3)$$

In general

$$Pr [X_{N+M} = i | X_N = j] = p_{ij}^{(M)} \quad (M = 1, 2, \dots) \quad (4)$$

It follows that

$$p_{ij}^{(0)} = \delta_{ij} = \begin{cases} 1 & i = j \\ 0 & \text{otherwise} \end{cases} \quad (5)$$

For a physical interpretation of the Markov chain, we can consider a group of particles being mixed in a mixer. Let the mixer or the mixture blended by passing through motionless mixers be divided into a finite number of cells denoted by $1, 2, \dots, w$, and let $p_{ij}^{(M)}$ be the probability of transition of the number of particles from cell i (state i) at time $M\tau$ to cell j (state j) at time $(M + N)\tau$ (for any M). $p_{ij}^{(N)}$ is called the N -step transition probability, conditional probability, or correlation coefficient. The probabilities $p_{ij}^{(N)}$ can be expressed in terms of p_{ij} as follows

$$p_{ij}^{(N)} = \sum Pr [X_{M+N} = i | X_{M+N-1} = i_{n-1}] \times \dots \times Pr [X_{M+2} = i_2 | X_{M+1} = i_1] Pr [X_{M+1} = i_1 | X_M = j] = \sum_{(i_1, i_2, \dots, i_{n-1})} p_{ii_{n-1}} p_{i_{n-1}i_{n-2}} \dots p_{i_2i_1} p_{i_1i} \quad (6)$$

Hence for a homogeneous Markov chain, eq 6 becomes

$$p_{ij}^{(N)} = \sum_k p_{ik} p_{kj}^{(N-1)} \quad (7)$$

The matrix

$$P = [p_{ij}] \quad (8)$$

is called a stochastic matrix. From eq 7, we see that

$$P^{(N)} = P^N \quad (9)$$

Furthermore, the matrix identity

$$P^{M+N} = P^M P^N \quad (10)$$

yield

$$P^{(M+N)} = P^{(M)} P^{(N)} \quad (11)$$

or

$$p_{ij}^{(M+N)} = \sum_k p_{jk}^{(M)} p_{ki}^{(N)} \quad (M, N \geq 0) \quad (12)$$

This is the well-known Chapman-Kolmogorov equation.

The system under consideration is an ensemble of parti-

cles, which is contained in a collector or container. The state of mixing or mixing characteristics of the system are examined through the observation of relative displacement of the particles in the ensemble located in the collector or container. The helical mixing elements of motionless mixers should be simply considered as mechanical devices which provide the agitation needed to induce such relative displacement when the ensemble of particles is passed through the mixer. In other words, the mixing system under consideration is regarded as being of the batch type. It is worth noting that for any batch mixer, e.g., drum mixers and V-type mixers, mixing is attained through relative replacement of particles in the batch of particles which is placed in the mixer. Thus the term "mixer" and "mixture" can be used interchangeably for batch mixers. This is, of course, not permissible in the case of continuous mixing.

For a mixer of an arbitrary configuration, the entire space in the mixer can be divided into a finite number of cells of any convenient shape for analysis and measurement. The size of each cell should be large enough to represent the average conditions over a region involving every component, yet small enough to assure the continuity of the particle distribution. This condition is significant in that the number concentration of each component inside the cell is assumed to be evenly distributed. The size and shape of the cells need not be equal. For example, the cells adjacent to the walls of the mixer do not necessarily have to be the same size and shape as those far from the walls. Let w be the total number of cells in the mixer and n_i be the number of particles in cell i . Then

$$\sum_{i=1}^w n_i = n \quad (13)$$

where n is the total number of particles in the mixer. With the assumption that pulverization and coagulation do not take place, n is a constant. According to eq 12, the fraction of particles in cell j , ϵ_j , at a certain observation, which are eventually found in cell k , ϵ_k , after a finite time interval of observations (τ), can be described by the transition probability from state E_j to state E_k in one transition step, i.e.

$$p_{kj}^{(N)} = P\{X_N = \epsilon_k | X_{N-1} = \epsilon_j\} = p_{kj} \quad (14)$$

The last equality in the above equation is based on the assumption of a homogeneous Markov chain. Since p_{kj} 's are transition probabilities, they must satisfy the following conditions.

$$\sum_{k=1}^w p_{kj} = 1 \text{ and } 0 \leq p_{kj} \leq 1 \quad (j, k = 1, 2, \dots, w)$$

