THE MIXING OF POWDERS

IN THE HORIZONTAL ROTATING DRUM MIXERS

BY

PAUL CHEUNG MAU YEUNG

SUBMITTED FOR A DEGREE OF DOCTOR OF PHILOSOPHY

OF LOUGHBOROUGH UNIVERSITY OF TECHNOLOGY

1968

SUPERVISOR :

P. J. LLOYD

DIRECTOR OF RESEARCH :

PROF. D.C. FRESHWATER

SUMMARY

The literature published on powder mixing is reviewed and the conclusion drawn that the operation is misunderstood and the situation confused because insufficient attention has been paid to the properties of particles being mixed. As an aid to simplication, powders are classified as free flowing powders, where the particle motion is individual and non-free flowing powders, where the mixing is more concerned with the shuffling and subsequent breaking down of large agglomerates.

This thesis considers the mixing of free flowing powders only. A 'diffusion' type equation is obtained by stochastic analogy after two postulates are made. Thses postulates are concerned with the packing of the particles and the momentum transfer. The application of this equation to four cases are discussed in relation to these postulates. The four cases are

(1) The mixing of components consisting of physically identical particles

(2) The mixing of components of same density and different particle size

(3) The mixing of components of same particle size but different density

(4) The mixing of components of different density and particle size.

The results of a large series of experiments of mixing in horizontal drum are presented in the discussion of these four cases. The effect of mixer dimensions and the speed of rotation is further discussed in relation to the mixing of physically identical particles. When particles of different size and/or density are mixed segregation occurs. It is possible to predict the type of segregation that will occur and in some cases to predict the correct operating condition which will enable a pseudo-randomisation of the mixture obtained.

ACKNOWLEDGEMENT

CONTENTS

CHAPTER   ONE

INTRODUCTION

The mixing of powders is one of the oldest unit operations. Powders have been mixed to a line scale of scrutiny in many fields of technology over many hundreds of years and one might expect that the operation would be well understood. Yet it is an operation frequently neglected in textbooks on Chemical Engineering because in most industrial applications, the mixing of powders is an art more than a science.

One of the reasons why the technology of powder mixing is not as advanced as it might have been is due to the complexity of the operation. Powders can consist of materials of widely different densities and particle sizes. Available powder mixers show as much variation in their geometry and method of impelling the material. To put the wide range of particle properties into a diverse selection of machinary and expect a simple unified theory is perhaps asking a little too much. Furthermore the question of how one can assess the progress of the mixture has lead to a multitude of different methods. All using statistics as a reasonable method of handling the bulky experimental data. Unfortunately the arguments over which statistic gives the more reliable assessment has led to the overlooking of the process which is proceeding within the machine.

Attempts have been made to describe the mechanisms of powder mixing, but here again one can be easily mislead by taking the obvious analogy that powder mixing is like liquid mixing and mechanisms which apply in liquids should apply in powders. This is not so, as a powder consists of small particles and by this essence is very different from a liquid.

The aim of this study is to study powder mixing as a particulate problem and by so doing put a little order into the confused state of knownledge. There are many possible approaches to study this problem. The most direct one is to consider the fundamental property of the materials and to classify the powders to be considered under this heading. Powdered materials can be generally classified into free flowing powders, in which the interparticle forces are small compared with the inertia forces and the particles can be considered as separate entities and non-free flowing or cohesive powders, where interparticle forces can not be ignored. This study is concerned with the first class of powders.

Since in free flowing powders, the particles can move individually, the movement of the particles can be considered by a stochastic analogy. Assumptions have to be made to make the analogy applicable but deviation from the degenerate case enabel a closer insight into the behaviuor of the mixtures to be obtained. It is hoped that this approach will provide a better understanding of the mechanisms of mixing and segregation of free flowing powders.

The experimental work consisted entirely of a study of the simplest powder mixer – the horizontal rotating drum, since the picture of mixing mechanisms can be confused by using to complicated a machine. The experimental results both qualitative and quantitative are presented in full detail since they might be of use in developing new and better theories. This is felt to be important because as is seen in the literature survey, too much experimental evidence has been proposed which can not be used to test new theories of mixing because insufficient data has been presented.

CHAPTER TWO

A LITERATURE SURVEY

2.1. INTRODUCTION

The measurement of mixing progress is generally described
by a degree of mixing or a mixing index of which many forms have
been proposed. No general form, however, has been proposed which
can cover all kinds of powder mixing. The concept of a degree of
mixing is necessary to obtain a degree of comparison between
mixers in a particular system, but has it been necessary for each
investigator to propose a new definition?

The rate of mixing has been considered as first order
differential equation by a majority of investigators, but others
have proposed a second order relation. So the reported experimental
results can not be summarised in a universal rate equation re-
presenting all the mechanisms of powder mixing.

Attempts to investigate the mechanisms of powder mixing
are important for many reasons. Firstly, if the mixing mechanism
was known and understood, it should lead to a method for calculat-
ing the mixing time in various mixers. The amount of time necessary
for testing could be then reduced considerably, and the final
states of mixedness predicted. Secondly, the rate of mixing is
dependent on the mechanisms of mixing, therefore, a study of these
mechanisms should point the way in which machine and technique
could be improved. However, the published literature is very
deficient on this point.

Although many papers have been published on the mixing of
powders,   comparison between the work of different investigators
is difficult because the results are very dependent on the material
used and the techniques employed. It is difficult for one to survey
all the literature in a form which would cover all aspects of
powder mixing, e.g., degree of mixing, theory of mixing, experi-
mental work, etc. Therefore, for convenience, the survey has
reviewed the literature under three headings, namely, the degree
of mixing and experimental details, the theory of mixing-rate and
mechanisms, and the equipment and materials used. These are
presented in chronological order. It is hoped that the survey
will provide an easier form for the comparison of different
proposals from the available literature, without adding further
confusion. Since most of the work done by Japanese researchers

had not been reviewed previously (mainly because it was written
in Japanese), a more detailed discussion of this work is given.

2.2. THE DEGREE OF MIXING AND EXPERIMENTAL DETAILS

Oyama published a series of papers from 1933, entitled
'the motion of granular or pulverulent materials in a horizontal
rotating drum'. These studies contained the investigations of the
motion and mixing of granules in the horizontal rotating drum. In
here, only those papers regarding to the investigation of mixing
will be reviewed,

Initially, Oyama (57) studied the mixing with components
of equal sized granules only distinguished by colour, in a hori-
zontal cast iron drum (200mm I.D., 400mm in length) on which a
glass panel was fixed on one end. The drum was driven by a vari-
able speed electri motor. Black and white limestone granules (1.5mm
dia.) were loaded in such a way that gave two parallel layers, one
on top of the other. On starting the drum, the progress of mixing
was recorded with a cine camera which was placed on outside of the
glass panel. The cine film which recorded the motion of the granu-
les, was used for analysing the experimental results. A light beam
was projected through the film to a phototube which in turn was
connected to a fibre electrometer. With moving of the film at a
steady speed, the movement of the granules could be projected on
to the phototube. The movement of the fibre of the electrometer
recorded the changing intensity of the film and was recorded con-
tinuously.

A formula was given for the intensity of light received
by the film from a group of granules,

$$1 = \frac{1}{t} \left(\frac{0}{0'}\right)^{1/r} \tag{2.1}$$

Where,   1 = intensity of light,

0 = opacity of the photographic image,

0' = opacity of the fog (background),

t = time of exposure, and

r = a constant depending on the film used and the
developong conditions (for Oyama's case was 1.53)

Thus the intensity of light in terms of progress of mixing could be measured. This may be seen in Fig.(2.1). Before the mixing
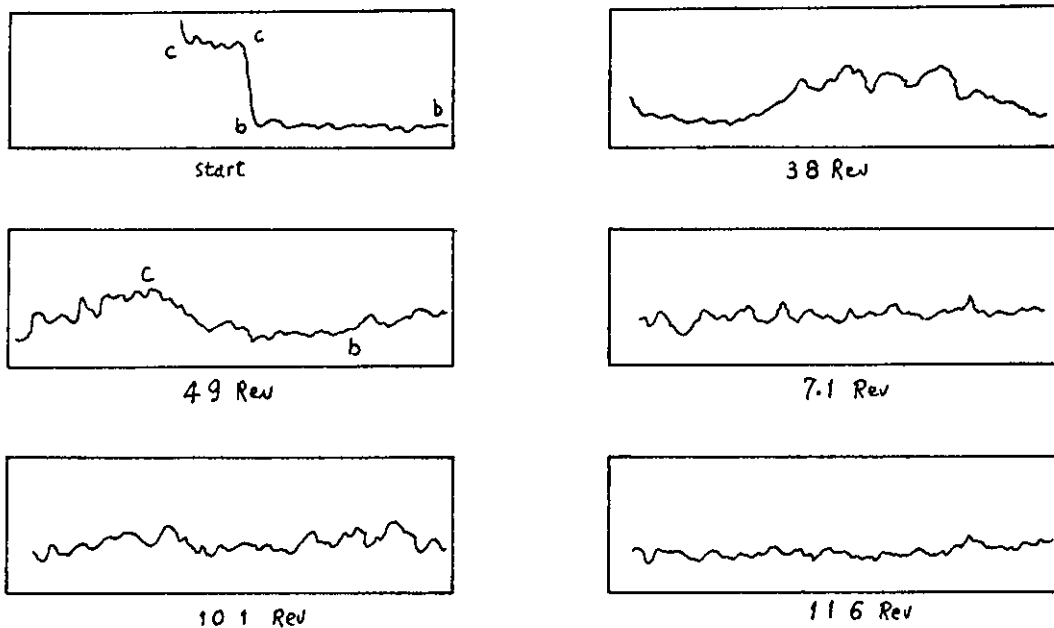

Fig.(2.1) The changes of intensity of light as mixing progress.

started, it had two parts, one due to the black granules which was loaded on the bottom, and one due to the white ones on the top. A sharp jump occurred in the gap as the light passed through the borderline between the two layers. At 4.9 revolutions, the difference between band c was less than that for the initial state. Also the layer number of the small peaks and valleys as compared to the initial state indicated that alternate layers of differently coloured granules were beginning to form. At any speed of rotation, however, the intensity became more uniform . For the purpose of comparing the degree of mixing at each speed of rotation the formula for the degree of mixing was proposed as;-

$$M = 1 - \frac{1_{max\ c}}{1_{max\ c_0}} \qquad (2.2)$$

Where, M = degree of mixing,

$1_{max\ c}$ = intensity of light i corresponding to the maximum density difference,

$1_{max\ c_0}$ = intensity of light i corresponding to the pre-rotation maximum density difference.

The degree of mixing covered the ranges from the initial state $(M = 0)$ to the complete mixed state $(M = 1)$.

The relationships between i and revolutions of drum at different states were shown in Fig.(2.2). Fig.(2.3) shows that the relationships between degree of mixing and mixing time at different speeds of rotation.
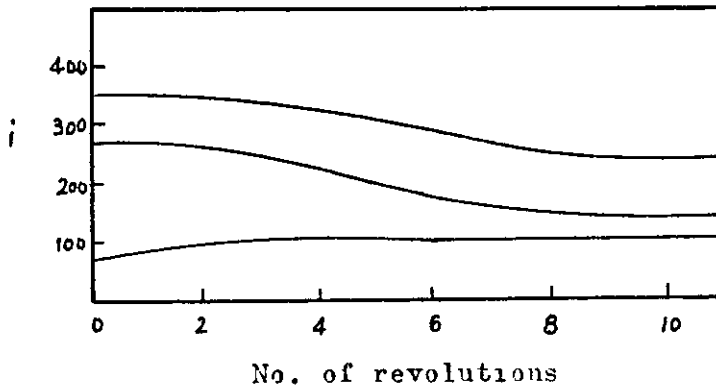
Fig.(2.2) The relationships between i and revolutions.

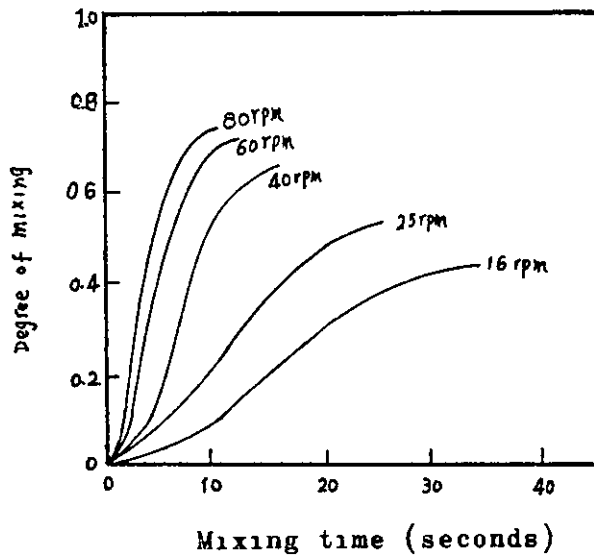Fig.(2.3) Degree of mixing vs. time of mixing.

Oyama concluded that at higher speed of rotation, up to 80rpm, a higher degree of mixing was obtainable, as could be seen in Fig.(2.3). The most satisfactory results were obtained when the speed of rotation lay between the equilibrium state and the critical state which was around 60rpm. The proposed degree of mixing only applied to the observed end plane.

Later, Oyama (58, 59) investigated mixing of granules with different sizes, the degree of mixing was developed on the basis of the packing theory. The degree of mixing for two differently sized granules ~~mixture~~ involved a comparison of the specific volumes of the individual materials, with that obtained when the two materials were packed as closely as 'practically attainable'. The relation between specific volume and weight percent of the two components was plotted as in Fig.(2.4) in accordance with the data determined by Westman and Hugill (86), who had used a specially designed machine to determine the minimum value attainable, this could be represented by the straight line AB in Fig.(2.4), and this line may be written by an equation as,

$$V_s = \frac{x}{S_1(1 - v_1)} \qquad (2.3)$$

Where, V_s = specific volume of the mixture,

S_1 = true specific gravity of the large particles,

v_1 = fraction of voids of large particles,

x = weight fraction of large particles.

Fig.(2.4)

It was supposed that a weight fraction x of large particles is introduced and distributed at random, through the bed of fixed components without leaving any voids between the adjacent large particles. Then, for a unit weight of the system, the specific volume was represented by a straight line CBD, or

$$V_s = \frac{1 - x}{S_2(1 - v_2)} + \frac{x}{S_1} \qquad (2.4)$$

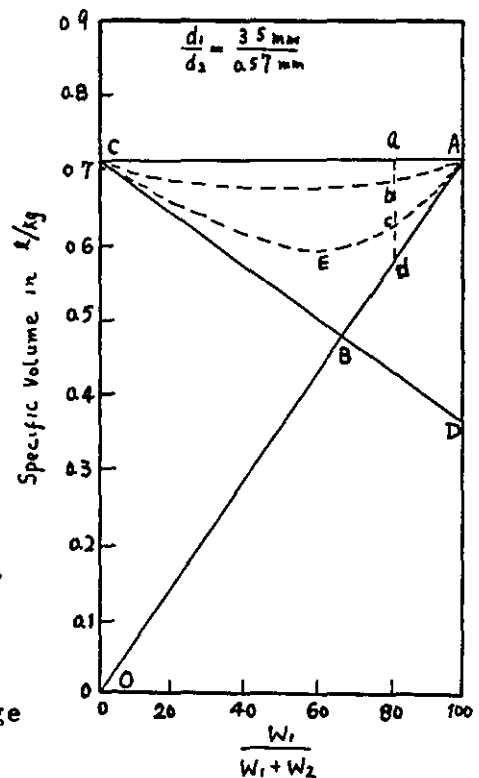If two components of different sizes were introduced

separately into a container without action of mixing, the total volume would be the sum of each individual apparent volume. So that the specific volume could be expressed by an equation as,

$$V_s = \frac{x}{S_1(1 - v_1)} + \frac{1 - x}{S_2(1 - v_2)} \qquad (2.5)$$

or the line AC in Fig.(2.4).

(Subscript 1 referring to the large particles and 2 to the small particles)

In order to obtain an equation which represented the specific volume of packing of a binary mixture, having any diameter ratio, Oyama developed an empirical equation from a general equation which was based on the theory informed by Westman and Hugill. That the general equation is,

$$X^2 + 2GXY + Y^2 = 1 \qquad (2.6)$$

When $G = 1$, Eq.(2.6) becomes,

$X + Y = 1$

$G = \infty$ Eq.(2.6) becomes,

$XY = 0$, G is a function of diameter ratio.

By making appropriate substitutions for X and Y, from Eqs.(2.3), (2.4) and (2.5), then Eq.(2.6) could be converted into an equation which becomes Eq.(2.5) for $G = 1$, and either equations of (2.4) or (2.5) for $G = \infty$. Consequently X and Y obtained,

$$X = S_2(1 - v_2)(V_s - \frac{x}{S_1(1 - v_1)})$$

$$Y = \frac{S_1(1 - v_1)}{v_1} (V_s - \frac{x}{S_1} - \frac{1 - x}{S_2(1 - v_2)})$$

for Eq.(2.6).

The degree of mixing was determined from Fig.(2.4), if (ac) is the difference between 'non-mixed' specific volume and closest packed specific volume, and (ab) the difference between 'non-mixed' specific volume and mixed specific volume, the the degree of mixing could be defined as,

$$M = (ab)/(ac) \times 100 \ (\%) \qquad (2.7)$$

The experimental work was performed in the same drum mixer which was used in the previous paper (57), however, the materials for this studies involved three different sizes, 3.5mm, 1.3mm and 0.57mm of limestone granules.

Oyama concluded from the experimental results that the highest degree of mixing would occur at 60-80rpm regardless of the size ratio of particles or the proportion of each component. At this speed the state of motion of particle changed from 'cascade' to 'cateract' and may be called the 'critical' state. A further increase in speed of rotation to that corresponding the equilibrium state at 88rpm lowered the degree of mixing. The degree of mixing is also affected by the weight ratio of two components. It was seen that a higher degree of mixing would be obtained when the weight ration approaching the extreme values of zero and infinity, and the poorest value when the proportion was unity.

Continuing these experiments in the same horizontal rotating drum mixer, Oyama (58, 60) found that separation of the granules occurred after a certain time of mixing. This separation phenomena was observed after the drum was stopped, the mixing bed had appeared several clear strips (the author did not give clear description of the strips, whehter they were formed by large or small granules, it is understood that the 'strip' has the same meaning as the 'band' discussed by Roseman (69)). The strips were formed perpendicular to the axis of the drum and appeared to be a change in structure of the granular material. The structure, number and the widths of these strips was very dependent on the operating conditions, but they were reproducible under the same conditions. The strips soon appeared and became more sharp and stable as the mixing continued. The strips even appeared when two components well mixed before the experiment were rotated in the drum. The pattern of these strips was affected by the weight ratio of the two components, but not the total weight of the materials being mixed.

Oyama concluded that the mixing mechanisms of two components of different sizes are largely influenced by the different physical properties of the materials. The cause of the appearance of

the strips was principally attributed to the different collapse
of the components in the mixed bed at the starting of rotation (
it is understood that the 'different collapse' means the different
angles of repose on which Oyama had done a considerable amount of
experiments to demonstrate the effects of the collapse of diferent
materials). Although a considerable amount of experimental in-
vestigation of this phenomena was made by Oyama. No reason why the
separation of large and small granules appeared to depend granule
size was given and the investigation remained qualitative.

Lacey (48) performed fundamental work on the degree of
mixing which involved the very laborious counting of all the parti-
cles (0.2" dia. of bullet topioca) in a mixture. The experimental
work was carried out in a wooden sphere, the degree of mixing was
proposed as :-

$$M = \left(\bar{\alpha}\bar{\beta}/n \right)^{1/2} \tag{2.8}$$

Where,   M = degree of mixing,

$\bar{\alpha}$ = fraction of A particles in the mixture of A and B,

$\bar{\beta}$ = fraction of B particles in the mixture of A and B,

n = number of particles in a cell (mixture completely
divided into cells).

The plots of M vs. $1/\sqrt{n}$ characterised the mixing. Lacey
thought that there is probably a family of curves of the similar
shape could be obtained for each combination of mixing machine and
materials.

Buslik (13) considered the mixing of multi-sized particu-
late materials, a formula was developed for computing the standard
deviation (σ_{δ}) for a given weight of a random sample which has a
known size distribution of the particles. The formula is,

$$\sigma_{p}^{2} = \frac{G(100 - G)w_{g} + G^{2}(\bar{w} - \bar{w}_{g})}{W} \tag{2.9}$$

Where, $\bar{w}_{g}$ = average particle weight in a size fraction being
investigated. The size generally contain a range
of sizes and is actually included but this frac-
tion is called w_{g} size (constant density being

assumed), the calculation of w_g was discussed by the author,

G = percentage by weight of the mixture of particles of size w_g,

w = average weight of all particles of all sizes in the whole mixture which is calculated from,

$$w = 1/100 \left(A w_a + B w_b + \ldots\ldots \right)$$

where, A, B, ... are the percentage by weight of the size w_a, w_b,.... in the mixture.

W = total weight of sample.

Buslik followed Oyama in considering the mixing of different sized materials, but it is felt that this proposed 'degree of mixing' is too complicated for practical purposes.

Coulson and Maitra (21) studied the mixing of powder experimentally. The experimental work was performed in a simple rotating drum mixer which could be inclined at an angle. Various angles of inclination and materials prop-erties were investigated in terms of rate of mixing and segregation tendencies. A simple method for finding the degree of mixing was proposed. 30 spot samples were withdrawn at each time for analysing the mixedness, if n samples out of 30 were found to have approximately the same composition as in the whole system then the degree of mixing was defined as,

$$M = \frac{n}{30} \times 100 \ \%$$ (2.10)

percent mixed.

In their analysis, an 'unmixedness' index X was used for the experimental plots which measured the segregation of the mixture. This 'unmixedness' index X was defined as,

$$X = (100 - M) \ \%$$ (2.11)

It was concluded that a inclined angle of 23° of the rotating drum, optimum mixing performance would be achieved.

Beaudry (8) presented a means for determining how will a continuous blender was operating, as compared with how well it is capable of operating. This takes into account the type of blending cycle used, the ratio of blender volume to batch volume,

and for certain cycles, the ratio of the inflow rate to the outflow rate.

$$\text{Blending Efficiency (B.E)} = \frac{(V_b/V_a) - 1}{\gamma - 1} \times 100\% \quad (2.12)$$

Where, V_b = variance among the batches before blending,

V_a = variance among the batches actually obtained after blending,

the variance was defined as,

$$V = \frac{\sum(C_i - C_{av})^2}{N}$$

where, C_i = the value of component A for the i-th batch,

C_{av} = the average value of component A,

N = number of batches sampled.

γ = the limiting ratio of V_b/V_p, V_p is the variance at perfect blending.

Application of this blending efficiency was only considered in the blending of liquids.

Visman and Van Krevelen (81) replotted Coulson and Maitra's results on a probability paper, as they considered that this method of plotting would give a better interpretation of the 'unmixedness' index of Coulson and Maitra.

Dukes (27) developed a means for testing the randomly mixed powders, as called the equivalent agglomerated size (E.A.S) which was defined as,

$$\text{E.A.S} = \left(\frac{P \cdot g}{x - \bar{x}}\right)\left(\frac{\sum(x - \bar{x})^2}{n - 1}\right) \quad (2.13)$$

Where, P = number of particles of each sample,

g = number of grammes weight in each sample,

n = number of samples drawn and analysed,

x = proportion of a determinable constituent C in each sample, and

$\bar{x}$ = mean proportion of C in the entire powder.

The smaller the E.A.S, the better was the degree of mixing.

15

In the limit, a mixture which was completely randomised in the statistical sense with respect to individual particles should have an equivalent agglomerate size of unity under ideal conditions. The limitation of experimental technique and analytical accuracy made this limit unattainable in practi e. The experimental results obtained from mixing of coal and magnesium oxide with an average of 3×10^6 particles per gramme, gave an E.A.S. of 352,000 after 0.5 minute mixing, however, an index decreased to 1,040 after 16 minutes of mixing.

Theoretical study of the mixing process was discussed by Danckwerts (23, 24), who defined the 'goodness of mixing' by two statistically definable quantities, the scale and the intensity of segregation .

Danckwerts considered the scale of segregation as analogous to the 'scale of turbulence' used in the statistical theory of turbulence. He defined this property as

$$R(r) = \frac{\overline{(a_1 - \bar{a})(a_2 - \bar{a})}}{\overline{(a - \bar{a})^2}} = \frac{\overline{(b_1 - \bar{b})(b_2 - \bar{b})}}{\overline{(b - \bar{b})^2}} \qquad (2.14)$$

Where, a_1, a_2 (or b_1, b_2) are concentrations measured at two points in the mixture at a distance r apart,
$\bar{a}$ or $\bar{b}$ are the mean concentration of a or b in the mixture as a whole.
$R(r)$ is called the coefficient of correlation between the values of a (or b) at points separated by a distance r.

Then he stated two measures of this scale of segregation, the linear scale S and the volume scale V. The linear scale was defined as,

$$S = \int_0^\infty [R(r)] \, dr = \int_0^\epsilon R(r) \, dr \qquad (2.15)$$

where, ϵ is the value of r for which $R(r)$ falls to zero. The volume scale is 2π times the area under the curve of $r = f(r^2 R(r))$ so that,

$$V = 2\pi \int_0^\infty r^2 [R(r)] \, dr = 2\pi \int_0^\epsilon r^2 R(r) \, dr \qquad (2.16)$$

Danckwerts gave several methods for measuring the scale of segregation.

solids mixing, which involves analysis of a number of samples, a
formula was developed for computing the scale of segregation V,
as follows :-

$$V = \frac{\sigma_\kappa^2}{2 \, v \, \sigma_a^2} \qquad (2.17)$$

where, σ_κ^2 is the variance among the spot sample measurements (the
content of A in each sample of size v is denoted by κ), σ_a^2 is the
variance of point concentrations of component A from the overall
mean concentration $\bar{a}$.

The determination of point concentration by special tech-
niques such as electrical, optical methods, following the progress
of a chemical reaction, was discussed by Danckwerts. Although these
ideas are very interesting, their adaptation to a dry powder mixing
process would require considerable investigation of practical
means of powder mixing. The measuring techniques must not so dis-
turb the mixing patterns, that the picture obtained is not truly re-
presentative of the mixer in normal operation.

The intensity of segregation was defined as ,

$$I = \frac{\sigma_a^2}{\bar{a} \, \bar{b}} = \frac{\sigma_b^2}{\bar{a} \, \bar{b}} = \frac{\sigma_a^2}{\bar{a}(1-\bar{a})} = \frac{\sigma_b^2}{\bar{b}(1-\bar{b})} \qquad (2.18)$$

where, σ_a^2 (or σ_b^2), $\bar{a}$, $\bar{b}$, are denoted as above. I is the intensity of
segregation.

Although I had been represented 'in total', however, it
was the ratio of the variances.

Blumberg and Maritz (9) discussed the theoretical concepts
of complete and incomplete mixing from a view point of statistical
hypothesis. The experimental data was given from the mixing of two
materials having the same particle size differing only by colour.
For the degree of mixing of an incomplete mixed batch, the follow-
ing equation was proposed,

$$\phi = v \sum (z_1 - \zeta_\bullet)^2 \qquad (2.19)$$

Where, $\quad v =$ the average number of particles per sample,

$\quad Z = 2 \sin^{-1}\sqrt{x_i}$, $x_1 = \sigma/n$, which equals the fraction
of real particles in a spot sample of n particles,

$\quad \zeta_\bullet = 2 \sin\sqrt{\sigma 5}$, as the fraction of real particles in the

whole mixture.

Since ϕ is also a chi-square variable, the probability that a value as large as the computed value would occur by chance, which could be looked up in the chi-square tables. If this was less than the level that had been set (say one time in twenty), then the mixing was considered to be incomplete. If it turned out, however, that this large value could occur by chance of more than one in twenty, it was then considered the mixing was complete, i.e., a randomly mixed batch had been obtained.

Lacey (49) further studied the mixing of particulate solids theoretically. He set up the theory of mixing mechanisms which showed the relationship of few systematic practical investigations. He proposed the degree of mixing from Kramer's original formula to,

$$M = \frac{\sigma_o^2 - \sigma^2}{\sigma_o^2 - \sigma_R^2} \qquad (2.20)$$

Where, σ^2 = true variance of the samples,

σ_o^2 = variance at the completely unmixed state, and

σ_R^2 = expected variance of completely randomised state.

Lacey stated that this degree of mixing is more satisfactory statistically, since σ^2, unlike σ, has additive properties. Then he further developed the expression to,

$$1 - M = \frac{\sigma^2 - \sigma_R^2}{\sigma_o^2 - \sigma_R^2} \qquad (2.21)$$

which was called a fundamental equation for expressing the state of a mixture.

One of the outstanding papers was published by Stange (76), who formulated the measurement of the random variance of a binary and different sized mixing system with the known size distribution. This random variance could be represented by a general formula, as,

$$\sigma_R^2 = \frac{PQ}{g} \left[P \bar{v}_A^2 (1 + c_a^2) + Q \bar{v}_P^2 (1 + c_P^2) \right] \qquad (2.22)$$

Where, σ_R^2 = theoretical random variance,

P, Q = weight fractions of the mixing components,

$\bar{v}_P, \bar{v}_Q$ = average particle weight for P and Q,

C_P, C_Q = variation coefficients for P and Q. Referring to the frequency distribution of particles by weight, the variation coefficients could be calculated as,

$$C = \bar{\sigma}_P / \bar{r}_P \quad \text{and} \quad C = \sigma_Q / \bar{r}_Q ,$$

where σ_P and σ_Q are the standard deviations of weight distribution of components P and Q, and $\bar{r}_P$, $\bar{r}_Q$ are the mean particle weights of P, Q.

g = weight of sample.

This equation could be simplified in some special cases.

(1) in the binary system, if one of the components Q in the mixture is much less than other component P, as P $\simeq$ 1, hence Eq.(2.22) becomes,

$$\sigma_R^2 = (Q/g)\bar{r}_Q (1 + C_Q^2) \tag{2.23}$$

(2) when the initial distributions consist of almost equal particle size, the squares of the coefficients of variation C_P^2 and C_Q^2 become very small compare with unity and therefore could be neglected. Then Eq.(2.22) is simplified to,

$$\sigma_R^2 = \frac{PQ}{g} (P\bar{r}_Q + Q\bar{r}_P) = \frac{PQ}{g} (\bar{r}_P \bar{r}_Q / r) \tag{2.24}$$

(3) when both components of the mixture have been comminuted to about the same extent, $\bar{r}_P = \bar{r}_Q = \bar{r}$ and $C_P^2 = C_Q^2 = C^2$, then Eq.(2.22) becomes,

$$\sigma_R^2 = \frac{PQ}{g} \bar{r} (1 + C^2) \tag{2.25}$$

(4) when component P in the mixture is over whelmingly predominant, say P $\geqq$ 90%, and the mean particle weights $\bar{r}_P$ and $\bar{r}_Q$ and the coefficient of variation C_P^2 and C_Q^2 are for both components at least 'of the same order', then Eq.(2.22) becomes,

$$\sigma_R^2 = \frac{PQ}{g} P\bar{r}_Q (1 + C_Q^2) \tag{2.26}$$

(5) when the weight fractions of P and Q are of the same order, P $\simeq$ Q, but component Q is much finely divided than P, $\bar{r}_P << \bar{r}_Q$, then Eq.(2.22) becomes,

$$\sigma_R^2 = \frac{PQ}{g} Q\bar{r}_P (1 + C_P^2) \tag{2.27}$$

Later, Stange (77) extended the formulae developed to

evaluate three or more components in the mixture. To accomplish this, the initial approach was to combine two real components into a fictitious component and to treat the mixture as a binary system. The mixing quality could be computed for all three components.

Weidenbaum and Bonilla (82) investigated the fundamental of mixing of powders in a horizontal rotating drum mixer. The materials used were salt and sand having the same particle size ($-44/+54$ U.S. mesh). Various theoretical aspects of the subject were discussed and the degree of mixing was described as the ratio of the random standard deviation to the measured standard deviation, as

$$M = \frac{\sigma_R}{\sigma} \tag{2.28}$$

Where, $\sigma_R = \left[P(1 - P)/n \right]^{1/2}$, P is the true particle fraction of one of the components in the mixture, or the mean binormial distribution, n is the total number of particles in a spot sample,

$\sigma = \left[\dfrac{(x/n)_i^2 - (\bar{x}/n)^2}{N} \right]^{\frac{1}{2}}$, $(x/n)_i$ is the particle fraction of the sample in i-th spot sample, $(\bar{x}/n)$ is the arithmetical mean particle fraction of a component for N spot samples, N is the total number of spot samples taken from the mixture.

Graphs of σ vs. number of revolutions were plotted. Also, the use of chi-square test was illustrated for comparing distributions to determine whether a batch was randomly mixed.

Smith (75) studied the mixing of soil with chemicals for the stabilisation of soil. The strength of stabilised soil was dependent on the uniformity of the mixture. A mixing coefficient M was proposed which was defined as the ratio the standard deviation of completely unmixed state to the standard deviation of the samples taken from the mixture, as

$$M = \frac{\sigma_0}{\sigma} \tag{2.29}$$

Where, $\sigma = (x_i - u_0)^2/N$, x is the content of the additives of any spot sample, u_0 is the mean fraction of the additives, N is number of spot samples taken from the mixture.

$$\sigma_0 = \left[u_*(1 - u_*) \right]^{1/2}.$$

M is unity at no mixing state, and theoretically approach to infinity as mixed product become more and more uniform.

Forscher (32) theoretically studied the continuity of one powder (A) in a mixture of two powders (A and B), by means of using the stochastic process. It was assumed that the two powders are well mixed, so that particles of powder A are randomly distributed in the powder B. The continuity condition for powder A is expressed by the probability that the separation between two particles of this phase vanishes. For spherical particles of unequal sizes the continuity condition can be stated as a probability that the distance between the centres of any two particles of powder A equals the sum of the radii. The analysis methods (3 steps) and general equations were developed. A powder mixture of equal spherical particles was discussed by generating the random number, in such a way that they had a random distribution which the author claimed that as a special case of the general method. The author first explored the mixture of powder which relating to packing and stochastic considerations, however, the proposed method seems limited to the equal size spherical powder mixture.

A method based on accepted statistical laws was derived to show how the mixing of particulate ingredients could be measured, and how the degree of mixing may be indicated by an index of probability, was given by Adams and Baker (1). They studied the mixing operation in four types of mixers, using natural polyethylene granules (white) and carbin blacked polyethylene granules (black). Three sets of test were carried out in the mixers. The first two sets of test required the graphs of the blacks vs. the sequence number of samples ; one test looked for the number of samples outside certain control limits or confidence limits, and the other looked for the number of consecutive samples on one side of the mean. The third test required computing

$$b = \sigma^2/\bar{x} \tag{2.30}$$

Where, $\sigma = \Sigma(x - \bar{x})^2/(N - 1)$, x is the number of black granules in a sample, $\bar{x}$ is the mean of black granules in the samples, N is total number of samples.

From t'e graphs, the probability of obtaining this large value of b from a poisson distribution was obtained. This probability was used as a measure of mixing in a manner analogous to that of a statistical test of significance.

Oyama and Ayaki (62) studied the powder mixing by considering mathematically the degree of mixing and some other theoretical aspects on the progress of mixing. The stochastic process was introduced with particular interests on the Markov chain technique. The analysis of transition probabilities in a matrix of a Markov chain lead to the equations similar to those of Brownian motion, on the assumptions that the mechanism of mixing is independent of time and position in the mixer. The authors stated that these assumptions seemed to be satisfactory for measuring the longitudinal mixing in the horizontal rotating drum mixer, with materials of similar physical properties. A simple degree of mixing was proposed, as the measured sample standard deviation, or, its square, the variance,

$$\sigma^2 = \frac{1}{N} (x_i - \bar{x})^2 \tag{2.31}$$

if the true particle fraction in the mixture is known, then,

$$\sigma^2 = \frac{1}{N} (x_i - a)^2 \tag{2.31'}$$

Where,   a is the true fraction of components in the whole mixture, x_i is particle fraction in a sample and $\bar{x}$ is the average fraction of samples, N is number of samples.

An index of mixing which is the form proposed by Lacey (Eq.(2.20) and Eq.(2.21)) was used for the plotting of the experimental results.

The authors considered that the degree of mixing could be composed of two parts, σ_m^2 and σ_r^2. It was described that σ_m^2 is the σ^2 due to local composition variations which would be independent of sample size, and σ_r^2 is the variance due to sample size, which were referred that Oyama's earlier theory as whole and local mixing.

Experimental work was performed in a horizontal rotating drum mixer with a constant speed of rotation, sands of same size were used. The main emphasis of this paper was on the rate of mixing and the operating conditions of the mixing process.

Since 1956, a series of studies were published by Yano et al., who investigated the mixing of powders in several mixers, such as V-type (V), double cone (D.C.), rotating cube (R.C.), horizontal drum (H.D.) and ribbon mixer. The performances of these mixers were tested carefully, under various mixing conditions, as well as the physical properties of the materials used. Finally, the authors concluded that the experimental results could cover most of the mixing process in the tumbling mixers, however, in the case of ribbon type mixer, it should belong to another catagory.

Since these papers are very comprehensive studies of the subject, separate reviews are needed.

(1). The first paper by Yano et al (89) was a detailed study of the mixing of powders in V-type mixers. Two sizes of the mixers were used (actual mixer sizes were 0.7 and 5.5 litres resp.). PVC and anhydrous sodium carbonate powders of the same particle size (-100/+200 standard mesh) were used. The sample standard deviation was proposed as the degree of mixing.

$$\sigma = \left[\frac{1}{N} \ (C_i - C_0)^2 \right]^{\frac{1}{2}} \tag{2.32}$$

Where,

σ = between sample standard deviation, vol.%,

C_0 = concentration of Na_2CO_3 in the mixture in a perfectly mixed state, vol.%,

C_i = concentration of Na_2CO_3 in the spot sample, vol.%,

N = number of spot sample taken at each sampling.

The sample was analysed in such a way that each spot sample was weighed, its weight w gm. contained both Na_2CO_3 and PVC powders. The powder was placed into water, Na_2CO_3 dissolved, then PVC can be separated by filtration. 0.1 N HCl solution was used to titrate the Na_2CO_3 solution. The use of 0.1 N HCl could be converted to the quantity of Na_2CO_3 in the solution by weight. The weight of Na_2CO_3 in each spot sample was denoted by w_a, so that the weight of PVC (w_b) could be computed.

The concentration of Na_2CO_3 in each spot sample is then,

$$C = \frac{100}{1 + (w_b/w_a)(\rho_a/\rho_b)} \qquad\qquad 0 \le C \le 100 \ (vol.\%)$$

and,

$$C_o = C_a = \frac{100}{1 + (w_b/w_a)(\rho_a/\rho_b)} \quad , \quad 0 \le C_o \le 100 \quad (vol.\%)$$

where,

w_a and w_b are the weights of Na_2CO_3 and PVC powders,

ρ_a and ρ_b are the densities of Na_2CO_3 and PVC powders.

A standard deviation of 13.2% was computed which was based on 30 spot samples at each sampling taken at the specificed location in the mixer, comparing with the standard deviation of 13.9% which was based on 7 spot samples taken at the representive places under the same operating conditions. Therefore, it was decided that the standard deviation would be based on 7 spot samples for each sampling throughout all the experiments.

The time required to achieve the minimum standard deviation was influenced by the speed of rotation, the proportion of materials loaded to the total volume of the mixer, the ratio of the two components and the method of loading. The minimum standard deviation was shown to occur at the extreme ratio of the two components, i.e. 0 or ∞, the highest value of standard deviation occurred at unity. The authors emphasised the importance of the method of loading, good loading shortened the mixing time. Different optimum operating conditions were obtained for different sized mixers. For small mixer the best speed of rotation was found to be 60rpm and for large one the optimum value was at 50rpm. A 13.6% of the volume ratio (equivalent to about 30 vol.% of loose packed powder to the total volume of the mixre), the best mixing results could be obtained.

(2) The second paper was published a year later (1957) by the same group of authors (90). They investigated the performance of various powder mixers, such as double cone (D.C.), rotating cube (R.C.), horizontal drum (H.D.) and the V-type mixer which was used in the previous study. Two sizes (0.7 and 5.5 litres) of each type of mixers were used. The materials and the testing techniques were the same as discussed previously.

Various opertating conditions such as speed of rotation, fullness of loading were studied for each mixer regarding to the degree of mixing and the mixing time required to reach the minimum standard deviation (not necessary the best degree of mixing) for all mixers remained fairly constant, but st speeds greater than 50rpm,

d̶i̶f̶f̶e̶r̶e̶n̶t̶ ̶r̶e̶s̶u̶l̶t̶s̶ ̶w̶e̶r̶e̶ ̶o̶b̶t̶a̶i̶n̶e̶d̶ The optimum speed of rotation for the best mixing conditions (lowest standard deviation and shortest mixing time required) was found to be 60rpm for D.C.-1, R.C.-1,V-1 and 50rpm for D.C.-2, R.C.-2, V-2, but for H.D.-1 and H.D.-2 the higher speed of rotation of 115rpm and 90rpm were needed (-1, referring to the small mixers and -2 for the large mixers). The optimum volume ratio for all mixers was 13.6 %.

(3). The third paper by Yano et al (91) contained a study of the effects of the physical properties of the powders on the optimum mixing conditions in the various mixers which were used previously.

The experiments were investigated into six different powders, such as Na_2CO_3, PVC, glass powder, sand, iron powder and alumdun which were formed five pairs of binary-component, namely, PVC-Na_2CO_3 (P-N), glass-Na_2CO_3(G-N), sand-Na_2CO_3(S-N), iron-Na_2CO_3(F-N) and alumdun-Na_2CO_3(A-N). However, all these powders were in the same size range (-100/+200 standard mesh).

The physical properties of each material was determined carefully under various conditions. The experimental results indicated that,

1), for powder of a fixed size range, although the powder combination was different,but little difference was evinced in the optimum mixing conditions on the type and size of mixer.

11), in order to examine the effect of the physical properties of the powders on the degree of mixing and the rate of mixing, a kinetic characteristic constant of powder C_R should be used,

$$C_R = \frac{\bar{u}}{(Q_2 \sin\alpha)^q} = \left\{ \frac{b}{f[2 + (b/h)]} \right\}^q \tag{2.33}$$

Where, $q = 1/(1 +m)$, m is the inte-gral number,

$\bar{u} = (Q_1/Q_s) \times L'$ (cm/sec),

Q_1 = flow rate of powder, mass flow (gm/sec),

Q_s = weight of powder in the interval of the length,

$Q_2 = Q_1/b\,h$, h is the thickness of layer and the coefficient of friction, b is the width of the pipe for testing the powder (cm),

L' = interval distance,(cm) and

α = angle of inclination of the pipe for testing.

It was concluded that the degree of mixing was highly dependent on the difference of C_R of the two powders, as ΔC_R increased with increasing the minimum standard deviation. For geometrical reasons, V-, R.C. mixers are not suitable for the mixing of materials having large ΔC_R, where as H.D. and D.C. and D.C. mixers may be recommended to handle a mixture having large ΔC_R.

(4) Yano et al (92) published the fourth paper in 1959 which investigated the influence of particle size of powder on the optimum operating conditions, the degree of mixing and the rate of mixing in the four types of mixers which were described previously. Six different mono-sized pairs of Na_2CO_3 - sand system were used, the size ranges were, -42/+48, -48/+65, -65/+80, -80/+100, -100/+150, and -150/+200.

Series of experiments were tested on the effects of the best mixing conditions with particle sizes. It was concluded that in general, the values of the optimum speed of rotation increased with increasing the mean particle size d_{av}. The relationship between the optimum speed of rotation N_{op} and d_{av} in a fixed size range could be expressed by

$$N_{op} = K.d_{av} \qquad (2.34)$$

Where, K = a proportional constant dependent on the shape
          and the size of the mixer, for V-, D.C.- R.C.-
          mixers are 180, and for H.D. is 380, d_{av} was
          measured in millimeters.

The degree of mixing decreased with increasing particle size. In the case of two different sized-component system, it was found that as the ratio of the mean particle size decreased, the mixture tends to be poorer. This effect was very pronounced when the ratio dropped below 0.7 - 0.8.

(5) Finally, Yano et al (93) summarised all the experimental results from it was attempted to establish a general expression for the rate of mixing in tumbling mixers. It was stated that three stages of mixing would occur in the whole mixing process,

namely, initial, mixing and equilibrium stages. The original paper did not name the stages. It was only described the mixing stage as the region which could be represented by a straight line on plotting the logarithm of degree of mixing vs. time of mixing. The slope of this straight line was determined by different factors investigated in the previous papers. It was stated that the general equation so developed could be used to describe the mixing of powders in different conditions by varying of the coefficients in accordance with the properties involved.

(6) Meanwhile, Yano et al (94) also studied the mixing of powders in the ribbon type mixer. The experimental procedures and testing techniques were the same as those used in the previous experiments. Four pairs of same sized Na_2CO_3 and sand combinations, with the size ranges -35/+48, -48/+65, -65/+80 and -80/+100 were used. The materials were equally loaded end to end. The mixing curves obtained in this type of mixer were basically similar to those obtained in tumbling mixers with respect to the optimum operating conditions for different materials properties, speed of rotation of the ribbon, ratio of loading volume and the degree of mixing. The best degree of mixing could be obtained at 80rpm (ribbon speed) and 38% of the volume ratio. The finer the powder particle size, the better the degree of mixing, however, a longer mixing time was needed.

Iiyama and Aoki (43) studied the mixing of powders in a vertical agitator tank. The main attribution was on the energy consumption of the paddle type agitator, and the material properties (Toyaura and Soma sand were used). The torque equations were developed. These equations could be classified into two ranges, at low and high speeds for powder mixing. The principal torque equation was analogous to the pumping theory including the effect of centrifugal force.

Otake et al (56) also studied the mixing of powders in a vertical agitator tank mixer. The mechanism of powder mixing in the vertical tank with paddle-type impeller was analogous to the theory of radial diffusion and solved by Bessel's function 1st kind. Two pairs of powders having the same particle size were used, sand - salt (-32/+42 mesh) and Na_2CO_3 - PVC (-100/+200). The degree of

mixing was proposed as,

$$M = \frac{\sigma}{\sigma_o} = \left[\sum_{i=1}^{k} \frac{P_i}{N_i} \sum_{j=1}^{N_i} \frac{(C_{ij} - C_0)}{C_0(1 - C_0)} \right]^{\frac{1}{2}}$$
(2.35)

Where, σ = standard deviation of C_{ij} from the C_0 in the total samples,

σ_o = standard deviation of the sample in a perfectly unmixed stated,

k = no. of powder particle layers,

N_i = no. of spot samples in i-th layer,

P_i = width due to size of layer $(\frac{r_i - r_{i-1}}{Ro})$, Ro is the radius of tank, r is the radial distance of the i-th powder particle layer,

C_{ij} = concentration of marked powder in j-th sample taken from the i-th layer (wt.%), and

C_0 = concentration of the marked powder in the mixture of perfectly mixed state (wt.%).

The relation between the degree of mixing and the operating conditions, such as impeller size, rotational speed, etc., were determined by dimensional analysis, as,

$$\frac{\sigma}{\sigma_o} = 0.676 \ (Nt)^{-0.48} \left(\frac{D_i N^2}{g}\right)^{-0.16} \left(\frac{D_T}{D_i}\right)^{0.69} \left(\frac{R_i}{D_i}\right)^{0.57}$$
(2.36)

and $$\frac{(\sigma/\sigma_o)}{(Nt)_c} = 6.18 \times 10^{-6} \left(\frac{D_i N^2}{g}\right)^{-0.52} \left(\frac{D_T}{D_i}\right)^{0.8}$$
(2.36')

Where, t = time of mixing,

g = acceleration due to gravity,

D_T = mixer tank diameter,

R_i = radius of the marked powder charged in the tank,

N = rotational speeds of impeller, and

D_i = impeller diameter.

By analogy to the diffusional process, the apparent diffusivities were evaluated from the concentration distribution of the two kinds of powder. The apparent diffusivities and the operating conditions were related by the equation,

$$D_s/N \ D_i^2 = 5.75 \times 10^{-3} (D_i N^2/g)^{0.18} (D_T/D_i)^{0.45}$$
(2.37)

Where, D_s = apparent diffusivity of the marked powder in the tank.

Furthermore, the theoretical degree of mixing was give in terms of apparent diffusivity,

$$\frac{\sigma}{\sigma_0} = \left[\frac{4R^2}{R_0^4} \sum_{k=1}^{\infty} \frac{J_1^2(m_k R)}{C_0(1 - C_0)m_k^2 J_0^2(m_k R_0)} \, e^{-2D_s m_k^2 t} \right]^{\frac{1}{2}} \quad (2.38)$$

Where, J_0 = zero order of Bessel function of 1st kind,

                  J = first order of Bessel function of 1st kind,

                  m_k = arbitrary constant.

Introducing the obtained value of apparent diffusivity into the above equation and comparing the theoretical degree of mixing with observed value, the authors found that good agreement was obtained between theory and the experiments.

A very special method for testing the performance of some common commercial mixers was developed by Gray (36) who used a re-flectivity probe which has a light and a photocell behind a glass window. The probe measured the intensity of light reflected from a light particles immediately outside the glass window. For mixture of light and dark particles and light source of constant intensity, the readings of the photocell meter are uniquelly related to the composition of the mixture. Eight commercial mixers had been tested with three pairs of powders selected to provide data for the comparison of the mixing performance of the equipment. The mixing progress was followed by determining the standard deviation of the microammeter readings obtained directly from the probe inserted in the mixture. The standard deviation used was :-

$$\sigma = \left[\frac{\Sigma (i - \bar{i})}{N - 1} \right]^{\frac{1}{2}} \quad (2.39)$$

Where,   N = no. of probe meter readings,

                  i = probe meter reading, (μA),

                  $\bar{i}$ = mean probe readings, (μA), and

                  σ = standard deviation of reflectivity probe meter reading, (μA).

A low σ corresponds to a high uniformity of the mixture. The mixing of different sized materials in several types of mixers were tested in this manner, however, no particular data was given for

the selection of mixers.

A three-component mixing system was studied by Gayle et al(33) The study was based on the mathematical treatment of chi-square. This method for testing mixing progress is not only applicable to binomial distributions but also to distributions consisting of any finite number of components distinguishable as discrete particles, while the standard deviation is generally limited to a binary-component system.

The chi-square is represented by the form, as,

$$\chi^2 = \sum \frac{(0 - E)^2}{E} \tag{2.40}$$

Where, χ^2 = chi-square,

0 = observed number of particles of any given colour in a sample, and

E = corresponding expected number, based on the average distribtution of components in the mixture.

The value of chi-square decreases with increasing the uniformity of the mixture. At the unmixed state, the value of chi-square is equal to the number of degrees of freedom multiplied by the number of particles counted for each sample.

From the authors' experiments, the degrees of freedom were 24 (3 components and 12 spot samples withdrawn at each sampling), and 30 particles contained in each spot sample, so that at the numixed state, chi-square was 24 x 30 = 720.

The standard deviation of the observed chi-square could be expressed as,

$$\sigma_{\chi^2} = \overline{\chi}^2 (2/N)^{1/2} \tag{2.41}$$

Where, $\overline{\chi}^2$ = average observed value of chi-square, and

N = degree of freedom.

Because the expected value of chi-square from a random mixture is equal to the number of degree of freedom, Eq.(2.41) could be reduced to that given by a random mixture in most tests, i.e., $\sigma_{\chi^2} = \sqrt{2N}$.

A formula similar to Lacey's (Eq.(2.20)) was proposed.

by combining the various chi-square values and the segregation
index S defined as,

$$S = \frac{X_o^2 - X_r^2}{X_s^2 - X_r^2} \qquad (2.42)$$

Where, X_o^2 = observed chi-square from any mixture,

X_r^2 = expected chi-square for random mixture, and

X_s^2 = expected chi-square for segregated mixture.

This segregation index varies from unity to zero as the
mixture varying from complete segregation to complete randomised
state.

Then, Gayle et al (34) further used this index to test the
performance of horizontal rotating drum mixer by using a four-com-
ponent system. The authors stated that the use of this segrega-
tion index was valid and its application for multi-component system
confirmed.

Shinner and Naor (73) describe a new statistical criterion
for testing the randomness of powders mixture. The proposed testing
method was based on the shortest distance between particles of the
component present in lower concentration. The properties of the
distribution of this characteristic were derived from poisson dis-
tribution. The proposed test was particularly sensitive in detecting
clusters of particles and had more advantages than X^2 test. Later
Shinner et al (74) applied this test to a particular powder mixing
system which dealing with coloured and white ceramic powders in a
V-type mixer. It had been found that the proposed test was applicable,
and the mixing time in V-type mixer was strongly dependent on the
particle size, the mixing time may be longer than that commonly
expected for complete randomness to be achieved.

Carley-Macauly and Donald (18) (19) and Carley-Macauly (20)
defined the degree of mixing as the systematic variance, or the
variance due to the position of a series of samples throughout the
mixture. The systematic variance was obtained by measuring the total
variance and subtracting from the final random variance, i.e.,

$$V_p = V - \sigma_R^2 \qquad (2.43)$$

Where, V = estimated value of the variance among sampling
compositions, the experimental estimate of σ_R^2,

$$\sigma_R^{-2} = \text{random variance at complete mixed state, equal}$$

to pq/n for a two-component mixture of equal
sized particles.

The main experimental work was performed in three different types of rotating mixers, namely, inclined drum, double cone and horizontal cylinder. The materials were Bedford sand of two grades, grade 1, was 850 - 600 μ, and grade 2, 500-420 μ. The binary-component system with mono-sized ingredients were distinguishable by colour.

Rosaman and Donald (67) (68) and Roseman (69) continued the theory of mixing from Carley-Macauly in the horizontal drum mixers. The experiments were mainly studied the mechanisms of mixing due to different size or density of the components. The experimental work was carried out in two different sized perspex mixers. However, most part of the experiments were assessed by observations , no detailed experimental conditions are given.

Rumpf and Müller (70) studied the mixing of powders in a centrifugal mixer. The mechanism of this centrifugal type mixing could be clarified by a model in which the distribution of the material resettled on the surface of the mixture after having been thrown out. The particle motion in the mixing drum by a rotating paddle or other elements was studied, and an approximated calculation for the trajectory of this motion could be made. The mixing patterns created by different rotating elements (4 types such as rod, paddle, flat-bar-with-cross-bar and ploughshare) were studied photographically, in order to determine the best mixing condition. The experimental work was also carried out to determine the effects of shape and speed of those rotating elements, the mixing time and the power consumption. It was concluded that the mixing time is not of decisive importance for the development and design of mixers. The centrifugal mixer required comparatively short time of mixing. The rotating element was used mainly to increase the radial mixing.

Lastov'tser (50) described an alternative method to the existing restrictive batch methods of powder mixing. He showed the method of large scale mixing of free flowing powders continuously by means of two types of mixer, namely, flow and circulation mixers. The degree of inhomogeneity of the resulting mixture was

determined from a number of samples analysed for their content of
the component present by weight which would be referred to the key
component, as,

$$I = \frac{100}{C_0} \sqrt{\frac{\Sigma(C_i - C_0) \, n}{N - 1}} \qquad (2.44)$$

Where,   I = degree of mixing,

C_0 = concentration of one of the components in a per-
        fectly mixed state,

C_t = actual concentration of the corresponding component,

n = number of samples of concentration C_t, and

N = total number of samples analysed.

The degree of inhomogeneity I suitable for mixing with
different proportions of the components, showed that the larger the
value of I the worse the uniformity of the mixture, at perfect mixing
I should approach to zero.

The variation of I in the course of mixing, as a function
of R_i (in the flow type) or R (in the circulation type), where R_i
and R_2 are the mixture's recirculation factors, which was defined,
as,

$$R = G_m/G_t \qquad (2.44')$$

Where, G_m = rate of materials fed into the hopper, kg/hr.,

G_t = total weight of materials charged into the mixer.

Talc (90% by wt.) and sugar (10% by wt.) having the same
particle size (300μ) were used for this investigation.

Kaye and Sparrow (46) investigated the surface diffusion as
a mixing mechanism. It was thought that all three mixing mechanisms
which Lacey (49) proposed namely convective, diffusive and shaering
are operating simultaneously in a running mixing system, however, a
quantitative study of the importance of one particular mechanism is
very difficult.   The authors decided to separate one mechanism
from the other two mechanisms and investigated in isolation with a
specially designed apparatus. The apparatus consisted of two troughs
on a stationary inclined plate, each trough was divided into 20
compartments each of one centimeter wide. Glass beads (No.5 grade)
of different colours were used, 200 red glass beads were put into
compartment 10, of the upper trough. Equal number of blue glass

beads were put into compartments 9 and 11 of the same trough. Then the beads were released simultaneously. The angle of inclination was maintained at 37°, so that the beads rolled downwards and collected in the lower trough. The beads from each collecting compartment were counted. However, only the lateral distribution of red beads was obtained. A graph of the data obtained from the distribution of red beads, drawn on an arithmatical probability paper gave a straight line. This was due to the symmetry of the lateral distribution of the red beads from compartment 10. It was concluded, therefore, that the results showed that after rolling under gravity down a so-called equilibrium surface, as the mechanism of diffusive mixing on the inclined surface – surface diffusion in a mixing machine, the lateral distribution of particles is normal.

Poole et al (63) studied the mixing of fine metallic powders in a centrifugal bowl batch mixer, on 3 kg scale. The definition of the final random mixture used Stange's relationship which had been varified with mixtures of copper and nickel powders, and this had provided a revealing basis of comparison for the results. After initial experiments with spheroidal metal powders, more complicated systems were studied which including ceramic oxide powders of differing shape factor and agglomeration characteristics. The measure of inhomogeneity of a mixture by means of the standard deviation of the samples σ ,

$$\sigma^2 = \frac{\sum_{i=1}^{n}(x_1 - \bar{x})^2}{n - 1} \tag{2.45}$$

Where, x_1 = concentration of the minor mixture component in the sample,

$\bar{x}$ = mean sample soncentration estimated from a finite group of samples,

n = number of particles in a class found in a one component sample of weight W.

The measured coefficient of variation of sample comcentration C was then introduced, this was equalled to $\sigma/\bar{x}$. And the mixing index was defined as,

$$M = C/C_R = (\sigma/\sigma_R) \times (m/\bar{x}) \tag{2.46}$$

Where, C_R = the theoretical value of C for fully randomised
                   mixture or σ_R/m , σ_R is the theoretical standard
                   deviation of sample concentration (Stange) and
                   m is the theoretical mean sample concentration.

It was found that the amount of mixing required to approach
random homogeneity not only increases markedly in the presence of
agglomerates but also could be increased proportionally with the
increase of minor constituent concentration. An approximate corre-
lation had been found between the dependence of homogeneity on
sample size and the degree of randomness attained in a mixture.
Such a relationship could provide an indication of the nearness to
random homogeneity in the absence of information on the particle
size.

Later, Poole et al (64) used the same measuring techniques
and materials to study the mixing characteristics of these spheroidal
metal and ceramic oxide powders in a continuous ribbon mixer, at a
throughput up to 24 kg/hr scale,   using the residence time be-
tween 5 to 50 minutes.

The homogeneity obtained from the powder mixture free of
segregation were as good as those obtained in the batch mixer. The
use of an impact wheel as a combined pre-mixer, and agglomerate
mill had also been investigated, a 30% improvement was found in the
homogeneity of a urania-thoria mixture, compared with that obtained
in the ribbon mixer alone.

Strel'Tsor et al (78) studied the kinetics of mixing of
powdered materials in a commercial mixer. The study was concerned
mainly with the rate of mixing in a laboratory scale centrifugal
mixer. They proposed for their degree of mixing, as,

$$M = \frac{\sigma}{\sigma_o} \tag{2.47}$$

Where, σ = standard deviation of spot samples, and
                   σ_o = standard deviation of non-mixed state.

The authors stated that when solid mixing occurred in a
high speed mixer (300rpm), the rate coefficient k in the common
first order rate equation was more constant, but it would change
and become a function of mixing time in a lower speed mixer. The

experimental materials used were salt and sand.

Rutgers (71)(72) studied the axial dispersion rate for continuous flow of long grain rice (red rice and ordinary rice had been used for the experiment) in the laboratory size rotating cylinder which could be inclined at an angle of 0° to 4°. A piston flow model with superimposed longitudinal diffusivity was developed. The Boderstain number was introduced for the axial 'diffusion' type process, this Boderstain (B_0) number could be simplified to,

$$B_0 = 2/\sigma^2 - 1 - \sigma^2/2 - (\sigma^2)^2/2 - 5(\sigma^2)^3/8 \qquad (2.48)$$

Where σ^2 is the variance.

This equation was used to calculate the axial dispersion coefficients. Variations in the properties of the solid particles had also been studied to correlate the effect of axial mixing.

Cahn et al (16) studied the geometric coordinate of the V-type mixer. The two arms of the V-type mixer could be rotated at an angle of 0° (standard type) to 180° (inclined cylinder). The rate of mixing was interpreted in terms of the flow between the arms of the mixer. $CaCO_3$ and SiO_2 were used for the experiments. The degree of mixing was used of the type proposed by Lacey and the sample variance σ^2 was defined as the functions of number of rotation N, and the variable angle of the mixer arms, which was given in an empirical equation, as,

$$\sigma^2 = (-11.77 \log N - 0.212\beta + 74.11)^2 \qquad (2.49)$$

Where β is the variable angle of the mixer arms.

Alternatively the whole mixing process in the V-type mixer could be separated into two parts, namely, the statistical criterion of mixing M and the geometrical (flow) criterion of mixing Q. The relation between these two parts was stated that,

$$M = Q + constant,$$

both were arbitrary parameters by definition, and that they should be independently derived.

Hill (41) examined the mixing of natural polyethylene and

carbon blacked masterbatch in the double cone mixers. Adams and Baker (2) had obtained the poor results in similar experiments. Hill, however, found that when the materials have a small size difference but have a 1.3 density ratio, a good mixture was obtained in a $250ft^3$ batch mixer. It was suggested that it was the size and not the density of the particles which was responsible for poor mixing. Poor mixing could be aviading either by providing the mixer with a deflector or by making the particle size of the small component more than than the large component.

A new degree of mixing was proposed by Ashton and Valentin (4). They pointed out that when the mixing proceeded numerical values of the type of degree of mixing proposed by Lacey are all between 0.95 to 0.99. This could be over come by representing σ_o^2 and σ_R^2 on a logarithm scale and the new degree of mixing A^2 on a linear scale, so that,

$$A^2 = 0 \text{ for } \sigma^2 = \sigma_o^2 , \text{ and}$$

$$A^2 = 1 \text{ for } \sigma^2 = \sigma_R^2 . \text{ Then,}$$

$$A^2 = \frac{\log \sigma_o^2 - \log \sigma^2}{\log \sigma_o^2 - \log \sigma_R^2} , \text{ or}$$

$$A = \left[(\log \frac{\sigma_o}{\sigma})/(\log \frac{\sigma_o}{\sigma_R}) \right]^{\frac{1}{2}} \tag{2.50}$$

This proposal showed good discrimination over the whole mixing range and was largely insensitive to the proportion of the composition of the mixture.

Experiments were performed in four small scale industrial mixers, such as air-mixer, double cone, vertical-screw and Z-blade, with three different ranges of size and specific gravity materials. The combination of materials were classified into two categories, easy and non-easy mix. Where an easy mix was used, the extent of final mixing was found to be the same for all mixers. However, non-easy mixes were produced whenever a difference in specific gravity or particle size occur between the two components.

Later, Ashton et al (3) used a light probe to measure the local composition of a binary mixture of powders. The light probe was first developed by Gray (36), however, Ashton et al used the direct reading from the light probe to a digital computer, and the

final results could be computed automatically.

Campbell and Bauer (17) reported the studies on the demixing tendency in the horizontal rotating cylinder which was called de-mixer, for mixing of components of different sized materials. Various physical conditions had been studied from sand and soda ash combinations, however, no detialed operating conditions were given. It was concluded that the random movement of individual particles in a moving bed of free flowing particles is the exception, rather than the rule, various physical factors and properties of particles create a tendency to overcome the random movement, particles tend to seek out and remain in segregated concentration with similar particles. Only particles differing in colour alone will yield a statistically random mixture. The most stable and efficient mixture is obtained when all particles are of same size. With the usual raw materials, density differences and difference in particle shape have a measurable but minor effect on the efficiency of mixing and stability of mixture, when compared with the effect of size difference.

kun and de Chazal (47) proposed a mathematical model of a random mixture consisting of large, medium and small sized particles. The proposed equations of variance were based on the assumption that all the particles are randomly distributed and have the same probability to occupy each unit volume in the mixture. The variances for three different sized components are expressed as,

$$\sigma_x^2 = \frac{X(1 - X)}{a} \quad , \qquad a = V_s/v_1$$

$$\sigma_y^2 = \frac{y'(1 - y')}{b} \quad , \qquad b = E(V_{m+s})/v_m \ , \ y' = \frac{Vs}{E(V_{m+s})Y}$$

$$\sigma_z^2 = \sigma_y^2 + \sigma_{ex}^2 \ , \ \sigma_{ex}^2 = \left[\frac{E(Vex)_1}{V_1}(4.25 + 8\omega + 4\omega^2)\right]^2 \sigma_x^2 \quad (2.51)$$

(the equations were derived on the basis of the volume fraction)

Where, $\sigma_x^2 , \sigma_y^2 , \sigma_z^2$ = variance, of large, medium and small particles respectively.

σ_{ex}^2 = variance of the excluded volume fraction,

v_1 = volume of a large particle,

V_s = sample volume,

V_1, V_m = volume, of large and medium particle respectively.

V_{m+s} = volume available to medium and small particles alike,

$E(V_{m+s})$ = expected value of V_{m+s} ,

X, Y = volume fraction, of large and medium particles in a mixture respectively,

ω = the ratio, diameter of medium particle to diameter of large particle.

In order to test the mathematical model, experiments were performed in a horizontal rotating drum mixer (9" long and 8" dia. of standard steel pipe, with the thin steel flights at the longitudinal direction), two types of particle were used – glass beads of sieve fraction –24/+28, –32/+35 and –42/+48, and silica gel of –14/+16 –20/+24 and –28/+30. The mixer was kept at a constant speed of 36 rpm. Samples were drawn after the mixer rotated 900 revolutions, it was claimed that the mixture reached the equilibrium state. Theoretical and experimental evaluations of sample variance were computed and shown good agreement, and significantly better than those results obtained from Buslik's model. It was concluded that the developed model could be extended to any number of sizes, however, the complexity of the equations increases rapidly.

Hogg, Cahn et al (42) studied the mixing of powders in a horizotal rotating drum mixer (4" diameter, 16" long made of perspex tube) with two identical materials – Cataflex '202' and yellow flash beads (75-105μ) with different refractive index. It was proposed that when two components loaded end to end in such a mixer, the mixing could occur only by diffusion. Therefore, Fick's 2nd law of diffusion was analogous to such an ideal mixing system. The degree of mixing was used of the type proposed by Lacey and the diffusion equation was solved for σ^2 as,

$$\sigma^2 = \frac{1}{2L} \left\{ \frac{L}{2} \ erf^2 \left(\frac{L}{\sqrt{4(DN)}} \right) - \frac{4(DN)}{(2\pi)^{1/2}} \ erf \left(\frac{L}{2(2DN)^{1/2}} \right) \right.$$
$$\left. \times \left[1 - \left(\frac{2}{\pi} \right) exp \left(\frac{-L^2}{16DN} \right) \right] \right\} \qquad (2.52)$$

Where,   N = number of revolutions of the mixing drum,

L = the length of the mixer, and

D = the diffusion coefficient (Fick's 2nd low).

The authors concluded that by application of the equation of diffusion, theoretical expressions for the kinetics of the process were derived. The progress of mixing was following by a sampling and assaying technique which allowed quite accurate determinetion of concentration with little disturbance of the charge. Excellent agreement was obtained in comparing the experimental data with the theoretical prediction.

A model which was completely analogous to the diffusional mixing mechanism was developed, for the simulation of the mixing process by Cahn (14) and Cahn and Fuerstenau (15). The experimental work was performed in a 8" long and 4" dia. barrel mixer with 1/4" black and white Lucite beads. The mixer was divided into 16 equal sections. The individual particle movements along the mixer axis were observed into twenty load-speed combinations (the loads and speeds were ; 30, 40, 50, 60% of the total mixer volume and 20, 30, 40, 50, 60% of the mixer critical speed); each for at least 500 revolutions. The relationships for the diffusion coefficient were derived from Fick's 2nd law and diffusion coefficients obtained from the simulation and from the actual mixing process were shown good correction. The authors stated that the probability distribution of particle movements may be accurately simulated by the use of Monte Carlo techniques and the digital computer. These techniques are the valuable tools for study the mixing as the computer allows a rapid analysing the effects on the variation of mixing parameters, meanwhile, leads to a better understanding of the mechanism of mixing itself.

Müller and Rumpf (55) studied the axial mixing in the horizontal drum mixers. The axial motion of particles could be generally represented by the Kolmogorov's 2nd equation. The experimental work performed in the 190mm dia. with various length mixers (from $L/D = 1$ to 2.5). The materials used were copper and nickel powders of the same particle size ($300-400\mu$) and the ration of the components was kept at $1:7.7x10^{-3}$ by wt. In conjunction of this the materials having the differences in size and density also studied by using iron powder, quartz sand and limestone with sizes of $150-200, 300-400, 630-750\mu$ combinations. The measured standard deviation was used for their degree of mixing. This study was an attempt to apply the Kolmogorov's 2nd equation into a mixing system for describing the axial motion of particles. Several cases were discussed, in particular, the Fick's

2nd law of diffusion was introduced and applied into their system.

Harnby (38) compared the performance of some industrial solid mixers using the 'segregating materials'. A'discharge profile'was proposed and described that it has advantage for dealing with strongly segregating materials over the traditional statistical methods for measuring the uniformity of mixture. The materials used were millet (2057-1676μ) and salt (699-178μ) in four types of mixer, namely, nautamixer, ribbon blender, rotocube and V-mixer with the range of working capacities of 0.41 to 1.77 ft^3. In addition to the theories of segregation proposed by earlier authors, such as, vibrational segregation by Brown (11), pouring segregation by Williams (88) and percolation, free flight segregation by Ashton et al (4), Harnby proposed that the segregation occurs by the similar mechanisms as mixing, i.e. diffusive, shearing and convective segregations. Of these mechanisms, that convective was described most likely to minimise segregation whilst mixing in progress.

