Newell, R. B., "Multivariable Computer Control of an Evaporator," Ph.D. thesis, Univ. Alberta, Edmonton, Canada (1971).

and D. G. Fisher, "Experimental Evaluation of Op-

velop., 11, 213 (1972b).

Ogata, K., "State Space Analysis of Control Systems," Prentice-Hall, Englewood Cliffs, N. J. (1967).

Paraskos, J. A., and T. J. McAvoy, "Feedforward Computer Control of a Class of Distributed Parameter Processes,"

AIChE J., 16, 754 (1970).
Porter, B., "Assignment of Closed-Loop Eigenvalues by the Direct Method of Liapunov," Intern. J. Control, 10, 153

Rosenbrock, H. H., "State Space and Multivariable Theory," Thomas Nelson, London (1970).

Sobral, M., and R. G. Stefanek, "Optimal Control of the Linear Regulator Subject to Disturbances," I.E.E.E. Trans. on Auto. Control, AC-15, 498 (1970).

Solheim, O. A., and A. Saethre, "Optimal Control of Continuous and Discrete Multivariable Systems by Feedforward and Feedback," IFAC Conf. Multivariable Control Systems, Dusseldorf (1968).

West, H. H., and M. L. McGuire, "Optimal Feedforward-Feedback Control of Dead Time Systems," Ind. Eng. Chem. Fundamentals, 8, 253 (1969a).

"A Limitation of Predictive Controllers," paper pre-

sented at the 62nd Ann. Am. Inst. Chem. Engrs. Meeting, Washington, D.C. (1969b).
Wonham, W. M., "On Pole Assignment in Multi-Input Controllable Concerns," I.E.E.E. Trans. Auto. Control, AC-12, 660 (1967).

Manuscript received September 10, 1971; revision received March 21, 1972; paper accepted March 24, 1972.

The Mixing of Solid Particles in a Motionless Mixer-A Stochastic Approach

A Markov chain model was used to model the axial mixing of solid particles in a motionless mixer having no moving parts. One step transition probabilities were determined experimentally for the model. Based on these transition probabilities, the model was able to predict spatial distribution of tracer particles up to seven steps of the Markov chain, which was equivalent to seven consecutive passes of the mixture through the mixer. Experimental results were in good agreement with those predicted from the Markov chain model.

S. J. CHEN L. T. FAN

Department of Chemical Engineering Kansas State University Manhattan, Kansas 66502

C. A. WATSON

U.S. Grain Marketing Research Center MQRD, ARS, USDA, Manhattan, Kansas 66502

SCOPE

Solids mixing, or solids blending, is an operation by which two or more solid materials in particulate form are scattered randomly by the movement of particles in a mixer. The random movement of particles is caused by the motion of the mixer when energy is supplied to the mixer externally or by effect of gravity on the particles. Such operations are common in many industrial processes and can be easily visualized but are difficult to describe quantitatively. Empirical approaches often dominate the design and operation of a mixer. Most of the research on solids mixing has centered around the application of statistics to the mixing problem (Fan et al., 1970; Butters, 1970). Concentration profiles of constituent particles after mixing are difficult to obtain. Therefore, statistical sampling techniques are usually employed to obtain information on the final mixture. Such information is often incomplete because of bias involved in sampling.

Recently the theory of stochastic processes has been applied to analyze and to understand the mixing of solid particles. Models have been proposed to simulate the mixing of particles in some mixers to predict the concentra-

the mixing of solid particles in a drum mixer but did not conduct experiments to verify the model. Inoue and Yamaguchi (1969) and Yamaguchi (1969) proposed models of Markov chains to describe solids mixing in a two dimensional V-mixer and in a pan mixer. Mixing experiments were conducted to obtain transition probabilities using a single particle tracing technique. Transition probabilities were difficult to obtain experimentally. However, the concentration profiles of tracer particles predicted by the model based on the experimentally determined transition probabilities were in good agreement with the measured profiles. Extension and application of the results of the two dimensional model to the actual three dimensional mixer, however, is open to question. Oleniczak (1962) postulated a Poisson process for interchange of particles between a volume element and the rest of the mixture. He obtained a stochastic model for the V-mixer. The distribution of tracer particles was found to be bimodal at a low number of revolutions. As the number of revolutions was increased to 30, the bimodal distribution approached the normal distribution. The model did not apply when the number of revolutions exceeded 30.

tion profiles of constituents of the mixture. Oyama and

Ayaki (1956) proposed a Markov chain model to describe

S. J. Chen is with Kenics Corp., One Southside Road, Danvers, Mass. 01923.

The objective of the present work is to study the axial mixing of solid particles in an unconventional motionless mixer by applying the theory of Markov chains. Transverse mixing of solid particles in the mixer was reported

previously (Chen et al., 1971). The mixer can be used as a continuous mixer or as a semibatch mixer. Unlike conventional mixers, the mixer has no moving parts. In this study solid particles passed through the mixer under gravity.