Suppose that particles of a homogeneous polynary system containing r components, which are identical except for their colors, are to be mixed. Let the number of particles of component j in cell i be m_{ij} , and the total number of particles in cell i be n_i . Then

$$n_i = \sum_{j=1}^r m_{ij} \quad (i = 1, 2, \dots, w) \quad (15)$$

The total number of particles of component j in the mixer, m_j , is related to m_{ij} as

$$m_j = \sum_{i=1}^w m_{ij} \quad (j = 1, \dots, r) \quad (16)$$

and the total number of particles in the mixer, n is related to n_i , m_j , and m_{ij} as

$$n = \sum_{i=1}^w n_i = \sum_{i=1}^w m_j = \sum_{j=1}^r m_{ij} \quad (17)$$

Figure 1 illustrates these relationships.

When mixing is theoretically perfect, we expect that the number concentration of each component through the mixer will reach the final equilibrium concentration given by

$$(C_j)_\infty = \frac{m_j}{n} \quad (j = 1, 2, \dots, r) \quad (18)$$

This means that the concentration of any given component in each cell will also approach the equilibrium concentration $(C_j)_\infty$. Let $C_{kj}(N)$ be the concentration of component j in cell k at $t = N\tau$ ($N = 0, 1, 2, \dots$) where N is the cumulative number of mixing operations. Then, for nonsegregating r -component homogeneous particles

$$\lim_{N \rightarrow \infty} C_{kj}(N) = (C_j)_\infty \quad (j = 1, 2, \dots, r; k = 1, 2, \dots, w) \quad (19)$$

Based on the assumption of a first order homogeneous Markov chain given by eq 11 in describing the solids mixing process, the number of particles of component i moving from cell j to cell k , $Q_{j \rightarrow k}$, during time τ can be written as

$$Q_{j \rightarrow k}(i) = p_{kj} m_{ji} \quad (i = 1, 2, \dots, r; k \neq j; j = 1, 2, \dots, w) \quad (20)$$

$Q_{j \rightarrow k}(i)$ is simply the flow rate of particles of component i from cell j to cell k . When $k = j$

$$Q_{j \rightarrow j}(i) = p_{jj} m_{ji} \quad (i = 1, 2, \dots, r; j = 1, 2, \dots, w) \quad (21)$$

$Q_{j \rightarrow j}(i)$ is the number of particles of component i that remain in cell j . The flow rate of particles of all components moving from cell j to cell k is

$$Q_{j \rightarrow k} = \sum_{i=1}^r Q_{j \rightarrow k}(i) = p_{kj} \sum_{i=1}^r m_{ji} = p_{kj} n_j \quad (22)$$

$Q_{j \rightarrow k}$ is the total flow rate of particles (including all components) moving from cell j to cell k . The number of particles of component j in cell i at time $N\tau$ after the onset of the mixing operation is

$$m_{ij}^{(N)} = \sum_{l=1}^w p_{il}^{(N)} m_{lj}(0) \quad (i = 1, 2, \dots, w; j = 1, 2, \dots, r) \quad (23)$$

where $p_{il}^{(N)}$ is the N th order transition probability from cell l to cell i . The number fraction of component j in cell i is then

$$C_{ij}(N) = \frac{m_{ij}^{(N)}}{n_i} = \frac{1}{n_i} \sum_{l=1}^w p_{il}^{(N)} m_{lj}(0) \quad (24)$$

The number of particles in cell i after transition in N steps is

$$n_i = \sum_{j=1}^w Q_{j \rightarrow i} = \sum_{j=1}^w p_{ij}^{(N)} n_j = p_{i1}^{(N)} n_1 + p_{i2}^{(N)} n_2 + \dots + p_{iw}^{(N)} n_w \quad (25)$$

Dividing both sides of eq 25 by the total number of particles in the mixer, we have

$$\pi_i = \sum_{j=1}^w p_{ij}^{(N)} \pi_j \quad (i = 1, 2, \dots, w) \quad (26)$$

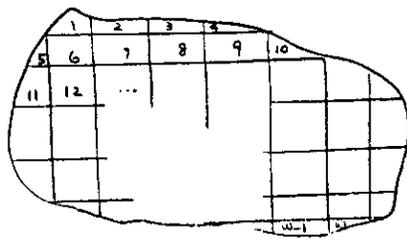
where π_i is the number of fraction of particles (in cell i) to the total number of particles. For steady-state operation, both $p_{ij}^{(N)}$ and π_i can be assumed to be constant with respect to time. In practice, the number of particles in each cell will fluctuate with time. We can assume that π_i ($i = 1, 2, \dots, w$) is the time average of the distribution ratio of the particles in cell i . π_i has the following properties.