Lloyd and Yeung (51) proposed the between sample variance is a simpler and universal definition of the degree of mixing. This proposal has the virtue of simplicity, but the disadvantage that there is no convenient index on a scale between 0 and 1 as other more complicated froms such as Lacey's proposal.

Many other proposals on the degree of mixing had been published before 1958, which were surveyed by Weidenbaum (84). It seems little need for one to review those proposals here again.

2.3. THE THEORY OF MIXING – RATE AND MECHANISMS

Although Oyama (57) did not put forward a rate equation for powder mixing, but he did state the mechanisms of mixing in the horizontal drum mixer. He considered three stages of mixing ;
(1) by the shearing produced in the materials by the motion of the drum,
(2) by the shearing produced by either the parabolic fall or turbulent drift of particles which have departed from the circular motion, and
(3) by the shearing resulting from spiral motion of the particles.

These proposals by Oyama can only apply to mixing in a radial direction and are clearly not universal.

A three dimensional shuffling mixing operation was considered by Brothman et al (10). When a mechanical agitation acted on two separates material layers, the surface of separation between the layers

would be extented. Let S_p be the maximum theoretical possible surface of separation, and a proportion S/S_p had been developed a time t. Then any given increment of t would be proportional to the difference between the instantaneous magnitude of the surface of separation S at $(t + 1)$ time. The relation could be written as :-

$$\left(\frac{S}{S_p}\right)_{t+1} = \left(\frac{S}{S_p}\right)_t + \Phi\left[1 - \left(\frac{S}{S_p}\right)_t\right] \tag{2.53}$$

This equation had been mathematically treated and the final equation is given,

$$(P_t)_E = 1 - \left\{\exp\left[- kS_p(1 - e^{-tc})\right]\right\}^{\nu/V_o} \tag{2.54}$$

Where, $(P_t)_E$ = proportion of equal size volume units containing mixture after time t at satisfactory mixing end point,

k, c, Φ = proportional constants in Eq.(2.53) and Eq.(2.54)

ν = No. of units of volume containing at least one element of one of the components, and,

V_o = volume of sample.

If Eq.(2.54) applied to a real case, the time required for perfect mixing could be estimated. However, the authors did not verify the validity of the equation.

Coulson and Maitra (21) extended the theory of Brothman et al for mixing of binary components. The rate equation proposed was analogous to the diffusion process, hence,

$$\frac{dw}{dt} = k \, dc = k(c_o - c) \tag{2.55}$$

The unit weight w could be replaced by unit surface S transferred (diffusion), so that Eq.(2.55) becomes,

$$\frac{dS}{dt} = k(S_o - S) \tag{2.56}$$

or, $S = S_o(1 - e^{-kt}) \tag{2.56'}$

Then let, $n_o = S_o/s = V_o/v$, and $n = S/s = V/v$

so that, $n = n_o(1 - e^{-kt}) \tag{2.56''}$

Where,   s = minimum boundary surface which is measurable,

v = volume of a sub-volume.

Hence nv/V would be the fraction of the system which is mixed,

the fraction unmixed is : $n_0 v/V - nv/V = x/100$,

so that $$v/V(n_0 - n) = v/Vn_0(1 - e^{-kt}) = x/100$$

or,

$$t = \frac{1}{k} \ln \frac{100}{x} \qquad (2.57)$$

Where x is the percentage of unmixed.

The plot of $100/x$ vs. t was nonlinear at low values of time and so the theory still leaves much to be desired before the theory can be accepted.

Visman and Van Krevelen (81) reploted the data of Coulson and Maitra. Using $(100 - x)$ on the probability scale and time t on the arithmetic scale. Thus, if there was a straight line from such a plot, its equation would be,

$$(100 - x) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{t} (e^{-y^2/2}) dy \qquad (2.58)$$

Where, y is a dummy variable.

Differentiating this equation with respect to t gives,

$$\frac{d(100 - x)}{dt} = \frac{1}{\sqrt{2\pi}} e^{-t^2/2} \qquad (2.59)$$

Which was the rate equation giving a straight line on such a graph.

Lacey (49) pointed out that simply finding an exponential law of mixing rate in practice must not be taken by itself as substantiation of a theory without considering its range of agreement . The theory of mixing based on the mixing mechanisms were described as,

1). Convective mixing, transfer of groups of adjacent particles from one location in the mass to another,

11). Diffusive mixing, the distribution of particles over a freshly developed surface.

111). Shearing mixing, the setting up of slipping planes within the mass.

In all cases, since mixing tends towards an equilibrium state, an exponential approach to ideal mixing may involve one or more of the three mechanisms outlined above. It was assumed that

$$M = 1 - e^{-kt} \qquad (2.60)$$

Where k is the slope which depends on the effectiveness of the machin and the physical properties of the materials. If the rate of mixing was due to diffusive mixing mechanism, it should be independent of sample size; and if the rate was depended greatly on sample size, convective mixing predominated.

Fig.(2.5)

The diffusive mixing had been considered to occur when particles were spread over the freshly exposed surface within the mixer. This is clearly similar to molecular or thermal diffusion. The phenomena of the particles random motion within the mixing plane is analogous to the Fick's second law of diffusion for gases and liquids.
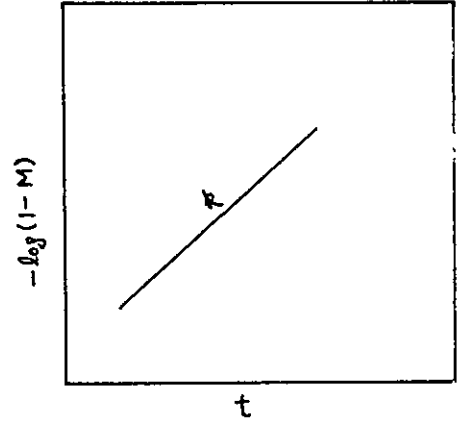
$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2} \qquad (2.61)$$

From this equation, a rate equation can be developed for the case of a horizontal cylinder rotating on its own axis. The materials were loaded end to end, with a vertical common surface. The mixing mechanism could be considered as analogous to the motion of molecules of two different gases. Therefore Eq.(2.61) can be solved to yield:-

$$(c - c_0) = \frac{2}{\pi} \sum_{n} \exp\left(- \frac{n^2 \pi^2 Dt}{L^2}\right) \frac{\sin(nc_0 \pi)}{c} \cos(n\pi x) \qquad (2.62)$$

from the first moment of the mean and the second moment of the variance, Eq.(2.62) can be expressed, with n = 1;

$$\sigma^2 = \frac{2}{\pi} \exp\left(- \frac{2\pi^2 Dt}{L^2}\right) \sin^2 c_0 \qquad (2.62')$$

It is clear that $-\log \sigma^2$ vs. t will give a straight line. The initial portion of the plot will be curved. Lacey attributed this to transients due to one of the components having

to travel to the middle of the mixer. Lacey stated that the straightness of such a plot implied that either D is independent of position, or else there is a good circulation which evens out variations in D. Curvature of the graph would indicate regions of poor mixing which would lengthen the mixing time. This would be due to poor circulation and such a region could then be looked for and the defect remedied.

Weidenbaum and Bonilla (82) by comparing solid mixing to diffusion in gases and liquids, obtained the initial mixing rate equation:-

$$\frac{d(\sigma/\sigma_e)}{dt} = k \left(1 - \frac{\sigma}{\sigma_e}\right) \qquad (2.63)$$

or,

$$\ln\left[\frac{1}{1 - (\sigma/\sigma_e)}\right] = \ln\left[\frac{1}{1 - (\sigma/\sigma_e)}\right]_0 + kt \qquad (2.63')$$

The graph of $\ln\left[1/(1 - \sigma/\sigma_e)\right]$ vs. t is plotted from experimental results:-
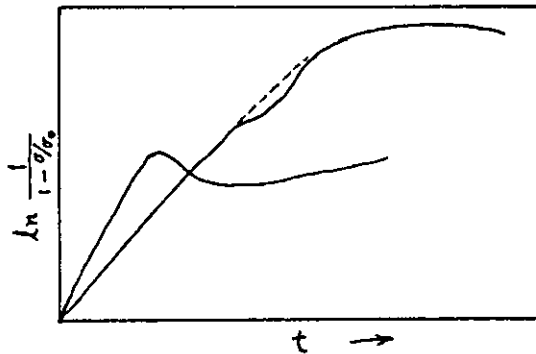
Fig.(2.6)

The intercept on the ordinate was found to depend on the loading conditions. The graphs in Fig.(2.6) were initially straight lines after which the curves tend to be irregular. The authors suggested that straight line part of the curve corresponded to randomisation, since the derivation of the equation assumed a driving force proportional to the separation from the current to the equilibrium state. The later irregularities in the curves indicated that some driving force which caused unmixing was becoming more important. This was believed to take place when large quantities of either components reached the end walls. This was

observed as axial segregation which opposed mixing. Therefore a rate mechanism based on the randomisation could not hold.

Oyama and Ayaki (62) assumed that the mixture could be divided into N cells of equal volume. These cells were numbered from 1 to N, and a Markov chain process could be applied. When the mixer had operated for (t-1) intervals, the particle fraction of cell i for the A particles is $x_i^{(t-1)}$ and after one more interval the fraction is $x_i^{(t)}$. Hence if the transition probability of the particles transferred from cell j to cell i is $P_{i,j}^{(t)}$, $x_i^{(t)}$ and $x_j^{(t-1)}$ ($j = 1,2,\ldots,N$) are related by the expression

$$x_1^{(t)} = P_{1,1}^{(t)} x_1^{(t-1)} + P_{1,2}^{(t)} x_2^{(t-1)} + \ldots\ldots\ldots + P_{1,N}^{(t)} x_N^{(t-1)}$$

to express this for the whole mixer, the transition probability matrix $\left[D_{i,j}^{(t)}\right]$ could be used. Then the particle fraction in each of the cell after time t could be expressed as

$$
\begin{bmatrix} x_1^{(t)} \\ x_2^{(t)} \\ \cdot \\ \cdot \\ \cdot \\ x_N^{(t)} \end{bmatrix}
=
\begin{bmatrix} x_1 \\ x_2 \\ \cdot \\ \cdot \\ \cdot \\ x_N \end{bmatrix}_{(t)}
=
\left[D_{i,j}^{(t)}\right]
\begin{bmatrix} x_1 \\ x_2 \\ \cdot \\ \cdot \\ \cdot \\ x_N \end{bmatrix}_{(t-1)}
\qquad (2.64)
$$

Where,

$$
\left[D_{i,j}^{(t)}\right] =
\begin{bmatrix}
P_{1,1}^{(t)} & P_{1,2}^{(t)} & \ldots\ldots\ldots & P_{1,N}^{(t)} \\
P_{2,1}^{(t)} & P_{2,2}^{(t)} & \ldots\ldots & \ldots P_{2,N}^{(t)} \\
\cdot & \cdot & & \cdot \\
\cdot & \cdot & & \cdot \\
\cdot & \cdot & & \cdot \\
\cdot & \cdot & & \cdot \\
P_{N,1}^{(t)} & P_{N,2}^{(t)} & \ldots\ldots\ldots & P_{N,N}^{(t)}
\end{bmatrix}
$$

and, $$\sum_n P_{i,j}^{(t)} = 1$$

The transition probability $P_{i,j}$ showed the mixing characteristic in each of the cells and this was usually only related to the particle distribution at a previous time, but not to the previous state of mixing.

When the particles have the same physical properties and

the mixer used a constant speed, then the transition probability $P_{1,J}$ becomes independent of time. Hence,

$$\left[D_{1,J}^{(1)} \right] = \left[D_{1,J}^{(t)} \right] = \left[D_{1,J}^{(N)} \right] = \left[D_{1,J} \right]$$

$$= \begin{bmatrix} P_{1,1} & P_{1,2} & \cdots\cdots & P_{1,N} \\ P_{2,1} & P_{2,2} & \cdots\cdots\cdot & P_{2,N} \\ \cdot & \cdot & & \cdot \\ \cdot & \cdot & & \cdot \\ \cdot & \cdot & & \cdot \\ P_{N,1} & P_{N,2} & \cdots\cdots\cdot & P_{N,N} \end{bmatrix}$$

$$\sum_{1=1}^{N} P_{1,J} = 1, \qquad \sum_{J=1}^{N} P_{1,J} = 1$$

Therefore, the particle fraction in each cell becomes,

$$\begin{bmatrix} x_1 \\ x_2 \\ \cdot \\ \cdot \\ \cdot \\ x_N \end{bmatrix}_{(t)} = \left[D_{1,J} \right] \begin{bmatrix} x_1 \\ x_2 \\ \cdot \\ \cdot \\ \cdot \\ x_N \end{bmatrix}_{(t-1)} = \left[D_{1,J} \right] \begin{bmatrix} x_1 \\ x_2 \\ \cdot \\ \cdot \\ \cdot \\ x_N \end{bmatrix}_{0}$$

Thus the particle distribution along the mixer at any time t could be computed if the initial mixing condition is known. However, no practical application of this theory was attempted.

Oyama and Ayaki also considered the mixing mechanism as analogous to one-dimensional Brownian motion.

The rate equation (2.62) proposed by Lacey was considered. It could be expressed simply as :-

$$\sigma^2 \propto e^{-t} \tag{2.65}$$

The authors stated that it would be better to use the difference of $\sigma^2 - \sigma_R^2$ instead of σ^2, thus

$$(\sigma^2 - \sigma_R^2) \propto e^{-t} \tag{2.65'}$$

The rate of change with time of the variance of the particle

fraction x could be considered as proportional to the difference between σ^2 and the σ_R^2, as

$$\frac{\partial(\sigma^2)}{\partial t} = -\phi(\sigma^2 - \sigma_R^2) \tag{2.66}$$

Hence,

$$\ln\frac{\sigma^2 - \sigma_R^2}{\sigma_o^2 - \sigma_R^2} = \ln(1 - M) = -\phi t$$

or

$$\log(1 - M) = -0.4343\,\phi t$$

$$\sigma^2 = (\sigma_o^2 - \sigma_R^2)e^{-\phi t} + \sigma_R^2$$

$$= C\,e^{-\phi t} + \sigma_R^2 \tag{2.67}$$

It was assumed that after a certain time of mixing, the frequency distribution of the particle fraction and each point in the mixture followed approximately a normal distribution. The graph of $\log(1-M)$ vs. time t was a straight line for these cases for which the proposed rate equation applied, as shown in Fig.(2.7)
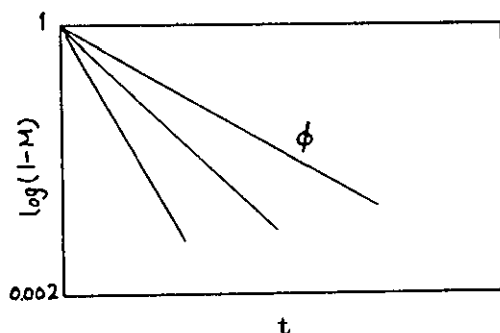
Fig.(2.7)

However, where two difference materials were mixed, the rate equation did not hold any more, as shown in Fig.(2.8)
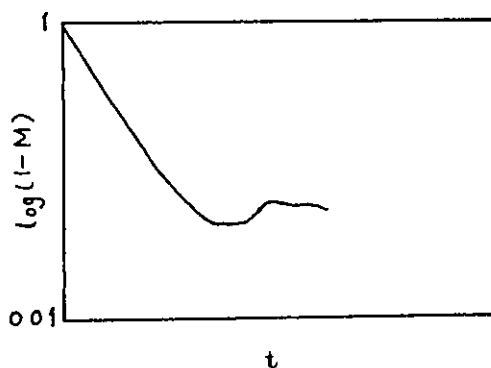
Fig.(2.8)

The authors pointed out that the radial mixing in such a drum is extremely rapid in comparison with axial mixing, so that the controlling factor in determining of mixing time is the coefficient of axial mixing.

Yano et al (93) summarised the results of mixing in various mixers. It was suggested that the mixing mechanism could be classified into three stages as shown in the Fig.(2.9) (which was also mentioned under Section 2.2). A curved section is found at the initial stage of mixing (region 1), followed by a section where with σ proportional to $e^{-\phi t}$ (region II), after that there is a
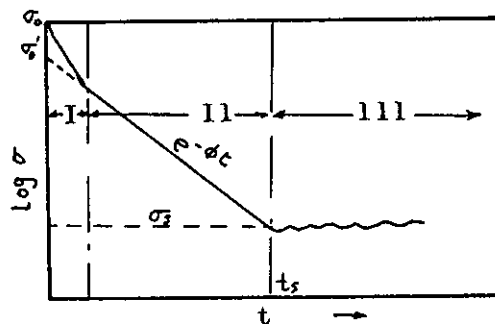
Fig.(2.9)

section where σ takes on its minimum value σ_s within a certain finite mixing time t_s, beyond this point, mixing does not proceed any further even if the mixing time is extended. The value of σ rises and falls (region III)

It was found that the mixing of powders proceeded by means of overall mixing, brought about by the movement of the powder particles in small clumps (convective mixing), and local mixing brought about by positional interchange (diffusion) of the particles. It may be assumed that overall mixing is dominant in region 1 and that in region II localised mixing action is dominant.

In region III, if there is no difference between the particle size of the components, the mixture is in a state of oscillatory equilibrium controlled by the mixing action. Any separating action would be repeated cyclicly. The value of σ consequently showed a rise or fall.

When the components have different particle sizes, region II is very short, the separating action works the more strongly

in region III, a good state of mixing could not be obtained.

In general the rate equation could be experssed with a new σ_o' , so that,

$$\sigma = \sigma_o' e^{- \phi t} \tag{2.68}$$

Where, $\phi =$ coefficient of mixing velocity, and

$\sigma_o' =$ standard deviation at t=0 from Fig.(2.9).

The empirical equation obtained from the experimental results was given as :-

$$\phi (F/V) = k_1 \exp -k_1(\ln\frac{N}{N_{\phi M}}) \tag{2.69}$$

$$\phi (N) = K_2 \exp -k_2(F/V)x10^2 \tag{2.70}$$

Combining Eqs.(2.69) and (2.70),

$$(N,F/V) = K \exp\left[-k_1(\ln\frac{N}{N_{\phi M}}) - k_2(F/V)x10^2\right] \tag{2.71}$$

Where, $N_{\phi M} =$ speed of rotation at maximum ϕ, and

K_1 , K_2 , k_1 , k_2 = constants.

(other symbols were cited in Section 2.2.)

Equation (2.71) only considered materials having the same physical properties. If materials having different physical properties are used, the characteristics of powder C_R of both components should be considered. Therefore Eq.(2.71) was modified to,

$$\phi = K'(\Delta C_R)^n \exp\left[-k_1'd_{av}(\ln\frac{N}{N_{\phi M}}) - \frac{k_2'}{d_{av}^{1/2}} (F/V)x10^2\right]$$
$$\tag{2.72}$$

Where, K', k_1', k_2' are modified proportional constants.

From the experimental results, the authors claimed that the proposed empirical equations were valid.

Gayle et al (33) proposed the rate equation of mixing as the rate of change of the degree of segregation, as,

$$-\frac{dS}{dt} = k (S)^n \tag{2.73}$$

Where, n is a constant indicating the order of the process, k is the rate constant.

The authors explained that most of the workers have stated or assumed the mixing as following a first order equation with n=1, or Eq.(2.73) becomes,

$$\frac{dS}{dt} = -kS \qquad (2.74)$$

$$\ln S = -kt + k_1 \qquad (2.74')$$

However, the plot of lnS vs. t did not yield a straight line over the entire range as shown in Fig.(2.10),
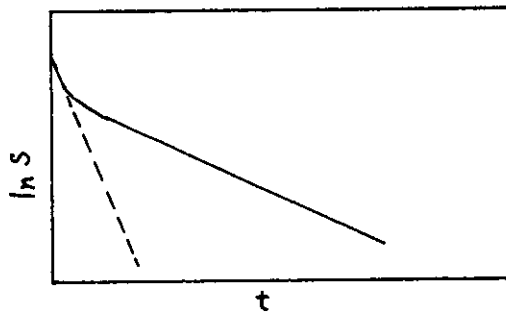
Fig.(2.10)

It appeared to indicate an initial period of rapid mixing, after which the rate dropped somewhat and the slope became apparently linear.

When n=2, Eq.(2.73) becomes,

$$\frac{dS}{dt} = -kS^2 \qquad (2.75)$$

$$1/S = kt + k_1 \qquad (2.75')$$

The plot of 1/S vs. t failed to yield a straight line over the entire range, giving instead an initial linear portion followed by a markedly nonlinear portion, as shown in Fig.(2.11).
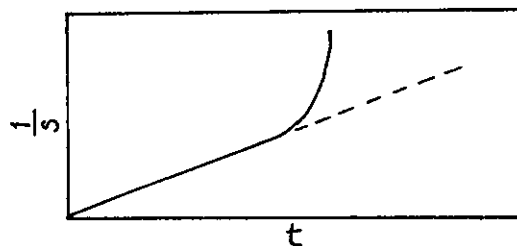
Fig.(2.11)

It was considered that taken together, those plots could be interpreted to indicate the particular mixing mechanism used. The mixing process was initially a second-order process, followed by a first-order process. This, the authors stated, seemed reasonable in view of the nature of the mixing process. However, the authors explained further that another possible interpretation is that the process is actually first order and rapid mixing observed during the early stage is at least partly the result of taking a small number of the samples from fixed locations within the mixer.

Gayle et al (34) further studied the mixing of powders in the horizontal rotation drum mixers. An empirical rate equation was proposed which involving the geometric relations of the drums. Thus,

$$\frac{dS}{dt} = -k'D^{1.45} S^2/L^2 \tag{2.76}$$

Where, D and L are the inside diameter and the length of the drum mixer in inches, k' is a constant as before.

Solving for S, Eq.(2.76) becomes,

$$\frac{1}{S} = 1 + k'D^{1.45}t/L^2 \tag{2.76'}$$

The proposed rate equation showed good agreement with the experiments, a straight line plot could be obtained from $1/S$ vs. $D^{1.45}t/L^2$.

Rose (66) considered that mixing and demixing may occur at the same time. The rate of mixing should combine these two potentials – mixing and demixing. As the rate equation was considered to be the first order, it was stated that,

$$\frac{dM}{dt} = A(1 - M) - B\phi \tag{2.77}$$

Where,   A = constant which depends on all the geometrical and physical parameters of the mixer, and the powders on which the rate of mixing depended.

B = also a constant which dependent on densities, size distributions and shape of particles of the two components.

ϕ = constant which depends on the distribution of

the two components in the mixture,

M = the degree of mixing.

From Eq.(2.77), A(1 - M) leads the mixing potential and
B leads the demixing potential. The difference will be the net
mixing rate in a particular system. The demixing potential could
be positive or negative. The sign largely depended on the method of
loading for the two different sized components. Rose showed in
the sketches the methods for loading,

Dense component below   Dense component above
ϕ + Ve (A) ϕ - Ve (B)

Thus, Φ is positive for one fully segregated condition,
passes through zero for the completely mixed condition. This could
be summaried as,

$$M = 0, \qquad \Phi = 1$$

$$M = 1, \qquad \Phi = 0$$

$$M = 0, \qquad \Phi = -1$$

A simple equation which fulfil these conditions, as

$$M = 1 - \Phi^2 \qquad (2.78)$$

Differentiating this equation with respect to t,

$$-2\Phi \frac{d\Phi}{dt} = \frac{dM}{dt} \qquad (2.78')$$

Substituting Eq.(2.77) and Eq.(2.78) to Eq.(2.78') becomes,

$$-2\phi\frac{d\phi}{dt} = A\phi^2 - B\phi \quad ,$$

or,

$$-2\frac{d\phi}{dt} = A\phi - B \qquad (2.79)$$

Solving for ϕ,

$$A\phi - B = Ce^{-At/2} \qquad (2.80)$$

Again from Eq.(2.78)

$$\pm (1 - M)^{1/2} = C/Ae^{-At/2} + B/A \qquad (2.81)$$

As demixing potential can be positive or negative, there-

53

fore two equations are obtained:-

$$\phi + Ve \qquad M = 1 - \left[(1 - B/A)e^{-At/2} + B/A \right]^2$$

$$\phi - Ve \qquad M = 1 - \left[B/A - (1 - B/A)e^{-At/2} \right]^2 \qquad (2.82)$$

Curves corresponding to these equations are plotted as Fig.(2.12).

Although these equations explain the behaviour of the material in a mixer with fair precision, they are not completely
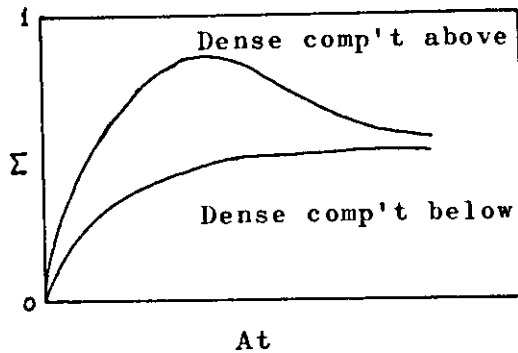
Fig. (2.12)

satisfactory for application to the whole process. The author realised that no real machine leads to a perfect mixture as M = 1, therefore he introduced an 'intrinsic efficiency' of the mixer, in to the equation. This intrinsic efficiency η is a measure of the perfection of the machine as a mixing device and is independent of the nature of the charge in the mixer. Thus, the equations become,

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

$$\text{and} \qquad M = \eta \left\{ 1 - \left[B/A - (1 + B/A)e^{-At/2} \right]^2 \right\} \qquad (2.82)$$

Finally Rose noticed that when the mixing process is carried out for an infinite time, the degree of mixing attained an equilibrium value Me, given by,

$$Me = \eta \left[1 - (B/A)^2 \right] \qquad (2.83)$$

Weydanz (87) considered the mixing process is a shuffling operation. The mixing model was given as in Fig.(2.13).

The materials are separated by a vertical line and the volumetric proportion of one of the constitents is equal to y on the lift-hand side of that line and to (1-y) on the right-hand side.
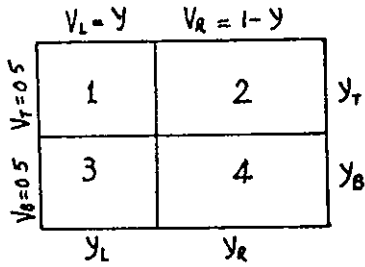
Fig.(2.13)

Weydanz then considered mixing effect combined with demixing effect. In the vertical plane, a small volume v is moved upwards and is exchanged with an equal volume from the upper region. At the same time a volume of the pure heavier component from the upper region exchanged with an equal amount of the other component in the lower region. If these two processes are combined and the resulting equation is sol-ved, then the standard deviation of the mixture is given by the equation,

$$\sigma_v = 0.5(c/b)(1 - e^{-4bt}) \tag{2.84}$$

and
$$\sigma_{v_o} = 0.$$

$$\sigma_{v_\infty} = \sigma_{M\alpha} = 0.5(c/b)$$

Where, $b = dv/dt$, the rate of mixing,

c = a measure of the demixing rate.

In the horizontal plane, the demixing is absent and only mixing takes place. In this case the standard deviation would be,

$$\sigma = \left[y(1 - y) \right]^{1/2} e^{-at/y(1-y)} \tag{2.85}$$

and
$$\sigma_o = y(1 - y), \tag{2.85'}$$

$$\sigma_{\infty} = 0.$$

Where, a = the rate of mixing for this particular case,

y = the distance from the dividing line.

Combining Eq.(2.85) and Eq.(2.85'), then a new form is obtained,

$$M = \frac{\sigma}{\sigma_o} = \left[e^{-2At} + \sigma_M^2 (1 - e^{-BAt})^2 \right]^{1/2} \tag{2.86}$$

Where, $A = (1/\sigma_o^2)a,$

$B = 4\sigma_o^2(b/a),$ and

$\sigma_M = (0.5/\sigma_o)(c/b)$

It can thus be seen that mixing here is an exponential process in which the standard deviation may pass through a minimum, i.e., the degree of mixing through a maximum. If $B = 2$, then the resulting equation can be simplified to,

$$M = \left[e^{-2At} + \sigma_M^2 (1 - e^{-2At})^2 \right]^{1/2} \tag{2.87}$$

This was the equation that Weydanz had used to measure the rate of mixing.

Otake et al (56) studied the mixing of solid in the vertical tank with paddle type agitator. The mixing mechanism was considered to be analogous to the radial diffusion or radial heat transfer. This was based on the equation,

$$\frac{\partial C}{\partial t} = D_s \left(\frac{\partial^2 C}{\partial r^2} + \frac{1}{r} \frac{\partial C}{\partial r} \right) \tag{2.88}$$

(symbols used as before)

with certain boundary conditions Eq.(2.88) can be solved, as:-

$$C = C_0 + \frac{2R}{Ro^2} \sum_{k=1}^{\infty} \frac{J_1(m_k R) J_0(m_k r)}{m_k J_0^2(m_k R_0)} \exp(-D_s m_k^2 t) \tag{2.89}$$

The theoretical variance can be expressed as:-

$$\sigma^2 = \frac{\int_0^{R_0} (C - C_0)^2 2\pi r \rho H \rho \, dr}{\int_0^{R_0} 2\pi r \rho H \rho \, dr} = \frac{\int_0^{R_0} (C - C_0)^2 r \, dr}{\int_0^{R_0} r \, dr} \tag{2.90}$$

Substituting Eqs.(2.89) into (2.90), with using the condition of Bassel function first kind zero order, the final solution obtained:-

$$\sigma^2 = \frac{4R^2}{R_0^4} \sum_{k=1}^{\infty} \frac{J^2(m_k R_c)}{m^2 J_0^2(m_k R_0)} \exp(-2D_s m_k t) \tag{2.91}$$

It was described that Eq.(2.91) can be used to measure the rate of mixing, however, no practical work was applied to this equation. Further, The authors used dimensional analysis to define the relationship between the variables,

$$\frac{\sigma}{\sigma_0} = \Psi(Nt, \, D_1 N^2/g, \, D_T/D_1, \, H_p/D_1, \, R_1/D_1) \tag{2.92}$$

Where,   N = the number of revolutions of the agitator, and

t = the mixing time.

From the experimental results the empirical equation was given as:-

$$\frac{\sigma}{\sigma_0} = 0.676(Nt)^{-.48}(D_1 N^2/g)^{-.16}(D_T/D_1)^{.69}(R_1/D_1)^{.57} \tag{2.93}$$

If Nt is greater than 500, the empirical equation becomes,

$$\left(\frac{\sigma}{\sigma_\bullet}\right)\left(\frac{1}{Nt_c}\right) = 6.18 \times 10^{-6}(D_1 N^2/g)^{-.52}(D_T/D_1)^{.8} \qquad (2.94)$$

In the same manner, another empirical equation for D_S developed that,

$$\frac{D_S}{ND_1} = 5.75 \times 10^{-3}(D_1 N^2/g)^{.18}(D_T/D_1)^{.46} \qquad (2.95)$$

Good agreement was obtained between the empirical equation and the experimental results.

Fisher (31) studied the rate of diffusive mixing of powders in a horizontal rotating drum mixer. He found that the radial mixing was very rapid and the mixing rate was controlled by axial mixing. The diffusion equation in cylindrical coordinate,

$$\frac{\partial c}{\partial t} = D_x \frac{\partial^2 c}{\partial x^2} + D_r\left(\frac{\partial^2 c}{\partial r^2} + \frac{1}{r}\frac{\partial c}{\partial r}\right) \qquad (2.96)$$

could then be simplified :-

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2} \qquad (2.97)$$

Eq.(2.97) is a standard equation and can be solved if the boundary conditions are known :-

$$\frac{c}{c_0} = \frac{d}{L} + \frac{4}{\pi}\sum\left[\frac{1}{n}\left(\cos\frac{n\pi x}{L}\cos\frac{n\pi}{2}\sin\frac{n\pi d}{2L}\right)\exp\left(\frac{-n^2 D\pi^2 t}{L^2}\right)\right]$$

$$(2.98)$$

Where d is the thickness of the tracer at the initial condition, L is the total length of the mixer.

The variance could be defined as :-

$$\sigma^2 = \mu_2' - \mu^2 \qquad (2.99)$$

Where, $\quad \mu_2' = \int x^2\left(\frac{c}{c_0}\right)dx$

$\quad\quad \mu = L/2$

The variance was obtained for $L = 1$,

$$\sigma^2 = 0.08333 + \frac{4}{\pi^2}\sum_n\frac{1}{n^2}\exp(-n^2 x^2 Dt)\cos\frac{n\pi}{2}\cos(n\pi) \quad (2.100)$$

This was used to calculate the diffusion coefficient D, however, Fisher noticed a lack of agreement between the observed and the calculated results. This was thought to be due to inaccurate application of the diffusion equation to a discontinuous medium with the presence of convective current at the ends of the mixer.

Carley-Macauly and Donald (18,19) and Carley-Macauly (20) proposed a simple rate equation dominated by diffusive mixing,

$$\log V_p = -\frac{t}{\tau} + C \qquad\qquad (2.101)$$

Where, $\quad \tau$ = the inverse mixing rate,

$\quad\quad\quad$ t = the number of revolutions, and

$\quad\quad\quad$ C = a constant. $(V_p = V - \sigma_R)$

They proposed that the total mixing rate in a given mixing body could be compounded from the rate measured in separate directions. The individual values of V_p, each of which obeys a law of the type of equation (2.101) could be added. The fastest of these individual rates is that in the plane of particle circulation. The mixing rate also varies with the speed of rotation of the mixer, materials, type of mixer, and method of loading. Straight line plots from the experimental results were obtained in most conditions except at the initial stage, even in the mixer with a baffle where the mixing action was predominately that of shear.

Roseman and Donald (67, 68) and Roseman (69) stated that demixing will accompany the mixing process, when the mixture was composed by two or more materials of different properties. If the ratio of differently sized components exceed 1.2 (for binary components), the small particles might slip into the voids of the large particles and cause serious demixing. The tendency increases rapidly with increased size ratio until the 'critical ratio' was attained, where there was no mixing at all. The similar demixing also existed when the density difference exceeded 1.2

The two dimensional mixing theory as named radial and axial mixing. The radial mixing is very rapid, and the mixing is controlled mainly by the axial mixing. The demixing tendency caused the radial bands to be formed along the axis of the mixer, the formation of bands was thought to be due to the different angle of repose

of the components.

Strel'tsor et al (78) proposed a first order rate equation
as :-

$$M = e^{-At} \qquad (2.102)$$

Where,   A = a constant which depends on the physical properties
           of materials and mixing conditions, this was called
           the rate coefficient.

The experimental results showed that during the starting up
of the mixer, the mixing rate was lower than when it was running
at constant speeds. The rate coefficient A was shown not
to be constant, but a function of t, as proposed :-

$$A = A_{st}(1 - e^{t/t_s}) \qquad (2.102')$$

Where A_{st} is the value of A under stable mixing conditions, t_s
is a multiple of the start up period.

As A_{st} decreased, t_s increased with increases on the degree of
loading of the mixer. A_{st} increased and t_s decreased when the rotor
velocity of the mixer (n) increased to 300rpm, but it remained
constant at high-er n. A_{st} was almost independent of the particle
size.

Hogg, Cahn et al (42) used Fick's 2nd law of diffusion in
the mixing of an ideal system. This was given as :-

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

or,

$$\frac{\partial c(x,N)}{\partial t} = D \frac{\partial^2 c(x,N)}{\partial x^2} \qquad (2.103)$$

Where N is the number of revolution of the mixer.

Eq.(2.103) with the boundary conditions can be solved as :-

$$c(x,N) = \frac{1}{2} + \frac{2}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{(2n-1)^2} \exp\left[\frac{-(2n-1)^2 \pi^2 DN}{L^2}\right]\left[\sin\frac{(2n-1)\pi x}{L}\right] \qquad (2.104)$$

With the relation used by Fisher, the variance σ^2 can be obtained
as,

$$\sigma^2(N) = \frac{2}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{(2n-1)^2} \exp\left[\frac{-2(2n-1)\pi^2 DN}{L^2}\right] \qquad (2.105)$$

It may be simplified by taking n = 1 to,

$$\sigma^2(N) = \frac{2}{\pi^2} \exp\left(-\frac{2\pi^2 DN}{L^2}\right)$$

or,

$$\ln\left[\sigma^2(N)\right] = \ln(2/\pi^2) - (2\pi^2 DN/L^2) \qquad (2.106)$$

The was the general theory which describe the diffusional mixing of ideal solids.

An experimental system was designed so that it approximated very closely to the ideal case of solid mixing and would be analogous to the gases and liquids mixing system. The validity of the theoretical arguments was shown by the excellent agreement with the experimental results.

Muller and Rumpf (55) stated that the actual mixing process may be described as a Markov process, as a stochastic process without after effect. A constant Markov process may be applied by the Second kolmogorov equation. This could represent the general mixing process, as:-

$$\frac{\partial q(x,t)}{\partial t} = -\frac{\partial}{\partial x}\left[T(x,t).q(x,t)\right] + \frac{\partial^2}{\partial x^2}\left[D(x,t).q(x,t)\right]$$

$$(2.107)$$

Where, $q(x,t)$ = the probability distribution of particles,

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

$T(x,t)$ = the transport coefficient.

However, when the components have the same physical properties or differ only by colour, the transport coefficient becomes insignificent, and diffusion coefficient becomes a constant, independent of time and position. The 2nd Kolmogorov equation was simplified to Fick's 2nd law of diffusion, giving,

$$\frac{\partial q(x,t)}{\partial t} = D\frac{\partial^2 q(x,t)}{\partial x^2} \qquad (2.108)$$

With certain boundary conditions Eq.(2.108) was solved as:-

$$q(x,t) = 2\cos\pi x\left[\exp(-Dt/L^2)\right] \qquad (2.109)$$

The variance of the concentration distribution was defined in a similar was to Fisher, and Hogg et al in the form,

$$\sigma(t) = \left[\int_0^\ell q(x,t)\,dx \right]^{\frac{1}{2}} = \sqrt{2}\, \exp(-\pi^2 Dt/L^2) \qquad (2.110)$$

The total standard deviation was taken the sum of the theoretical standard deviation and the random standard deviation, as

$$\sigma'_{ys}(t) = \left[2\,\exp(-2\pi^2 Dt/L^2) + \sigma'^2_{\xi}(t) \right] \qquad (2.111)$$

Where, $\sigma'_{ys}(t)$ = the relative total standard deviation,

$\sigma'_{\xi}(t)$ = the relative random standard deviation.

Eq.(2.111) was used to determine the diffusion coefficient D. Although the authors had considered the general mixing cases, however, for simplicity of the investigation, the experiments only considered the case in which Fick's 2nd law applies, and the diffusion coefficient D were determined in terms of the ratio of the length and diameter of the mixers. It was found that the diffusion coefficient D is almost constant and independent of the dimensions of the mixer.

The 2nd Kolmogorov equation was also considered in a very general way, by assuming the transport coefficient as an independent variable of x, t, and the probability density q(x,t) as subject to the nature of the condition applied. However, no conclusion was drawn on the validity of the equation.

2.4. MIXING EQUIPMENTS AND MATERIALS

(I) Mixing Equipments

In a recent paper by Lloyd and Yeung (51) classified the dry powder mixing equipments into three categories by their nature of motions, namely, tumbling, tumbling with shear and shearing mixers. The general recommendation for uses of those mixers in terms of material properties, was given and listed in Table 2.1.

Adams and Baker, Gray, Yano et al, Ashton and Valentin and Harnby have all tested various types of mixers. The equipment that has been investigated and reported in the literature has been summarised by Lloyd and Yeung and is listed in Table 2.2.

Most of the research work was concerned with mixers on a very scale, so far no actual relationships have been proposed on the

scale up problem. The relationships of capacity, dimension and power consumption for different types of mixers which could only be obtained from the manufacturer's specifications.

Table 2.1. Recommendations for Applications

Mixer	Powder
Tumbling mixer	Free-flowing-components of similar particle size
Tumbling mixer with shear	Slightly cohesive powder,free-flowing powders
Shear mixer	Cohesive powders,free-flowing,segregating mixtures

Table 2.2. Classificarion of Mixers

General	Type	Reference
Tumbling mixer	A. Horizontal cylinder	1,14,17,18,19,30,33,34 35,42,44,54,56,61,62,67 68,79,82,90,91,92,93
	B. Inclined cylinder	9,18,19,21
	C. Rotating cube	1,36,38,91,92,93
	D. Double cone	1,18,19,36,41,90,91,92 93
Tumbling mixer with shear	A. Horizontal cylinder with baffles or paddle	44,54
	B. Twin shell V-type	1,36,38,40,85,89,90,91 92,93
	C. Concrete mixer	36
	D. Hammer	36
Shear mixer	A. Ribbon	1,36,38,64,90
	B. Finger prog,pugmill type	3,28,53
	C. Kneadex	75
	D. Müller	36,75
	E. Centrifugal	75
	F. Reducing helix extruder	75
	G. Reciprocating conveyor	75
	H. Vertical agitator	43,56
	I. Propellent	41
	J. Lödige	63
	K. Vertical helical impeller	3,4,37,38
	L. Z-blade	3
Others	A. Continuous ribbon type	64
	B. Continuous flow type	50
	C. Circulation type	50
	D. Air mixer	4

(II) Materials

The materials used in over 50 published papers were summarised by Lloyd and Yeung, and is tabulated in Table 2.3. It is shown that the majority of the experimental work had consisted of a study of the mixing of two components of same narrow size ranges and was limited to free-flowing materials. The mixing of components with different sizes had been the subject of very few investigations.

Table 2.3.   Materials Used

Size (μ)		Material		Shape		Mixer	Ref
Component 1	Component 2	Component 1	Component 2	Component 1	Component 2		
7 000 2 100 1 700	7 000 2 100 1 700	red rice	rice			continuous type horizontal cylinder	72
5 000	5 000	bullet tapioca	bullet tapioca			wooden sphere	48
4 000	3 000	carbon black in natural polyethylene	natural polyethylene	cube	cube	double cone ribbon blender V type, rotating cube	1
3 500	1 350	limestone	limestone			square box horizontal cylinder	60, 58
3 500	570	limestone	limestone			square box horizontal cylinder	60 58
3 000	2 000	carbon black in natural polyethylene	natural polyethylene	cube	cube	double cone	40
2 380/1 450	1 000/500	sand	sand			continuous type horizontal cylinder	79
2 380/1 410	1 000/700	sand	sand			continuous type horizontal cylinder	79
2 380/1 410	500/350	sand	sand			continuous type horizontal cylinder	79
2 057/1 407	699/127	millet seed	salt	ellipsoid	cube	vertical helical impeller ribbon blender rotating cube V-type	38
1 500/800	1 500/800	witherite	sand			double cone air mixer vertical helical impeller Z blade	4
1 500/800	180/110	witherite	pyrites			double cone air mixer, vertical helical impeller Z blade	4
1 500/800	180/110	calcite	sand			double cone air mixer, vertical helical impeller Z-blade	4
1 300	1 300	limestone black	limestone white			horizontal cylinder with or without baffles	57
1 168/833	1 168/833	roofing granules	roofing granules			horizontal cylinder	33,34
991/701	991/701	sand	sand			horizontal cylinder	62
833/701	833/701	sand	sand			horizontal cylinder	62
850/600	850/600	sand	sand			horizontal cylinder, double cone inclined cylinder	18,19
833/175	833/175	sand	salt			vertical cylindrical tank with paddle agitator	80
833/175	833/175	sand	$K_2Cr_2O_7$			vertical cylindrical tank with paddle agitator	80
600/420	600/420	glass beads	glass beads	sphere	sphere	horizontal cylinder double cone inclined cylinder	18,19
600	160	coal	salt			inclined cylinder	21
600/150	600/150	coal	salt			inclined cylinder	21
600	210	coal	salt			inclined cylinder	21
600	300	coal	salt			inclined cylinder	21

* 3-component system   • 4-component system

Table 2.3.—continued

Size (μ)		Material		Shape		Mixer	Ref
Component 1	Component 2	Component 1	Component 2	Component 1	Component 2		
589 417	589 417	CaCO₃	sand			V-type	16
800–420	500 420	sand	sand			horizontal cylinder double cone inclined cylinder	18 19
495 351	495–351	sand	salt			vertical tank with paddle agitator	56
417 295	417–295	sand	salt			horizontal cylinder V type	82 85
417 295	417–295	sand	sand			inclined cylinder	9
417 295	417 295	sand	Na CO₃			ribbon	94
417 351	417 351	sand	sand			horizontal cylinder	62
351 295	351–295	sand	Na₂CO₃			V type double cone rotating cube and horizontal cylinder	97
351–295	208 175	soda ash	sand			horizontal cylinder	11
295–208	295–208	sand	Na₂CO₃			V-type double cone rotating cube and horizontal cylinder ribbon	93 94
208 175	208 175	sand	Na CO₃			V type double cone rotating cube and horizontal cylinder ribbon	92
400–100	850 600	sand	sand			vertical tank with paddle agitator	73
295 246	295–246	sand	sand			inclined cylinder	9
250 200	250 200	quartz sand	Na SO₄			horizontal cylinder with paddle agitator	54
208 175	208–175	sand	Na CO₃			ribbon	94
208–74	208–74	Ba SO₄	Ilmenite B			V type double cone Muller ribbon hammer mill and concrete mixer horizontal cylinder	36
208 74	74	sand	Ilmenite A			V type double cone Muller ribbon hammer mill and concrete mixer horizontal cylinder	36
295–41	295–41	silver sand	silver sand			horizontal cylinder double cone inclined cylinder	18,19
250–150	250–150	witherite	pyrites			vertical helical impeller	3
250–150	250–150	white sand	red sand			vertical helical impeller	3
175–147	175–147	sand	sand			horizontal cylinder	62
175–147	175–147	sand	Na CO₃			V type double cone rotating cube horizontal cylinder, ribbon	92
147–104	147–104	sand	Na CO₃			V type double cone rotating cube horizontal cylinder ribbon	92
147–74	174–74	pvc	Na CO₃			V type double cone rotating cube horizontal cylinder	89 90 91
147–74	147–74	Al₂O₃	Ilmenite ore			V-type double cone	36
147–74	147–74	sand	Na CO₃			vertical tank with paddle agitator	56
177–105	177–105	white pottery bisque	black pottery bisque			V type	74
180–110	180–110	sand	calcite			double cone air mixer vertical helical impeller Z blades	4
105 75	105–75	yellow glass beads	white glass beads			horizontal cylinder	42
104–74	104–74	sand	Na CO₃			V type, double cone, rotating cube horizontal cylinder	92
85–10	45–15	copper	nickel	spheroidal	spheroidal	Loedige continuous ribbon	63, 64
25	45–15	UO₂ (II)	nickel	rough	spheroidal	Loedige, continuous ribbon	63, 64
35	10	UO₂	ThO₂	rough	Sgnera plate	Loedige continuous ribbon	63 64

2.5. CONCLUSION AND DISCUSSION

The many methods for determining and expressing the degree
of mixing have been summarised. These vary from a photometric
technique through to a truly statistical representation of the
randomisation of mixture. It is felt that there is little need
for each paper produced on the subject to propose a new, and su-
pposedly better definition of the degree of mixing. Over 30 differ-
ent degrees of mixing have been proposed and as Lloyd and Yeung
have pointed out, 'this state of affairs makes research into the
subject extremely difficult, as comparison between the results of
different authors in impossible'.

The majority of experimental work has consisted of a study
of mixing two components within narrow size ranges. Free-flowing
materials have provided the bulk of the experimental investigations,
and the number of reported experiments on mixing of particles below
100 μ is few.

The mixing of components of differing sizes has been the
subject of very few investigations and has no satisfactory theory
on the segregation had been published. Oyama (1933) and Roseman (
1962) have both worked on the mixing of differing sizes and have
proposed similar theory of segregation, however, there is a lack
of evidence to justify the theory.

More attention has been given to the effects of material
properties and the performance of mixers in recent years. The most
comprehensive experimental study on these relationships has been
given by Yano et al (1956-1960). These have been discussed in de-
tial in the text. Their attempted to develop a quantitative picture
of mixing by grouping the experimental parameters, is inherently
dangerous if the mechanisms of mixing are not fully understood.
A more recent study by Ashton et al (1966) also compared the per-
formance of number of mixers but no general theory was attempted.

Most mechanisms of powders mixing are postulated as analo-
gous to other processes. Lacey (1954) first pointed out three po-
ssible mechanisms and Weidenbaum (1955) experimentally proved that
the mixing of same sized-particle materials was followed by a first
order rate equation or exponential decay law (diffusion type),

Forscher (1955) showed that a fully randomised mixture could be measured by a stochastic process, and Oyama (1956) stated that the mechanism of particle mixing could be described by a one-dimensional Brownian motion and introduced the Markov chain process into the subject theoretically. The first order rate equation on the mixing of same sized materials has been adopted by many other research workers, with insufficient explanation as to how and why the 'law' could be applied. In very recent years, number of research workers; Fisher (1963), Hogg, Cahn et al (1966), Müller and Rumpf (1967) have studied the diffusive mixing alone by analogous to Fick's second law of diffusion by using uniformly sized materials. Further, Cahn and Fuerstenau (1967) developed a random walk model for the simulation of the diffusion mixing process, as thought the mixing of powders could be studied by the use of Monte-Carlo method and computer. It is obvious when the process is assumed as a diffusion problem, the probability distribution of particle movements could be obtained by Monte-Carlo method. This method for solving diffusion problems was first studied by king (46') in 1951. It is felt that analogy alone can not be used to propose a greater understanding of the process of mixing, since the results may only be curve fitting. It is apparent that the mixing process may follow a relationship similar to Fick's law. The concept of 'diffusion' must be used with care, since the materials which have been studied have too large size of particles to have a purely diffusive nature under the conditions of mixing. Since a powder is a particulate system, the particulate nature of the mixing problem should be considered before any assumption is made. Then and only then will a better understanding nature of the mixing process be obtained.

Even though the mixing of powders has been studied over a period of many years, it remains a highly empirical unit operation, and it has really received very little systematic investigation, particular with reference to material properties. If a clearer, fundamental and universally aceptable theory, based on material properties, could be developed, the state of mystery and art of the operation of the mixing of powders would disappear, and the process designers could confidently predict all the process variables required for the good design.

CHAPTER THREE

THE PRELIMINARY EXPERIMENTS

3.1. INTRODUCTION

As pointed out in the last Chapter, a systematic study particularly with reference to the material properties is needed. The planning of a reasonable area of investigation is difficult without a brief understanding of the general picture of 'mixing' of components with differences in physical properties and of the motion of particles in a dynamic system. Therefore qualitative experiments were made in order to obtain some information quickly, relating to the mixing in a simple system.

The preliminary experiments fell into two parts. The first part in an ordinary sampling bottle with a horizontal rotating motion. The 'states of mixedness' were observed with three combinations of materials, these were, sand – sand, sand – gypsum and sand – glass powder. The results were obtained purely by observing the surface of the mass, during 'mixing' or after the machine was stopped. This study was initiated with the intention of establishing a general picture of the behaviour and characteristic of mixing with materials of different physical properties.

The second part was to study the radial motion of particles in a horizontal rotating drum using a photographic technique. The states of motion and particle packing of moving bed were illustrated at different speeds of rotation. This study might provide some information of mixing in the radial direction, where segregation occur and the best operating conditions in the horizontal rotating drum mixer.

These preliminary experiments resulted in the establishment of when 'mixing' and 'segregation' occurred in the mixing of <u>powders</u> in the horizontal rotating drum mixer and thus made it possible to develop the quantitative experimental programme and the theories described in later chapters.

3.2. THE PRELIMINARY EXPERIMENT I – MIXING IN A SAMPLING BOTTLE

3.2.1. EXPERIMENTAL

Each material was carefully sieved and the physical properties were determined, and shown in Sections 3.3. and 5.2.

The procedure of the experiments is described as follows :-
The bottle was an ordinary sampling bottle of 600 ml. capacity. A
standard method of loading was employed to prevent side effects
arising from different methods of loading. The sampling bottle
was held at 45° from the horizontal position and one of the mate-
rial was loaded. The material usually formed an angle of $20-25^o$
with the wall of the bottle when the bottle was returned to horizon-
tal position. The second material was then charged on the top of the
first. The finer material was usually placed below the coarse one.

The materials were loaded into the bottle to 40% of its
capacity and the binary system was kept at the 50/50 volume ratio.
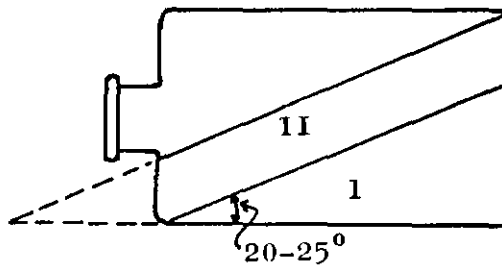
Fig. 3.1. The sampling bottle mixer

3.2.2. EXPERIMENTAL RESULTS

For the easy comparison of the observed results between the
sand – sand and sand – gypsum combinations at different size ratios,
the results are presented in a tabulated form

Size Ratio	Results	
	Sand – sand.	Gypsum – sand
1 (particle size, passing through 85 and retain- ed on 120 B.S.S., i.e -85/+120)	A uniform mixture was very easily obtained, even at very low speed of rotation. Increased the speed of ro- tation decreased the time of mixing, but the relation was not arithmetical. Pro- longing the time of mixing did not alter the results.	A good mixture was obtain- ed, no particular segrega- tion was observed. Increa- sing the time of mixing, cause little change in the mixture. At higher speeds of rotation (above 40rpm), the mixture reached the state of equilibrium very

Size Ratio	Results	
	Sand – Sand	Gypsum – Sand
(cont.)	Various speeds of rotation were also tested, and similar results were obtained.	quickly and no change was observed with prolonged mixing.
1.27 (particle size, -72/+85, -85/+120)	At low speed of rotation (7.2rpm), a band (axial segregation) of large particles formed in the middle of the bottle after a short time of mixing. Initially this band was quite wide but gradually narrowing down until an equilibrium width was established. Further increases in the mixing time did not change the width of the band. Similar results were obtained with increasing speeds of rotation up to 25rpm, at speeds above 25rpm, it was found that no band was formed.	At low speeds of rotation (7.2 to 16rpm), the mixing progress was very slow, after mixing for half an hour, the uniformity of the mixture was still very low, but no band was observed. At the higher speed of rotation of 35rpm, the mixture became quite uniform at the initial stage. Other speeds of rotation were tested, the behaviour of the mixture was similar to the lower speeds of rotation.
1.50 (particle size, -60/+72, -85/+120)	A band was easily formed at the low speeds of rotation (10-25rpm). The speed at which the band disappeared was above 35rpm. The mixing time required to obtain a reasonable mixture at this speed was a bit longer. A higher speed, above 55rpm, was required to obtain a good mixture.	No band was formed at low speed of rotation although radial segregation existed. After mixing for 45 minutes the mixture became more uniform, but further increases in the time of caused no change in the uniformity of the mixture. The uniformity of mixture seemed to have reached a

Size Ratio	Results	
	Sand – Sand	Gypsum – Sand
(cont.)	Prolonged mixing did not change the mixture obtained.	steady state. At higher speeds of rotation, the rate of mixing was much faster, and the equilibrium state was quickly reached. The other phenomena were the same as at lower speed of rotation.
1.80 (particle size, -52/+60, -85/+120)	Similar results were observed as in the case of the size ratio 1.50. The band was formed quickly. It narrowed with increasing time of mixing until a fixed widht of band was reached. By increasing the speed to about 40rpm, the band gradually disappeared, and the mixture appeared to be more uniform.	At low speed of rotation axial segregation (but not with a clearly defined band) was formed in some places. This segregation appeared mainly as a gathering of the coaser particle (gypsum) covered the liner particles (sand). At higher speeds of rotation (above 36rpm), the mixture appeared to be much uniform. No conspicuous band appeared at various speeds of rotation, although radial segregation still appeared.
2.35 (particle size, -36/+52, -85/+120)	No mixing effect was observed at low speeds of rotation, prolonged mixing caused a band to form. With a speed of 14rpm, the band appeared rapidly. Further increases in speed to about 50rpm, caused the	Observation of mixing process was extremely interesting, after mixing for 12 minutes at 7.2rpm, the materials separated into two complete parts, occupying one side each end to end. It was found that

Size Ratio	Results	
	Sand – Sand	Gypsum – Sand
(cont.)	band to slowly disappear. Serious radial segregation of both materials existed. A speed of 100rpm was required to obtain a good mixture. The mixture did not become more uniform with increases in the mixing time.	while a very few of gypsum particles were found in the sand region none of sand particle could be found in the gypsum region. This phenomena continued to exist until the speed of rotation increased to 24rpm. The completely separated regions then became the normal band form (with large particles in the middle). When the speed of rotation was increased above 45rpm, the band gradually disappeared but radial segregation still existed.
3.00 (particle size, –30/+36, –85/+120)	No mixing effect was observable at low speed of rotation. With a speed of 8rpm, a band soon appeared. It separated the materials into two complete parts which could not be broken down by prolonged mixing time. When the speed was increased to 55rpm, the band disappeared after for 2 minutes, after which time the mixture showed no improvement. Radial segregation still existed.	At low speeds of rotation, the band formed quickly after a very short time of mixing. At a speed of 60 rpm, the mixture became a little better, but the band still existed. This state was observed at speeds up to the critical speed.

Size Ratio	Results	
	Sand — Sand	Gypsum — Sand
(cont.)	At 75rpm, the uniformity of the mixture seemed to be better, but no improvement was observable with increased time of mixing.	
4.42 (particle size, -18/+30, -85/+120)	The band formed at a speed of 16rpm. At this speed, the band appeared extremely quickly. After mixing for 30 seconds, the band was clearly formed. When the speed was increased to 75rpm, the band disappeared, but it reappeared after a few minutes of mixing. When the speed of rotation was increased to 130rpm, centrifugal force predominated. The particles were distributed uniformly throughout the bottle, but serious radial segregation existed, especially at the ends. At this size ratio, mixing in the simple bottle was extremely difficult to obtain and only a very low uniformity of the mixture could be obtained.	It was found that mixing could not be obtained even at high speeds of rotation. Axial segregation—a band appeared at low speeds of rotation, and radial segregation appeared at the high speeds of rotation. At this size ratio, a uniform mixture could not be obtained.

Additional experiments were made with a sand – glass powder combination. The size ranges were –120/+200 for sand and –200/ for glass powder. It was found that no band appeared at any speed of rotation, though serious radial segregation occurred. In particular the glass powder formed layers. These layers could not be dispersed even at a fairly high speed of rotation. The shape of the glass powder was spherical, giving it a highly free flowing property. At a higher speed of rotation, the mixture seemed to be much uniform, eventhough, the materials were very hard to mix.

3.2.3. CONCLUSIONS

As already stated, the aim of these experiments was to obtain some qualitative information concerning the behaviour of the powder mixing under various conditions. Of particular interest were the size ratios and density difference of the materials being mixed. These experiments can be summarised into two parts ; the importance of the materials being mixed and the mixing process. As far as the materials are concerned, if they have the same physical properties, size and size distribution, the experiments indicated that they are easily mixed and a uniform mixture could be rapidly obtained. When the materials are different in size, and a mean size ratio is greater 1.3, segregation in form of a axial band and radial core can easily exist in the mixture. Initially the band appeared quite wide but gradually narrowed until an equilibrium width was established. In general this phenomena could be plotted as
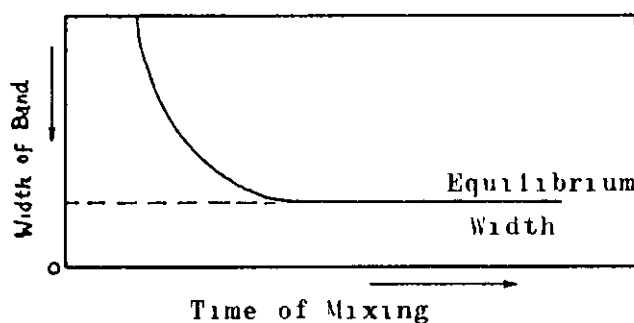
Fig. 3.2

Extreme difficulty is found in obtaining a good mixture when the mean size ratio is over 3. A reasonably uniform mixture can not then be obtained in the simple bottle mixer. When the materials differ

in density alone, good mixing can be obtained when the size ratio is 1.
When the components differ in size and density, both radial and axial
segregation can occur. The sand and glass powder combination showed
very similar results to those obtained with sand and gypsum combination.

In the mixing process of prime importance is the rate at which
a desired uniformity is reached. The speed of rotation of the machine
influences not only the time required for the mixing, but also affects
the final uniformity of the mixture. At low speeds of rotation, the
experiments indicated that low uniformity with serious segregation can
be obtained when mixing with particles of different size, moreover
even mixing with particle of same size a longer time of mixing will be
required. In general, a speed of under 20rpm is unsuitable for good
mixing. The speed of rotation should therefore be above 20rpm but be-
low the critical speed for reasonable results to be obtained.

Higher speeds of rotation give more movement to the particles.
As the particles have more chance to change their relative positions
and so distribute more uniformly throughout the whole particle assem-
ply, the possibilities of segregation are reduced.

Secondly, these experiments indicated that there is a tendency
for an equilibrium to be established after a certain time has elapsed,
regardless of the ease or difficulty of the mixing. The uniformity
becomes independent of time, in fact mixing beyond this limit only
causes segregation to become serious again. The rate of mixing might
be indicated generally by the following diagram,

Fig. 3.3.

Thirdly, the end wall effects the axial mixing. A shearing
force is created by the friction of the end wall and the rotated
particles, and gives the particle a compared velocity in the axial
direction.

Although the simple bottle is not a perfact drum, the most si-
gnificant phenomena in the powder mixing due to changes in the physical
properties of the materials and the patterns of segregation – axial

in density alone, good mixing can be obtained when the size ratio is 1.
When the components differ in size and density, both radial and axial
segregation can occur. The sand and glass powder combination showed
very similar results to those obtained with sand and gypsum combination.

In the mixing process of prime importance is the rate at which
a desired uniformity is reached. The speed of rotation of the machine
influences not only the time required for the mixing, but also affects
the final uniformity of the mixture. At low speeds of rotation, the
experiments indicated that low uniformity with serious segregation can
be obtained when mixing with particles of different size, moreover
even mixing with particle of same size a longer time of mixing will be
required. In general, a speed of under 20rpm is unsuitable for good
mixing. The speed of rotation should therefore be above 20rpm but be-
low the critical speed for reasonable results to be obtained.

Higher speeds of rotation give more movement to the particles.
As the particles have more chance to change their relative positions
and so distribute more uniformly throughout the whole particle assem-
ply, the possibilities of segregation are reduced.

Secondly, these experiments indicated that there is a tendency
for an equilibrium to be established after a certain time has elapsed,
regardless of the ease or difficulty of the mixing. The uniformity
becomes independent of time, in fact mixing beyond this limit only
causes segregation to become serious again. The rate of mixing might
be indicated generally by the following diagram,

Fig. 3.3.

Thirdly, the end wall effects the axial mixing. A shearing
force is created by the friction of the end wall and the rotated
particles, and gives the particle a compared velocity in the axial
direction.

Although the simple bottle is not a perfect drum, the most si-
gnificant phenomena in the powder mixing due to changes in the physical
properties of the materials and the patterns of segregation - axial

or radial may be demonstrated simply from these simple tests an extensive quantitative study into the importance of material properties of the powder mixing was launched and the main aim of these preliminary experiments obtained.

3.3. THE PERLIMINARY EXPERIMENT II – THE STUDY OF RADIAL MOTION OF PARTICLES IN A HORIZONTAL ROTATING PERSPEX DRUM

3.3.1. EXPERIMENTAL

Gypsum and sand were used for this study, the physical properties are listed in Table3.1.(PVC granules, 0.125" diam. and 0.125" long were used for taking the still photographs).

The experimental apparatus used was a perspex drum with 5" outside diameter and $4\frac{3}{4}$" inside diameter, $6\frac{3}{4}$" in length, and directly driven by a variable speed electric motor as shown in Photo (3.1)

TABLE 3.1.

PHYSICAL PROPERTIES OF GYPSUM AND SAND

	Gypsum	Sand (Moneystone)
size (BSS)	-8/+10	-18/+30
density (gm/ml)	2.14	2.585
apparent density stationary (gm/ml)	1.27	1.34
shape	granular	granular
angle of repose	28.5°	29.5°

The motions of particles at various speed of rotation were recorded by a high speed cine camara (32 fps), through the transparent end of the drum. Still photograph-s were also taken at fixed speeds of rotation with various exposure times. Representative prints from the cine film obtained under different experimental conditions are shown in Photos (3.2) to (3.5).

3.3.2. ANALYSIS OF THE CINE FILMS

When the drum is moving at a certain high speed, a kidney-shaped moving mass is created in the drum. Two types of motion were observed in the drum, these may be named 'cascade' and 'cataract' (61,68). At a relative low speed of rotation, the particles travel upwards on circular arcs, concentric with the wall of the drum, until a point of instability is reached, after which they roll down a surface which is inclined at an angle to the horizontal, this type of motion as shown in Photo (3.2.), the still photograph is shown in Photo (3.6). At a higher speed of rotation, the particles no longer roll down the surface of the charged mass, instead, at a certain point they are projected into space, and fall essentially as projectiles. When the particles meet the charged mass, they resume the upward path, this type of motion as shown in Photos(3.4,3.5), the still photograph is shown in Photo (3.7). In any of those motions a boundary such as ABC in Photo (3.6) or Photo (3.7), can easily be seen, this line separates the charged mass in two states of motion, i.e. upward movement and downward movement.

In general, the motions of particles in a rotating drum can be classified into three distinct regions, upward movement (region I), downward movement (region II) and the boundary (region III), this is sketched in Fig.(3.4).

