

**ELUCIDATION OF LOCAL AND GLOBAL STRUCTURAL
PROPERTIES OF PACKED BED CONFIGURATIONS**

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for the degree of Doctor of Philosophy**

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by

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ABSTRACT

Some of the most common unit operations being used in the chemical industry are based upon packed bed configurations. Multi-tubular catalytic reactors are a prominent example of such systems which are believed by many experts to be the heart of the chemical profession and frequently the most expensive units of the plant. They offer extensive surface area between phases of the matter in a compact physical manner, and also are relatively easy to construct and maintain. The successful design of such systems depends on models, which should describe the processes occurring within the fixed bed and must also be able to accurately represent the intrinsic behaviour of the reactor quantitatively. It is therefore necessary to have good estimates of the associated transport parameters, namely the wall heat transfer coefficient, effective radial and axial thermal conductivity coefficients and radial and axial dispersion coefficients, in order to design efficiently. Unfortunately, the accuracy of the design data for the prediction of these parameters are in doubt. They are considered to be unreliable because of being based on poorly defined packed bed models in terms of structure of the packing. The non-uniformities of voidage in the packing matrix must be appreciated and hence be included in descriptive models of the physical system in order to establish reliable design data.

As the roles of modelling and experimentation are complementary, a number of beds comprised of equilateral cylindrical particles were prepared to examine their global and local structural properties, with a view to identifying and characterising features which could be used for prediction purposes. The micro-structural details within packed beds were studied by means of an image analyser, so that the local variations of voidage in angular, axial and radial directions could be portrayed. It is only on the basis of this kind of information that a well-defined description of the bed structure becomes accessible. This research activity has been concerned with the study of several factors influencing the packing structure

such as entrance/exit and wall effects, reproducibility and scaling problems. Also succeeding the accurate data acquisition stage, a number of correlations have been developed for a wide scope of diameter ratios covering the industrial range. These correlations allow the mean and local voidage distributions to be predicted reliably.

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CHAPTER 1

INTRODUCTION

CHAPTER 1

INTRODUCTION

1.1 General Background

Packed bed type devices are widely used in industrial practice to promote fluid mixing as well as to provide extended interfacial area between phases. For economical production of large amounts of product, fixed-bed catalytic reactors are usually the first choice, particularly for gas-phase reactions. Many catalysed gaseous reactions are amenable to long catalytic life (1 - 10 years) and as the time between catalytic changeouts increases, annualized replacement costs decline dramatically, largely due to savings in shut-down costs. It is not surprising therefore, that fixed-bed reactors now dominate the scene in large-scale chemical product manufacture. Many special fixed-bed designs have been developed, but all types of fixed-bed reactors fall into one of two major categories: adiabatic or non-adiabatic. The adiabatic design is always preferred when acceptable conversion and selectivity to the desired product is possible. It is simpler and less costly than the non-adiabatic design, which requires heat transfer along the bed between the reaction side and a cooling or heating medium. For such heat transfer to be effective, a high ratio of heat transfer surface to reactor volume is required, usually in the form of multiple small diameter tubes containing the catalyst and with the heat transfer medium flowing exterior to the tubes. Figure 1.1 shows a single adiabatic bed which can be used for moderately exothermic reactions such as a mild hydrogenation process. Figure 1.2, however, shows a multi-tubular non-adiabatic reactor which is employed for highly endothermic or exothermic reactions requiring close temperature control to ensure high selectivity such as ethylene oxidation to ethylene oxide. Nowadays, a considerable part of solid catalysed gas-phase processes is

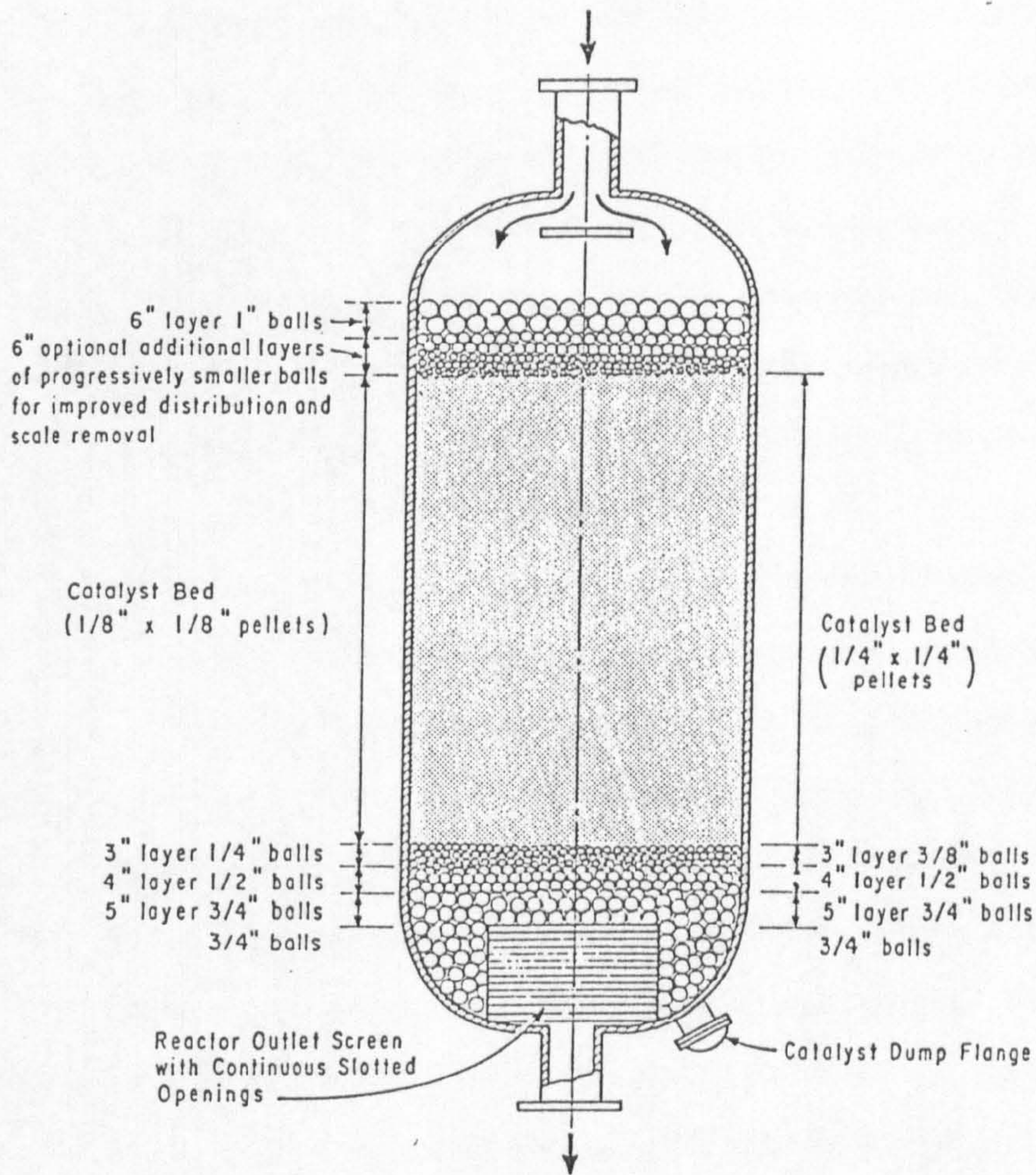


FIGURE 1.1: Single Adiabatic Bed

(Source of Extraction: H. F. Rase, "Fixed-bed reactor design and diagnostics", Copyright 1990, Butterworth Publishers)

carried out in multi-tubular reactors, which as the most expensive units of the plant, exert a large influence on the total production costs. The equipment possesses a mechanical configuration with two basic components. These are a bundle of tubes forming an array of a definite geometrical pattern and a shell, usually equipped with baffle plates of special design, in order to ensure the most effective flow distribution in the inter-tubular space and thus the most efficient heat transfer to or from the tube bundle. The number of tubes in the bundle can reach as many as 20,000 to 30,000. In nearly all types of multi-tubular reactors used today, the catalyst is inserted inside the tube and the heat transfer fluid flows through the shell side of the unit. The tube bundle may be arranged in different ways. In most industrial designs, the tubes form equilateral triangular arrays (staggered or in-line), although other solutions such as square arrangements are sometimes used.

As the reaction proceeds on the catalyst surfaces, gradients of temperature and concentration (or partial pressure) are produced and constitute the driving potential for the transport processes of mass and heat between the interior of the catalyst and the flowing fluid. In addition to these complex transport processes, flow patterns of the bulk fluid as it passes through the bed are tortuous and unpredictable, since a priori knowledge of particle arrangement in a bed is not possible. Figure 1.3 shows the movement of fluid in a packed bed. The designs of such systems as multi-tubular catalytic reactors, thermal regenerators and packed distillation and absorption towers heavily depend upon models which adequately describe the transport of mass, heat, and fluid as well as the pressure drop of the fluid through the bed. These mechanisms in turn are all sensitive to the size and shape of the free paths available for flow, i.e. voidage of the packing. For instance, non-uniformities of voidage in the bed structure can influence the velocity of fluid passing through the packing, this has a direct effect on the film heat transfer coefficient and the pressure drop of the fluid across the bed. Also the contacting time between the fluid and the solid is influenced by the above changes and could hence have a significant effect on the

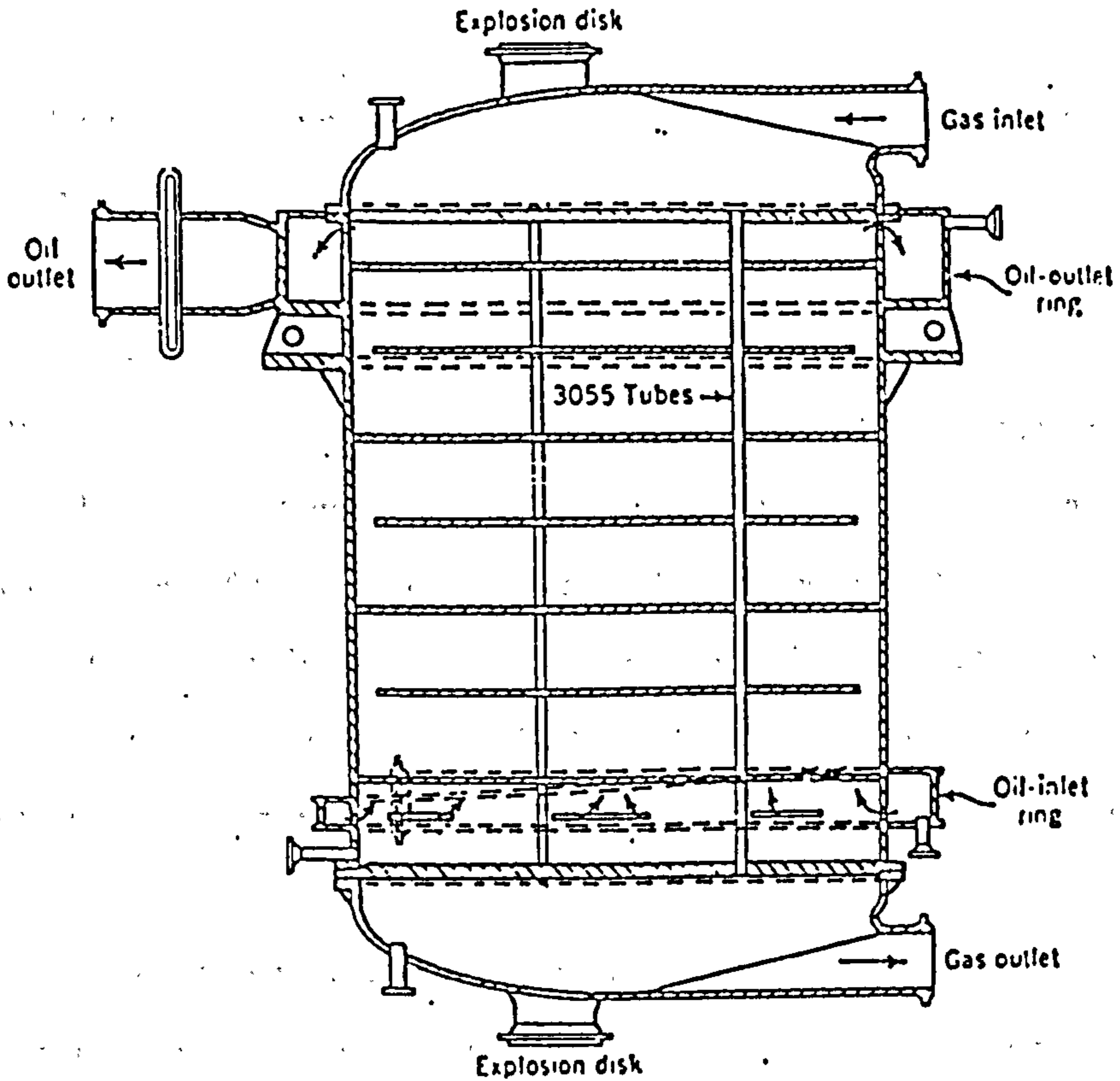


FIGURE 1.2: Multitubular non-adiabatic reactor

(Source of Extraction: H. F. Rase, "Fixed-bed reactor design and diagnostics", Copyright 1990, Butterworth Publishers)

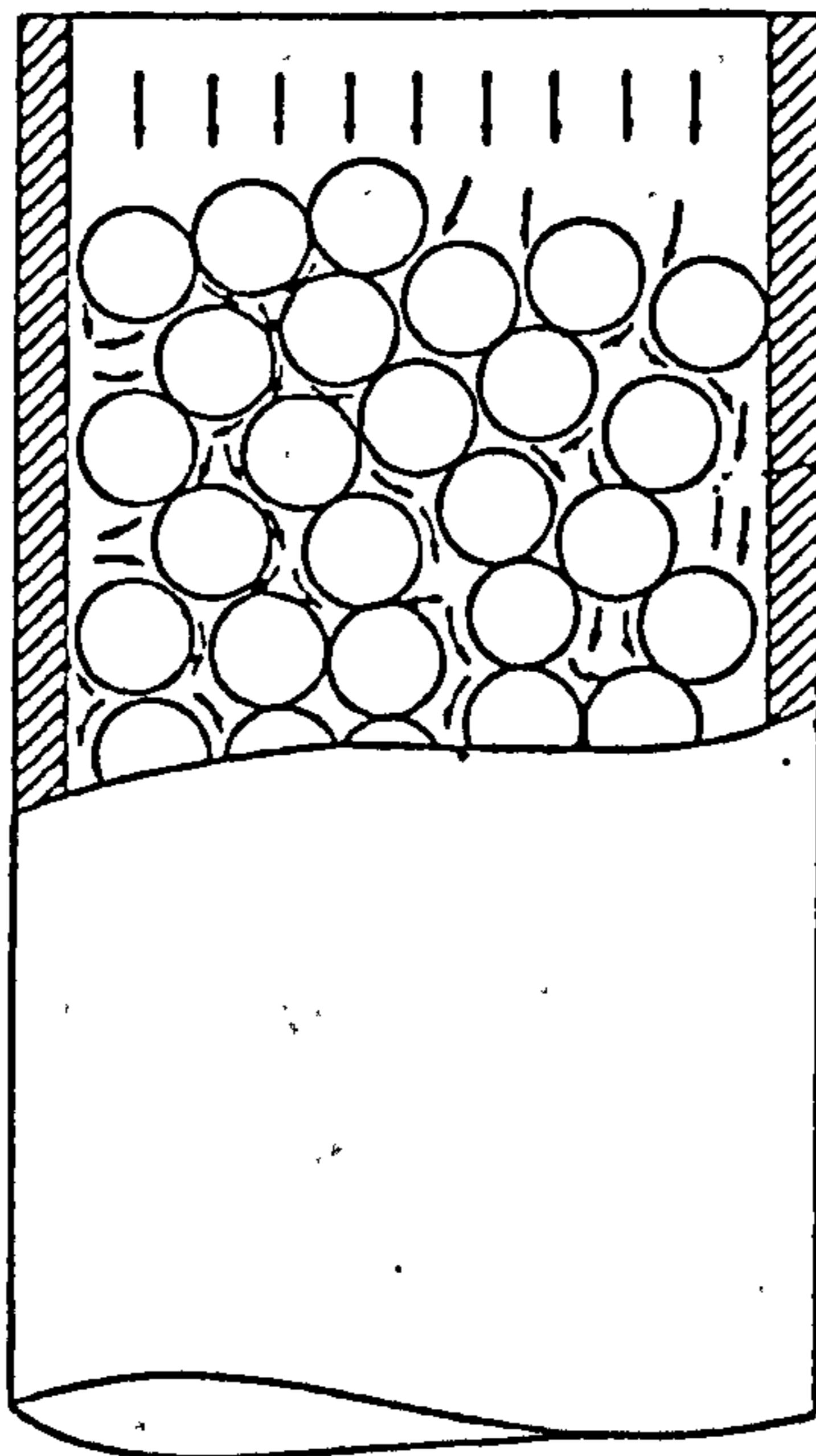


FIGURE 1.3: Movement of fluid in a packed bed

production rate and yield of the catalytic reactor. Therefore, the structural features of the packing play an important role in the performance and overall characteristics of particulate systems.

1.2 Objectives of the Present Study

Reliable design of any unit operation depends heavily upon the accuracy of the design data. Any attempt to quantify the phenomena occurring in a fixed bed must take account of the transport of heat and mass between the catalyst and the fluid and within the catalyst, the pressure drop across the bed, and the certainty of the conservation of energy and mass. It is therefore vital to have good estimates of transport parameters, in particular the wall heat transfer coefficient, effective radial and axial thermal conductivity coefficients, and radial and axial dispersion coefficients. Despite extensive effort over the past few decades, there is still a great deal of uncertainty as to the reliability of the available data for the prediction of these parameters. The published correlations are regarded as unreliable because of being based on poorly defined models in terms of structural characteristics of the bed, such is the plug flow assumption which indicates uniform distribution of voidage across the bed. This is not a realistic representation of the structure of the system since the existence of enhanced flow passage in the vicinity of the tube wall has been verified by numerous investigators. Therefore there is a need for developing refined predictive models which incorporate non-uniformities associated with bed structure.

Figure 1.4 illustrates a flow chart for developing more descriptive models for design and performance prediction of packed bed systems. This path is currently taken by the department to achieve the above goals, and the present study is concentrated on the top section of the chart, i.e. the characterisation of the packing structure. Detailed study of the global and local structural properties of the packing gives a clear indication of the extent of the entrance/exit and wall effects on the

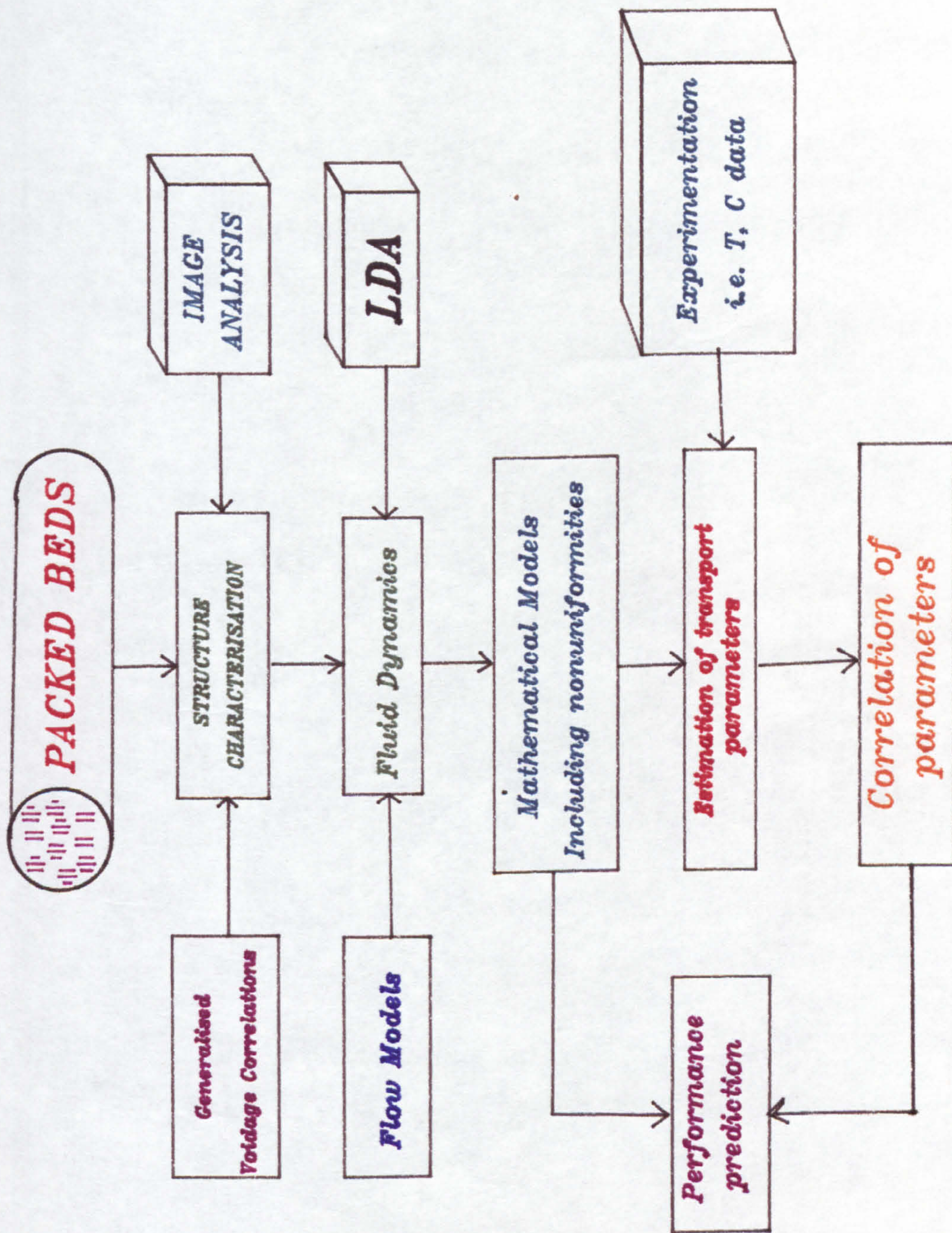


FIGURE 1.4: A Schematic Diagram for the Prediction of Performance in Packed Beds

particle assembly. The generalized voidage correlations derived from such investigations can be used in appropriate flow models, e.g. Brinkman type, to establish the fluid dynamics of the system. This approach in conjunction with suitable mathematical representation of the physical system could be used to extract the values of the transport coefficients of a given set of experimental data. This information can then be used to formulate design correlations in terms of descriptive parameters, e.g. Re , Pr , of the system.

Most studies into dispersion, pressure drop and chemical reaction in packed beds have been concerned with spherical particles, and yet in many industrial applications of catalysis the particles are cylindrical. One reason for this is convenience of manufacture, also there are often a number of distinct process advantages in the use of cylindrical pellets of which the most important are probably uniformity of size. The application of cylindrical particles is hampered because accurate information necessary for the design is not readily available. Literature lacks adequate data which enables structural properties of such beds to be established predictively.

Owing to the rapid developments in the field of image analysis it is now possible to study the micro-structural details within packed beds and hence identify and characterise the desired local properties. This is a process for deriving numerical information from images and quantifying their parameters - size, shape and number - for subsequent identification and characterisation. The technique has become an indispensable tool and has found application in such diverse fields as medicine, science and engineering. Void fraction of a number of beds consisting of full equilateral cylindrical particles have been examined by a high resolution Image Analyser. Generalised correlations which allow the distribution of voidage in axial and radial directions of such beds to be reliably predicted are hence developed from the data obtained. The effects of reproduction and scaling of these beds have also been examined.

1.3 Structure of the Thesis

In order to report on the approach and tackling of the aforementioned objectives of this research activity, the thesis has been structured in an orderly and systematic fashion. Chapter 2 contains a comprehensive literature review on the structural properties of regular and randomly packed beds of spherical and cylindrical particles.

Chapter 3 discusses the hardware and software of the image analysis systems, together with the way the sample materials are chosen and prepared.

Chapter 4 explains the functional dependency of the structural characteristics on the geometrical and dimensional properties of the system. Also developed correlations which allow the bulk void fraction and the distribution of voidage in axial and radial directions of packed beds of cylinders to be predicted are included.

Chapter 5 compares the resulting correlations and data of this study with the published ones.

Chapter 6 concludes the findings of this research project and includes recommendations for future work on this subject.

CHAPTER 2

LITERATURE SURVEY

CHAPTER 2

LITERATURE SURVEY

2.1 Introduction

The existing information on the global and local structural properties of packed bed systems is reviewed in this chapter. Also a critical appraisal of the reported techniques for obtaining the desired data and the published correlations for the subsequent prediction of the structural behaviour are given.