CONCLUSIONS AND SIGNIFICANCE

A homogeneous Markov chain model can be used to describe the axial mixing of solid particles in a motionless mixer. The model is able to predict the distribution of the tracer particles after mixing. In such a mixer, axial mixing is far less appreciable than the transverse mixing. This information is important when the mixer is used as a continuous inline mixer. In this case maximum transverse mixing with some axial mixing is often desired to ensure precise process control and uniform property of the mixture.

One-step transition probabilities of the Markov chain model were determined experimentally. Mixing mechanisms in the mixer were observed by means of high speed photography. To verify the model, semi-batch operations of the mixer were performed. Distributions of the tracer particles after their passage through the mixer were determined and compared with those calculated from the model. They were in good agreement. The model, while still rudimentary, appears to describe adequately the complex mixing process in the mixer. In the present study distributions of the tracer particles were simulated up to

seven steps of the Markov chain, which were equivalent to seven consecutive passes of the mixture through the mixer. Considerable static electricity was generated on the particles after seven passes. This gave rise to deviations of the prediction from the experimental results.

Unlike the deterministic approach which has been employed mainly to describe the axial mixing of solid particles in a drum mixer, the stochastic approach makes no assumptions as to the use of mathematical equations in the development of the model. Mathematical intractability was thus avoided. End effects were automatically taken into account in the model. In this respect the stochastic approach seems to be a better approach than the deterministic approach in describing the mixing of solid particles in a mixer. While the determination of the transition probabilities through experiment is time-consuming, faster sampling techniques could expedite the determination and improve the accuracy of the model. A similar approach can be used to describe the mixing of particles in a drum mixer, ribbon blender, and Z-Blade mixer.

A MARKOV CHAIN MODEL FOR THE MOTIONLESS MIXER

Consider the experimental scheme shown in Figure 1. Particles in section A-B are to be mixed when they flow under gravity through the section of the motionless mixer B-C. The mixture is to be collected in section C-D which is of the same length as section A-B. This mixing process is equivalent to mixing the particles in section A-B by passing the mixer through the section assuming that the mixing mechanisms can be reproduced. Sections A-B and C-D are divided into r segments. These segments are considered as the states of a Markov chain. After each step of a Markov chain a particle either stays in the same segment or moves to any of the r segments. The particle whose position is observed after each step will be called the tracer particle. Using a tracer particle many times in repeated experiments may be considered equivalent to using many tracer particles in one experiment. A step of a Markov chain for the mixing process is considered as one pass of the particles through the mixer under gravity. A homogeneous Markov chain is used to describe this mixing process. Mathematically it is written as (Parzen, 1962)

$$p_j(n) = \sum_{i}^{r} p_i(0) p^{n_{ij}}, \quad j = 1, 2, ..., r$$
 (1)

where $p_i(0)$ = initial probability that the tracer particle is in state i and p_{ij} = probability of a transition from state i to state j. Equation (1) can be written in vector and matrix notation as

$$p(n) = p(0) P^n (2)$$

where

$$p(n) = [p_1(n) \ p_2(n) \dots p_r(n)]$$
 (3)

$$p(0) = [p_1(0) \ p_2(0) \dots p_r(0)] \tag{4}$$

$$P = \begin{pmatrix} p_{11} & p_{12} & \dots & p_{1r} \\ p_{21} & p_{22} & \dots & p_{2r} \\ \vdots & \vdots & \ddots & \vdots \\ \vdots & \vdots & \vdots & \vdots \\ p_{r1} & p_{r2} & \dots & p_{rr} \end{pmatrix}$$
 (5)

If a slug of tracer particles is placed to occupy the whole *i*th segment before mixing, then the *i*th component of the initial probability vector is

$$p_i(0) = \frac{n_i}{n_i} = 1 \tag{6}$$

where n_i is the total number of particles which occupy one segment. Thus

$$p(0) = [0 \ 0 \dots 0 \ 1 \ 0 \dots 0] \tag{7}$$

Substitution of Equation (7) into Equation (2) gives the probability distribution of the tracer particles in all the segments in the mixer after any number of passes through the motionless mixer. Since n_i tracer particles occupy the whole segment i before mixing, the number of tracer particles in segment i after n steps is

$$m_i(n) = n_i p_i(n), \quad i = 1, 2, ..., r$$
 (8)