$$\sum_{i=1}^w \pi_i = 1$$

and

$$0 \leq \pi_i \leq 1 \quad (i = 1, 2, \dots, w) \quad (27)$$

The particle number balance for component j in the mixer after N transition steps is



An arbitrary configuration of a mixer

Cell Number	1	2	3	...	w
Number of particles in each cell	n_1	n_2	n_3	...	n_w
Number of particles of component 1 in each cell	m_{11}	m_{21}	m_{31}	...	m_{w1}
Number of particles of component 2 in each cell	m_{12}	m_{22}	m_{32}	...	m_{w2}
...
Number of particles of component r in each cell	m_{1r}	m_{2r}	m_{3r}	...	m_{wr}
Total number of particles in each cell	$n_1 = \sum_{j=1}^r m_{1j}$	n_2	n_3	...	n_r

Total number of particles in the mixer
 $n = \sum_{i=1}^w n_i$

Total number of particles of component 1 in the mixer
 $m_1 = \sum_{j=1}^w m_{1j}$

Total number of particles of component 2 in the mixer
 $m_2 = \sum_{i=1}^w m_{i2}$

...

Total number of particles of component r in the mixer
 $m_r = \sum_{i=1}^w m_{ir}$

Figure 1. Illustration of an arbitrary configuration with a solid mixer.

$$m_j = \sum_{i=1}^w C_{ij}(N)n_i = \sum_{i=1}^w C_{ij}(0)n_i = \text{constant} \quad (j = 1, 2, \dots, r) \quad (28)$$

Equation 24 now can be written as

$$C_{ij}(N) = \frac{1}{\pi_i} \sum_{l=1}^w p_{il}^{(N)} \pi_l C_{lj}(0) \quad (i = 1, 2, \dots, w; j = 1, 2, \dots, r) \quad (29)$$

where

$$C_{lj}(0) = \frac{m_{lj}(0)}{n_l} \quad (30)$$

Let

$$C(N) = \begin{bmatrix} C_{11}(N) & C_{12}(N) & \dots & C_{1r}(N) \\ C_{21}(N) & C_{22}(N) & \dots & C_{2r}(N) \\ \vdots & \vdots & \ddots & \vdots \\ C_{w1}(N) & C_{w2}(N) & \dots & C_{wr}(N) \end{bmatrix}$$

$$C(0) = \begin{bmatrix} C_{11}(0) & C_{12}(0) & \dots & C_{1r}(0) \\ C_{21}(0) & C_{22}(0) & \dots & C_{2r}(0) \\ \vdots & \vdots & \ddots & \vdots \\ C_{w1}(0) & C_{w2}(0) & \dots & C_{wr}(0) \end{bmatrix}$$

$$\Pi = \begin{bmatrix} \pi_1 & & & 0 \\ & \pi_2 & & \\ & & \ddots & \\ 0 & & & \pi_w \end{bmatrix}$$

$$P = \begin{bmatrix} p_{11} & p_{12} & \dots & p_{1w} \\ p_{21} & p_{22} & \dots & p_{2w} \\ \vdots & \vdots & \ddots & \vdots \\ p_{w1} & p_{w2} & \dots & p_{ww} \end{bmatrix}$$

Equation 24 can then be written in matrix notations as

$$C(N) = \Pi^{-1} P^N \Pi C(0) \quad (31)$$

Similar results for C in a vector form instead of a matrix form have been obtained for two component systems by Inoue and Yamaguchi (1969). If the initial concentration distribution and the transition matrix are known, the concentration distribution at time $N\tau$ can be calculated from the above equation, with the constraint given in eq 28. Once the concentration distribution is known, the concentration variance σ_N^2 of the mixture at time $N\tau$ is given by

$$\sigma_N^2 = \frac{1}{r} \sum_{j=1}^r \sum_{i=1}^w \pi_i [C_{ij}(N) - (C_j)_\infty]^2 \quad (32)$$