Oyama (61) has said the boundary can be described by a logarithmic-spiral curve, the general equation can be represented as

$$\rho = ae^{-bk\alpha} \qquad (3.1)$$

where ρ is the distance from the fixed point to the particle, α is angular displacement from the horizontal direction, and a, b, k are constants
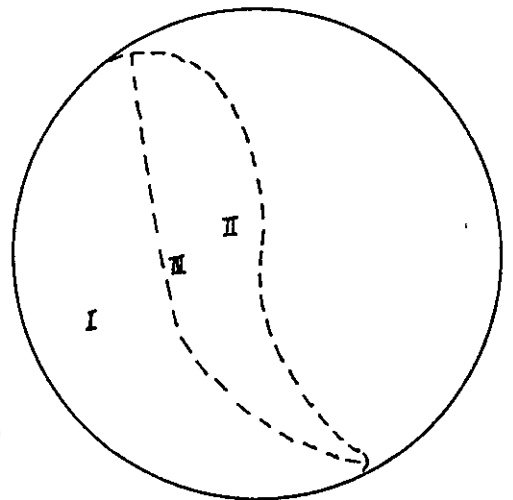
Fig.3.4

Along this boundary, sketched as region III, there is the minimum velocity gradient, due to the particles changing their direction of motion, therefore shearing takes
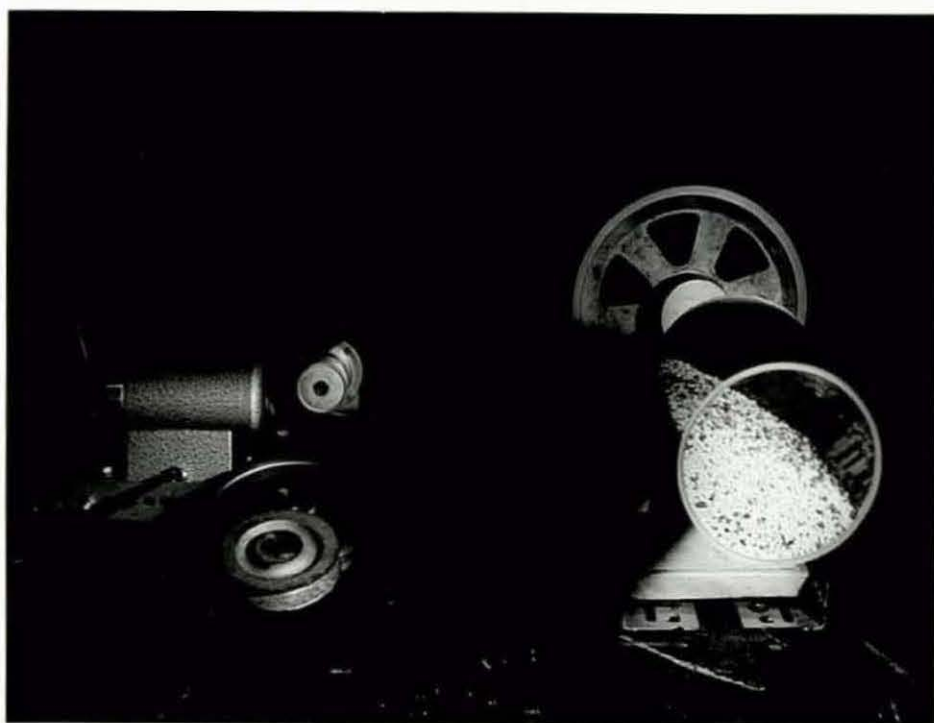
PHOTO 3.1. EXPERIMENTAL APPARATUS
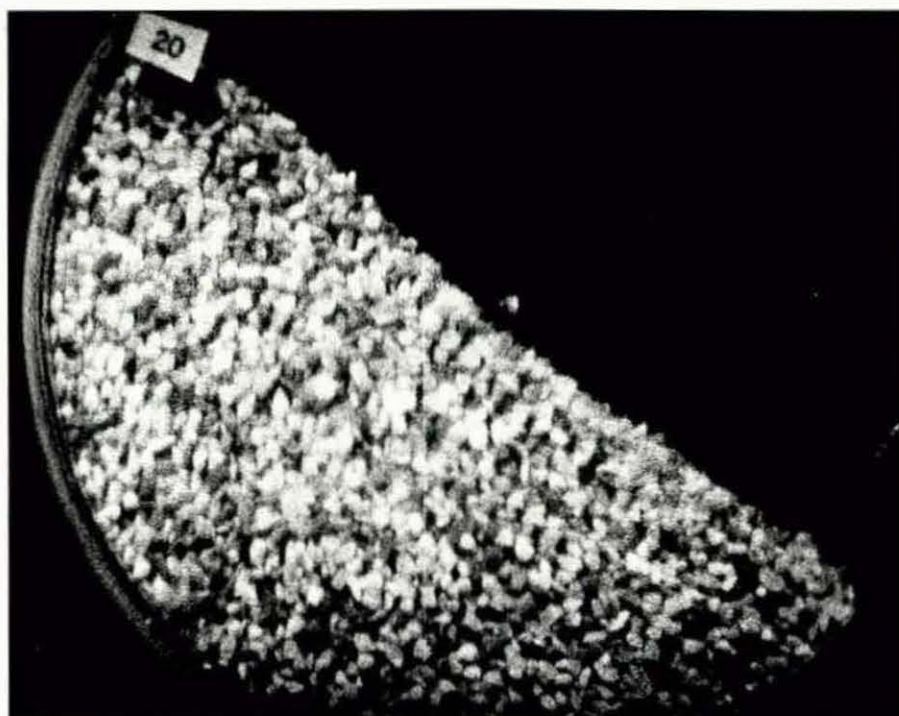
PHOTO 3.2. THE MOTION OF PARTICLES IN THE
ROTATING DRUM AT 20 rpm
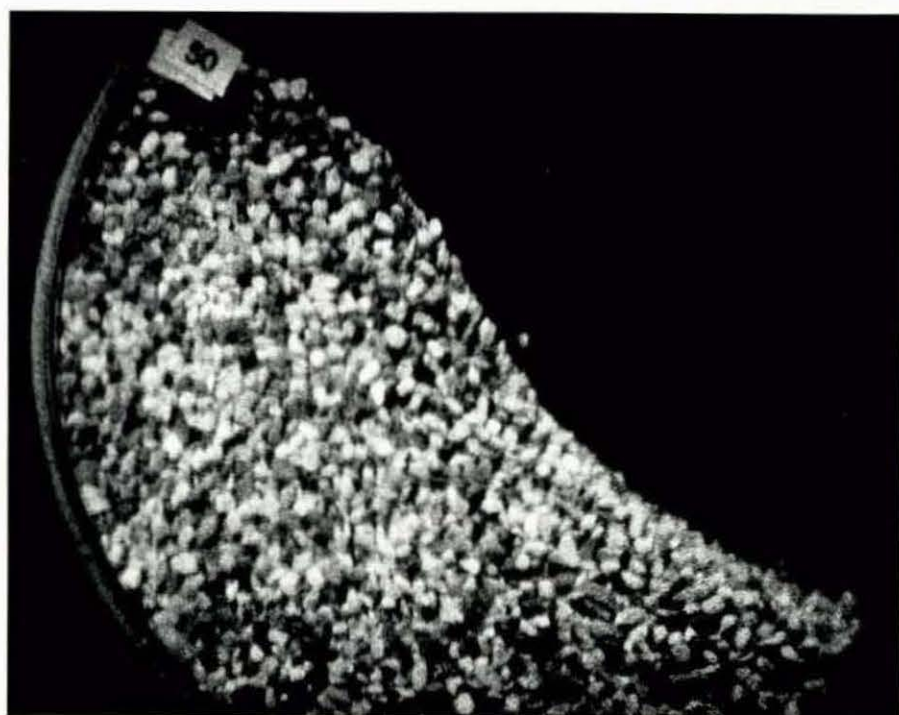
PHOTO 3.3. THE MOTION OF PARTICLES IN THE
ROTATING DRUM AT 50 rpm
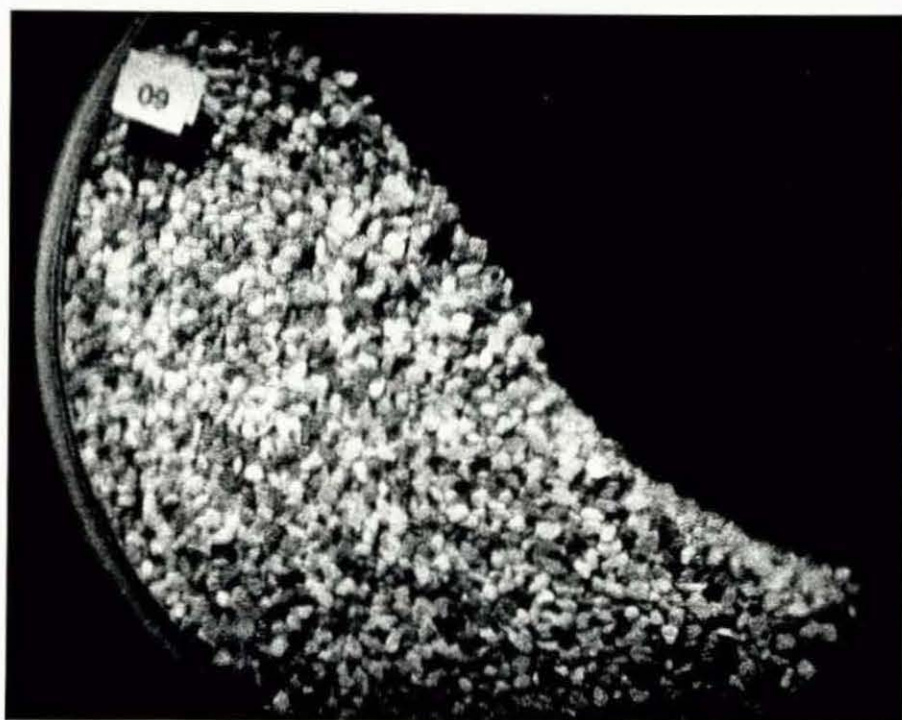
PHOTO 3.4. THE MOTION OF PARTICLES IN THE
ROTATING DRUM AT 60 rpm
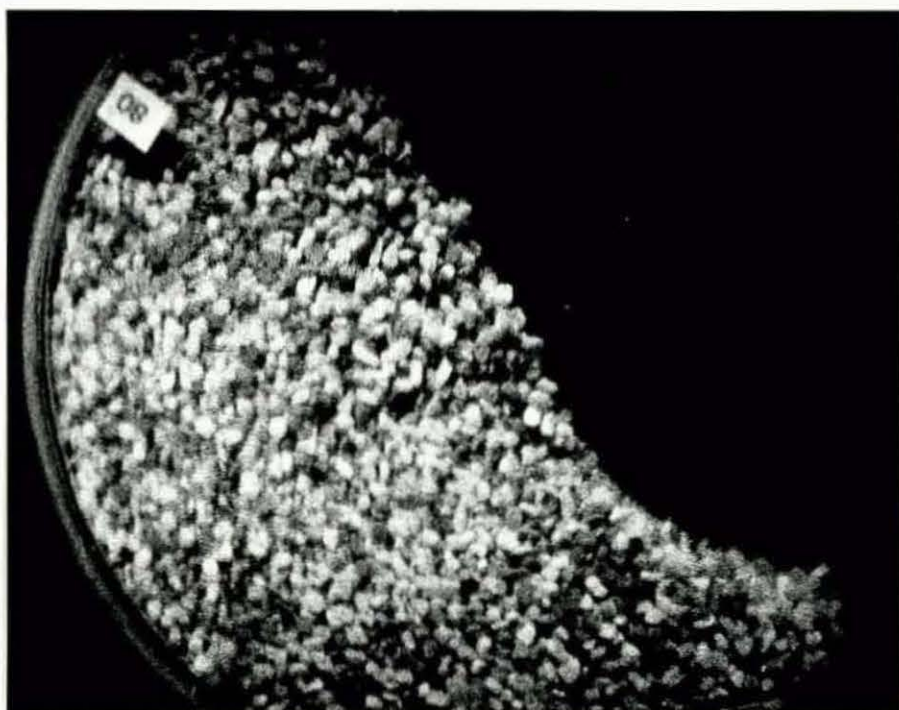
PHOTO 3.5. THE MOTION OF PARTICLES IN THE
ROTATING DRUM AT 80 rpm

PHOTO 3.6. THE MOTION OF PARTICLES IN THE ROTATING
DRUM AT 20 rpm (STILL PHOTOGRAPH)
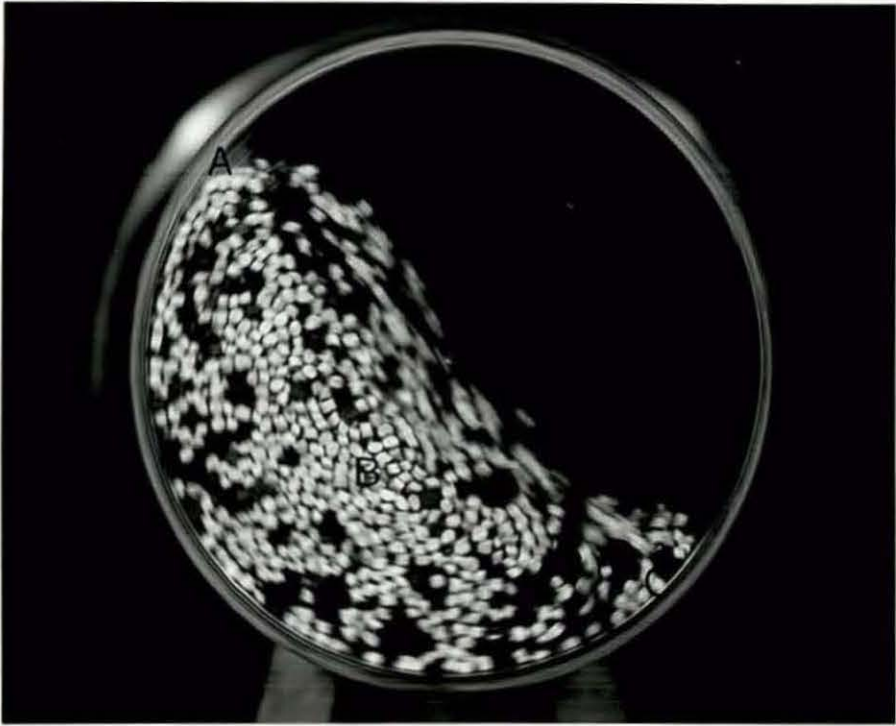
PHOTO 3.7. THE MOTION OF PARTICLES IN THE ROTATING
DRUM AT 60rpm (STILL PHOTOGRAPH)

PHOTO 3.8. THE MOTION OF PARTICLES IN THE ROTATING
DRUM AT 20rpm (STILL PHOTOGRAPH 1/10 SEC. EXPOSURE TIME)

place between particles. In the centre of the mass, as B in Photo
(3.6) or Photo (3.7), at this fixed point, the particles are sta-
tionary (or at best move only slightly).

A better description of the motion, however, would be par-
ticulate, contrasting with the bulk behaviour of material in region
I. On reaching the bottom of the slope, the particles form a loose
packing, which is carried unchanged to the top of the slope. There
the packing dilates before breaking up and individual particle be-
gin to fall. In forming the loose packing, individual particles may
take up different radial positions on each travese, and so give the
rapid radial mixing which has been noted in these mixers. The sta-
bility of the loose packing could be seen from individual particle
velocities. The angular velocity of particles at different radial
positions at a given rotational speed of the drum is shown in Fig.
(3.5A). All curves show a tendency for the angular velocity of
particles reach a maximum at 0.7R The results were taken along
the radius in the centre of the powder mass.
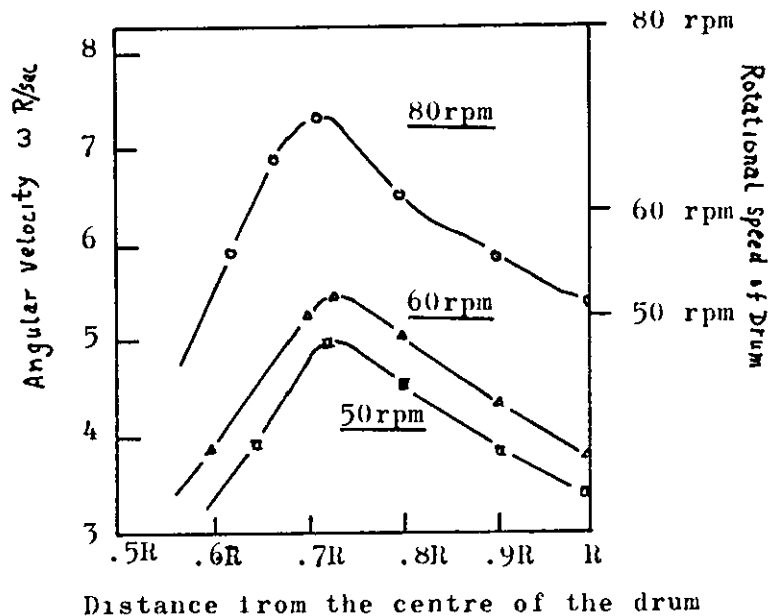
Fig.3.5A   Radial Angular velocity distribution
of gypsum particles

The linear velocity of the particles, however, is constant
at radial distance from the outside to 0.7R even at the highest
angular velocity. This is shown in Fig.(3.5B). This phenomena
could only occur if the packing is stable. The extrapolation of
line BC in Fig.(3.5B) to v = 0, gives the position of the particles
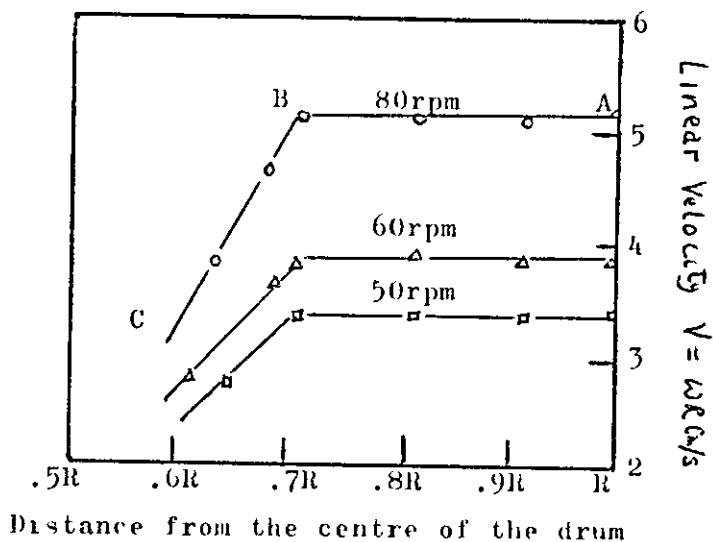
Fig.3.5B   Radial linear velocity distribution
of Gypsum particles

which change their direction of motion.

The trajectory region (region Il) of the particles can
be observed from the photographs. In this region, particles will
have less freedom to exchange their axial position as the depth
increases. The region has been studied by Kaye and Sparrow, Cahn
et al, and was thought to constitute surface diffusive mixing.

3.3.3.   CONCLUSION AND DISCUSSION

The boundary separating the mass of particles into two
states of motion can be observed clearly and might be described
by a log-spiral curve. If the local velocity of particles can

be obtained the highest point of the path at each layer could be computed from Eq.(3.17). The rapid radial motion may be further demonstrated in Photo (3.8), which was taken at 20rpm with 1/10 second of exposure time.

It has been noticed that the velocity of individual particles near the wall is lowder than the actual speed of rotation of the drum. This is caused by the material slipping. A loose packing is formed at high angular rotations whilst at low velocities the packing may be relatively tight. From the loose packing, the interchange of particles can easily occur, on the other hand, where the packing is tight mixing is less probable. Thus one would expect a higher rate of mixing at high particle velocity in region II.

By considering this two-dimensional (plane) and one-component system, a new concept of the optimum speed of rotation and fullness of charge for the drum mixer may be obtained.

The optimum speed of rotation is the speed at which resultant forces acting on a particle on trajectory path are at a minimum. The particles could very gently fall down the charged mass in a 'partially fluidised state' with a maximum time for the particles travel from the top to the bottom of the inclined surface. A particle could then receive a maximum chance of displacement in the axial direction.since it would receive an increase in the probability of collisions with other particles and the axial rate of mixing would be increased.

The regions could be measured from the photographs. The proportion of region I to region II is a function of speed of rotation. Since region I is a form of stable packing, the axial interchange of particles is most unlikely to be taking place so the mixing must take place mainly in region II.

By observing the motion at 20rpm (Photo 3.6), it can be seen that the depth of region II is only 2-3 particle diameters thick and the particle are not projected, but are simply rolling down the inclined surface. Under these conditions it might be expected that it would be difficult to change the axial position of the particles.

At 80rpm (Photo 3.5), the proportion of I and II is almost
equal, however the paricles are mainly under centrifugal motion, and
the time during which the particles are following trajectory will
be too short to give a maximum probability of changing positions,
so the axial mixing rate would be low.

The optimum speed of rotation must therefore be obtained
between these two speeds - about 50rpm say. If a method could be deve-
loped which measures the actual velocity of the trajectory, then
the resultant forces of a particle might be computed and a quanti-
tative picture for different conditions be obtained, from which
a optimum value could be selected.

Setting **aside** the question of economics, the optimum fullness
of charge will depend upon the states of motion. This can be ex-
plained by discussing flow regions for these different charges.-
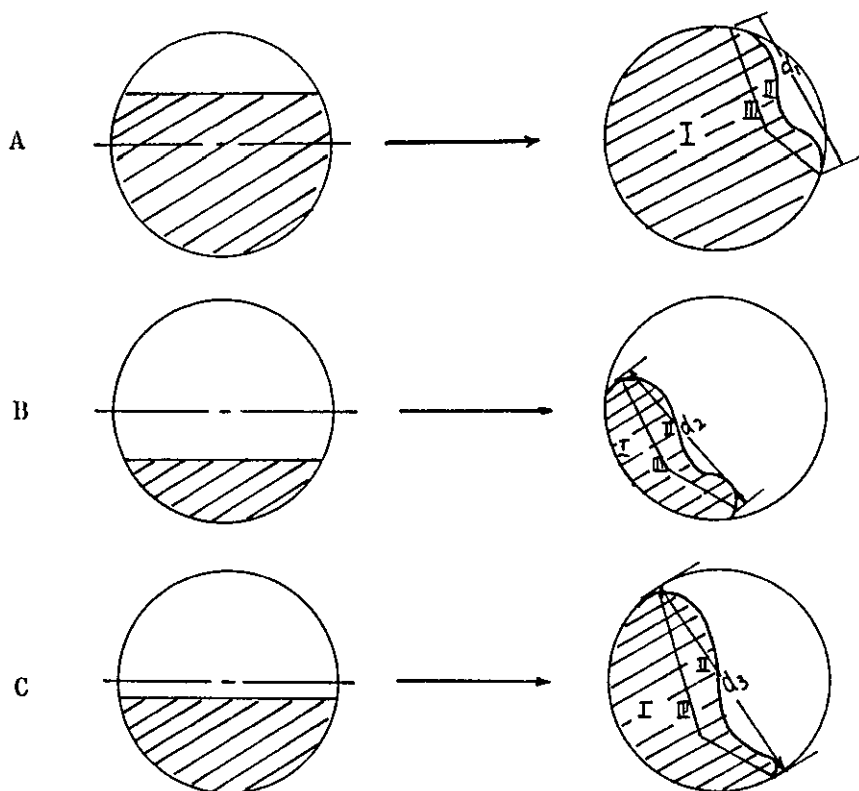
Fig. 3.8

When the material is loaded more than half full as sketched
in Fig.(3.8A), the trajectory path is short and the proportion of
region I to region II is large, therefore, particles spend the majority
of the mixing in the stable packing and have limited axial dis-

placements. In this case the mixing efficiency is unlikely to be a maximum.

In the second case (Fig.3.8B), the material is loaded much less than half full. Whilst the proportion of region I to region II is now reasonable, the distance down which the particles roll is clearly not a maximum.

This will occur in the third case (Fig.3.8C) where the charge is such that the trajectory path is equal to the diameter of the drum. This occurs when the loading is such that the expanded mass of the particles occupies half the drum, particles then have more chance to displace their axial position.

From these considerations, it is clear that to obtain the fastest mixing rate, both the speed of rotation and the fullness of charge must be selected to give the longest time for a particle to travel down the slope within the mixer. This prediction seems reasonable when compared with the experimental results obtained by previous investigators (18) (57)(90).

CHAPTER   FOUR

THE DEVELOPMENT OF A SUITABLE THEORY FOR

MIXING OF FREE FLOWING POWDERS

4.1. INTRODUCTION

There are many possible approaches to the investigation of the theory of powder mixing. It seems reasonable to suppose that in the mixing of particulate matter the first step must be the consideration of the particle properties. The mixing machine is needed to perform the mixing but it is the behaviour of the particles within the machine which can or cannot lead a good mixing. If this was understood, a suitable machine could be designed without much difficulty.

However, since there are too many kinds of particulate materials to be investigated individually, one must simplify the problem by classifying the powders. A logical division is into two main groups, the first is concerned with free flowing powders and the second with non-free flowing powders (12), since then in each group a different mechanism to cause mixing is required. In the present thesis, the object of the investigation has been restricted the consideration of free flowing powders.

The mechanism of mixing process for free flowing powders can be described by the exchange of individual particles within the whole mixture. For instance, one wants to mix 100 black particles with 100 white particles, as sketched in Fig.(4.1)
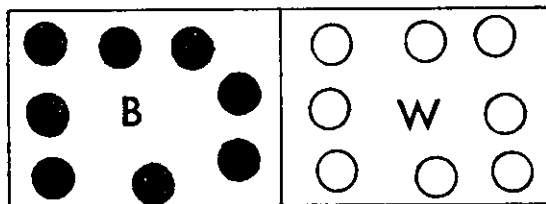
Fig.4.1.

Suppose one picks a particle from B and replaces it with a particle from W, and the process is repeated until it is possible to pick a particle from either side with the same chance of picking a black or white particle, then this is when the mixing process is complete. The problem is reduced to one of a simple probability. When 50 black particles have been exchanged with 50 white particles, the chance of finding a black particle in any position is equal to that of picking a white particle.

This is the approach of the present investigation. The application of probability theory to particle movement in the mixing

system with particular reference to the behaviour of free flowing
powders in a horizontal rotating drum mixer with a constantly
applied mechanical force, without which application of energy
no powder mixing could occur.

4.2. THE THEORY

Since the particles with a free flowing powder are moving
individually in the horizontal rotating drum mixer (referring to
Chapter 3.3.). The mixing process can be described by a stochastic
movement. However 'stochastic analogy' has mainly been concerned
with microscopic (biological) system, or a system which does not
suffer instability over changes of the parameters (i.e. birth-death,
waiting time, telephone call, etc.). The powder mixing system deals
with finite sized particles in a dynamic situation. It is therefore
a further study of the application of the stochastic process to the
particulate mixing system and one must consider the particulate
behaviour involved. To do this two postulates must be proposed
which will take into account the finite values of particle size and
momentum.

Postulate 1

All particles of the same size and shape have the same
probability of occupying each unit volume within the mixture (25).
(This postulate covers the packing of the particles).

Postulate 2

All particles of the same mass have the same probability
of undergoing unit movement after collision with other particles
(i.e. a constant mechanised force is applied). (This postulate is
concerned the momentum of the particles).

In general, the mixing of free flowing powders can be
divided into three distinct classes.

(1) The material obeys both postulates. This can only occur
when the process is mixing physically identical particles, differing
only by colour. (An example of this is the mixing of masterbatch
coloured granules into the full batch).

(2) The materials which obey the first postulate and not the
second, e.g. the particles have the same size but differ in density.

(3) The materials which do not satisfy either postulate, e.g. where both a size difference and a density difference occurs.

These three classes will be discussed separately in the following:

(1) Mixing of components with physically identical particles.

'A randomised mixture is a mixture in which the probability of finding a particle of one component at any position within the mixture is a constant which equals to the proportion of that component in the whole system'(84). It is clear therefore that only the present class can fulfill this requirement.

Consider initially the mixing of two free flowing powders in a horizontal rotating drum mixer, at a constant speed of revolution. As the mixer rotates, a particle will suffer a displacement ΔS which is some function of ΔX, ΔY, and ΔZ in a small interval of time Δt. Many workers have shown that mixing in the radial direction is very fast when compared to the slow drift of the particles along the axis. Thus it is the axial motion of particles which controls the overall rate of mixing. Thus it can be assumed that the radial mixing is infinitely fast and attention need only be paid to the axial movement. The problem is resolved therefore to the one dimensional case in which the displacement $\Delta S = \Delta X$.

Since the materials under present consideration satisfy each of the postulates, each particle in the system will have an equal probability of undergoing a displacement. The motion of a single particle can thus be considered as a series of equal length steps in either direction along the axis. Each step will be in a positive or negative direction with equal probability of $\frac{1}{2}$. After N such steps the particle will be at any of the position within

$$-N, \ -N+1, \ -N+2, \ \ldots\ldots\ -2, \ -1, \ 0, \ 1, \ 2, \ \ldots\ldots N-2, \ N-1, \ N$$

The probability of particle arrived at the position m after N displacements is $P(m, N)$. It is clear that $P(m, N)$ will be equal to $(1/2)^N \left[= (p)^{\frac{1}{2}(N+m)} (q)^{\frac{1}{2}(N-m)}, \text{since } p = q = 1/2 \right]$ times the number of distinct sequences of steps which would lead to the position m after N steps. In general, there will be $(N + m)/2$ steps in the positive direction and $(N - m)/2$ steps in the negative direction.

So that the total number of such distinct sequences will be

$$\frac{N!}{\left[\frac{1}{2}(N+m)\right]!\left[\frac{1}{2}(N-m)\right]!}$$

The probability of arriving at position m after N steps is thus

$$P(m,\ N) = \frac{N!}{\left[\frac{1}{2}(N+m)\right]!\left[\frac{1}{2}(N-m)\right]!}\ (\frac{1}{2})^N \tag{4.1}$$

In terms of the binomial coefficients $C_r^{n'}$s, so Equation 4.1 can be rewritten as the form of Bernoullian trial,

$$P(m,\ N) = C_{1/2(N+m)}^{N}\ (\frac{1}{2})^N \tag{4.1'}$$

In other words, the probability distribution is Bernoullian.

If N is large, Stirling's formula (29) can be used

$$n! = (2\pi)^{1/2}\ n^{n+1/2}\exp(-n) \tag{4.2}$$

and

$$\ln n! = (n+\frac{1}{2})\ln(n) - n + \frac{1}{2}\ln(2\pi) \tag{4.2'}$$

Let $\quad N'! = \frac{1}{2}(N+m)!\quad$ and $\quad N''! = \frac{1}{2}(N-m)!$

Therefore Eq.(4.1) becomes,

$$P(m,\ N) = \frac{N!}{N'!\ N''!}\ (\frac{1}{2})^N \tag{4.3}$$

Take logarithm both sides of Eq.(4.3), then

$$\begin{aligned}
\ln P(m,N) &= \ln N! + N\ln\frac{1}{2} - \ln N'! - \ln N''! \\
&= (N+\frac{1}{2})\ln N - N + \frac{1}{2}\ln(2\pi) + N\ln\frac{1}{2} \\
&\quad - (N+\frac{1}{2})\ln N' + N' - \frac{1}{2}\ln(2\pi) \\
&\quad - (N''+\frac{1}{2})\ln N'' + N'' - \frac{1}{2}\ln(2\pi) \\
&= (N+\frac{1}{2})\ln N - N - \frac{1}{2}\ln(2\pi) + N\ln\frac{1}{2} \\
&\quad - \frac{1}{2}(N+m+1)\ln(\frac{N+m}{2}) + \frac{1}{2}N + \frac{1}{2}m \\
&\quad - (\frac{N+m+1}{2})\ln(\frac{N-m}{2}) + \frac{1}{2}N - \frac{1}{2}m \\
&= (N+\frac{1}{2})\ln N - (\frac{N+m+1}{2})\ln\frac{N}{2}(1+\frac{m}{N}) \\
&\quad - (\frac{N-m+1}{2})\ln\frac{N}{2}(1-\frac{m}{N}) - \frac{1}{2}\ln(2\pi) - N\ln 2.
\end{aligned}$$

Take $\qquad \ln(1\pm\frac{m}{N}) = \pm\frac{m}{N} - \frac{m^2}{2N^2} \pm \frac{m^3}{3N^3} + \cdots$

for large N,

$$\ln \left(1 \pm \frac{m}{N}\right) = \pm \frac{m}{N} - \frac{m^2}{2N^2}$$

So that,

$$\ln P(m, N) = \left(N + \frac{1}{2}\right) \ln N - \frac{1}{2} \ln(2\pi) - N \ln 2$$
$$- \frac{1}{2}(N + m + 1)\left(\ln N - \ln 2 + \frac{m}{N} - \frac{m^2}{2N^2}\right)$$
$$- \frac{1}{2}(N - m + 1)\left(\ln N - \ln 2 - m/N - m^2/2N^2\right)$$
$$= N\ln N + \frac{1}{2}\ln N - \frac{1}{2}\ln(2\pi) - \frac{1}{2}(N+m+1)\ln N$$
$$+ \frac{1}{2}(N+m+1)\ln 2 - \frac{1}{2}(N+m+1)\frac{m}{N} + \frac{1}{2}(N+m+1)\frac{m^2}{2N^2}$$
$$- \frac{1}{2}(N-m+1)\ln N + \frac{1}{2}(N-m+1)\ln 2 + \frac{1}{2}(N-m+1)(m/N)$$
$$+ \frac{1}{2}(N-m+1)(m^2/2N^2)$$

$$\ln P(m, N) = -\frac{1}{2}\ln N - \frac{1}{2}\ln(2\pi) + \ln 2 - \frac{m^2}{2N} + \frac{m^2}{2N^2}$$

or

$$P(m, N) = \left(\frac{2}{N\pi}\right)^{1/2} \exp\left(-\frac{m^2}{2N}\left(1 - \frac{1}{N}\right)\right)$$

Since $N \gg 1$, so $1 - \frac{1}{N} \rightarrow 1$, then,

$$P(m, N) = \left(\frac{2}{N\pi}\right)^{1/2} \exp(-m^2/2N) \tag{4.4}$$

Now, since N is very large, it is convenient to introduce the net displacement x instead of m from the starting position as the variable,

$$x = m\delta \tag{4.5}$$

Where δ is the length of each step. Thus Eq.(4.4) becomes,

$$P(x/\delta, N) = \left(\frac{2}{N\pi}\right)^{1/2} \exp(-x^2/2N\delta^2) \tag{4.6}$$

Let, $N = nt$ and $D = \frac{1}{2}n\delta^2$ \hfill (4.7)

Where n is the number of displacement per unit time, t is the time which will be taken at each interval and D is a certain physical quantity. Then Eq.(4.6) becomes,

$$P(x, t) = (1/\sqrt{4\pi Dt}) \exp(-x^2/4Dt) \tag{4.8}$$

The equation has been obtained by considering the displacement of a single particle. It represents the probability of finding the particles at a position x after time t. Clearly if a large number of particles are considered the probability function will represent the proportion of one species throughout the volume

and to understand the dispersion of particles throughout the mixer volume, one must now consider how this function varies with time.

Suppose that during an interval of time Δt which is long enough for a particle to produce a large number of displacements but still short enough for the net mean square increment $\overline{\Delta x}^2$ in x to be small, under those considerations the probability that a particle undergoing of a net displacement Δx in the time Δt is given by Eq.(4.8), hence,

$$Q(\Delta x, \Delta t) = \frac{1}{\sqrt{4\pi D \Delta t}} \exp(-\frac{\overline{\Delta x}^2}{4D\pi t}) \tag{4.9}$$

This function is independent of the starting position of the particles. The derivation of the probability distribution of $P(x, t + \Delta t)$ at $(t, + \Delta t)$ from the distribution $P(x, t)$ at the earlier time t, from Eq.(4.9), the probability distribution of $P(x, t + \Delta t)$ could be expressed by a integral equation (5)(26)(45), thus,

$$P(x, t + \Delta t) = \int_{-\infty}^{\infty} P(x - \Delta x, t) \, Q(\Delta x, \Delta t) \, d(\Delta x) \tag{4.10}$$

Since $\overline{\Delta x}^2$ is assumed to be small enough, the expansion of each side of the equation can be taken under the integral sign by Taylor's theorem :

$$\int_{-\infty}^{\infty} P(x - \Delta x, t) \, Q(\Delta x, \Delta t) \, d(\Delta x)$$

$$= \int_{-\infty}^{\infty} \frac{1}{\sqrt{4\pi D\Delta t}} \exp(-\frac{\overline{\Delta x}^2}{4D\Delta t}) \left[P(x - \Delta x, t) \right] d(\Delta x)$$

$$= \frac{1}{\sqrt{4\pi D\Delta t}} \int_{-\infty}^{\infty} \exp(-\frac{\overline{\Delta x}^2}{4D\Delta t}) \left[P(x, t) + (-\Delta x)\frac{\partial P}{\partial x} \right.$$
$$\left. \frac{1}{2!}(-\Delta x)^2 \frac{\partial^2 P}{\partial x^2} + \frac{1}{3!}(-\Delta x)^3\frac{\partial^3 P}{\partial x^3} + .. \right] d(\Delta x)$$

$$= \frac{1}{\sqrt{4\pi D\Delta t}} \int_{-\infty}^{\infty} \left[\exp(-\frac{\overline{\Delta x}^2}{4D\Delta t}) \, P(x,t) - \exp(-\frac{\overline{\Delta x}^2}{4D\Delta t}) \cdot x \cdot \frac{\partial P}{\partial x} \right.$$
$$+ (\frac{\Delta x}{2})^2 \exp(-\frac{\overline{\Delta x}^2}{4D\Delta t})\frac{\partial^2 P}{\partial x^2}$$
$$\left. - \frac{\Delta x^3}{6} \exp(-\frac{\overline{\Delta x}^2}{4D\Delta t}) \frac{\partial^3 P}{\partial x^3} + ... \right] d(\Delta x)$$

$$= \frac{1}{\sqrt{4 \pi D \Delta t}} \left\{ \sqrt{4 \pi D \Delta t} \ P(x, \ t) - \left[2D\Delta t \ \exp(- \frac{\overline{\Delta x}^2}{4D\Delta t}) \right]_{-\infty}^{\infty} \right.$$

$$\left. + (D\Delta t)\sqrt{4 \pi D \Delta t} \ \frac{\partial^2 P}{\partial x^2} \right\} + R([\Delta t]^2)$$

$$= P(x, \ t) + D \frac{\partial^2 P}{\partial x^2} \Delta t + R([\Delta t]^2) \qquad (4.11)$$

Accordingly,

$$P(x, \ t + \Delta t) = P(x, \ t) + \frac{\partial P}{\partial t} \Delta t + \frac{1}{2!} (\Delta t)^2 \frac{\partial^2 P}{\partial t^2}$$

$$+ \frac{1}{3!} (\Delta t)^3 \frac{\partial^3 P}{\partial t^3} + \dots \dots$$

$$= P(x, \ t) + \frac{\partial P}{\partial t} \Delta t + R'([\Delta t]^2) \quad (4.12)$$

Combine Eq.(4.11) and Eq.(4.12), yield

$$P(x, \ t) + \frac{\partial P}{\partial t} \Delta t + R'([\Delta t]^2) = P(x, \ t) + D \frac{\partial^2 P}{\partial x^2} \Delta t + R([\Delta t]^2);$$

$$(4.13)$$

Where the functions $R([\Delta t]^2)$ and $R'([\Delta t]^2)$ are functions of t to power higher than 2, so that in the limit where $\Delta t \rightarrow 0$, equation (4.13) becomes

$$\frac{\partial P}{\partial t} = D \frac{\partial^2 P}{\partial x^2} \qquad (4.14)$$

This equation can be used to describe the process of mixing with components of physically identical materials.

(2) The mixing with components of the same size but differ in density.

In this class, the materials obey the first postulate but no the second. As the particles undergo displacements the probability will not be symmetrical i.e. p and q are no longer equal to $\frac{1}{2}$. It is therefore necessary to modify Eq.(4.1) for this requirement, hence

$$P(m, \ N) = \frac{N !}{\frac{1}{2}(N + m) ! \ \frac{1}{2}(N - m) !} (p)^{\frac{1}{2}(N+m)} (q)^{\frac{1}{2}(N-m)} \quad (4.15)$$

This equation can be rewritten in form

$$P(m,\ N)\ =\ C^{N}_{\frac{1}{2}(N+m)}\ (p)^{\frac{1}{2}(N+m)}\ (q)^{\frac{1}{2}(N-m)} \tag{4.15'}$$

clearly the present problem has a poisson distribution.

Suppose $p = \frac{1}{2} - \frac{\xi m}{2}$ and $q = \frac{1}{2} + \frac{\xi m}{2}$ (4.16)

Where p and q are the probabilities of a particle moves forward and backward direction, ξ is a certain physical parameter.

Rewritten Eq.(4.15), becomes,

$$P(m,\ N)\ =\ \frac{N\ !}{\left[\frac{1}{2}(N+m)\right]!\left[\frac{1}{2}(N-m)\right]!}\ \left(\frac{1}{2}-\frac{\xi m}{2}\right)^{\frac{1}{2}(N+m)}\left(\frac{1}{2}+\frac{\xi m}{2}\right)^{\frac{1}{2}(N-m)}$$

$$\tag{4.17}$$

Let $N' = \frac{1}{2}(N + m)$ and $N'' = \frac{1}{2}(N - m)$

Thus Eq.(4.17) becomes,

$$P(m,\ N)\ =\ \frac{N\ !}{N'!\ N''!}\ \left(\frac{1}{2}-\frac{\xi m}{2}\right)^{\frac{1}{2}(N+m)}\left(\frac{1}{2}+\frac{\xi m}{2}\right)^{\frac{1}{2}(N-m)} \tag{4.18}$$

This equation can be simplified with using of Stirling's formula , and similar to Eq.(4.3)

$$\ln P(m,\ N)\ =\ \left(N+\frac{1}{2}\right)\ \ln N\ -\ N\ +\ \frac{1}{2}\ \ln\ (2\pi)\ -\ N'\ln N'$$

$$+\ N'\ -\ \frac{1}{2}\ln(2\pi)\ -\ N''\ln N''\ +\ N''\ -\ \frac{1}{2}\ln(2\pi)$$

$$+\ \frac{1}{2}(N+m)\ln\left(\frac{1}{2}-\frac{\xi m}{2}\right)\ +\ \frac{1}{2}(N-m)\ln\left(\frac{1}{2}+\frac{\xi m}{2}\right)$$

$$=\ -\ \frac{1}{2}\ln\ N\ -\ \frac{1}{2}\ln\ 2\pi\ +\ \ln\ 2\ -\ \frac{m^{2}}{2N}\left(1-\frac{1}{N}\right)$$

$$+\ \frac{2\xi m^{2}N}{2N}\ -\ \frac{\xi^{2}m^{2}N^{2}}{2N}$$

Since $N \gg 1$ then $1 - \frac{1}{N} \rightarrow 1$, thus

$$\ln P(m,\ N)\ =-\ \frac{1}{2}\ \ln\ N\ -\ \frac{1}{2}\ \ln\ 2\pi\ +\ \ln\ 2\ -\ \frac{m^{2}}{2N}\ +\ \frac{2\xi mN}{2N}\ +\ \frac{\xi^{2}m^{2}N^{2}}{2N}$$

or

$$P(m,\ N)\ =\sqrt{\frac{2}{\pi N}}\ \ \exp\left[-\ \frac{(m-\xi mN)^{2}}{2N}\right] \tag{4.19}$$

Let, $x = \delta m$, $N = n t$ and $D = \frac{1}{2} n\delta^{2}$ as before,

so Eq.(4.19) becomes,

$$P(x,\ t)\ =\ \frac{1}{\sqrt{4\pi Dt}}\ \ \exp\left[-\ \frac{(x-\xi nxt)^{2}}{4Dt}\right] \tag{4.20}$$

Further let $\xi n\ x = \beta$, so that,

$$P(x,\ t) = \frac{1}{\sqrt{4\pi Dt}}\ \exp\left[-\frac{(x-\beta t)^2}{4Dt}\right] \qquad (4.21)$$

A differential equation can be derived by a similar consideration as before and the probability of any displacement Δx at time Δt will be given by

$$Q(\Delta x,\ \Delta t) = \frac{1}{\sqrt{4\pi D\Delta t}}\ \exp\left[-\frac{(\Delta x-\beta\Delta t)^2}{4D\Delta t}\right] \qquad (4.22)$$

Hence analogous to Eqs.(4.10), (4.11), (4.12), the probability distribution of $P(x,\ t+\Delta t)$ is,

$$P(x,\ t+\Delta t) = \int_{-\infty}^{\infty} P(x-\Delta x,\ t)\ Q(\Delta x,\ \Delta t)\ d(\Delta x) \qquad (4.23)$$

so,
$$P(x,\ t+\Delta t) = P(x,\ t) + \frac{\partial P}{\partial t}\Delta t\ + R'([\Delta t]^2)$$

$$\int_{-\infty}^{\infty} P(x-\Delta x,\ t)\ Q(\Delta x,\ \Delta t)\ d(\Delta x)$$

$$= \frac{1}{4\pi D\Delta t}\int_{-\infty}^{\infty}\exp\left[-\frac{(\Delta x-\beta\Delta t)^2}{4D\Delta t}\right]\left\{P(x,t)\ + (-\Delta x)\frac{\partial P}{\partial x}\right.$$

$$\left. +\ \frac{1}{2!}(-\Delta x)^2\frac{\partial^2 P}{\partial x^2}\ +\ \frac{1}{3!}(-\Delta x)^3\frac{\partial^3 P}{\partial x^3}\ +\ \dots\right\}d(\Delta x)$$

$$= P(x,\ t)\ -\ \frac{\partial P}{\partial x}\Delta t\ +\ D\ \frac{\partial^2 P}{\partial x^2}\Delta t\ +\ R([\Delta t]^2)$$

so that
$$P(x,\ t) + \frac{\partial P}{\partial t}\Delta t\ + R'([\Delta t]^2) \qquad (4.24)$$

$$= P(x,\ t)\ -\ \beta\ \frac{\partial P}{\partial x}\Delta t\ +\ D\ \frac{\partial^2 P}{\partial x^2}\Delta t\ +\ R([\Delta t]^2)$$

Passing to the limit $\Delta t \rightarrow 0$, therefore the solution is obtained as:-

$$\frac{\partial P}{\partial t}\ =\ D\ \frac{\partial^2 P}{\partial x^2}\ -\ \beta\frac{\partial P}{\partial x} \qquad (4.25)$$

Eq.(4.25) may be used to describe the mixing process when components contains the same size but differ in density.

(3) The mixing with components of differ in size and density.

In this class, the materials do not obey both postulates, thus it seems unreasonable and difficult for one to make two modifications at the same time. As it involves not only the change of

momentum but also the particle packing problem. One may demonstrate this situation in the following example :-

In Fig.(4.2), there are 10 particles all different in size on a one-dimensional plane.
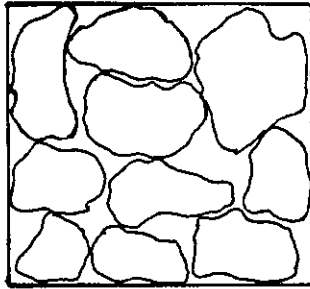
Fig. 4.2.

Each particle occupies its own projected unit area which are all different. One particle picked from the system can not be replaced by any other particle without changing the original form. As the mixing problem is a dynamic situation, when all these particles undergo displacements, it is clear that they can not move randomly. Thus the stochastic analogy can not be applied to the system. If all particles are arranged as shown in Fig.(4.3),
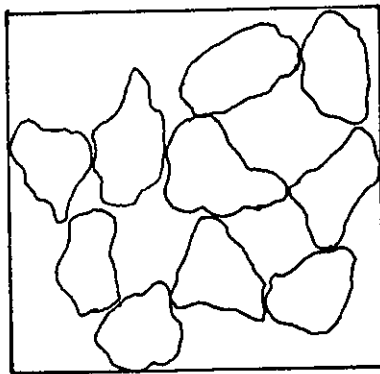
Fig. 4.3.

although they are not equal sized but the particles may change their positions without causing any change to the system. In this condition, the particles in the mixing system could freely undergo displacements, thus the mixing process may be decribed by a 'pseudo-stochastic' (con-

ditional) process. This class will be considered further after the experimental results are obtained, then a definitive conclusion may be drawn.

4.3. CONCLUSION AND DISCUSSION

The mixing of free flowing powders may be described by Eq.(4.14) and Eq.(4.25) dependent on the nature of the material properties.

Eq.(4.14) has the same form of Fick's second law of diffusion and demonstrates the validity of the earlier conclusions. It further demonstrates the proposal by Lacey that the obtaining of this type of equation does not necessary mean that the process is diffusion (49). The value of obtaining the equation in this rigorous way is seen in the introduction of the parameter $D = \frac{1}{2} n\delta^2$. This parameter must depend therefore on both material and mixer properties. As this equation has been developed from Bernoullian distribution, it can only be applied to a homogeneous system and only such a system can achieve a state of randomisation. Provided that both basic postulates are satisfied an equation of the type Eq.(4.14) (i.e. similar to Fick's equation) will be obtained regardless of the type of mixing equipment used.

Eq.(4.25) has a form similar to an equation which Müller and Rumpf proposed, namely the Kolmologov second equation. This equation was claimed to represent genera-lly the powder mixing process. From the present derivation of this equation, it shows that the equation may be suitable to describe the mixing of components which differ in density but have the same size and so the equation is not completely general. Initially, this equation has shown a feature of the Poisson distribution. So the probability distribution will be asymmetric. Thus this mixing system can not achieve a state of randomisation and segregation will occur. If the density difference is very samll, so that the symmetrical displacement of particles may not be affected, as $\xi \longrightarrow 0$, then $p \cong q$ and $\beta \longrightarrow 0$. In this circumstance, Eq.(4.25) becomes Eq.(4.14) and the mixing process may achieve a state of randomisation.

For mixing with components which differ in size and density, all particles undergo displacements according their surroundings,

therefore the movement of particles is not stochastic (random). Unlike systems of gases and liquids in which the volume of the particles is often ignored, in a particulate solid system the actual volume of each particle plays an important part in the whole process.

It could be concluded from the above study that the fully randomised mixture could only be obtained by mixing physically identical components, whenever the components differ in size or density some inhomogeneity will occur. The causes of the in-homogeneity, or segregation, are different when mixing two components differing only in density and when mixing components of different size. In former case segregation is mainly caused by the momentum difference (or by gravitational force), once segregation is established it becomes stable and the application of further energy will not change the pattern obtained. Segregation is caused in the latter case by the rearrangement of particles. This involves both the change of the occupied volume by indi-vidual particles and by the change of momentum. These changes would probably continue until all similar particles are together then the system will become stable. However any external force may affect the stability and the pattern of segregation may change. This was also note by Brown (11).

This theoretical analysis is now tested experimentally in the following chapter.

CHAPTER   FIVE

EXPERIMENTAL   WORK

TECHNIQUE,   RESULTS,   DISCUSSION AND CONCLUSION

103

5.1. INTRODUCTION

The experimental work was designed as a systematic investigation of four series of material combinations. These are :- (a) the same size distribution and same density, (b) same density, different size distribution, (c) same size distribution different density (in this series the effect of particle shape was also investigated) and (d) different size distribution and density. The experimental programme was envisaged as having three main objects.

The first object was to test the theory developed in Chapter 4 with the actual mixing system.

The second object of the experiments was to study the mixing and segregation with components of different physical properties and attempt the prediction of the final state of the mixture.

The final object of the experimentation was concerned with the operating conditions and geometric dimensions of the mixers.

It was impossible to plan a comprehensive experimental programme for all existing mixing problems within the present period of investigation. However, it is hoped that the results presented provide a broad picture and sufficient evidence on which general theories of mixing and segregation may be based.

For easy reading of the experimental results, all diagrams will appear with the text and the representative numerical data will be tabulated in the Appendix I.

5.2. EXPERIMENTAL EQUIPMENT, MATERIALS AND TECHNIQUE

5.2.1. MATERIALS AND METHOD OF PREPARATION

The materials used in the experiments should be representative of the general physical behaviour of granular solids. Therefore, sand (Photo 5.1A) (Moneystone A1, supplied by B.I.S.) with different size ranges were used for most of the experiments, and gypsum (Photo 5.1B) (from B.G.L.), glass beads (Photo 5.1C) (from E.G.C.) and zicron sand (Photo 5.1D) (from Podmore & Son Ltd.) were

also used to form a binary mixing system with differences in density, shape and size combinations. The main physical properties and size ranges of those materials are listed in Table 5.1.

TABLE 5.1. PHYSICAL PROPERTIES OF MATERIALS

Materials Physical Parameters	Sand		Gypsum	Zicron		Glass beads
	S-1	S-2		Z-1	Z-2	
Type	Moneystone		Commer-cial	Industrial		Commer-cial
Size (B.S.S.)	-36/+52	-52/+60	-36/+52	-36/+52	-52/+60	-36/+52
Density (gm/ml.)	2.58		2.14	4.66		2.89
Shape	Granular		Granular	Granular		Sphroid

The size distribution curves between a limited sieve range are shown separately in Fig.(5.1) to Fig.(5.3). The size range were selected so that the particles were easily visible under a magnifying glass and could be separated with miniature sieves. The particle size was also selected so that a reasonable number of particles were obtained in a small sample.

Sand was coloured with a dye to provide particles differing only by colour. The dyeing process presented no special problem. Sand was sieved to give the required cut in particle size. Half of the quantity produced was washed and dyed with a black household dye, 'Dynol' by trade name. The dye was dissolved in boiling water, the material was slowly added into the dye solution and stirred frequently for 15 minutes. It was then placed in a laboratory oven to be dried and sieved again before use. The dyed material was always used as one of the components whenever a binary mixing system was studied.

5.2.2. DESCRIPTION OF MIXING APPARATUS

5.2.2.1. MIXERS

The mixers were of the horizontal rotating drum type, consisting of four different geometric dimensions. The bodies of the

FIG. 5.1. SIZE DISTRIBUTION OF SAND   O −52/+60
△ −36/+52
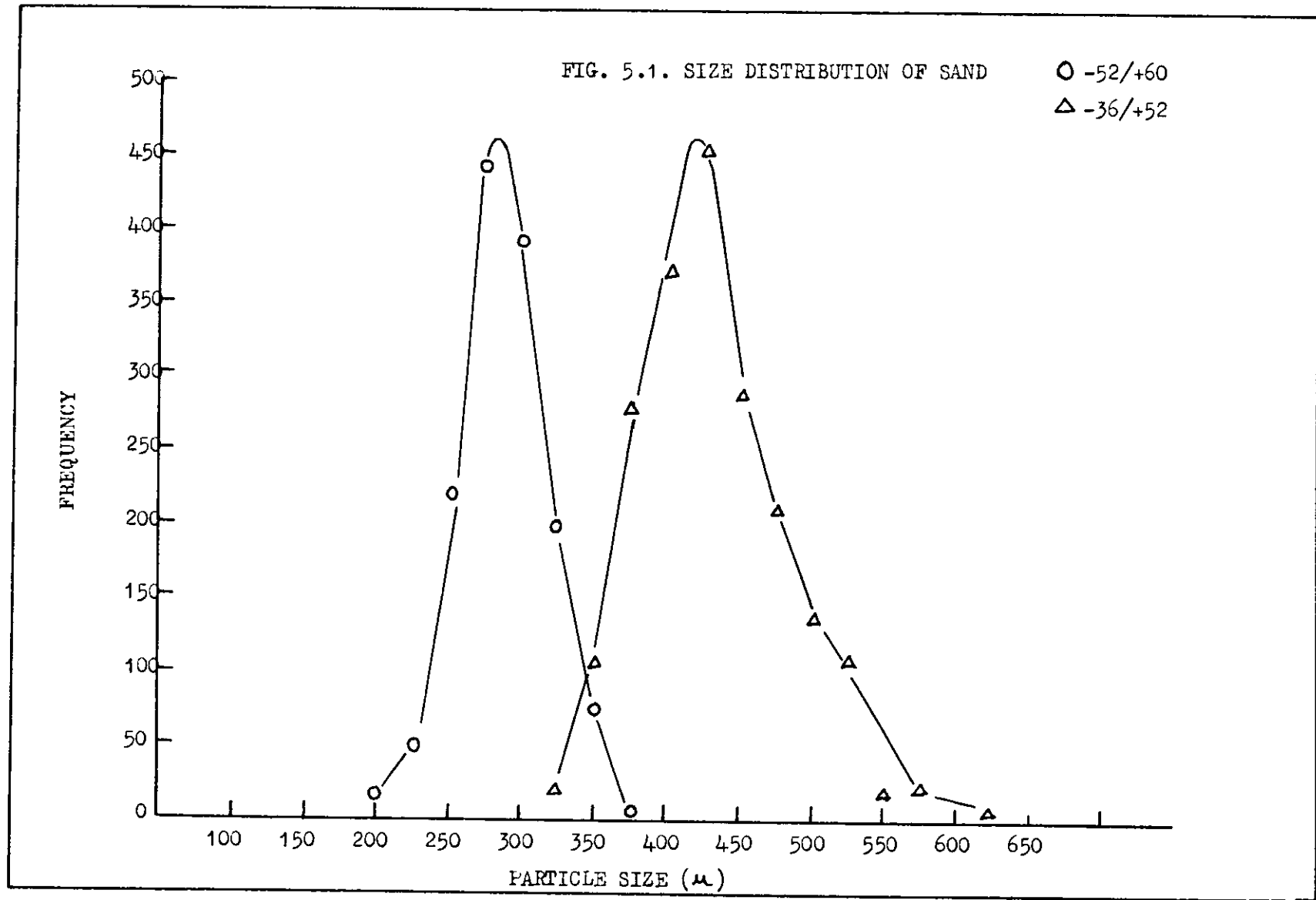
FIG. 5.2. SIZE DISTRIBUTION OF GLASS BEADS AND GYPSUM
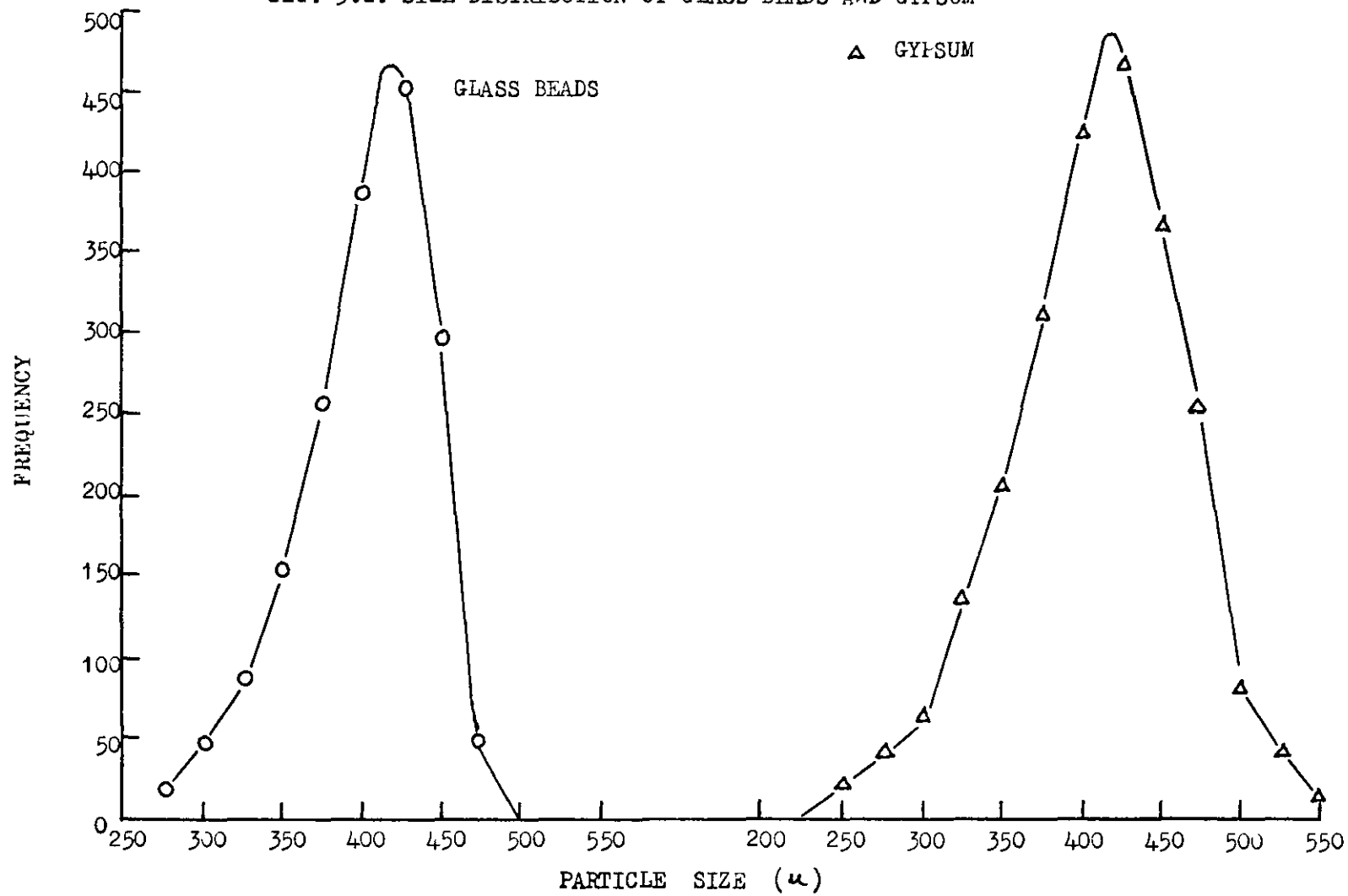
FIG. 5.3. SIZE DISTRIBUTION OF ZICRON SAND
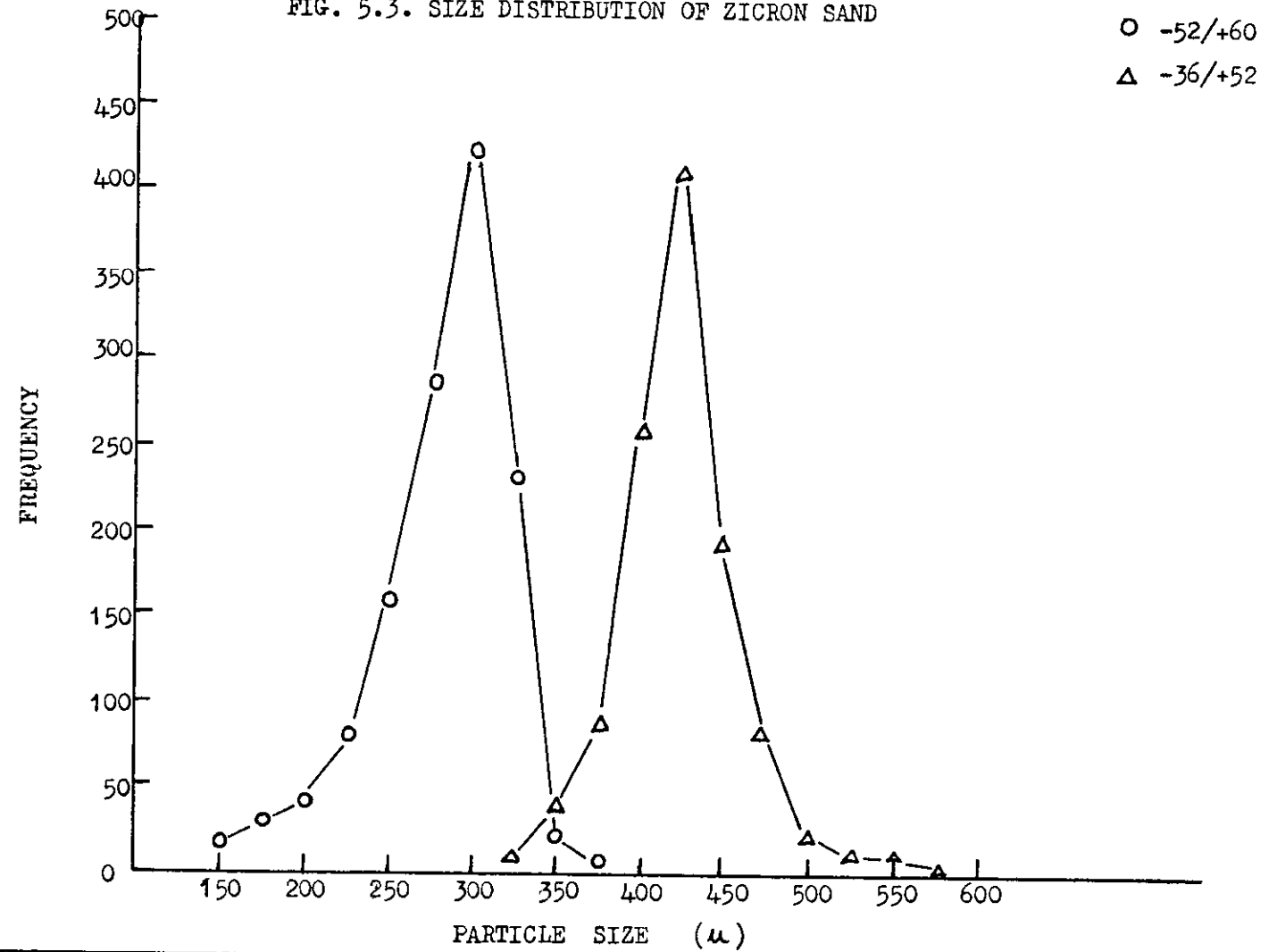
mixer were constructed of perspex cylinder or sheet with the removable lids. The dimensions are given is Table 5.2., and the outline

TABLE 5.2.   DIMENSIONS OF MIXERS

Type	L/d	Length L (cm)	Diameter d (cm)	Capacity (litres)
M-1	2	29.5	14.5	4.7
M-2	1	15	14.5	2.8
M-3	3	44.5	14.5	7.5
M-4	1	18.2	18.2	4.6

drawing is given in Fig.(5.4). Photo (5.2) shows the various sized mixers and Photo (5.3) shows that the mixer while in used.

For thorough sampling across the mixing bed, one of the end walls of the mixer contained 9 sampling holes sealed with leak proof rubber pluges. The general arrangement of those sampling hole is shown in Fig.(5.4).

5.2.2.2. DRIVING APPARATUS

The driving apparatus consists of a variable width ball mill rollers and an electric motor. The speed of the electric motor is controlled by a coupled reducing gear box of Synchromic single phase type motor (1/3 Hp) (supplied by Allspeed Ltd.) which allows a desired speed to be set accurately over a range from 30 to 280 rpm.

Photo (5.4) shows the complete arrangement of the apparatus.

5.2.3. GENERAL METHOD OF LOADING OF THE MATERIALS

The mixer was first placed on a special mixer holder, a special semi-circluar plate was placed at the middle of the mixer. Materials were loaded into both compartments separately and leveled. The thin plate was then removed slowly without disturbing the material. The lid of the mixer was closed. The material   arrangement is shown in Photo (5.5). The mixers were always filled to 40% of the total volume.

5.2.4. THE SAMPLERS AND SAMPLING TECHNIQUE

Modified thief samplers were used which consist of an inner rod of brass rod (BS 249 rod) and the outer sleeve of Alrecs tele-
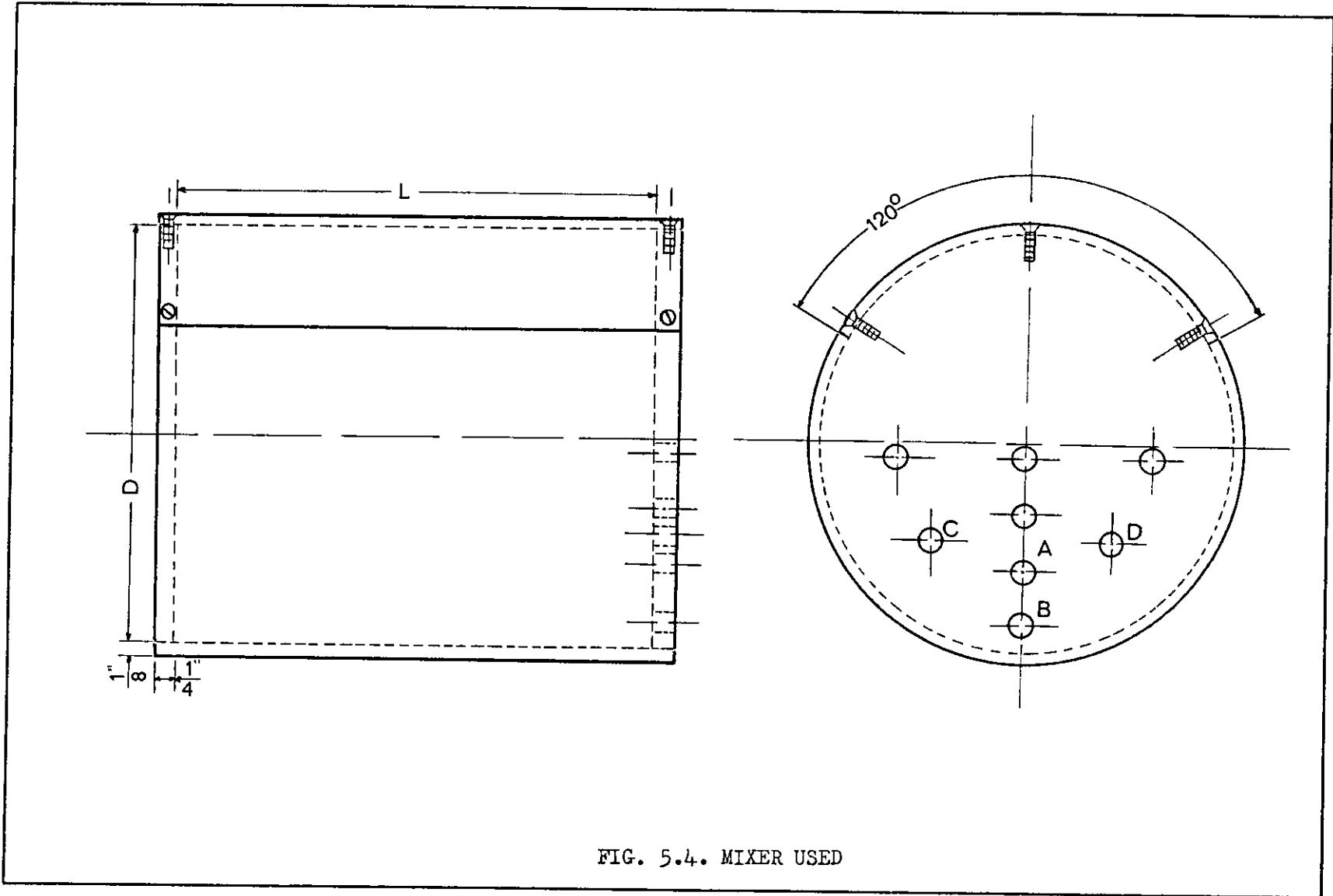
FIG. 5.4. MIXER USED

FIG. 5.5. SAMPLER USED

mark lined up with holes

A—A

B—B
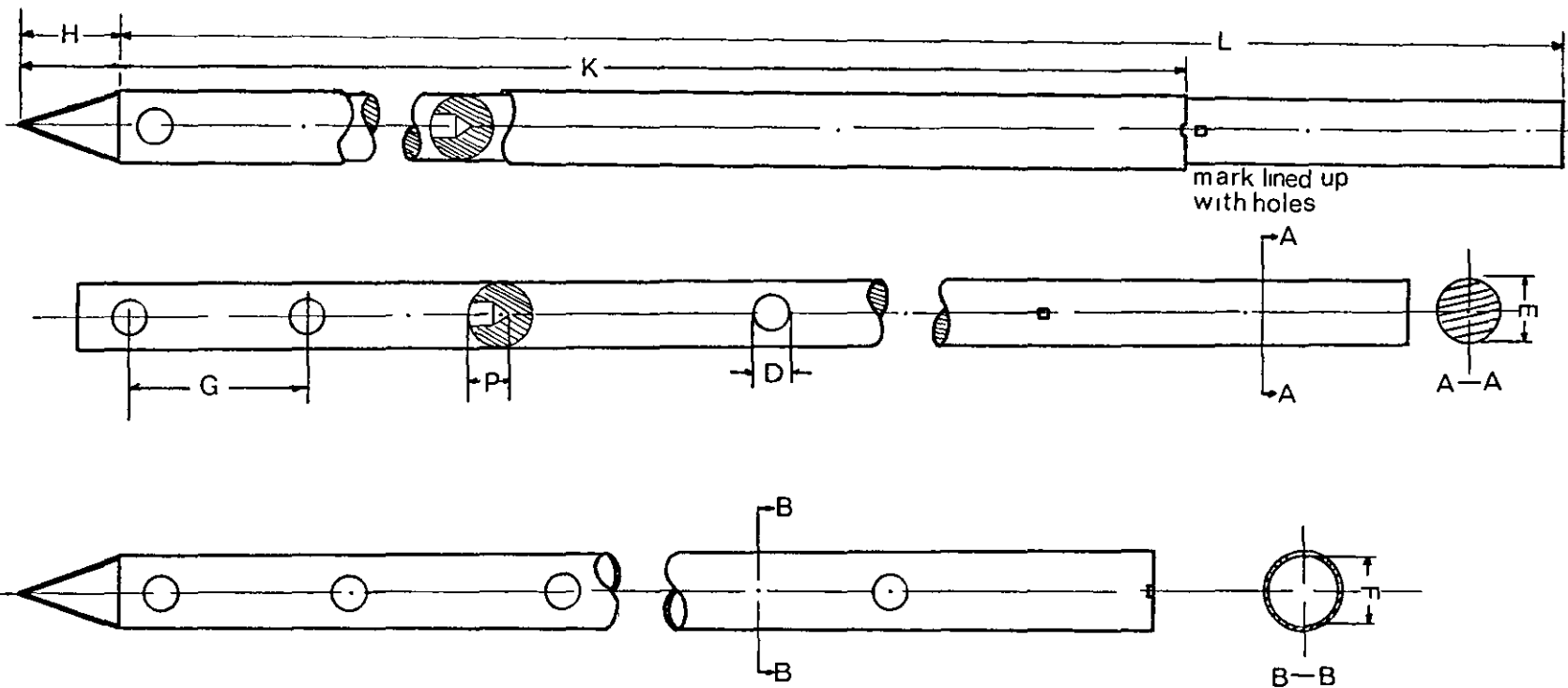
TYPE	DIMENSION								TYPE	DIMENSION							
No.1	D	E	F	G	H	K	L	P	NO 2	D	E	F	G	H	K	L	P
	3/32"	5/32"	0.167"	7/8"	1 3/16"	14"	18"	1/8"		7/32	9/32"	0.291"	7/8"	1 3/16"	14"	18"	7/32"
	3/32"	5/32"	0.167"	0.875"	0.8125"	14"	18"	1/8"		7/32	9/32"	0.291"	7/8"	13/16"	14"	18"	7/32"
	3/32"	5/32"	0.167"	0.875"	15/16"	14"	18"	1/8"		7/32	9/32"	0.291"	1 3/8"	15/16"	21"	30"	7/32"
										7/32	9/32"	0.291"	7/8"	15/16"	14"	18"	7/32"

scopic brass tubing (supplied by RZT Metals). The rod is drilled to give sampling holes at 7/8" intervals. The sleeve has a sharp point at one end, holes at 7/8" intervals corresponding to the holes in the rod.