Substituting Equation (1) into Equation (8) yields

$$m_i(n) = n_i \sum_{i=1}^r p_i(0) p_{ii}^n, \quad i = 1, 2, ..., r$$
 (9)

The final distribution of the tracer particles at any step of the Markov chain can be calculated from Equation (1)

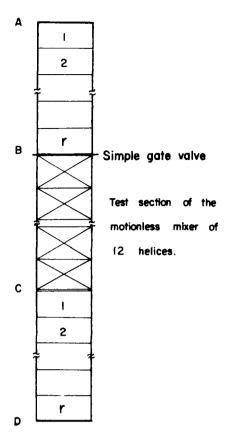


Fig. 1. Schematic setup of the experiment for determining the transition probabilities.

and the actual number of tracer particles in each segment from Equation (9). The Markov chain is completely determined if the initial and transition probabilities are given. The transition matrix, represented by Equation (5), possesses several properties. The diagonal elements of the matrix are self-loop transition probabilities, that is, the probabilities of a particle to remain in the same state after a transition. Row elements of the matrix are transition probabilities of a particle to leave the state corresponding to the row number. Column elements of the matrix are transition probabilities of a particle to enter the state corresponding to the column number. Figure 2 summarizes these properties of the transition matrix of a system with r states.

EXPERIMENT

The Motionless Mixer

A motionless mixer similar to one reported recently (Armeniades et al., 1965 Pattison, 1969) was constructed for this study. 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. in length were cut to give the helices a 180° twist. 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 from the tube. A photograph of the mixer of two helices is shown in Figure 3.

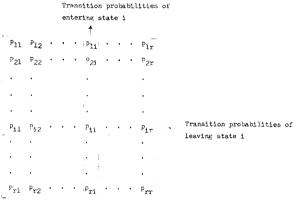
A stream of solid particles flowing through the tube under the effect of gravity was split in two streams and twisted clockwise by the first helix. At the entrance to the second helix each split stream was split again and was mixed with the split stream of the other stream with the mixed streams twisted counterclockwise. This behavior of the stream of particles repeated itself for each two helices. Although the mechanism of shear mixing may not be as prominent as it is in mixing of fluid forced through the mixer, it does occur as a result of the difference in velocities of particles flowing along the surfaces of helices. It was observed by using high speed photography that there are several mixing mechanisms operating in the motionless mixer studied. They are

- 1. multiple divisions and combinations of the flow of particles.
- 2. interaction of the particles among themselves and interaction of the particles with the helices and with the wall of the mixer,
 - 3. change in the direction of the flow of particles, and
 - 4. difference in the velocities of particles.

These mechanisms cause the random movement of the particles.

Determination of the Transition Matrix for the Model

A schematic representation of the experimental set-up for determining the transition matrix is shown in Figure 1 as mentioned previously. The premixing section had the same length as the postmixing section. The premixing section was separated from the mixer section by a simple gate valve. Each section



> Self-loop transition probabilities

Fig. 2. Summary of the properties of the transition matrix

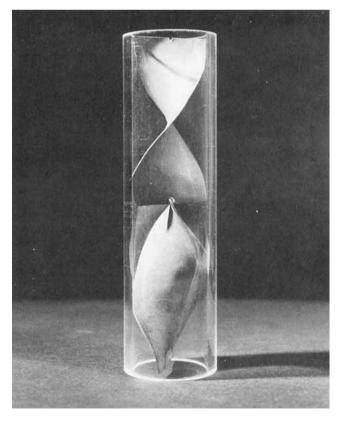


Fig. 3. The motionless mixer of two helices.