$C_{ij}(N)$ can be simplified further as follows

$$\begin{aligned} C_{ij}(N) &= \frac{1}{n_i} \sum_{l=1}^w p_{il}^{(N)} m_{lj}(0) \\ &= \frac{1}{\pi_i} \sum_{l=1}^w p_{il}^{(N)} \frac{m_{lj}(0)}{n} \\ &= \frac{1}{\pi_i} \sum_{l=1}^w p_{il}^{(N)} \frac{m_j m_{lj}(0)}{n m_j} \\ &= \frac{(C_j)_\infty}{\pi_i} \sum_{l=1}^w p_{il}^{(N)} q_{lj}(0) \end{aligned}$$

where

$$q_{lj}(0) = m_{lj}(0)/m_j \quad (33)$$

is the number fraction of component j in cell l at the beginning of the mixing operation (i.e., $N = 0$). The corresponding concentration variance is

$$\sigma_0^2 = \frac{1}{r} \sum_{j=1}^r \sum_{i=1}^w \pi_i [C_{ij}(0) - (C_j)_\infty]^2 = \frac{1}{r} \sum_{j=1}^r \sum_{i=1}^w [(C_j)_\infty]^2 \pi_i \left[\frac{C_{ij}(0)}{(C_j)_\infty} - 1 \right]^2 \quad (34)$$

$$\frac{C_{ij}(0)}{(C_j)_\infty} = \frac{m_{lj}(0)}{n_i} \frac{n}{m_j} = \frac{m_{lj}(0)}{m_j} \frac{n}{n_i} = \frac{q_{lj}(0)}{\pi_i} \quad (35)$$

and therefore

$$\sigma_0^2 = \frac{1}{r} \sum_{j=1}^r \sum_{i=1}^w [(C_j)_\infty]^2 \pi_i \left[\frac{q_{lj}(0)}{\pi_i} - 1 \right]^2 \quad (36)$$

Substitution of eq 33 into eq 36 yields

$$\sigma_0^2 = \frac{1}{r} \sum_{j=1}^r \sum_{i=1}^w [(C_j)_\infty]^2 \pi_i \left[\frac{1}{\pi_i} \sum_{l=1}^w p_{il}^{(N)} q_{lj}(0) - 1 \right]^2 \quad (37)$$

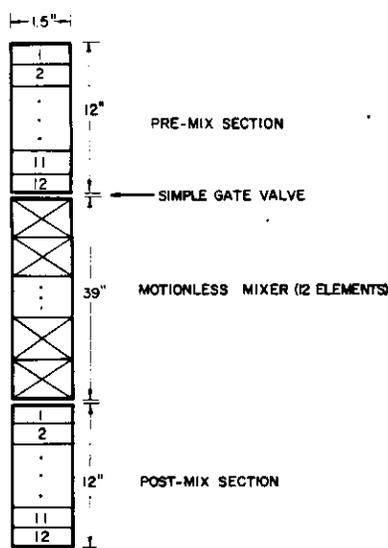


Figure 2. Schematic setup of the experiment.

The variance reduction ratio ψ can be written as

$$\psi = \frac{\sigma_N^2}{\sigma_0^2} = \frac{\sum_{j=1}^r \sum_{i=1}^w [(C_j)_\infty]^2 \pi_i \left[\frac{1}{\pi_i} \sum_{l=1}^w p_{li}^{(N)} q_{lj}(0) - 1 \right]^2}{\sum_{j=1}^r \sum_{i=1}^w [(C_j)_\infty]^2 \pi_i \left[\frac{q_{ij}(0)}{\pi_i} - 1 \right]^2} \quad (38)$$

The degree of mixedness may be defined as (Lacey, 1943).

$$M = 1 - \psi$$

Since

$$\lim_{N \rightarrow \infty} \sigma_0^2 = 0 \quad (39)$$

M is unity when mixing is complete and M is zero at the onset of mixing. According to eq 39, the degree of mixedness is a function of the final equilibrium concentration $(C_j)_\infty$, fraction of the number of particles in each cell π_i , and initial ratio distribution $q_{ij}(0)$. Note that for a two-component system for which $r = 2$, the degree of mixedness is not affected by the final equilibrium concentration $(C_j)_\infty$. This can be easily shown by setting $r = 2$ in eq 38, namely

$$\psi = \frac{\sum_{i=1}^w \pi_i \left[\frac{1}{\pi_i} \sum_{l=1}^w p_{li}^{(N)} q_{l1}(0) - 1 \right]^2}{\sum_{i=1}^w \pi_i \left(\frac{q_{i1}(0)}{\pi_i} - 1 \right)^2} \quad (40)$$

This is the equation derived by Inoue and Yamaguchi (1968) for the binary homogenous particle system.