Different sized samplers were used (Photo 5.6) for various sized mixers. The outline drawing of the sampler and the principle dimensions are given in Fig.(5.5). Sampler No.1 was used for taking small samples (for particle counts). The sampling hole contains approximately 200 sand particles (sand S-1). No.2 samplers were used for large samples, the hole containing approximately 1.0g of sand.

Sampling positions were selected in such a way that they were given systematic sampling throughout the mixture. In general the sampler was inserted at two horizontal levels marked A & B on Fig.(5.4) and also alternatively at the levels marked C & D.

For the mixing of same sized components, particles counts of each components were made on each spot sample with a illuminated magnifier. The particle fraction of dyed and not dyed sand (or other components) was computed for each sample and the result noted.

For the mixing of different sized components, a miniature sieve (3" diameter) was used to separate the components. Each component was weighed on a Oerters-O-Matics balance which could weigh to 0.0001g. The weight fraction of each component was calculated and the corresponding position of the sample noted.

5.2.5. ANALYSIS OF THE EXPERIMENTAL RESULTS

5.2.5.1. THE SELECTION OF A DEGREE OF MIXING

Herdan (39) stated that "There is no such thing as a complete mixture in the sense that all the samples taken from the mixture, no matter how small, will be exactly alike. Since there will be random fluctuations in the sample quality even in the most homogeneous mixture". From this statement, it is clear that the samples taken from the mixture implied a deviation from the true mean. Therefore the uniformity of the mixture will proportional to this deviation. Thus a degree of mixing can be proposed which is based on this deviation, the standard deviation. When the mixing is 'complete', the standard deviation becomes a minimum. If the mixing process could be

described by a Bernoullian distribution, clearly, the minimum standard deviation is the random standard deviation.

Most proposals for a definition of the degree of mixing include the standard deviation of a random binary mixture. The random standard deviation or its square, the variance, for mixture distinguished only by colour is given by

$$\sigma_R^2 = \frac{p(1-p)}{n} \tag{5.1}$$

where p is the particle fraction of one of the components and n is the total number of particles per sample.

For mixture of components of different size distribution

$$\sigma_R^2 = \frac{(PQ)}{g}\left[P\overline{V}_Q(1 + C_Q^2) + Q\overline{V}_P(1 + C_P^2)\right] \tag{5.2}$$

(for nomencleture see p.17)

However, from the last Chapter, it is understood that the mixture could reach a state of randomisation when components distinguished only by colour. Therefore Eq.(5.2) may not valid in the actual mixing system. The selection of a degree of mixing for all cases, therefore the between sample variance would be most adequate, i.e.

$$\sigma^2 = \frac{\Sigma(p - \bar{p})^2}{N - 1} \tag{5.3}$$

or

$$\sigma^2 = \frac{\Sigma(p - a)^2}{N} \tag{5.3'}$$

where p is the measured fraction, and $\bar{p}$ the mean fraction of N samples, a is the true fraction of the component in the whole mixture.

This degree of mixing has the virtue of simplicity, but the disadvantage that there is no convenient index on a scale between 0 and 1 (such as obtained in the equation proposed by Lacey).

The between sample variance σ^2 was based on the particle fraction (for same sized components) and weight fraction (for different sized components).The relationship between these two values of variance may be obtained (83)

Let a set of samples A in which the particle fraction is p and the weight fraction is w

$$p - w = \delta$$

Taking the mean value of p and w so,

$$\bar{p} = \bar{w} + \bar{\delta}$$

$$(p_1 - \bar{p}) = (w_1 - \bar{w}) + (\delta_i - \bar{\delta})$$

$$(p_1 - \bar{p})^2 = (w_1 - \bar{w})^2 + 2\,(w_1 - \bar{w})(\delta_i - \bar{\delta}) + (\delta_i - \bar{\delta})^2$$

$$\frac{\sum (p_i - \bar{p})^2}{N} = \frac{\sum (w_i - \bar{w})^2}{N} + 2\,\frac{\sum (w_i - \bar{w})(\delta_i - \bar{\delta})}{N} + \frac{\sum (\delta_i - \bar{\delta})^2}{N}$$

or

$$\sigma_p^2 = \sigma_\omega^2 + \sigma_\delta^2 + \frac{2\sum (w_1 - \bar{w})(\delta_i - \bar{\delta})}{N}$$

Let,

$$p_1 = g_{A_1}\rho_A / (g_{A_1}\rho_A + g_{B_1}\rho_B)$$

$$w_1 = g_{A_1} / (g_{A_1} + g_{B_1})$$

Where

g_A is g.of A-component in a sample

g_B is g. of B-component in a sample

ρ_A is particles of A-component per g,   and

ρ_B is particles of B-component per g.

Let

$$\rho_A = k\,\rho_B$$

Then

$$\delta_i = K\,g_{A_i}\rho_B / (K\,g_{A_1}\rho_B + g_{B_1}\rho_B) - g_{A_i}/(g_{A_1} + g_{B_1})$$

$$= K\,g_{A_1}/(K\,g_{A_i} + g_{B_1}) - g_{A_i}/(g_{A_1} + g_{B_1})$$

Thus δ would depend on the value of fraction and K, it could be determinated for various combinations of these two parameters.

1) For components of same density

as K = 1, $\delta_i = 0$, so,

$$\sigma_p^2 = \sigma_\omega^2 \qquad \text{for all cases}$$

ii) For components of different density

as $K \neq 1$

a) $g_A/(g_A + g_B) = 0$, $\delta = 0$

so $\sigma_p^2 = \sigma_\omega^2$

b) $g_A/(g_A + g_B) = 1$ $\delta = 0$, so $\sigma_p^2 = \sigma_\omega^2$.

c) $g_A/(g_A + g_B) = k$, then $g_A = kg_B + kg_A$

or $g_A = kg_B/(1 - k)$

thus

$$Kg_{A_1}/(Kg_{A_1} + g_B) = K(\frac{k}{1 - k})/\left[K(\frac{k}{1 - k})g_B + g_B\right]$$

$$= Kk/(Kk - k + 1)$$

so that $\delta = Kk/(Kk - k + 1) - k = \dfrac{Kk(1 - k) + k(k - 1)}{Kk - k + 1}$

Hence $\sigma_p^2 \neq \sigma_\omega^2$

5.2.5.2. STATISTICAL TESTS

Statistical tests were often made. The Student t test was used for testing a set of samples withdrawn at a particular mixing time to test the difference between the measured mean and the ture mean. The F test was used for testing the duplicated runs. These tests are recommended by Am. I. Chem. Eng. solid mixing equipmemt test (2).

5.2.6. EXPERIMENTAL PROCEDURE

The mixer was placed on a specially made holder and materials were loaded as described. The mixer was then replaced on the roller machine. The electric motor was set a fixed speed of rotation. The time was recorded by a stop watch. At each interval of mixing the machine was stopped and mixer turned by hand to the position required for convenient sampling. It was then replaced on the holder and samples taken. The interval of each mixing time was varied from 30 seconds to two hours. Generally during the initial stage, very short intervals were taken. The intervals were increased as mixing tended to near the equilibrium state.

Since only few points could be taken and analysed each day. Each run of experiment extended over many days. The mixture was left undisturbed between sampling.
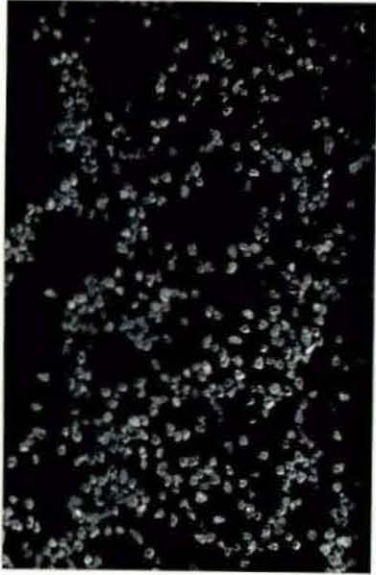
A. MONEYSTONE SAND

B. GYPSUM
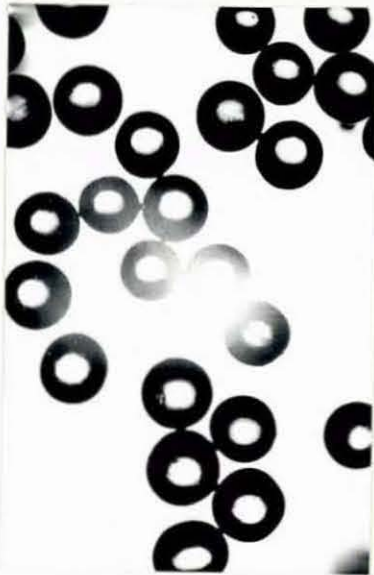
C. GLASS BEADS

D. ZICRON SAND

PHOTO 5.1. MATERIALS USED (NOT TO SCALE)
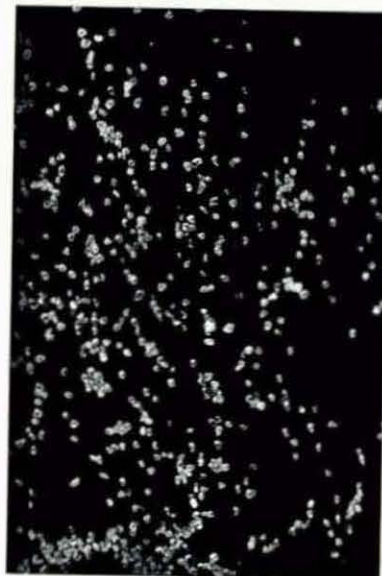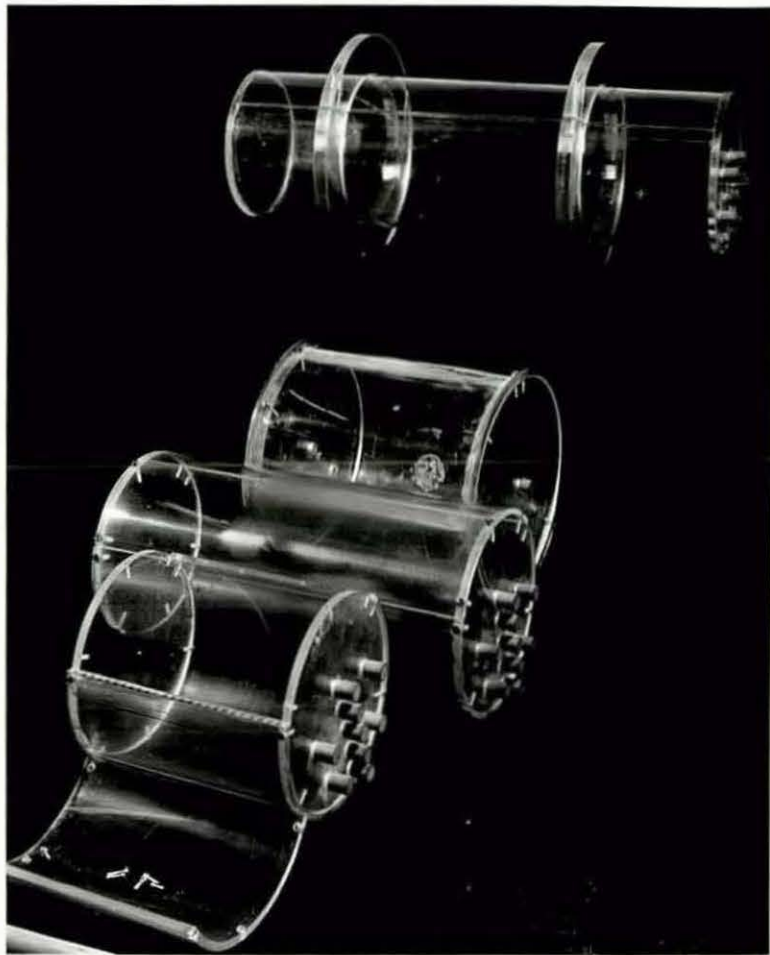
PHOTO 5.2. MIXERS USED
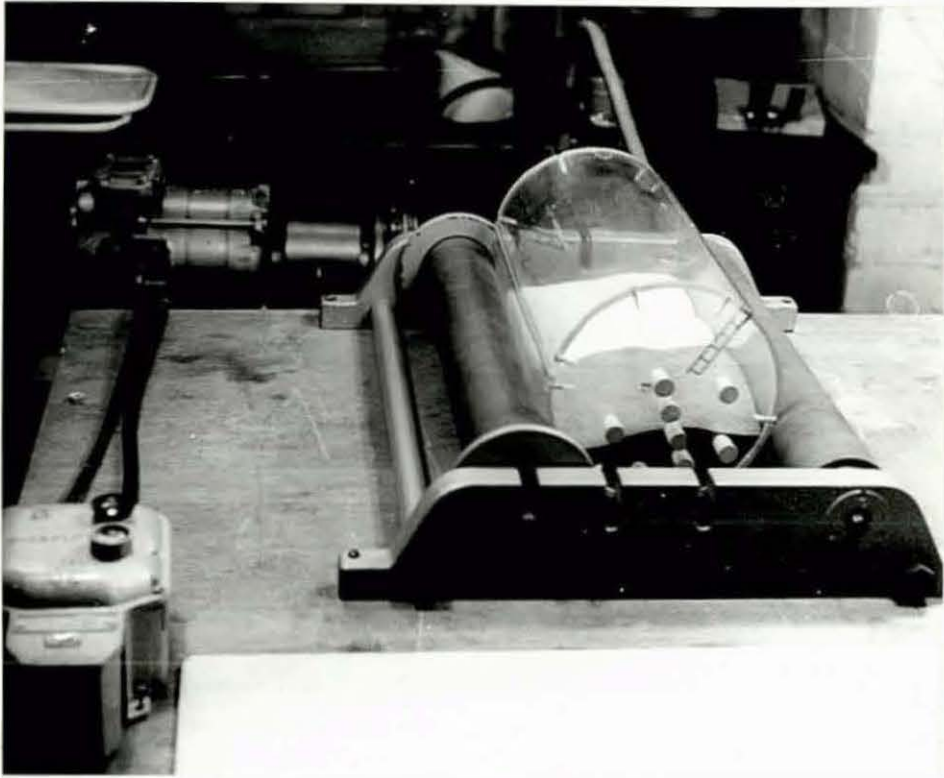
PHOTO 5.3. MIXER WHILE IN USE
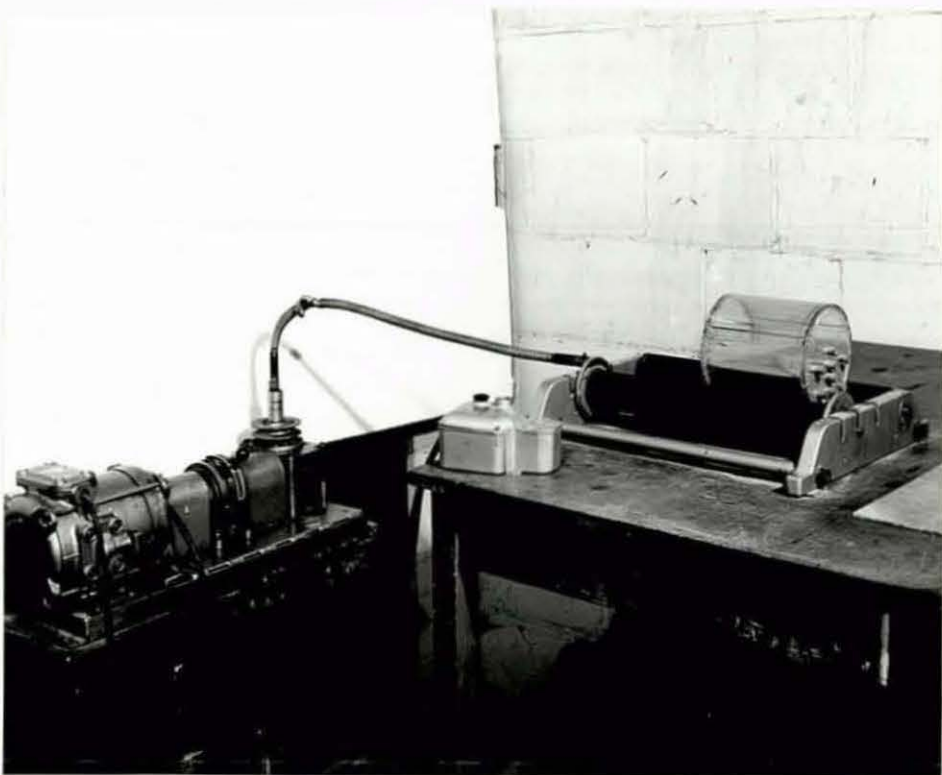
PHOTO 5.4. COMPLETE ARRANGEMENT OF THE APPARATUS
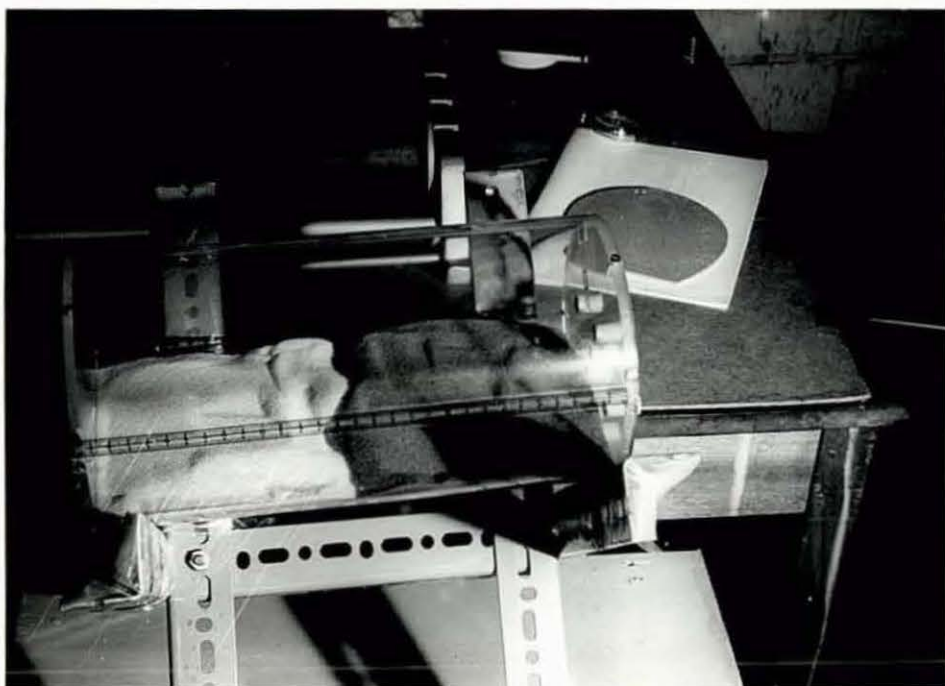
PHOTO 5.5. LOADING OF THE MATERIALS
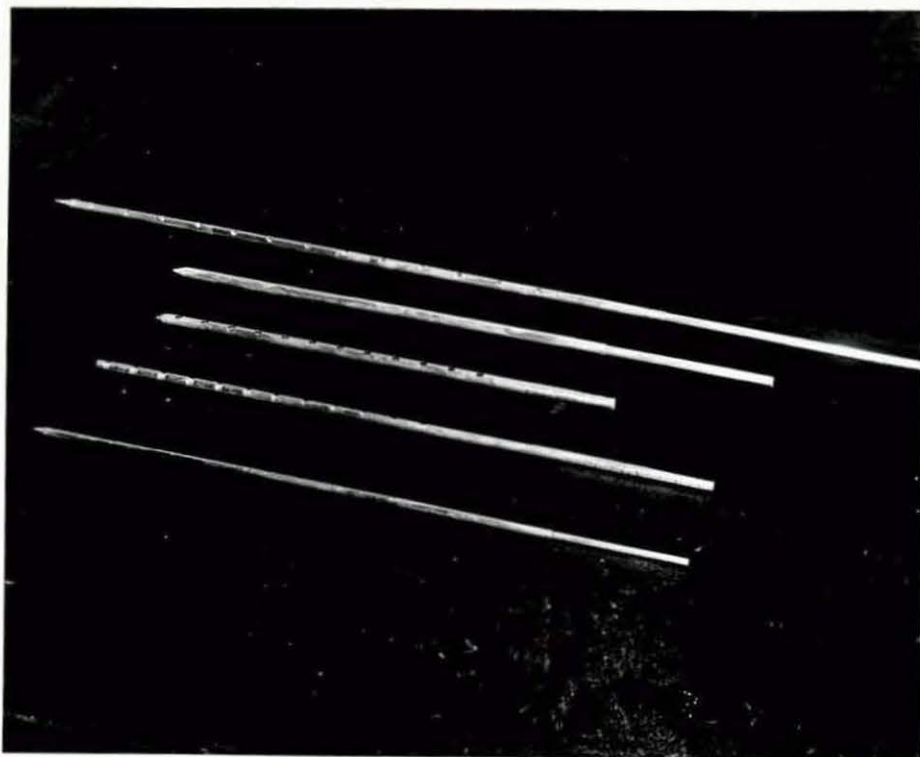
PHOTO 5.6. SAMPLERS USED

5.3. SOLUTION OF THE DEVELOPED MIXING EQUATION

The mechanisms of mixing with physically identical components and the components differ in density only, they may be described by the equations

$$\frac{\partial P}{\partial t} = D\frac{\partial^2 P}{\partial x^2} \tag{4.14}$$

and

$$\frac{\partial P}{\partial t} = D\frac{\partial^2 P}{\partial x^2} - \beta\frac{\partial P}{\partial x} \tag{4.25}$$

The solutions of these equations can be used to determine the rate of mixing and the distribution profiles.

Eq.(4.14) has shown a characteristic of a Bernoullian distribution, the end point of the mixture in such system may be assumed to be that of the randomised mixture. Thus the solution can be easily obtained. However Eq.(4.25) represents a Poisson distribution and it would be unreasonable to assume an end point of the mixture in such system. Therefore, Eq.(4.25) will not be solved in the present section, and it will not be considered until the experimental results have been obtained in the later section.

Solution of equation (4.14)

When time of mixing is approaching to infinity, the concentration (probability) distribution is expected to be normal, but for the mathematical reason, this may be assumed that the end point of the mixture equal to a unique constant, i.e. the proportion of overall components. The equation can be solved for the initial conditions described in Section 5.1.

For the sake of simplicity, let a new probability function G be defined such that it varies from an initial value P_0 to a final value $\frac{1}{2}P_0$ where P_0 is the proportion of one component at time $t = 0$.

Hence

$$G = 2\left(\frac{P}{P_0} - \frac{1}{2}\right) \tag{5.4}$$

Rewritten Eq.(4.14), becomes

$$\frac{\partial G}{\partial t} = D\frac{\partial^2 G}{\partial x^2} \tag{5.5}$$

Let $\qquad G = yz$

where y is function of x only, and z is function of t only, as

$$y = f(x) \quad \text{and} \quad z = f(t)$$

Hence Eq.(5.5) becomes,

$$\frac{\partial (yz)}{\partial t} = D \frac{\partial^2 (yz)}{\partial x^2}$$

Differentiating,

$$\frac{1}{D z} \frac{\partial z}{\partial t} = \frac{1}{y} \frac{\partial^2 y}{\partial x^2} \qquad (5.6)$$

When x varies, left handside of the equation remains constant, as z is function of t only, similarly, when t varies, right handside of the equation remains constant. Let $-a^2$ be the common constant, and the equation can be solved in two parts. Hence

$$\frac{\partial^2 y}{\partial x^2} = -a^2 y \qquad (5.7)$$

and $\qquad \dfrac{1}{D z} \dfrac{\partial z}{\partial t} = -a^2 \qquad (5.8)$

The general solutions of Eq.(5.7) and Eq.(5.8) are

$$y = K_1 \cos ax + K_2 \sin ax \qquad (5.9)$$

$$z = k_3 \exp(-Da^2 t) \qquad (5.10)$$

Thus, $\qquad G = yz = K_3 \exp(-a^2 Dt)(K_1 \cos ax + K_2 \sin ax) \qquad (5.11)$

As the materials were loaded end to end as sketched in Fig.(5.6), the boundary conditions can be fixed.
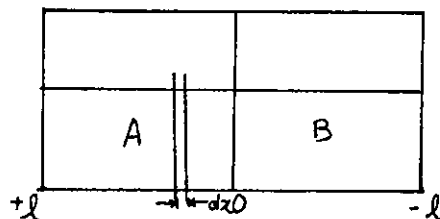
Fig.(5.6)

The boundary conditions are;-

(1) $t = 0$, $G = 1$, $P = P_0$

(2) $t = \infty$, $G = 0$, $P = \frac{1}{2} P_0$

(3) $x = 0$, $G = 0$

(4) $\dfrac{\partial G}{\partial x} = 0$, $x = \ell$

The particular solution will be,obtained as follows,

Using boundary condition (2) for Eq.(5.11), so that

$K_1 = 0$,

this also satisfies (3), and the general solution becomes

$$G = K_4 \exp(-a^2 Dt)\sin ax \qquad (5.12)$$

Where $K_4 = K_2 K_3$.

Differentiation of Eq.(5.12) with using boundary condition (4)

$$\dfrac{\partial G}{\partial x}\Bigg]_{x\,=\ell} = 0 = K_4 \exp(-a^2 Dt)\cos a\ell \qquad (5.13)$$

But $\quad K_4 \exp(-a^2 Dt) \neq 0$, so

$\cos a\ell = 0$,

thus

$$a = (2n - 1)\dfrac{\pi}{2\ell} \qquad (5.14)$$

where n is any positive integral number $(1, 2, 3, \ldots\ldots)$

Put Eq.(5.14) into Eq.(5.12), then becomes,

$$G = K_4 \exp\left[-(2n-1)^2\dfrac{\pi^2 Dt}{4\ell^2}\right]\sin(2n-1)\dfrac{\pi x}{2\ell} \qquad (4.15)$$

For n number of results, Eq.(4.15) should be repressed as,

$$G = A_n \exp\left[-(2n-1)^2\dfrac{\pi^2 Dt}{4\ell^2}\right]\sin(2n-1)\dfrac{\pi x}{2\ell} \qquad (4.16)$$

Using boundary condition (1) for equation(5.16), so,

$$1 = A_1 \sin\dfrac{\pi x}{2\ell} + A_2 \sin\dfrac{3\pi x}{2\ell} + A_3 \sin\dfrac{5\pi x}{2\ell} + \ldots \qquad (4.17)$$

Eq.(5.17) compared with

$$1 = \frac{4}{\pi} \left(\sin \frac{\pi x}{2\ell} + \frac{1}{3} \sin \frac{3\pi x}{2\ell} + \frac{1}{5} \sin \frac{5\pi x}{2\ell} + \ldots \right) \quad (5.18)$$

so that,

$$A_n = \frac{4}{\pi(2n-1)} \quad , \quad n = 1,2,3,\ldots$$

Thus

$$G = \sum \frac{4}{\pi(2n-1)} \left\{ \exp\left[-\left(\frac{2n-1}{2\ell}\right)^2 \pi^2 Dt \right] \sin\left(\frac{2n-1}{2\ell}\right)\pi x \right\}$$

or

$$G = \frac{4}{\pi} \left\{ \exp\left[-\left(\frac{\pi^2 Dt}{4\ell^2}\right) \right] \sin \frac{\pi x}{2\ell} + \frac{1}{3} \exp\left[-9\left(\frac{\pi^2 Dt}{4\ell^2}\right) \right] \sin \frac{3\pi x}{2\ell} \right.$$

$$\left. + \ldots \ldots \right\} \quad (5.19)$$

Using the relation of Eq.(5.4), therefore the particular solution for the mixing system is obtained, as

$$P = \frac{P_0}{2} (G + 1) \qquad \text{or for } P_0 = 1 \text{ then}$$

$$P = \frac{1}{2} + \frac{2}{\pi} \left\{ \exp\left[-\left(\frac{\pi}{2}\right)^2 \frac{Dt}{\ell^2} \right] \sin \frac{\pi x}{2\ell} + \frac{1}{3} \exp\left[-9\left(\frac{\pi}{2}\right)^2 \frac{Dt}{\ell^2} \right] \sin \frac{3\pi x}{2\ell} + \ldots \right\} \quad (5.20)$$

Now let F be the fraction of the particles of one component remaining in the same half of the mixer after mixing time t,

$$F = \frac{1}{2} + \frac{1}{2\ell} \int_0^{+\ell} G \, dx$$

$$F = \frac{1}{2} + \frac{4}{\pi^2} \left\{ \exp\left[-\left(\frac{\pi}{2}\right)^2 \frac{Dt}{\ell^2} \right] + \frac{1}{9} \exp\left[-9\left(\frac{\pi}{2}\right)^2 \frac{Dt}{\ell^2} \right] + \ldots \right\} \quad (5.21)$$

and F' be the fraction of those particles which have moved to the other side after the same time.

$$F' = \frac{1}{2} + \frac{1}{2\ell} \int_0^{-\ell} G \, dx$$

$$F' = \frac{1}{2} - \frac{4}{\pi^2} \left\{ \exp\left[-\left(\frac{\pi}{2}\right)^2 \frac{Dt}{\ell^2} \right] + \frac{1}{9} \exp\left[-9\left(\frac{\pi}{2}\right)^2 \frac{Dt}{\ell^2} \right] + \ldots \right\} \quad (5.22)$$

The difference between the fractions is the rate of particle movement from one side to the other. Thus,

$$R(t) = F - F'$$

$$R(t) = \frac{8}{\pi^2} \left\{ \exp\left[-\left(\frac{\pi}{2}\right)^2 \frac{Dt}{\ell^2} \right] + \frac{1}{9} \exp\left[-9\left(\frac{\pi}{2}\right)^2 \frac{Dt}{\ell^2} \right] + \ldots \right\} \quad (5.23)$$

The variance of the probability distribution will be

$$\sigma^2(t) = \frac{1}{2\ell} \int_0^\ell (P - \frac{1}{2})^2 dx$$

$$\sigma^2(t) = \frac{2}{\pi^2} \left[\exp(-\pi^2 Dt/2\ell^2) + \frac{1}{3} \exp(-9\pi^2 Dt/2\ell^2) + \dots \right] \qquad (5.24)$$

Both Eq.(5.23) and Eq.(5.24) contain the parameter D and so can be used to determine its magnitude, and the rate of mixing may be obtained.

5.4. EXPERIMENTAL RESULTS, DISCUSSIONS AND CONCLUSIONS

5.4.1. COMPONENTS WITH THE SAME SIZE AND DENSITY DISTINGUISHED ONLY BY COLOUR

Series of experiments were carried out to test the theory of mixing using dyed and not dyed Moneystone sand (S-1) in three differently dimensioned mixers.

5.4.1.1. COMPARISON OF DIFFERENTLY DIMENSIONED MIXER AT THE SAME SPEED OF ROTATION

Concentration distributions obtained at representative intervals of mixing time are plotted in Fig.(5.7) to Fig.(5.9) for mixers M-1, M-2 and M-4 at 52% of the corresponding critical speed of rotation.* In general, the shape of those curves are similar, apart from the initial period, the profiles of the concentration distribution showed a linear progression. Fig.(5.10) shows the variation of concentration with the fixed positions. From the concentration distribution curves the rate of particle movement $R(t)$ and the variance $\sigma^2(t)$ can be calculated and are plotted as function of mixing time in Fig.(5.11) and Fig.(5.12). Both of these sets of curves have a similar shape and indicate three distinct regions. An initial drop in the main function which may be thought of as a transient stage occuring before the regime, discussed earlier, is established. A straight line showing that the derived theory for such system is applicable and the third region represents an equilibrium value in each function. These regions have also been discussed previously by Yano et al (93). It is also noted that mixer M-4, the large mixer with length equal to diameter gives the faster mixing.

---

*The critical speed of rotation is a speed of rotation at which, according to the laws of centrifugal force, particles on the outer circle do not leave the wall of the drum, that is the centrifuge (35). It also can be computed by the force balance, by equating the force of gravity tending to cause the particles to fall to the centrifugal force tending to carry the particles around

$$N \ (rpm) = \sqrt{\frac{60^2 g}{2\pi^2 d}}$$

where d is the diameter of the drum

$g = 980 \ cm/sec^2$

5.4.1.2. RATE OF MIXING AS A FUNCTION OF SPEED OF ROTATION

Since a powder is a particulate system, the packing and the porosity of the mixing bed can affect the rate of mixing. The variation of the porosity of sand as a function of mixer speed of rotation is shown in Fig.(5.13). The results were obtained from two different diameter mixers, showed as only a function of the proportion of critical speed of rotation and is independent to the mixer diameter when the loading is maintained constant. The porosity was that obtained when the mixer is stopped after rotating for 5 minutes and is not the porosity obtained when the mixer is rotating. A maximum porosity is obtained at 33% of the critical speed. The concentration distribution for mixer M-2 at this speed is plotted in Fig.(5.14), from these curves the rate of particle movement R(t) and the variance $\sigma^2(t)$ can be obtained and are plotted in Fig.(5.11) and Fig.(5.12). Thus the rate of mixing can be compared.

5.4.1.3. DETERMINATION OF THE RATE DETERMINING PARAMETER D AND CONCLUSION

By considering only the first term of Eq.(5.23) and Eq.(5.24), hence

$$R(t) = \frac{8}{\pi^2} \exp\left(- \frac{\pi^2 Dt}{4 \ell^2}\right) \tag{5.23'}$$

$$\sigma^2(t) = \frac{2}{\pi^2} \exp\left(- \frac{\pi^2 Dt}{2 \ell^2}\right) \tag{5.24'}$$

Graphs of ln R(t) vs. time t and ln $\sigma^2(t)$ vs. time t should yield atraight lines. From the slope of each it is possible to determine the values of the determining parameter D hereafter called the migration coefficient. If the length of mixer is measured in centimeters and the mixing time in seconds, the dimension D can be expressed as cm^2/sec. The values of D obtained from the experimental runs are presented in Table 5.3.

TABLE 5.3.   VALUES OF MIGRATION COEFFICIENT   D

Run	Mixer	% of critical speed of rotation	D (cm/sec^2)	
			from R(t)	from $\sigma^2(t)$
R-SI-11	M-1	52	0.0261	0.0257
R-SI-12	M-1	52	0.0264	0.0260
R-SI-13	M-1	52	0.0259	0.0257
R-SlII-2	M-2	52	0.0256	0.0254
R-SV-1	M-4	52	0.0268	0.0272
R-ASI-1	M-2	33	0.0291	0.0295

It can be seen from the results in this table, the migration coefficient D is independent of size of the mixer provided the speed of rotation is maintained at a constant fraction of the critical speed. This finding is similar to that obtained by Müller in his investigation into drum mixers.

From the results presented in this section, the equation (Eq.4.14) can be confidently applied to the mixing of free flowing particles of the same size and the same density where the particles have an equal probability of undergoing displacements. Rate of mixing is found to depend on a migration coefficient which is a function of the speed of rotation, and particle characteristics but which is independent of mixer dimensions.

FIG. 5.7. AXIAL CONCENTRATION DISTRIBUTION OF S-1 AT 52% C. S.R.
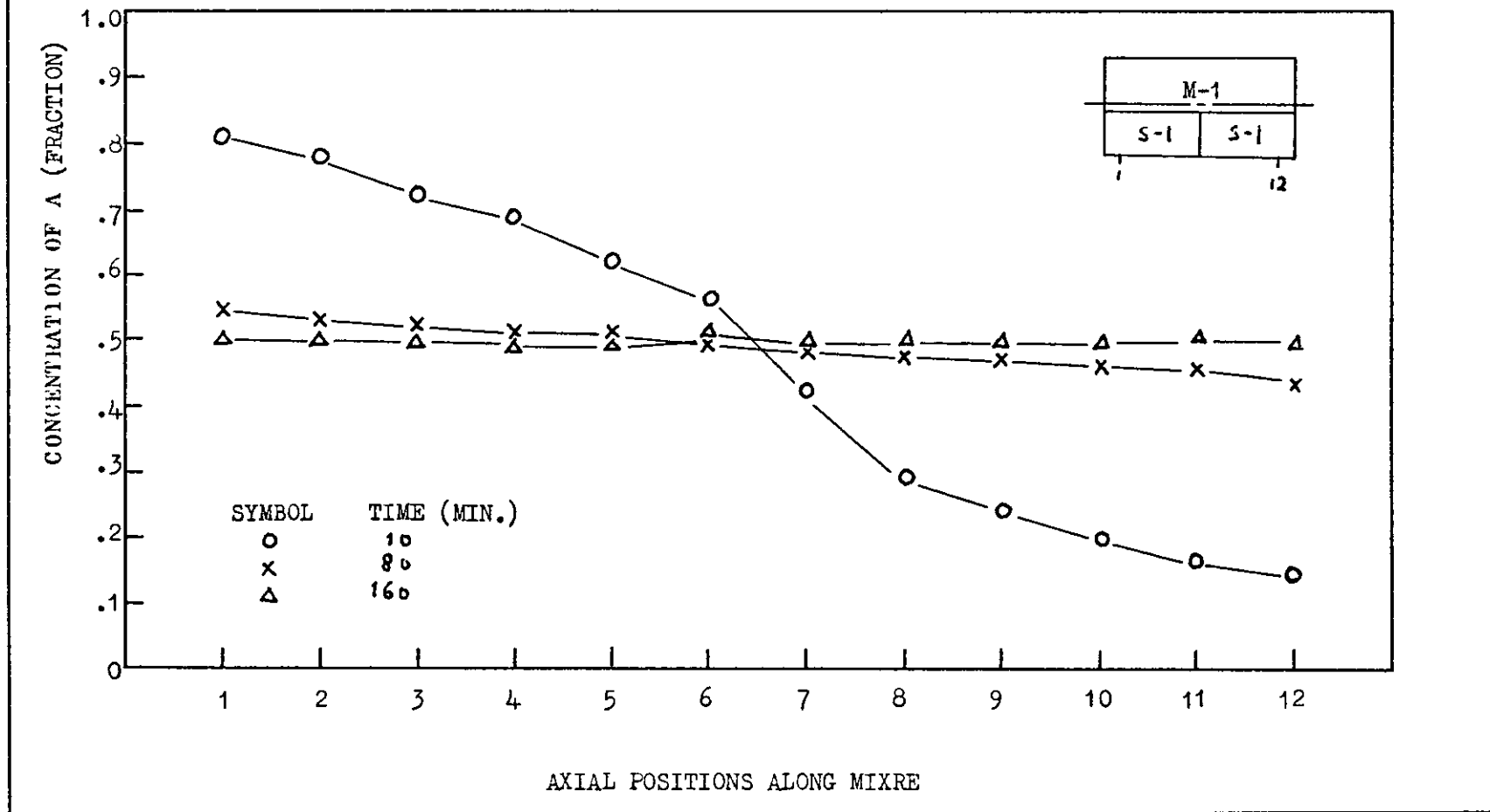
FIG. 5.8. AXIAL CONCENTRATION DISTRIBUTION OF S-1 AT 52% C. S. R.
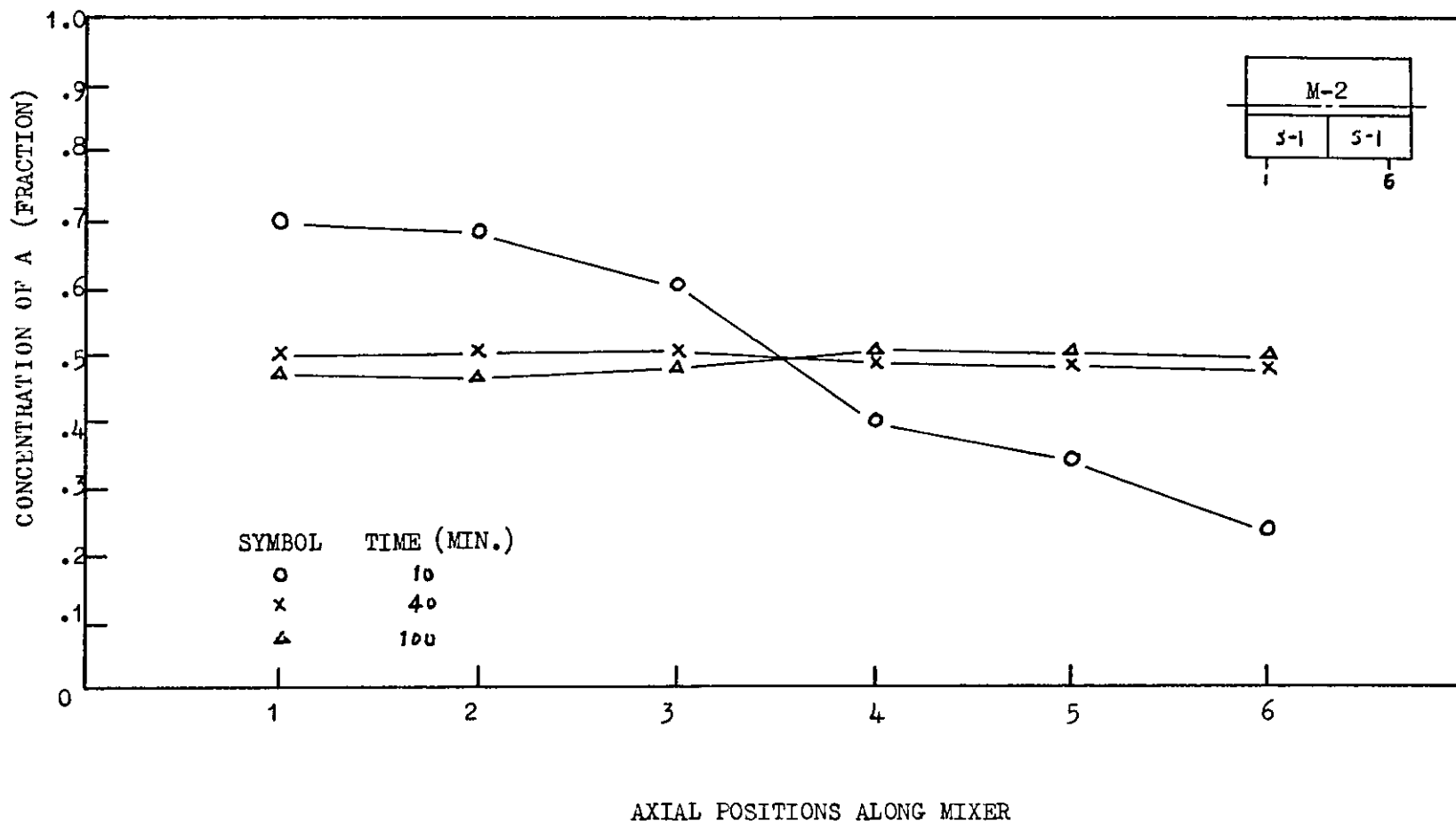
FIG. 5.9. AXIAL CONCENTRATION DISTRIBUTION OF S-1 AT 52% C. S. R.
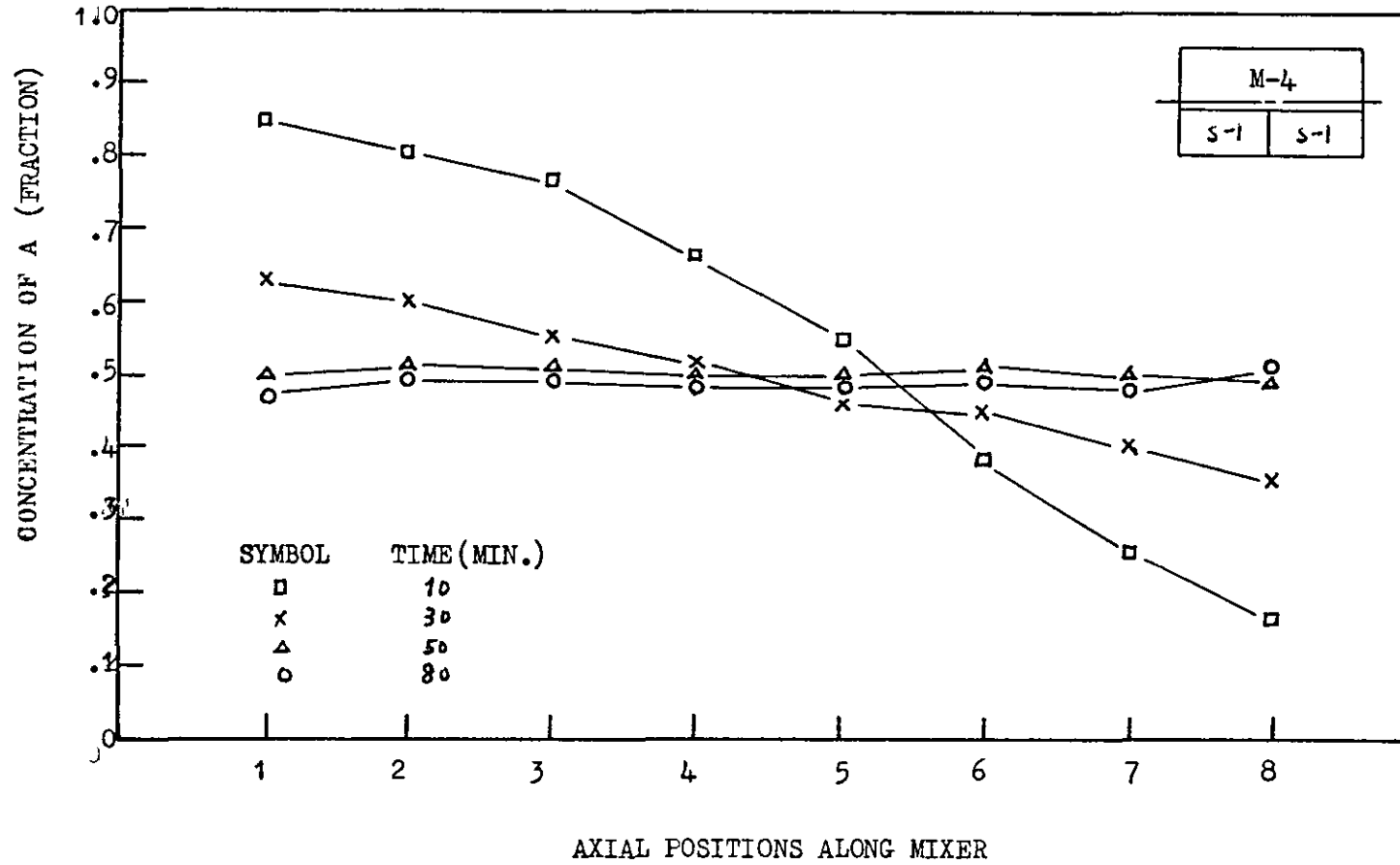
FIG. 5. 10 CONCENTRATION VARIATION S-1 AT 52% C. S. R.
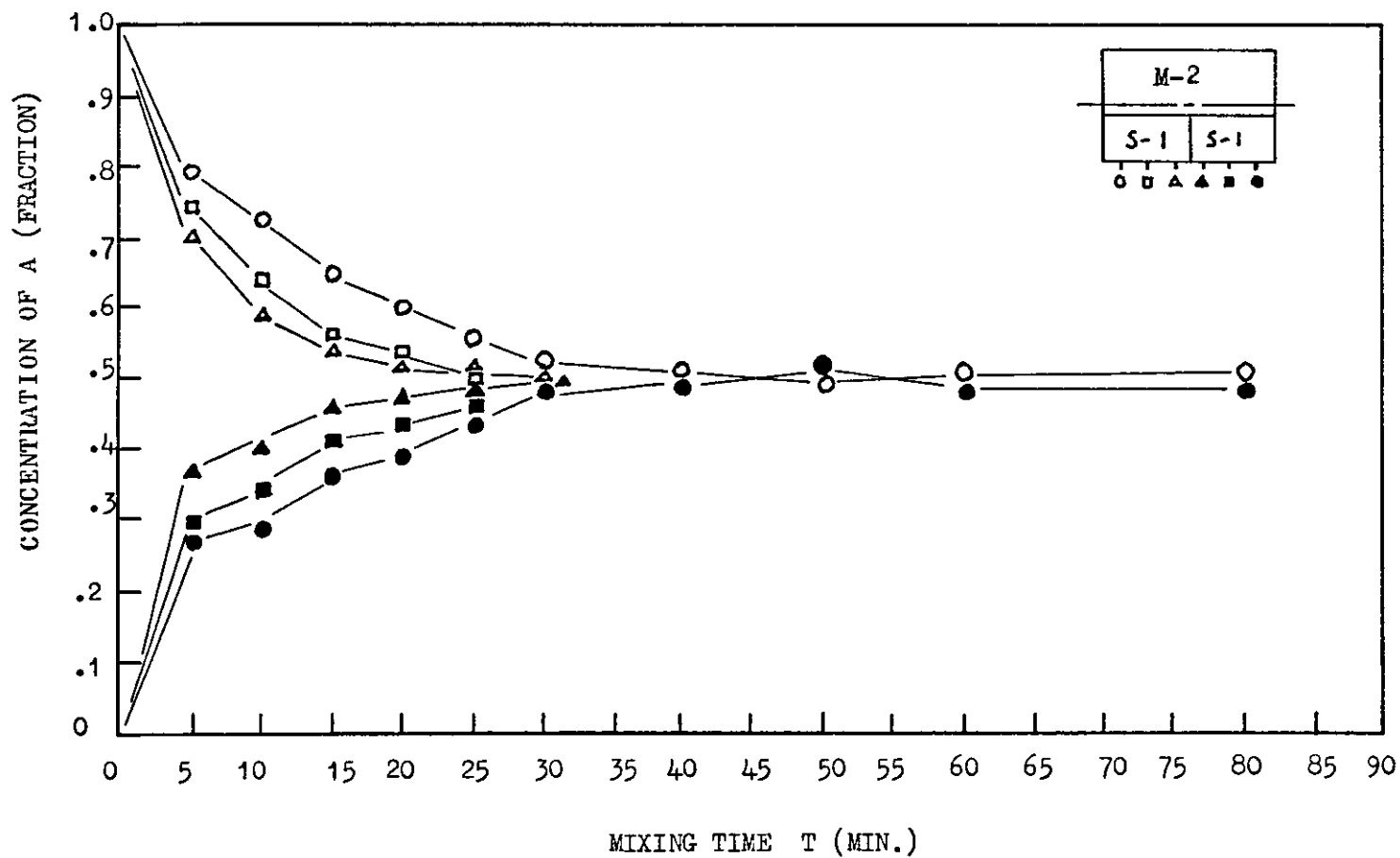
FIG. 5.11. THE RATE OF PARTICLE MOVEMENT R(t) VS. T

133
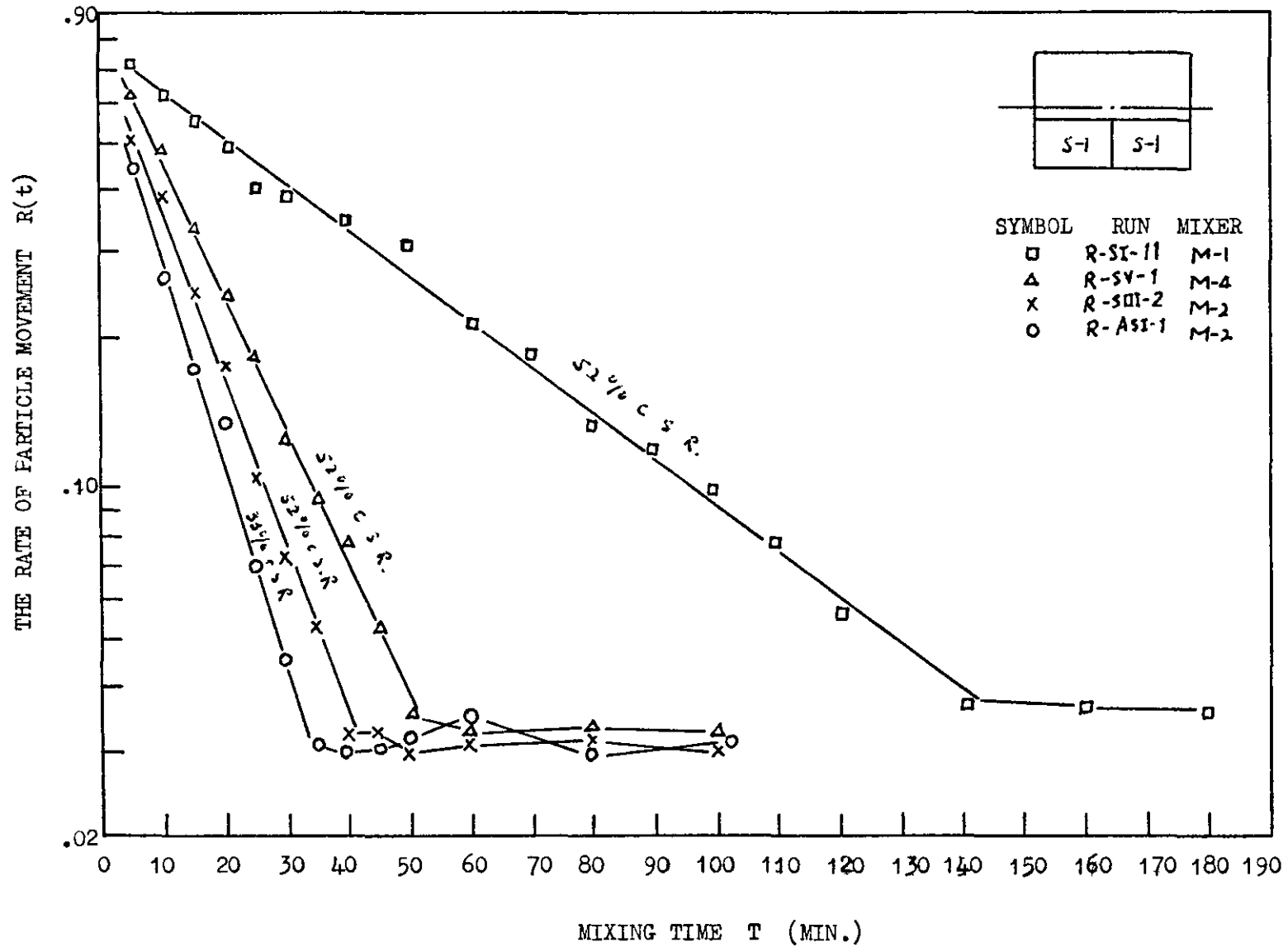
FIG. 5.12. SAMPLE VARIANCE $\sigma^2(t)$ VS. T
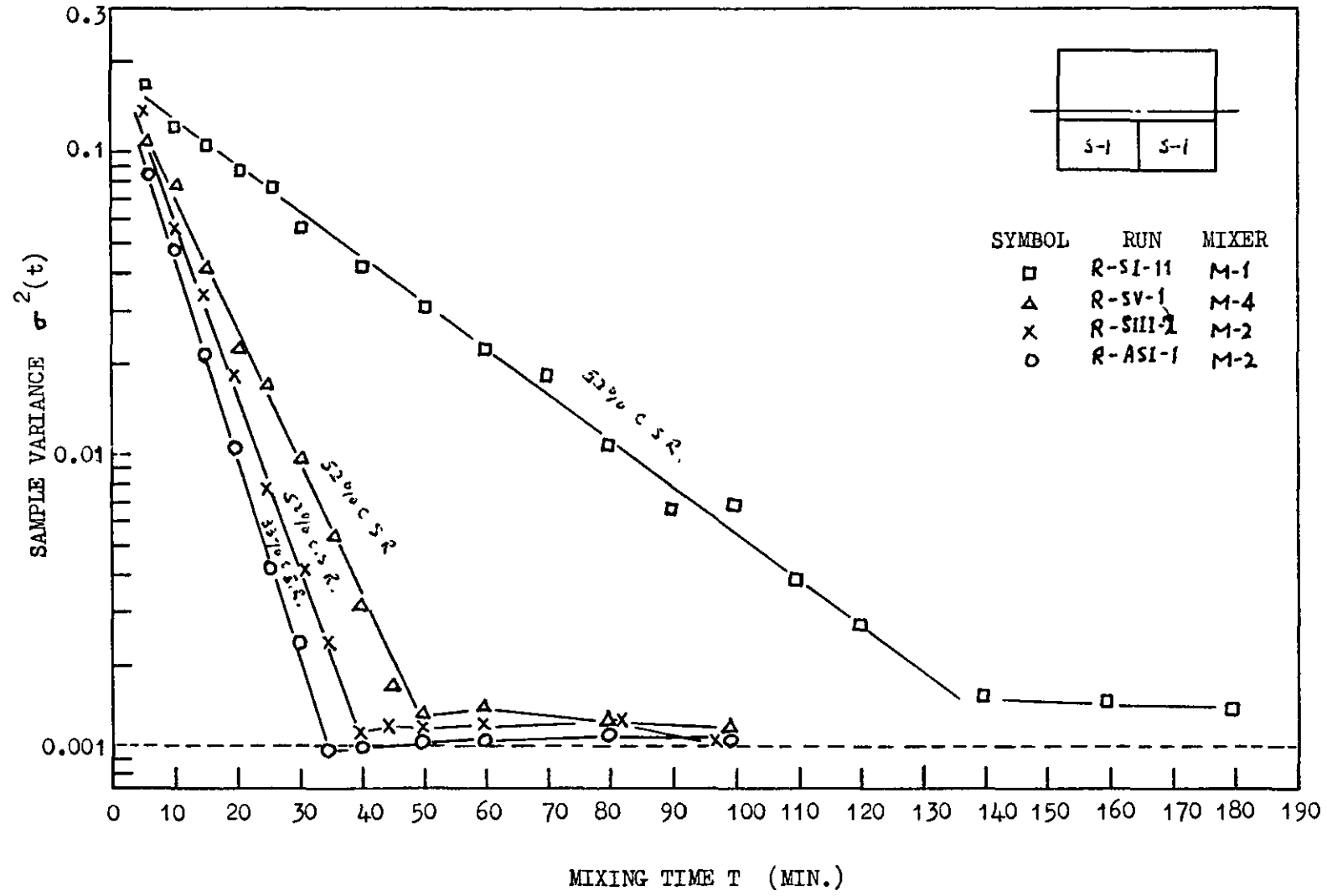
FIG. 5.13. POROSITIES OF SAND (%) VS. PERCENT OF CRITICAL SPEED OF ROTATION

PERCENT OF CRITICAL SPEED OF ROTATION
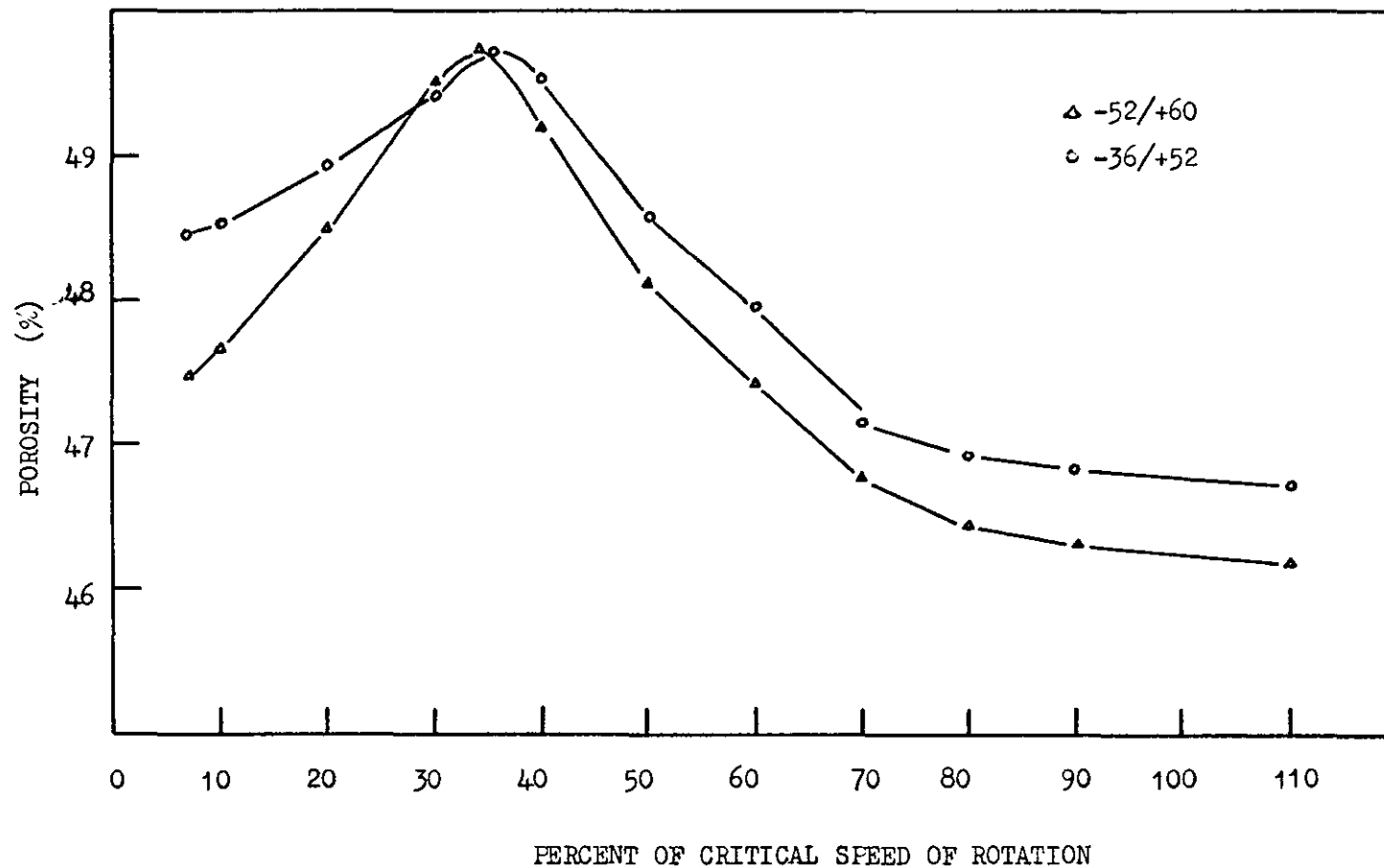
FIG. 5.14. AXIAL CONCENTRATION DISTRIBUTION OF S-1 AT 33% C. S. R.
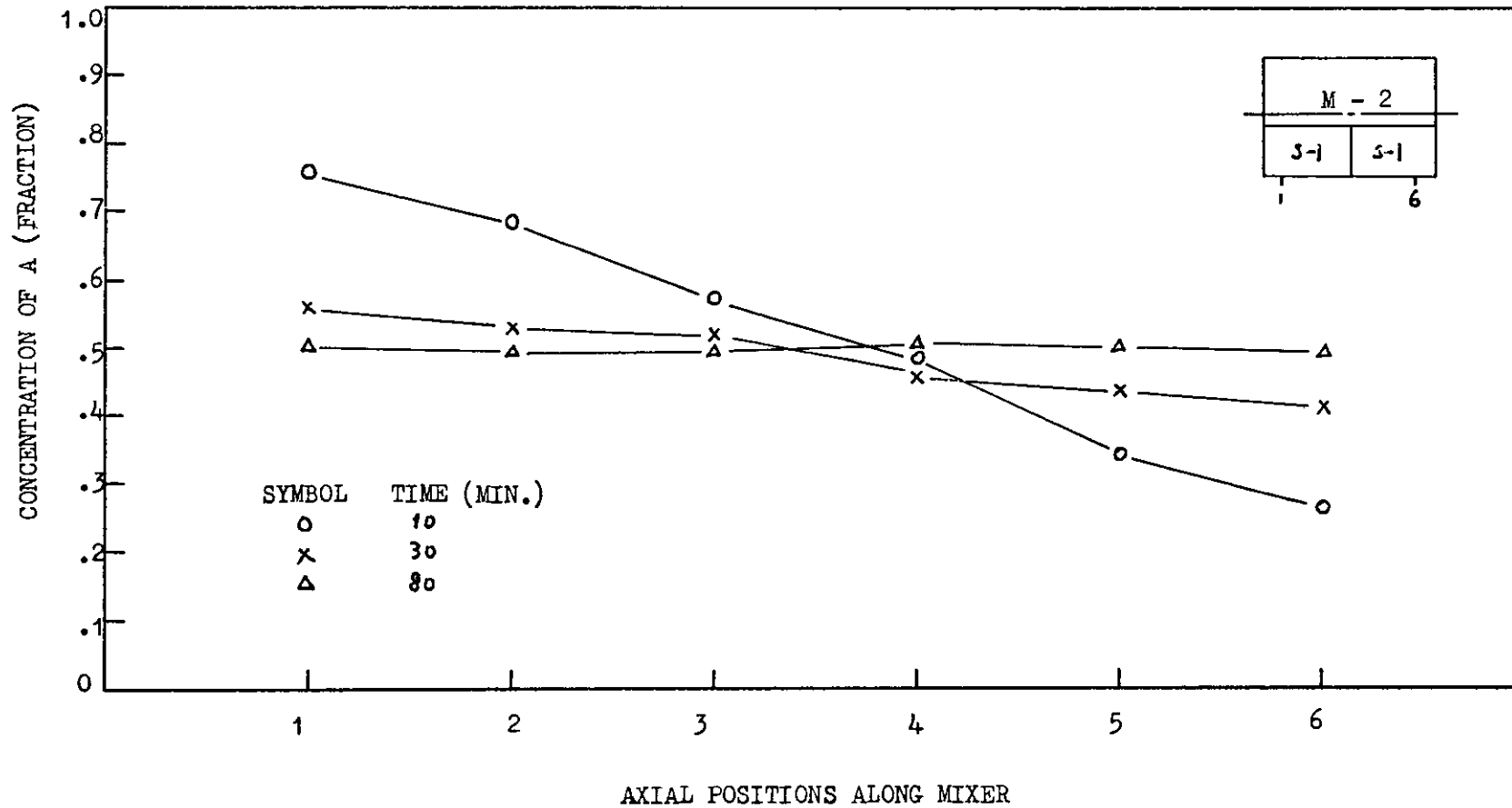
5.4.2. COMPONENTS WITH DIFFERENT SIZE DISTRIBUTION BUT SAME DENSITY

This series of experiments was carried out to investigate the mixing and segregation mechanism with components of differently sized Moneystone sands - S-1 and S-2 in four differently dimensioned mixers.

5.4.2.1. COMPARISON OF THE STATES OF MIXEDNESS IN FOUR MIXERS AT THE SAME SPEED OF ROTATION

The concentration distribution curves of the larger particles (S-1) are plotted in Fig.(5.16) to Fig.(5.19) for mixers M-1, M-2, M-3 and M-4 at 20% of the corresponding critical speed of rotation. The general shape of those curves are similar. After a very short initial period, the profiles of the distribution curves show a separation of large and small particles and indicate segregation of large particles in the middle of the mixers. Fig.(5.20) shows the variation of concentration with time of large particles at fixed positions. These curves indicate the separation of large and small particles at the specified location in terms of time. The sample variance $\sigma^2(t)$ is plotted as a function of mixing time for all mixers in Fig.(5.21). The shape of those curves are generally similar an initial transient period, a very short mixing stage and a region in which separation of large and small particles occurs. After this stage, an equilibrium state is established.

5.4.2.2. THE STATES OF MIXEDNESS AS A FUNCTION OF SPEED OF ROTATION

The speed of rotation of mixer can affect the state of mixedness. Further experiments were performed into differently dimensioned mixers at 35% and 52% of the corresponding critical speed of rotation. The concentration distribution curves for these conditions are plotted in Fig.(5.22) to Fig.(5.25) and Fig.(5.26) to Fig.(5.28). The shape of these sets of curves are distinctively different. For the case of a 35% of the corresponding critical speed of rotation, apart from the initial period the profiles of the concentration distribution showed a linear progression. But in the case of 52% of the corresponding critical speed of rotation, the profiles show the appearance of small particles in the middle of the mixer. The

sample variance $\sigma^2(t)$ can be calculated for both sets of curves and plotted as function of mixing time in Fig.(5.29) and Fig.(5.30) for all mixers. From those sets of curves, the curves in Fig.(5.29) showed the three-stage mixing process, as described in the last section. The increase in $\sigma^2(t)$ after the minimum shows when segregation occurred. The concentration with time profiles for both cases are illustrated in Fig.(5.31) and Fig.(5.32) for mixer M-2.

5.4.2.3. DISCUSSION OF THE RESULTS, FURTHER EXPERIMENTS ON A SINGLE WIDER SIZE DISTRIBUTION MATERIAL ROTATING IN THE MIXER, AND CONCLUSION

It is found from the experimental results that the separation of large and small particles is a tendency which was controlled by the speed of rotation. For a given speed of rotation, the patterns of the final mixture (equilibrium) seem to be definite and independent of the size of mixer. The experimental results can be summarised and presented in Table 5.4.

TABLE 5.4.

EQUILIBRIUM CONCENTRATION OF LARGE PARTICLES

AT THE CENTRE OF MIXER WITH SPEED OF ROTATION

Type of Mixer	L/d	Equilibrium concentration of large particles at the centre of the mixer with fraction of critical speed of rotation			size ratio	density ratio
		0.20	0.35	0.52		
M-1	2	0.70	0.51	0.32	1.27	1
M-2	1	0.72	0.49	0.32	1.27	1
M-3	3	0.68	0.50	---	1.27	1
M-4	1	0.73	0.51	0.36	1.27	1
Ave.	--	0.71	0.50	0.33	--	-

The different concentration distribution profiles obtained at three different speed of rotation may be explained as follows.

At the lower speeds of rotation, consider the momentum

transfer on a collision between a large particle and a small particle. The small particle will tend to obtain a greater velocity than the large particle. Thus the central region of the mixer will be characterised by a deficiency in small particles, or an increase in the concentration of large particles. The concentration distribution will be a result of the dynamic equilibrium. Individual particles will be moving continually but the net concentration profile will remain sensibly constant. As the speed of rotation is increased, the momentum of the particles is also increased and a pseudo homogeneity is formed because all the particles are changing their axial position rapidly. At even higher rotational speeds, the large particles receive more energy than the samll particles at the end walls of the mixer. This is transferred to the small particles by momentum exchange and the small particles migrate to the centre of the mixer. So that now the centre is dificient in large particles.