2.2 Regularly Packed Particles Review

It is convenient to consider regular packings as assembled from layers and rows. Among the earliest work is that of Barlow, (1883) and Slichter, (1899) since when the theory of packing has been reviewed, refined and extended by Graton and Fraser, (1935); Hrubisek, (1941); Ackermann, (1945); Foord, (1945); Martin, et al., (1951); Wells, (1962). Hrubisek, (1941), in his comprehensive work considered the fundamental unit to be a row of contacting spheres. These rows can be arranged in the same place, parallel to each other and touching, to form a layer. Any number of special rhombic layer forms with intermediate intersection angles between rows are possible, but the most common packings are built from one or other of the limiting forms. These are the square layer with a 90° angle and the triangular or simple rhombic layer with an angle of 60° . Haughey and Beveridge, (1969); showed three stable ways in which two square or two triangular layers may be stacked:

- 1) each sphere in the second layer is placed vertically above a first layer sphere;

- 2) each second layer sphere is placed in the saddle point formed between two contacting first layer spheres;
- 3) each second layer sphere is placed in the hole formed by the contacting spheres of the first layer.

While an infinite number of regular forms can be produced by varying the angle of row intersection and the degree of layer displacement, the most common regular packings are of third and lower order. The following are the four basic patterns formed as a result of the above together with the evaluated value for the bulk mean voidage:

- i) Cubic, $\epsilon_{\text{mean}} = 0.4764$;
- ii) Orthorhombic, $\epsilon_{\text{mean}} = 0.3954$;
- iii) Tetragonal-spheroidal, $\epsilon_{\text{mean}} = 0.3019$;
- iv) Rhombohedral, $\epsilon_{\text{mean}} = 0.2595$.

Higher ordered packings have also been considered by Menzer, (1938); who interpreted high ordered packing as a systematic multiple twinning of the closed packed array. Manegold and von Engelhardt, (1933); described particle arrangements with points of contact between 3 and 12. Bagley, (1965); discussed a dense regular packing with five fold symmetry but it is shown by Bernal, (1966); and Clarke, (1966); that the packing is equivalent to a twinned structure. Broch, (1932); stated that while some variations will not affect bulk properties such as mean voidage they can, however, influence orientation and shape of solid or pore paths.

Various open-structured regular packings have been described. These are relatively loose arrangements with at least some of the particles free to move with respect to each other if they are not constrained by some bonding force at each point of contact, as in beds formed by crushing solids, sedimentation (Meissner et al.,

(1964)), or flocculation (Hutchinson and Sutherland, (1965)). The loosest forms (Hilbert and Cohn-Vossen, (1932)); and Heesch and Laves, (1933); have only four points of contact and a voidage greater than 0.877.

2.3 Randomly Packed Particles Review

Random packings are formed by the haphazard positioning of the particles to form an assembly or bed. Particles pack into mechanically stable or quasi-stable arrangements having at least three points of contact with the neighbouring particles. The average properties of such beds are largely dependent upon the mode of assembly. Several widely different models have been proposed based upon regular elements of particle packings to represent the randomly packed beds of particles:

i) Polyhedral models -

Bernal, (1960); interpreted a random packing of identical spheres in terms of polyhedra whose vertices are at the midpoints of the lines joining the base and neighbouring sphere centres. A predominance of pentagonal forms was found. Levine and Chermick, (1965); used polyhedra formed only by touching spheres.

ii) Coupled sphere models -

Blum and Wilhelm, (1965); used a unit cell in which an integral number, such as two or three, of touching spheres are coupled to form a cohesive unit.

iii) Local sphere shell models -

Models in which spheres are centred in shells at certain fixed distances from a base sphere have been used by Haughey and Beveridge, (1966); with triangular close packed and triangular equi-spaced models describing the limits of possible values of the base sphere co-ordination number.

iv) Monte Carlo models -

In the simplest case proposed by Bernal, (1960); spheres are assumed to be placed randomly one by one throughout the specified space, rejecting any location which causes overlapping. Griffith, (1962); and Smalley, (1962); have applied this to the packing of spheres along a straight line, but is in general rather cumbersome, producing a large number of unsuccessful trials when the packing is almost complete due to overlapping. In two dimensions, this computational problem has been avoided by displacing any overlapping sphere to accommodate new spheres, according to Round and Newton, (1963). Another application to the equation of state for hard sphere molecules, starts with a regular packing in unit space and then attempts random displacements of arbitrarily selected spheres. Salsburg and Wood, (1962); simulated an effectively infinite packing by applying periodic boundary conditions which were equivalent to the unit space being surrounded by identical units. This has been applied to two dimensions by Alder et al., (1955); and in three dimensions by Craker and Ray, (1966).

v) Void models -

Models of the void structure are mainly employed in the estimation of flow properties. Scheidegger, (1954); has reviewed the straight and serial types of capillary models. Foster and Butt, (1966); Petersen, (1957); Dodd and Kiel, (1959); used random networks of different sized capillaries. The pore size distribution has been presented differently by various investigators. Harris, (1965); used the rows and columns of a Latin square; whereas Lamb and Wilhelm, (1963); employed a size distribution of interconnected spherical holes instead; and a set of tubes with recessed pockets were used by Turner, (1959);

to represent the distribution. Epstein, (1958); Deans and Lapidus, (1960); Crider and Foss, (1965); have also considered voids as equivalent to interconnected mixing cells. Salsburg, (1966); showed that the Voroni polyhedra provides a convenient means of finding and size classifying all voids.

Just as each regular packing mode has a characteristic bulk mean voidage, it has been found that in the random packing of identical spheres a characteristic range of bulk mean voidage values is associated with a particular method of formation. Four modes can be distinguished:

i) Very loose random packing -

Ergun and Orning, (1949); reported that a fluidised bed at the minimum fluidisation velocity has a bulk mean voidage, ϵ_{mean} , of 0.46 to 0.47. Lamb and Wilhelm, (1963); continued that when the fluid velocity is slowly reduced the spheres settle to a ϵ_{mean} of about 0.44. Steinour, (1944); obtained similar values by sedimentation of spheres; also Oman and Watson, (1944); and Happel, (1949); by inversion of the bed container.

ii) Loose random packing -

Scott, (1960); Bernal and Mason, (1960); Sonntag, (1960); Macrae and Gray, (1961); Rutgers, (1962); Epstein and Young, (1962); Parsick et al., (1966); Susskind and Becker, (1966); Bernal and Finney, (1967); have all reported a bulk mean voidage value of 0.40 to 0.41 by packing the spheres so that they roll individually into place over similarly placed spheres; by individual random hand packing; or by dropping the spheres into the container as a complete loose mass. Eastwood, et al., (1969); obtained an average value for voidage of uniformly sized smooth macrospheres to be 0.43 ± 0.02 . Levine and Chernick's

model, (1965); gives a bulk mean voidage of 0.391. The Monte Carlo models usually produce values representative of these packings since no shaking down is taken into account. Tory et al., (1973); developed a computer program called PACS, to simulate very slow settling of rigid equal spheres into a randomly packed bed. An experimental void fraction of 0.424 was obtained from fairly crude measurements on a bed which was not free from wall and end effects. The simulated value was 0.419.

iii) Poured random packing -

When spheres are poured into a container, which is a common industrial practice, bulk mean voidage values between 0.375 and 0.391 are found (Carman, (1937); Wadsworth, (1962); Debbas and Rumpf, (1966)). The value of 0.25 is reported by Roblee et al., (1958); for ϵ_{mean} of randomly packed beds of cylindrical slugs. Parrish, (1961); and Weirusz, (1967); reported a mean value of 0.369 and 0.37, respectively, while Rozanski, (1964); quoted $\epsilon_{\text{mean}} = 0.375$. Peebles, (1965); found that $\epsilon_{\text{mean}} = 0.377$, while Benenati and Brosilow, (1962); had revealed that $\epsilon_{\text{mean}} = 0.39$ and Denton et al., (1963); reported that $\epsilon_{\text{mean}} = 0.395 \pm 0.005$. Beavers et al., (1973); reported the average value of the porosity for all the beds having diameter ratios greater than 15 to be 0.368. Beasley and Clark, (1983); confirmed Beavers et al., data. Dixon, (1988); reported $\epsilon_{\text{mean}} = 0.4$ for diameter ratios greater than 10. The experimental investigation of Moallemi, (1989); indicated that for monosize spherical particles, the magnitude of the ϵ_{mean} of packed bed configurations is only a function of the tube to particle diameter ratio and for relatively large beds, i.e. $d_t/d_p \geq 10$, it takes a constant value of 0.39. A three-sphere coupling model proposed by Blum and Wilhelm, (1965); gives an estimated value of

0.388.

iv) *Dense random packing -*

The minimum voidage values of 0.359 to 0.375 are obtained when the bed is vibrated or vigorously shaken down according to reports from Ranz, (1952); Sonntag, (1960); Macrae and Gray, (1961); Rutgers, (1962); Scott, (1962); Bernal et al., (1962).

Comparing the values of the bulk mean voidage reported for regular and random packings, it can be concluded that a smaller overall range 0.36 to 0.44 is found with random packings compared to 0.26 to 0.47 for the regular packing.

2.4 Mean Voidage Correlations Review

The bulk mean voidage is the quantity most frequently used to characterise the packing structure in a fixed bed. The ϵ_{mean} is affected by the presence of the more ordered packing adjacent to the tube wall. Correlations for this effect are few. Most unit operations texts present graphical correlations over a limited range of d_t/d_p , following Leva and Grummer, (1947). For cylinders there are even fewer predictive equations.

It is generally concluded that the wall effect is negligible if $d_t/d_p > 10$, with a somewhat higher limit for irregular particles. Verman and Banerjee, (1946); derived the following relationship:

$$\epsilon_o = 1 - \left(1 - \frac{d_p}{d_t}\right)^3 (1 - \epsilon) - \left(1 - \left[1 - 2 \delta_w \frac{d_p}{d_t}\right]^3\right) (\epsilon_w - \epsilon) \quad (2.1)$$

where, δ_w = wall layer width in d_p and ϵ_w = voidage of the wall layer. The overall voidage, ϵ_o , and the bulk mean voidage, ϵ , are equal when the wall effects become insignificant as $d_t/d_p \rightarrow \infty$.

Aerov, (1951); applied the above to spherical packings, resulting in:

$$\epsilon_o = 0.39 + 0.07 \frac{d_p}{d_t} + 0.54 \left(\frac{d_p}{d_t} \right)^2 \quad (2.2)$$

while Sonntag, (1960); and Jeschar, (1964); found:

$$\epsilon_o = \epsilon + 0.34 \frac{d_p}{d_t} \quad (2.3)$$

where ϵ is 0.36 for dense random and 0.39 for loose random packings.

Ayer and Soppett, (1964); also proposed the following:

$$\epsilon_o = 0.365 + 0.216 e^{(-0.313 d_t/d_p)} \quad (2.4)$$

$$\text{for } 3 \leq \frac{d_t}{d_p} \leq 32.$$

Beavers et al., (1973); proposed to predict the variation of void fraction with diameter ratio by the following:

$$\epsilon = \epsilon_\infty \left(1 + 2 \frac{d}{D_e} \left[\frac{\epsilon_w}{\epsilon_\infty} - 1 \right] \right) \quad (2.5)$$

where ϵ_w and ϵ_∞ are the void fractions at the wall zone and the core zone, respectively. It is assumed that $\epsilon_\infty = 0.368$, and $\epsilon_w = 0.476$.

At lower diameter ratios Haughey and Beveridge, (1969), proposed an empirical formula based on Aerov's experimental data:

$$\epsilon = 0.39 + 0.07 \frac{d_t}{d_p} + 0.54 \left(\frac{d_t}{d_p} \right)^2 \quad (2.6)$$

Griffiths, (1986), arrived at the following relationship for diameter ratios of greater than 3.5:

$$\epsilon = 0.38 + 0.035 \left(\frac{d_t}{d_p} - 3.5 \right)^{-0.27} \quad (2.7)$$

Dixon, (1988), derived a set of correlations, by both geometrical arguments and empirical treatment of the data, for the prediction of ϵ_{mean} in fixed beds. The void fraction has been correlated as a function of particle to tube diameter ratio for packings of spheres, equilateral cylinders and hollow cylinders. The correlations proposed for full cylinders are:

$$\epsilon = 0.36 + 0.1 \frac{d_{pv}}{d_t} + 0.7 \left(\frac{d_{pv}}{d_t} \right)^2 \quad (2.8)$$

$$\text{for } \frac{d_{pv}}{d_t} \leq 0.6$$

$$\epsilon = 0.677 - 9 \left(\frac{d_{pv}}{d_t} - 0.625 \right)^2 \quad (2.9)$$

$$\text{for } 0.6 \leq \frac{d_{pv}}{d_t} \leq 0.7$$

$$\epsilon = 1 - 0.763 \left(\frac{d_{pv}}{d_t} \right)^2 \quad (2.10)$$

$$\text{for } \frac{d_{pv}}{d_t} \geq 0.7$$

where d_{pv} is the diameter of equivalent volume sphere. The correlation for hollow cylinders is:

$$1 - \epsilon_{hc} = \left[1 + 2 \left(\frac{a}{b} - 0.5 \right)^2 \left(1.145 - \frac{d_{pv}}{d_t} \right) \right] \left(1 - \frac{a^2}{b^2} \right) (1 - \epsilon_{sc}) \quad (2.11)$$

$$\text{for } \frac{a}{b} \geq 0.5$$

where subscripts hc and sc denote hollow and solid cylinders, respectively. Also a and b are the inside and outside diameters of hollow cylinders.

Moallemi, (1989), developed a mathematical expression based on his experimental data to predict the mean voidage of packed beds of spherical particles:

$$\epsilon = 1 - \frac{2}{3} \left(\frac{1}{d_r} \right)^3 \frac{1}{\sqrt{\frac{2}{d_r} - 1}} \quad (2.12)$$

$$\text{for } 1 \leq d_r \leq 1 + \frac{\sqrt{3}}{2}$$

$$\epsilon = 0.383 + 0.254 d_r^{-0.923} \frac{1}{\sqrt{0.723 d_r - 1}} \quad (2.13)$$

$$\text{for } d_r > 1 + \frac{\sqrt{3}}{2}$$

$$\text{where } d_r = \frac{d_t}{d_p} .$$

The first expression was obtained from a theoretical approach based on the geometrical arrangement of spherical particles in a cylinder.

Most of the published correlations for the prediction of ϵ_{mean} in beds of spherical particles, and the very few in beds of cylindrical particles suffer from one or more of the following deficiencies. There is either lack of sufficient data points within the reported range; or no information is available on the end effect; or there is considerable scatter of the experimental data; or the application of the expression is limited to a very narrow range.

2.5 Local Voidage Review

The experimental measurements of Roblee et al., (1958); Benenati and Brosilow, (1962); Ridgway and Tarbuck, (1966); Thadani and Peebles, (1966); Kondelik et al., (1968); Pillai, (1977); Lerou et al., (1980); among others have clearly indicated the presence of oscillatory radial variations of the void fraction in packed beds which is because of the effect of the confining wall of the bed.

The microscopic structural details of packed beds have been given noticeable consideration over the last three decades, but were never fully explored before. In recent years, however, there has been a trend towards a more accurate description of the bed's structure to explain and to simulate the behaviour of fixed beds. Lerou and Froment, (1977); showed how predicted temperature and concentration profiles in reactors may differ depending upon whether non-uniform voidage profiles were accounted for or not. Carbonell, (1980); and Change, (1982); used the structural

properties of the bed to explain the dispersion phenomena. Kalthoff and Vortmeyer, (1980); introduced structural characteristics of the bed to improve their models for creeping flow in reactors, also Vortmeyer and Winter, (1982), incorporated such details in a 2 dimensional model of fixed beds and examined the significance of descriptive models. Cohen and Metzner, (1981); accounted for non-uniformities of the bed in the pressure drop relationship for both Newtonian and non-Newtonian fluids. Beasley and Clark, (1984); found that spatial variation in void fraction significantly influence the dynamic response of both fluid and solid temperatures in packed beds. Snaddon and Dietz, (1984); took the structure of the bed into account when they measured the interstitial flow intensification within packed granular bed filters.

There are generally four different methods described in the literature for the determination of local voidage distribution in packed beds.

i) Incremental filling -

Shaffer, (1952); measured the voidage of a packed bed of spheres by rotating the column 90° to a horizontal position and introducing a known amount of water into this section, raising the liquid level in increments. By assuming that the voidage would be the same everywhere at the same radial position, and introducing imaginary annular sections of the same width as the height of the liquid level increments, the void fraction was estimated as an average value for an annular area. This method was improved by Ridgway and Tarbuck, (1966); through rotating the cylindrical bed rapidly along its axis while filling the voids incrementally. The void fraction was deduced from the increase in level and volume. The accuracy of the data depends heavily upon the smoothness of the rotation of the cylindrical bed. A major disadvantage of this method is the inability to measure small changes in the thickness of the rotating water layer. Griffiths, (1986);

improved the technique even further by applying a speed motor to increase the speed of rotation to 3000 rpm. Although the experimental rig was improved but the accuracy of the radial void fraction measurements were limited due to the quantity of water added and the position of the water meniscus.

ii) *Solidification and incremental removal -*

This is the most commonly used method which involves immobilization of the packing with a slowly solidifying fluid such as wax or epoxy resin and then progressively reducing it by turning. The method originated with the work of Kimura et al., (1955); where crushed calcium carbonate was loaded into a glass tube and the void spaces filled with paraffin wax. The results indicated that irregular particles produce voidage patterns with a unity value at the wall which decreases gradually to constant bulk value of 0.4 as the bed centre is approached. Roblee et al., (1958); improved the method by putting emphasis on weighing the annular fractions to find the solids volume. After solidification the bed was sawed into circular slabs which were then machined into concentric annuli. The wax was then separated from the packing by dissolving it in benzene. Their work indicated that the radial voidage profiles for spheres and cylinders are distinctly different from those of irregular particles. They reported loss of some particles due to weak bonding with the wax during the machining operation, which has a significant effect on the accuracy of their data. Benenati and Brosilow, (1962); improved the bonding by using lead shots and epoxy resin which produce a more resilient bed. After each annular cut, the collections were weighed to calculate the voidage of that particular annulus by means of a mass balance on the density of the cuttings produced. They exhibited the same profiles for the radial

variation of voidage as Roblee et al., (1958); but with less scatter of data points. There are a number of drawbacks associated with their technique such as no attempts were made to obtain any specified mode of packing into the container, nor was the container vibrated to get a more settled bed which regularly occurs in industrial practice. They also reported loss of lead shots in every machining stage due to weak bonding which affects the validity of the data. They only considered two beds with aspect ratios, d_t/d_p , within the industrial range. They were faced with broken beds while approaching the core zone, therefore vital information was lost in beds with small diameter ratio. More importantly, in using such an approach local structural variations of voidage in the axial as well as the angular directions together with the influence of the end effect cannot be identified. Korolev et al., (1971); Propster and Szekely, (1979); adopted the same procedure and established data which agree well with the data of Benenati and Brosilow, (1962). Goodling et al., (1983); added iron filings to the epoxy resin to improve the bonding and produced similar profiles to those discussed earlier for beds of mono-sized spheres. No systematic trend with diameter ratio of the system geometry was observed.

Moallemi, (1989); improved the bonding by sand blasting the spherical lead shot and obtained very accurate data points by means of an image analyser. A large number of beds spanning the industrial range were examined. One of the major advantages of his work was the ability to examine the local variation of voidage in the axial as well as the radial direction.

iii) Individual particle measurement -

Pillai, (1977); constructed a perspex bed with a measuring grid scribed on its outside face. After the introduction of wooden discs into the bed

and random shaking, the distribution of the discs at the wall was quantified by counting the number of the particles having their centres between successive grid lines. Counts were made up to 3 particle diameters away from the wall, and from each count mean solid cross sectional area along the plane is calculated by means of an expression which will eventually give the area void fraction at any point within the bed. Therefore, the spatial location of the individual particles in the fixed bed are determined and the local distribution of voidage is derived by integration. Schuster and Vortmeyer, (1980); removed particles of a packing assembly from the top with a sticky probe. Limited information is available on their report regarding this technique, therefore no conclusive remark can be made on the reliability of their findings.

iv) *Projection of the bed -*

There are various possibilities of passing light or other electromagnetic radiation through a fixed bed. These can be used to produce projections in which the varying transparency of the bed can be attributed to fluctuations in the voidage. In this way, photographs of the local average density can be produced. Thadani and Peebles, (1966); used X-rays which could shine straight through horizontal slabs of a packing. The differential adsorption produced by the particles and resin is analysed by photomicrographic scanning to give a point radial voidage profile which is then azimuthally averaged. They observed a more damped oscillatory pattern for packed beds of mono-faced spheres, by about 10%, when compared to previous reports. This could be due to the irregular nature of the particles used, which could culminate in the reduction of the radial ordering and damping of the void fraction profiles. Buchlin et al., (1977); used the projection effect

to obtain an optically homogeneous bed. They then illuminated segments of the bed containing a fluorescent fluid with a laser in order to reveal the location of the individual spheres in one cross section. Schneider and Rippin, (1988); developed an optical method to demonstrate the voidage distribution in a packed bed of glass spheres based on the refractive index matching of the glass spheres and a fluid with the same optical properties as the solid. The voidage distribution was shown by colouring the liquid or using coloured glass particles on projection of the bed. The brightness of any point on the image is a measure of the light absorption by the colourant along the projection axis and therefore gives the average local voidage of the bed along this line. This method, however, is only applicable to shallow beds of particles, since increasing the height would increase the light scattering which will lead to loss of definition in the image. There is also the problem of resolution due to projection. They sacrificed the accuracy of the data for the simplicity and speed of the technique.

The work of most of the investigators using the four main techniques described above suffer from one or more of the following deficiencies:

- a) cannot measure either the influence or the extent of the entrance/exit effect;
- b) cannot account for variation of voidage in axial direction;
- c) cannot consider thin sections of the bed in axial and radial directions;
- d) result in total destruction of the test specimen;
- e) only systems with radial symmetry can be examined;
- f) cannot produce the full profile of radial voidage distribution.

2.6 Local Voidage Correlations Review

The local voidage variation gives rise to the so-called wall effects in the flow of fluids through the bed, and in the dispersion of heat and mass in the bed, and thus becomes a very important factor in the analysis and design of industrial packed beds such as reactors, absorbers, distillation columns, adsorbers, heat transfer equipment, and in the analysis and interpretation of data from laboratory and pilot plant beds. There have been several reports on this subject to establish expressions for prediction of such variations.

Schwartz and Smith, 1953; concluded that the velocity profile for gases flowing through a packed bed is not flat, but has a maximum value approximately one pellet diameter from the tube wall. In order to test their theory, the experimental velocity profiles near the wall were used to predict how the void space varies with radial position. The following equation was derived to define the velocity gradient in terms of void fraction:

$$\left| \frac{du}{dr} \right| \frac{du}{dr} = \frac{9}{4} k^2 \left(\frac{1}{R} \right)^3 \left[u^2 \frac{(1-\delta) \delta_o^3}{(1-\delta_o) \delta^3} - u_o^2 \right] r \quad (2.14)$$

where,

$$k \left(\frac{r}{R} \right)^{3/2} = \ln \left[\frac{u + (u^2 - u_o^2)^{1/2}}{u_o} \right] \quad (2.15)$$

r = radius of particle, R = radius of tube

u = point velocity at radius r

u_o = point velocity at centre of pipe

δ = void fraction at radius r

δ_o = void fraction at centre of bed.

A numerical stepwise integration procedure was then employed in Equation

(2.14), starting the process at a position two pellet diameters from the wall. For each increment of radial position, the value of δ was determined that would give exact agreement with their experimental velocity profile. These calculations lead to a series of values of void fraction and corresponding radial position for each combination of packing size and pipe size. Equation (2.14) however, does not hold for r/R values much greater than those corresponding to the peak velocity. To carry the calculations closer to the wall it would be necessary to revise the equation to take into account a variable mixing length.

Ridgway and Tarbuck, (1968); proposed a model for randomly packed beds of spheres located on a wall, based on the assumption that the bed can be subdivided into discrete but interpenetrating layers. The voidage for each layer could be calculated, and by adding the voidages of adjacent layers where they overlap, an analysis of the voidage variation could be made. The following relationship was derived using Benenati and Brosilow's (1962) experimental data for a flat wall:

$$1 - \epsilon_x = \sum_0^P \frac{F_1 \pi}{2 \sqrt{3} r^2} (x - 2\sqrt{2/3} p r F_2) (2r - x + 2\sqrt{2/3} p r F_2) \quad (2.16)$$

where,

$$F_1 = \frac{2\sqrt{3}}{\pi} (0.62 + 0.18 e^{-0.36p}) \quad (2.17)$$

and

$$F_2 = 0.991^p \quad (2.18)$$

ϵ_x = voidage at distance x from the wall,

p = layer number, 0 at wall,

r = sphere radius,

x = distance from the container wall.