IV. Experimental Procedure

The motionless mixer (Armeniades et al., 1965; Pattison, 1969) employed in this study was originally constructed by Chen et al. (1971, 1972) for their studies of the mixing of a two-component homogeneous system. A thin band of yellow brass, 0.025 in. thick and 1.5 in. wide, was twisted uniformly both clockwise and counterclockwise to make the helices. Bands of 3.25 in. length were cut to give the helices a twist of 180° . The clockwise and counterclockwise helices were inserted in a Pyrex tube of 1.5 in. inside diameter. Friction between the tube and the helices prevented the helices from slipping down the tube. Inside the tube, the helices appear alternately clockwise and counterclockwise. The schematic setup of the experiment is depicted in Figure 2.

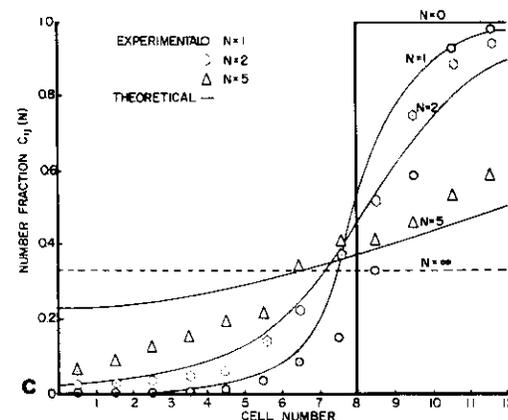
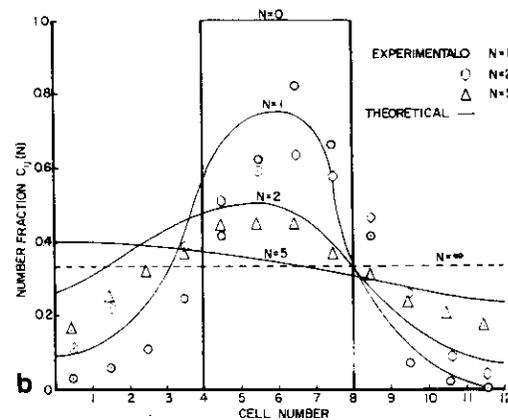
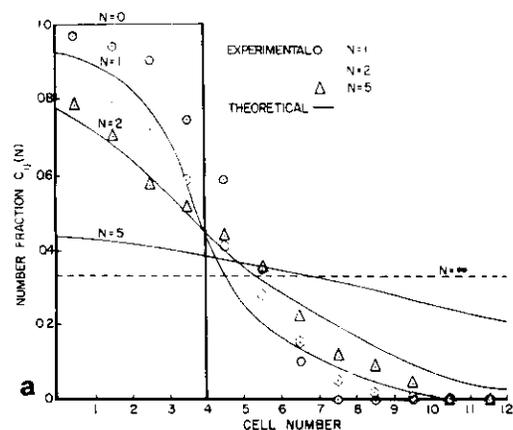


Figure 3. Number concentrations distribution of each cell: (a) component $j = 1$; (b) component $j = 2$; (c) component $j = 3$.

Spherical Lucite particles of a diameter of $\frac{5}{32}$ in. with three different colors (green, red, and white) were used in the experiments. The green particles were designated as component 1, the red particles component 2, and white particles component 3. In the pre-mix section, component 1 (green) occupied cells $i = 1, 2, 3,$ and 4 , component 2 (red) cells $i = 5, 6, 7,$ and 8 , and component 3 (white) cells $i = 9, 10, 11,$ and 12 . The particles were collected in the post-mixer section after they were passed through the mixer. The post-mixer section was placed in the pre-mix position for another pass. Determination of concentration distribution was carried out after a desired number of passes were performed.