The stability of packing as has been shown to occur when particles have the maximum number of contact points (22). This will occur when particles of the same size congregate together. Thus the segregating tendencies described above as a result of momentum transfer are enhanced by the stabilising of the particulate system when particles of the same size are together.

The segregation of particles into separate bands appears thus to be an intrinsic property of a bimodal distribution with a size ratio between the means of each part greater than 1.27. Using drums with a large length to diameter ratio Roseman (69) was able to obtain many bands.(See Table 5.5)

Two further points are of interest. Table 5.4. indicates that the concentration of large to small particles depends solely on the speed of rotation of the mixer and is independent of mixer size if the speed of mixer is presented as a proportion of the critical speed. In another experiment all the material in the segregated bands was removed by a suction device after mixing had proceeded for 680 minutes. Each band was then size analysed. The results are presented in Fig.(5.33). It is seen that the size distributions of the large particles; as analysed on a microscope, are distinctly different in each region.

140

TABLE 5.5. RESULTS OBTAINED BY ROSEMAN

Mixer length inches	large particles	small particles	No. of large particle band	No. of small particle band
2 - 4	glass beads	glass beads	2	1
5	" "	" "	3	2
7	" "	" "	3	2
10	" "	" "	4	3
14	" "	" "	5	4
$17\frac{1}{2}$	" "	" "	6	5
2	" "	salt	2	1
4	" "	"	3	2
8	" "	"	4	3
14	" "	"	6	5
$17\frac{1}{2}$	" "	"	7	6
8	" "	sand	4	3
3	sand	"	3	2
8	"	"	4	3

More results have also been obtained with bimodal distributions a further experiment with a wide size distribution was 'mixed! The sand covered the range -30/+60 and individual sieve cuts were dyed red, black and natural. After mixing for 180 minutes, 5 bands were observed. Each band was size analysed and the results shown in Fig.(5.34). This adds to the conclusions above that both the dynamic (momentum exchange) and the static (packing) of the particles lead to segregation - independently of the size distribution of the powder.

The importance of particle packing in the mixing of free flowing powders can be further seen from the results obtained at 35% of the corresponding critical speed of rotation. At this speed, both components have a maximum porosity as shown in Fig.(5.13). Since each mixture has a loosest form of particle packing. The

form is unstable and any external force will disturb the system and segregation is less likely to occur.

The stability of a mixture may be explained by a thermodynamic aspect. A stable system in which the entropy is a maximum. Particle movement in the rotating drum mixer might be considered as a single motion, the mechanical drive is providing rotational and such a process might be considered as a natural process in the thermodynamics. The powder mixing system deals with finite sized particles in a dynamic situation. Such system could be considered as a thermodynamically unstable since the entropy of system is always increasing.

The equation of principle of increasing entropy could be expressed as

$$f(dS) \geqq 0$$

The fact that entropy tending to a maximum is a trend towards the equilibrium of a natural process can be seen from the mechanical analogy in Fig.(5.35)
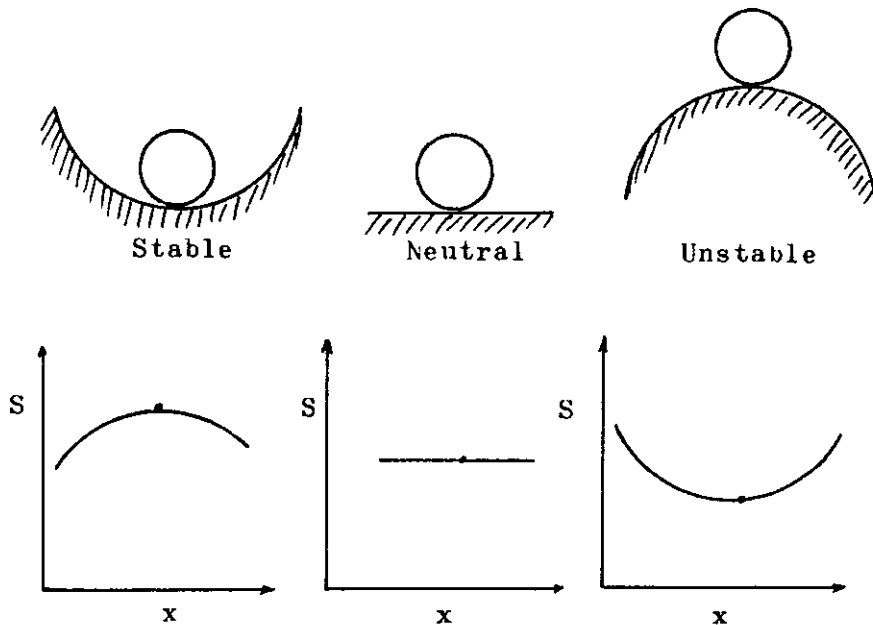
Fig.(5.35) Stability of Equilibrium

An equilibrium state of a system could be characterised as

Stable equilibrium	$d^2S < 0$	$S = S_{max}$
Neutral equilibrium	$d^2S = 0$	
Unstable equilibrium	$d^2S > 0$	$S = S_{min}$

To extend the analogy the relationships of the thermodynamic parameters of the particulate mixing system might be considered as

$$f(dS) - g(dU) - h(dV) > 0$$

where $f(dS)$, $g(dU)$ and $h(dV)$ are the function of the changes of entropy, internal energy and volume of the system.

For the maximum value of entropy the changes of internal energy and volume must be minimum. In the case of a particulate powder mixing system, the change of internal energy in the system may be less significant. Thus

$$g(dU) = 0$$

but the volume change (apparent volume) must be important whenever a mixing process is concerned as individual particles are actually occupying units of volume in the system. Hence the condition for stable equilibrium will depend on

$$f(dS) - h(dV) > 0$$

For the mixing of the same particle size components, as described in Chapter 4 (postulate 1), thus changing of places of particles within the system will not cause any volume change. Thus in this case

$$h(dV) = 0$$

so that the entropy is maximum. It is confirmed that mixing with components of same particle size is always in a stable system, so that the mixing process could be considered by a stochastic process.

However, for the mixing with components of different particle size, the position exchange of particles will cause a volume change, and the entropy of the system will not be a maximum. For achieving a stable equilibrium system, therefore all the similar sized particles must congregate together, so that the position exchange of particles within the segregated regions will not affect the volume. Then the entropy of the system will become maximum.

A good mixture may be obtained when the porosity of the material is a maximum. This mixture may be described as in a pseudo-equilibrium system. Such a system can be considered by the mechanical

analogy as shown in Fig.(5.36)
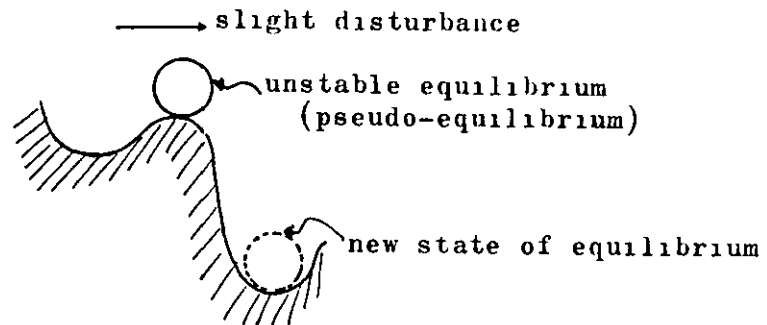
Fig.(5.36) Change from one equilibrium to another

Thus although a good mixture may be obtained which consists of different sizes, segregation will reappear during handling and transportation.

The explanations stated here, by considering the thermodynamic aspect, is only a brief thought on 'why' segregation must occur during mixing with components of different size. This rough idea might be worthy of further investigation.

Although a considerable amount of work has been done for mixing with components of different size, it is felt that at the present stage a quantitative rate mechanism is still cannot be developed. It can be concluded, from the experimental results that the states of mixedness can be predicted under certain circumstance. Segregation will usually occur but a pseudo-randomised mixture may be obtained by rotating the mixer at the speed at which the porosity of each component is a maximum.

FIG. 5.16. AXIAL CONCENTRATION DISTRIBUTION OF S-I AT 20% C. S. R.
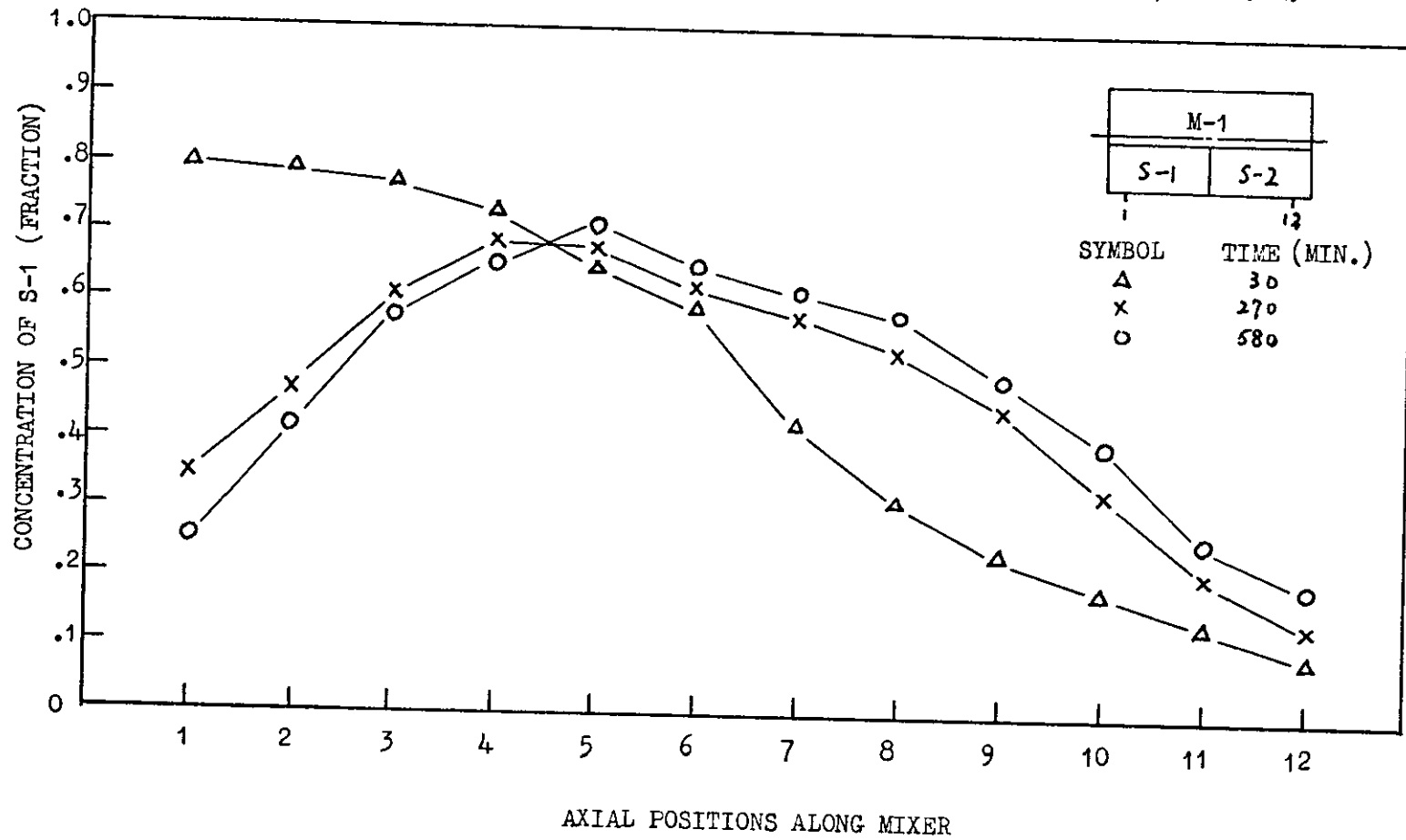
FIG. 5.17. AXIAL COMPOSITION DISTRIBUTION OF S-1 AT 20% C. S. R.
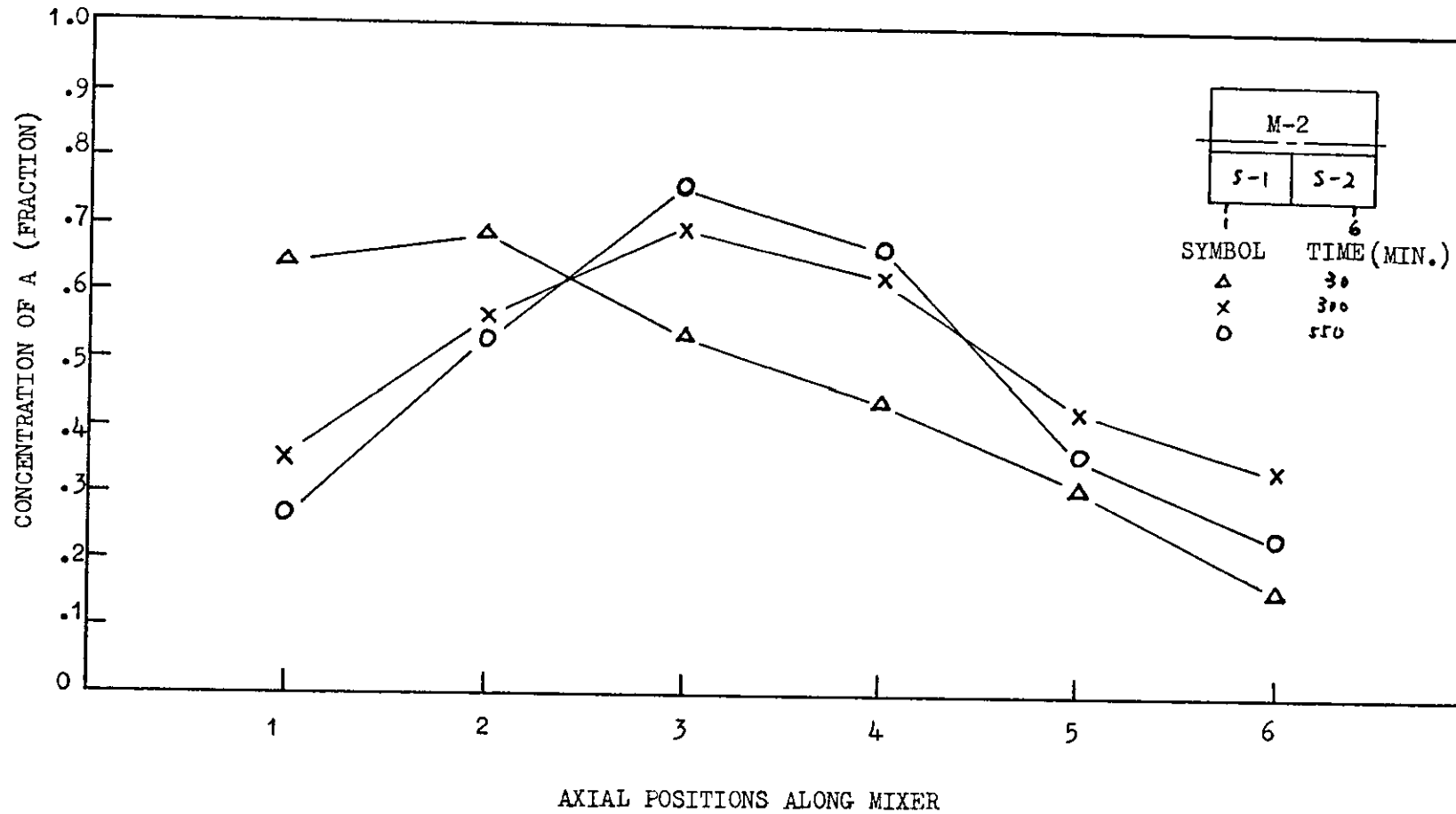
FIG. 5.18. AXIAL CONCENTRATION DISTRIBUTION OF 3-1 AT 20% C.S.R.
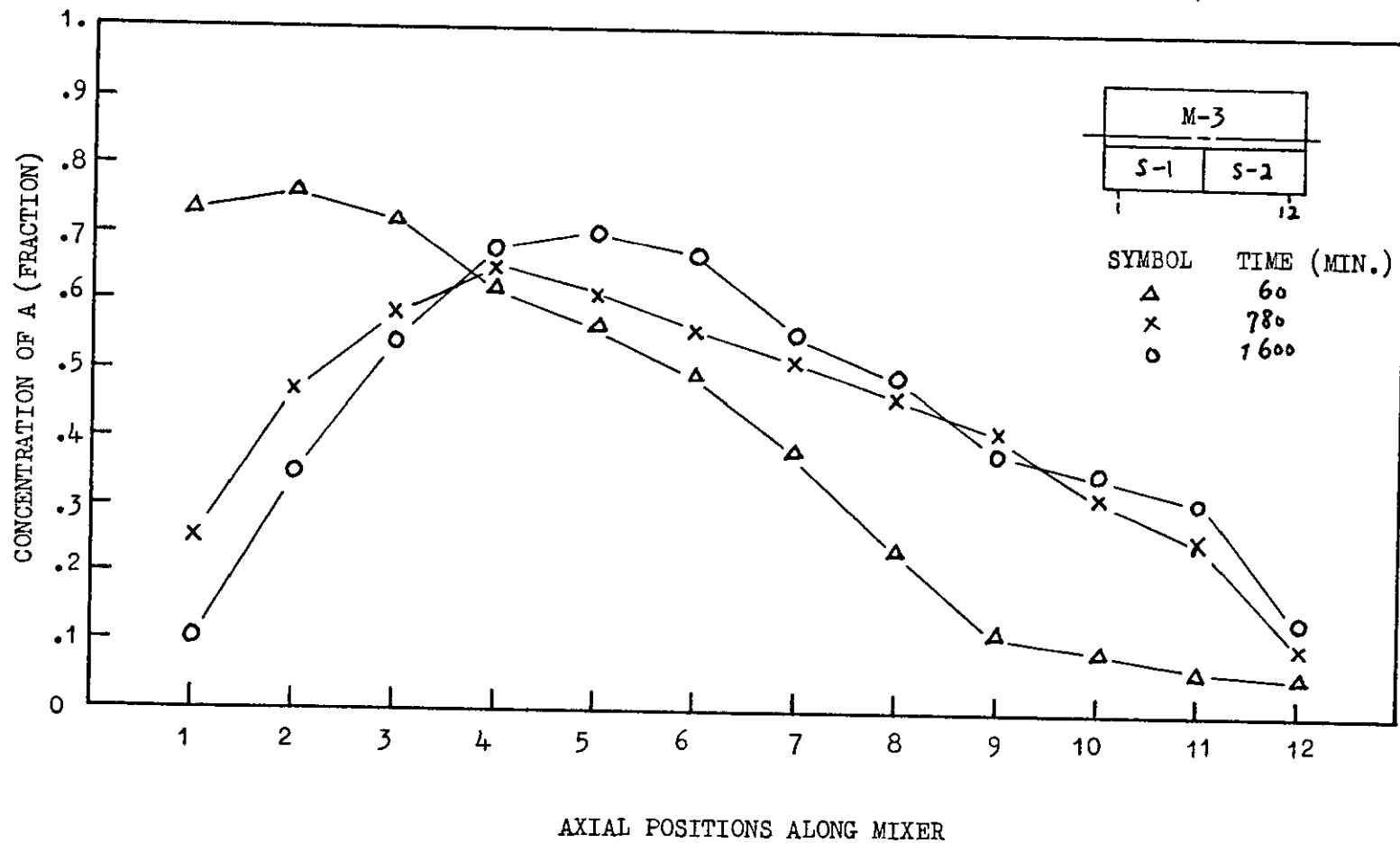
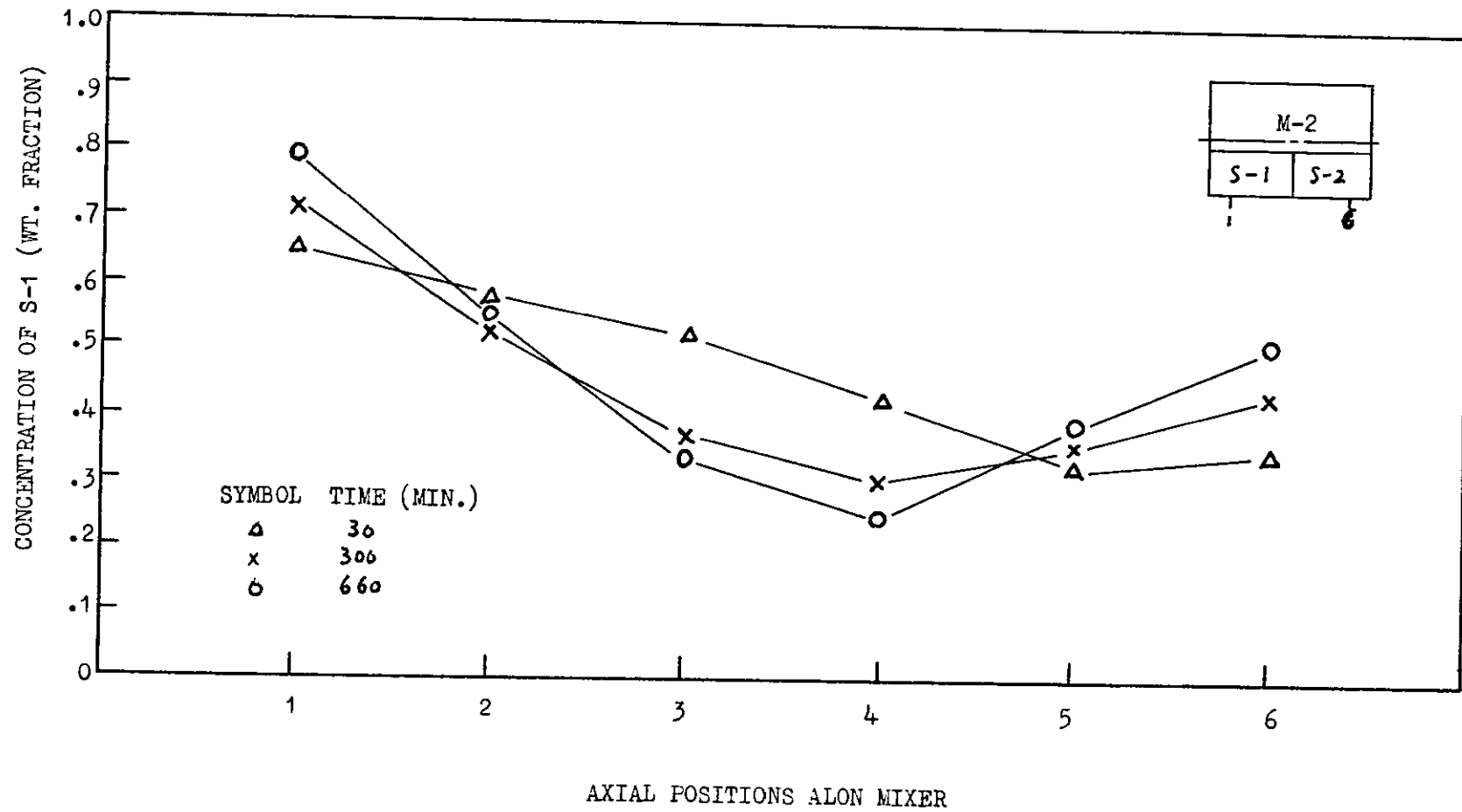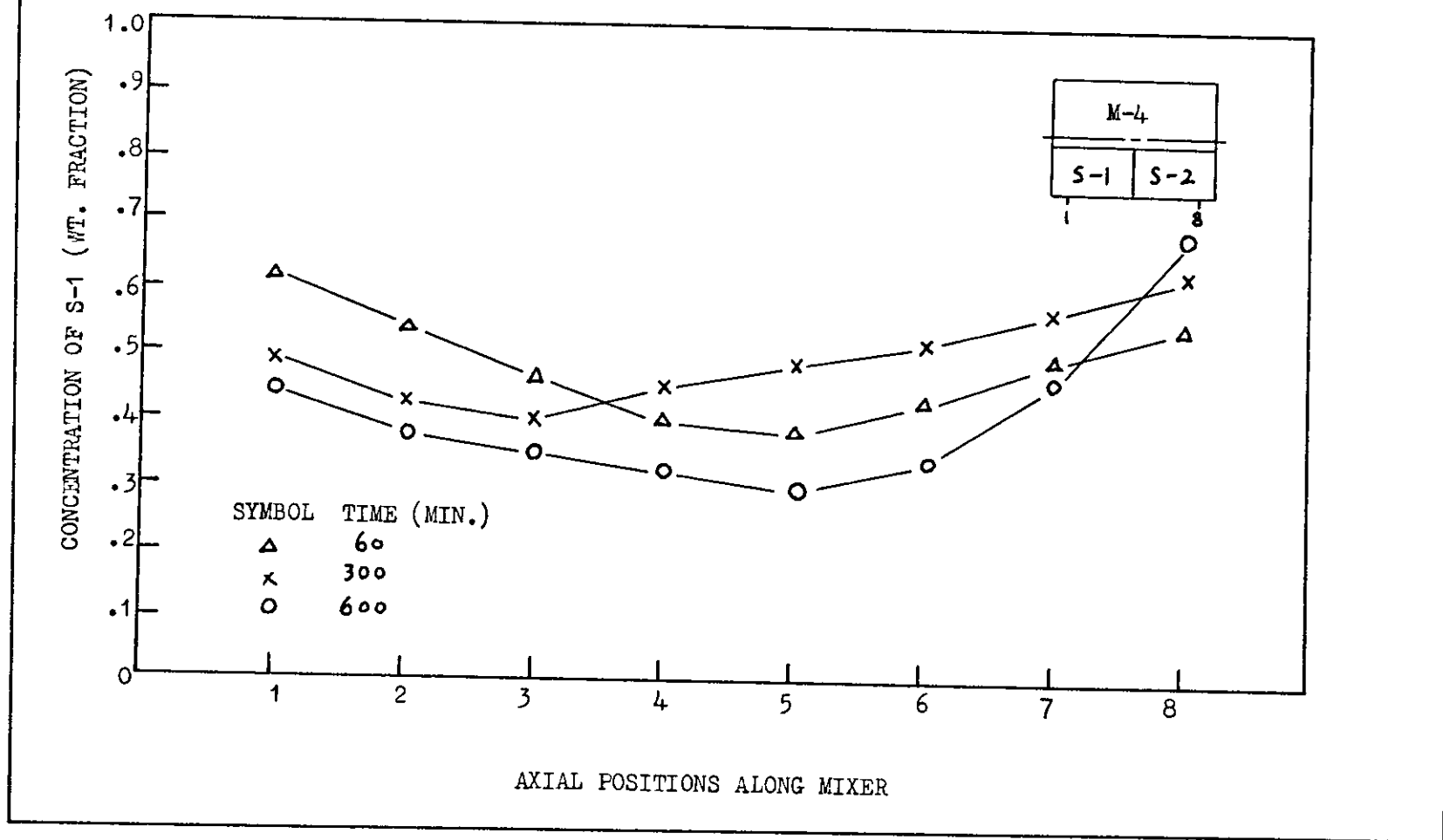FIG. 5.19. AXIAL CONCENTRATION DISTRIBUTION OF S-1 AT 20% C.S.R.
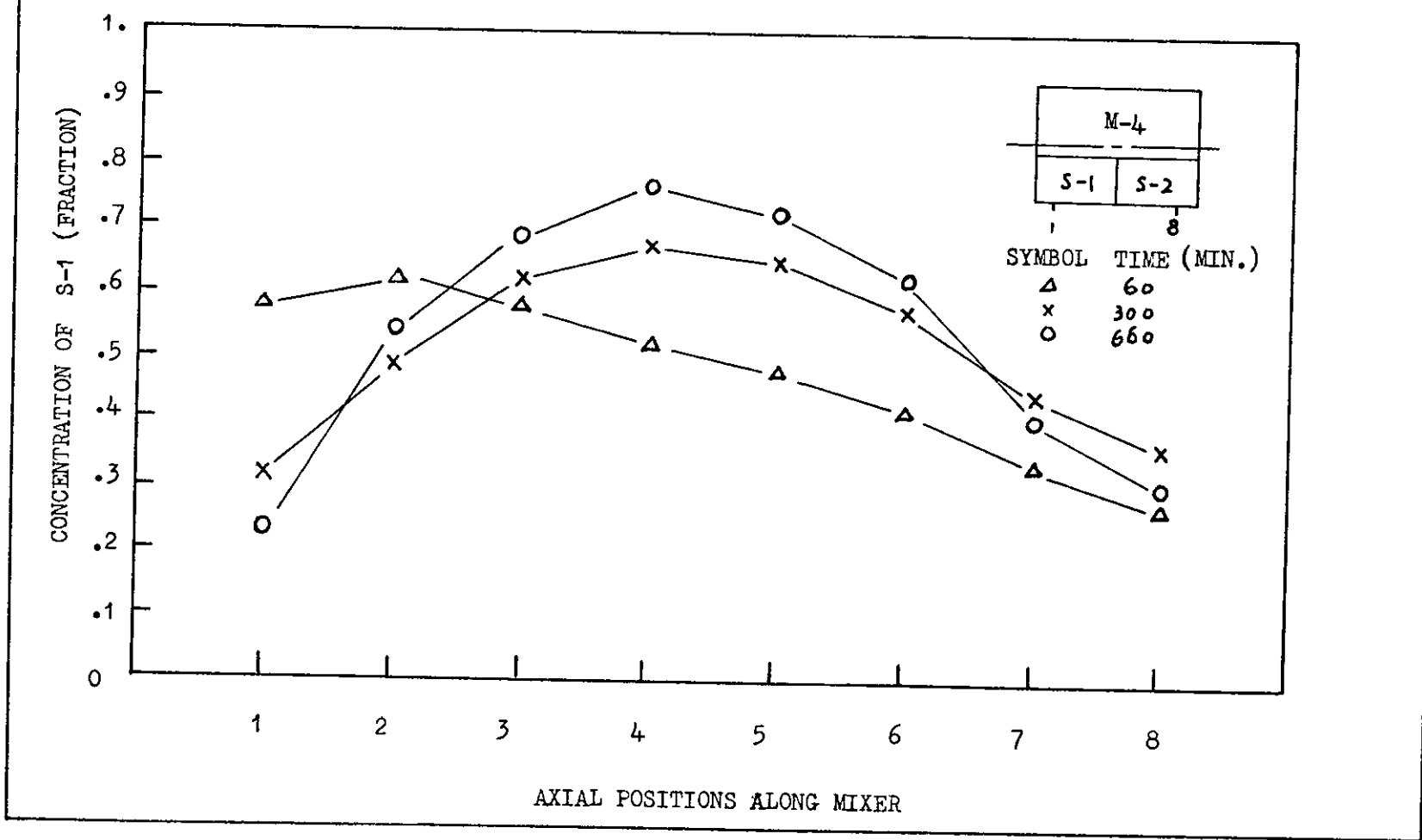
FIG. 5.20. CONCENTRATION VARIATION S-1 AT 20% C. S. R.
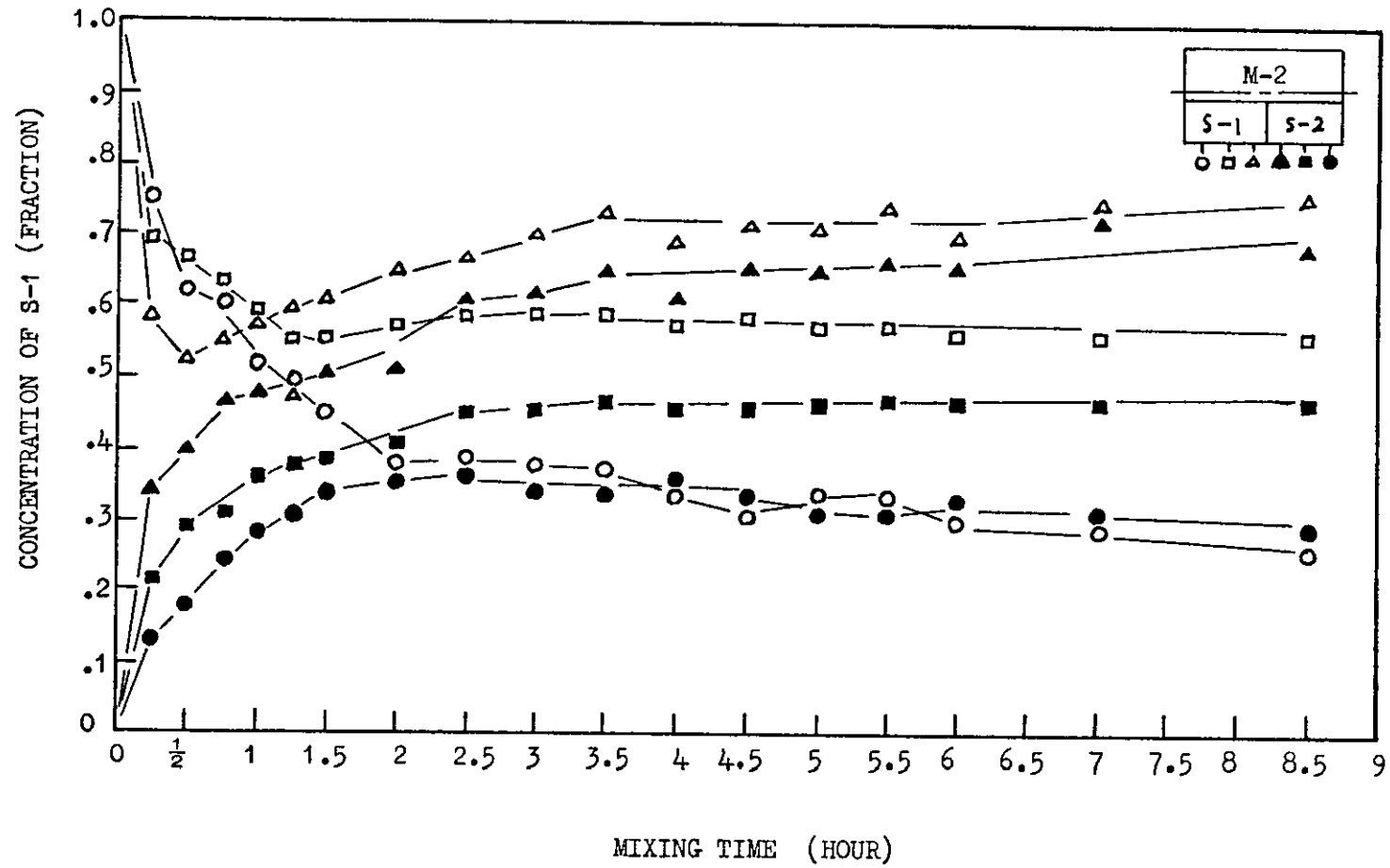
FIG. 5.21. SAMPLE VARIANCE $\sigma^2(t)$ VS. MIXING TIME
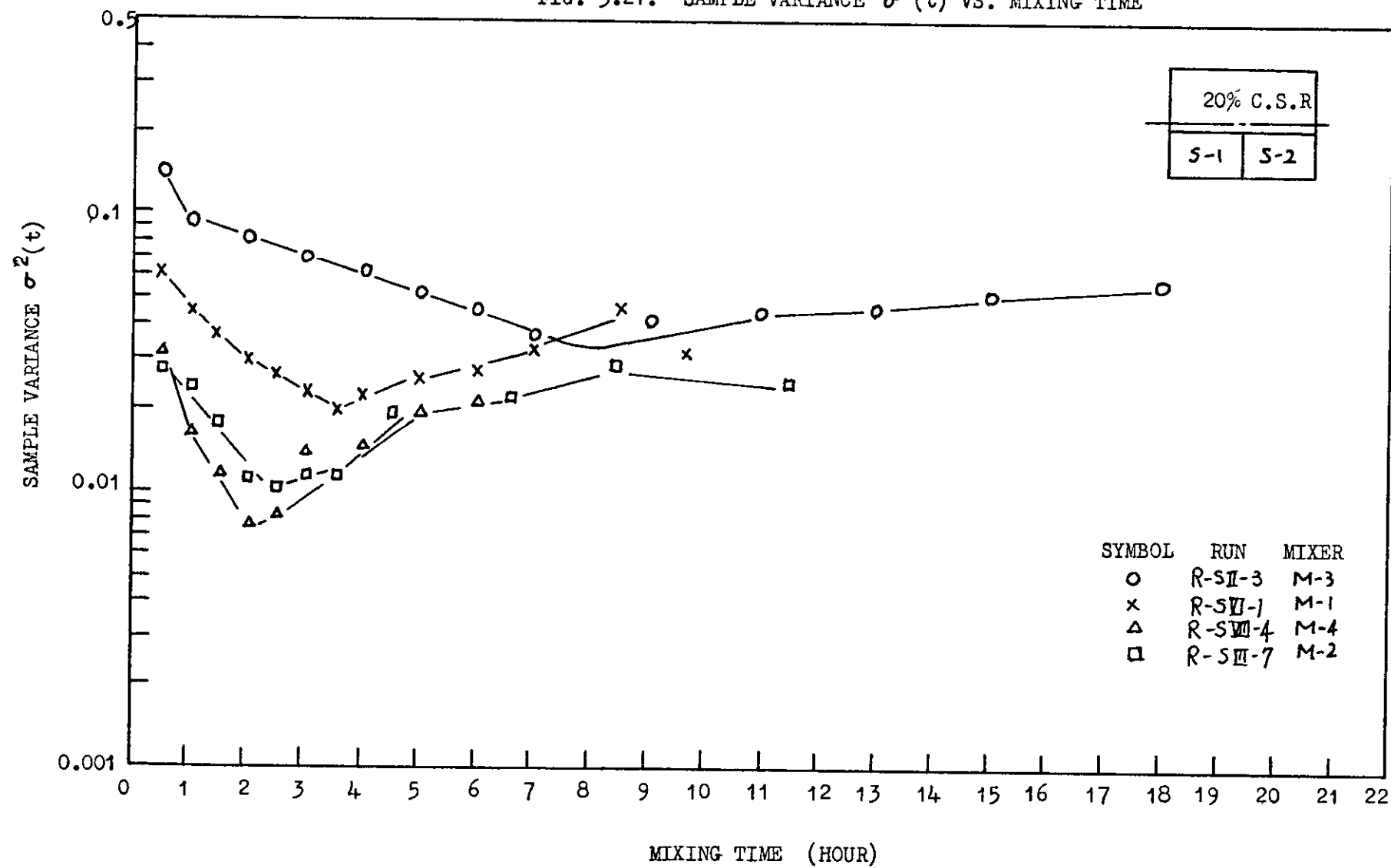
FIG.5.22. CONCENTRATION DISTRIBUTION OF S-1 AT 35% C. S. R.
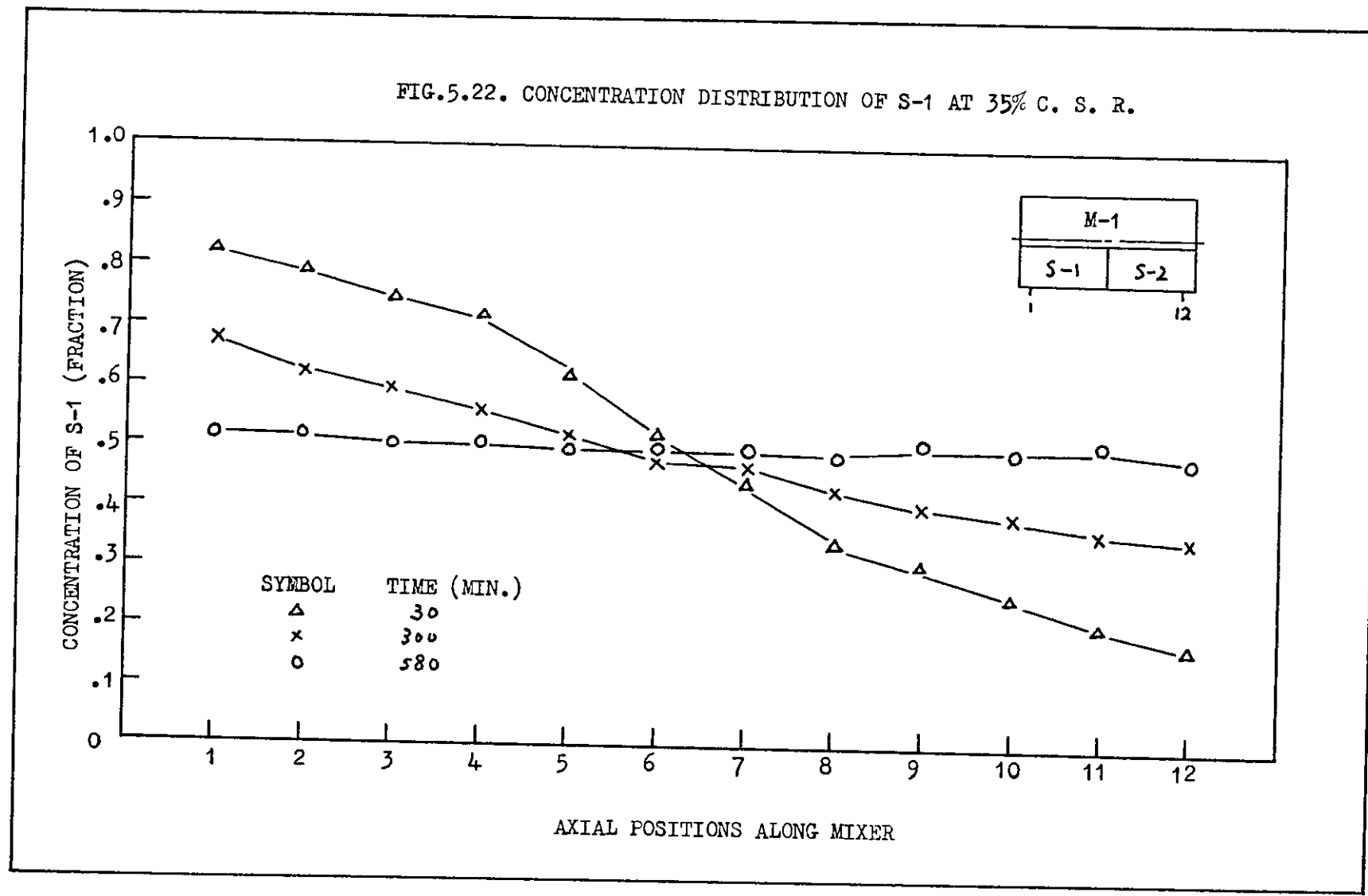
FIG. 5.23. AXIAL CONCENTRATION DISTRIBUTION OF S-1 AT 35% C.S.R.
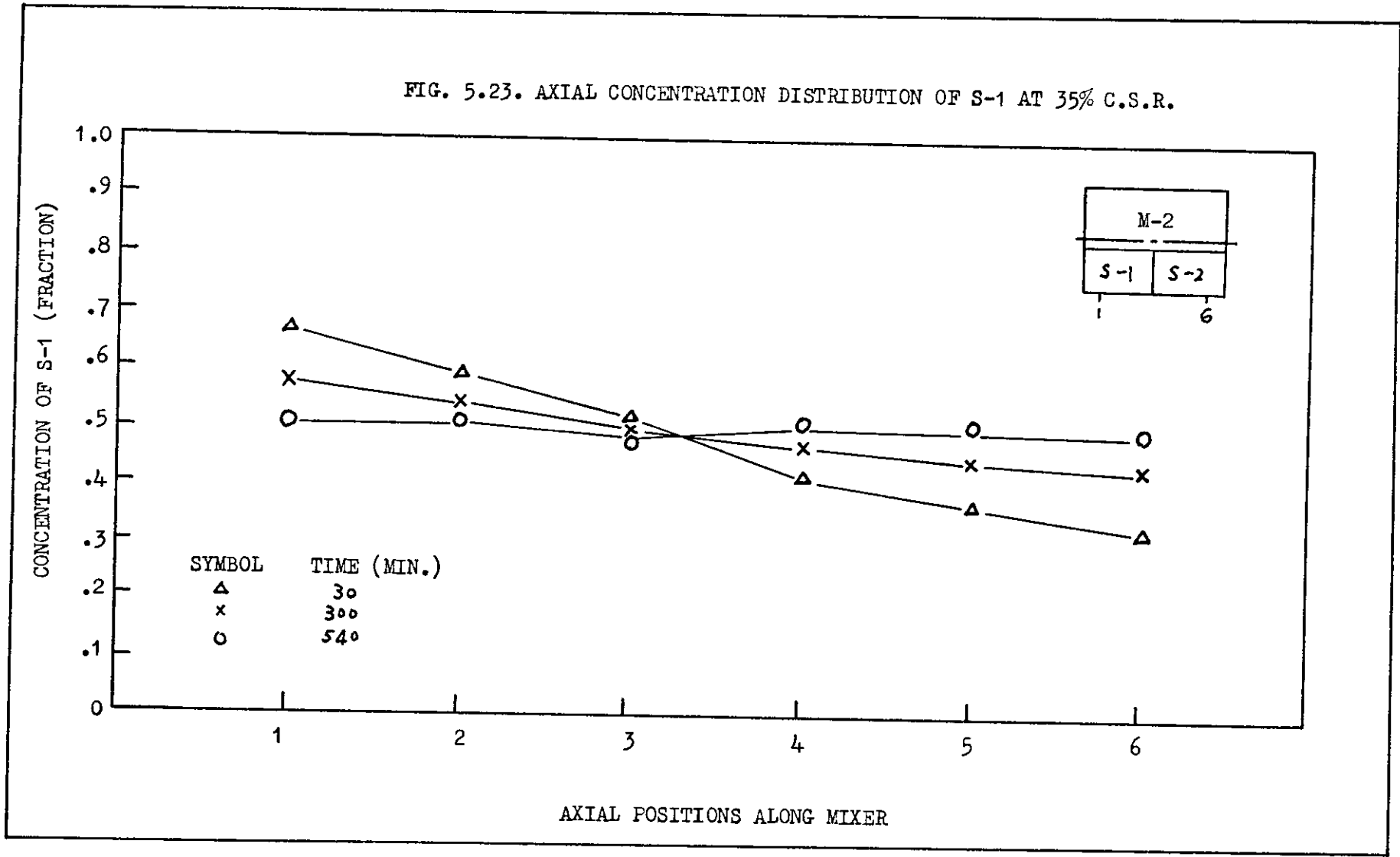
FIG. 5.24. AXIAL CONCENTRATION DISTRIBUTION OF S-1 AT 35% C.S.R.
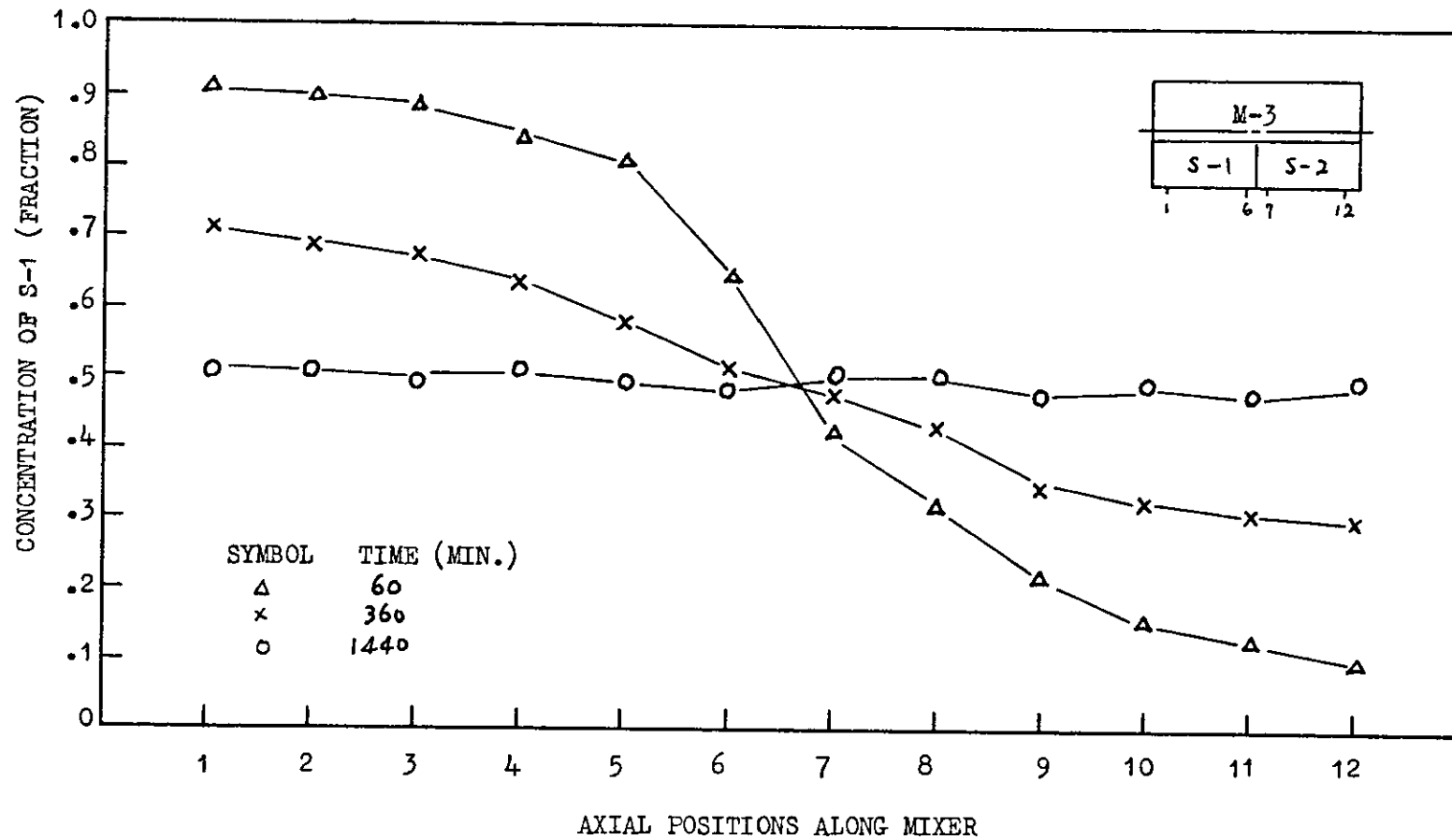
FIG. 5.25. AXIAL CONCENTRATION DISTRIBUTION OF S-1 AT 35% C.S.R.
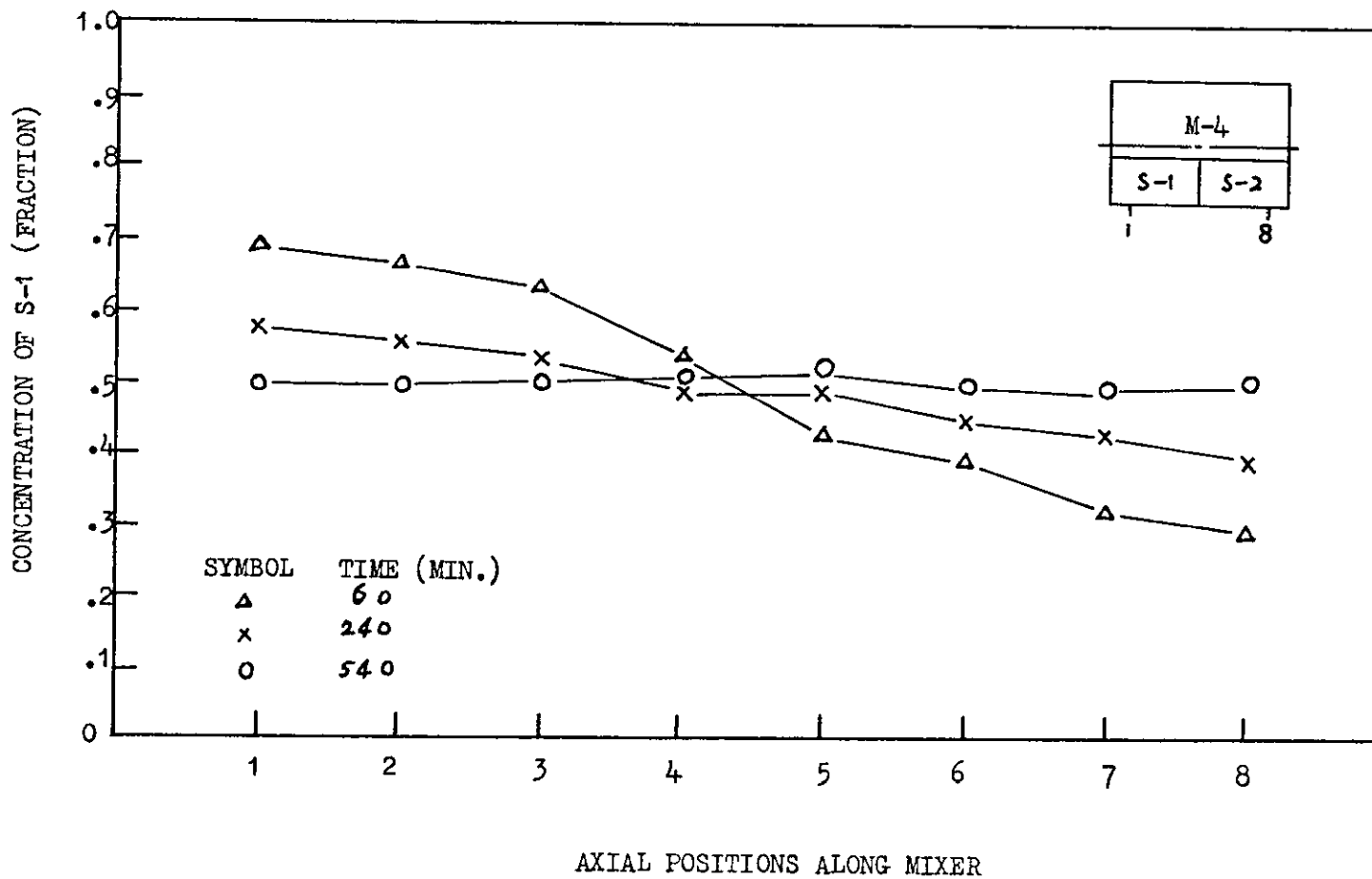
FIG. 5. 26. AXIAL CONCENTRATION DISTRIBUTION OF S-1 AT 52% C.S.R.
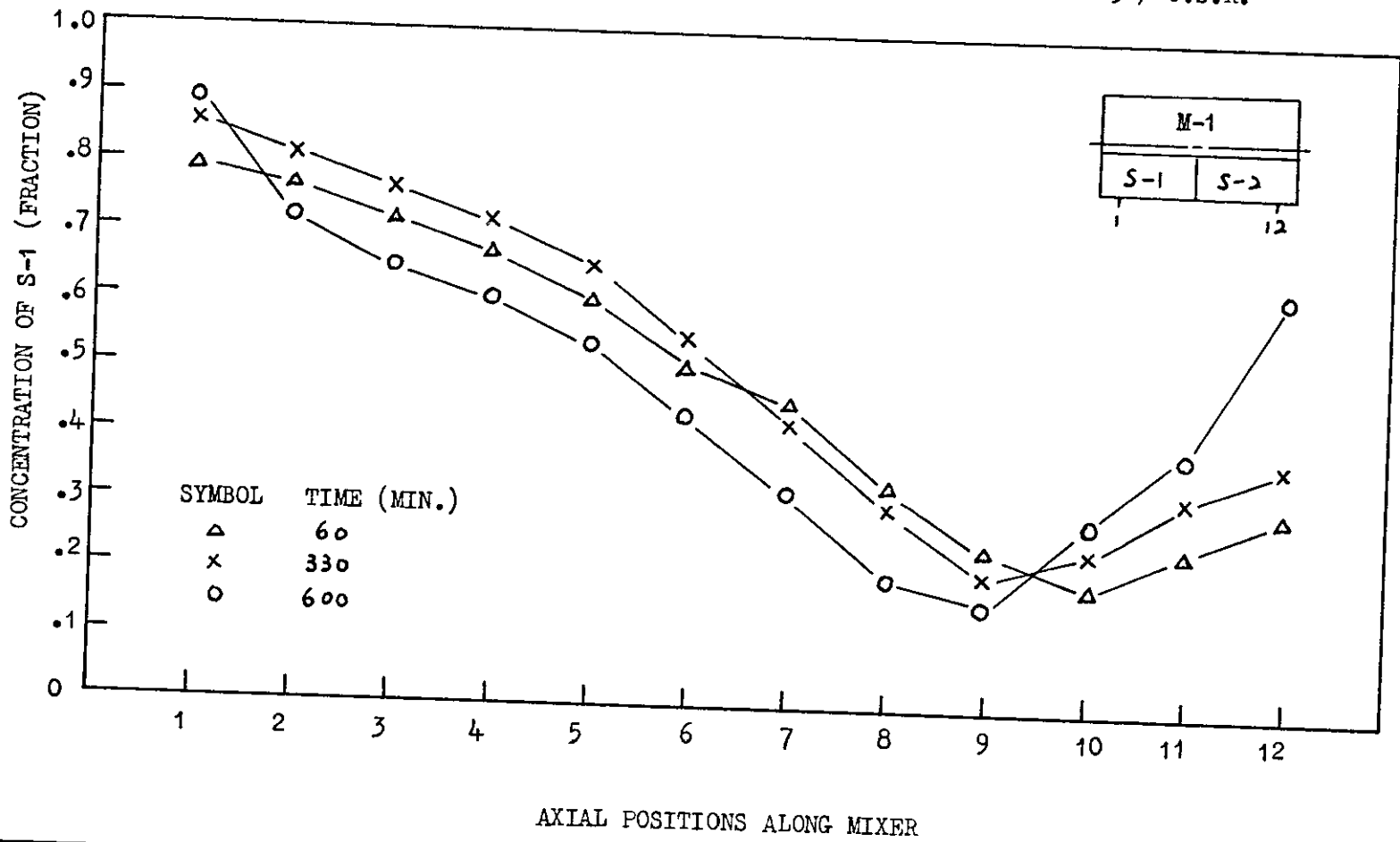
FIG. 5.27. AXIAL CONCENTRATION DISTRIBUTION OF S-1 AT 52% C.S.R.

FIG. 5.28. AXIAL CONCENTRATION DISTRIBUTION OF S-1 AT 52% C.S.R.

SYMBOL   TIME (MIN.)
△   60
✕   300
○   600

AXIAL POSITIONS ALONG MIXER

FIG. 5.29. SAMPLE VARIANCE $\sigma^2(t)$ VS. T.
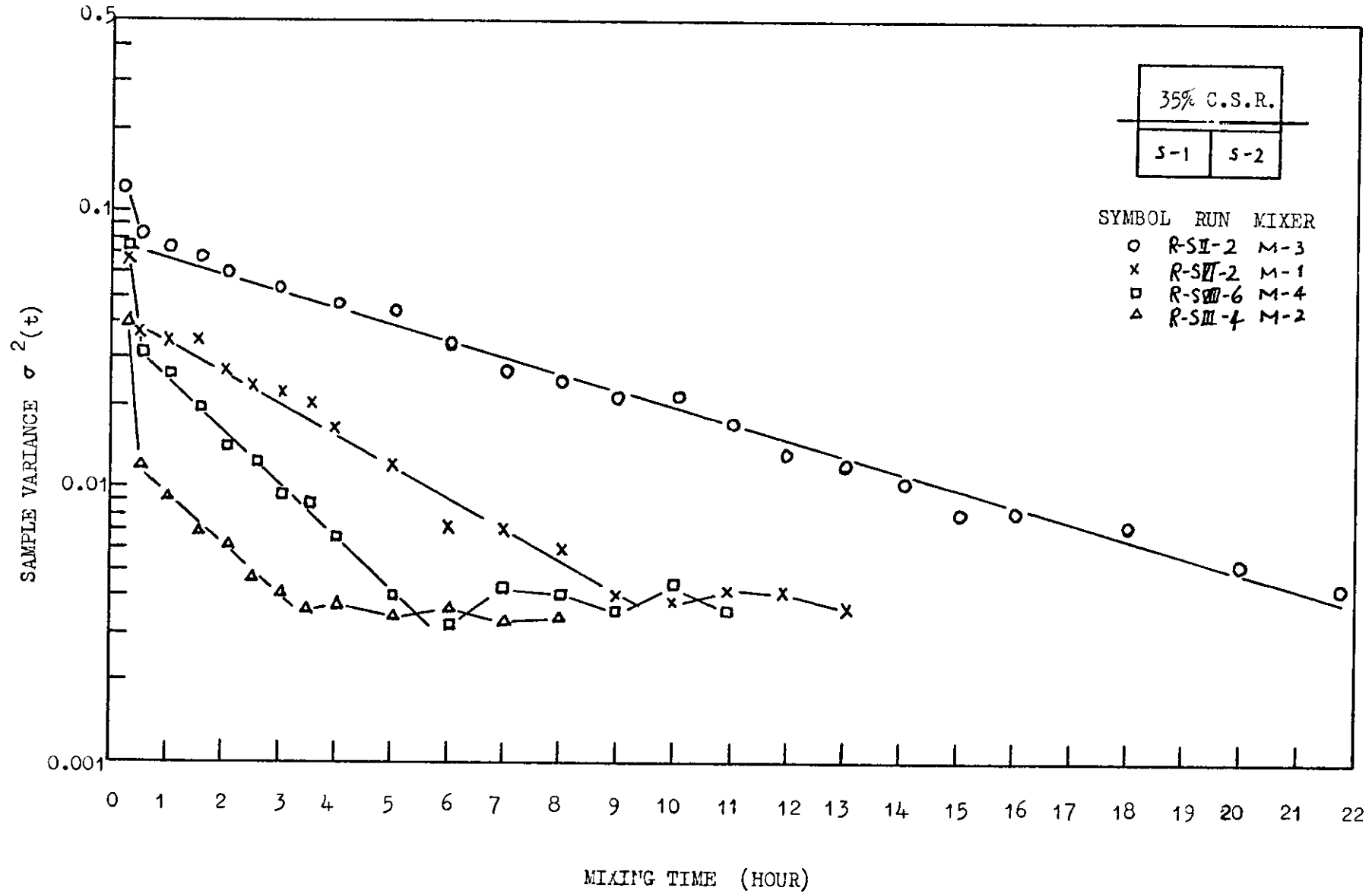
FIG. 5.30. SAMPLE VARIANCE $\sigma^2(t)$ VS. T.
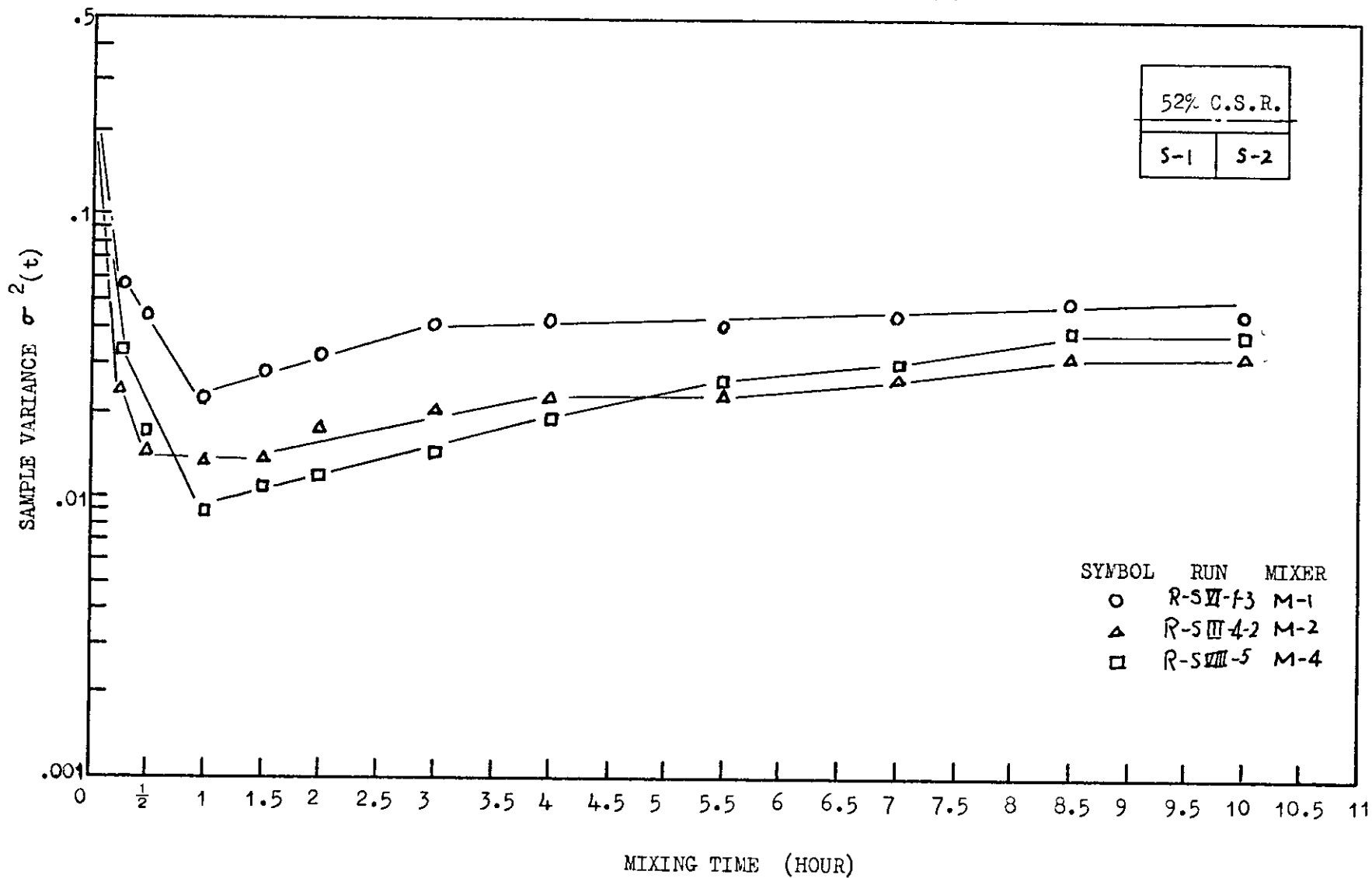
FIG. 5.31.CONCENTRATION VARIATION' S-1 AT 35% C.S.R.
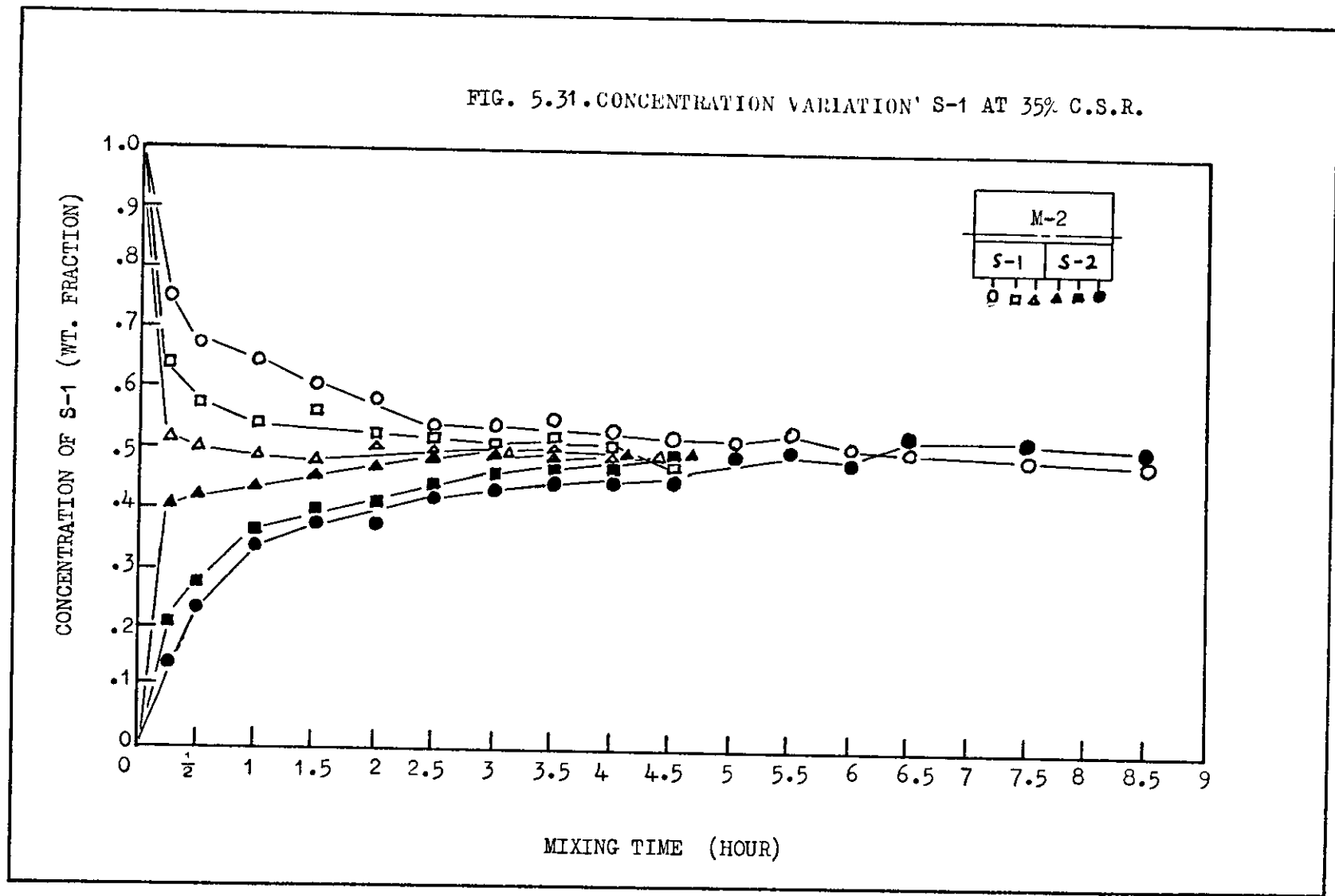
FIG. 5.32. CONCENTRATION VARIATION S-1 AT 52% C.S.R.
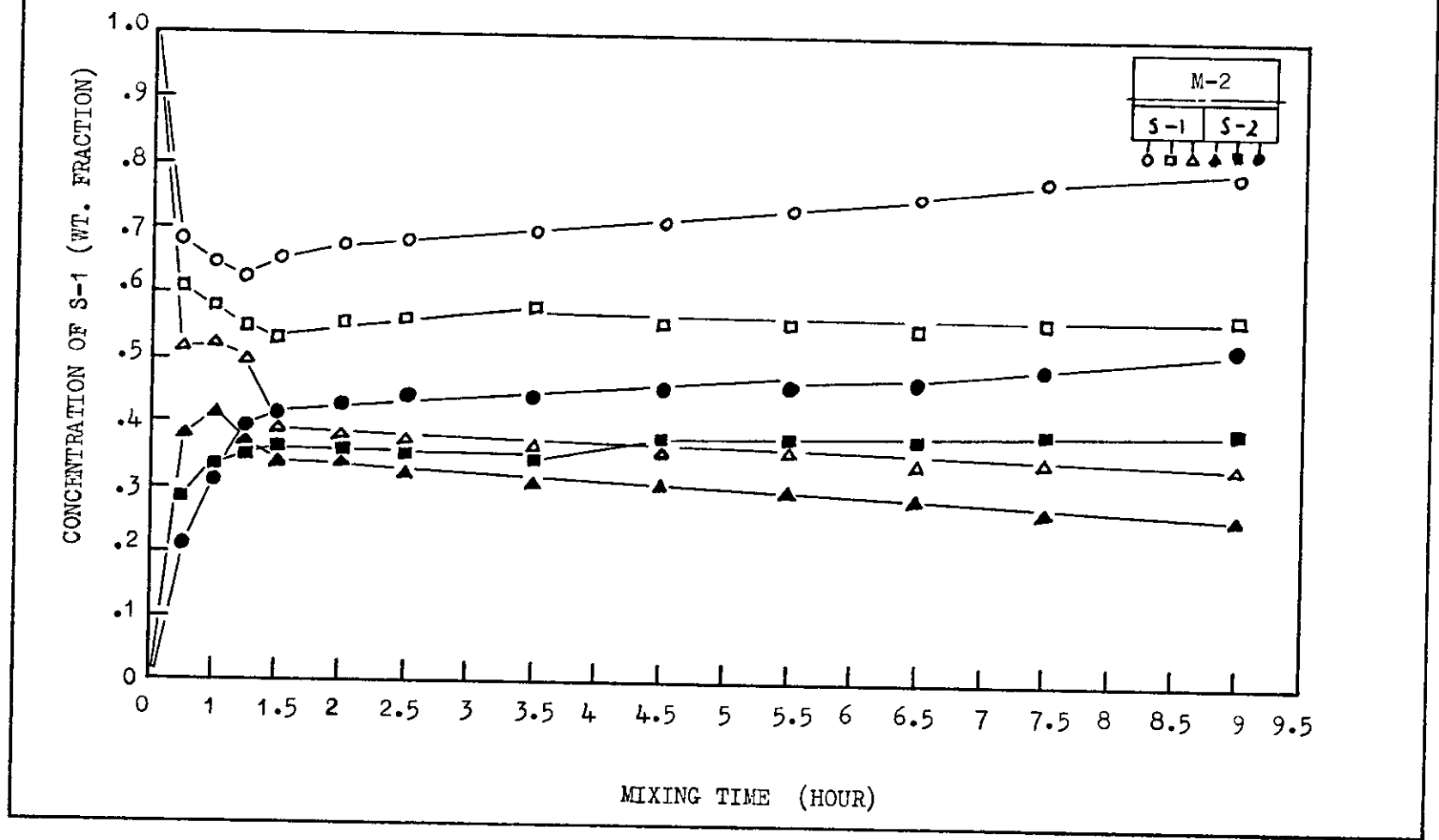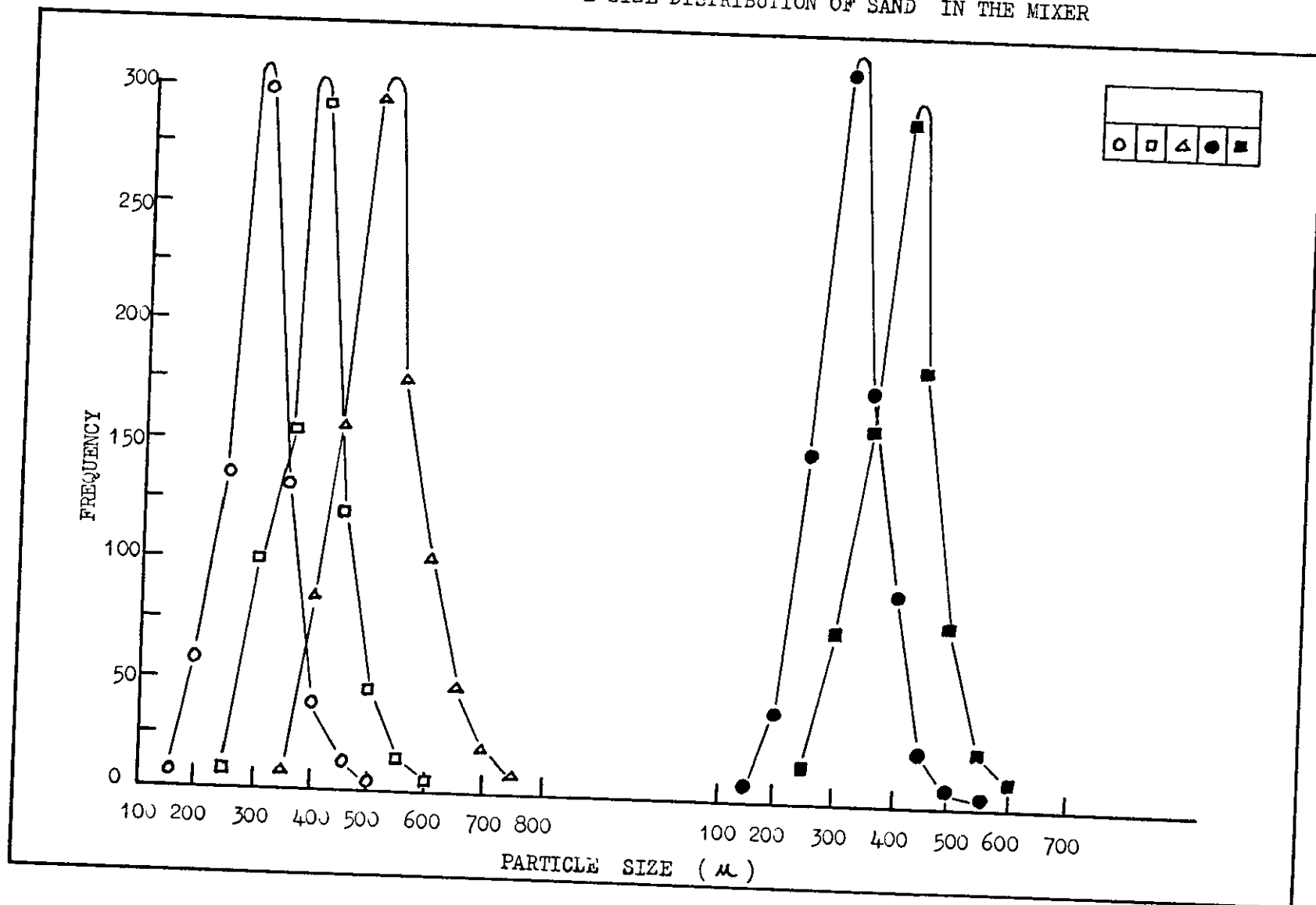
FIG. 5.33. PARTICLE SIZE DISTRIBUTION OF SAND IN THE MIXER
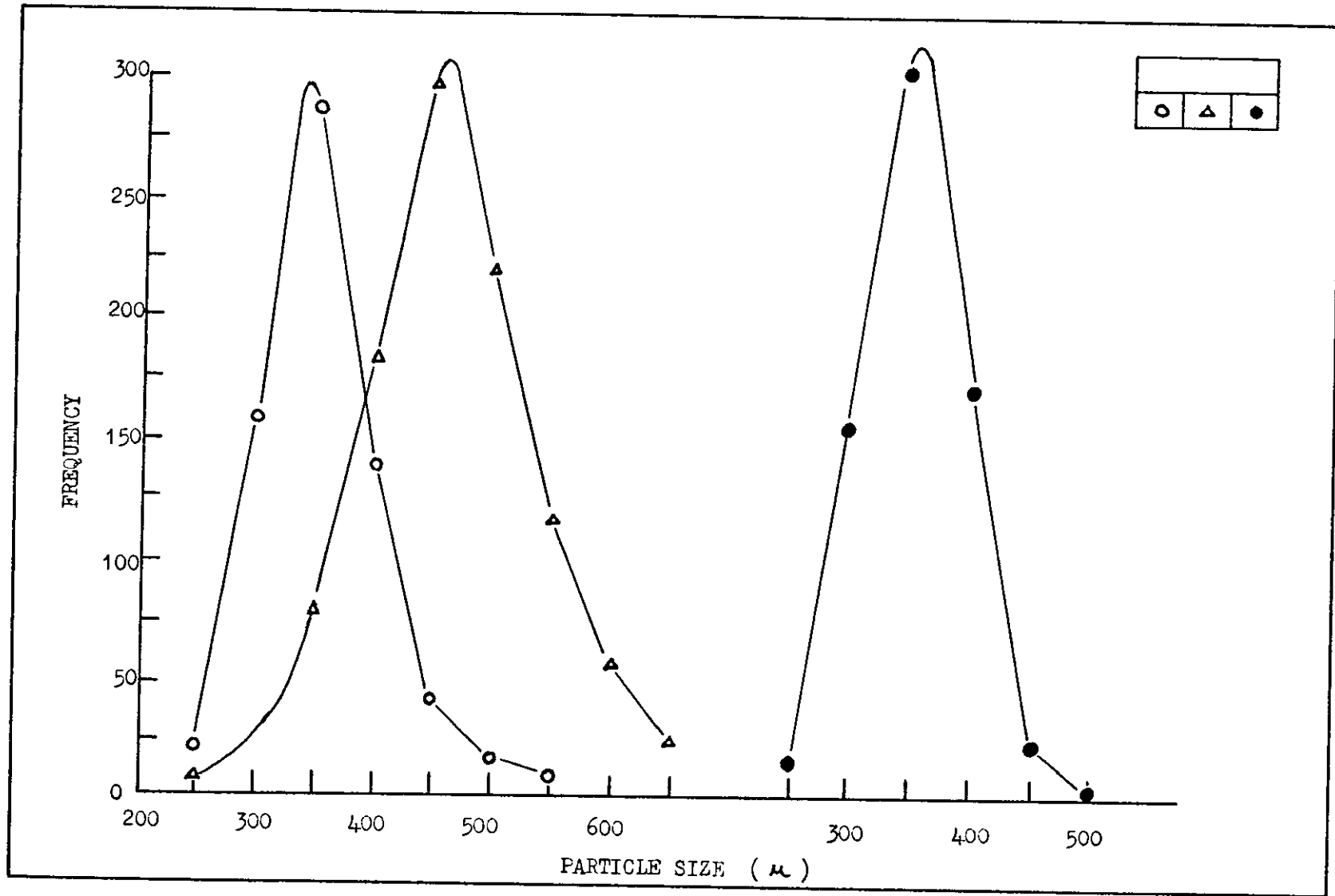
FIG. 5.34. PARTICLE SIZE DISTRIBUTION OF SAND IN THE MIXER