However, for a cylindrical wall, the relationship is as follows:

$$1 - \epsilon_z = \sum_0^P \frac{F_1 \pi}{2 \sqrt{3}} (z - 2\sqrt{2/3} p F_2 - \Delta F_2) (2 - z + 2\sqrt{2/3} p F_2 + \Delta F_2) \quad (2.19)$$

where,

$$z = x/r \quad (2.20)$$

ΔF_2 = difference between a cylindrical and a flat wall boundary. The introduction of the term ΔF_2 is difficult to justify theoretically, though it is quantitatively necessary. The agreement between Equation (2.19) and the experimental data is not so good. The application of this expression is limited to $d_t/d_p = 17.3$ and the format is cumbersome.

Kondelik et al., (1968); investigated the dependence of local void fraction in beds of uniform equilateral cylinders upon the distance from the wall on a series of randomly packed beds of different d_t/d_p ratios. They determined that this dependence is of a periodic nature and the minimum void fraction is between 0.1 and 0.2. The distance of this minimum from the wall is found to be greater in beds of cylinders than in beds of spheres by the following expressions:

$$2(\delta - 1/2) = (\psi - 1) - \sqrt{(\psi - 1)^2 - 3} \quad (2.21)$$

for $\psi \geq 3.309$

and

$$2(\delta - 1/2) = (\psi - 1) - \sqrt{\psi^2 + 4(1 - \sqrt{\psi^2 - 1})} \quad (2.22)$$

for $\psi < 3.309$

where,

$$\psi = d_t/d_p$$

δ = distance of 1st minimum from wall in terms of cylinder diameters.

They have therefore concluded that the wall region in beds of cylinders is wider than in beds of spheres.

Johnson, (1970); derived the following expression based on the data from Benenati and Brosilow, (1962):

$$\epsilon(r) = a_1 + a_2 e^{a_3(r/d_p)^{a_4}} \cos(a_5(r/d_p)^{a_6}) \quad (2.23)$$

for $r/d_p \leq 4$

and

$$\epsilon(r) = 0.38 \quad \text{for } r/d_p > 4 \quad (2.24)$$

where

$$\begin{aligned} a_1 &= 0.38, & a_2 &= 0.62, & a_3 &= -1.7 \\ a_4 &= 0.434, & a_5 &= 6.67, & a_6 &= 1.13 \end{aligned}$$

The expression is based on poor experimental data and its application is limited.

Marivoet et al., (1974); measured the velocity profile above a packed bed of spheres and included them in a mathematical model to predict the temperature profiles of the bed. They concluded that the gas flowing through the packing follows paths of least resistance between adjacent layers, so that the maxima of its radial velocity profile correspond to the maxima of the radial voidage profile. The radial temperature profiles also revealed the influence of the periodic structure of the packing. Their findings are, however, limited to a diameter ratio of 10.

Kubie, (1974); proposed a geometrical function for the prediction of radial variation of voidage in packed beds of spheres up to a distance of a particle diameter away from the wall:

$$\epsilon = 1 - [3(1 - \epsilon_b)(x - 2/3 x^2)] \quad (2.25)$$

for $x \leq 1$

and

$$\epsilon = 0.38 \quad \text{for } x > 1 \quad (2.26)$$

where

$$x = r/d_p$$

r = radius of the tube,

d_p = diameter of particle.

Lerou and Froment, (1977); measured velocity and temperature profiles perpendicular to the flow direction at the exit of a packed bed of spheres. The radial temperature profile exhibited two pronounced humps which were modelled by means of an energy equation containing a velocity profile inversely proportional to the void fraction profile and point values of the effective radial thermal conductivity. Their heat transfer model containing the local void fraction and velocity leads to values of wall heat transfer coefficient and radial effective thermal conductivity.

Pillai, (1977); calculated the area void fraction based on measurements of the number of particles having their centres lying between two successive planes.

Martin, (1978); improved on the work of Ridgway and Tarbuck, (1968); and presented empirical expressions to represent the void fraction in packed beds of spheres:

$$\psi(z) = \psi_{\min} + (1 - \psi_{\min}) z^2 \quad (2.27)$$

$$\text{for } -1 \leq z \leq 0$$

where

$$z = 2(y/d) - 1 \quad (2.28)$$

and

$$\psi(z) = \psi_{\infty} + (\psi_{\min} - \psi_{\infty}) e^{-z/4} \cos \frac{\pi z}{T} \quad (2.29)$$

$$\text{for } z > 0$$

with $\psi_{\min} = 0.23$ and $\psi_{\infty} = 0.39$ and $T = 0.816$ for $d_t/d_p = \infty$

y = distance from the wall, m

d = particle diameter, m.

Since the above are based on a limited set of data, its general application cannot be guaranteed.

Chandraskhara and Vortmeyer, (1979); presented the radial variation of voidage to the integrated void fraction data of Benenati and Brosilow, (1962) by:

$$\epsilon = \epsilon_0 (1 + b e^{-c\gamma/d_p}) \quad (2.30)$$

where,

ϵ_0 = void fraction at the centre of the bed,

γ = distance from the wall,

b and c = constants = 1 and 3 for $d_t/d_p = 14$ and 21.

The application of their expression is limited to diameter ratios of 14 and 21. Kalthoff and Vortmeyer, (1980); measured temperature profiles in a wall cooled fixed bed reactor and compared it with the solutions of a pseudohomogeneous 2-dimensional model which took into account the radial variation of voidage using Equation 2.30 .

The conversion rate, r , near the wall is represented by:

$$r = r(\bar{\epsilon}) \frac{1 - \epsilon}{1 - \bar{\epsilon}} \quad (2.31)$$

where $\bar{\epsilon}$ = mean voidage.

They concluded that the employed radial porosity and velocity functions improved the credibility of their model considerably. Cohen and Metzner, (1981), reported the effect of the structural non-uniformities on the flow characteristics of packed beds. They proposed the following to represent the variations:

$$\frac{1 - \epsilon}{1 - \epsilon_b} = 4.5 \left(x - \frac{7}{9} x^2 \right) \quad (2.32)$$

for $x \leq 0.25$

and

$$\frac{\epsilon - \epsilon_b}{1 - \epsilon_b} = a_1 e^{-a_2 x} \cos (a_3 x - a_4) \pi \quad (2.33)$$

for $0.25 < x < 8$

and

$$\epsilon(x) = \epsilon_b \quad \text{for } 8 \leq x \leq \infty \quad (2.34)$$

where,

ϵ_b = bulk porosity,

x = distance from the wall in terms of particle diameter

$a_1 = 0.3463$, $a_2 = 0.4273$, $a_3 = 2.4509$, $a_4 = 2.2011$.

The reported expressions do not show any trend with the diameter ratio of the system.

Govindarao and Froment, (1986); proposed a method to describe voidage variations in randomly packed beds of spheres in terms of the number of fractions with centres lying in prescribed concentric layers of the bed. They derived correlations for estimating the number fractions from a given aspect ratio.

$$\epsilon_i = 1 - \frac{h}{g_i} \left[n_{i+m} \left(m - \frac{1}{4} \right) + 3 \sum_{j=m+1}^{i+m-1} n_j b_{ij} \right] \quad (2.35)$$

for $i = 1$ to $2m$

and

$$\epsilon_i = 1 - \frac{h}{g_i} \left[(n_{i-m} + n_{i+m}) \left(m - \frac{1}{4} \right) + 3 \sum_{j=m+1}^{i+m-1} n_j b_{ij} \right] \quad (2.36)$$

for $i \geq 2m$

where

$$b_{ij} = m^2 - i^2 + j(2i - j) - 1/6 \quad (2.37)$$

$$g_i = 2am - 2i + 1 \quad (2.38)$$

$$h = N \Delta r / 3L \quad (2.39)$$

m = number of cylindrical concentric layers (CCL),

N = total number of spheres in the bed,

n_j = number fraction in the j th CCL,

Δr = thickness of a CCL,

a = aspect ratio,

L = length of the bed.

The correlation, however, estimates the voidage variations up to two particle diameters away from the wall, beyond which the predictions are only approximate.

A couple of semi-empirical correlations were proposed by Griffiths, (1986); for the prediction of radial voidage in the wall and the core zones of a packed bed of spherical particles:

$$1 - \epsilon_y = \frac{(1 - \epsilon_b) \{6 - 2.38 (d_t/d_p)^{-0.43}\} (2 y_m y - y^2)}{2 y_m d_p} \quad (2.40)$$

for $0 \leq y \leq y_m$

$$\epsilon_y = 0.38 + (\epsilon_m - 0.38) \cos \left(2.27 \pi \frac{y - y_m}{d_p} \right) e^{\frac{-0.26(y - y_m)}{d_p}} \quad (2.41)$$

for $y_m < y \leq d_t/2$

where ϵ_m is given by Equation (2.40) at $y = y_m$.

The range of the application of the above correlations is very narrow, i.e. $d_t/d_p < 8$.

Moallemi, (1989); derived a general correlation for the prediction of radial voidage in beds of spheres based on his experimental data:

$$\epsilon = 1 - \beta_1 [1 - e^{-\beta_2 x^{\beta_4}} \cos(\beta_3 x^{\beta_5})] \quad (2.42)$$

where,

x = distance from the wall.

The values of coefficients β_1 to β_5 were optimised and presented in a table with respect to diameter ratio, d_t/d_p . His correlation is applicable to a wide range of aspect ratios and based on accurate data.

Kufner and Hofmann, (1990); proposed the following relationship based on Vortmeyer and Schuster, (1983); to describe the oscillations of the voidage near the wall region:

$$\epsilon = \bar{\epsilon} \left[1 + c \exp \left(1 + \frac{R-r}{d_p} \right) \cos \left(\frac{R-r}{d_p} 2\pi \right) \right] \quad (2.43)$$

where,

$$c = \frac{1 - \bar{\epsilon}}{\bar{\epsilon} \exp(1)} \quad \text{for } r = R$$

r = distance from the wall,

R = radius of the bed,

ϵ = mean voidage.

The accuracy of the experimental data used for deriving these correlations is, however, under question.

2.7 Concluding Remarks

In order to obtain detailed information on the local and global structural properties of packed beds, a comprehensive literature review has been carried out. In all the reports on this subject, the voidage variations were accounted for by using either the data reported by Benenati and Brosilow, (1962); or by empirical equations, ranging from a simple power-law type or exponential equation to more complicated expressions involving trigonometric functions. An analysis of the voidage variations can be done by modelling either the structure of the void spaces, or the location of the particles. The reliability of the few reports available on the data is under question as the techniques employed for the measurements of local voidage variations have their own weaknesses. Nearly all the reports consider beds packed with unisized spheres, whereas nearly two thirds of the particles being used in industry are of cylindrical nature. Even there the range of dimensionalities of the beds considered are very limited. There is therefore, a need to overcome the existing deficiencies by employing more sophisticated techniques to ascertain reliable data and hence derive correlations for the prediction of the local and global structure of packed beds of cylindrical particles.

CHAPTER 3

EXPERIMENTAL METHOD

CHAPTER 3

EXPERIMENTAL METHOD

3.1 Introduction

The previous chapter established the importance of knowing the local variation of voidage in packed bed systems for the design and performance prediction purposes. It is therefore essential to ascertain accurate and representative data in order to characterise the packing structure. The deficiencies associated with the techniques adopted by earlier investigators (Shaffer, 1952; Roblee et al., 1958; Benenati and Brosilow, 1962; Thadani and Peebles, 1966; Ridgeway and Tarbuck, 1968; Buchlin et al., 1977; Schuster and Vortmeyer, 1981; Goodling et al., 1983; Schneider and Rippin, 1988) have already been discussed. Rapid development in computer technology has led to a considerable evolution in image analysis techniques. This is a process for deriving numerical information from images and quantifying their parameters, e.g. size, shape and number - for subsequent identification and characterisation. The technique has become an indispensable tool and has found application in such diverse fields as medicine, science and engineering. A sophisticated, high resolution Image Analyser is used here to study the microstructural details within packed beds and hence identify and characterise the desired local properties. The hardware and software of the Image Analysis, IA, system together with the way the sample materials are chosen and prepared are described in this chapter.

3.2 System Hardware

The hardware of the system which consists of a video digitizer, VDU camera, colour and monochrome video monitors, micro computer together with a

framestore and a video printer is illustrated in Figure 3.1; the connections between them are shown in Figure 3.2. The micro computer with the framestore installed in it is the focal point of the system. It gets an input line direct from the video camera and has four output lines to the keyboard, printer, monochrome and colour monitors which in turn is connected to the video printer. Figure 3.3 shows a photograph of the system setting with the way a sample bed is placed underneath the video camera. In the case of monochrome video input, the camera is connected to the top phono socket on the framestore connection panel at the rear of the computer. The colour monitor is connected via the SCART cable to the multi-way D-type connector located beneath the phono connectors also on the framestore connection panel. The DATAKEY software lock is connected to the serial port of the computer, and the mouse to the input side of the datakey device. Finally, the dot matrix printer is connected to the computer parallel port. Moallemi, 1989, has described the hardware of the system in more detail.

3.3 System Software

The software of the system allows an image to be captured by the video camera and stored onto the memory of the system and also on a floppy diskette by option, so that the image can be recalled at any stage for further analysis. Therefore, the principal stages of image analysis based techniques are image acquisition, image refinement, measurement and analysis. In general, desired images are captured and subsequently digitised and stored as arrays of pixels, ready for the necessary manipulations. Resolution of the system and capability of the software are the most important considerations in selecting a suitable image analysis device. The system used here is the one marketed by Oxford Framestore Applications Ltd.

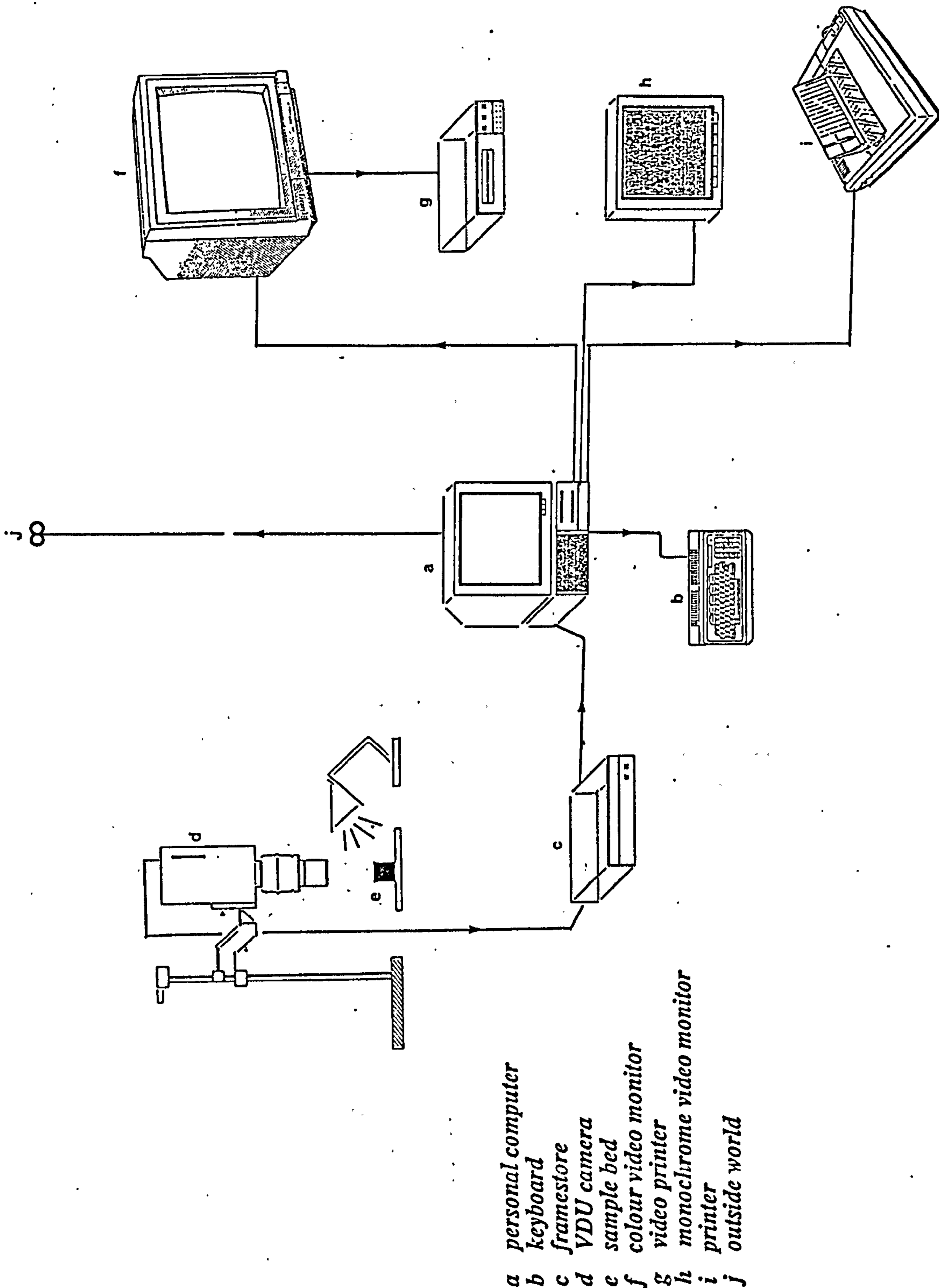


FIGURE 3.1: The Hardware of the Image Analysis System

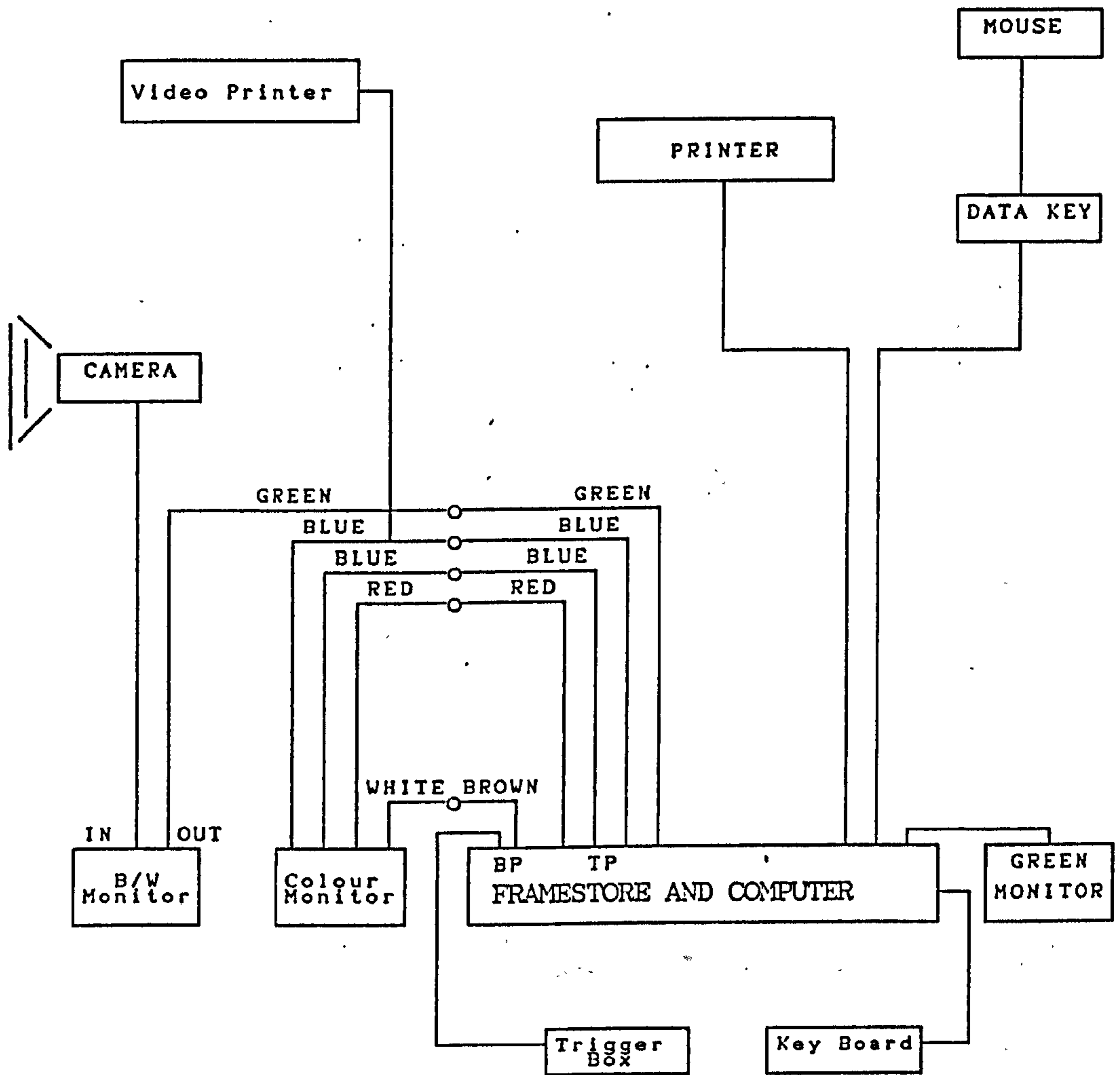


Fig. (3.2) Image Analysis Connection Diagram



FIGURE 3.3: Photograph of the System Setting

The memory can be divided into four segments, each capable of holding a single 256 x 256 element image, or two segments each of 512 x 256 pixels or one of 512 x 512 pixels depending on the resolution required. The framestore can thus hold multiple images, and the software allows images to be copied from one area to another, and display the desired segment. The main menu and all the sub-menus contain options to call these image housekeeping routines.

The video memory is dual-ported, that is to say that there are two independent data channels to the data stored in memory. One such channel drives a digital to analogue converter, and provides continuous video output to a monitor. The contents of the video memory are thereby displayed at all times. The other channel is dedicated to communications with the host micro-computer, giving the programme running in the processor access to the frame data for image processing and analysis.

Once the image is stored, analysis of the image can take place independently of the video input. The system is therefore well suited to single event capture and data taking, with subsequent data analysis.

The framestore could also be configured to perform certain real time operations at full video rate. This includes image subtraction, and image averaging. In addition, there is dedicated hardware on the framestore board to enable convolutions and histograms to be performed extremely rapidly. The video input to the framestore is via a digitiser, which converts the analogue video voltage to a digital signal, and a look-up table (LUT) which maps the digitiser output to the grey level bit pattern which is stored as the image information. The output from the framestore is via a separate look-up table and digital to analogue converter which rebuilds the signal to the appropriate video standard.

The LUT mapping functions are implemented in hardware, this makes possible certain video rate enhancement techniques. One obvious application of the output LUT is for the generation of false colour. The options available through the

input LUT include real-time image enhancement and negation and are controlled through a sub-menu of the system parameters menu.

Some routines, such as the line cross-section measurement, calculate parameters from the image and display the result on the computer monitor. A new image can then be captured, and the computer display updated. Such operations are particularly useful for on-line systems monitoring and diagnostics. Other processes, such as contrast stretching, averaging, subtraction and false colour can operate at true video rate.

Software may be driven by the mouse using the menus, with additional keyboard input, for file names entered from the keyboard in response to prompts. The menus are based on a tree structure, the top level being referred to as the 'MAIN MENU', with sub-menus for the control of system hardware, image input/output, image modification etc. Prompts are issued when the user is required either to make a simple choice, e.g. horizontal or vertical processing, in which case a dedicated menu would be inappropriate; or when detailed information is required such as the choice of the range of contrast enhancement.

Menu options are used where the operator is guiding control of the system through the software library, and options are most easily selected by the use of the mouse, alternatively the appropriate function keys could be used. The mouse has other important functions in addition to menu control. The interface to set the required window for windows processing uses the mouse for the movement of window boundaries; cursor and cross movement is implemented via the mouse, as is entry of user defined values within convolution matrices.

3.3.1 Measuremental approach

The surface of a cross section of a test sample is magnified to the maximum available size of the VDU screen to accomplish the highest possible resolution. The image is then captured by the video camera and stored on a diskette so that it can be

retrieved for the purpose of analysis at a later stage. The analysis is based on the colour contrast principle. The area of interest on the captured image can be framed rectangularly and then circularly enabling further operations to be performed on the selected region. After choosing a grey-level, the software displays the image in binarised form, therefore the image is segmented into two intensity states. After the image is calibrated, the software can assemble up to 300 consecutive annular rings corresponding to the pre-calibrated area of the image and work out the number of white and black pixels on each ring. The ratio of the number of black pixels to the white ones in a given annulus gives the local area voidage, $\epsilon(x,r)$. Axially averaging local area voidage of consecutive annular rings of each and every corresponding section results in radial voidage profiles, $\epsilon(r)$, for the whole of the sample bed.

This approach provides a 3-dimensional history of voidage, $\epsilon(x,r,\theta)$, in axial, radial and angular directions. Suitable averaging can provide other relevant data such as $\epsilon(x,r)$, $\epsilon(x)$, $\epsilon(r)$ or ϵ_{mean} . The uniqueness of this approach is mainly due to the quality of the data. One of the major advantages of this technique over the published ones is its ability in retrieving and subsequently analysing the stored images, plus the fact that very accurate information regarding the area of interest can be obtained by this modern, sophisticated and highly resolved system.