V. Experimental Results

The experimental results are summarized in Figures 3-6. Figures 3a, 3b, and 3c show, respectively, the number con-

Table II. The Transition Matrix of the 12-Elements Motionless Mixer (Chen et al., 1971)

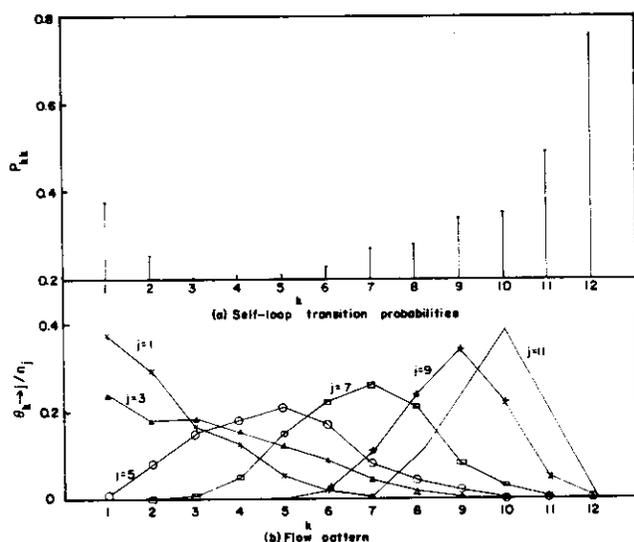
$$P = \left\| \left\| p_{ij} \right\| \right\| = \begin{bmatrix} 0.3756 & 0.3000 & 0.2400 & 0.0808 & 0.0096 & 0.0 & 0.0 & 0.0 & 0.0 & 0.0 & 0.0 & 0.0 & 0.0 \\ 0.2936 & 0.2571 & 0.1846 & 0.1692 & 0.0840 & 0.0115 & 0.0 & 0.0 & 0.0 & 0.0 & 0.0 & 0.0 & 0.0 \\ 0.1647 & 0.1807 & 0.1923 & 0.2038 & 0.1583 & 0.0910 & 0.0090 & 0.0 & 0.0 & 0.0 & 0.0 & 0.0 & 0.0 \\ 0.1250 & 0.1160 & 0.1692 & 0.2096 & 0.1885 & 0.1397 & 0.0506 & 0.0013 & 0.0 & 0.0 & 0.0 & 0.0 & 0.0 \\ 0.0519 & 0.0673 & 0.0929 & 0.1564 & 0.2109 & 0.2295 & 0.1506 & 0.0372 & 0.0032 & 0.0 & 0.0 & 0.0 & 0.0 \\ 0.0353 & 0.0372 & 0.0583 & 0.09167 & 0.1776 & 0.2288 & 0.2237 & 0.1256 & 0.0218 & 0.0 & 0.0 & 0.0 & 0.0 \\ 0.0077 & 0.0179 & 0.0237 & 0.0423 & 0.0897 & 0.1865 & 0.2699 & 0.2449 & 0.1083 & 0.0090 & 0.0 & 0.0 & 0.0 \\ 0.0026 & 0.0071 & 0.0096 & 0.0237 & 0.0385 & 0.0827 & 0.2160 & 0.2801 & 0.2410 & 0.0962 & 0.0025 & 0.0 & 0.0 \\ 0.0006 & 0.0026 & 0.0038 & 0.0128 & 0.0173 & 0.0429 & 0.0859 & 0.2058 & 0.3410 & 0.2372 & 0.0500 & 0.0 & 0.0 \\ 0.0 & 0.0 & 0.0006 & 0.0038 & 0.0019 & 0.0128 & 0.0314 & 0.0763 & 0.2199 & 0.3949 & 0.2487 & 0.0096 & 0.0 \\ 0.0 & 0.0 & 0.0 & 0.0 & 0.0 & 0.0019 & 0.0038 & 0.0135 & 0.0571 & 0.1981 & 0.4891 & 0.2365 & 0.0 \\ 0.0 & 0.0 & 0.0 & 0.0 & 0.0 & 0.0 & 0.0 & 0.0019 & 0.0083 & 0.0276 & 0.1968 & 0.7654 & 0.0 \end{bmatrix}$$


Figure 4. Self-loop transition probabilities and flow pattern in a motionless mixer.

concentration distributions of components 1, 2, and 3 in the axial direction of the mixer as a function of cell numbers. The initial number concentration distribution for each component is a step function. As the number of passes through the mixer increases, the concentration distribution approaches the final equilibrium state. It is interesting to note that the experimental results in Figures 3a-c show considerable random scattering. As pointed out by King (1968), in continuous flow systems the inherent randomness of the process will produce a stochastic output from a deterministic input.