5.4.3. COMPONENTS WITH DIFFERENT DENSITY BUT SAME SIZE

This series of experiments was carried out into the mixing of three combinations of sand-gypsum, sand-glass beads and sand-zicron sand. These components have the same particle size distribution (-36/+52) but different in density and were mixed in mixer M-2. The particle shape of those materials are shown in Photo (5.1.)

5.4.3.1. SAND AND GYPSUM (DENSITY RATIO 1.21)

The concentration distribution curves are plotted in Fig. (5.37) at 52% of the critical speed of rotation. The profile of the curve is similar to those shown in Fig.(5.7). From this set of curves the rate of particle movement $R(t)$ and the variance $\sigma^2(t)$ can be computed and plotted as functions of mixing time in Fig.(5.38) and Fig.(5.39). The shape of these curves are similar to those shown in Fig.(5.11) and Fig.(5.12). These results indicate that even though there is a density difference and the materials do not obey one of the basic postulates, the results obtained are similar to those expected if there were no density difference and a good degree of randomisation was obtained. The time required to achieve the minimum $\sigma^2(t)$ is found longer that the case shown in Fig.(5.12). This may be thought of as caused by the difference in material properties which may affect the net displacement of individual particles. The variation of concentration with time at specific location is shown in Fig.(5.40). Further experiment was performed in mixer M-1 at the same operating conditions. The particle movement $R(t)$ and the variance $\sigma^2(t)$ are plotted in Fig.(5.38) and Fig.(5.39).

The variation of the porosity of gypsum is shown in Fig. (5.41) as a function of speed of rotation. This shows that maximum porosity is obtained at about 40% of the critical speed of rotation. Additional experiments were carried out at 35% of the critical speed of rotation and the concentration distribution curves are plotted in Fig.(5.42) and the rate of particle movement $R(t)$ and the variance $\sigma^2(t)$ are further drawn in Fig.(5.38) and Fig.(5.39). The rate of mixing can be compared and is shown as a function of speed of rotation. The variation of concentration with time at fixed positions is plotted in Fig.(5.43).

5.4.3.2. SAND AND GLASS BEADS (DENSITY RATIO 1.11)

The concentration distribution curves are plotted in Fig. (5.44) at 52% of the critical speed of rotation. The profile of the distribution curves shows segregation of sand in a band in the middle of the mixer with glass beads close to both the end walls. This form of segregation may be described by the momentum interchange which appears in Section 5.4.2.3. The variation of concentration with time at specific positions of glass beads plotted in. Fig.(5.45). These curves show the separation of the glass beads and sand particles at specific locations in terms of time. The variance $\sigma^2(t)$ vs. mixing time is plotted in Fig.(5.46). The shape of this curve is similar to those shown in Fig.(5.30). Although there is a smaller density difference than sand and gypsum, but the mixing was achieved far from random. The segregation of glass beads was further observed qualitatively from the experiments which performed in mixer M-2 at 20% of the critical speed of rotation. The concentration distribution curve of glass beads at the equilibrium state (after mixing for three hours) is shown in Fig.(5.47). The profile of this curve is similar to those shown in Fig.(5.44). The form of segregation seems definite and independent of the speed of rotation.

5.4.3.3. SAND AND ZICRON SAND (DENSITY RATIO 1.81)

450ml.of each component (640 gm.of sand and 1170 gm. of zicron sand, this gives almost equal number of particles per unit volume) were used for each experimental run. The concentration distribution curves at 52% of the critical speed of rotation is plotted in Fig.(5.48). The profile of the concentration distribution shows the increase concentration of zicron sand in the middle of the mixer and an equilibrium state is rapidly established. The radial concentration distribution along the diameter of the drum is shown in Fig. (5.51). This concentration distribution was obtained from the middle of the mixer after mixing for 180 minutes. The profile of this radial concentration distribution is shown to be a function of position. It was found that the lighter particles (sand) were also concentrated near the end walls. A three dimensional concentration distribution is attributed to such system. The variance $\sigma^2(t)$ vs. mixing time is plotted in Fig.(5.46). The shape of this curve is similar to that obtained from mixing of sand and glass beads. The

variation of concentration with time is shown in Fig.(5.50).

Fig.(5.49) shows the concentration distribution at 20% of the critical speed of rotation and after mixing for 200 minutes. Generally, the profile of this curve is similar to those shown in Fig.(5.48). The radial distribution at this speed is shown in Fig.(5.52). In general, the state of mixedness is stable, regardless of the speed of rotation.

5.4.3.4. DISCUSSION OF THE RESULTS AND CONCLUSION

The results obtained from three combinations of materials have shown a great variety from one combination to the other. The theory set up in Chapter 4 to describe the mixing process with components of different density but same sized particles is clearly not generally applicable.

Without considering any mixing process, let us consider this equation in general. By assuming the mixing is allowed to extend towards infinity so that an equilibrium state may be achieved

$$\lim_{t \to \infty} \left(\frac{\partial P}{\partial t} \right) \longrightarrow 0$$

so

$$D \frac{\partial^2 P}{\partial x^2} - \beta \frac{\partial P}{\partial x} = 0$$

The concentration distribution as the mixing time approaches infinity could be obtained by solving this equation. Viz :-

$$P_{t \to \infty} = C \frac{D}{\beta} \exp\left(\frac{D}{\beta} x\right) + C_1$$

Assuming D and β are constants, the concentration distribution is an exponential function of axial position. Thus axial segregation must be expected in such system.

In the case of sand and gypsum, the equilibrium concentration distribution presented no exponential relationship with the axial positions, and a good degree of randomisation was obtained. This may be assumed that the momentum difference between particles is very small, so that each particle in the system will have an equal probability of undergoing displacement. Thus Eq.(4.14) can be used to interpret this kind of mixing process.

By using Eq.(5.23') and Eq.(5.25') the rate determining

parameter D – migration coefficient can be calculated from the experimental runs and are presented in Table 5.6. Here the migration coefficient obtained in two mixers appears to be constant.

TABLE 5.6. VALUES OF MIGRATION COEFFICIENT D

FOR SAND AND GYPSUM SYSTEM

Run	Mixer	% of critical speed of rotation	D (cm/sec^2)	
			from $R(t)$	from $^2(t)$
R–SVI1–3	M–1	52	0.00804	0.00801
R–SVI1–6	M–2	52	0.00797	0.00793
R–SAVI1–1	M–2	35	0.0146	0.0144

However the segregation obtained from mixing sand and glass beads where density ratio is less than the sand –gypsum mixture indicates that some other influence such as particle shape may be a contributing factor to segregation.

In the case of large density difference, as sand mix with zicron sand, the system has shown a three dimensional concentration distribution profile. In such system the neglection of the displacement of particles in Y– and Z– directions may be unreasonable, so that a three dimensional equation should be used. Moreover the end walls of the drum may play an important part on the concentration distribution, as the particles arrive the ends could have changed their original probability of undergoing displacement. Thus different distribution near the end walls would be expected.

It can be concluded from these experimental results that for the particles of similar shape, the effects of small density difference on the final state of mixedness would be very minor and a good degree of mixing can be obtained.

When the components have large density difference, segregation occurs both axially and radially and a three dimensional stochastic equation might be able to describe the mixing process. The form of such segregation in the mixer can not be predicted and is independent of the speed of rotation.

When particles have large difference in shape. Segregation must be expected even when the density difference is small. The final state of the mixture is independent of speed of rotation.

FIG. 5.37. AXIAL CONCENTRATION DISTRIBUTION OF S-1 AT 52% C.S.R.
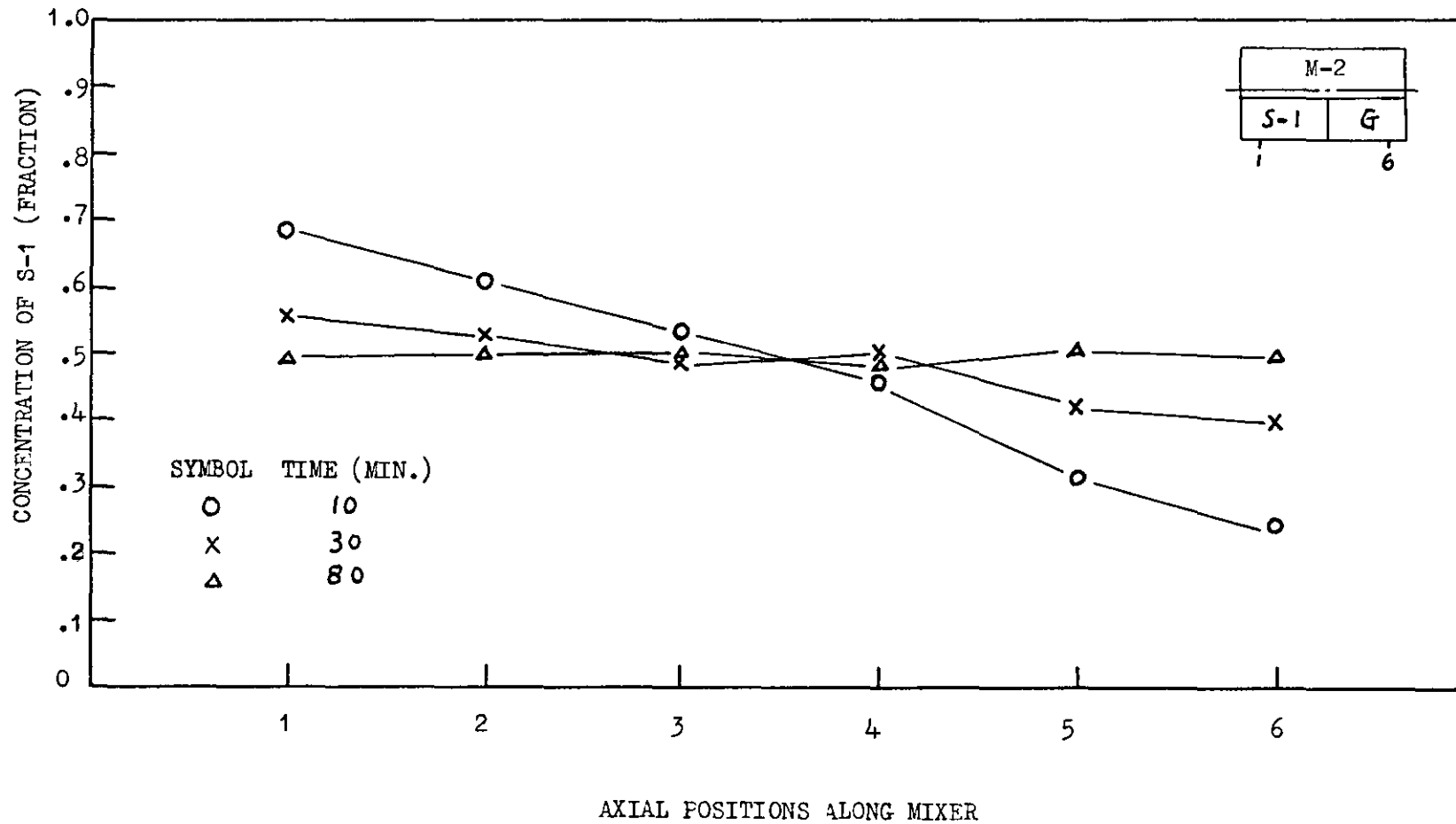
FIG. 5.38. THE RATE OF PARTICLE MOVEMENT R(t) VS. T.

MIXING TIME T. (MIN.)
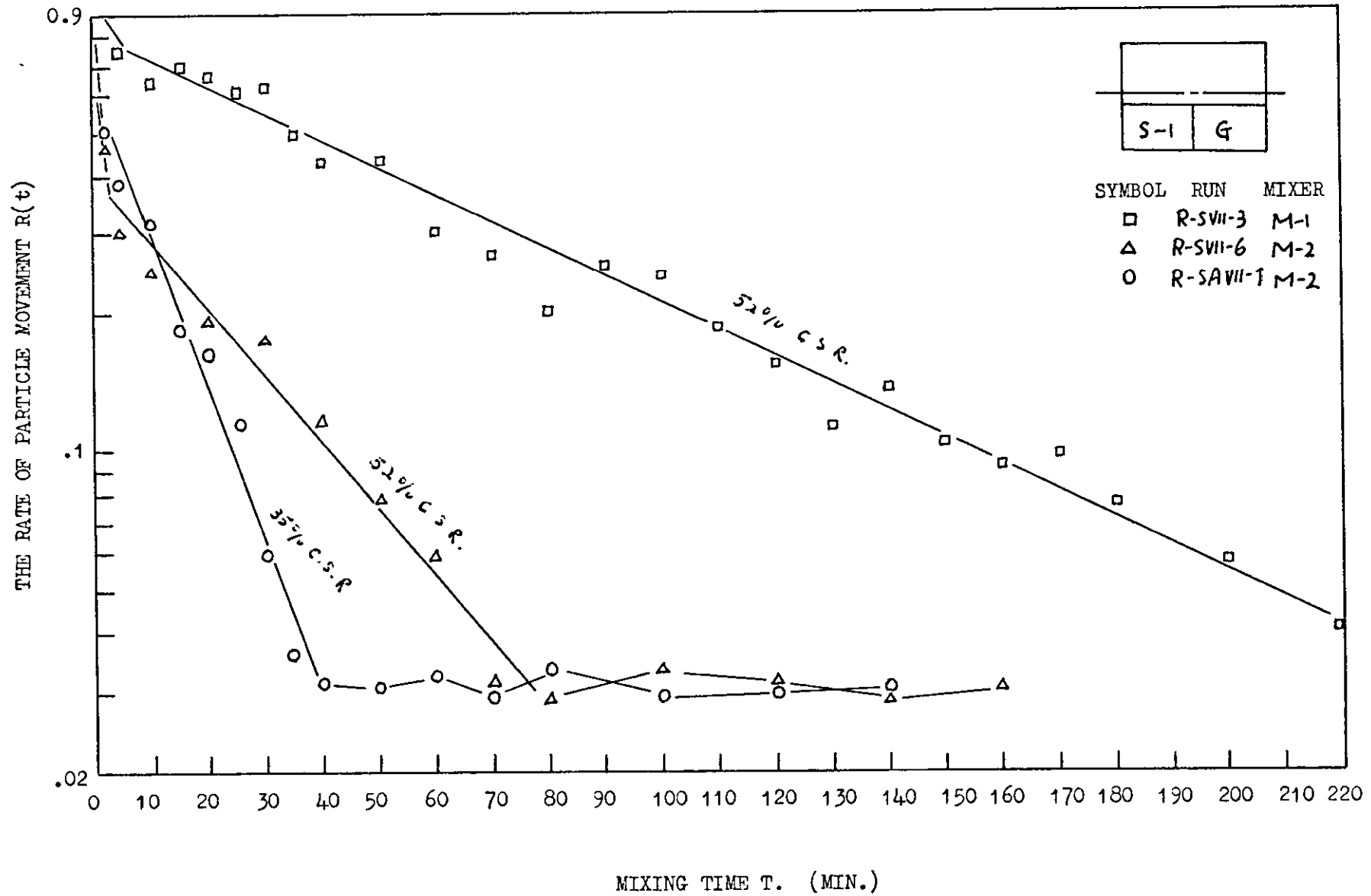
FIG. 5.39. SAMPLE VARIANCE $\sigma^2(t)$ VS. T.
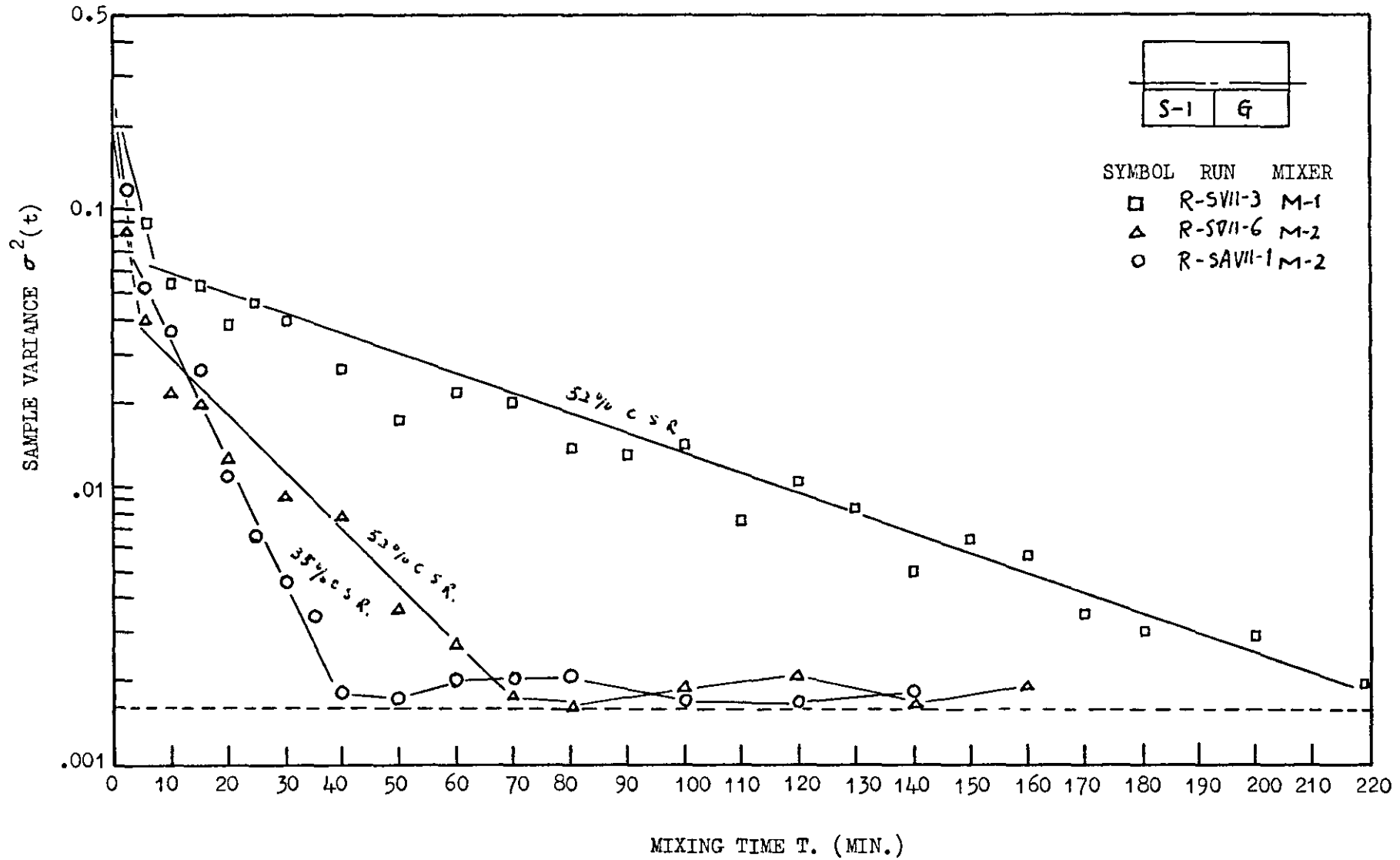
Fig. 5.40 CONCENTRATION VARIATION S-1 AT 52% C.S.R.
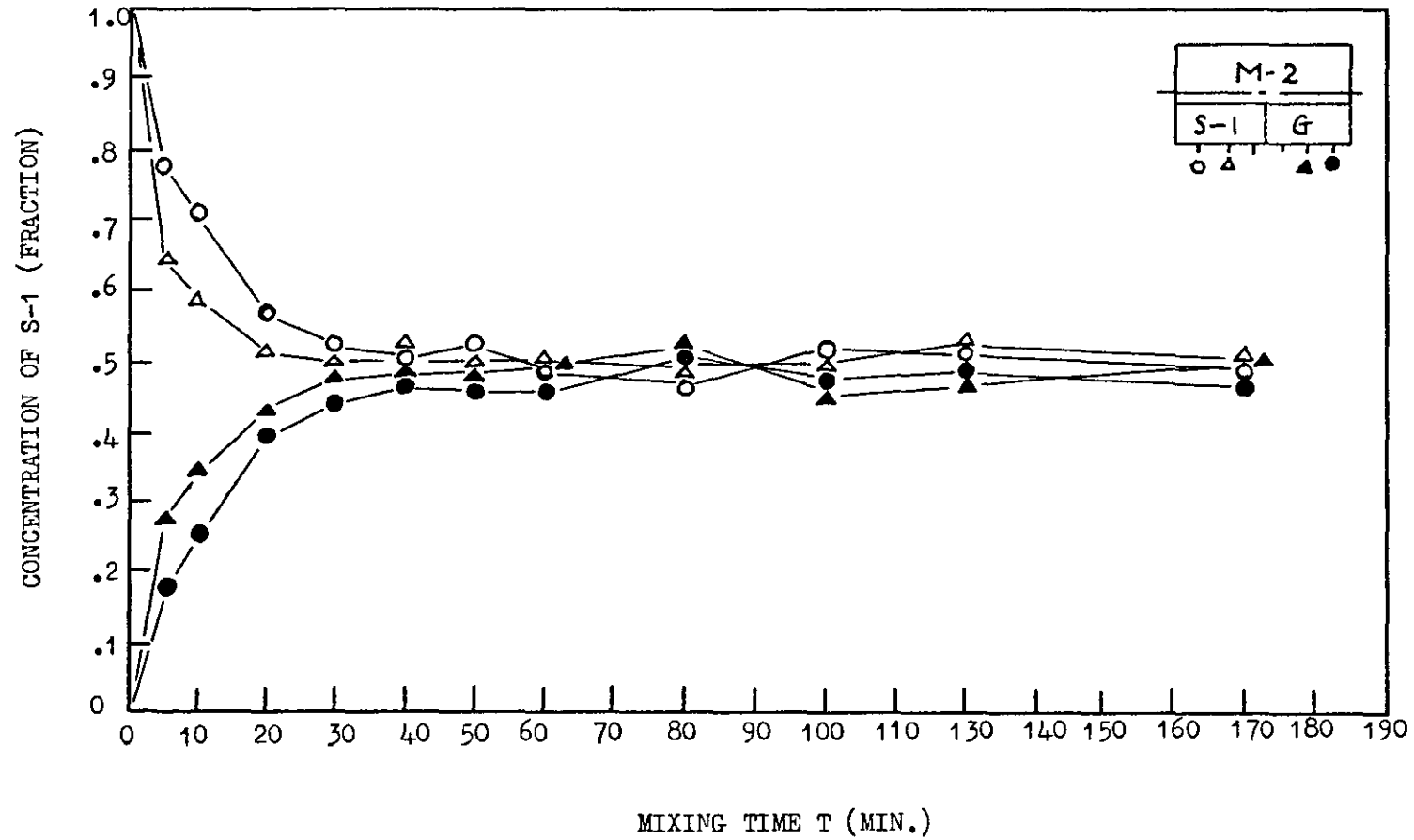
FIG. 5.41. POROSITY OF GYPSUM (%) VS. PERCENT OF CRITICAL SPEED OF ROTATION
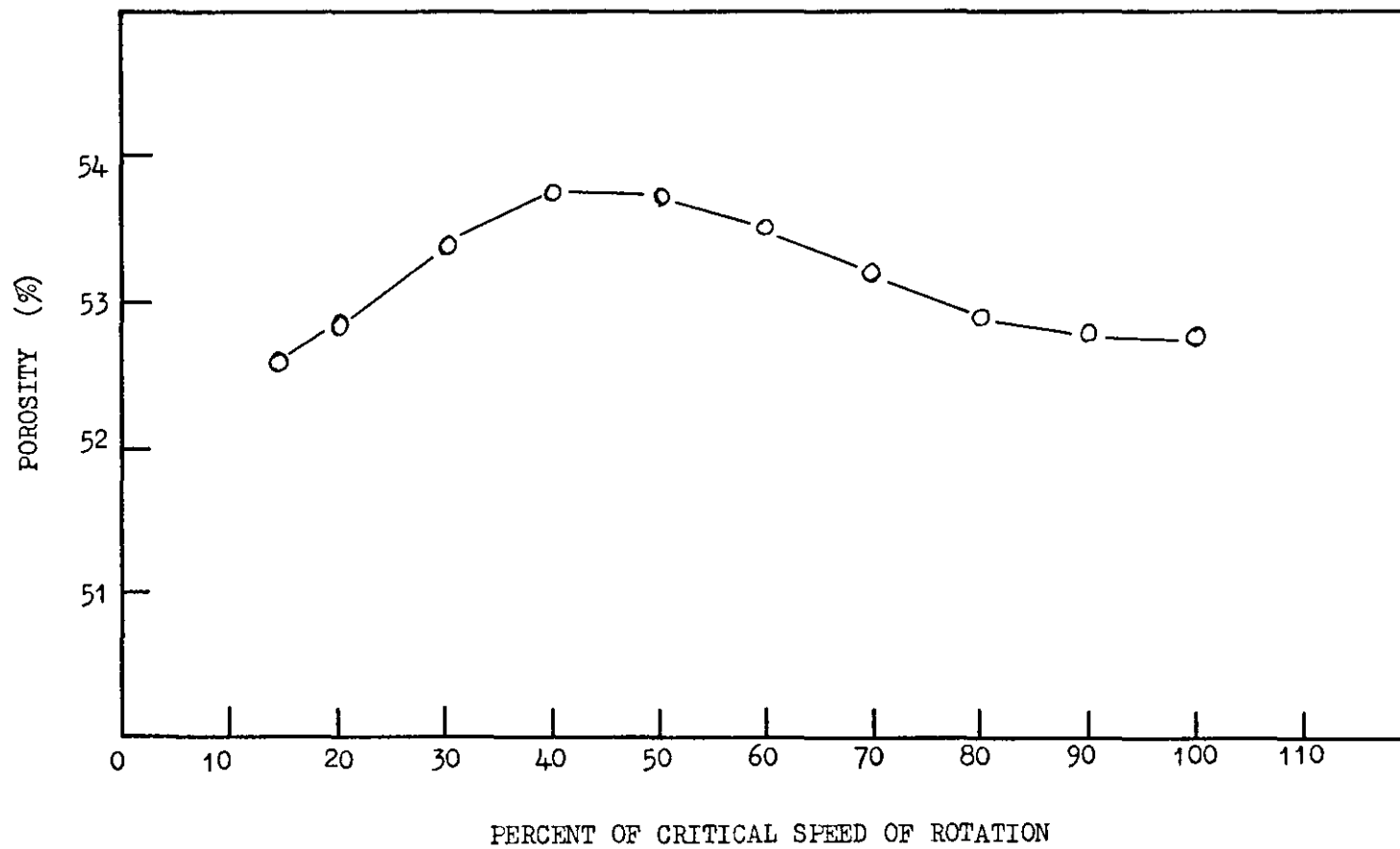
PERCENT OF CRITICAL SPEED OF ROTATION

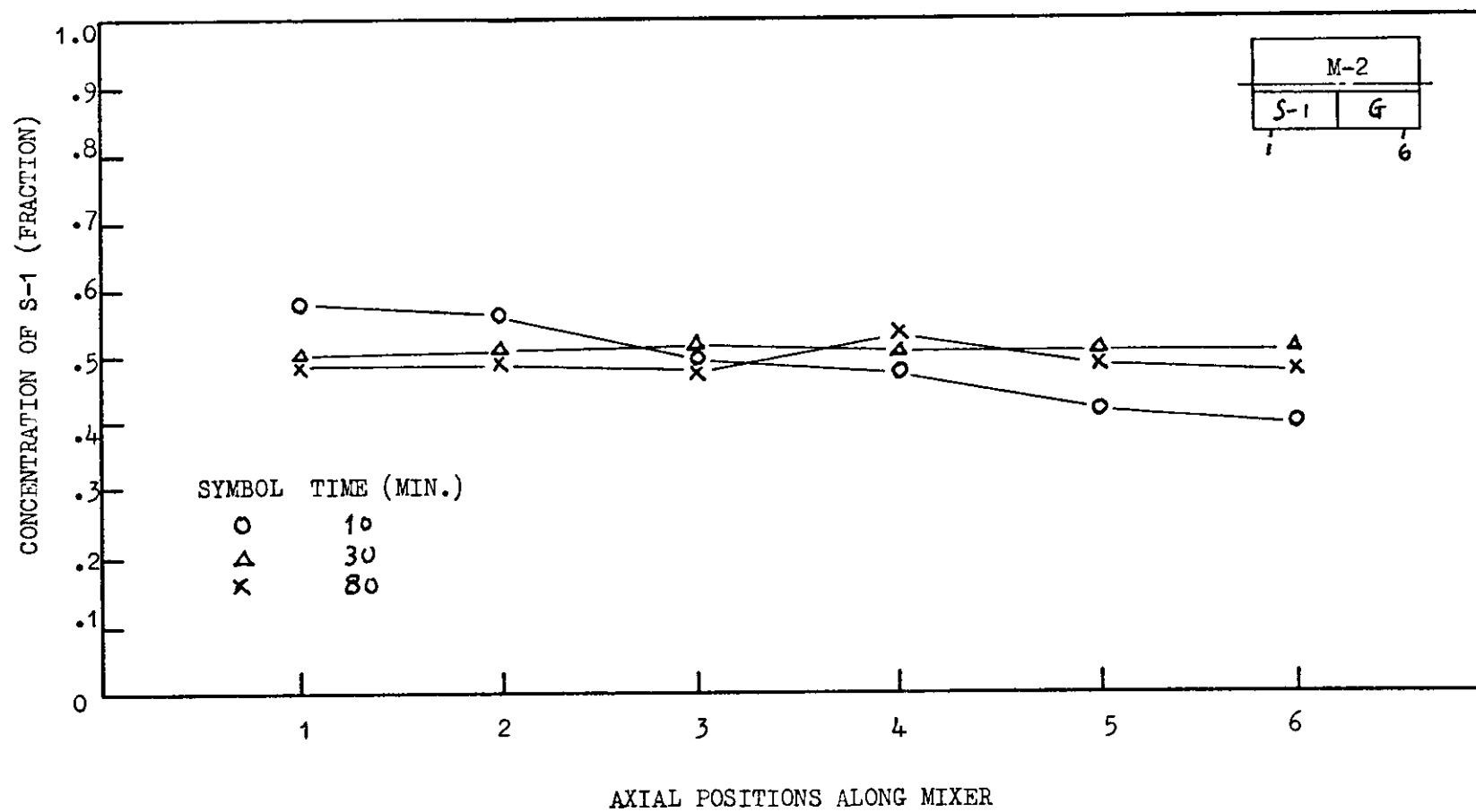FIG. 5.42. AXIAL CONCENTRATION DISTRIBUTION OF S-1 AT 35% C.S.R.

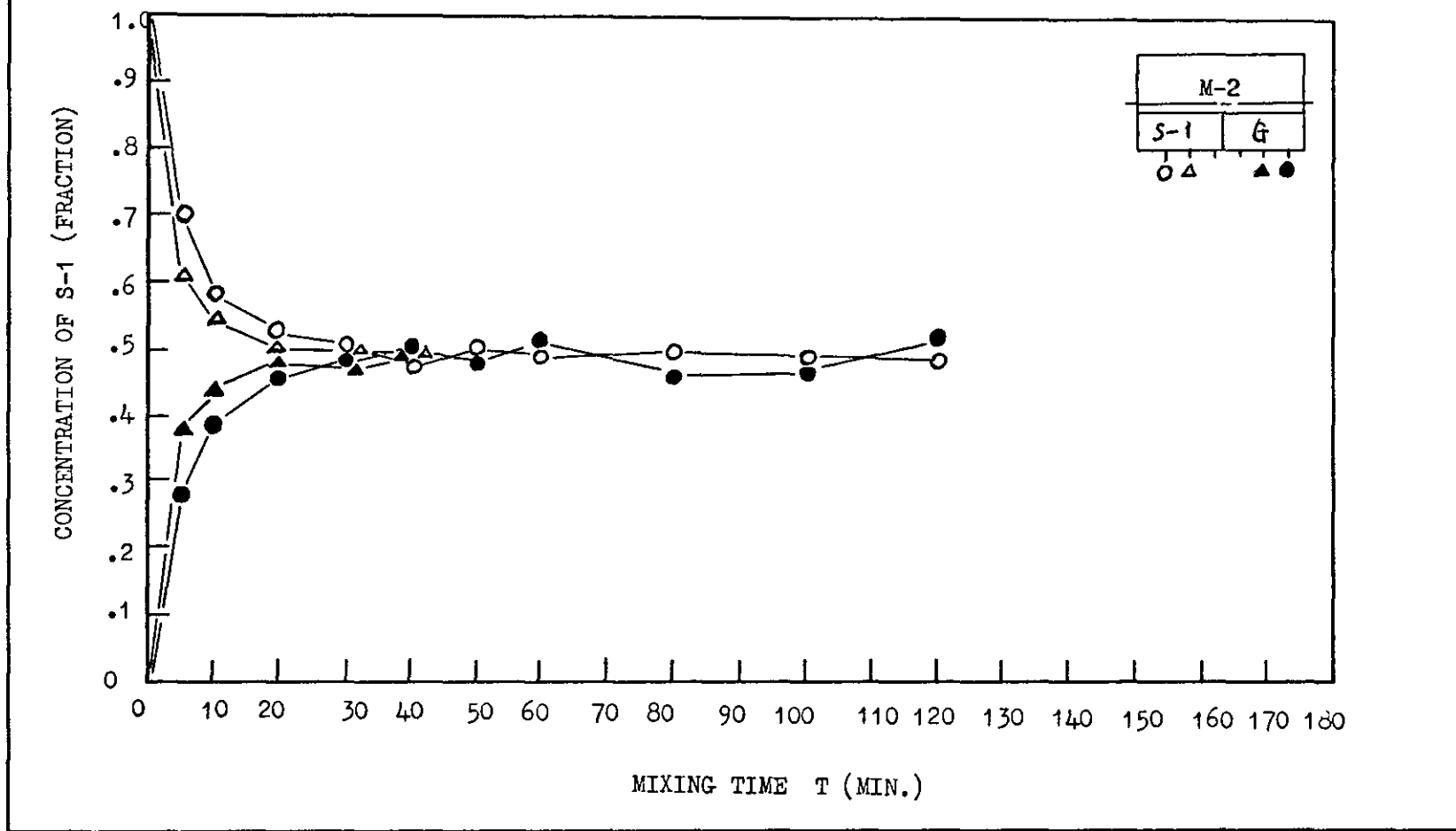FIG. 5.43 CONCENTRATION VARIATION S-1 AT 35% C.S.R.

FIG. 5.44. AXIAL CONCENTRATION DISTRIBUTION OF GLASS BEADS AT 52% C.S.R.
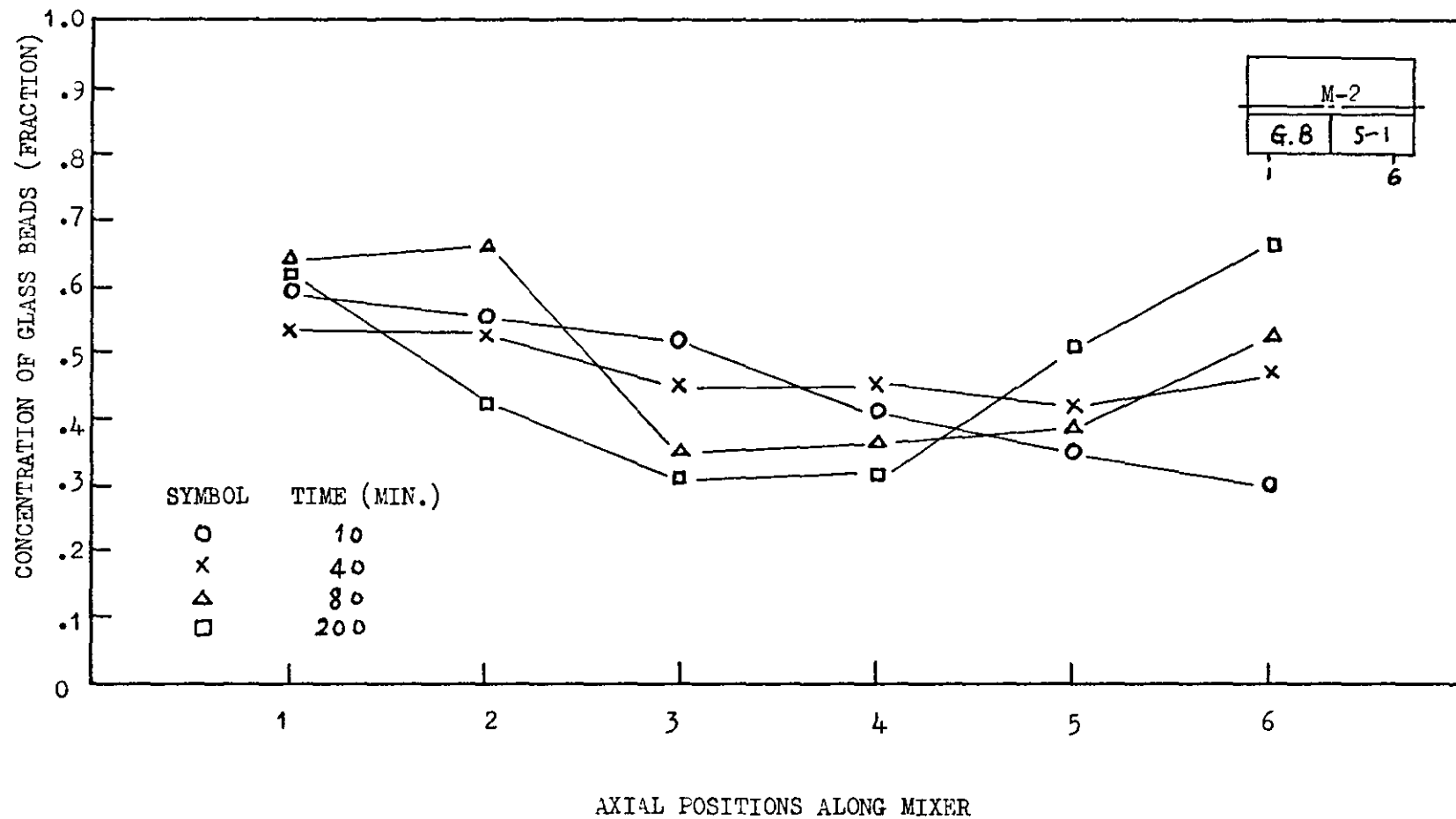
FIG. 5.45. CONCENTRARION VARIATION GLASS BEADS AT 52% C.S.R.
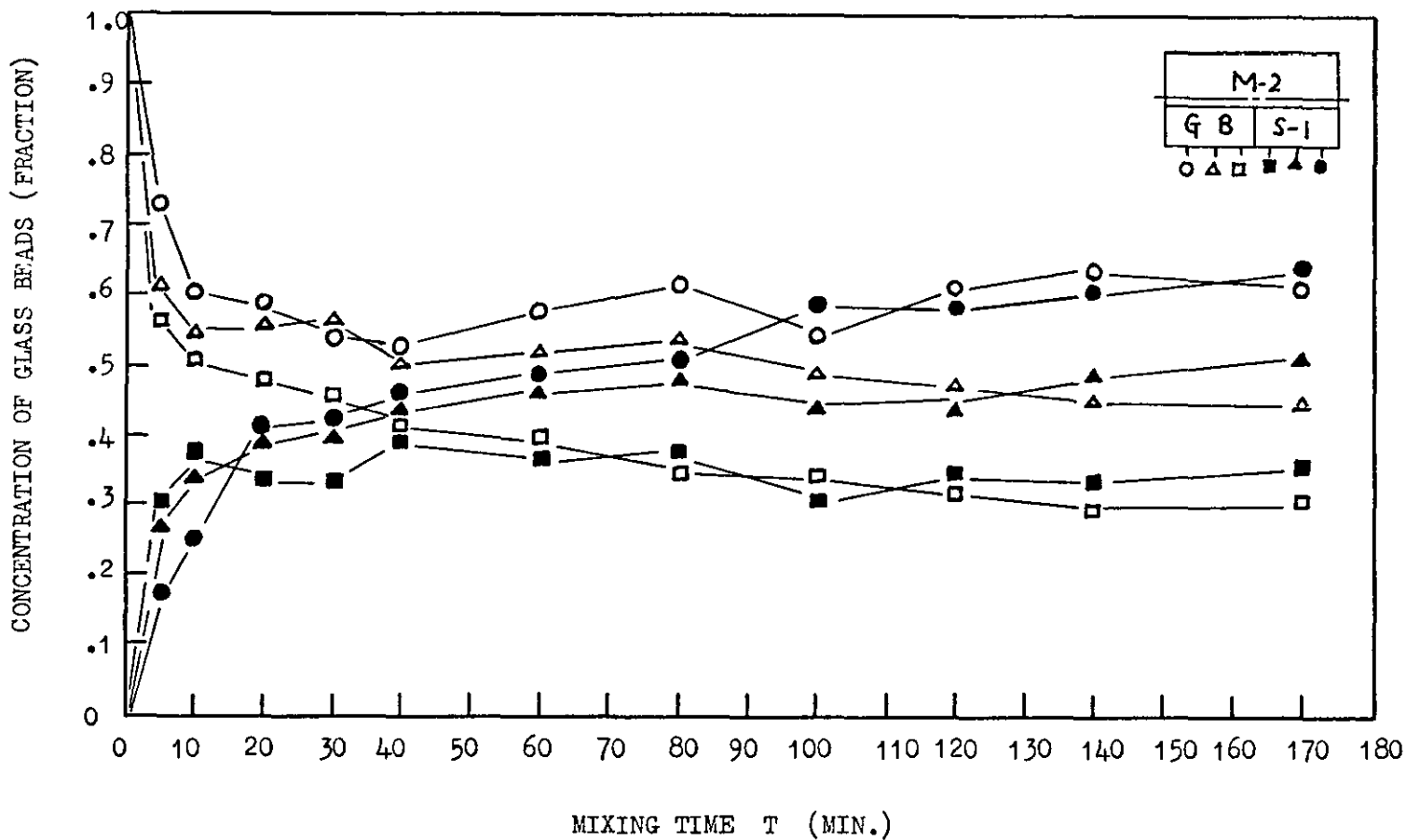
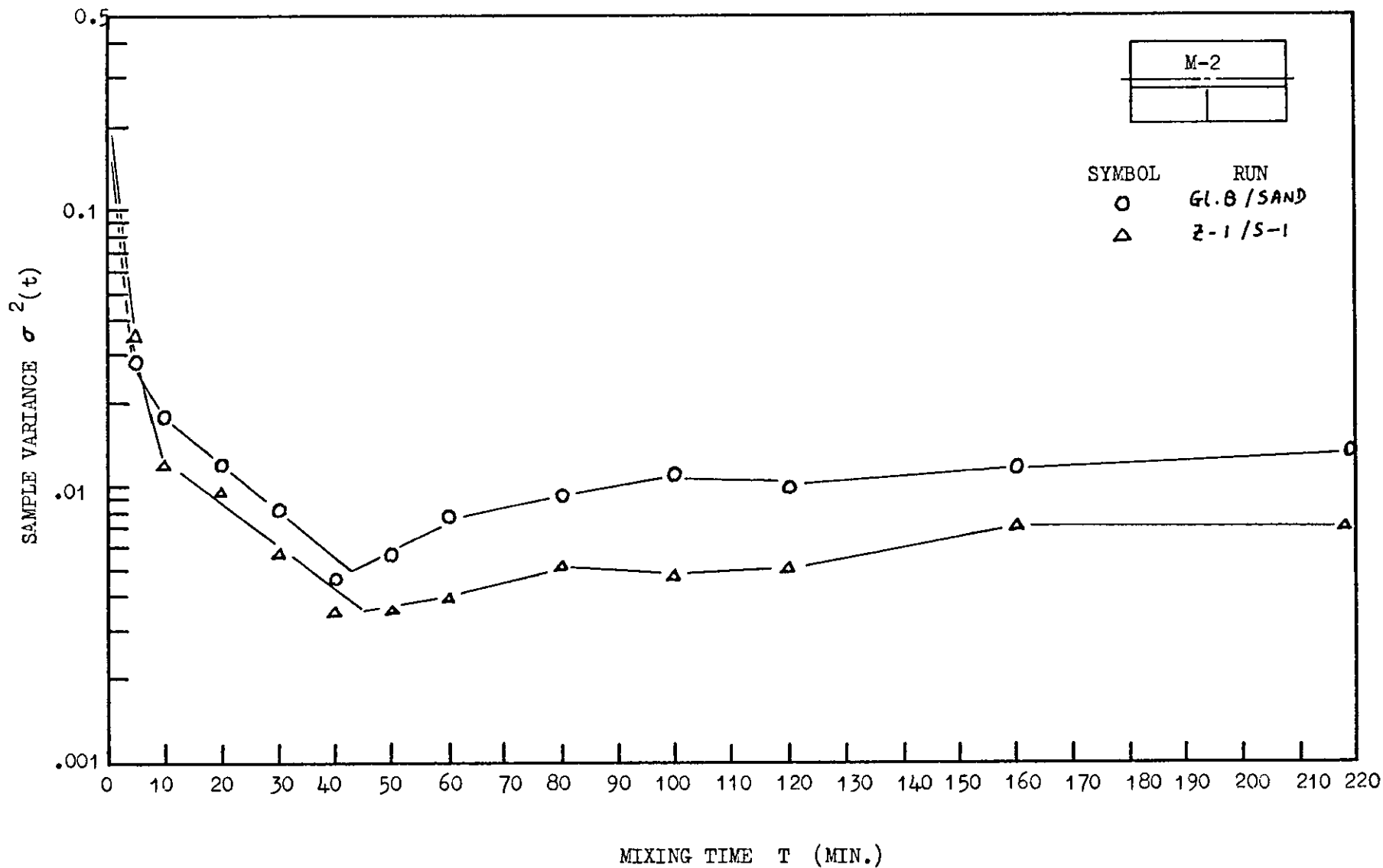FIG. 5.46. SAMPLE VARIANCE $\sigma^2(t)$ VS. T AT 52% C.S.R.

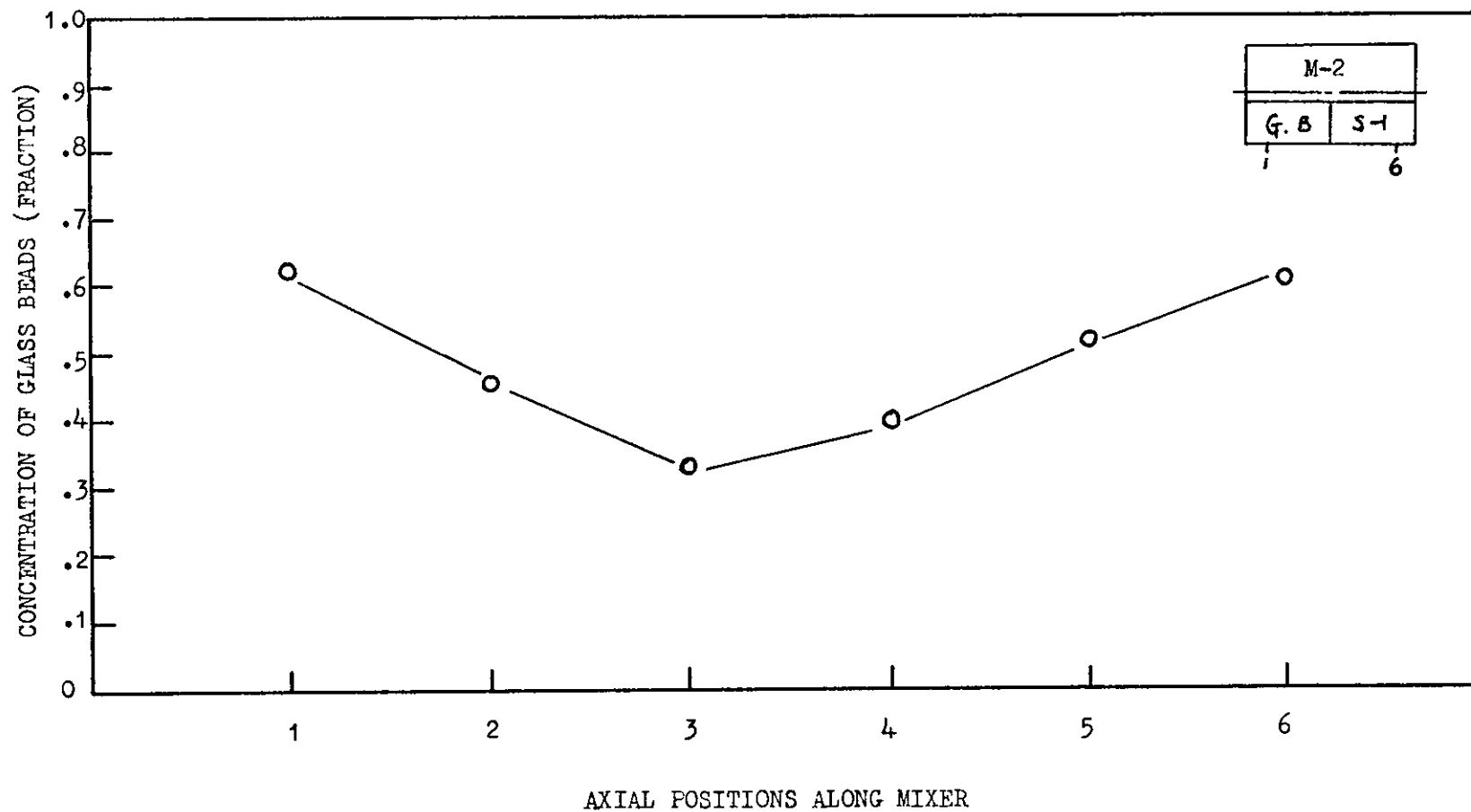FIG. 5.47. AXIAL CONCENTRATION DISTRIBUTION OF GLASS BEADS (EQUILIBRIUM) AT 20% C.S.R.

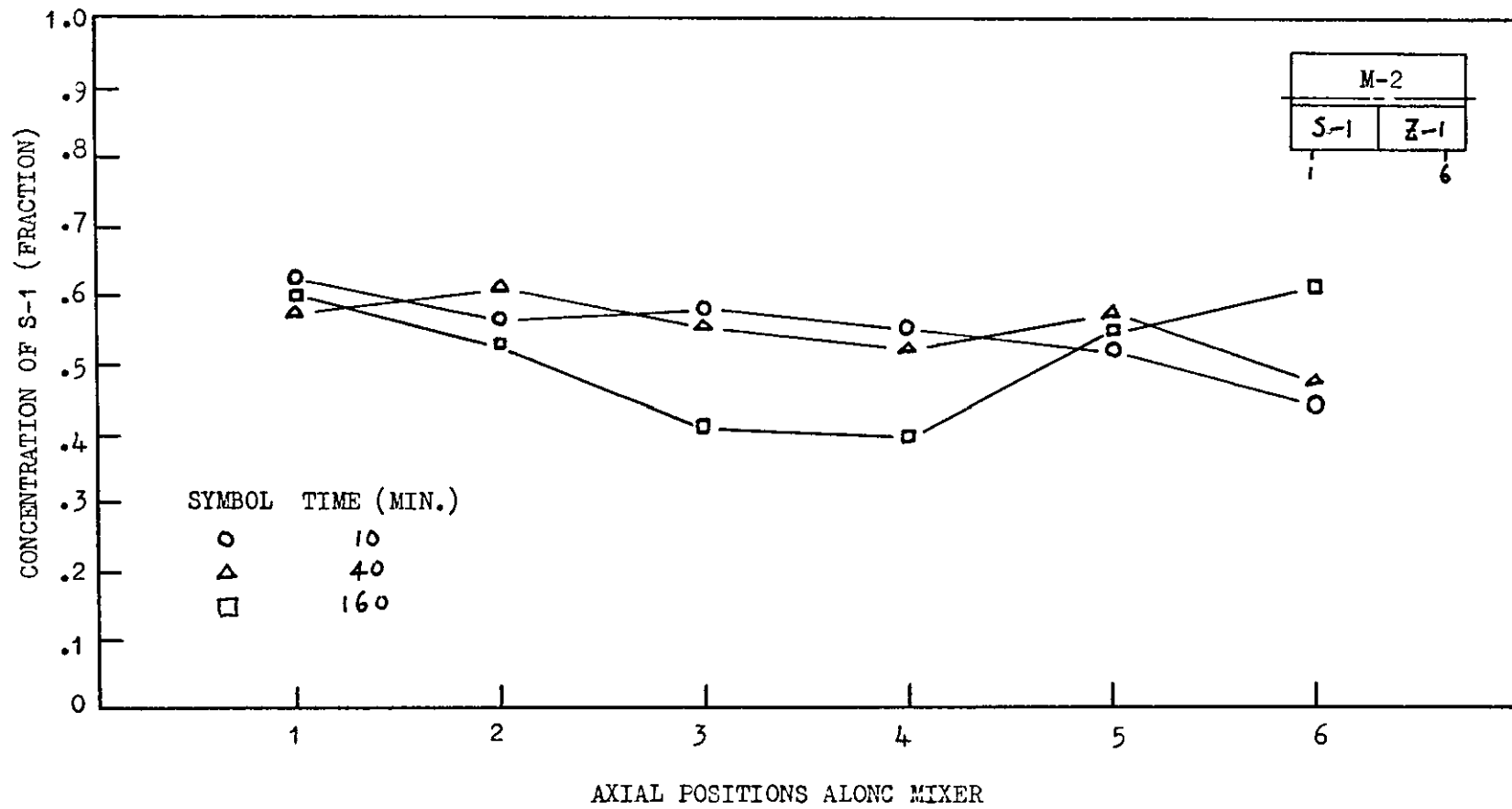FIG. 5.48. AXIAL CONCENTRATION DISTRIBUTION OF S-1 AT 52% C. S. R.

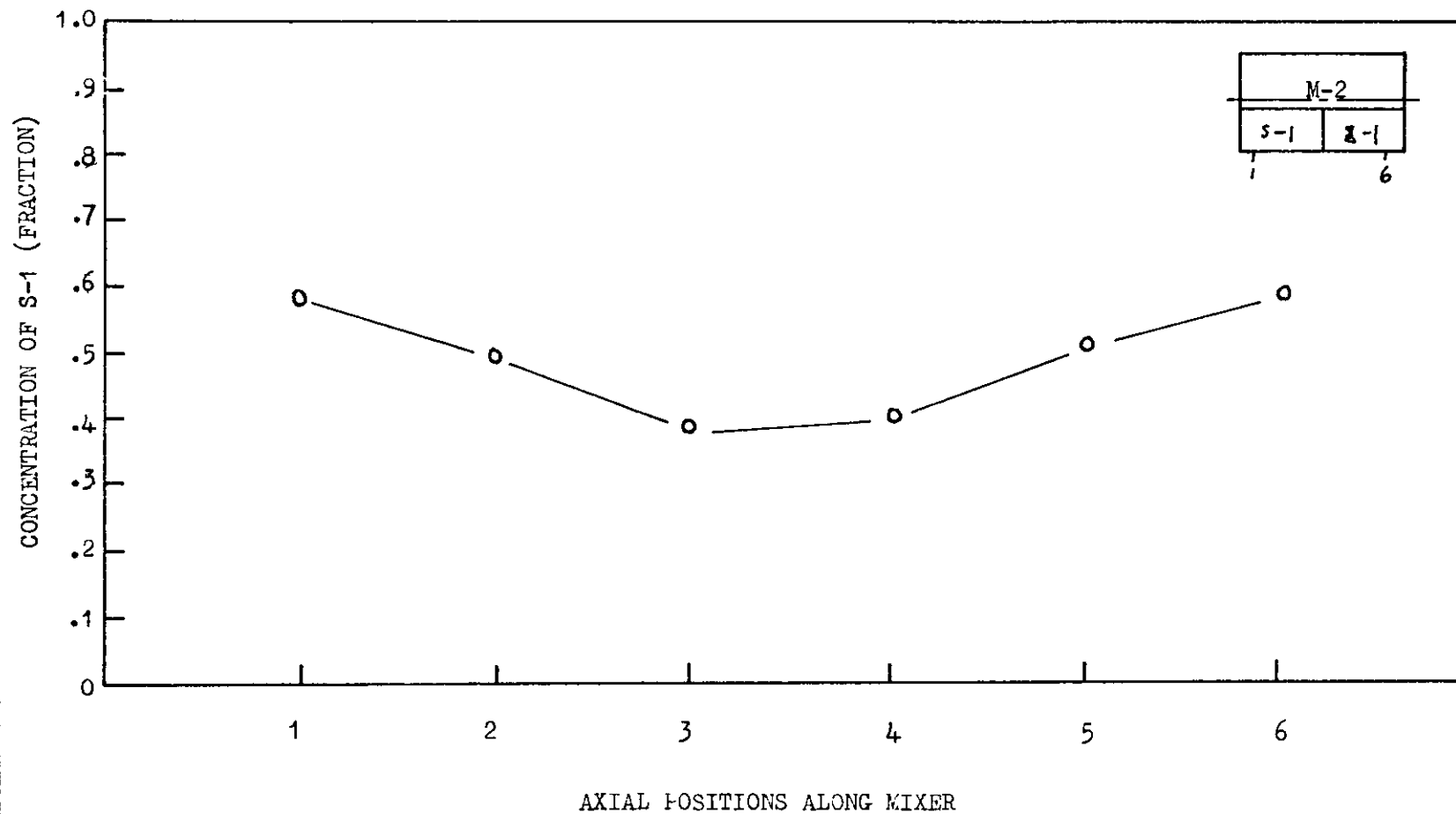FIG. 5.49. AXIAL CONCENTRATION DISTRIBUTION OF S-1 (EQUILIBRIUM) AT 20% C.S.R.

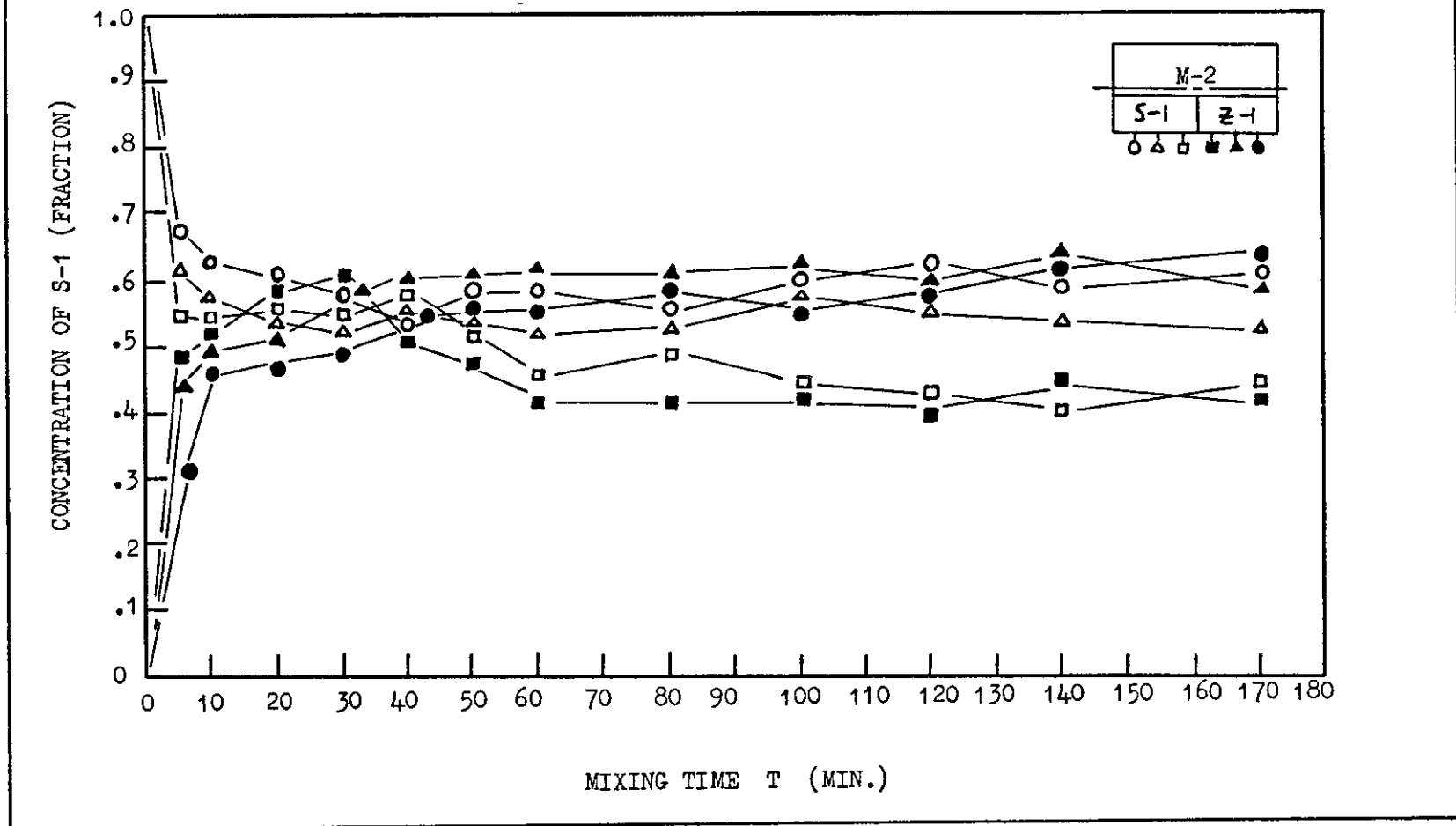FIG. 5.50. CONCENTRATION VARIATION S-1 AT 52% C. S. R.