3.4 Preparation of Test Beds

The technique used is that of solidification and step-wise removal of the packed bed. Rods of lead were accurately cut by a designer cutter to obtain sharp-edged equilateral cylindrical particles. These were then poured into a PVC cylindrical container and the interstices were subsequently filled with dyed liquid epoxy resin. The bonding material is of low viscosity and high strength properties. To prepare the liquid resin, 100 parts by volume of the epoxy resin is added to 45 parts of Araldite hardener and thoroughly mixed to lower the viscosity of the liquid

prior to its pouring into the PVC mould. The resulting liquid is then dyed so that the colour contrast between the shiny surface of the lead particles and the dark epoxy resin filling the interstices is distinctive. The bonding liquid is then poured into the particulate bed. The entire casting is then placed under a vacuum or to remove any air pockets created within the voids.

After a successful de-aeration operation the casting is left to solidify. Removing the PVC mould resulted in a solidified packed bed, ready to be machined. A standard lathe machine running at its maximum RPM of 2500 is used for shaving off the surface of the cross section of the sample bed at equal intervals, 1 mm to be precise. In order to remove the desired height off the top of each bed, i.e. 1 mm, between 1 to 5 shavings were performed. The depth of each cut was in connection with the size of the cylindrical particles used in the bed, i.e. the beds with smaller particles had to be cut in smaller portions. It was important to use a sharp bit in taking axial cuts to produce a smooth surface for polishing. Upon completion of an axial cut, the surface was polished by silicon carbide papers and diamond paste wheels of various sizes in order to achieve the best possible contrast between the two areas under the video camera. Figures 3.4 and 3.5 show the cross section of a couple of packed beds of equilateral cylinders. The orientation of the cylindrical particles is responsible for such varied shapes. The cutting and polishing procedure is repeated for each and every cross-section of the entire bed.

3.5 Experimental Approach

In order to establish a better insight into the local variation of voidage, a number of test beds with different diameter ratios, d_t/d_{pe} have been prepared in the same manner as mentioned earlier. The dimensionality of the physical beds, i.e. diameter of the tube, d_t , and the equivalent diameter of the particle, d_{pe} , have been selected to cover the diameter ratios being used in practice. A total of ten beds have been prepared spanning a range of diameter ratios, d_t/d_{pe} , from 3 to 30. The

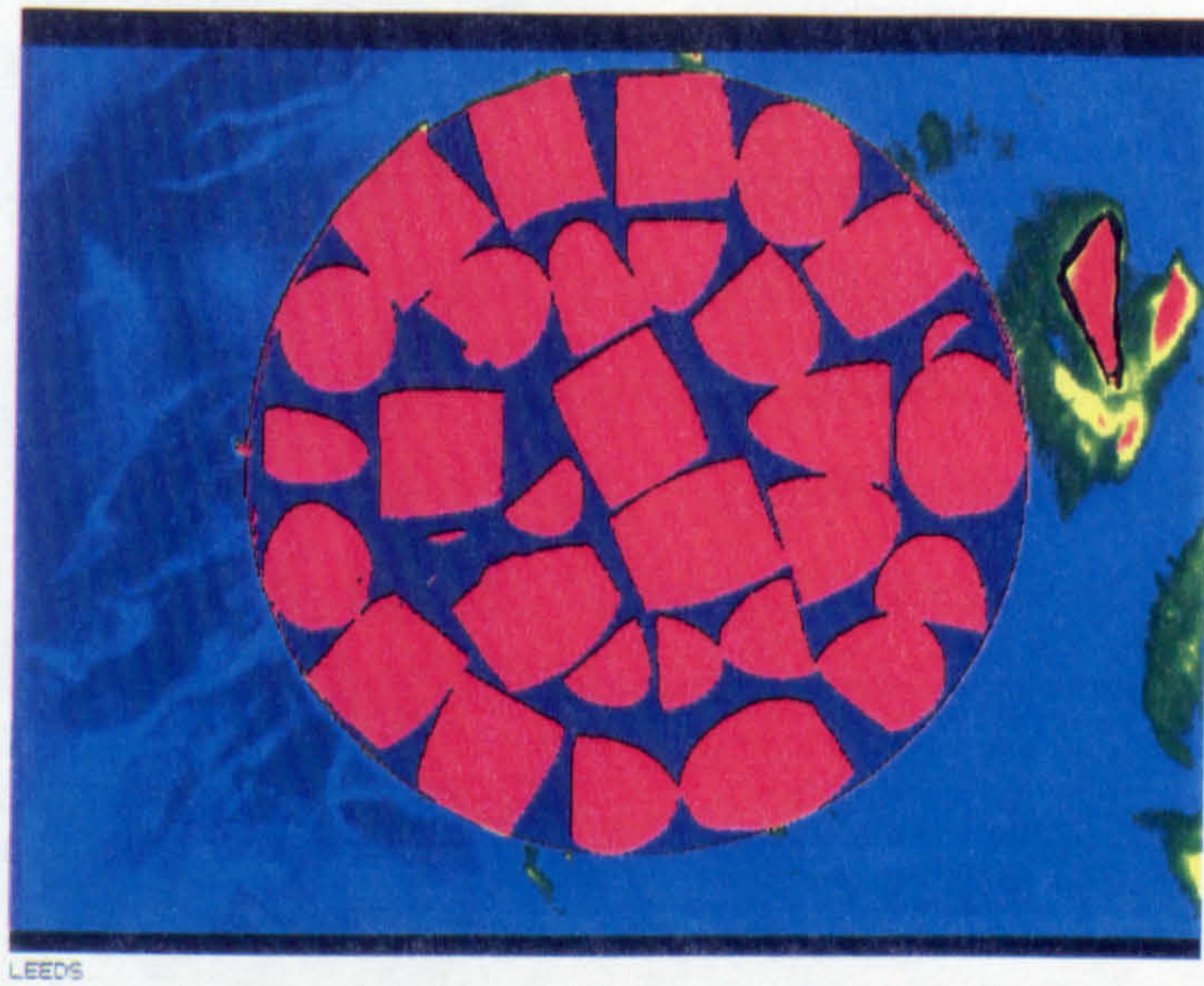


FIGURE 3.4: A Cross-Section of the Bed EC



FIGURE 3.5: A Cross-Section of the Bed HC

purpose of the first three beds made were to examine the effect of reproduction and scaling-up of packed beds on their structural properties. Two identical beds of 39 mm in diameter comprised of equilateral cylinders of 4.58 mm, i.e. beds AC and BC, Table 3.1, have been prepared to examine the reproducibility of bed structure, while bed CC with a scale-up factor of 2, i.e. $d_t = 78$ mm and $d_{pe} = 9.16$ mm, is constructed to provide the necessary information regarding the scaling of the particulate systems. The dimensionalities used give an equivalent diameter ratio, d_{re} , of 8.5 for all three beds.

Another seven solid beds have been made with the size of tube and particle varying between 49 - 128 mm and 4.58 - 14.54 mm, respectively. Inspection of these beds will generate information necessary for the structural characterisation of the packed bed. A detailed account of the beds under investigation is given in Table 3.1. To make the analysis more comprehensive and easier, four of the beds were made having the same d_{pe} and four with the same d_t while varying the other parameter. This reduces the number of variables and hence an element of complexity in the analysis stage.

In order to obtain detailed and representative information on the local properties of each bed, no cut deeper than 1 mm was taken. This will give a clear picture of the end effect. Detailed treatment and analysis of the data obtained from these beds is presented in the later chapters.

TABLE 3.1
Range of the Test Beds

ID tag	d_t mm	d_{pe} mm	d_{re}	Bed Height mm	Number of Cuts*
AC	39	4.58	8.5	80	74
BC	39	4.58	8.5	80	74
CC	78	9.16	8.5	100	89
DC	49	14.54	3.37	85	75
EC	49	9.16	5.35	78	68
FC	49	6.98	7.02	74	70
GC	49	4.58	10.7	62	61
HC	66	4.58	14.41	57	56
YC	101	4.58	22.05	39	38
ZC	128	4.58	27.95	41	40

*cutting at 1 mm intervals

CHAPTER 4

IDENTIFICATION AND CHARACTERISATION OF VOIDAGE PATTERNS

CHAPTER 4

IDENTIFICATION AND CHARACTERISATION OF VOIDAGE PATTERNS

4.1 Introduction

Since the extent of heat and mass transport is based upon the area of the interphase boundary provided and the packed beds can provide maximum area in a given volume, compact devices can be constructed to bring discrete phases of matter in contact with each other. One such device is the catalytic chemical reactor, the design of which is based upon mechanisms of heat and mass transfer, fluid velocity, and pressure drop of fluid. These mechanisms are subsequently sensitive to the porosity of the bed. Therefore, knowledge of the voidage distribution within a packed bed is important to any rigorous analysis of the transport phenomena in the bed.

It is now generally accepted that the velocity profile near the wall of a fixed bed is strongly affected by the variation in bed porosity caused by the wall, although there is some difference of opinion on the extent of the region over which the variation is significant. Detailed velocity measurements very close to the fixed bed show a very large variation in fluid velocities from point to point in the bed, particularly near the wall (Vortmeyer and Schuster, (1983)). The corresponding interstitial velocity within the bed requires a knowledge of porosity as a function of radius. The true mean velocity may then be calculated from the flowrate and the experimentally established dependence of porosity upon radial position. An adequate understanding and evaluation of packed bed performance thus requires an appreciation of the local as well as bulk packing structure. Knowledge of the size and shape of the free paths available for flow within a packed bed is therefore essential to the development and solution of descriptive mathematical representations of such systems.

Global and local structural properties of packed beds comprised of equilateral cylinders have been examined. The intention of this chapter is to explain the functional dependency of the structural characteristics to the geometrical and dimensional properties of the system. Finally, included in this chapter are the developed correlations which allow the bulk void fraction and the distribution of voidage in axial and radial directions of such beds to be reliably predicted.

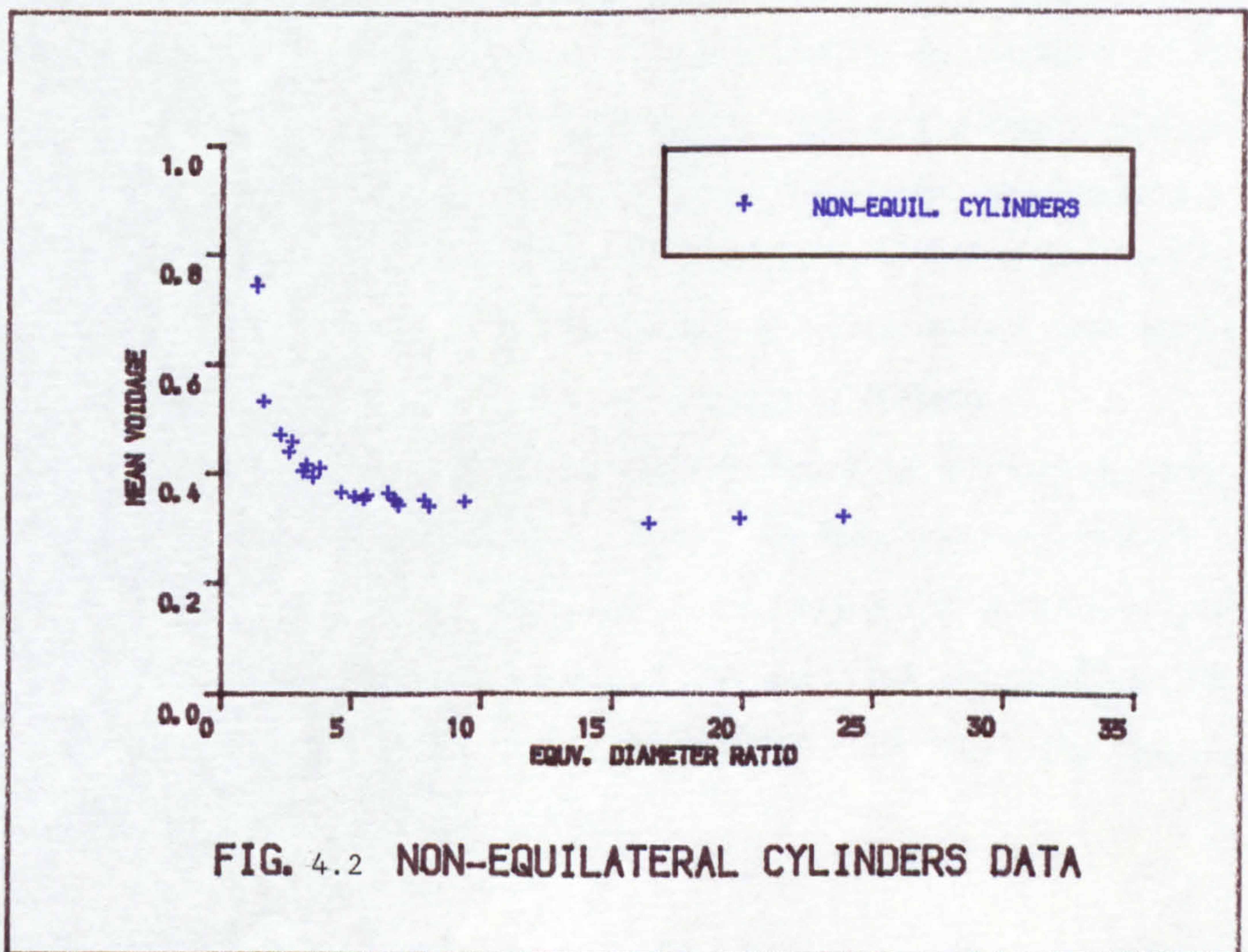
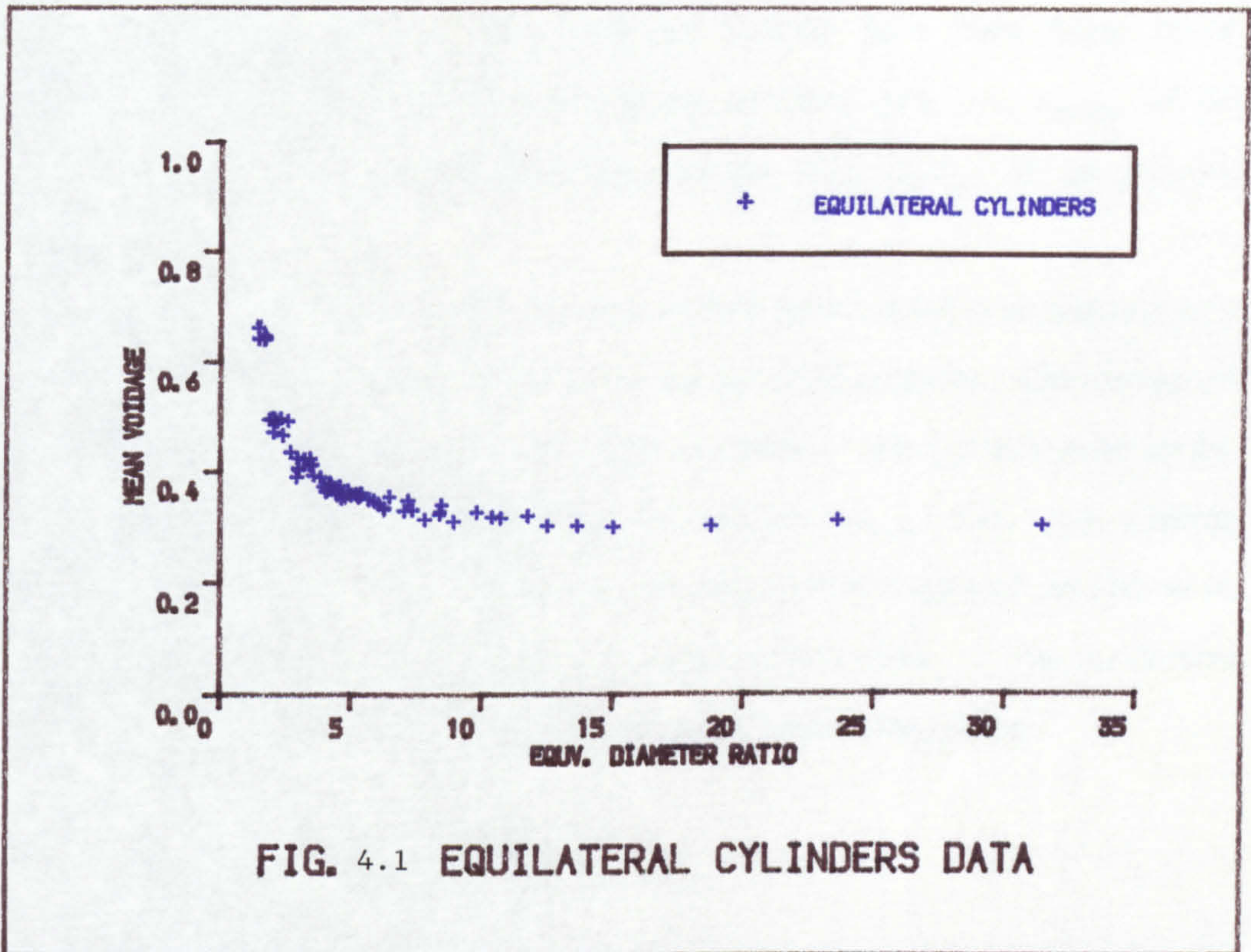
4.2 Global Properties of the Structure

The bulk void fraction or the mean voidage, ϵ_{mean} , which is defined as the ratio of the void volume to the total volume of the packed bed, is a global structural property of packed bed configurations and hence is the quantity most frequently used to characterise the overall space available for flow. Mathematical models of packed beds heavily depend upon having an accurate value for the bulk void fraction as this index is commonly used in making pressure drop as well as heat and mass transfer calculations. Unfortunately, literature fails to provide sufficient information in order to allow this global property to be predicted for a variety of particle shapes and sizes. The few reports available by Leva and Grummer, (1948); Beavers et al., (1973); Beasley and Clark, (1984); and Moallemi, (1989); are mainly concerned with beds comprised of unisized spherical particles. Since nearly two thirds of the packing particles used in industry are of cylindrical nature, and not enough information is available on the structural properties of such beds, then a series of experiments have been carried out in an attempt to ascertain the desired data. For this a wide range of sizes for tube, d_t , and particle, l_p and d_p , are considered, $3 < d_t/d_p < 30$ in order to cover the size ratio being used in practice.

To prepare a test bed, particles were simply poured into a measuring cylinder and gently vibrated to get a more settled bed. The bed mean voidage was then measured by filling the voids with a recorded quantity of water. Care was taken to eliminate any air pockets as this would have had an effect on the accuracy of the

data. Twelve different sizes of very accurate equilateral and varied length-to-diameter ratio cylinders ranging from 4 - 14 mm were used to obtain the mean voidage of over 80 cylindrical packed beds. The employed particles were made of stainless steel having precise dimensions and virtually no size distribution. To ensure a minimum end effect, the aspect ratio of each bed was taken as $L/d_t > 3.0$. Mean voidage, ϵ_{mean} , of each bed was measured at least three times and an average value was used for analysis purposes. Figure 4.1 shows the distribution of the observed mean voidage for equilateral cylinders over a wide range of d_t/d_{pe} , where d_{pe} represents the diameter of spheres of equivalent volume. The displayed results indicate that the mean voidage of such beds decreases exponentially as the diameter ratio increases and approaches a constant mean voidage of 0.30 for $d_t/d_{pe} \geq 10$. It is interesting to note that the criterion for constant mean voidage, i.e. $d_t/d_{pe} \geq 10$, appears to be the same for spherical and cylindrical particles. However, it should be noted that the corresponding beds of such particles take mean voidage values of 0.40 and 0.30, respectively. The difference is due to the compactness of beds formed by cylindrical particles. The enhanced voidage region of the ϵ_{mean} versus d_t/d_{pe} curve is due to the noticeable presence of the container wall. As the diameter ratio increases the wall effect becomes less pronounced. For the extreme condition corresponding to just one particle fitting inside a tube, the mean voidage will be either 0.0 or 0.5, depending on whether the relative axes of the tube and the particle are parallel, $d_t = d_p$, or orthogonal, $d_t = \sqrt{2} d_p$, respectively. For these cases, the corresponding diameter ratios, d_t/d_{pe} , will be 0.87 and 1.23.

The information discussed so far only dealt with equilateral cylindrical particles. To rectify this, the mean voidage of a number of cylinders with different length-to-diameter ratios, $l_p/d_p = 0.5; 1.5; 2.0$, were measured in the manner described earlier. Length of the particles used were 5, 6, 12, mm with the corresponding diameter values of 10, 4, 6, mm, resulting in d_t/d_p ratios mentioned above. The particles were made of stainless steel with precise dimensions. Figure



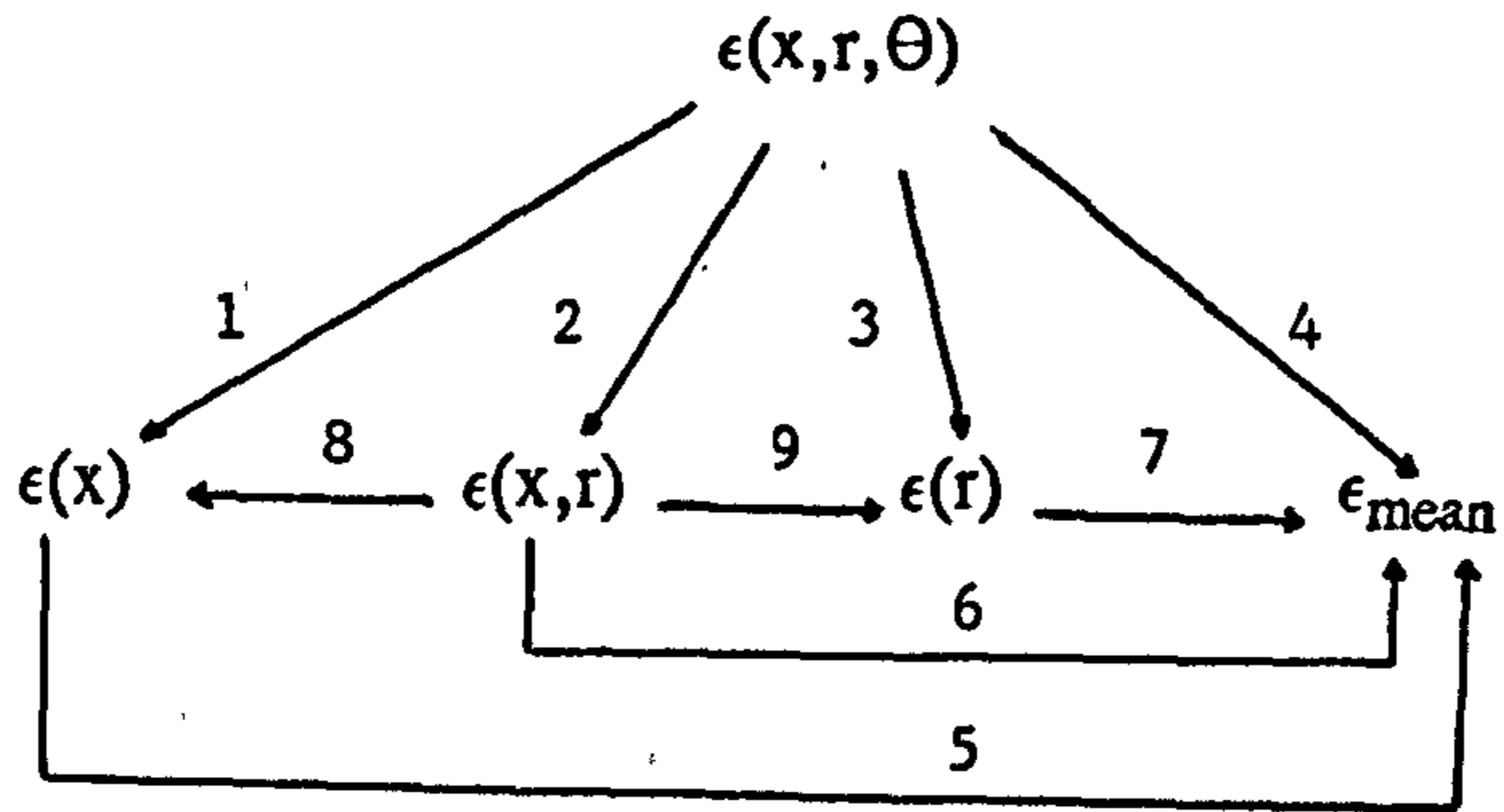
4.2 demonstrates the observed data which follows the same path as that of equilateral cylinders, this data is compared with the published data available in the literature in Chapter 5. These trends and features have been found to be reproducible and it is evident that the global structural property, ϵ_{mean} , of the packed beds is only dependent upon the diameter ratio, d_t/d_{pe} , of the physical system.