Figure 4a shows the self-loop transition probabilities in the cells of the mixer. The schematic representation of the experimental setup for determining the transition probability p_{kj} of the particles for one pass through the mixer is shown in Figure 2. The transition probability p_{kj} can be calculated on the basis of the accumulated records of motion by the following equation.

$$p_{kj} = \frac{M_{kj}}{\sum_{k=1} M_{kj}}$$

where M_{kj} is the number of transition of one component of particles from cell ϵ_k for one pass. Since all particles are identical, except in color, we expect p_{kj} to be the same for all components of particles ($j = 1, \dots, r$). The transition probability for binary homogeneous particles thus obtained by Chen et al. (1972) is shown in Table II. The point of in-

terest is that the flow of particles in the mixer is more vigorous in the upper cells than in the lower ones. For example, the number of flow of particles in cell 1 (an upper cell) is about twice that of cell number 12 (a lower cell). In cell 1, about 30% of the particles stay in the cell after each pass and the remaining 70% of the particles are exchanged with the particles in the other eleven cells. This is in contrast to cell 12, where 75% of the particles stay in the cell and only 25% of the remaining particles are exchanged with the particles in the other eleven cells. This is consistent with the expected entrance and exit effects for cells near both ends of the mixer.

Figure 5 shows the number concentration distributions as a function of number of passes at cells 1, 2, and 3. It is noted that the concentrations fluctuate around the final equilibrium concentration for the first few passes.

Figure 6 shows the degree of mixedness as a function of the number of passes. Equation 32 can be used to determine the degree of mixedness for mixtures consisting of various numbers of components. Regardless of the number of components or the number of cells in the mixer, the degree of mixedness varies from zero in a completely segregated state to unity in a completely mixed state. For illustrative purposes, we can assume two of the three components in the tertiary homogeneous particle system to be color blind; i.e., we cannot distinguish two of the three colors. In this way, the experimental data for the resulting two-component system can be obtained from that of the three-component system without actual experimentation. Data obtained in this manner for the two-component system are also shown in Figure 6.

VI. Analysis and Discussion

The initial concentration distributions of components 1, 2, and 3, respectively, are shown in Figures 3a-c. The corresponding initial concentration matrix is

$$C(0) = \begin{bmatrix} 1 & 0 & 0 \\ 1 & 0 & 0 \\ 1 & 0 & 0 \\ 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 1 & 0 \\ 0 & 1 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \\ 0 & 0 & 1 \\ 0 & 0 & 1 \\ 0 & 0 & 1 \end{bmatrix}$$

The containers and the mixer used in this study are cylindrical in shape. The cells within the container are chosen so

particle systems, giving a single curve of theoretical predictions for the first few passes. This implies that the degree of mixedness is substantially the same for the two- and three-component systems and depends mainly on mixer and particle characteristics. This also indicates that the transition matrix (Table II) can be applied to homogeneous particle systems in the mixer regardless of the number of components present. We can further assert that the degree of mixedness of a multicomponent homogeneous particle system can be theoretically predicted from the transition matrix of the two-component system contained in it. The only experiment required in this prediction is simply the experiment of obtaining the transition matrix of the two-component system. After the mixture passes through the mixer more than five times, the discrepancies between the theoretical predictions and the experimental data increase. This suggests strongly that the segregation effect begins to dominate. A careful examination of the particles used in the experimental studies indicates that a difference in size of as much as 0.0776 in. is found in the $\frac{5}{32}$ -in. particles.

It is interesting to note that based on the concept of entropy in thermodynamics, the entropy of the solid mixture at cell i can be defined as

$$S_i(N) = - \sum_{j=1}^r C_{ij}(N) \ln C_{ij}(N) \quad (i = 1, 2, \dots, w) \quad (41)$$

and the total entropy of the system is then

$$S(N) = \sum_{i=1}^w \pi_i S_i(N) \quad (42)$$

If the system is in the segregated state at the beginning of the mixing process, we have

$$S_i(0) = 0 \quad (i = 1, 2, \dots, w)$$

and, therefore

$$S(0) = \sum_{i=1}^w S_i = 0 \quad (43)$$

The total entropy of the system as a function of the number of steps of Markov chains together with the experimental data is shown in Figure 7. Due to the segregation effect, experimental data are not entirely consistent with theoretical predictions. For a constant number of particles, the total entropy of the system at the completely mixed state will approach a constant value, i.e.