0.3756	0.3000	0.2400	0.0808	0.0096	0.0	0.0	0.0	0.0	0.0	0.0	0.0
0.2936	0.2571	0.1846	0.1692	0.0840	0.0115	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
0.1250	0.1160	0.1692	0.2096	0.1885	0.1397	0.0506	0.0013	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
0.0353	0.0372	0.0583	0.0916	0.1776	0.2288	0.2237	0.1256	0.0218	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
0.0026	0.0071	0.0096	0.0237	0.0385	0.0827	0.2160	0.2801	0.2410	0.0962	0.0025	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	0.0006	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	0.0019	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	0.0019	0.0083	0.0276	0.1968	0.7654

was one foot in length, and was graduated every inch along the section. Every inch of the section was considered as a segment or as a state of the Markov chain. The motionless mixer of 12 helices was installed and used in the test section because the highest transverse degree of mixedness could be achieved as reported previously (Chen et al., 1971). Therefore, only the axial spread of the particles needs to be considered in this study. Spherical Lucite particles of 5/32 in. were used. Tracer particles were identical to the rest of the particles except for color. 520 particles occupied one segment. To determine the transition probabilities p_{ij} , $j=1,2,\ldots,r$, tracer particles were placed at segment i in the premixing section. The rest of the premixing sections were occupied by nontracer particles. The simple gate valve was opened to let the particles pass through the mixer section. The particles remained in a fixed position after they passed through the mixer section. Tracer particles which occupied one segment before mixing were dispersed among other particles after mixing. Particles in each segment of the postmixing section were removed by suction to avoid disturbance to the whole mixture. Tracer particles were separated and counted. The one-step transition probability is the fraction of the tracer particles originally in segment i, which moved to segment j (j = 1, ..., r) after one pass through the mixer section. It can be written as

$$p_{ij} = \frac{n_{ij}}{n_i}, \quad i = 1, ..., r$$
$$j = 1, ..., r$$
$$n_i = \sum_{j=1}^r n_{ij}$$

Twelve similar one-step experiments had to be conducted to determine all transition probabilities. The experiment was repeated three times to obtain the average values of the transition probabilities. Table 1 presents these one-step transition probabilities.

An Experiment for the Markov Chain of Several Steps

An experiment was conducted to illustrate the Markov chain of several steps. The tracer particles were placed to occupy the whole 7th segment in the premixing section. After the particles passed through the mixer section, they remained in a fixed position. This postmixing section could be exchanged with the premixing section (without disturbing the spatial distribution of the particles) and the particles could then be allowed another pass through the mixer section. Each pass corresponded to one step of the Markov chain. Sampling was done after the desired number of passes was performed. The same sampling procedures as used in determining the transition probabilities were followed after each step of the Markov chain. The distributions of the tracer particles for seven steps of the Markov chain are shown as solid lines in Figures 4 through 9.

RESULTS AND DISCUSSIONS

It is desirable to compare the distributions of the tracer particles determined experimentally with those calculated from Equation (2) in which the experimentally obtained one-step transition matrix appears. The initial distribution vector is

$$p(0) = [0 \ 0 \ \dots \ 0 \ 1 \ 0 \ \dots \ 0]$$

7th state

It can be seen from Equation (2) and the above equation that the 7th row of the matrix P^n is the distribution of the tracer particles after the nth step. If the tracer particles are uniformly distributed, the uniform concentration in

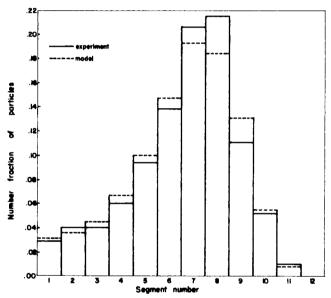


Fig. 4. Results of the 2nd step.

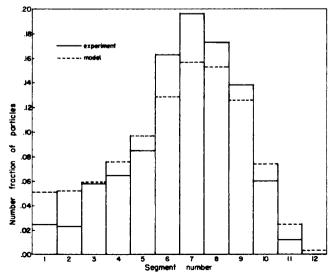


Fig. 5. Results of the 3rd step.

each segment is 1/12. Dotted lines in Figures 4 through 9 are the computed values. Computation of P^n was performed by using an IBM 360 computer in double precisions. The calculated results are in good agreement with the experimental data. However, further steps of the Markov chain showed deviation from the experimental results. This was probably caused by a considerable amount of static electricity generated after a few consecutive steps were performed, and/or by the fact that only three repeated experiments were conducted to determine the transition matrix. To obtain good agreement for further steps, it may be necessary to remove static electricity and to perform additional repeated experiments to determine the transition matrix. To determine the fluctuations of the distributions of the tracer particles in each segment after the repetitive mixing, the variances were calculated by using the following equation.

$$s^{2}(n) = \frac{1}{r} \sum_{j=1}^{N} \left(p_{j}(n) - \frac{1}{r} \right)$$
 (10)

If the tracer particles occupy one whole segment before mixing, Equation (10) can be rewritten by using Equations (2) and (7) as

$$s_i^2(n) = \frac{1}{r} \sum_{j=1}^r \left(p^{n_{ij}} - \frac{1}{r} \right)$$
 (11)