Having a good estimate for the value of bulk void fraction is an essential prerequisite to the modelling and design of packed bed configurations. Calculations of pressure drop which influences the pumping cost index is strongly influenced by the value of ϵ_{mean} . This chain dependency between ΔP , ϵ_{mean} , d_t/d_p , and packing shape and size distribution necessitates the development of a general correlation to predict the global structural property of the packed beds reliably. The correlation derived based on the data obtained will be discussed later in this chapter.

4.3 Local Properties of the Structure

Despite intensive work over the past few decades, there is still a great deal of uncertainty as to the reliability of the available data for the prediction of the transport parameters, in particular wall heat transfer coefficient, effective radial and axial thermal conductivity coefficients and radial and axial dispersion coefficients. The published correlations are regarded as unreliable because of being based on poorly defined models in terms of structural characteristics of the bed, such models assume plug flow, i.e. uniform distribution of voidage across the bed.

The assembly of particles that make up a packed bed has structural properties in axial, radial and angular directions, $\epsilon(x,r,\theta)$. However, pattern of voidage in angular direction, $\epsilon(\theta)$, would not be a desirable feature since there is no real use for such information and moreover, experimental studies by Moallemi, (1989), reveal that the nature of this variation is highly stochastic. Therefore, recognising an identifiable pattern for $\epsilon(\theta)$ is not feasible.



$$1. \quad \epsilon(x) = \frac{\int_0^R \int_0^{2\pi} \epsilon(x, r, \theta) d\theta dr}{2\pi R}$$

$$2. \quad \epsilon(x, r) = \frac{\int_0^{2\pi} \epsilon(x, r, \theta) d\theta}{2\pi}$$

$$3. \quad \epsilon(r) = \frac{\int_0^L \int_0^{2\pi} \epsilon(x, r, \theta) d\theta dx}{2\pi L}$$

$$4. \quad \epsilon_{\text{mean}} = \frac{\int_0^R \int_0^L \int_0^{2\pi} \epsilon(x, r, \theta) d\theta dx dr}{2\pi LR}$$

$$5. \quad \epsilon_{\text{mean}} = \frac{\int_0^L \epsilon(x) dx}{L}$$

$$6. \quad \epsilon_{\text{mean}} = \frac{\int_0^R \int_0^L \epsilon(x, r) dx dr}{LR}$$

$$7. \quad \epsilon_{\text{mean}} = \frac{\int_0^R \epsilon(r) dr}{R}$$

$$8. \quad \epsilon(x) = \frac{\int_0^R \epsilon(x, r) dr}{R}$$

$$9. \quad \epsilon(r) = \frac{\int_0^L \epsilon(x, r) dx}{L}$$

The study of axial variations of voidage, $\epsilon(x)$, gives a clear indication of the extent of the entrance/exit effect. Nobody has yet established the existence of $\epsilon(x)$ in packed beds of equilateral cylindrical particles, and hence requires more attention. The radial variations of voidage, $\epsilon(r)$, have been examined and studied by Roblee et al., (1958); Benenati and Brosilow, (1962); Ridgway and Tarbuck, (1966); Thadani and Peebles, (1966); Kondelik et al., (1968); but only recently the importance of this variation was fully realised by Kalthoff and Vortmeyer, (1980); Leron and Froment, (1977); Cohen and Metzner, (1981); Chandrasekhara and Vortmeyer, (1979); Martin, (1978); and hence justifiably found its way into more descriptive and definitive models. They have all clearly indicated the presence of oscillatory radial variations of the void fraction in packed beds due to the wall effect. The voidage in beds of spheres is reported to fall from a value of 1.0 at the wall to a minimum value of 0.2 at $\frac{1}{2} d_p$ away from the wall and attaining a constant value for aspect ratios greater than 10 at a distance of $5 d_p$ from the wall.

The desired model of a packing structure should describe the characteristic properties of the bed in terms of the shape and size of the particles as well as the tube. Most studies into dispersion, pressure drop, and chemical reaction in packed beds have been concerned with spherical particles, and yet in many industrial applications of catalysis the particles are of cylindrical nature. One reason for this is the convenience of manufacture, also there are often a number of distinct process advantages in the use of cylindrical pellets of which the most important are probably uniformity of size. The applications of cylindrical particles is hampered because accurate information necessary for the design is not readily available. As mentioned in Section 3.5, several packed beds comprised of equilateral cylindrical particles were thence prepared to examine their local structural properties.

4.3.1 Reproduction and scaling

Validation would be obtained from the data of sample beds, they must be examined for the effect of reproduction. It is only then that one can continue with the closer examination of the structural properties with confidence. Also to interpret the data obtained from pilot plants with resilience for the full scale industrial application. The effect of scaling up of the beds needs to be investigated.

To examine the reproducibility of the data gathered from a test bed and also the effect of scaling up of the physical geometry of the system, three sample beds were prepared in the manner described in Section 3.4. Two identical beds with $d_t = 39$ mm and $d_{pe} = 4.58$ mm and a bed scaled up by a factor of 2 with $d_t = 78$ mm and $d_{pe} = 9.16$ mm were hence made. Figures 4.3 and 4.5 illustrate that the dimensionally identical beds show similar patterns for axial as well as radial variations of voidage, with the first maxima and minima occurring at the same co-ordination and the voidage becoming uniform at about 0.30. The features of the established profiles will be discussed in detail later.

It is a different story with the scaled up bed. Although the patterns of the variations are similar, the scaled up bed generally exhibits lower maxima and minima, suggesting a more condensed packing. This could be due to the weight of the particles, the heavier the particles, the more condensed the packing. Figures 4.4 and 4.6 show the difference in the axial and radial variations of voidage for the bed with $d_t/d_p = 8.5$ and the scaled up bed. The first minima and maxima still occurs at $\frac{1}{2} d_{pe}$ and $1 d_{pe}$ away from the wall respectively, and both profiles reach a constant value of voidage at about $4 d_{pe}$ away from the container's wall. The corresponding value of void fraction for the scaled up bed is about 0.25 at the core zone.

According to the above results, one can deduce that the data obtained for $\epsilon(x)$ and $\epsilon(r)$ are reproducible in packed beds of equilateral cylinders, but when it comes to scaling there is a significant difference in the voidage values due to the weight of the particles. Therefore, in order to understand the effect of scaling up or