$$S(N) \longrightarrow S_f \text{ as } N \longrightarrow \infty$$

From eq 42

$$S_f = \ln r \quad (44)$$

The final entropy of the system is always greater than zero, except in the case of a unitary particle system where S_f equals zero. This indicates that the entropy can be used equally as an indicator of the state of mixedness of a mixture. The degree of mixedness according to eq 39 is based on the variances of the mixture. In using the variances, the degree of mixedness of the mixture changes from zero in the completely segregated state to one in the complete random state. A scale of the extent of mixing of any mixture can thus be defined based on these two reference limits. If we use the entropy as defined - by eq 41, the entropy of a mixture will change from zero in the completely segregated state to S_f in the completely random state. Hence, the entropy of a mixture can be similarly defined as a measure of the degree of mixedness of a mixture. Entropies computed from the experimental data obtained in this work were plotted in Figure 7 as the function of the number of passes

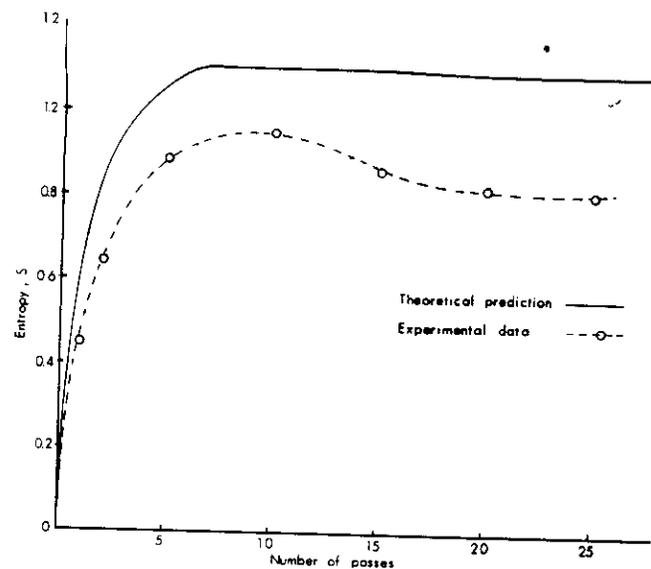


Figure 7. Increase of entropy as a result of mixing action.

through the motionless mixers. Theoretical predictions based on the model were also shown in Figure 7. It can be seen that the entropy gradually increased with the number of passes of the mixture through the motionless mixer. The experimental results were in good agreement with the theoretical predictions of the entropy of the mixture.

VII. Conclusions

A stochastic model is developed to study the mixing process of blending a multicomponent homogeneous particle system by passing it through a motionless mixer. The model can be applied to almost any class of mixer and the design complication of a mixer creates little difficulty in the use of this approach. By experimentally determining the transition probabilities of a binary homogeneous particle system, the model can predict the concentration distribution and the degree of mixedness of a multicomponent homogeneous particle mixture. This is of practical importance because the blending of multicomponent homogeneous particle systems is a necessary step in many process systems.

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Nomenclature

- $C_{ij}(N)$ = the number concentration of component j at cell i at $t = N\tau$
- $(C_j)_\infty$ = equilibrium concentration of component j
- M = degree of mixedness
- m_{ij} = the number of particles of component j in the cell i
- m_j = total number of component j in the cell i
- n = total number of particles in the mixer
- n_i = the number of particles in cell i
- p_{ij} = transition probability of the particles from cell j to cell i
- r = total number of components in the mixture
- S = total entropy of the system
- S_i = entropy in the cell i
- S_0, S_f = initial and final entropy of the system, respectively
- w = total number of cells in a mixer

Greek Letters

π_i = the number fraction of particles in cell i
 σ_N^2, σ_0^2 = mixture variances at $t = N\tau$ and $t = 0$, respectively
 ψ = variance reduction ratio

Literature Cited

Armeniades, C. D., Johnson, W. C., Raphael, T., mixing device, U.S. Patent 3,286,992 (1965).

Chen, S. J., Fan, L. T., Watson, C. A., *J. Food Sci.*, **36**, 688 (1971).
Chen, S. J., Fan, L. T., Watson, C. A., *AIChE J.*, **18**(5), 984 (1972).
Inoue, Y., Yamaguchi, K., *Kagaku Kogaku*, **33**, 286 (1969).
Lacey, P. M. C., *Trans. Inst. Chem. Eng.*, **21**, 53 (1943).
Lacey, P. M. C., *J. Appl. Chem.*, **4**, 257 (1954).
Pattison, D. A., *Chem. Eng.*, **11**, 94 (1969).
Parzen, E., "Stochastic Processes", Holden-Day, Inc., San Francisco, Calif., 1962.

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