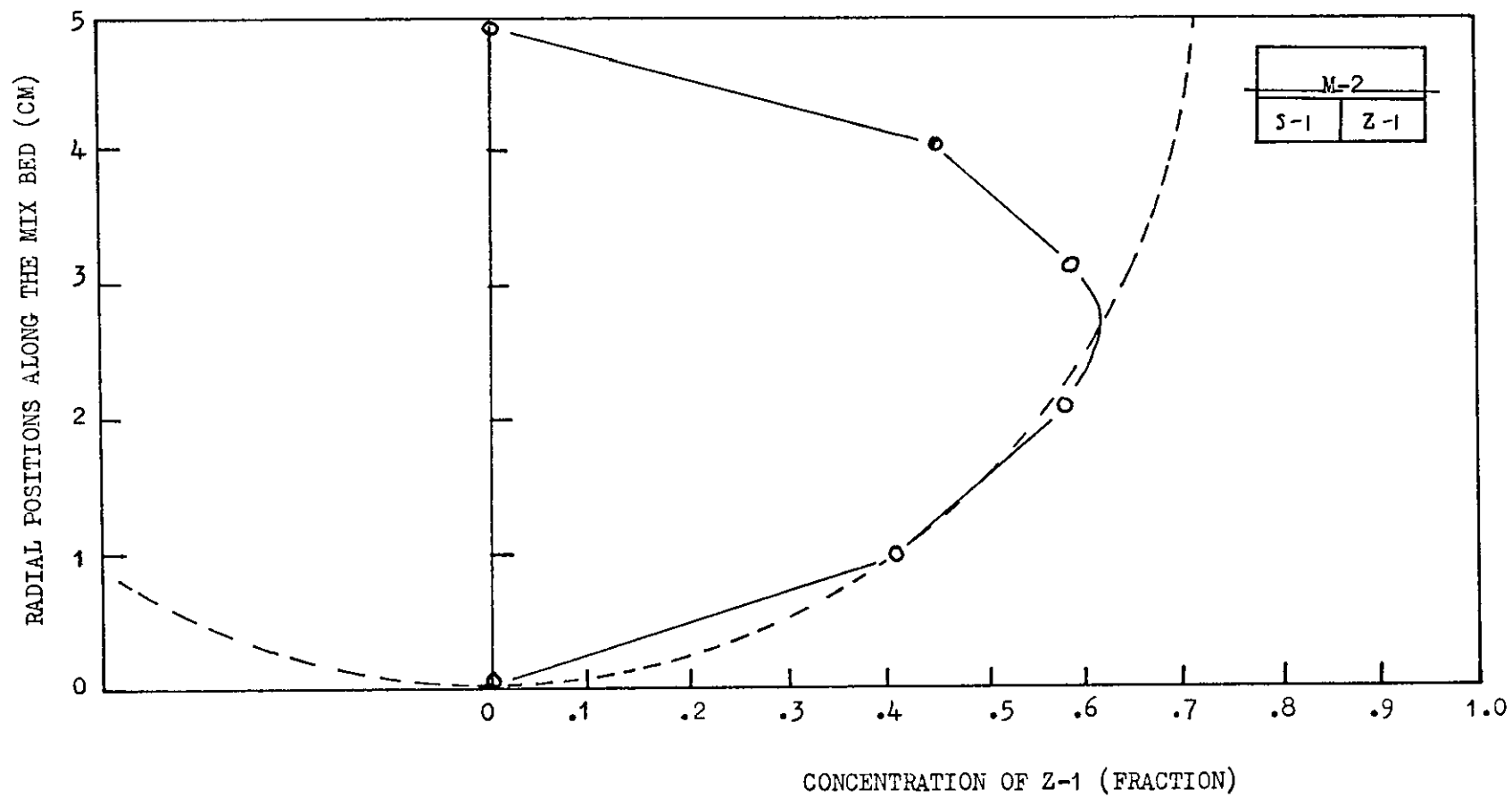FIG. 5.51. RADIAL CONCENTRATION DISTRIBUTION OF Z-1 AT 52% C. S. R.

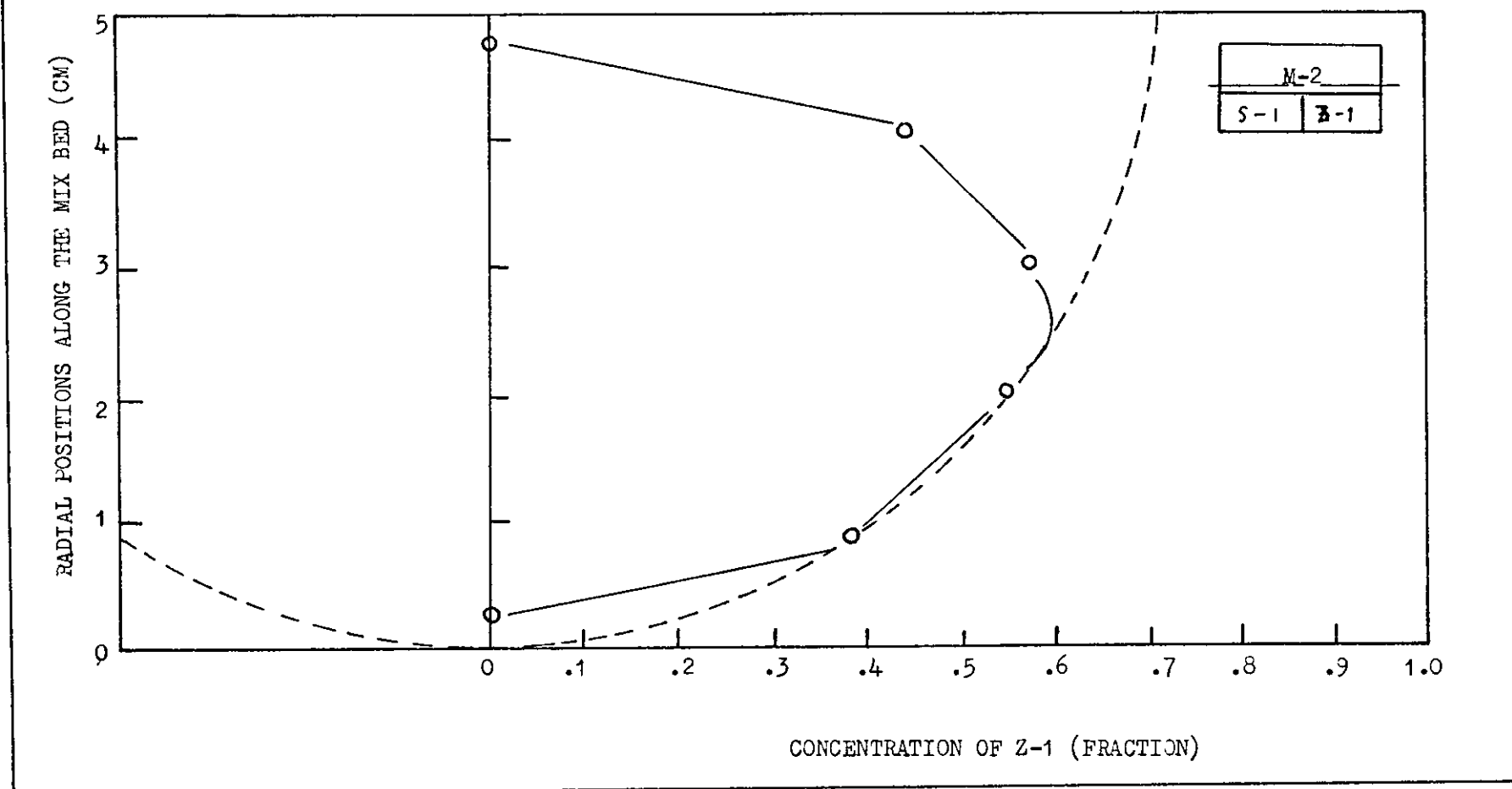FIG. 5.52. RADIAL CONCENTRATION DISTRIBUTION OF Z-1 AT 20% C. S. R.

5.4.4. COMPONENTS WITH DIFFERENT DENSITY AND DIFFERENT SIZE

In this series of experiments investigations were made into the mixing of combinations of sand(S-1) and zicron sand (Z-2) in mixer M-2. 470ml. of each component was used (700 gm. of sand and 1310 gm. of zicron sand). Samples were drawn at four horizontal levels as marked A, B, C, and D on Fig.(5.4).

5.4.4.1. EFFECTS ON THE SPEED OF ROTATION

The concentration distribution curves are plotted in Fig. (5.53) to Fig.(5.61) at 20%, 35% and 52% of the corresponding critical speed of rotation. These concentration distribution curves show that the segregation occurs axially and radially. The axial segregation is similar to that shown in Fig.(5.17). The concentration with time profile at fixed positions for three different speeds of rotation are plotted in Fig.(5.62) to Fig.(5.64). The variance $\sigma^2(t)$ is plotted as a function of mixing time in Fig.(5.65) for three different speeds of rotation. The shape of these curves is similar and showed that the process is dominated by the segregation. At 20% and 35% of the corresponding critical speed of rotation, the mixing process was operated at two stages and an equilibrium state rapidly established. At a higher speed of rotation the curve is similar to those shown in Fig.(5.21) and the radial mixing was improved.

5.4.4.2. DISCUSSION OF THE RESULTS AND CONCLUSION

The experimental results shown in the diagrams only represent the concentration of inner part of the mixture, as the outer part of the mixture could not be sampled during the mixing operation. In general, the mixing behaviour was observed qualitatively. The large particles (lighter in weight) moved rapidly from one side of the mixer to other side of the mixer, and the small particles (heavier) were sandwiched. This phenomena may be explained as that :- as lighter particles can expand the bulk volume more easily than the heavier particles under the same speed of rotation. Thus the lighter particles move faster on the inclined surface than the heavier particles and are then rolled with the whole mass

to form a 'sleeve'. This as shown in Fig.(5.66)

large particles (90%)
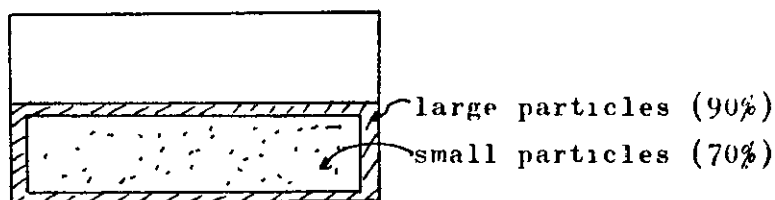
small particles (70%)

Fig.(5.66)

Clearly, this type of segregation is not that previously suggested (66, 67) as that the segregation occurs because the smaller heavier particles sink through the voids between the larger particles.

The axial segregation is independent of the speed of rotation, but radial mixing can be improved by increasing the speed of rotation. As can be seen in Fig.(5.61) at 52% of the critical speed of rotation, the profiles of the distribution curves, from four horizontal levels, are almost identical. In general, a higher speed of rotation is desired to achieve a better uniformity of the mixture. In such system, the mixing process is very complicated and will not generally be described by a stochastic process.

FIG. 5.53

AXIAL CONCENTRATION DISTRIBUTION OF Z-2 AT 20% C. S.R. FROM FOUR RADIAL POSITIONS   T = 10 MIN.
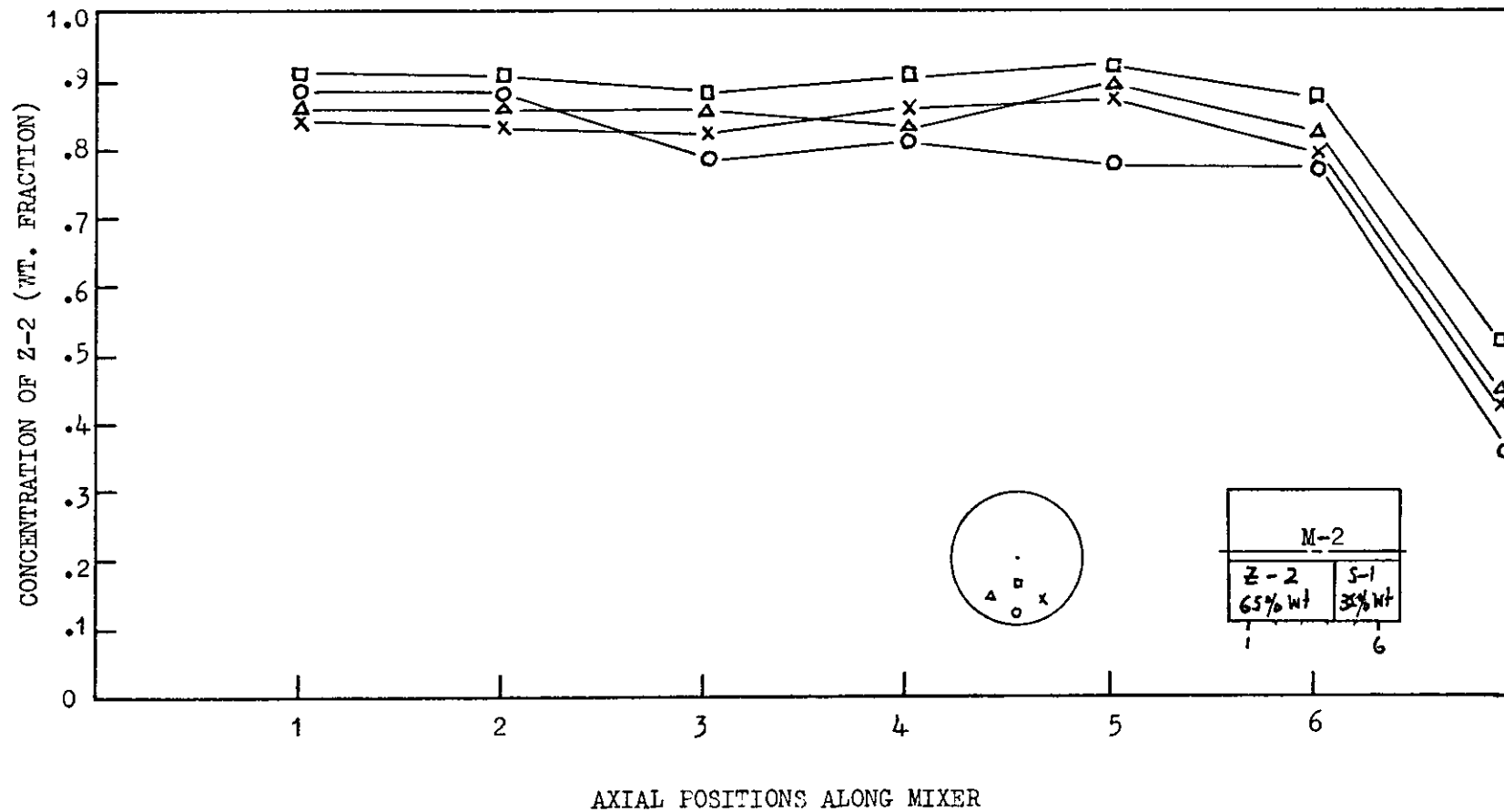
FIG. 5.54.

AXIAL CONCENTRATION DISTRATION OF Z-2 AT 20% C.S.R. FROM FOUR RADIAL POSITIONS T = 90 MIN.
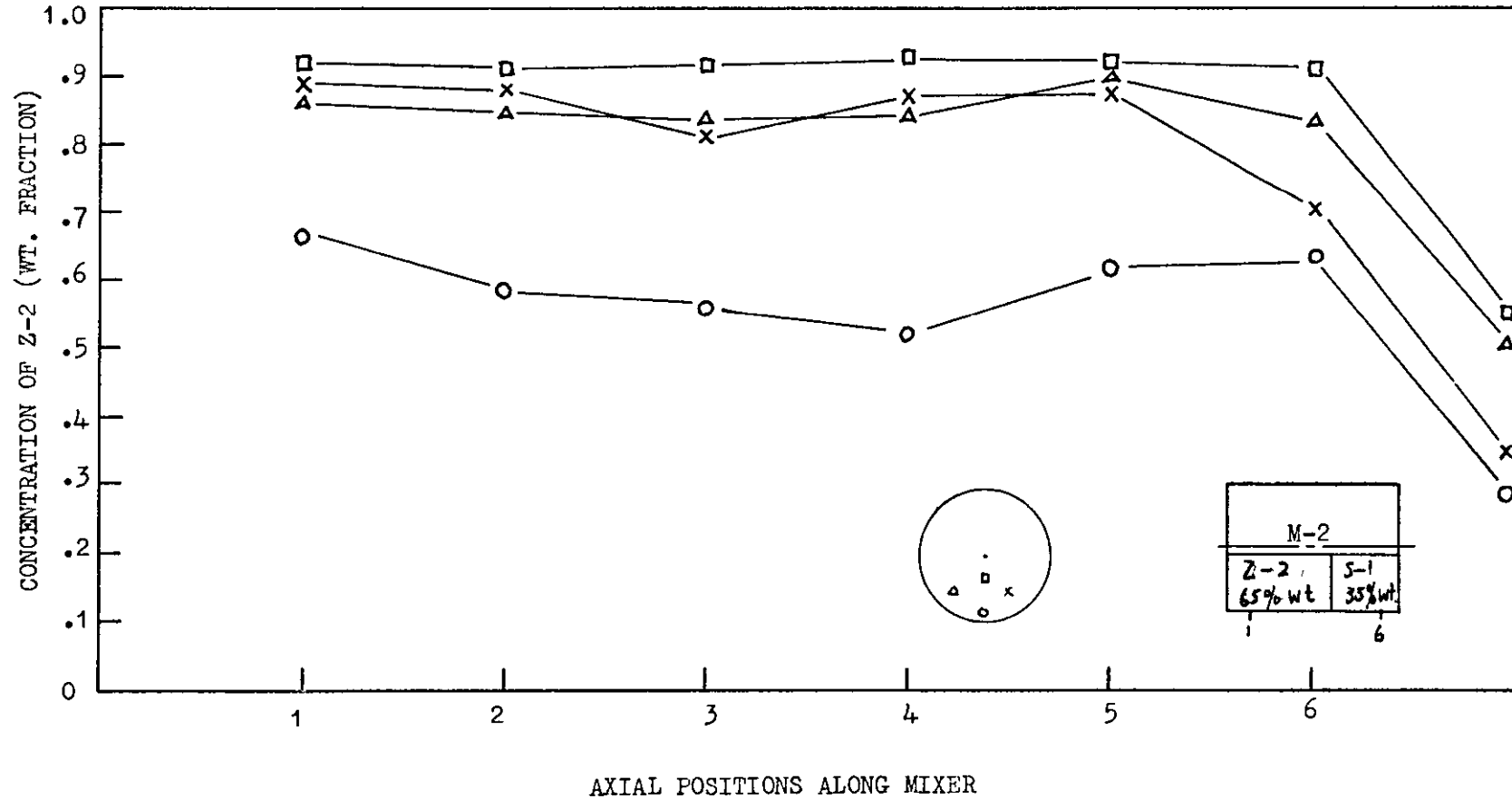
FIG.5.55.

AXIAL CONCENTRATION DISTRIBUTION OF Z-2 AT 20% C.S.R. FROM FOUR RADIAL POSITIONS   T = 600 MIN.
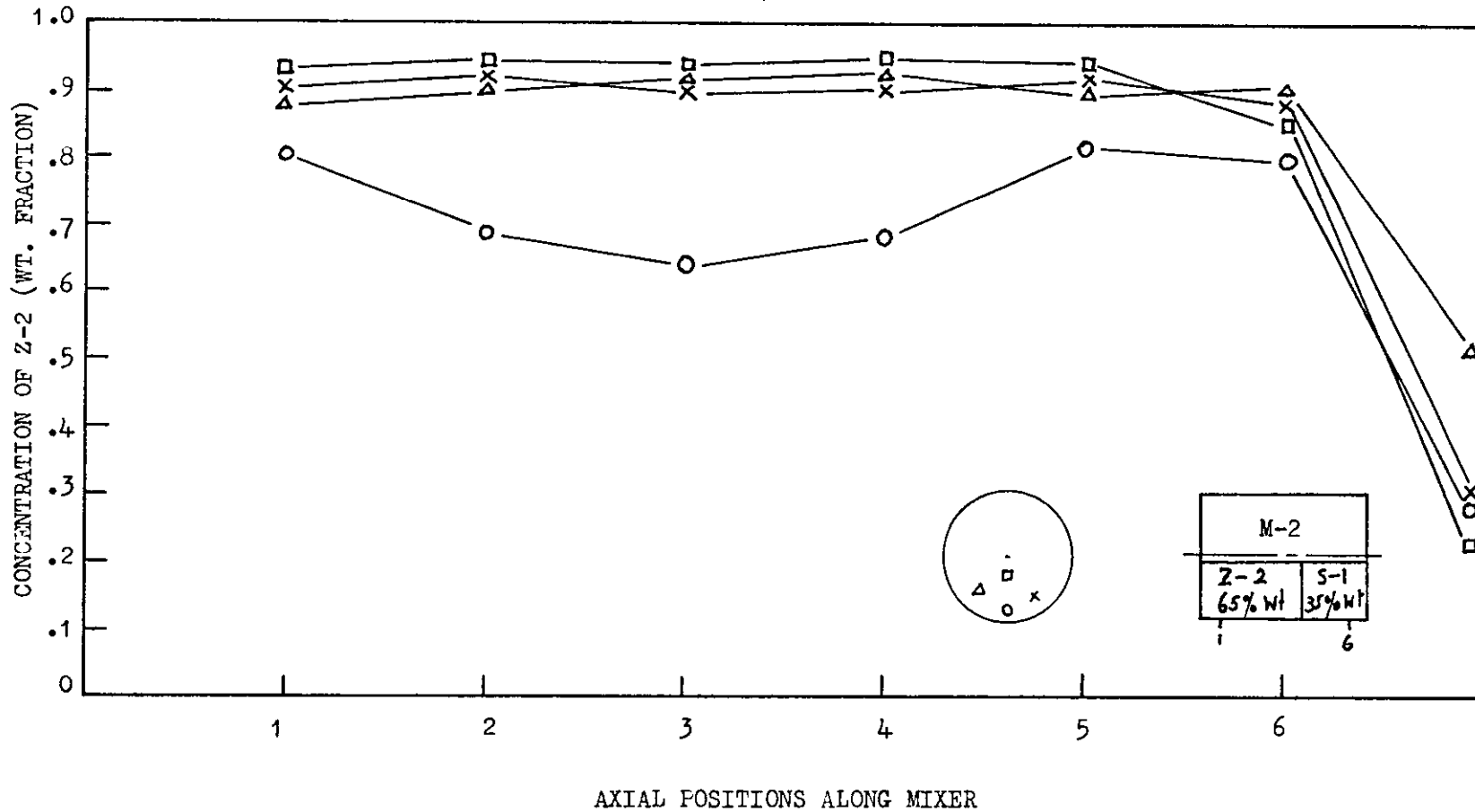
FIG. 5.56.

AXIAL CONCENTRATION DISTRIBUTION OF Z-2 AT 35% C. S. R. FROM FOUR RADIAL POSITIONS   T = 10 MIN.
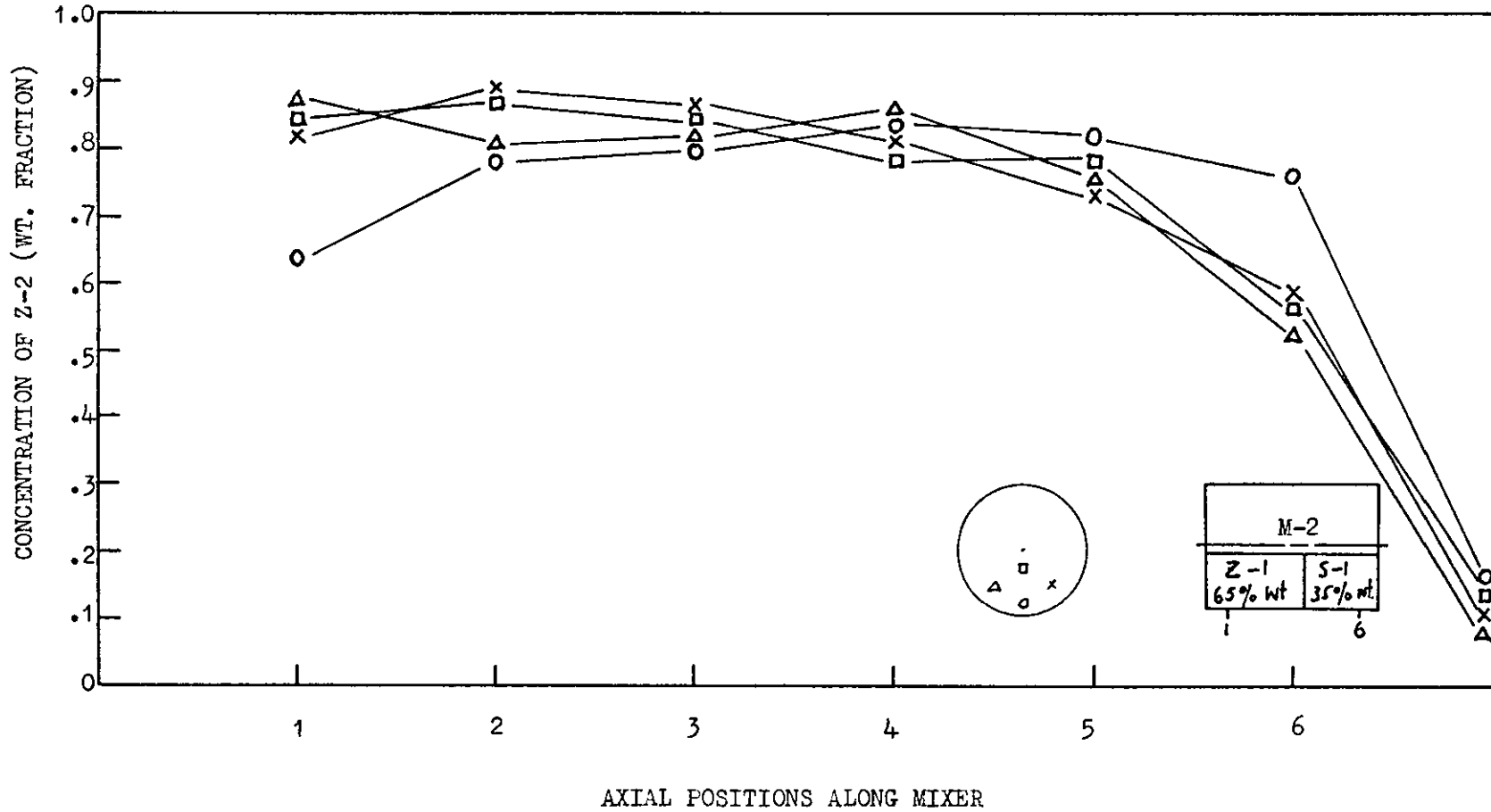
FIG. 5.5 7.

AXIAL CONCENTRATION DISTRIBUTION OF Z-2 AT 35% C. S. R. FROM FOUR RADIAL POSITIONS   T = 90 MIN.
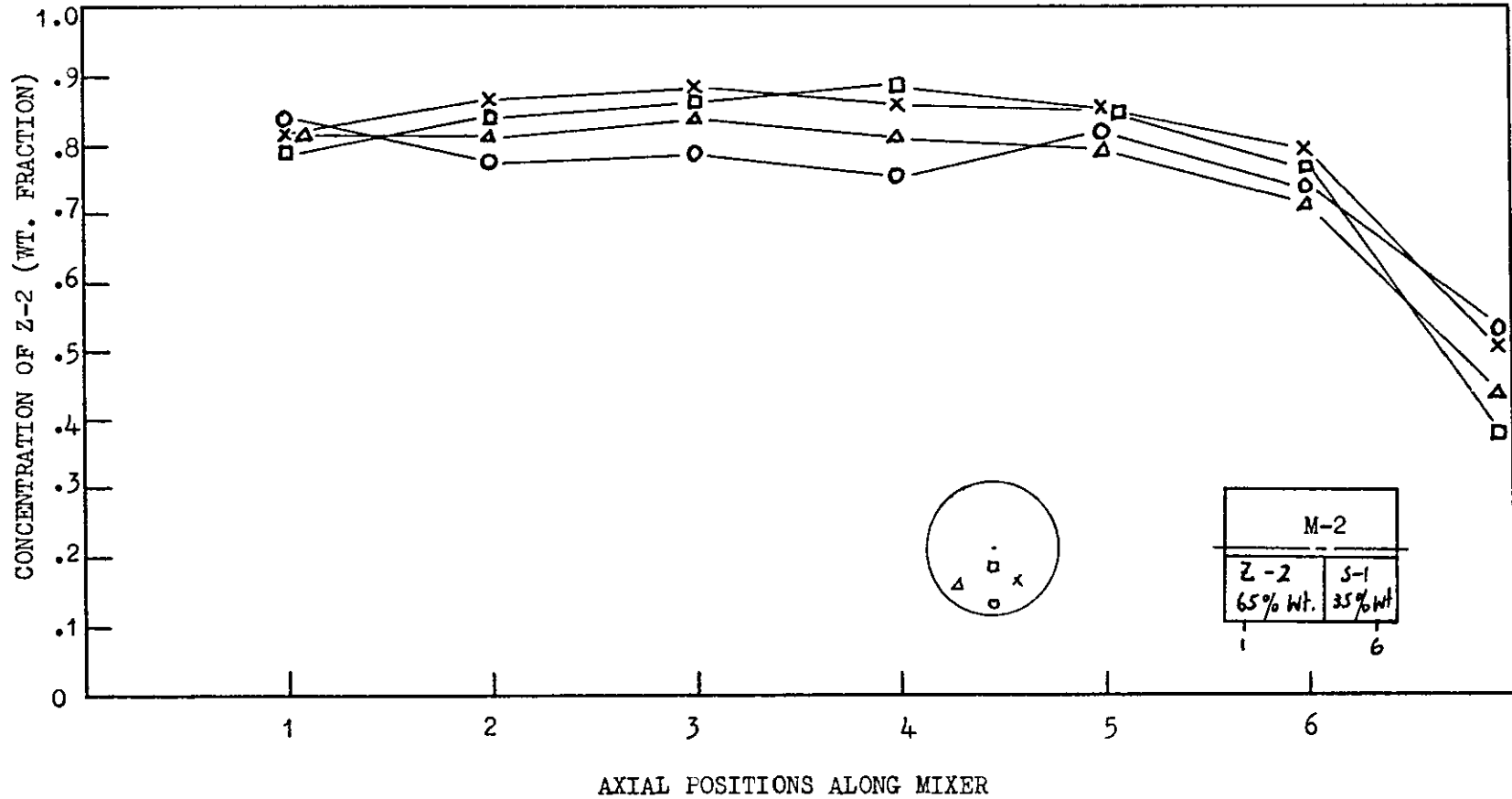
FIG. 5.58.

AXIAL CONCENTRATION DISTRIBUTION OF Z-2 AT 35% C.S.R. FROM FOUR RADIAL POSITIONS   T = 600 MIN.

191
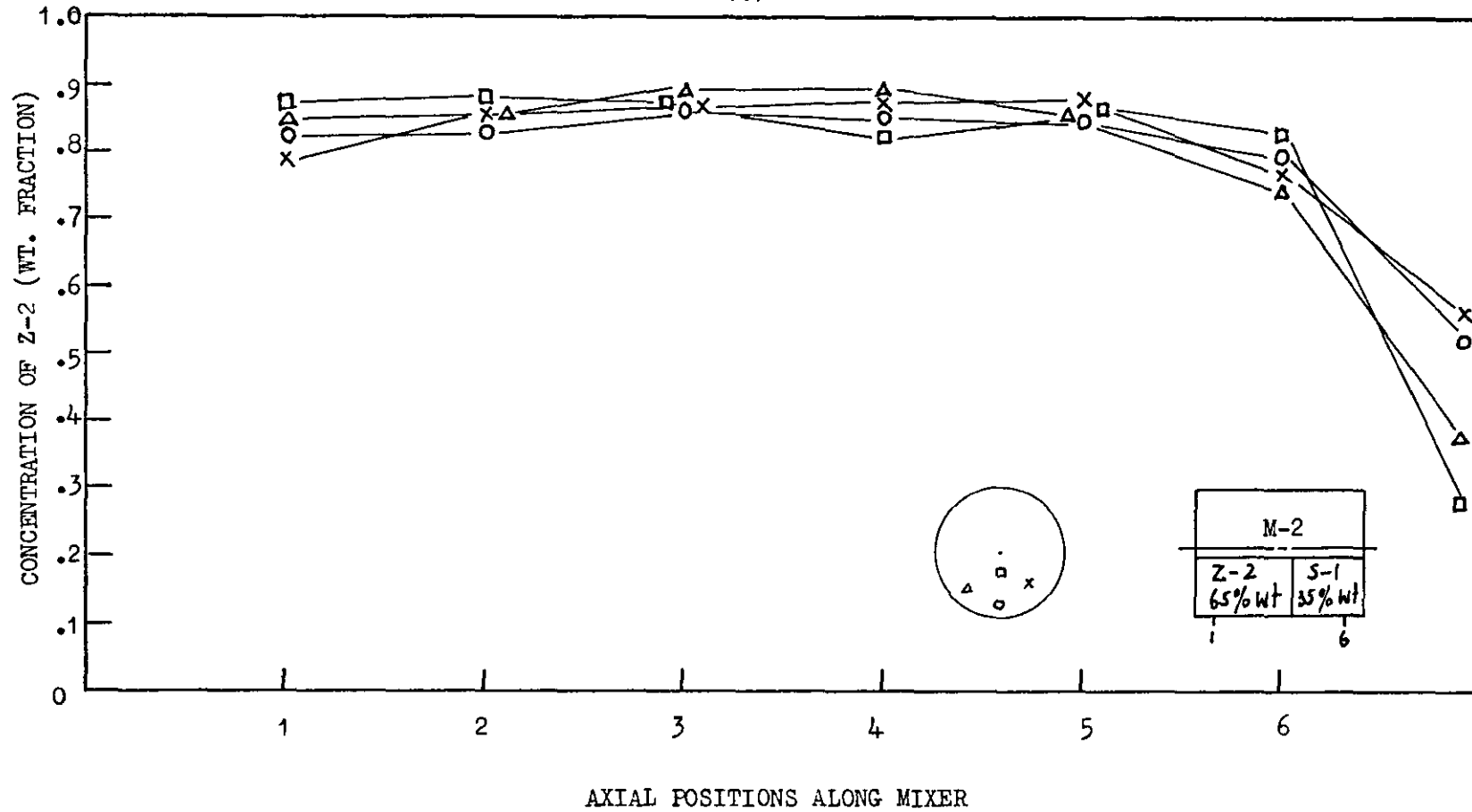
FIG. 5.59

AXIAL CONCENTRATION DISTRIBUTION OF Z-2 AT 52% C. S. R. FROM FOUR RADIAL POSITIONS T = 10 MIN.
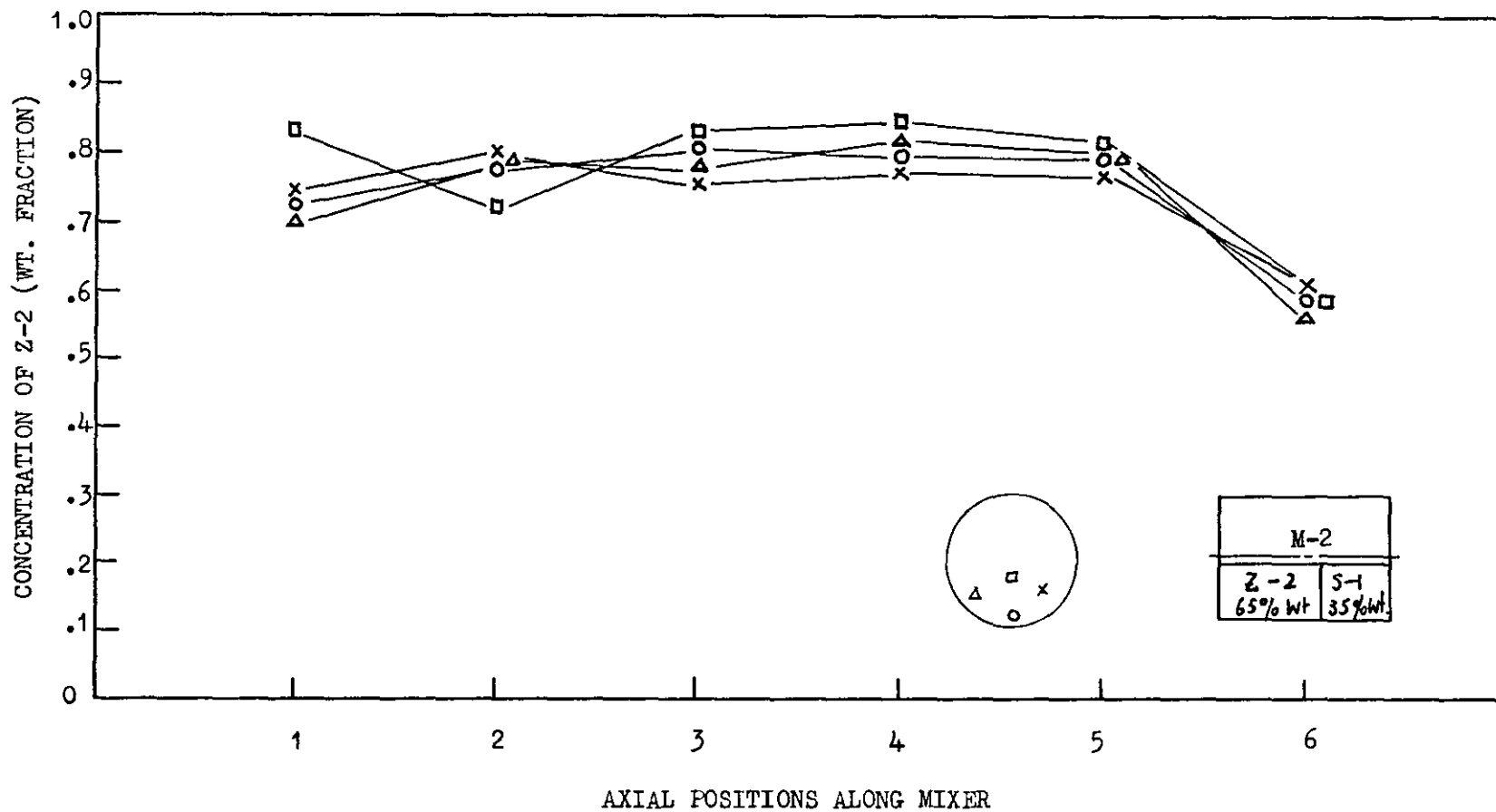
FIG. 5.60.

AXIAL CONCENTRATION DISTRIBUTION OF Z-2 AT 52% C. S. R. FROM FOUR RADIAL POSITIONS T = 90 MIN.
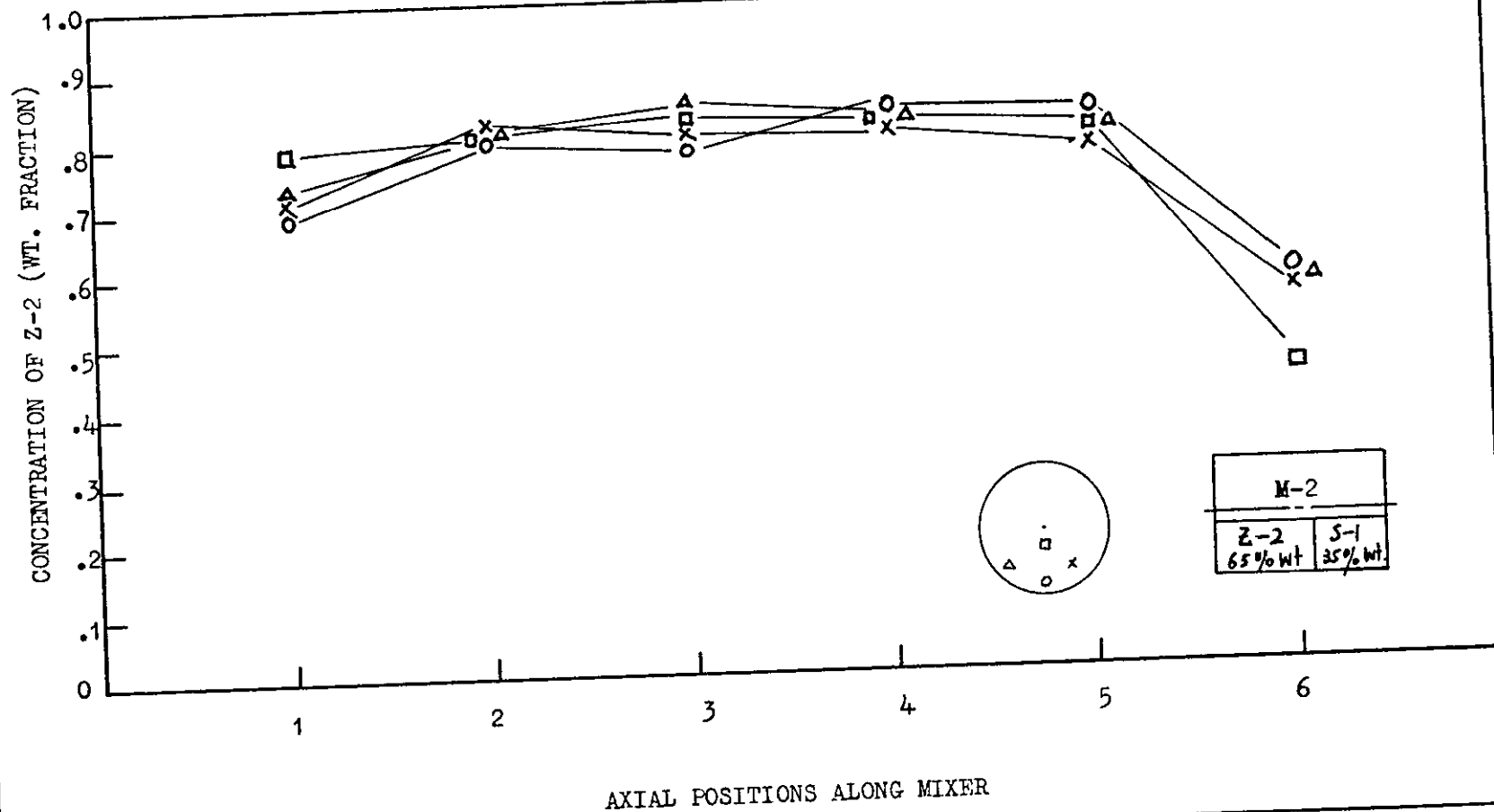
FIG. 5.61.

AXIAL CONCENTRATION DISTRIBUTION OF Z-2 AT 52% C.S.R. FROM FOUR RADIAL POSITIONS   T = 600 MIN.
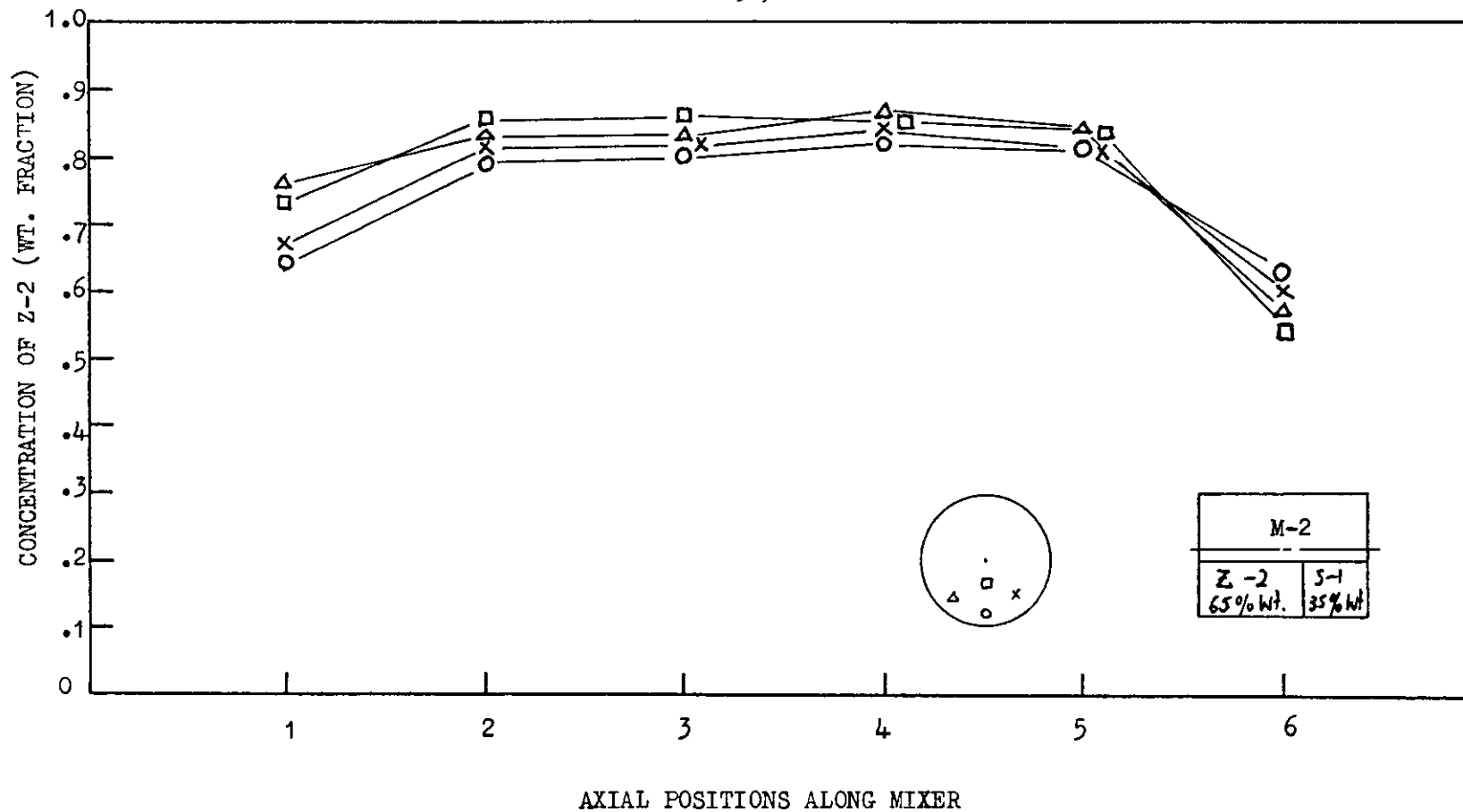
FIG. 5.62. CONCENTRATION VARIATION Z-2 AT 20% C.S.R. FROM POSITION o
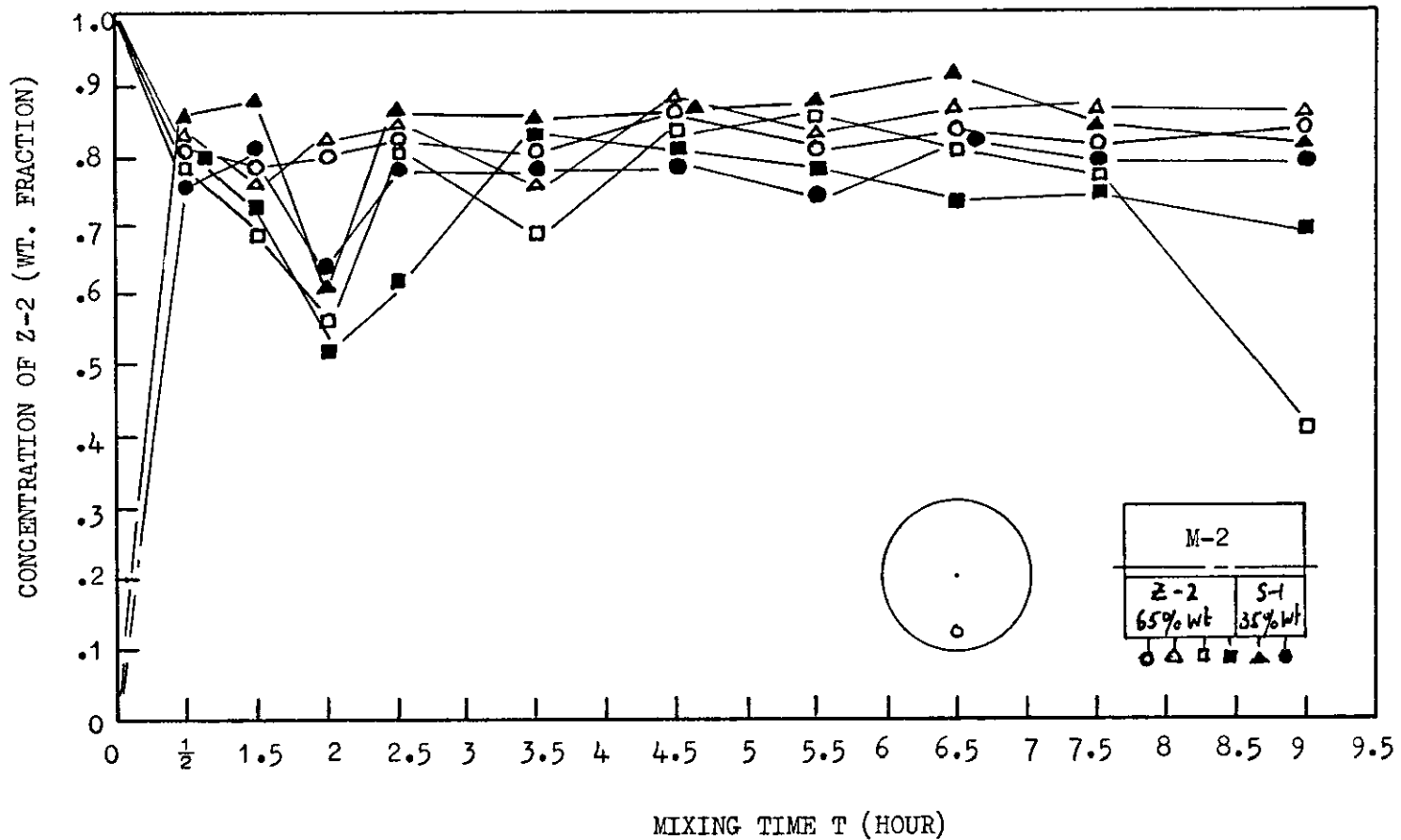
FIG. 5.63 CONCENTRATION VARIATION Z-2 AT 35% C.S.R. FROM POSITION □

CONCENTRATION OF Z-2 (WT. FRACTION)

AXIAL POSITIONS ALONG MIXER
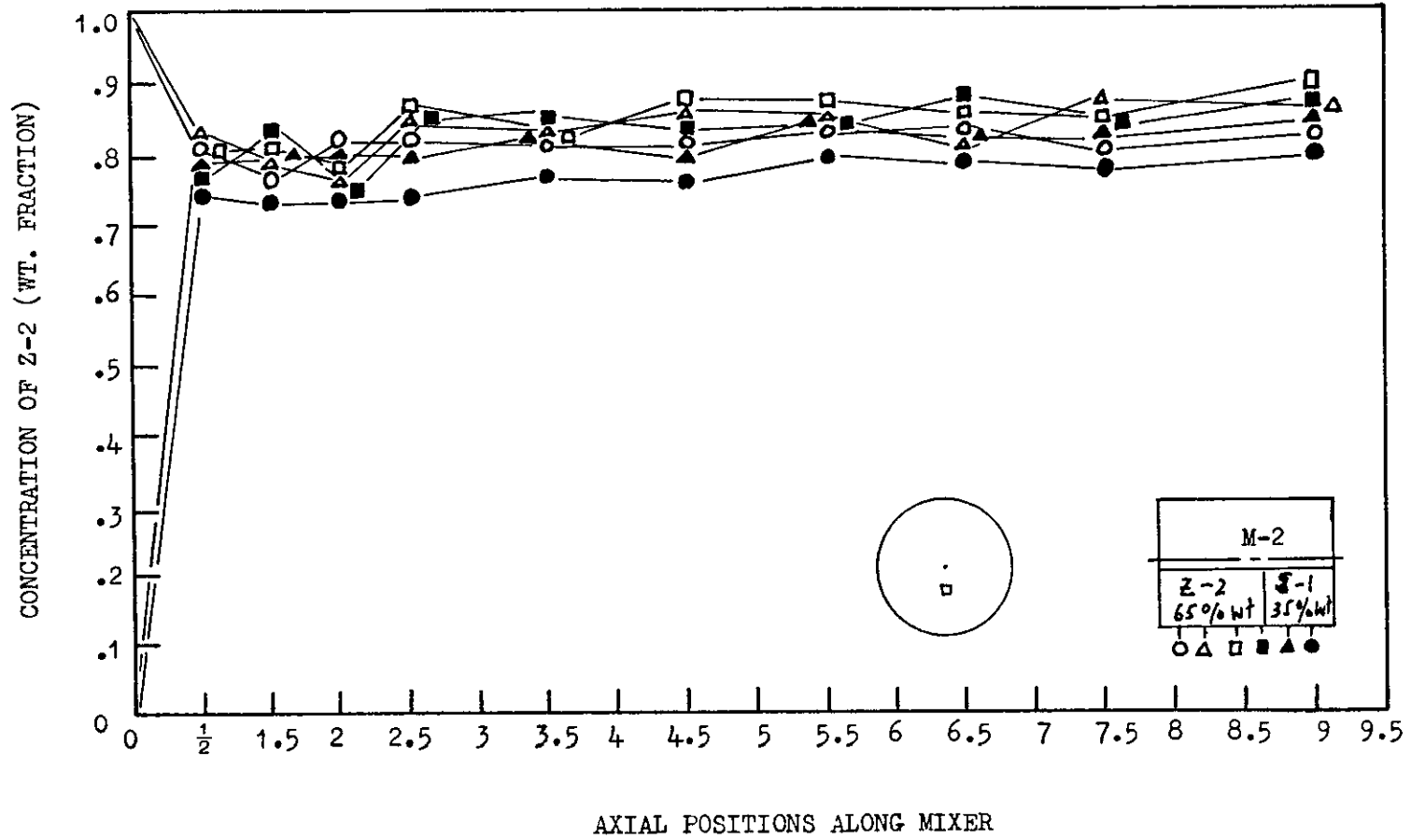
FIG. 5.64. VELOCITY DISTRIBUTION OF Z-2 AT 52% C.S.R. FROM POISTION o
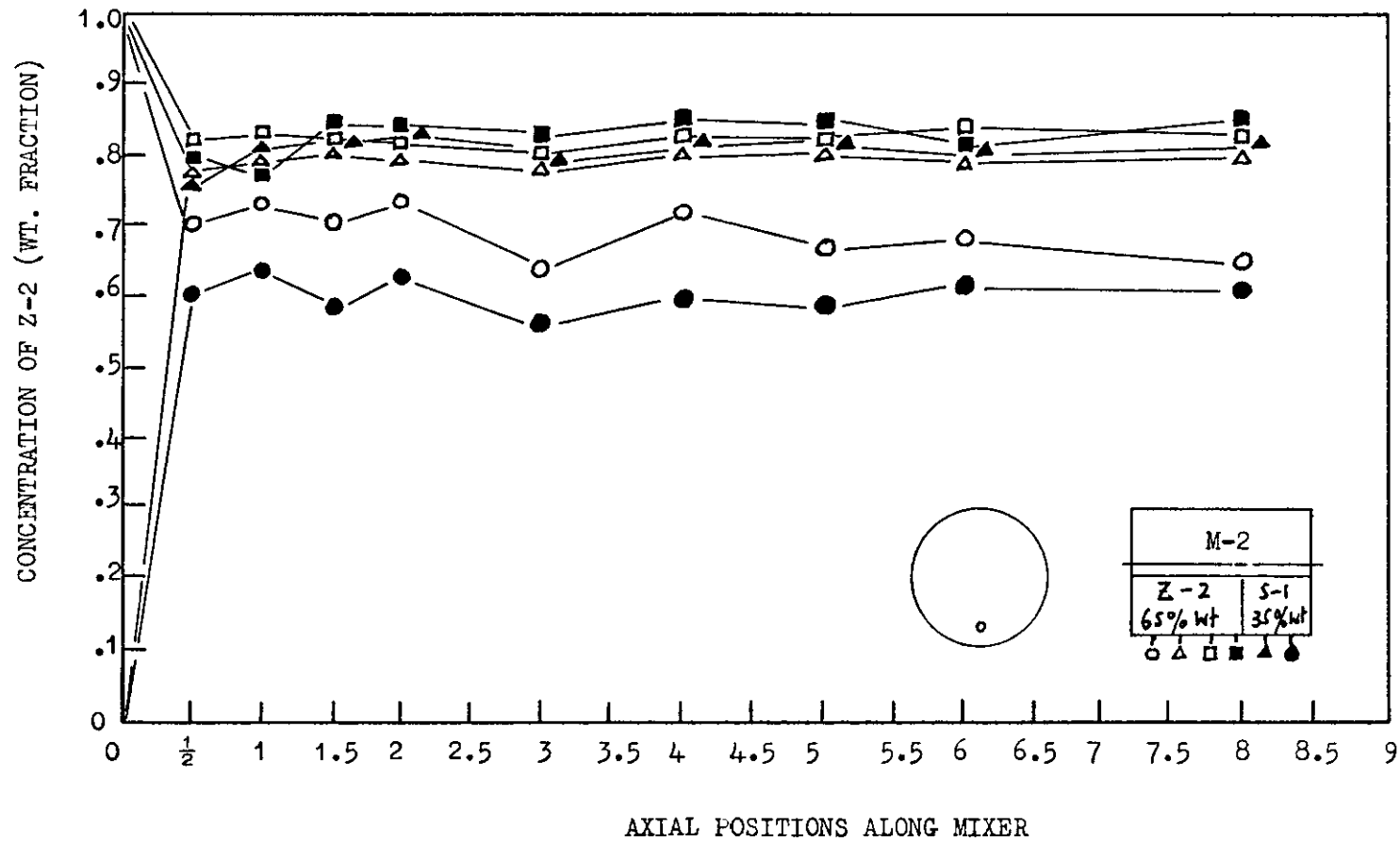
FIG. 5.65   SAMPLE VARIANCE $\sigma^2(t)$ VS. T

MIXING TIME T (HOUR)
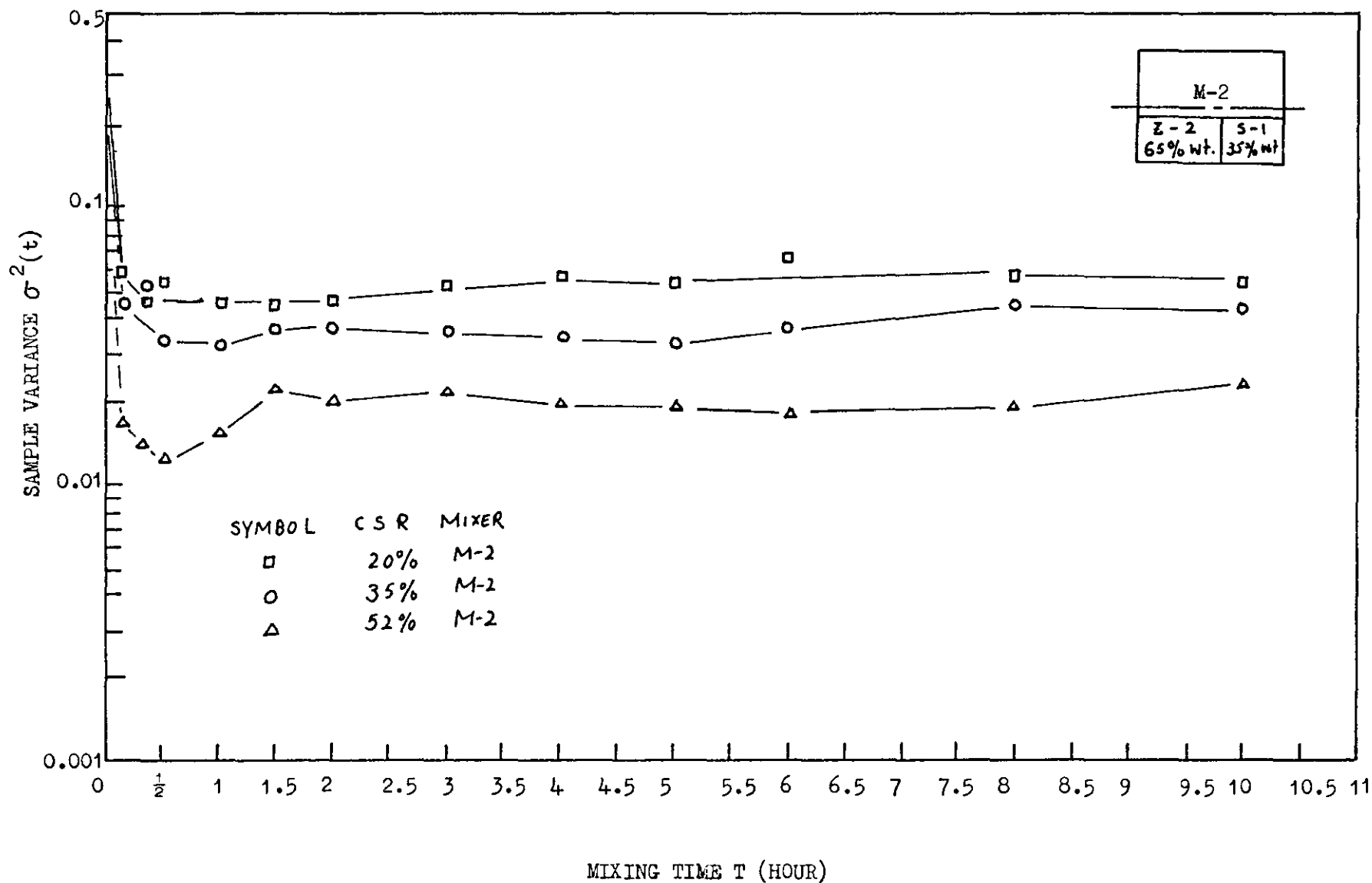
CHAPTER   SIX

FINAL   CONCLUSIONS

AND

SUGGESTIONS FOR FURTHER WORK

199

200

6.1. FINAL CONCLUSIONS

Conclusions have been drawn in each of the chapters and sections. They are summarised in the following :-

1. A stochastic analogy was used to study first principles of the nature of the mixing of free flowing particles. Two kinds of mixing equation have been developed to cover ;

    a.) mixing with components of physically identical particles,

    b.) mixing with components of same size and different density.

2. Sufficient experimental results have been obtained to show the first mixing equation is applicable to the mixing of components with physically identical particles. The rate of mixing is found to depend on the ratio of migration coefficient D to the square of the length of the mixer (D/ℓ^2). Moreover the migration coefficient D is independent of the size of mixer provided the speed of rotation it is kept at a constant proportion of the critical speed, but appears to be a function of this ratio.

3. When mixing components of different size but similar density a stable end condition is obtained in the form of bands of incomplete mixtures. These bands have near mono-size particle size distribution. The rotation of a wide size distribution showed that these bands are also formed when a continuous size distribution rather than a bimodal distribution is rotated in the drum.

4. A good mixture can be obtained when mixing components of different sizes, if the drum is rotated at angular speed at which porosity is a maximum.

5. When mixing particles of similar size and shape but small density difference, a good degree of randomisation can be obtained and the mixture tends to behave as if there was no density difference. However if the components have different shape, the end point of the mixture may be axial segregation.

6. When mixing components of large density difference, segregation occurs not only axially but also radially. A three dimensional stochastic equation might be able to describe such

a mixing system. This segregation is thought to be due to momentum difference discussed earlier.

7. Mixing with components of different size and different density. The mixing process is more complex than the system single variable change (different size or different density). But the pattern of segregation obtained is definite and is almost independent of the speed of rotation.

8. It has been found that the final state of mixedness obtained for any material combination is independent of mixer size.

It is believed that the present investigation has provided a better understanding and clearer definition on the mixing of free flowing powders in the horizontal drum mixer. Although a quantitative equation to cover mixing of all cases has not yet been developed, it is felt that sufficient experimental results have been obtained to provide a clue to justify the final states of mixedness.

6.2. SUGGESTIONS FOR FURTHER WORK

The present investigations have developed a general view of mixing and segregation of free flowing powders. Owing to the limited period of the present study, it was impossible to investigate the problem extensively and to develop a more general equation. Suggestions for further work is given in the following.

I. Extension of present investigation

1. The developed equations for mixing of free flowing powders have no limitation on what type of machine is used. As long the materials obey the basic postulates the equation may be applied. It is suggested, therefore that :-

i.) More experimental work is required in other types (other than rotating drum mixer) mixers for the materials which obey both postulates. The dependence of migration coeificient D on material properties. and machine types should be further investigated.

ii.) More experimental investigation is required on materials which obey first postulate but not the second. It has been shown that the final state of mixedness obtained depends largely on the shape of the particles. More work in this area would fulfil two purposes.

A.) The second equation has not been solved in the present work. This is mainly due to there being insufficient knowledge of particle movement to permit a firm boundary condition to be set up. If sufficient experimental results were available, this equation could be solved and the theory can be justified.

B.) Whilst it may be possible to control the sizes of different components to aid good mixing, it will not be possible always simultanously control the density of the material. So further experimental work is required to establish a more general approach which will enable the end condition to be realised and mixing time predicted.

2. Segregation mainly occurs in a mixture which consists of different sizes. However, the mechanism of separation and segregation in such a mixture was only postulated from the superficial

facts, the actual mechanism is still unknown. Therefore more work
is required to study the particle motion in such system. The
motion of particles may be traced by radioactive tracer or use
ultrasonic techniques. The pattern of motion of large and small
particles may be established. This can lead to greater understand-
ing of the mechanism of the separation and segregation in such
system. The second important area is the study of the stability
of particle ~~particle~~ packing in such systems, This is not only
important to particle mixing but also to many other aspects of
particle technology. The investigation may be studied theoretically
by two different routes, namely thermodynamics and statistical
mechanics. At the present time, the use of thermodynamics to study
a steady and mon-sized packing system has not yet been developed
but it is hoped that in time to come this approach will be beneficial.

II. Other fertile fields.

1. There are too few investigations on the continuous
mixing process. Since most physical-chemical processes have been
developed into continuous operation with automatic - on - line
control. It might be expected to be a providing development area,
more work is required on the continuous mixing process to develop
a working model, which will relate the material properties (e.g. size
density) feed rate and other factors, for an automatic control
system. One further advantage in the use of continuous mixing process
would be the avoiding of further handling of a 'seudo-randomised'
mixture.

2. The literature indicates that the mixing of fine powder
has been the subject of too few investigations. It is suggested
therefore a research programme (similar to the present study) should
be encouraged on the investigation of the mixing non-free flowing
powders,with particular reference to the material properties, to
obtain a general picture of the states of mixedness and to develop
a suitable mixing machine for such powders. It is also important
to develop a method which will enable easy analysis fine powder
samples.

3. It would be interesting to know the relationship be-
tween the final state of mixedness and power consumption in a

batch mixer. No reference to such study has been found in the literature.

4. It is also highly desirable to study the structure of mixture. This study may by useful to understand particles arrangements in a heterogeneous system and to test the definition of 'randomised' mixture. Sampling is generally employed to carry out such study. The samples taken from the mixture may not represent the true structure. Moreover the sampling probe may disturb the mixture. Methods must be desired which will enable the structure to be measured without disarranging the particles.

NOMENCLATURE

In the review ot the previous literature, the original authors' symbols have been used. Owing to use many mathematical formulae repeated the same letter may have made different meanings. Symbols have therefore seen defined as soon as they were introduced. It is felt that in this work such a system makes for a greater clarity and convenience.

REFERENCES

1. Adams, J.F.E.
   Baker, A.G.
   'An assessment of dry blending equipment'
   Trans.Instn.Chem.Engrs., 34, 1956

2. A. I. Chem. E.
   'Solid mixing equipment' A.I.Ch.E. Equipment testing procedure, 1966.

3. Ashton, M.D.,
   Valentin, F.H.H.
   'The mixing of powders and particles in industrial mixers' Trans.Inst.Chem.Engrs. 44, T166, 1966.

4. Ashton, M.D.,
   Schofield, C.,
   Valentin, F.H.H.
   'The use of a light probe for assessing the homogeneity of powder mixture' Chem. Eng. Sci., 21, 843/49, 1966.

5. Bailey, N.T.J.
   'The elements of stochastic processes with applications to the nature science' John Wiley & Sons Ltd., 1965.

6. Baker, C.N.,Jr.
   'Strength of soil-cement as a function of degree of mixing' Presented at 33rd annual meeting of Highway Research Board, Washington, D.C., Jan., 1954.