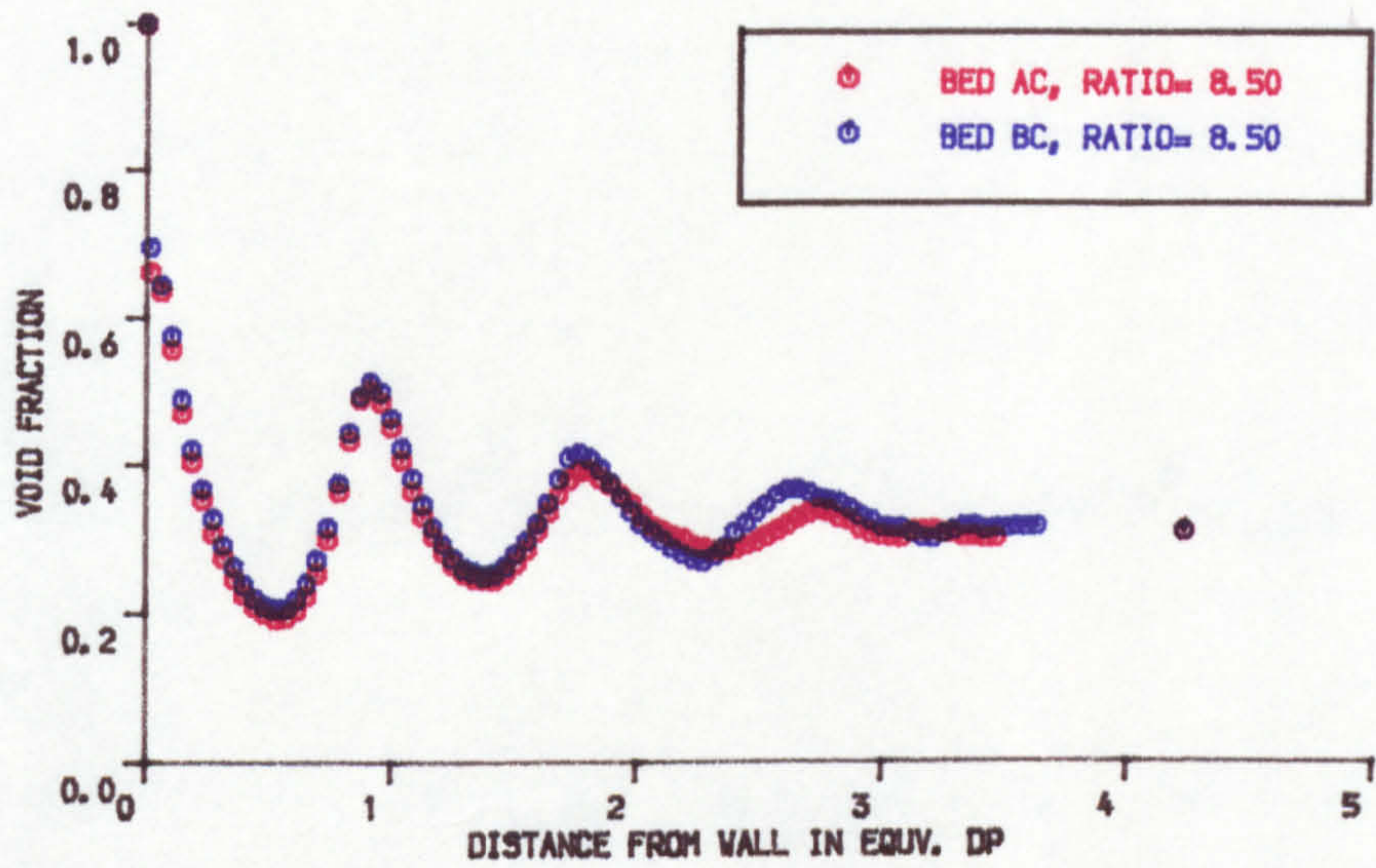


FIG. 4.3 RADIAL VOID PROFILES OF BEDS AC & BC

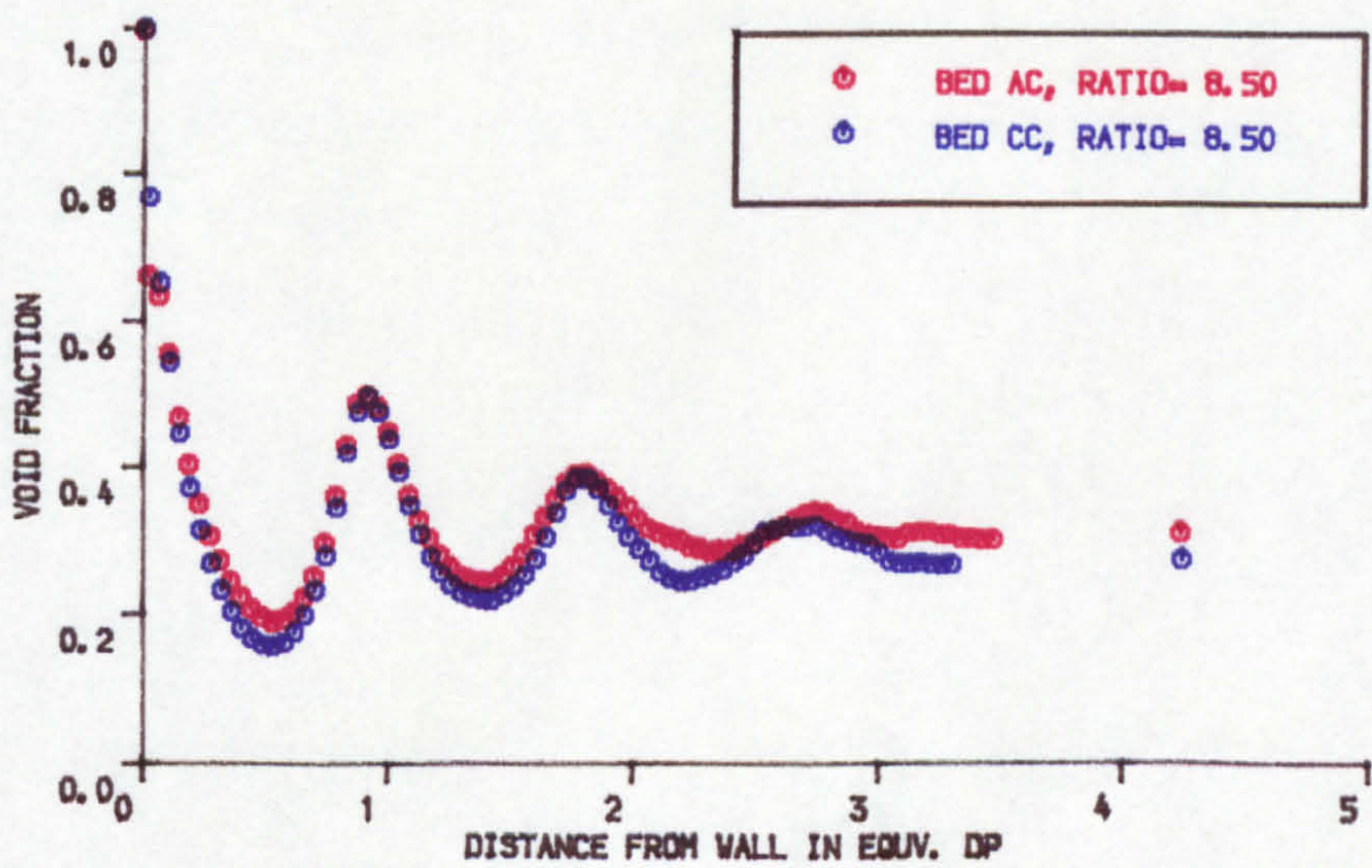


FIG. 4.4 RADIAL VOID PROFILES OF BEDS AC & CC

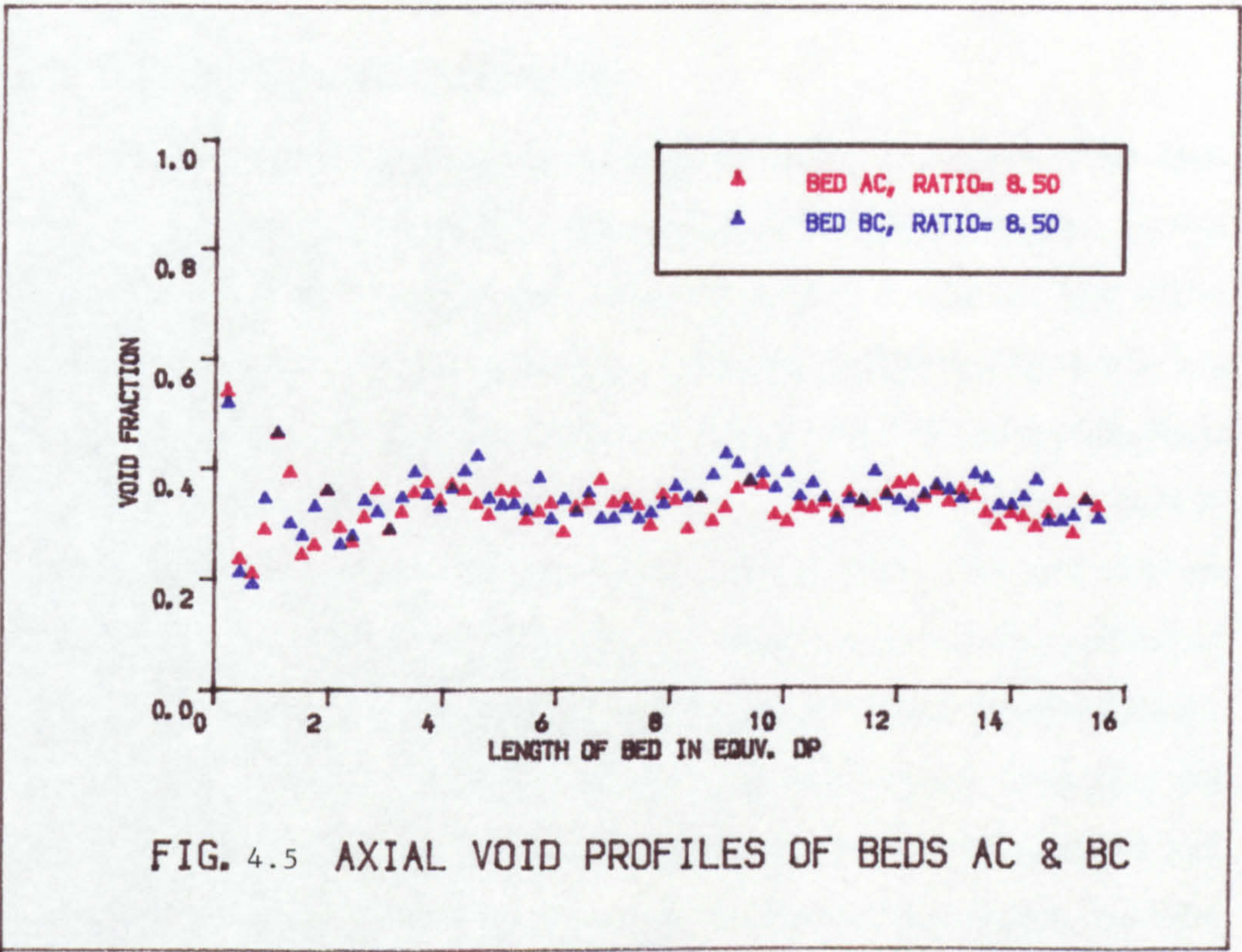


FIG. 4.5 AXIAL VOID PROFILES OF BEDS AC & BC

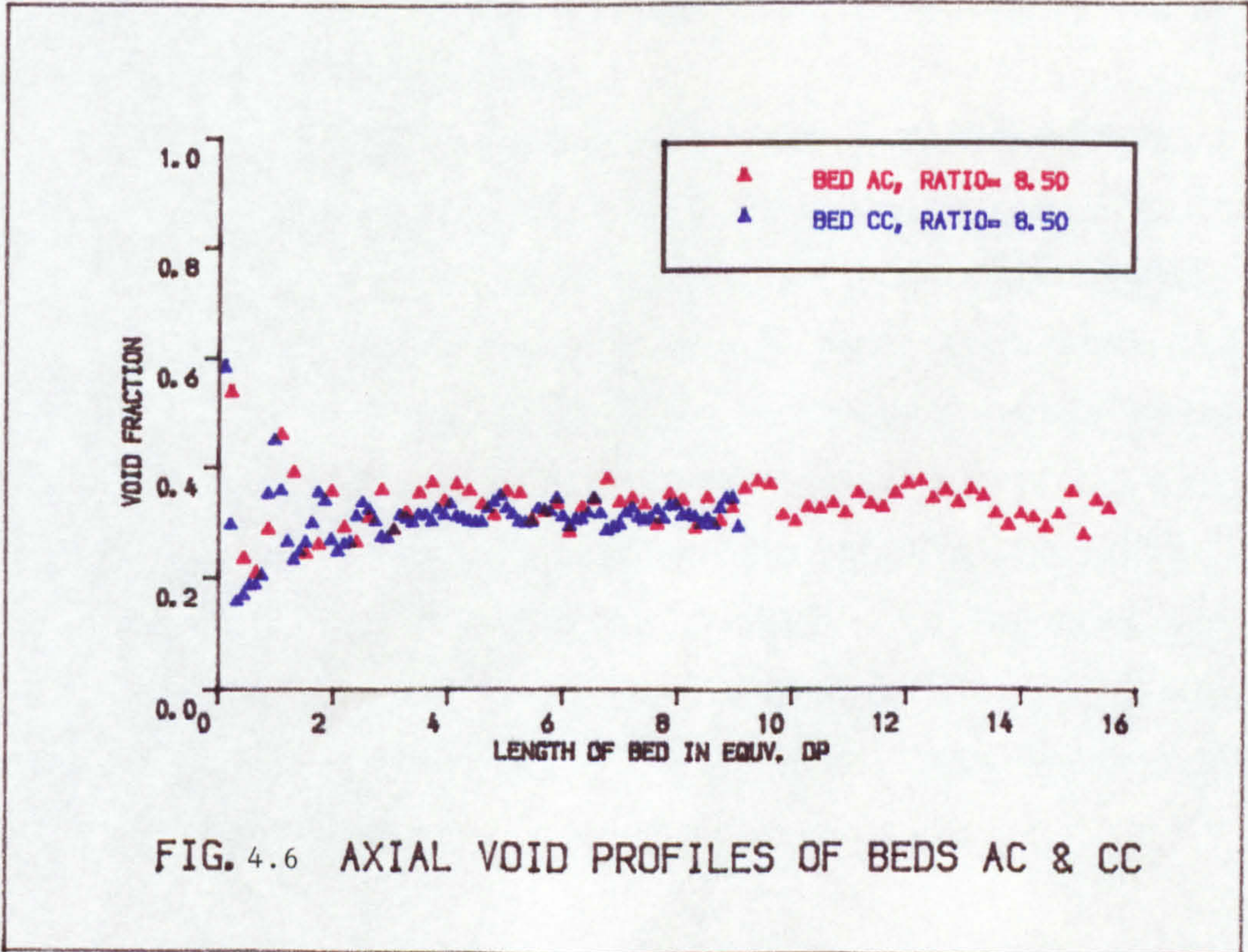


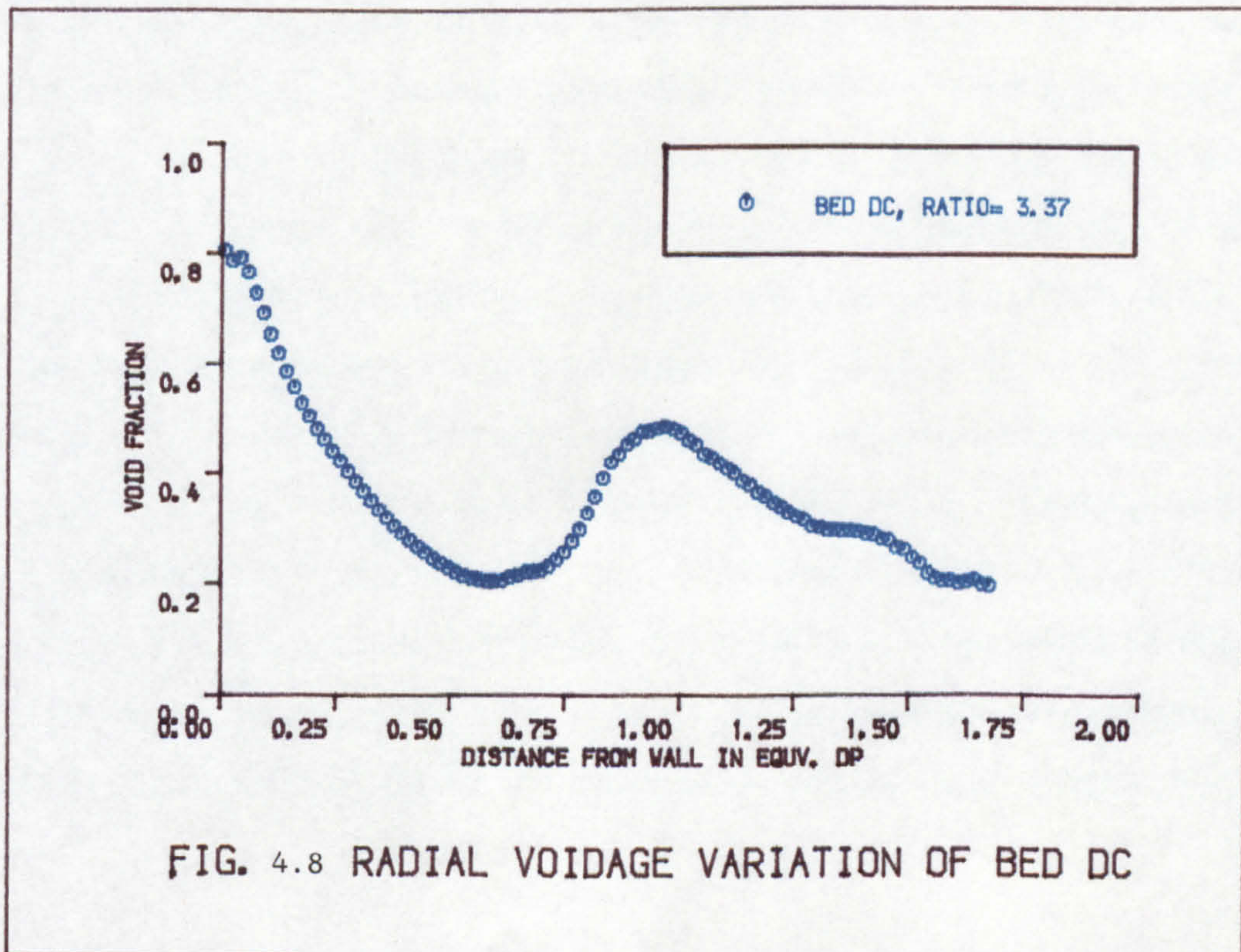
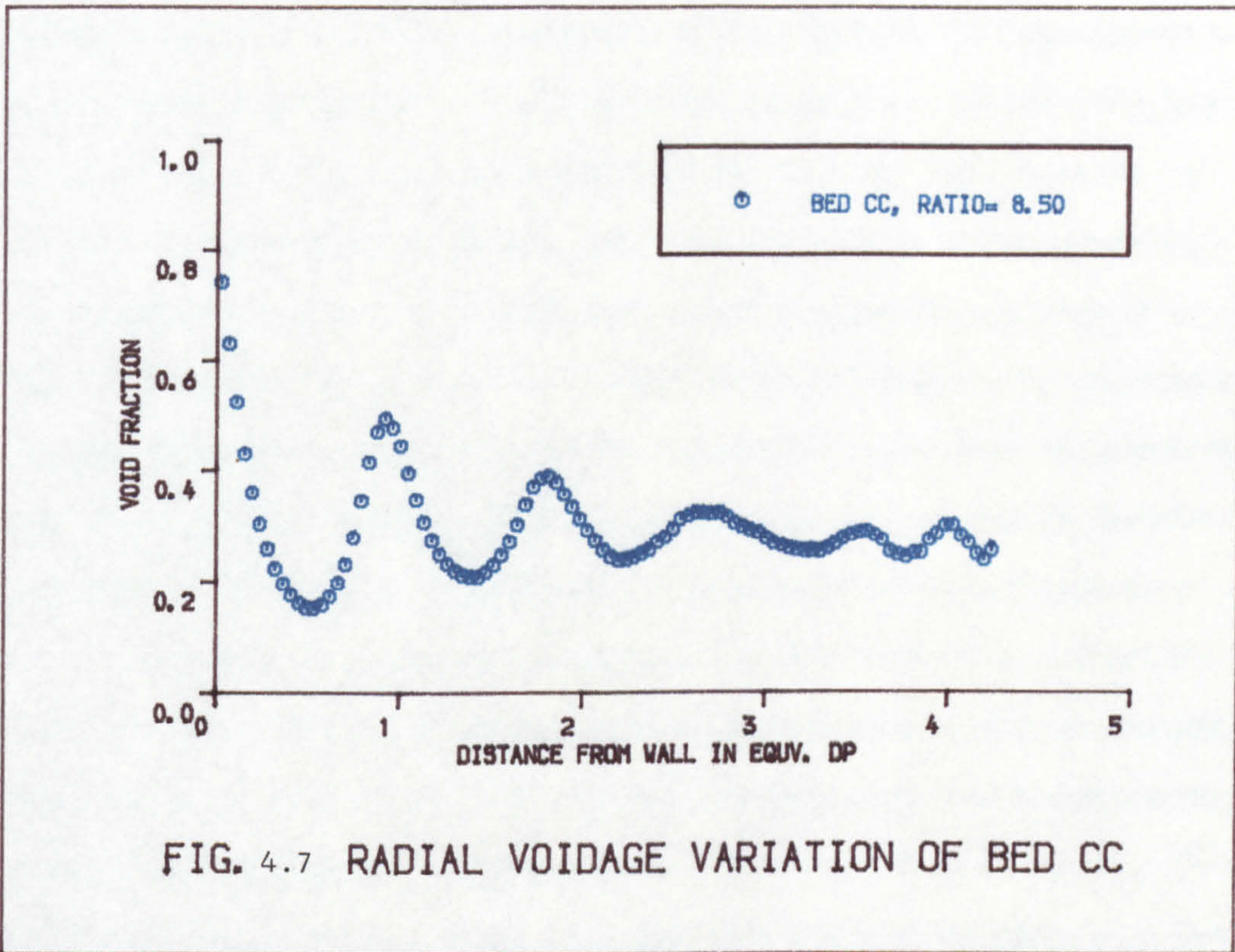
FIG. 4.6 AXIAL VOID PROFILES OF BEDS AC & CC

down the geometries of a packed bed better, more experiments need to be performed with various scaling factors.

4.3.2 Features of the experimental data

Following the machining and polishing operations, the resultant bed cross sections were photographed by the on-line camera of the Image Analyser. It was important to work out the right light settings in order to achieve the best colour contrast throughout the face of the sample. Each cross section of a bed could then be captured and retrieved from the corresponding floppy diskette, framed, binarized, and converted into 100 consecutive annular rings. The black and white pixels in every ring were then counted and presented in a tabular form. The data obtained were averaged in axial and radial directions in order to examine the variation of voidage in those directions accordingly. For example, in the case of radial voidage, $\epsilon(r)$, the mean voidage values of the corresponding annular rings of all the cross sections were axially averaged. The measuremental data were further processed and plots of radial variation of void fraction versus the dimensionless distance from the wall, r/d_{pe} , were produced for all the test beds to illustrate the significance of the boundary walls on the packing structure.

Considering Figure 4.7 as a radial voidage profile for a typical bed, $d_t/d_{pe} = 8.5$, a damped oscillatory pattern with enhanced voidage in the vicinity of the tube wall is recognised. The layer of the particles nearest to the wall tends to be highly ordered with most of the particles touching the wall. The next layer builds up on the surface of the first, in a less ordered fashion. The subsequent layers are less and less ordered, until a fully randomised arrangement is attained in regions far removed from the wall. The boundary walls therefore simply disturb the local packing in their vicinity and consequently increase the local voidage. This phenomenon could have a significant bearing on the flow as well as the heat and mass transfer characteristics of the beds. Appendix A contains the radial voidage profiles of all the sample beds examined.



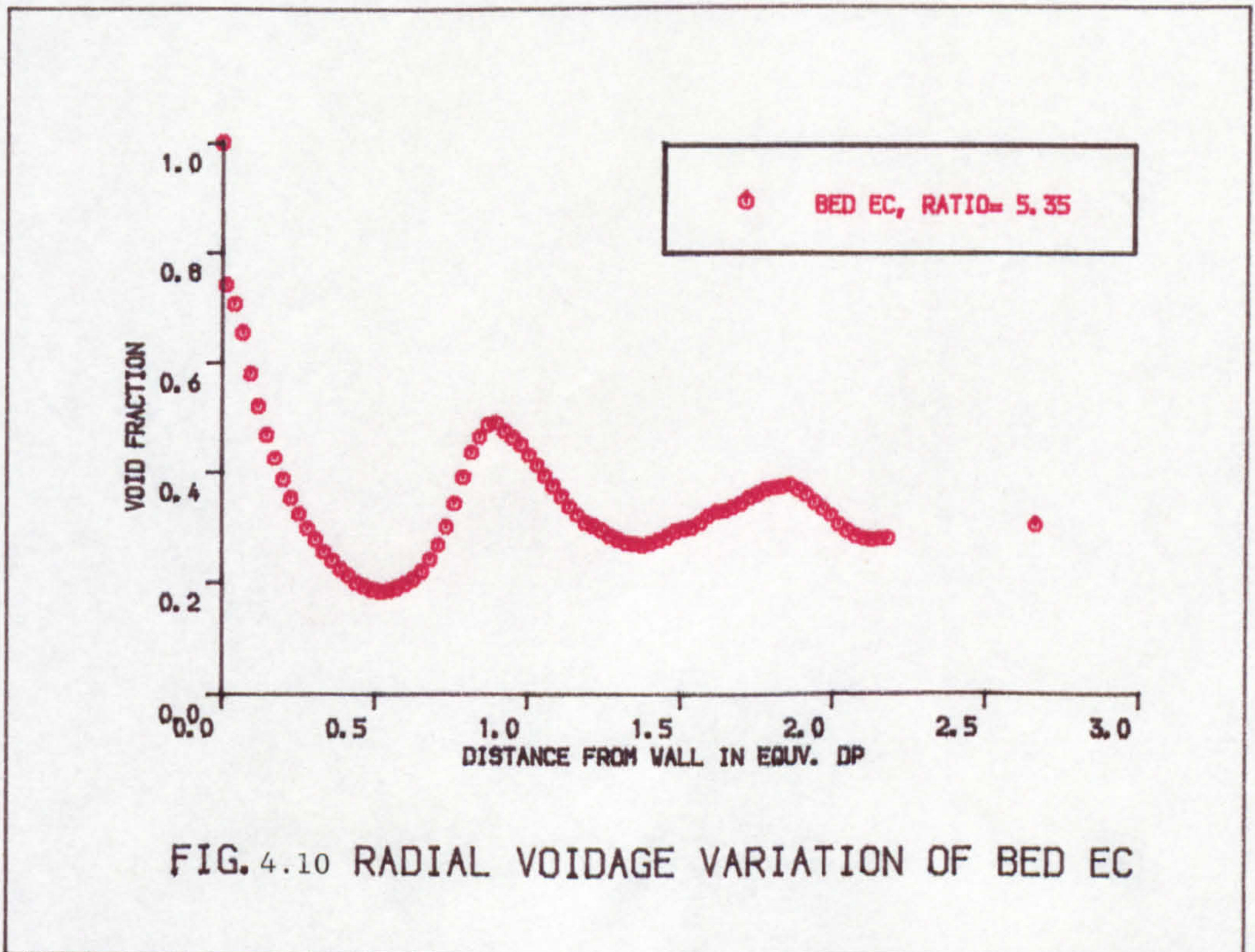
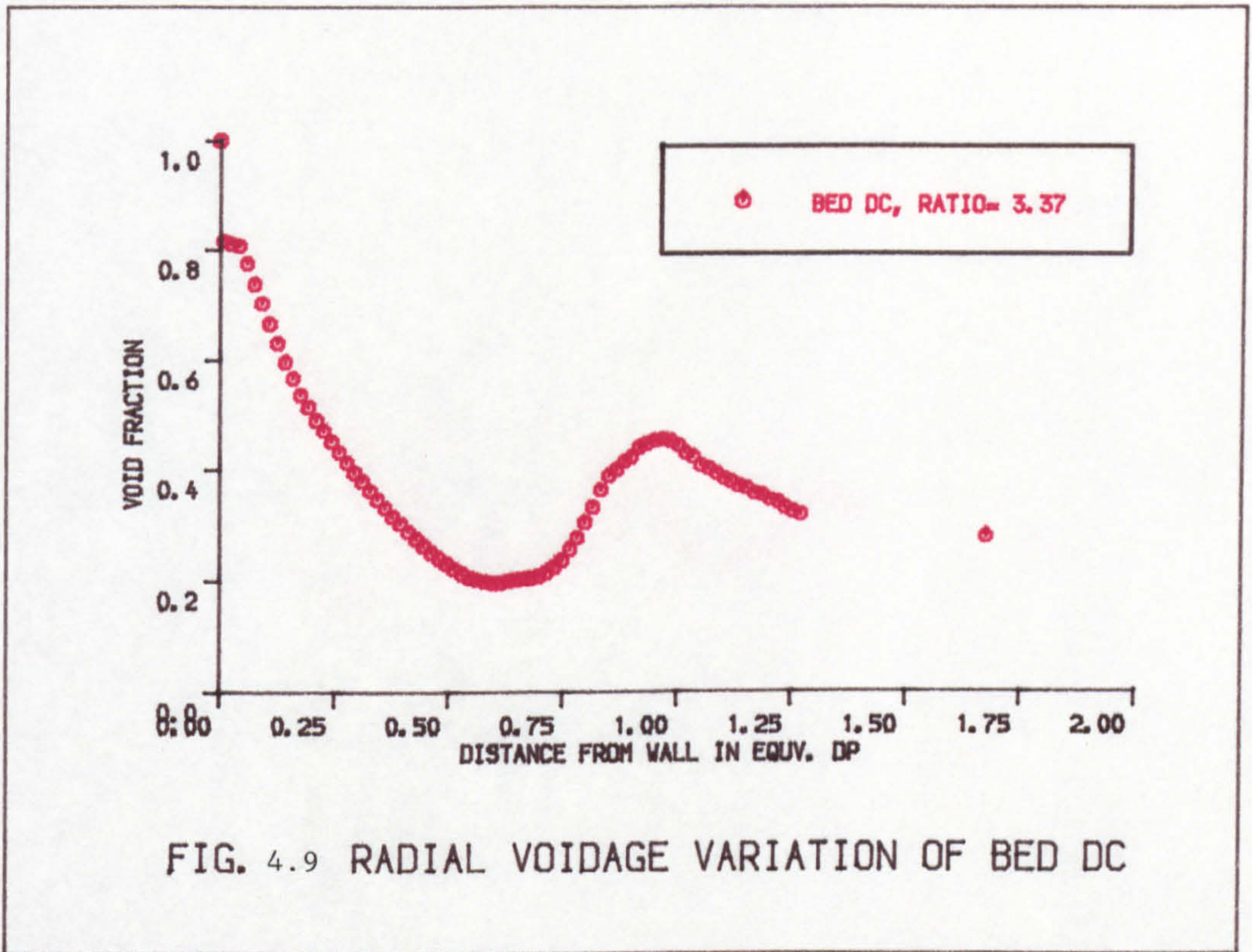
Careful examination of the results reveals that the radial voidage profiles are comprised of two distinctive zones, namely wall and core. In the wall zone, the voidage tends to be high with pronounced fluctuations while the core zone appears to have a rather steady pattern. Results show that the wall zone of beds with large d_t/d_{pe} occupy a distance equivalent to 3-5 particle diameters away from the wall, whereas for beds with small diameter ratios, the core zone is almost non-existent, but the enhanced voidage in the wall zone results in channelling of flow in such beds. This proves the influence of the dimensionality and shape of the constituents of the physical system, as the extent of the wall zone for packed beds of monosized spheres is reported (Moallemi, 1989), to be equivalent to 4 - 5 particle diameters from the wall. Orientation of cylindrical particles causes a more compact packing.

The radial voidage decreases oscillatory from 100% at the wall to about 28% at the bed centre. The pattern resembles that of monosized spheres with a noticeable difference in the value of core zone voidage. For beds comprised of spheres this value is observed to be about 38% compared to 28% for cylindrical particles. The difference between the two values arises from the shape of the particles. The location of the first minima and maxima which respectively appear $\frac{1}{2} d_{pe}$ and $1 d_{pe}$ away from the tube wall are the same in both configurations. However, in beds of cylinders the range of void fraction values for the first minima and maxima are between 0.1 - 0.2 and 0.4 - 0.5 respectively. Beds with small diameter ratios result in oscillatory profiles of very large magnitude which do not tend to damp out, Figure 4.8, shows the radial variation of voidage in a bed with $d_t/d_{pe} = 3.37$. The nature and frequency of the observed patterns tend to depend on the diameter ratio, d_t/d_{pe} , of the system. This is due to the continuous influence of the confining walls on the formation of the packing structure. The peculiar feature of the profiles shown at the centre of the beds is due to the smallness of the central annulus compared with the size of the particles. As the rings get smaller towards the centre of the bed, it becomes more difficult to obtain a meaningful local voidage data at

some point close to the centre. However, extensive study of all the obtained images revealed that by considering a larger core ring with a radius equal to one particle diameter, so that the whole of the core element is measured by a single scan, this peculiarity can be alleviated. Figures 4.9 to 4.15 show the refined radial voidage profiles resulted from the method described above.

By plotting mean voidage of each and every section of the test beds against normalised axial distance, L/d_{pe} , where L is the bed length, the end effect of each bed was examined. Figures 4.16 to 4.22 exhibit the axial variation of voidage of all the test beds prepared. Figure 4.20 is considered as a typical profile for such variations with $d_t/d_{pe} = 14.41$. It indicates a high value at the inlet which gradually decreases in a damped oscillatory manner to a constant value at some distance, $L/d_{pe} = 3.5$, away from inlet with the corresponding mean voidage value of about 30%. This signifies the presence of end effect which is similar to that of the boundary walls. This effect is common in all test beds examined, but the extent of its penetration is dependent on the geometrical factors, i.e. shape and size of the particles. Table 4.1 shows the extent of penetration of wall and end effects in the packed beds examined. It is worth noting that, in general, this effect is noticeable up to 4 equivalent particle diameters away from the bed end. It is interesting to note that the pattern of the axial variation of voidage for beds of cylinders does not resemble that of spheres. Although the voidage in both decreases exponentially to a constant value, the curve for cylinders is of oscillatory nature whereas for spheres is not as pronounced. Also the curve for spheres flattens at a mean voidage value of about 40% compared to 30% for cylinders. The first minima and maxima of the axial profiles of cylinders occur at $\frac{1}{2} d_{pe}$ and $1 d_{pe}$ away from the bed end, respectively.

The most important conclusion that can be drawn by studying features of the data obtained is that in the wall zone of cylindrical packed beds, the voidage is much higher than in the core, which results in considerable mal-distribution of fluid flow,