Table 2 shows the fluctuations of the distribution of tracer particles. Numbers in columns n are steps of the Markov chain and those in columns i are the segment numbers in which the tracer particles are placed before mixing. Columns of $s_i^2(n)$ provide the corresponding variances. The minimum variance for each step is denoted by a circle on the segment number. The minimum variance indicates that given a step number of the Markov chain a better mixture is obtained if the tracer particles are placed in the state indicated by the circle than if the tracer particles are placed in any other states. For the present experiment in which the tracer particles were placed in segment 7 before mixing, Table 3 presents the variances calculated from Equation (11) and the variances calculated from the experimental data. The variances decrease as the number of steps increase to spread the tracer particles. As explained previously, steps beyond seven increased deviation of the model from the experimental results. Moreover, the experimental variance for the 7th step was larger than that of the 6th step.

As reported previously (Chen et al., 1971), transverse

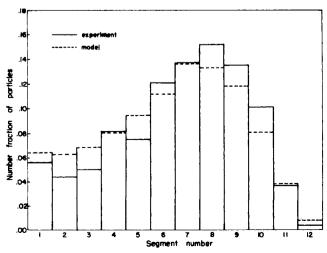


Fig. 6. Results of the 4th step.

mixing of solid particles in the motionless mixer can be readily achieved in one pass through the mixer when the particles to be mixed are fed parallelly into the mixer. Although the mixture of particles was passed through the mixer up to seven times consecutively, axial spread of the tracer particles did not increase appreciably. Figures 4 through 9 show that a distinct bell shaped curve was ob-

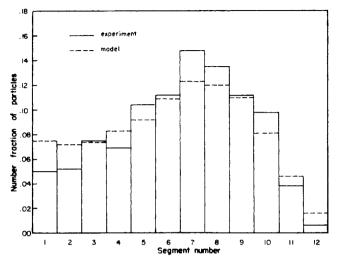


Fig. 7. Results of the 5th step.

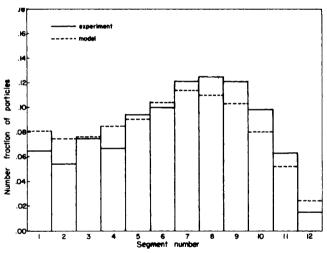


Fig. 8. Results of the 6th step.

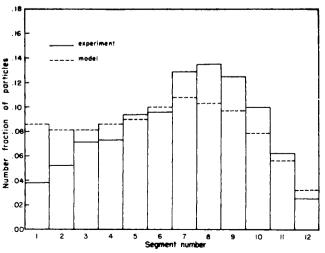


Fig. 9. Results of the 7th step.

Table 2. Variances and the Minimum Variances of Tracer Particles After Mixing

n	i	$s_i^2(n)$	n	i	$s_i^2(n)$	n	i	$s_i^2(n)$	n	i	$s_i^2(n)$
	1	1.74×10^{-2}		1	7.39×10^{-3}		1	4.60×10^{-3}		1	3.21×10^{-3}
	2	1.16×10^{-2}		2	6.11×10^{-3}		2	4.01×10^{-3}		2	2.87×10^{-3}
	3	7.37×10^{-3}		3	4.38×10^{-3}		3	3.11×10^{-3}		3	2.32×10^{-3}
	4 5	6.33×10^{-3}		4	3.46×10^{-3}		4	2.54×10^{-3}		4	1.94×10^{-3}
	5	6.52×10^{-3}		5	2.34×10^{-3}		5	1.63×10^{-3}		5	1.25×10^{-3}
1	6	6.78×10^{-3}	3	6	1.97×10^{-3}	5	6	1.13×10^{-3}	7	6	8.00×10^{-4}
	7	8.91×10^{-3}		7	$2.25 imes10^{-3}$		Ø	8.75×10^{-4}		7	4.20×10^{-4}
	8	9.84×10^{-3}		8	$2.80 imes 10^{-3}$		$\check{8}$	9.75×10^{-4}		8	3.40×10^{-4}
	9	1.20×10^{-2}		9	3.64×10^{-3}		9	1.49×10^{-3}		9	6.89×10^{-4}
	10	1.58×10^{-2}		10	5.59×10^{-3}		10	3.20×10^{-3}		10	2.14×10^{-3}
	11	2.12×10^{-2}		11	1.16×10^{-2}		11	8.45×10^{-3}		11	6.24×10^{-3}
	12	4.52×10^{-2}		12	2.49×10^{-2}		12	1.64×10^{-2}		12	1.15×10^{-2}
	1	1.03×10^{-2}		1	5.71×10^{-3}		1	3.81×10^{-3}			
	2	8.02×10^{-3}		2	4.88×10^{-3}		2	3.37×10^{-3}			
	3	5.39×10^{-3}		3	3.66×10^{-3}		3	2.67×10^{-3}			
	4	4.25×10^{-3}		4	2.94×10^{-3}		4	2.21×10^{-3}			
	5	3.29×10^{-3}		5	1.90×10^{-3}		5	1.42×10^{-3}			
2	6 7	3.19×10^{-3}	4	6	1.43×10^{-3}	6	6	9.38×10^{-4}			
	7	4.08×10^{-3}		7	1.36×10^{-3}		7	5.92×10^{-4}			
	8	4.91×10^{-3}		8	$1.65 imes 10^{-3}$		8	5.77×10^{-4}			
	9	6.10×10^{-3}		9	2.29×10^{-3}		9	9.96×10^{-4}			
	10	8.53×10^{-3}		10	4.09×10^{-3}		10	2.59×10^{-3}			
	11	1.42×10^{-2}		11	9.85×10^{-3}		11	7.26×10^{-3}			
	12	3.23×10^{-2}		12	2.00×10^{-2}		12	1.37×10^{-2}			