7. Barth, W.
   'Der Arbeitsverbrauch von Rohrmühlen' Techn. Mechan.u.Thermodynamik, 1, Nr. 9, 1930.

8. Beaudry, J.P.,
   'Blending efficiency' Chem.Eng., July, 1948.

9. Blumberg, R.,
   Maritz, J.S.
   'Mixing of solid particles' Chem.Eng.Sci., 2, 240, 1953.

10. Brothman, A,,
    Wollan, G.N.,
    Feldman, S.M.
    'New analysis provides formula to solve - mixing problem'. Chem.Mat.Eng., April, 1945.

11. Brown, R.L.
    'The fundamnetal principle of segregation' J.Inst.Fuel, 13, 15, 1939.

12. Burck, N.
    Chemistry and Industry, 21, May, 1966.

13. Buslik, D.
    'Mixing and sampling with special reference to multi-sized granular mixture' Bulletin ASTM, April, 1950 (TP92/99).

14. Cahn, D.S.
    'Simulation of diffusional mixing of particulate solids by Monte Carlo techniques' D.Eng. thesis, 1966, Univ. Calif. U.S.A.

30. Fischer, H.   'Der Arbeitvorgang in Kugelmühlen, insbe-
sondre in Rohrmühlen' V.D.I. 48, 437, 1904.

31. Fisher, R.K.   'Rate of diffusion mixing of particulate
solids' Ph.D. Thesis, Univ. Princeton, 1963.

32. Forscher, F.   'Analysis of continuity of one phase in a
powder mixture of two phases' J. Franklin Inst.
259, 1955.

33. Gayle, J.B.,
Lacey, O.L.,
Gray, J.H.   'Mixing of solids - Chi square as a criterion'
Ind.Eng.Chem. 50, 9, 1958, (1279/82).

34. Gayle, J.B.,
Gary, J.H.   'Mixing of solids with horizontal drum mixers'
Ind.Eng.Chem. 52, 6, 1960.

35. Gow, A.W.,
Campbell, A.B.,
Goghill, W.H.   'A laboratory investigation of ball milling'
Am.Inst.Min.Met.Eng. 326, 1930.

36. Gray, J.B.   'Dry solidz mixing equipment' Chem.Eng.Porgr.
53, 1, 1957.

37. Greathead, J.A.A.,
Simmonds, W.H.C.   'Mixing patterns in helical-flight dry solids
mixers' Chem.Eng.Progr. 53, 4, 1957.

38. Harnby, N.   'A comparison of the performance of industrial
solids mixers using segregating materials'
Powder Tech. 1, 2, 1967.

39. Herdan, G.,   'Small particle statistics' 2nd Ed. Butterworths.

40. Hill, F.P.,   'Mixing of polyethlene and carbon black master-
batch in double cone blenders' Trans. Instn.
Chem. Engrs. 43, 1965.

41. Hoffman, A.M.   'Radioactive tracer techniques in solid prope-
llant mixing' Ind.Eng.Chem. 52, 9, 1960.

42. Hogg, R., Cahn, D.S.
Healy, T.W.,
Fuerstenau, D.W.   'Diffusional mixing in an ideal system'
Chem.Eng.Sci. 21,1025/1038, 1966.

43. Iiyama, E.,
Aoki, R.   'On the torque characteristics of a paddle
type solid mixer' Kagaku Kogaku 24, 4, 1960.

15. Cahn, D.S., Fuerstenau, D.W., 'Simulation of diffusional mixing of particulate solids by Monte Carlo techniques' Powder Tech. 1, 3, 1967.

16, Cahn, D.S., Healy, T.W., Fuerstenau, D.W. 'Blender geometry in the mixing of solid' Ind. Eng.Chem. - Process Design and Dev. 4, 3, 1965.

17. Campbell, H., Bauer, W.C. 'Cause and cure of demixing in solid-solid mixers' Chem.Eng., Sept., 12, 1966.

18. Carley-Macauly, K.W. Donald, M.B. 'The mixing of solids in tumbling mixers' I Chem.Eng.Sci.,17, 1962, (493/506)

19. Idem, ibid., 19, 1964, (191/199)

20. Carley-Macauly, K.W. Ph.D. Thesis, Univ. London, 1958.

21. Coulson, J.M., Maitra, N.K. 'The mixing of solid particles' Ind.Chemist Feb., 1950.

22. Dallavalle, J.M. 'Micromeritics - the technology fine particles' 2nd Pitman Pub. Corporation N.Y., 1948.

23. Danckwerts, P.V. 'The definition and measurement of some characteristic of mixture' App.Sci.Res. A. 3, 1952.

24. Idem, 'Theory of mixture and mixing' Research (London) 6, 355, 1953.

25. Debbas, S., Rumpf, H. 'On the randominess of beds packed with spheres or irregular shaped particles' Chem.Eng.Sci. 21, 583/607, 1966.

26. Doob, J.L. 'Stchastic processes' John Wiley & Sons Inc. N.Y., 1960.

27. Dukes, J.A. 'An index of mixing' Intern.Chem.Eng. Nov., 521/523, 1951.

28. Earle, R.L. 'Some aspects of the mixing of plastic materials' Trans.Instn.Chem.Engrs. 37, T297, 1959.

29. Feller, W. 'An introduction to probability theory and its application' Vol 1, 2nd Ed. John Wiley & Sons Inc. N.Y., 1958.

44. Johnson, J.D. Discussion on the paper by Adams and Baker, Trans.Instn.Chem.Engrs. 34, T105, 1956.

45. Kac, M. 'Random walk and the theory of Brownian Motion' Am.Math.Monthly, 54, 369/391, 1947.

46. Kaye, B.H., Sparrow, D.B. 'Role of surface diffusion as a mixing mechanism in a barrel mixer' Ind.Chemist, April (part one) and May (part two) 1964.

46' King, G.W. 'Monte-Carlo method for solving diffusion problems' Ind.Eng.Chem. 43, 11, 1951.

47. Kun Sup Hyun, de Chazal, L.E.M. 'A statistical definition of perfect mixtures of solid of different sizes' Ind.Eng.Chem. Proc.Des.& Devel. 5, 2, 1966.

48 Lacey, P.M.C. 'The mixing of solid particles' Trans.Instn. Chem.Engrs. 21, 53, 1943.

49. Lacey, P.M.C. 'Development in the theory of particle mixing' J. Appl.Chem. 4, 257, 1954.

50. Lastovtsev, A.M. 'Mixers for free flowing solids' Translated by Bri.Chem.Eng., 8, 9, 1963.

51. Lloyd, P.J., Yeung, P.C.M. 'Mixing of powders' C.P.E. Oct., 1967.

52. Luikov, A.V., Mikhailov, Yu.A. 'The theory of energy and mass transfer' Pergamon Press, 1965.

53. Michaels, A.S., Puzinauskas, V. 'Evaluating performance characteristics of mechanical mixing process:-the dextrose-kaolinte -water system' Chem.Eng.Progr. 604/614, 1954.

54. Müller, Fr. 'Vermischung Körniger Faststoffmassen' Aufbereitungs – Technik, 5, 1966.

55. Müller, W., Rumpf, H. 'Das Mischen von Pulvern in Mischern mit axialer Mischkewegung' Chem.Ing.Tech. 39, 5/6, 1967.

56. Otake, T., Kitaoka, H., Tone, S. 'Mixing of solid particles in a cylindrical tank equipped with paddle type impellers' Kagaku kogaku, 25, 3, 1961. (In Japanese)

57.Oyama, Y.   'The motion of granular or pulverulent mate-
                                rials in a horizontal rotating cylinder,
                                report 2, mechanism of mixing and mixing rate'
                                Bull.Inst.Phys.Chem.Res.(Tokyo) 14,7, 570,
                                1935. (In Japanese).

58.Oyama, Y.   'Studies on mixing of solids' Sci Paper, Inst.
                                Phys.Chem.Res.(Tokyo) 37, 17, 1940.(In English).

59.Oyama, Y.   'Packing and mixing of broken solids by a
                                horizontal shaking motion, report 2, packing
                                of particles of two different sizes' Bull. Inst.
                                Phys.Chem.Res.(Tokyo) 19, 8, 1103, 1940. (In
                                Japanese).

60.Oyama, Y.   'The motion of granular or pulverulent ma-
                                terials in a horizontal rotating cylinder,
                                report 5' Bull.Inst.Phys.Chem.Res.(Tokyo)
                                18, 8, 600, 1939. (In Japanese).

61.Oyama, Y.   'The motion of granular or pulverulent mate-
                                rials in a horizontal rotating cylinder, report
                                6' Bull.Inst.Phys.Chem.Res.(Tokyo) 19, 8,
                                1070, 1940. (In Japanese).

62.Oyama, Y.,   'The mixing of particulate solids' Kagaku
    Ayaki, K.   Kogaku, 20,4, 148/155, 1956.(Translation
                                available from U.K.A.E.A.)

63.Poole, K.R.,   'Mixing powders to fine-scale homogeneity
    Taylor, R.F.,   studies of batch mixing' Trans.Instn.Chem
    Wall, G.P.   Engrs. 42, T305, 1964.

64.Poole, K.R.,   'Mixing powders to fine scale homogeneity
    Taylor, R.F.,   studies of continuous mixng' Trans Instn.
    Wall, G.P.   Chem.Engrs. 43, T261, 1965.

65.Rose, H.E.,   'A treatise on the internal mechanics of ball,
    Sullivan, R.M.E.   tube, and rod mills' Constable, London, 1958.

66.Rose, H.E.   'A suggested equation relating to the mixng'
                                of powders and its application to the study
                                of the performance of certain types of machine'
                                Trans.Instn.Chem.Engrs. 37, T47, 1959.

67. Roseman, B., Donald, M.B.   'Effects of varying the operating conditions of a horizontal drum mixer' I. Bri.Chem.Eng. 7, 10, 1962.

68. Idem   idib., 7, 11, 1962.

69. Roseman, B.   Ph.D. Thesis, Univ. London, 1962.

70. Rumpf, H., Müller, W.   'An investigation into the mixing of powders in centrifugal mixers' Trans.Instn.Chem. Engrs. 40, T272, 1962.

71. Rutgers, R.   'Lonitudinal mixing of granular material flowing through a rotating cylinder' Chem.Eng. Sci. 20, 1079/1087, 1965.

72. Idem.   idib., 20, 1089/1100, 1965.

73. Shinnar, R., Naor, P.   'A test of randomness for solid - solid mixture' Chem.Eng.Sci. 15, 220/229, 1961.

74. Shinnar, R., Katter, A., Stege, I.   'Experimental study of intimate mixing of solid particles' Chem.Eng.Sci. 18, 677, 1963.

75. Smith, J.C.   'Mixing chemicals with soil' Ind.Eng.Chem. 47, 11, 1955.

76. Stange, K.   'Gie Mischgute einer Zufallsmischung als Grundlage zur Beurteilung von Mischversuchen' Chem.Ingr.Tech. 26, 6, 1954. (Translation avialable from U.K.A.E.A.)

77. Stange, K.   'Die Mischgüte einer Zufallsmischung aus drei und mehr Komponenten' Chem.Ingr.Tech. 35, 8, 1963.

78. Strel'trov, V.V., et al   'Kinetics of mixing of granulated materials in commercial mixers' Khim.Prom. 11, 824,1964.

79. Sugimoto, M., Endoh, K.,   'Behaviour of granular materials flowing through a rotating cylinder' Kagaku Kogaku 30, 5, 1966. (In Japanese).

80. Tone, S., Otake, T.   'Mixing of granular solids in the cylindrical tank with paddle agitators' Kagaku Kogaku 30, 5, 1966. (In Japanese).

81.Visman, J.,   Ingenieur (Utrecht) 63, 42, 1944.
    Van Krevelen, D.W.

82.Weidenbaum, S.S.,   'A fundamental study of the mixing of parti-
    Bonilla, C.F.   culate solids' Chem.Eng.Progr. 51, 1, 1955.

83.Weidenbaum, S.S.   Ph.D. Thesis, Columbia University, 1953.

84.Weidenbaum, S.S.   'Mixing of solids' in Drew,T.B, J.W. Hooper,Jr.
                       'Advances in Chemical Engineering' Vol.11,
                       Acad.Press Inc. N.Y., 1958.

85.Weidenbaum, S.S.   'Mixing of solids ina twin shell blender'
    Corson, R.C.,   Ceramic Age, Aug., 1963.
    Miller, D.P.

86.Westman, A.E.R.,   'The packing of particles' J. Am. Ceram. Soc.
    Hugill, H.R.   13, 767, 1930.

87.Weydanz, W.   'Zeitlicher Ablauf eines Mischungsvorgange'
                       Chem.Ingr.Tech. 32, 343, 1960.

88.Williams, J.C.   Fuel Soc. J. Univ. Sheffield, 14, 29, 1963.

89.Yano, T.,Kanise,I., 'Mixing of powders by the V-type mixer'
    Tanaka, K.   Kagaku Kogaku 20, 4, 1956.(In Japanese)

90.Yano, T.,Kanise,I., 'Mixing of powders in several types of mixers'
    Hatano, Y.,   Kagaku Kogaku 21, 7, 1957. (Translation availa-
    Kurahashi, S.,   ble from U.h.A.E.A.).

91.Yano, T.,Kanise,I., 'Influence of the physical properties of
    Tanaka, K.,   powder on the optimum operating conditions,
    kurahashi, S.   mixing degree and mixing speed of several
                       types of mixers' Kagaku Kogaku 22, 12, 1958.
                       (Translation available from U.K.A.E.A.).

92.Yano,T.,Kanise,I., 'The influence of particle size of powders
    Sano,Y.,Tsutsumi,M.,on the optimum operating conditions, degree
    Okamoto, Y.   and rate of mixing in several types of mixers'
                       Kagaku Kogaku, 23, 9, 1959.(Translation avai-
                       lable from U.h.A.E.A.).

93.Yano, Y.,   'The effect exerted by various factors on the
    Kanise,I.,Sano,Y. coefficients of mixing velocity in powder
                       mixers' Kagaku Kogaku 24, 4, 1960.(Translation
                       available from U.K.A.E.A.).

94.Yano,T., Sano, Y.,   'Mixing of solid particles using a ribbon
   Shirakawa, U.,   mixer' Kagaku Kogaku 24,4, 1960. (Transla-
   Tominaga, T.   tion available from U.K.A.E.A.).

APPENDICES

APPENDIX I

EXPERIMENTAL RESULTS

Run : R-SIII-2

Conditions : Mixer : M-2   Speed of rotation : 52%
Material : Sand S-1   Size : -36/+52
Density : 2.58 gm/ml.   Loading : end to end
Fullness : 40%   Ratio : 50/50

Mixing Time (Min.)	Mean Concentration	R(t)	$\sigma^2(t)$
5	0.563	0.502	0.145
10	0.552	0.480	0.062
15	0.504	0.255	0.034
20	0.521	0.178	0.0185
25	0.507	0.0981	0.00756
30	0.501	0.0763	0.00388
35	0.503	0.0543	0.00232
40	0.492	0.0351	0.00120
45	0.498	0.0344	0.00124
50	0.502	0.0306	0.00125
55	-	-	-
60	0.489	0.0340	0.00138
80	0.497	0.0354	0.00122
100	0.487	0.0352	0.00143
120	0.504	0.0364	0.00146

Run :   R-ASI-1

Conditions :   Mixer : M-2   Speed of rotation : 33%
                  Material : Sand S-1   Size : -36/+52
                  Density : 2.58 gm/ml. Loading : end to end
                  Fullness : 40%   Ratio : 50/50

Mixing Time (Min.)	Mean Concerntration	R(t)	$\sigma^2(t)$
5	0.478	0.502	0.0845
10	0.489	0.268	0.0386
15	0.498	0.178	0.0210
20	0.4985	0.138	0.0103
25	0.500	0.0703	0.00410
30	0.502	0.0435	0.00240
35	0.502	0.0321	0.00106
40	0.504	0.0302	0.00108
45	-	0.0311	0.00135
50	0.511	0.0330	0.00124
60	0.520	0.0350	0.00135
80	0.504	0.0297	0.00134
100	0.497	0.0346	0.00125
120	0.506	0.0326	0.00130
140	0.504	0.0305	0.00138

Run :   R–SVI–1

Conditions :   Mixer : M–1   Speed of rotation : 20%
                Materials : Sand, S–1, S–2 Size : –36/+52, –52/+60
                Density : 2.58 gm/ml.   Loading : end to end
                Fullness : 40%   Ratio : 50/50 (wt.)

Mixing Time (Min.)	Mean Concentration of S–1 by wt. fr.	$\sigma^2(t)$
30	0.476	0.0612
60	0.493	0.0458
90	0.483	0.0362
120	0.502	0.0302
150	0.507	0.0281
180	0.473	0.0250
210	0.498	0.0212
240	0.479	0.0229
270	0.452	0.0259
300	0.490	0.0362
330	0.494	0.0303
360	0.483	0.0330
390	0.478	0.0342
420	0.498	0.0343
480	0.509	0.0366
580	0.473	0.0385

Run :   R-SVI -2

Conditions :   Mixer : M-1   Speed of rotation : 35%

Materials : Sand, S-1, S-2,   Size : -36/+52, -52/+60

Density : 2.58 gm/ml.   Loading : end to end

Fullness : 40%   Ratio : 50/50 (wt.)

Mixing Time (Min)	Mean Concentration of S-1 by Wt.fr.	$\sigma^2(t)$
15	0.476	0.054
30	0.483	0.0382
60	0.482	0.035
90	0.486	0.0372
120	0.498	0.0286
150	0.488	0.0247
180	0.512	0.0240
210	0.497	0.0226
240	0.478	0.0182
300	0.504	0.0160
360	0.511	0.0073
420	0.496	0.0070
480	0.488	0.0068
540	0.504	0.00502
600	0.500	0.00460

Run :   R-SVll-4-2

Conditions :   Mixer : M-2   Speed of rotation : 52%

Materials : Sand, S-1, S-2   Size : -36/+52, -52/+60

Density : 2.58 gm/ml.   Loading : end to end

Fullness : 40%   Ratio : 50/50 (wt.)

Mixing Time (Min)	Mean Concentration of S-1 by Wt. fr.	$\sigma^2(t)$
15	0.475	0.0251
30	0.496	0.0148
60	0.501	0.0132
90	0.457	0.0136
120	0.498	0.0172
180	0.463	0.0203
240	0.501	0.0230
330	0.476	0.0240
420	0.483	0.0272
510	0.495	0.0320
600	0.494	0.0330

Run : R-SVII-6

Conditions :   Mixer : M-2   Speed of Rotation : 52%
               Materials : Sand, S-1, Gypsum.Size : -36/+52
               Density : 2.58, 2.14 gm/ml. Loading : end to end
               Fullness : 40%   Ratio : 50/50 (vol.)

Mixing Time (Min.)	Mean Concentration of S-1	R(t)	$\sigma^2(t)$
5	0.517	0.302	0.0402
10	0.512	0.245	0.0214
20	0.502	0.207	0.0195
30	0.492	0.182	0.00905
40	0.498	0.134	0.00760
50	0.504	0.078	0.00360
60	0.503	0.0565	0.00272
70	0.506	0.0335	0.00183
80	0.500	0.031	0.00160
100	0.498	0.0362	0.00193
120	0.506	0.0345	0.00202
140	0.497	0.0302	0.00196
160	0.513	0.035	0.00198

Run :　R-SX-2

Conditions :　Mixer : M-2　　Speed of rotation : 52%
Materials : Sand S-1, Glass Beads. Size : -36/+52
Density : 2.58, 2.89 gm/ml.　Loading : end to end
Fullness : 40%　Ratio : 50/50 (vol.)

Mixing Time (Min)	Mean Concentration of Glass Beads	$\sigma^2(t)$
5	0.469	0.0385
10	0.492	0.0176
20	0.497	0.0122
30	0.486	0.0083
40	0.495	0.0046
50	0.499	0.00572
60	0.490	0.00788
80	0.486	0.00904
100	0.496	0.0105
120	0.476	0.00962
160	0.486	0.0110
220	0.484	0.0132

APPENDIX   II

II.1. Derivation of the most probable value of the mean square errors of an individual measurements $\sigma^2(1)$

In the derivation of the most probable value of a set measured quantity, it is assumed that the quantity σ was known. Therefore the probability of obtaining a certain set of errors (which is equal to the probability of obtaining a certain set of measures results) depend only on the hypothesis made regarding the variable a. For the generalised case, it considers that the quantity σ is also unknown, therefore to examine the probability of the set of errors, it needs to make hypothesis regarding the value of both a and . The probability of finding the results

$$x_1, \ x_2, \ x_3, \ \ldots \ x_N$$

under the given hypothesis may be expressed by formular

$$P(x \ x_1, \ x_2, \ \ldots \ x_N | a, \sigma \ ) = (\frac{\Delta\delta}{\sqrt{2\pi}})^N \sigma^{-N} \exp\left[\frac{-S(a)}{2\sigma^2}\right] \ (1)$$

Where,

$$S(a) = \sum_{i=1}^{N} (x_1 - a)^2 \tag{2}$$

Rewritten the quantity $S(a)$ as follows,

$$S(a) = \sum_{i=1}^{N} (x_i - a)^2 = \sum_{i=1}^{N}\left[(x_1 - \bar{x}) + (\bar{x} - a)\right]^2$$

$$= \sum_{i=1}^{N} (x_1 - \bar{x})^2 + 2(\bar{x} - a)\sum_{i=1}^{N} (x_1 - \bar{x}) + N(\bar{x} - a)^2 \tag{3}$$

The arithmetic mean of set of measurement,

$$\bar{x} = \frac{\sum_{i=1}^{N} x_1}{N} \tag{4}$$

So,

$$\sum_{i=1}^{N} (x_1 - \bar{x}) = 0 \qquad (\text{if } \bar{x} \text{ is exactlt computed}) \tag{5}$$

Therefore,

$$S(a) = \sum_{i=1}^{N} (x_i - \bar{x})^2 + N(\bar{x} - a) \tag{6}$$

The probability of the set of measurements could then be written in the form of

$$P(x \ x_1, \ x_2 \ldots \ x_N | \ a, \sigma \ )$$

$$= (\frac{\Delta\delta}{\sqrt{2\pi}})^2 \frac{\sqrt{2\pi}}{\sqrt{N}} \sigma^{-N+1} \exp\left[-\frac{\sum_{i=1}^{N}(x_1 - \bar{x})^2}{2\sigma^2}\right]\left\{\frac{1}{\sqrt{2\pi}\frac{\sigma}{\sqrt{N}}} \exp\left[\frac{-(a-\bar{x})^2}{2(\frac{\sigma}{\sqrt{N}})^2}\right]\right\}$$

$$\tag{7}$$

Where, the desired probability is a function of the two independent parameters a and .

To obtain a rule for finding the most probable value of that it is suitable for all value of a, let one assumes that all values of a are equally probable in some region say form $-\alpha$ to $+\alpha$ that is large enough for one to be able to consider it as practically equivalent to the interval from $-\infty$ to $+\infty$. For this, it is sufficient that α be a magnitude of the order of $3-4\,\sigma$.

Let one multiply both sides of Eq.(7) by d(a) and integrates with respect to a from $-\infty$ to $+\infty$. From the generalised theorem on the addition of probabilities, one obtains on the left side the probability of the set of measurements for all values of a with a fixed σ, on the right side, one needs to integrate only the last factor as it is the only one containing a, this factor is the normal probability density of the random variable a, so that, its integral from $-\infty$ to $+\infty$ is unity and obtained

$$P(x \quad x_1, x_2, \ldots x_N / \sigma) = \left(\frac{\Delta\delta}{\sqrt{2\pi}}\right)^N \frac{\sqrt{2\pi}}{\sqrt{N}} \exp\left(-\frac{\bar{s}}{2\sigma^2}\right) \sigma^{-N+1} \quad (8)$$

where,

$$\bar{s} = \sum_{i=1}^{N} (x_i - \bar{x})^2 \quad (9)$$

Accordingly this equation could be rewritten as,

$$P(\sigma \mid x_1, x_2, \ldots x_N) = C\, \sigma^{-N+1} \exp\left(-\frac{\bar{s}}{2\sigma^2}\right) \quad (10)$$

Where the coefficient C includes the proportionality constant appearing in the corallary of Bayer's formula and as well as all the factors which do not depend on .

The most probable value of   in a set of measurements is the value at which the function

$$P(\sigma) = \sigma^{-N+1} \exp\left(-\frac{\bar{s}}{2\sigma^2}\right) \quad (11)$$

has a maximum.

Logarithm of both sides of Eq.(11) and differentiate twice with respect to σ, one has,

$$\ln P = (1-N)\ln - \frac{\bar{s}}{2\sigma^2} \quad (12)$$

$$\frac{1}{P} \frac{dP}{d\sigma} = \frac{1 - N}{\sigma} + \frac{\bar{s}}{\sigma^3} \qquad (13)$$

$$-\frac{1}{P} \frac{dP}{d\sigma} + \frac{1}{P} \frac{d^2P}{d\sigma^2} = \frac{N - 1}{\sigma^2} - \frac{3\bar{s}}{\sigma^4} \qquad (14)$$

Let Eq.(13) equal to zero, so,

$$\sigma^2 = \frac{\bar{s}}{N - 1} \qquad (15)$$

Substitute Eq.(15) into Eq.(14), thus,

$$\frac{1}{P}\left(\frac{d^2P}{d\sigma^2}\right) = -2 \frac{(N - 1)^2}{\bar{s}} < 0$$

Where confirms that the obtained value of   is the most probable one, Hence,

$$\sigma^2 = \frac{\sum (x_1 - \bar{x})^2}{N - 1} \qquad (16)$$

II.2. Derivation of the randomised variance $\sigma_R^2 (2)$.

The derivation of a randomised variance of a set of measurements from a ideal mixture, one needs to assume few points. If the overall fraction of a binary component in the whole mixture is $\bar{p}$ of A-component particles and $\bar{q}$ of B-component particles $(\bar{p} + \bar{q}) = 1$, N samples withdrawn from the mixture at a time and each sample contains n particles of p fraction of A-component and q fraction of B-component in the sample, i.e. $(p + q) = 1$.

So that the variance of N samples will be Eq.(16) or,

$$\sigma^2 = \frac{\sum (p - \bar{p})^2}{N - 1} \qquad (16')$$

for large value of N, Eq.(16') becomes,

$$\sigma^2 = \frac{\sum (p - \bar{p})^2}{N} \qquad (17)$$

Rewritten Eq.(17) becomes,

$$\sigma^2 N = \sum (p - \bar{p})^2 = \sum p^2 - 2 \bar{p} \sum p + \bar{p}^2 \qquad (18)$$

For n particles containing p and q, the frequencies of successes in N samples are given by the successive terms in the binomial expension of $(q + p)^n$, i.e.,

$$N\left[q^n + n\, q^{n-1}p + \frac{n(n-1)}{1.2}\, q^{n-2}p^2 + \frac{n(n-1)(n-2)}{1.2.3}q^{n-3}p^3 +\ldots\ldots\right]$$

$$(19)$$

The possible value of p in a sample of n particles are

$$0,\ \frac{1}{n},\ \frac{2}{n},\ \frac{3}{n},\ldots\ldots$$

For obtaining of $\sum p^2/N$, is multiplying the succesive terms in the series of Eq.(19) by

$$0,(\tfrac{1}{n})^2,\ (\tfrac{2}{n})^2,\ (\tfrac{3}{n})^2,\ \ldots\ldots\ldots$$

and summing up the results, i.e.,

$$\frac{\sum p^2}{N} = \frac{1}{n^2}\left[n\, \bar{q}^{n-1}\bar{p} + 2n(n-1)\bar{q}^{n-1}\bar{p}^2 + \frac{3}{2}(n-1)(n-2)\bar{q}^{n-3}\bar{p}^3 +\ldots\ldots\right]$$

or, $\quad \dfrac{\sum p^2}{N} = \dfrac{\bar{p}}{n}\left[\bar{q}^{n-1} +, 2n(n-1)\bar{q}^{n-2}\bar{p} + \frac{3}{2}(n-1)(n-2)\bar{q}^{n-3}\bar{p}^2 +\ldots\right]$ (20)

Eq.(20) may be rewritten as,

$$\frac{\sum p^2}{N} = \frac{\bar{p}}{n}\left[\bar{q}^{n-1} + (n-1)\bar{q}^{n-2}\bar{p} + \frac{1}{2}(n-1)(n-2)\bar{q}^{n-3}\bar{p}^2 +\ldots\ldots\right]$$

$$+\ \frac{\bar{p}}{n}\left[0 + (n-1)\bar{q}^{n-2}\bar{p} + (n-1)(n-2)\bar{q}^{n-3}\bar{p}^2 +\ldots\ldots\right]\ (21)$$

Examining Eq.(21), one finds that the first series is containing a result of $(\bar{q} + \bar{p})^{n-1}$ and therefore its is 1, since $(\bar{p} + \bar{q}) = 1$. The second series is the same one with successive terms multiplied only by $0,1,2,\ldots$ i.e. by successive values of $(n-1)p$, and its sum is therefore the mean value of $(n-1)p$, i.e., $(n-1)\bar{p}$. Thus,

$$\frac{\sum p^2}{N} = \frac{\bar{p}}{n}\,(1) + \frac{\bar{p}}{n}\,(n-1)\bar{p} = \frac{\bar{p}}{n}\left[1 + (n-1)\bar{p}\right] \qquad (22)$$

Hence, $\qquad \sigma^2 = \sum p^2/N - \bar{p}^2 = (\bar{p}/n)\left[1 + (n-1)\bar{p} - n\bar{p}\right]$

$$= \bar{p}(1 - \bar{p})/n$$

or $\qquad \sigma_R^2 = \dfrac{\bar{p}\,\bar{q}}{n} \qquad\qquad (23)$

(Both of the derivations are based on the assumption of obtaining normal distribution).

APPENDIX   III

DESIGN A MOST ECONOMICAL MIXER (DRUM TYPE)

Let 2R be the diameter of the end wall of the drum, and L be the length of the mixer

The total surface of area of the mixer A is thus

$$A = 2\pi (R^2 + RL) = 2\pi R(R + L) \tag{1}$$

The volume of the mixer V is,

$$V = \pi R^2 L$$

so that

$$L = \frac{V}{\pi R^2}$$

Hence,

$$A = 2\pi (R^2 + V/\pi R) = 2\pi R^2 + 2V/R \tag{2}$$

Differentiate A with respect to R, and let it equals to zero, thus,

$$\frac{dA}{dR} = 4\pi R - 2V/R^2 = 0$$

$$2\pi R^3 = V$$

or

$$\pi R^2 L = 2\pi R^3,$$

$$L = 2R \tag{3}$$

Therefore, when the lenght of the mixer equal to the diameter, is thus most economical geometric dimensions.

APPENDIX   IV

USING A GAMBLER'S TECHNIQUE (STOCHASTIC) TO
PREDICT THE EFFECT OF MIXING ON THE RATIO OF THE COMPONENTS

The effect of powder mixing on the ratio of two components has been a subject of investigation by many workers, e.g., Oyama (3), Coulson and Maitra (4), Yano et al (5), Poole et al (6) and Ashton et al (7). However, this subject may be studied by analogous to another aspect of stochastic process, namely, the gambler's ruin technique. It will be demonstrated in the following the gambler's ruin towards a prediction of the mixing efficiency with comparison of the available experimental results by previous authors.

The actual gambler's method leads a problem as ; a gambler A who has an initial capital of n (units) against a banker B, whose initial capital is m (units). The gambling game proceeds by steps, each step the gambler has a chance of P of winning a unit from the banker or a chance of $Q = 1 - P$ of losing one unit to the banker. The actual capital X possessed by the gambler after a finite games is thus $0 \leq X \leq n + m$.

Thus, the mixing process may be analogous to this gambling method without taking the actual physical nature of the process into account. Considering a binary mixing system consists of A and B two components, the ratio of A to B varies from 0 to ∞. or the fraction of A in the whole system from 0 to 1. Now one wants to know how the ratio of the components affects the mixing efficiency. Clearly, when A/B = 0 or ∞ gives the prefect mixture, i.e., one component system. However, in general, it may be considered of a gambling method.

In the gambler's terminology that the gambler's ultimate ruin means component-A is completely'mixed' by B. Since the game is taking steps, therefore, the gambler can stop the game at any time if he wishes, this is also meant that the 'mixing process' could be stopped when it was only'partially mixed'.

Let q_n be the probability of A - component'loses'its n-particles (gambler's ultimate ruin) and p_n the probability of its 'winning! Therefore, q_n could be obtained with its initial condition

as follows (8) (9).

After the first trial the A-component contains (gambler's capital) either $n + 1$ or $n - 1$, according to its 'winning' or 'losing' that 'game', hence,

$$q_n = Pq_{n+1} + Qq_{n-1} \qquad 1 \leqslant n \leqslant m+n-1 \qquad (1)$$

If $n = 1$, then,

$$q_1 = Pq_2 + Q \qquad (2)$$

If $n = m + n - 1$, then,

$$q_{n+m-1} = Qq_{n+m-2} \qquad (3)$$

Thus the boundary conditions for (1) will be,

$$q_0 = 1$$
$$q_{m+n} = 0 \qquad (4)$$

Where, $\qquad n = 1, 2, 3, \ldots \ldots m+n-1$.

Let $q_n = W^n$ and put into Eq.(1), so that the auxiliary equation is obtained by a standard method (10),

$$PW^2 = W - Q = 0 \qquad (5)$$

This equation has the roots of $W = 1$, Q/P.

First, let one considers the general case, that the particles of A are not necessary have the equal chance of 'winning' or 'losing' (in gambler's terminology, the game is being considered 'unfair'). Thus, $P \neq Q$. With Eq.(5), so that Eq.(1) has two particular solutions. That :

$$q_n = 1,$$
or,
$$q_n = Q/P \qquad (6)$$

Hence, $\qquad q_n = (1)^n h + k(Q/P)^n \qquad (7)$

Where h, k are the arbitrary constants.

If Eq.(7) is held, that it must be satisfied the boundary condition Eq.(4), then,

$$h + k = 1,$$

and

$$h + k(Q/P)^{n+m} = 0$$

so that,

$$h = \frac{(Q/P)^{n+m}}{(Q/P)^{n+m} - 1} \quad , \quad k = \frac{1}{(Q/P)^{n+m} - 1} \tag{8}$$

The required solution is therefore obtained,

$$q_n = \frac{(Q/P)^{n+m} - (Q/P)^n}{(Q/P)^{n+m} - 1} \tag{9}$$

For the particular case, as the components A and B are under equal chance of 'winning' or 'losing' (the gambler is playing a 'fair' game), then $P = Q$. Therefore, the auxiliary equation (5) has two roots of $W = 1$, so,

$$q_n = h(1)^n + kn(1)^n \tag{10}$$

In order to satisfy the boundary condition Eq.(4), thus,

$$h = 1$$

and

$$h + k(n + m) = 0$$

so that the required solution for q_n is,

$$q_n = 1 - \frac{n}{n + m} \tag{11}$$

Equations (9) and (11) may be used for finding the probabilitied of particles to be 'mixed' after n movement of particles. It can be seen that the probability q_n is dependent on the magnitudes of n and m, if $n \gg m$, in gambler's theory, who would never ruin, only has the chance of winning, on the other hand, if $n \ll m$, then the gambler has no chance of winning. When $n = m$, then the chances of winning and losing will be equal (if $P = Q$). As the mixing is concerned that at $n \gg m$ or $n \ll m$, both cases have the higher probability to be 'mixed', only at $n = m$, then the materials have the lowest

probability of 'mixing'.

The second question, one wants to knows is that how long it takes to mix a batch when the ratio of the components are varified. It leads an expected duration of trail in gambler's terminology. Suppose the duration of the 'game' has a finite expectation D_n (n, m being fixed), if the first trial leads to a win for the gambler, the conditional duration from that point onward is D_{n+1}. The expected duration of the 'whole' game if the first trial is a win (occurring with probability P) is thus $(1 + D_{n+1})$. Similarly, the expected duration of the 'whole' game if the first trial is a loss (occurring with probability Q) is $(1 + D_{n-1})$. Viz.

$$D_n = P(1 + D_{n+1}) + Q(1 + D_{n-1})$$
$$= 1 + PD_{n+1} + QD_{n-1} \qquad 1 \leq n \leq n+m-1 \qquad (12)$$

The extreme value of n = 1 and n = n+m-1 are being included on the assumptions, thus,

$$D_o = 0$$
$$D_{n+m} = 0 \qquad (13)$$

Clearly, Eq.(12) is a non-homogeneous difference equation as being appeared of the term 1. Comparing with Eq.(1), it may be written as

$$D_n = \frac{n}{Q - P} + h + k(Q/P)^n \qquad (14)$$

with the boundary conditions from Eq.(13), hence,

$$h + k = 0,$$
$$\text{and} \qquad h + k(Q/P)^{n+m} = -\frac{n + m}{Q - P} \qquad (15)$$

Solving for h and k, obtained,

$$D_n = \frac{n}{Q - P} - \frac{n + m}{Q - P} \cdot \frac{1 - (Q/P)^n}{1 - (Q/P)^{n+m}} \qquad (16)$$

Similarly, when P = Q = 1/2, then one has to replace the term $n/(Q-P)$

in Eq.(14) by $-n^2$, so that the equation would be reasonable, thus

$$D_n = -n^2 + h + kn \tag{17}$$

with the same boundary conditions of Eq.(13), hence,

$$h = 0,$$

and

$$h + k(n + m) = (n + m)^2$$

So that the required solution for $P = Q$ is,

$$D_n = -n^2 + (n + m) = n.m \tag{18}$$

Eqs.(16) and (18) may be used for determining the duration of 'mixing' with known ratio of the components.

Now let one applies this gambler's technique into a practical mixing problem towards a prediction of mixing efficiency. Considering that n particles of A-component and m particles of B-component in a binary mixing system, as described before. Since the magnitudes of m and n are very large, for convenience, one may use the fractions rather than the actual numbers, so that when m varies from 100 to 0%, n correspondently varies from 0 to 100%, as m+n is constant.

For the sake of simplicity for demonstrating this gambler's technique is workable in the mixing problem, one only considers here, the probabilities of P and Q are equal, as Class 1 which stated in Chapter 4, so that it may be used to campare those results obtained experimentally by previous workers.

TABLE I

m (%)	n (%)	m+n (%)
0	100	100
1	99	100
5	95	100
10	90	100
20	80	100
30	70	100
40	60	100
50	50	100

TABLE   I   (CONT.)

m (%)	n (%)	m+n (%)
60	40	100
70	30	100
80	20	100
90	10	100
95	5	100
99	1	100
100	0	100

TABLE   11

P	Q	n	n+m	Probability to be mixed	Expected mixing time (units)
0.5	0.5	0	100	0	0
"	"	1	100	0.001	99
"	"	5	100	0.05	475
"	"	10	100	0.1	900
"	"	20	100	0.2	1600
"	"	30	100	0.3	2100
"	"	40	100	0.4	2400
"	"	50	100	0.5	2500
"	"	60	100	0.6	2400
"	"	70	100	0.7	2100
"	"	80	100	0.8	1600
"	"	90	100	0.9	900
"	"	95	100	0.95	475
"	"	99	100	0.99	99
"	"	100	100	1.0	0

The results of probability of 'to be mixed' and the 'expected mixing time' are plotted in Fig.(1) and Fig.(2).

Referring to the previous work, Oyama had noted that the degree of mixing may be affected by the weight ratio of the two components, although he did not particularly study this subject, however, he concluded that the better results would be obtained when the weight ratio approaching both extremes, i.e. zero or infinity, the poorest mixing results appeared at unity.

Yano et al studied the effect of mixing on the ratio of two components with great details. Their results may be useful for the comparison of present method. Using the original figure and shown in Fig.(3), from which the relationship of component ratio and mixing time can be compared. If one replaces the measured standard deviation σ by σ/σ_R , where σ_R is the calculated random standard deviation which based on the known sample size and particle size, and replotted in Fig.(4), and extrapolation of all curves to the randomised state, i.e., $\sigma/\sigma_R = 1$ (it is understood that not all mixture could achieve a state of randomisation, as one can see in the text, moreover, the materials which Yano used were too fine to be mixed in a tumbling mixer, however, in here one only demonstrates how the gambler's method may be used). From these curves, the ratio of components and mixing time can be plotted in Fig.(5), or replotted in form as Fig.(6). Both Figs.(1) and (6) have a similar shape, and indicate good agreement.

Poole et al obtained similar experimental results and summarised in Figs.(7) and (8). It was concluded that a low concentration mix reaches its state of limiting homogeneity much sooner than a higher concentration mix, so that, for example, increasing a mix ratio from 10:1 to 1000:1 may decrease the time required to approach random homogeneity by two order of magnitude.

Further experimental results had reported by Ashton et al who provided a bulky data on the mixing of components at different ratios. The results obtained in three out of four mixers have shown that the similar finding as Yano and Poole. These are plotted in Figs.(9), (10) and (11). Rearrangement of the data from Fig.(9) and replotted in Fig.(12), so that it may be used to compare with the gambler's method. Ashton et al stated that the degree of mixedness achieved for 1000:1 ratio of free flowing materials suggests that, at this ratio, the industrial technique of pre-mixing 'master' batches may not be necessary in certain circumstances provided that the only consideration involved is the dispersal of one part of A in 1000 parts of B.

From the reported results indicate that the gambler's method for predicting the effects of mix ratio of two components on the degree of mixing and mixing time is workable, this method may be

used not only qualitatively but also quantitatively, e.g., one tests
an experiment at a very low concentration level, say 1:1000 to obtain
the required mixing time for achieving the state of randomisation,
then compare with the gambler's method by correlating of the proba-
bilities of P and Q (this may also depend on the material properties),
from which the time requirement for higher concentration mixtures
may be obtained without doing further experiments. This method may
be particularly useful for mixing of free-flowing powders. This study
is only an attempt of using some available probability theory into
the particle mixing problem. It is hoped that the effect of mixing
on the ratio of components will need no further investigation ex-
perimentally, and making use of the available method on the similar
aspects. The gamble's method may also be extended into multi-component
system (as the banker is not restricted to face only one gambler),
the solution would lead a matrix (11).

* Remark : Poeple who work in the pharmaceutical industry may
appreciate the difficulty of mixing with a very low concentration
active ingredient and other inactive ingredients,so that pre-mixing
is usually employed. However, on the theoretical aspects, it has
shown that a very low concentration mixture and a high concentration
mixture for achieving the state of randomisation have no difference,
e.g., 4 black particles mix with 4000 white particles and 4000 black
particles with 4000 white particles (without taking the actual material
properties into account), now one divides each mixture into four
parts and asks what is the probability of finding equal number of
black particles in each part of the mixture. Clearly, in the former
case, the probability is 4/4004 or .000999 and later case is .5.
Therefore, it seems that when mixture at 50/50 level would be easier
to be mixed, however, when one considers further that the original
concentration of the former case is only .000999, and the later case
is .5, so that, finding a black particle at the randomised state
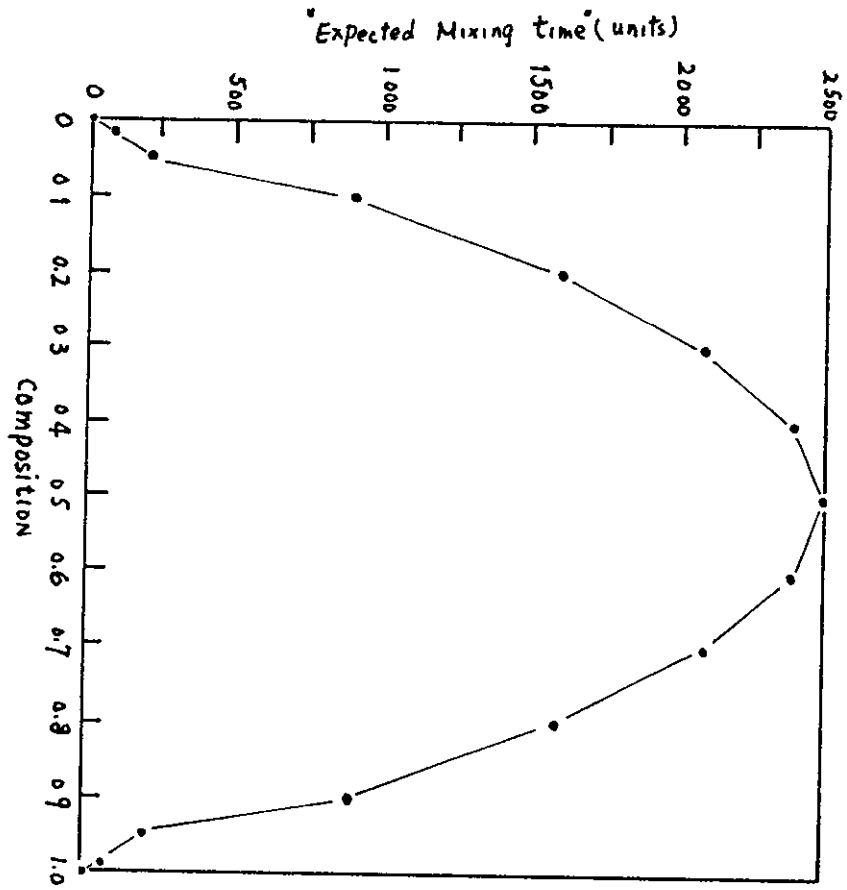will be the same. From the present point of view the pre-mixing is
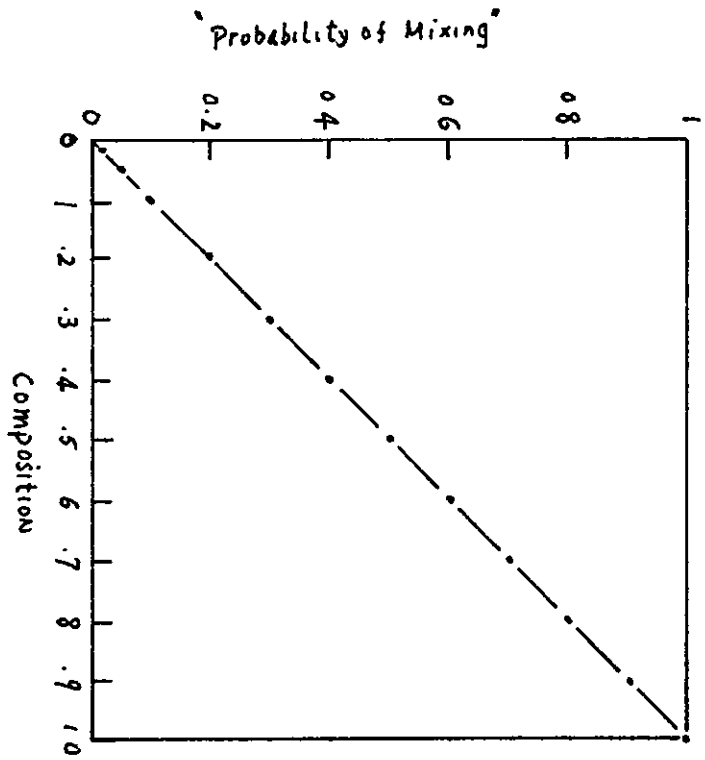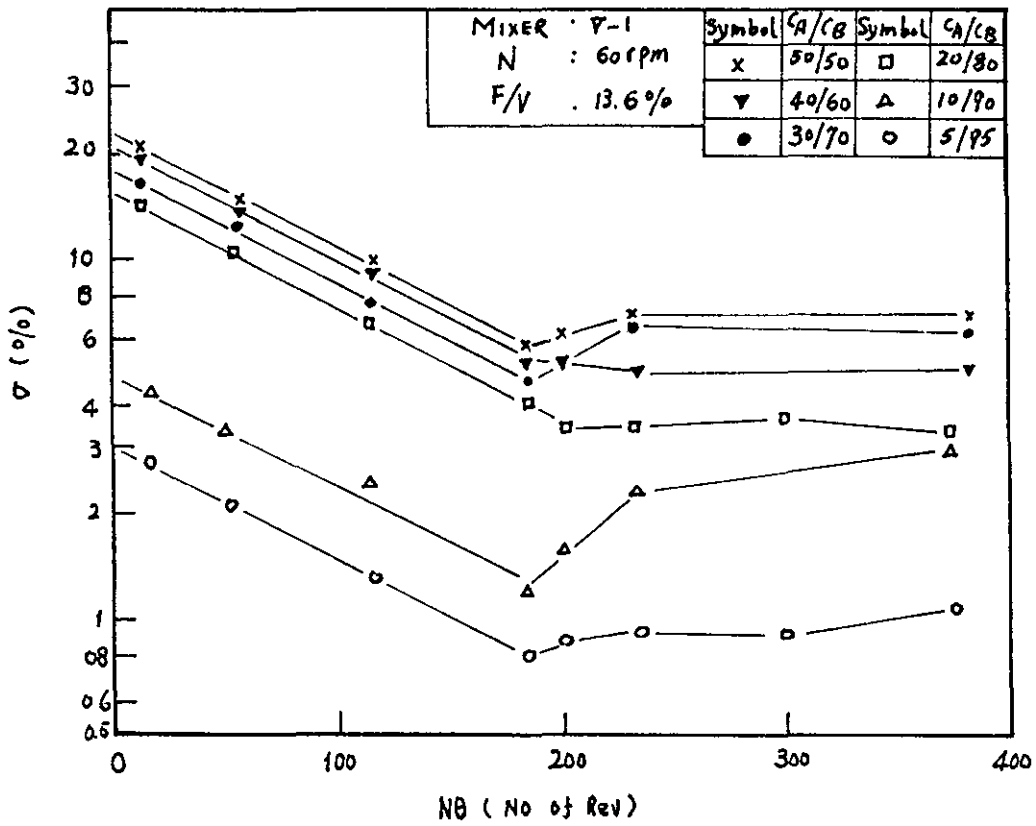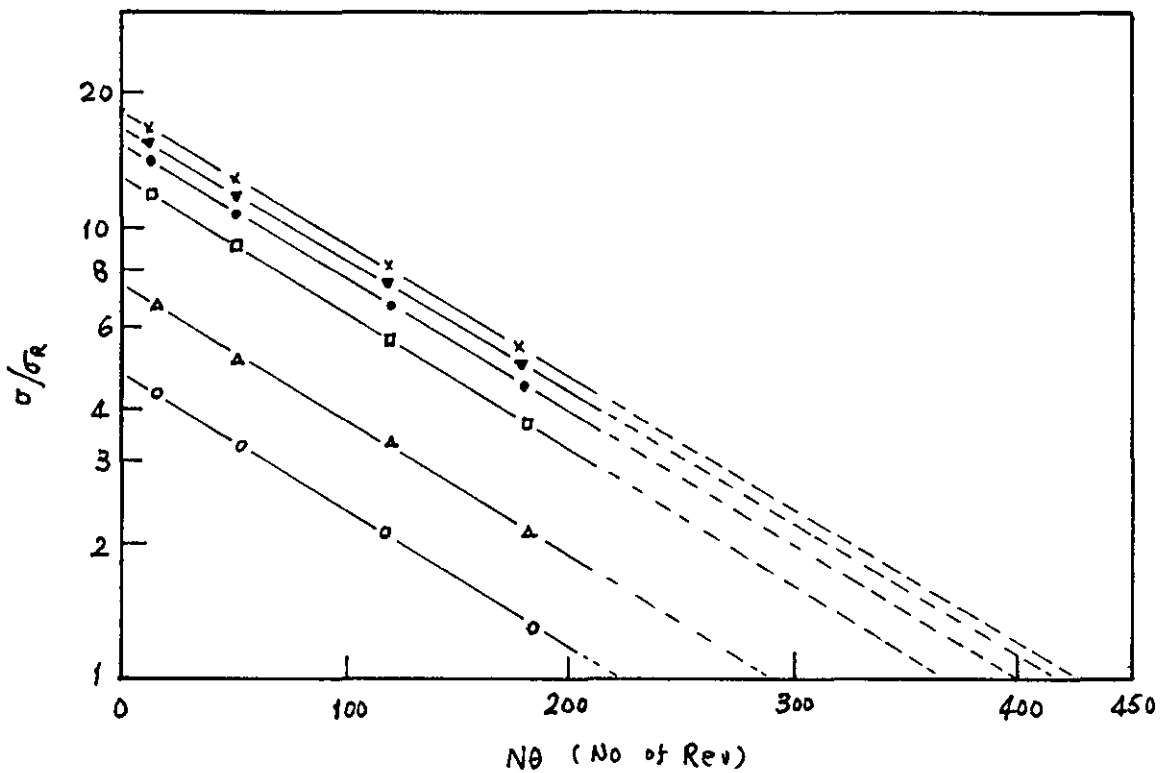unnecessary.

FIG. 1

FIG. 2

FIG. 3   NθVS.   σ(%)

FIG. 4   Nθ VS   %/σR

NΘ (No of Revolutions)
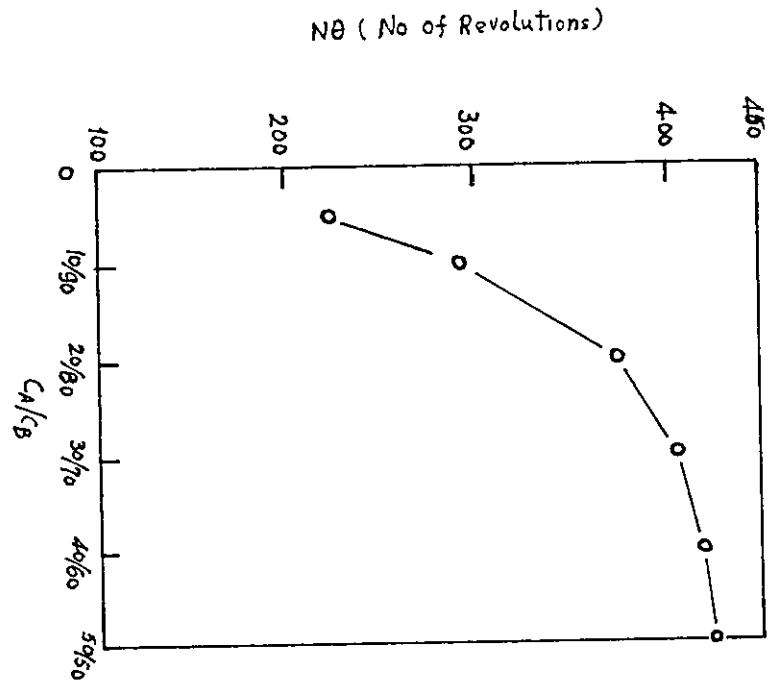
$\frac{C_A}{C_B}$

FIG 5

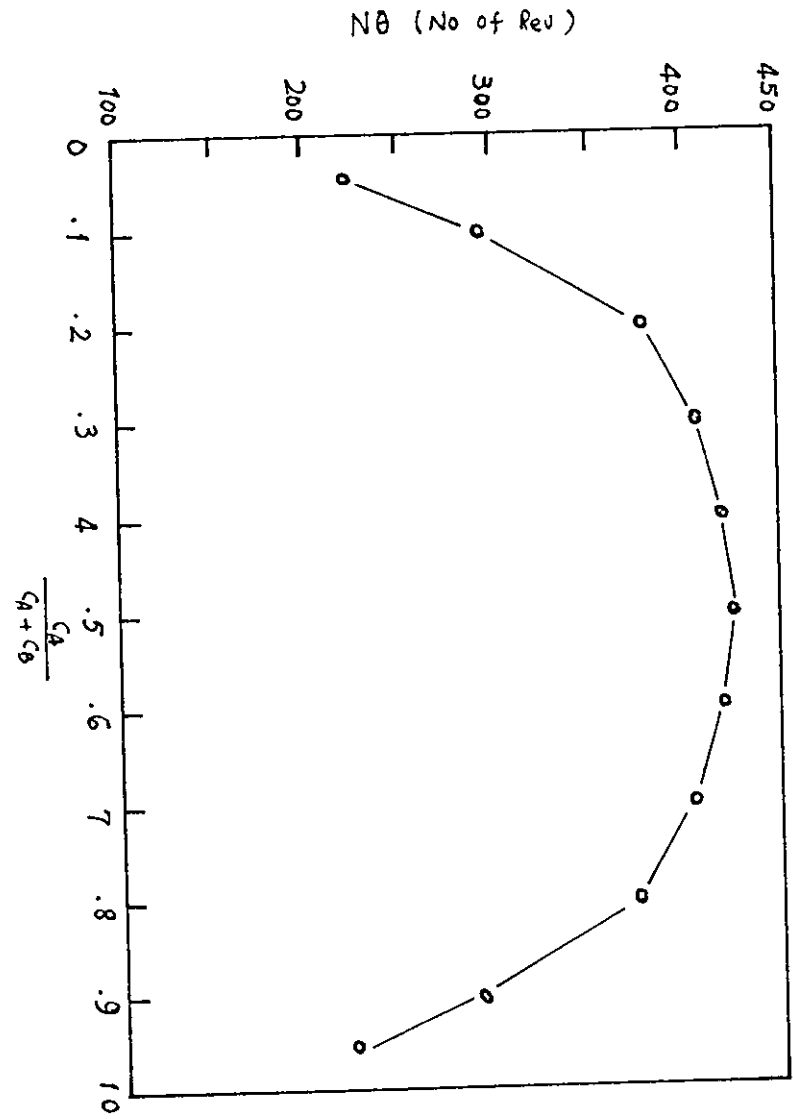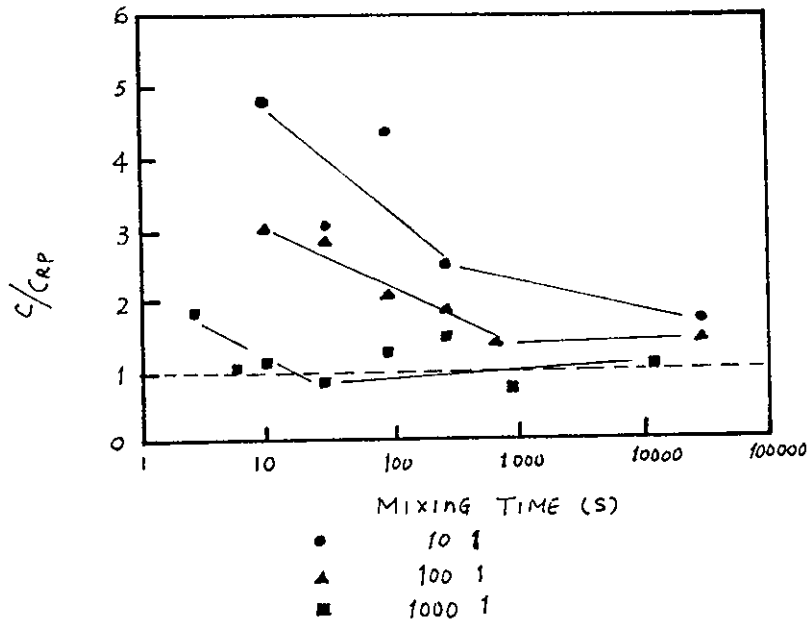NΘ (No of Rev)

$\frac{C_A}{C_A + C_B}$

FIG. 6

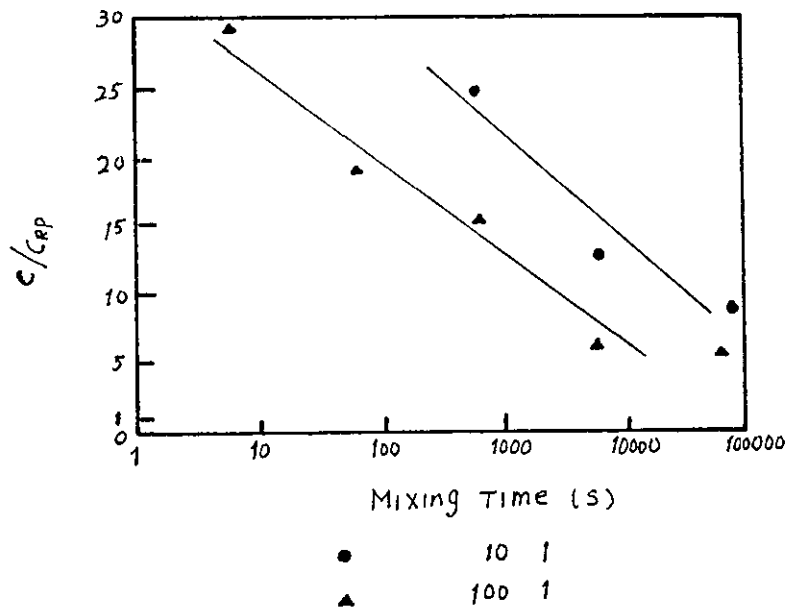FIG 7 Effect of Mixing time on relative homogeneity

FIG 8 The effect of mixing time on homogeneity relative to that for random mixed particles
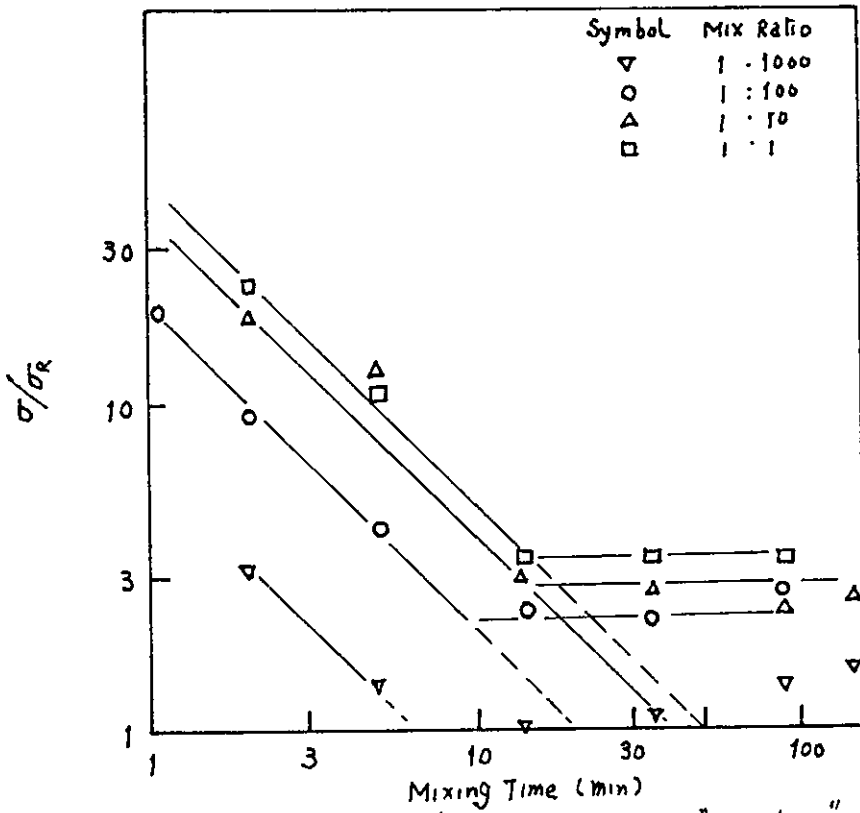
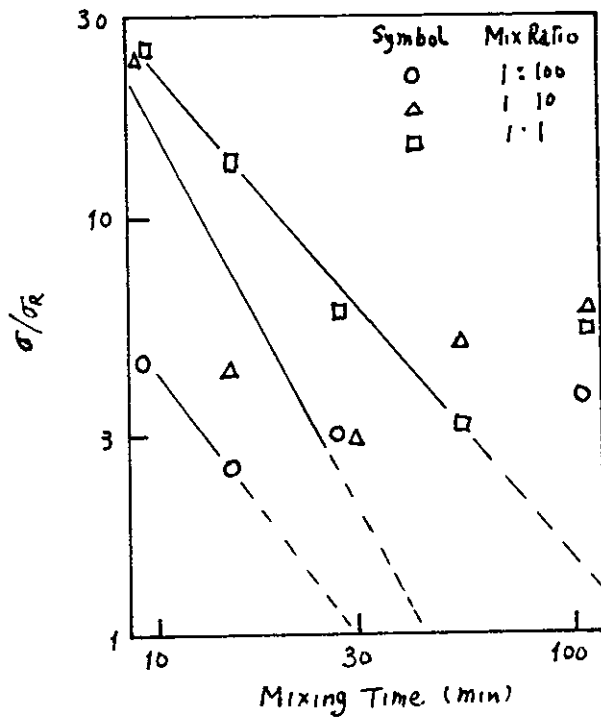FIG. 9 The effect of Mixture composition upon "Mixedness" in
the Z-Blade Mixer

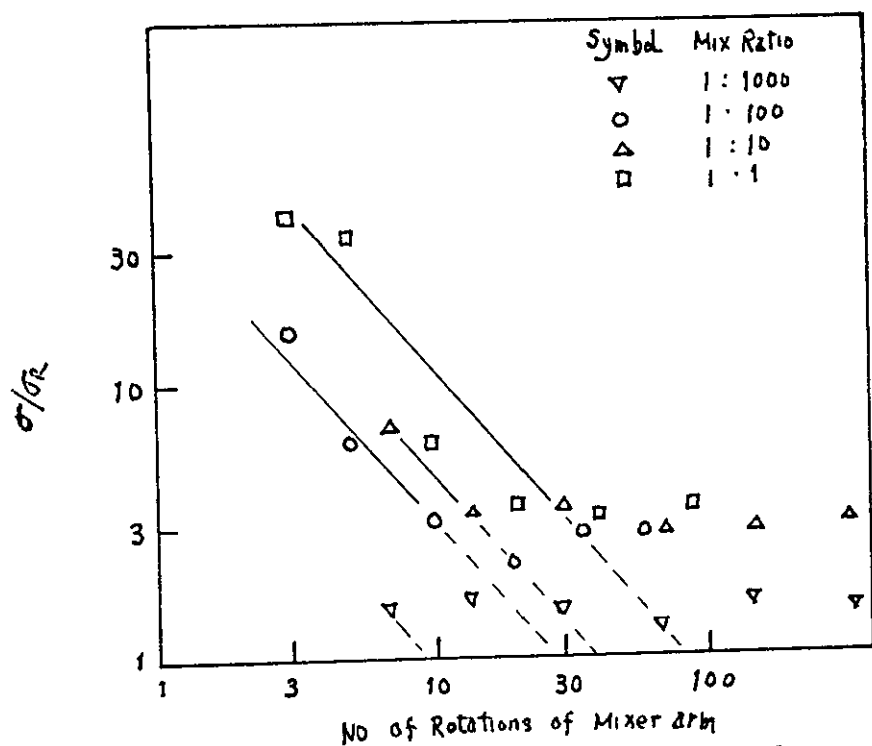FIG 10 The effect of Mixture composition upon "mixedness"
In the Air-Mixer

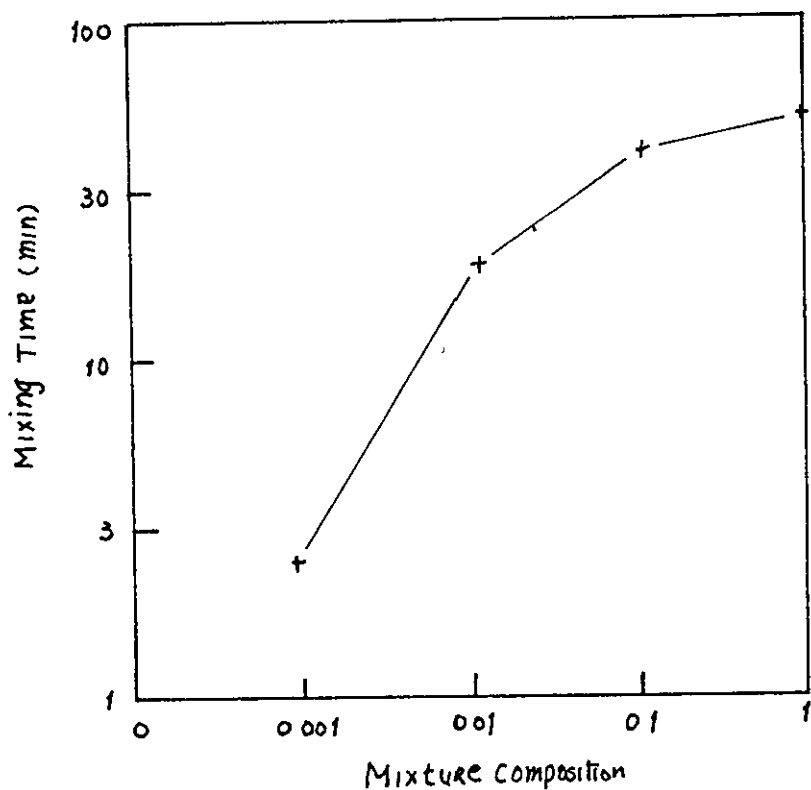FIG 11   The effect of Mixture Composition upon "mixedness" in the Nauta Mixer

FIG 12   The mixig time required for randomised state vs Mixture Composition in the z-blade Mixer

REFERENCES

1. Shchigoler, B.M.   'Mathematical analysis of observation' Iliffe Book Ltd., London, 1960.

2. Lacey, P.M.C.   'The mixing of solid particles' Trans.Instn. Chem.Engrs. 21, 53, 1943.

3. Oyama, Y.   'The motion of granular or pulverulent materials in a horizontal rotating cylinder, report 2 mechanism of mixing and mixing rate' Bull. Inst. Phys. Chem. Res.(Tokyo) 14, 7, 570, 1935.

4. Coulson, J.M., Maitra, N.K.   'The mixing of solid particles' Ind. Chemist Feb., 1950.

5. Yano, T.,Kanise,I., Tanaka, K.   'Mixing of powders by the V-type mixer' Kagaku Kogaku, 20, 4, 1956.

6. Poole, K.R., Taylor, R.F., Wall, G.P.   'Mixing powders to fine-scale homogeneity studies of batch mixing' Trans.Instn.Chem. Engrs. 42, T305, 1964.

7. Ashton, M.D., Valentin, F.H.H.   'The mixing of powders and particles in industrial mixers' Trans.Instn.Chem.Engrs. 44, T166, 1966.

8. Feller, W.   'An introduction to probability theory and its application' Vol. 1, 2nd Ed., John Wiley & Sons Inc., N.Y., 1958

9. Bailey, N.T.J.   'The elements of stochastic processes with applications to the nature science' John Wiley & Sons Ltd., 1965.

10. Gold-berg, S.   'Introduction to difference equations' John Wiley & Sons Inc., N.Y., 1958.

11. Level, M.   'Probability theory' 3rd Ed., D. Van Nortrend, Co., Inc. 1963.