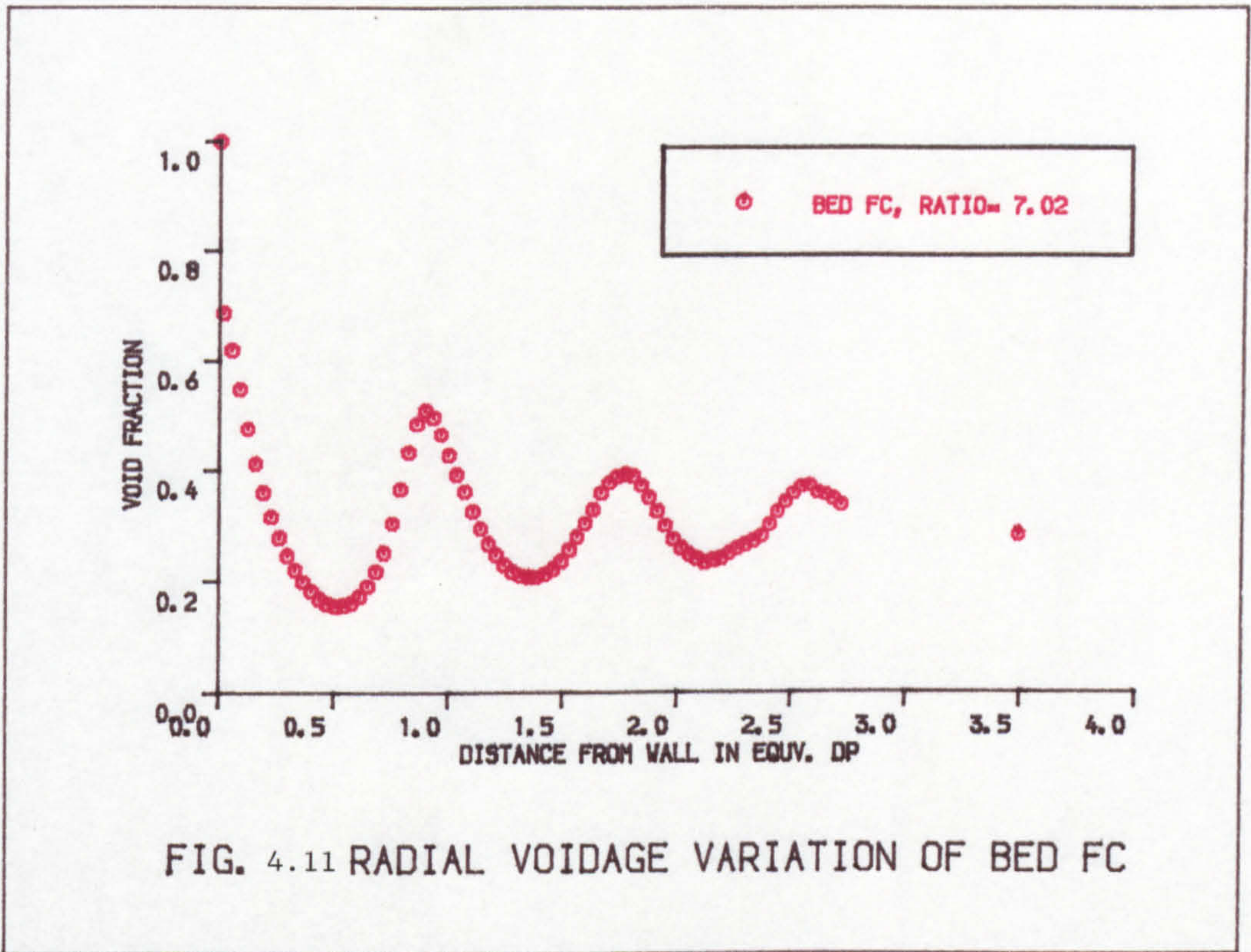


FIG. 4.11 RADIAL VOIDAGE VARIATION OF BED FC

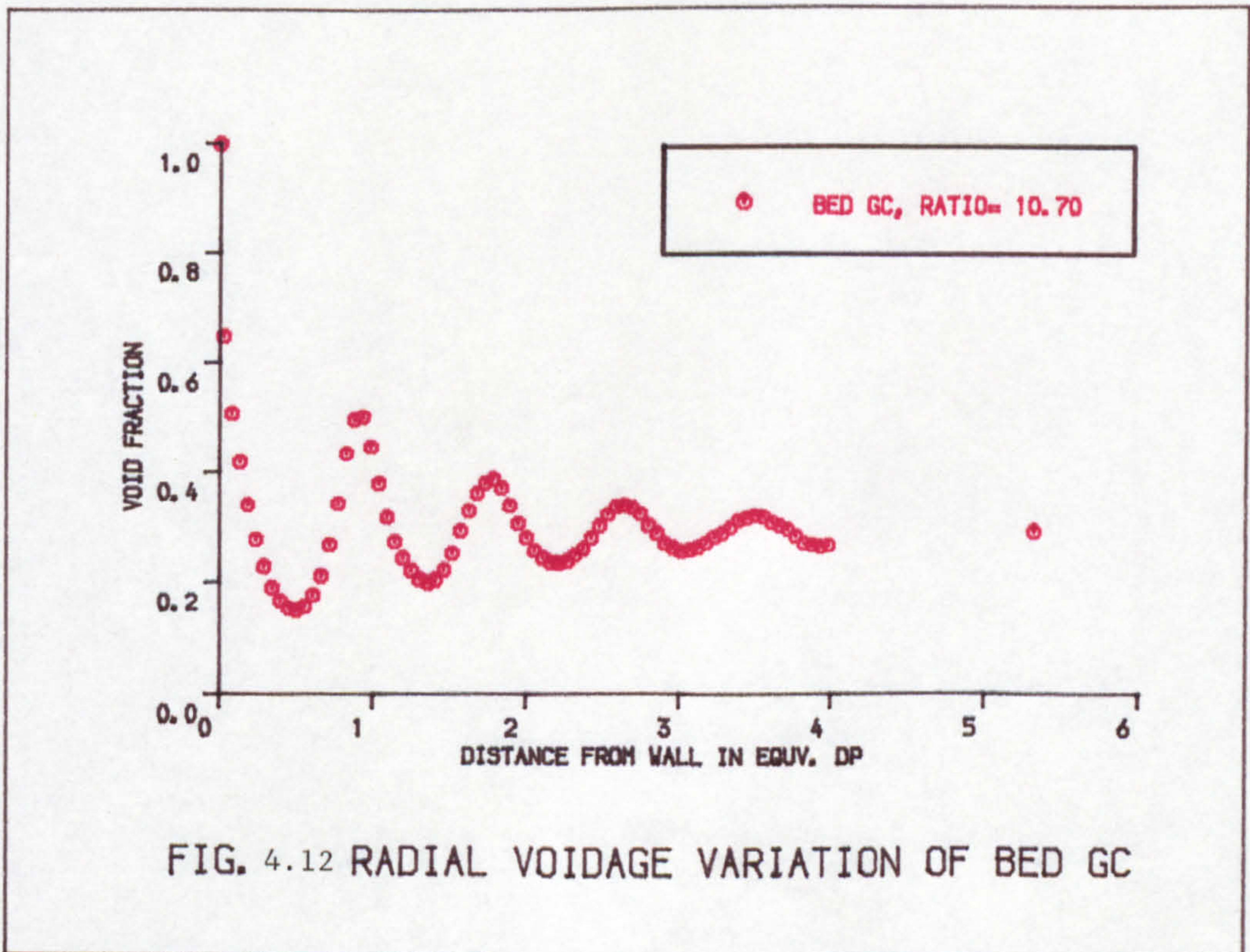


FIG. 4.12 RADIAL VOIDAGE VARIATION OF BED GC

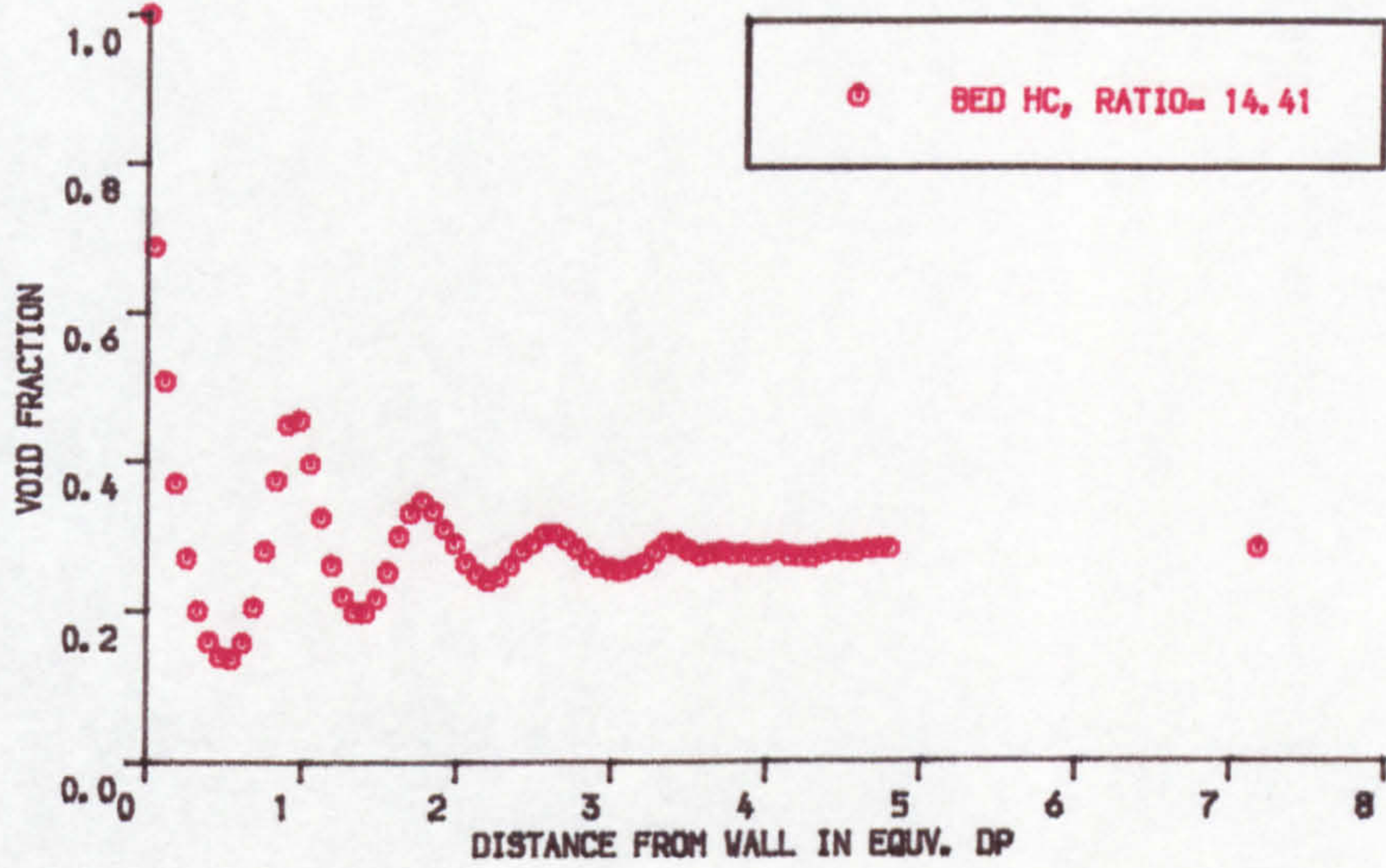


FIG. 4.13 RADIAL VOIDAGE VARIATION OF BED HC

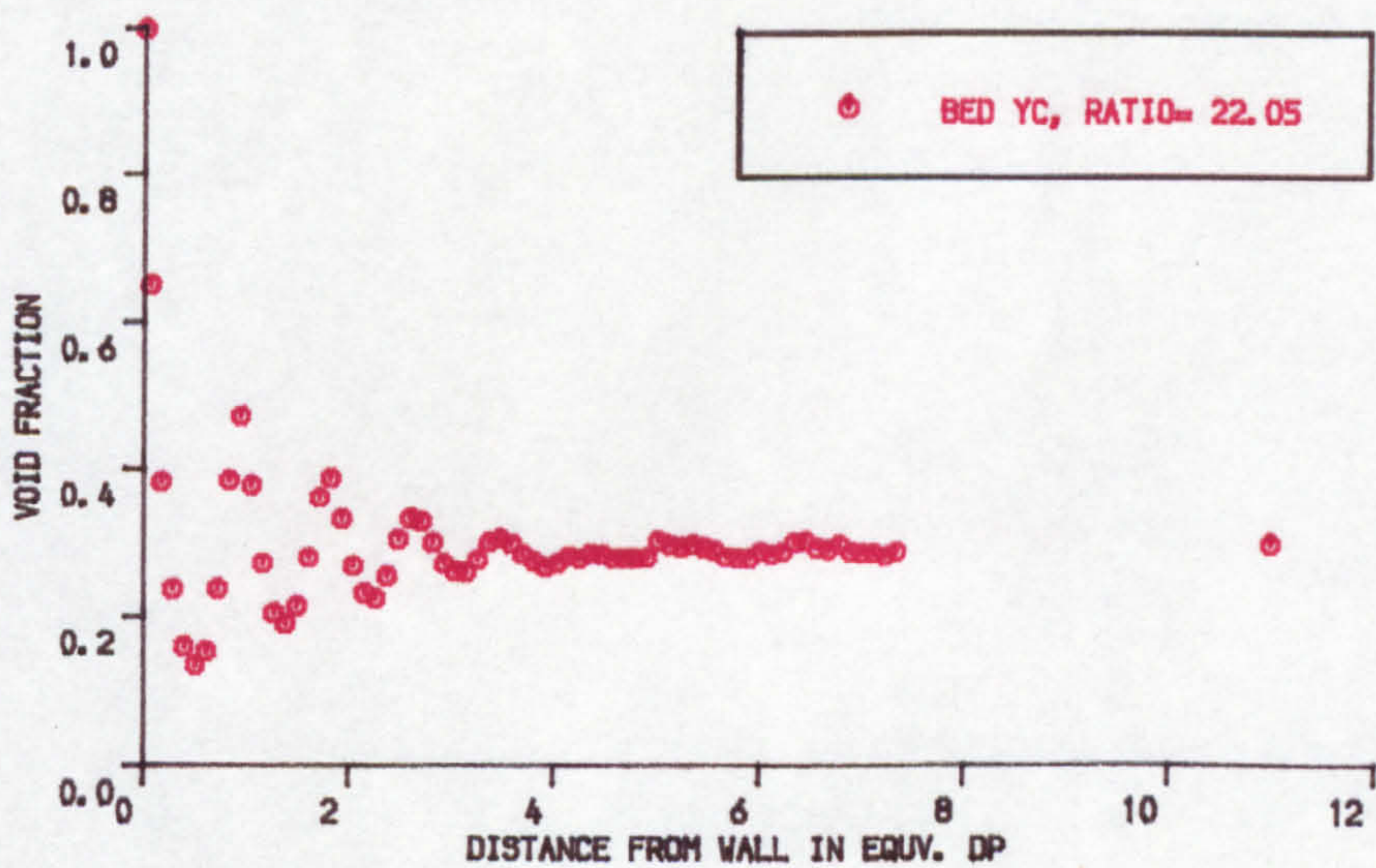


FIG. 4.14 RADIAL VOIDAGE VARIATION OF BED YC

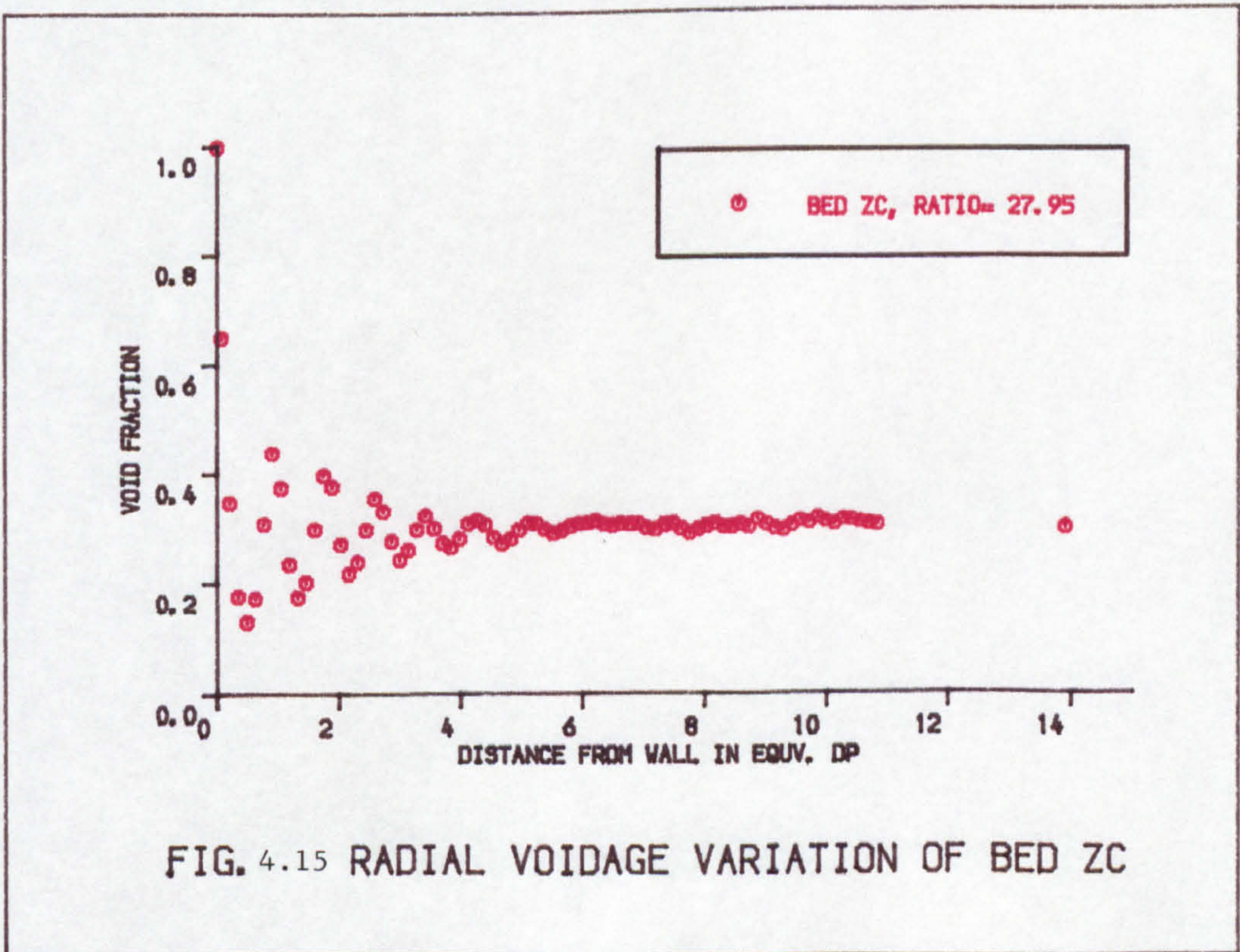


FIG. 4.15 RADIAL VOIDAGE VARIATION OF BED ZC

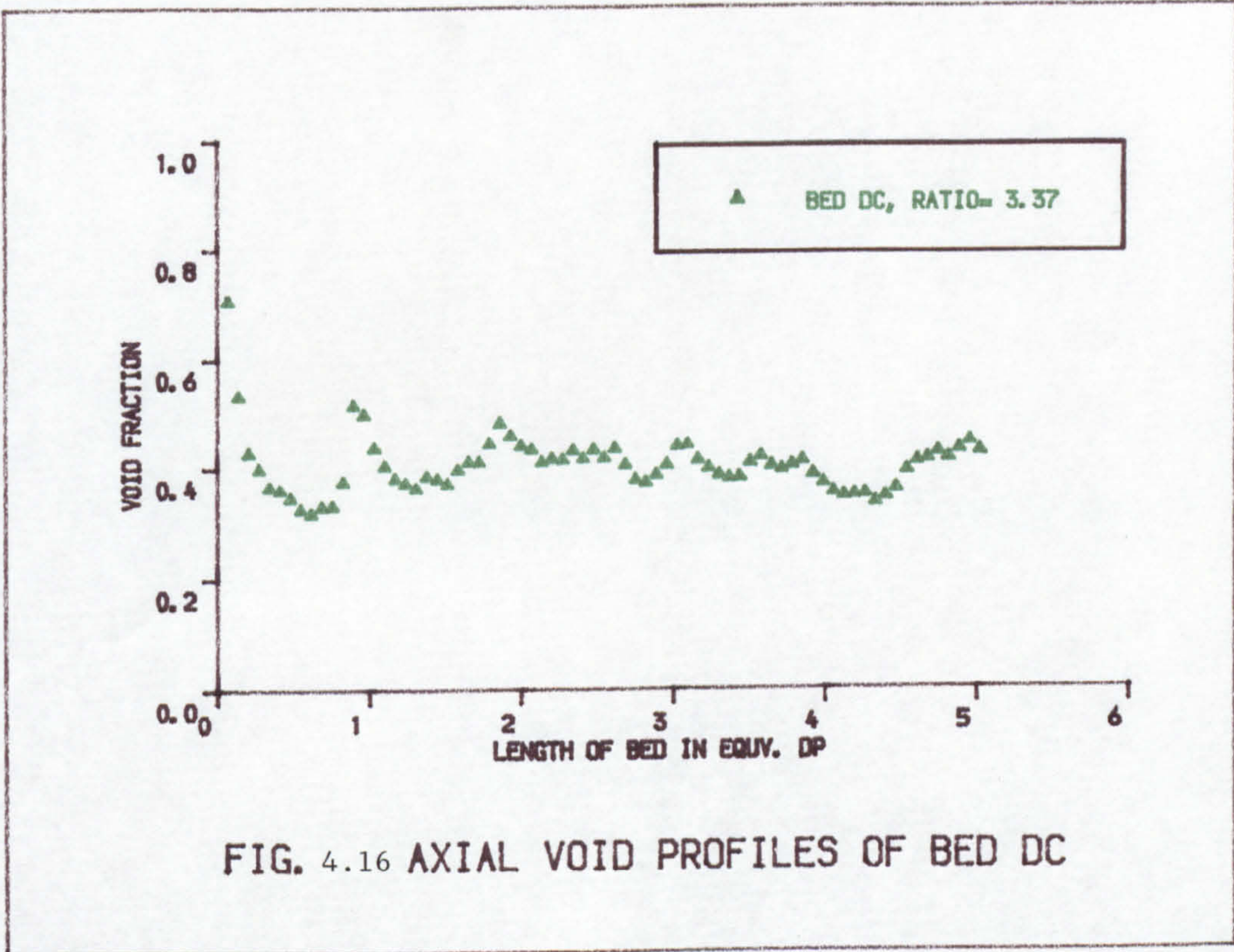


FIG. 4.16 AXIAL VOID PROFILES OF BED DC

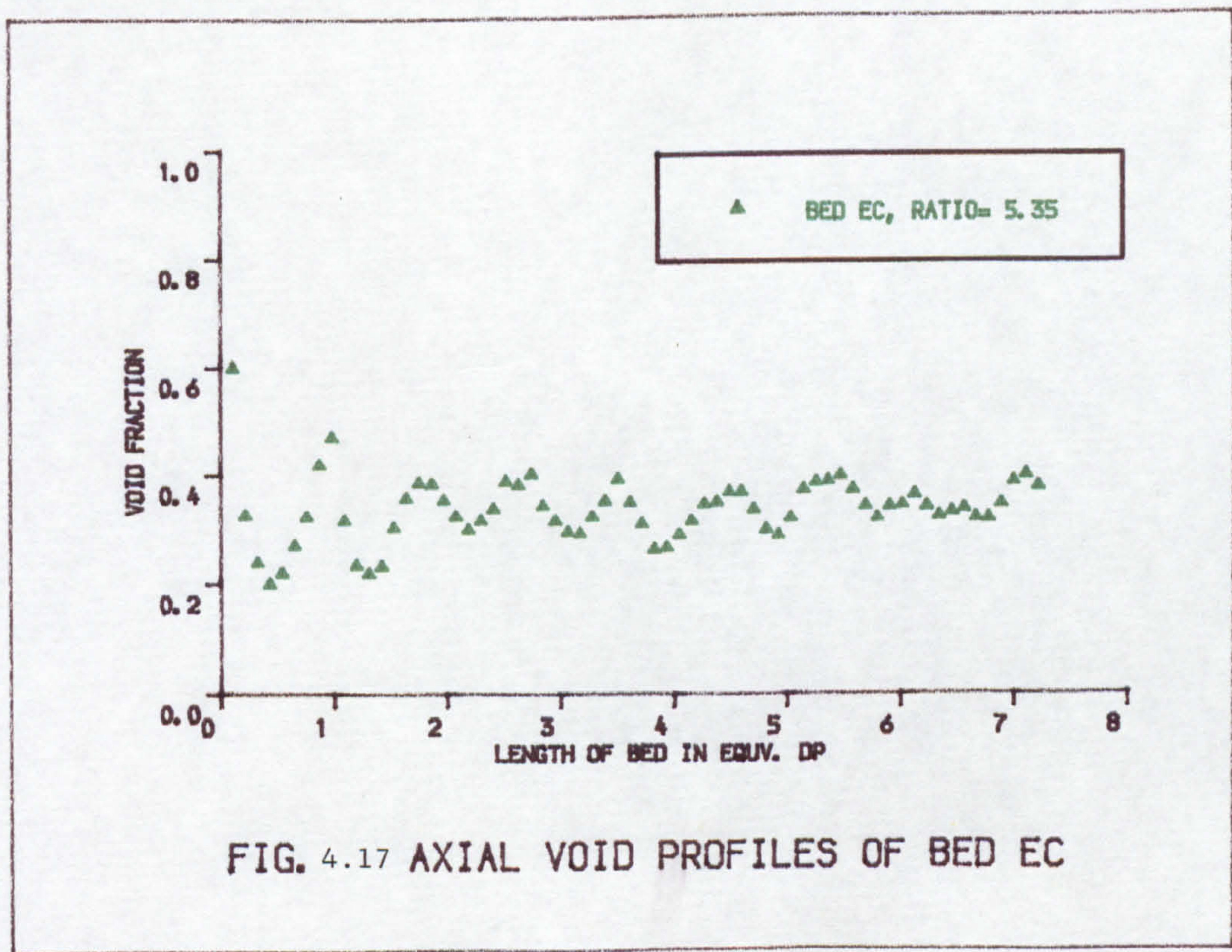


FIG. 4.17 AXIAL VOID PROFILES OF BED EC

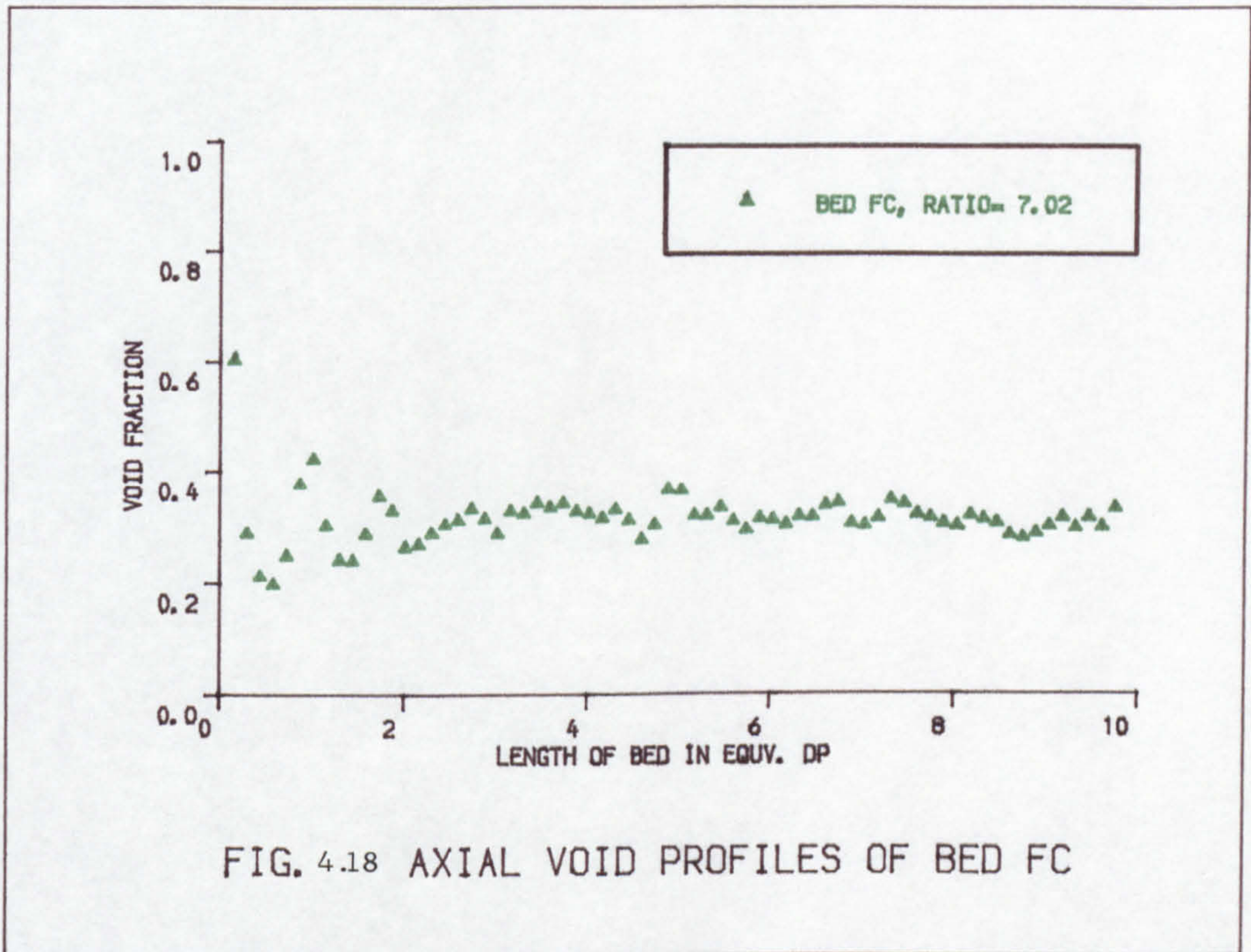


FIG. 4.18 AXIAL VOID PROFILES OF BED FC

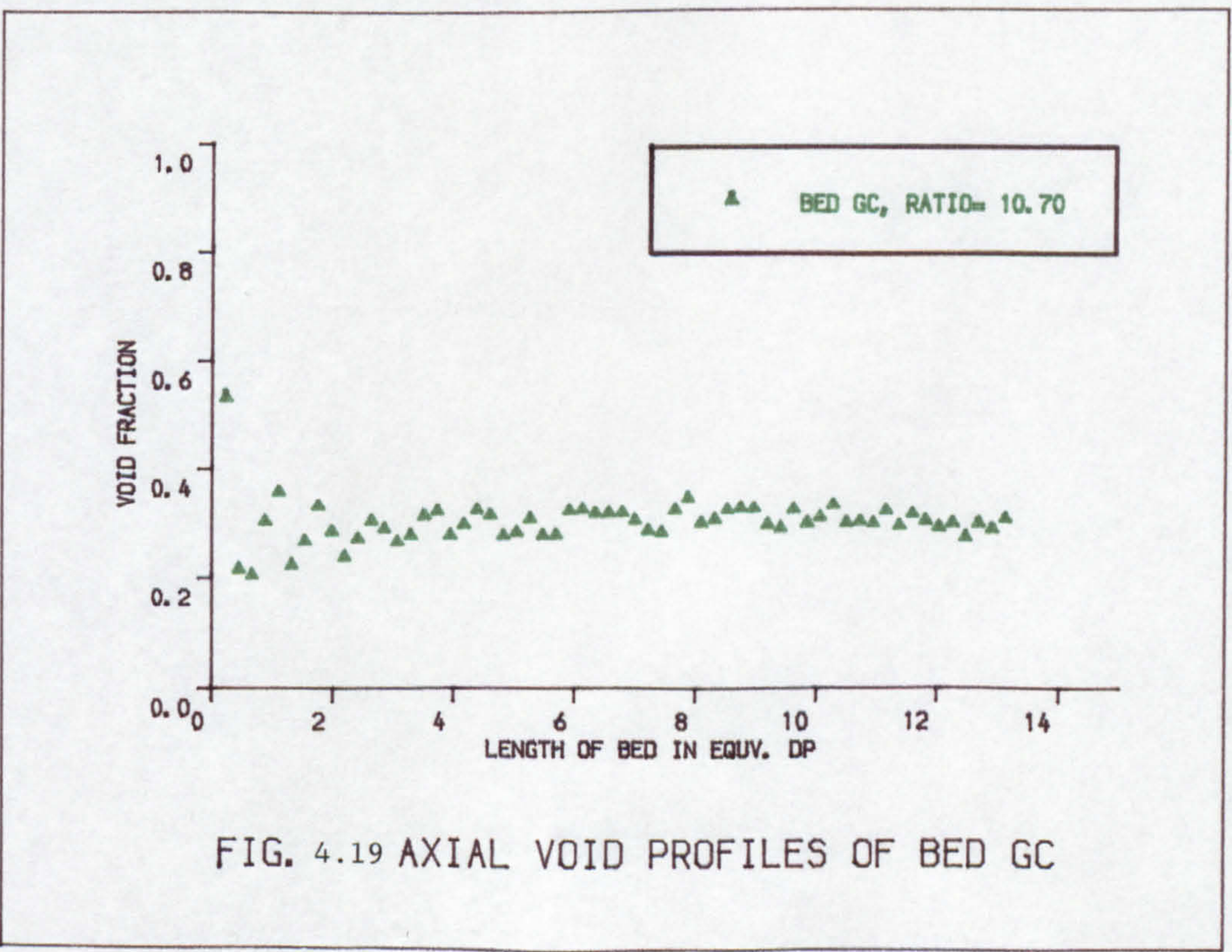


FIG. 4.19 AXIAL VOID PROFILES OF BED GC

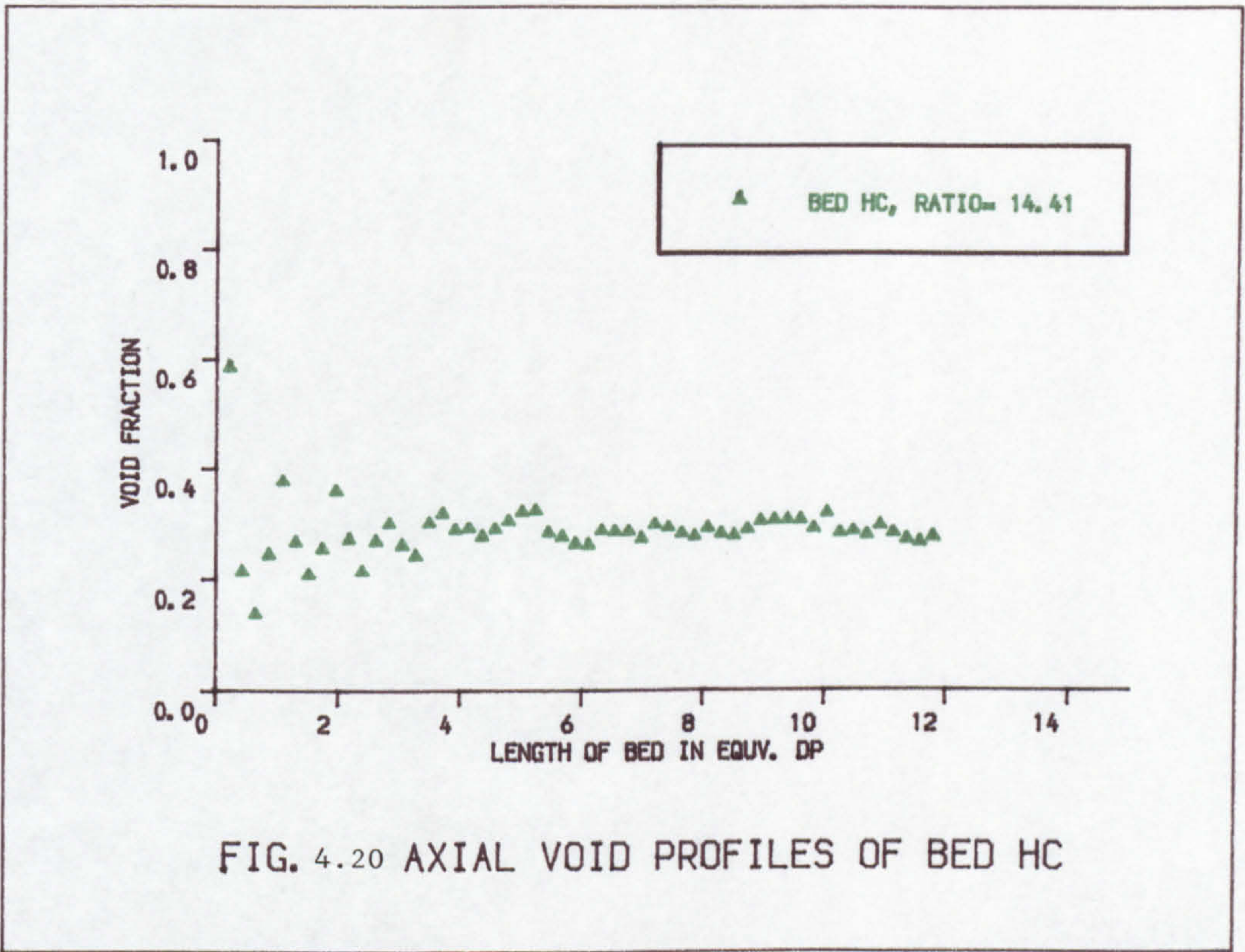


FIG. 4.20 AXIAL VOID PROFILES OF BED HC

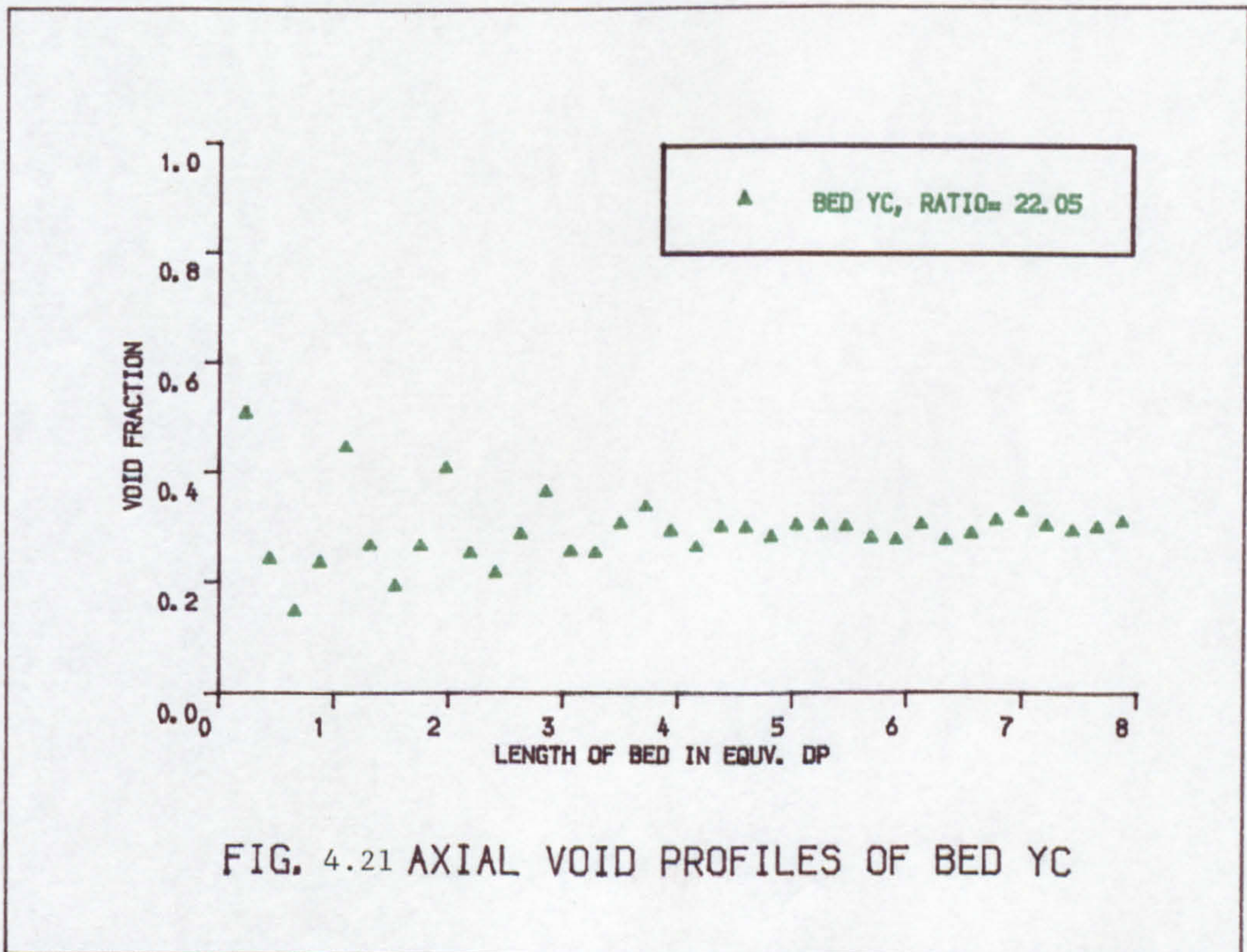


FIG. 4.21 AXIAL VOID PROFILES OF BED YC

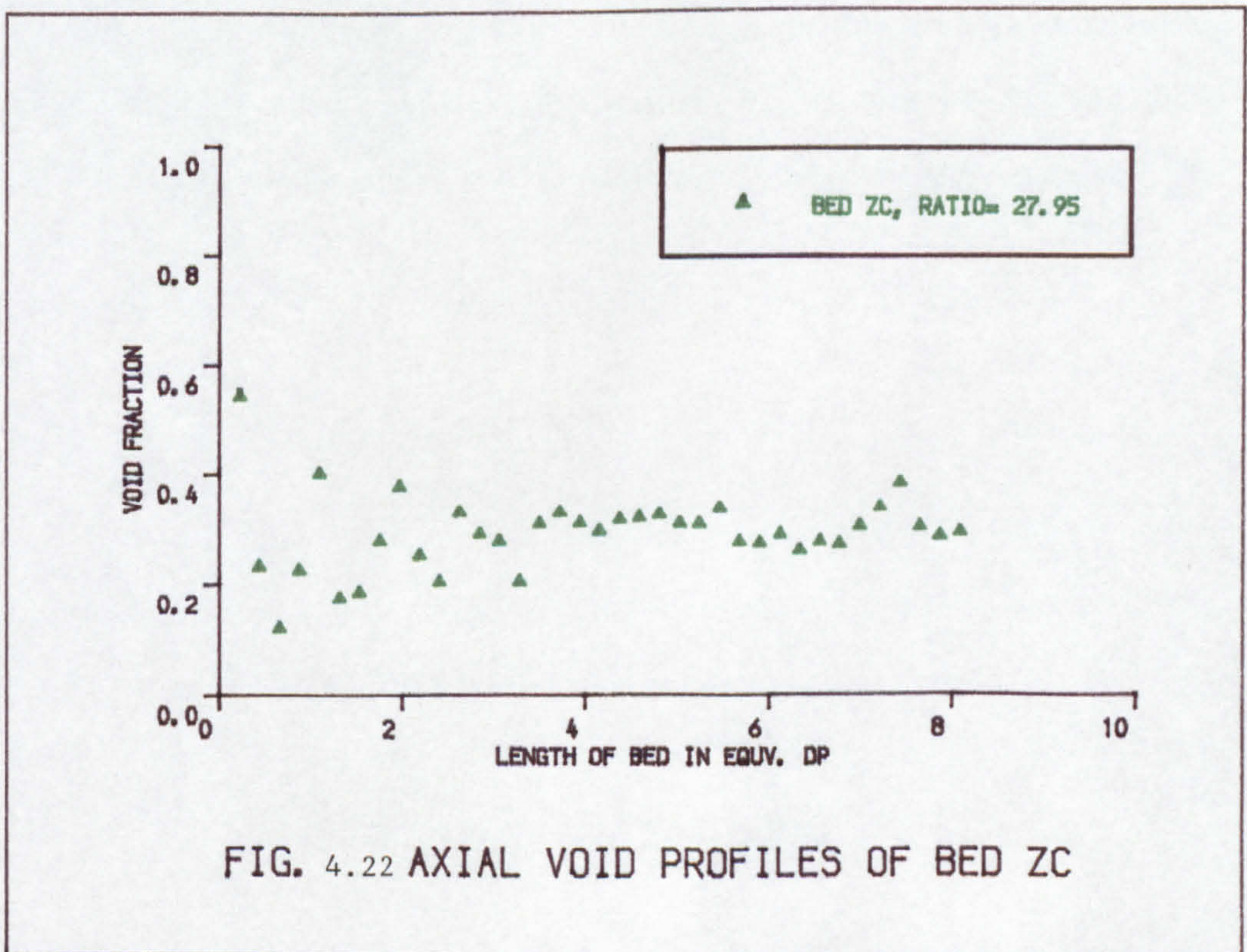


FIG. 4.22 AXIAL VOID PROFILES OF BED ZC

TABLE 4.1
The Extent of Penetration of Wall and End Effects
in Packed Beds of Equilateral Cylinders

ID	d_t/d_{pe}	Wall Effect, d_{pe}	End Effect, d_{pe}
AC	8.50	3	3.5
BC	8.50	3	3.5
CC	8.50	3	3.5
DC	3.37	1.7	3.5
EC	5.35	2.6	6
FC	7.02	3.5	3.5
GC	10.70	4	3.5
HC	14.41	3	3.5
YC	22.05	4	4
ZC	27.95	5	4

particularly along the radius of the cylindrical bed. Such variations of flow and structure could have a significant effect on the associated heat and mass transfer properties and in the presence of reaction it could influence the overall rate of conversion. In view of this possible scenario, the aforementioned features arising from the structural non-uniformities of the particulate bed ought to be taken into account if better designs are being sought. Because of the nature of the bed structure, the local fluid velocity inside the porous medium varies continuously in magnitude and direction. The tortuous nature of the bed and the variations in the shape of flow passage cause a lateral variation in the resistance to flow which has a marked effect on the velocity distribution. Therefore the model equations which incorporate the effective radial thermal conductivity, wall heat transfer coefficient and fluid-solid convective heat transfer coefficient are to be reviewed with respect to the variations in the magnitude of the fluid velocity. The above mentioned parameters are thus influenced by the operating conditions and the physical dimension of the system.

Having established that the presence of the confining wall constitutes a source of non-random radial variations in the void fraction of cylindrical packed beds, an attempt has been made to formulate a functional relationship between the radial voidage variations and the parametric properties of the physical system such as the tube diameter as well as particle size and shape. Such a relationship can thus permit the structural characteristic of the bed to be established predictively.

4.4 Pattern Characterisation