Table 3. Variances Obtained by Using Equation (10) and by Using Experimental Data

Steps							
Variances	1	2	3	4	5	6	7
Variances from the model	8.91×10^{-3}	4.08×10^{-3}	2.25×10^{-3}	1.36×10^{-3}	8.75×10^{-4}	5.92×10^{-4}	4.20×10^{-4}
Variances from experimental data	9.54×10^{-3}	4.79×10^{-3}	4.22×10^{-3}	2.42×10^{-3}	1.65×10^{-3}	9.83×10^{-4}	1.18×10^{-3}

tained. There is much less axial mixing in the mixer than the transverse mixing. This information is important when the mixer is used as an inline continuous mixer. In this case maximum transverse mixing with some axial mixing is often desired to ensure precise process control and uniform quality of the mixture.

ACKNOWLEDGMENT

Financial support provided by the Agricultural Research Service, United States Department of Agriculture, is gratefully acknowledged.

NOTATION

 $m_i(n)$ = number of tracer particles in segment *i* after *n* steps

= number of particles which occupy one segment

= number of tracer particles found in segment j that came from segment i in one step

= transition matrix

p(n) = probability vector after n steps of the Markov

 $p_i(n)$ = probability that the tracer particle will be in the state *j* after *n* steps

= probability of transition from state i to state j in

= ith row jth column element of the transition matrix Pn

= number of states or segments

 $s^2(n)$ = variance of the tracer particles after n steps of the Markov chain

 $s_i^2(n)$ = variance of the tracer particles after n steps of the Markov chain in which segment i is occupied only by the tracer particles before mixing

LITERATURE CITED

Armeniades, C. D., et al., "Mixing Device," U.S. Patent No.

3,286,992 (1965).

Butters, J. R., "Recent Development in the Mixing of Dry Solids," Brit. Chem., 15, 41 (1970).

Chen, S. J., et al., "Effect of Handling Methods on Bulk Vol-

ume and Homogeneity of Solid Materials," J. Food Sci., 36, 688 (1971).

Fan, L. T., et al., "Solids Mixing-Annual Review," Ind. Eng. Chem., 62, 53 (1970).

Inoue, I., and K. Yamaguchi, "Particle Motion in Mixer and Mixing Process—Mixing in a Two-Dimensional V-Type Mixer," Kogaku Kogaku, 33, 286 (1969).

Oleniczak, A., "A Stochastic Model of Ideal Dry Particle Mixing," Ph.D. thesis, Princeton Univ., N. J. (1962).

Oyama, Y., and K. Ayaki, "Studies on the Mixing of Particulate Solids," Kogaku Kogaku, 20, 148 (1956).

Parzen, E., Stochastic Processes, Holden-Day, San Francisco,

Pattinson, D. A., "Motionless Inline Mixer Stir up Broad Interest," Chem. Eng., 11, 94 (1969).

Yamaguchi, K., "Analysis of Flow in Solid Mixing," J. Res.

Assn. Powder Tech. (Japan), 6, 292 (1969).

Manuscript received November 22, 1971; revision received May 16, 1971; paper accepted May 17, 1972.