As mentioned earlier, the literature lacks in having a reliable set of correlations capable of facilitating representative characteristics of the packed bed structure. Having established distinguishing trends for the global and the local structural properties of several packed beds of full equilateral cylinders, the next step would be to develop generalised correlations capable of providing features of

interest predictively.

4.4.1 Mean voidage correlation

A number of expressions have been employed to provide the best fit for the established trend between the mean voidage, ϵ_{mean} , and the equivalent diameter ratio, d_t/d_{pe} . For this a multi-variable optimisation routine proposed by Marquardt, (1963); has been used to facilitate such a fit. Numerous equations of complex as well as simple nature have been tried and Equation (4.1) is found to be the mathematical expression that best represents the data obtained for equilateral cylinders:

$$\epsilon_{\text{mean}} = 0.293 + 0.684 d_r^{-0.85} \frac{1}{\sqrt{1.837 d_r - 1}} \quad (4.1)$$

where $d_r = d_t/d_{pe}$.

Figure 4.23 illustrates the closeness of the fit. Figure 4.24 demonstrates the observed data for the mean voidage of a packed bed of cylinders with different length-to-diameter ratios, ($l_p/d_p = 0.5, 1.5, 2.0$) together with the predicted profile of full equilateral cylinders. Inspection of the displayed information reveals the closeness of the fit which suggests the applicability of Equation (4.1) to all practical sizes of cylindrical particles, providing equivalent particle diameter is used. The corresponding mean voidage resulted from Equation (4.1) for all the test beds examined together with the calculated mean voidage of each bed using Simpson's rule are tabulated in Table 4.2. The discrepancy between the mean voidage data of the two approaches is within 10%.

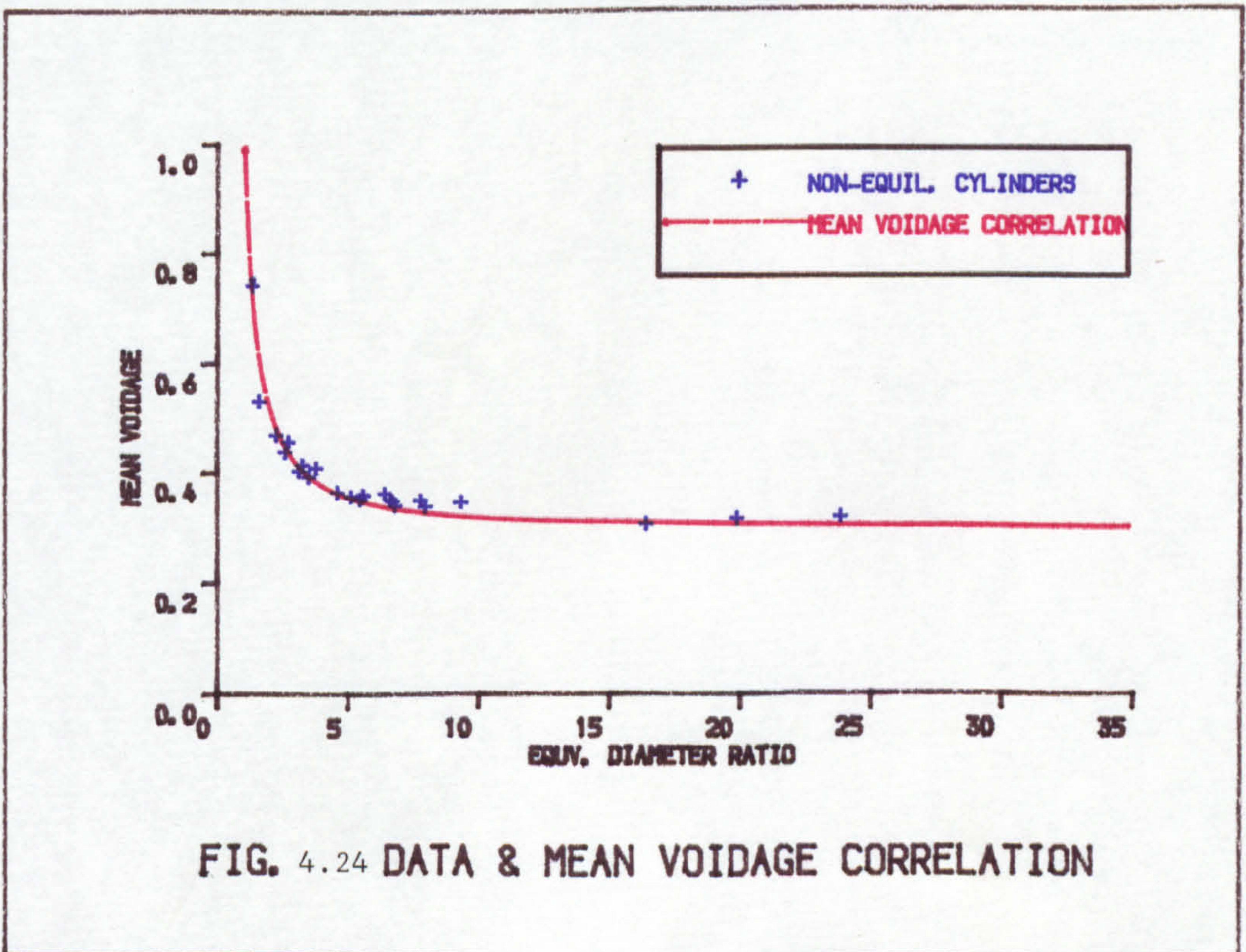
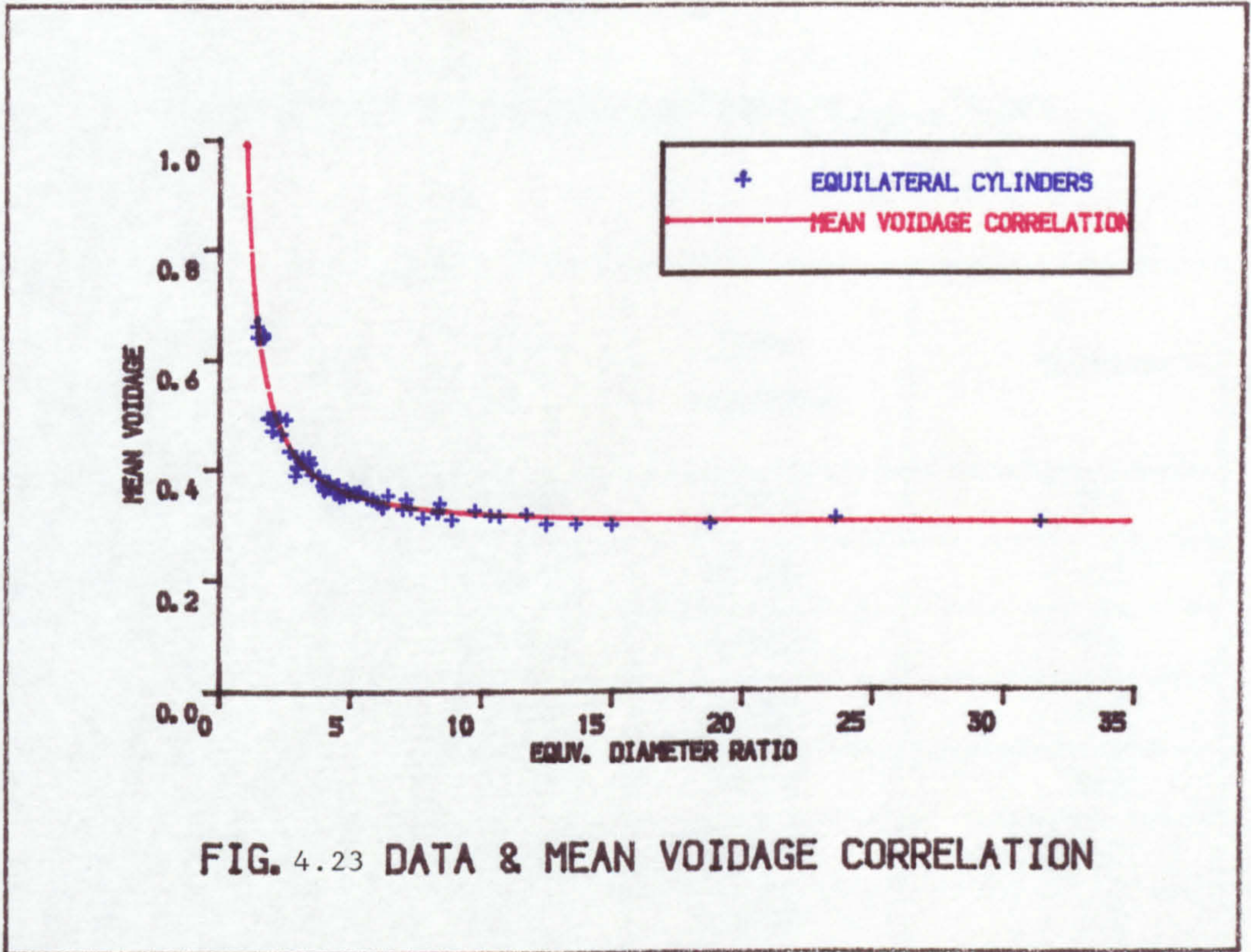


TABLE 4.2

Comparison of Experimental and Predictive ϵ_{mean} Values

d_t mm	d_t/d_{pe}	ϵ_{mean} data	ϵ_{mean} predicted	% Error
49	3.37	0.4027	0.4000	0.7
49	5.35	0.3340	0.3483	4.3
49	7.02	0.3098	0.3308	6.8
78	8.5	0.3062	0.3220	5.2
49	10.07	0.2940	0.3141	6.8
66	14.41	0.2785	0.3070	10.2
101	22.05	0.2833	0.3008	6.2
128	27.95	0.2868	0.2987	4.2

4.4.2 Local voidage correlations

The radial distribution of voidage produced by the analysis of the data were first examined for characterisation purposes. A mathematical expression, Equation (4.2), that defines the distinctive characters of the pattern, i.e. exponential, sinusoidal and steady, is chosen. The coefficients associated with the terms are responsible for the closeness of the fit.

$$\epsilon = 1.0 - \alpha_1 \{1.0 - \exp(-\alpha_2 x^{\alpha_4}) \cos(\alpha_3 x^{\alpha_5})\} \quad (4.2)$$

where $\alpha_1 \rightarrow \alpha_5$ are parameters associated with various characteristics of the pattern,

α_1 for the constant value of voidage in the core zone;

α_2 for the speed of damping of the oscillations;

α_3 for the frequency of the cycles;

α_4, α_5 are freedom parameters responsible for tighter fitting of the expression to the data points;

x is the dimensionless distance from the container wall.

In order to ascertain the optimized values of the above coefficients the Marquardt Algorithm was used again. This optimization routine exhibits rapid convergence even with relatively poor initial guesses.

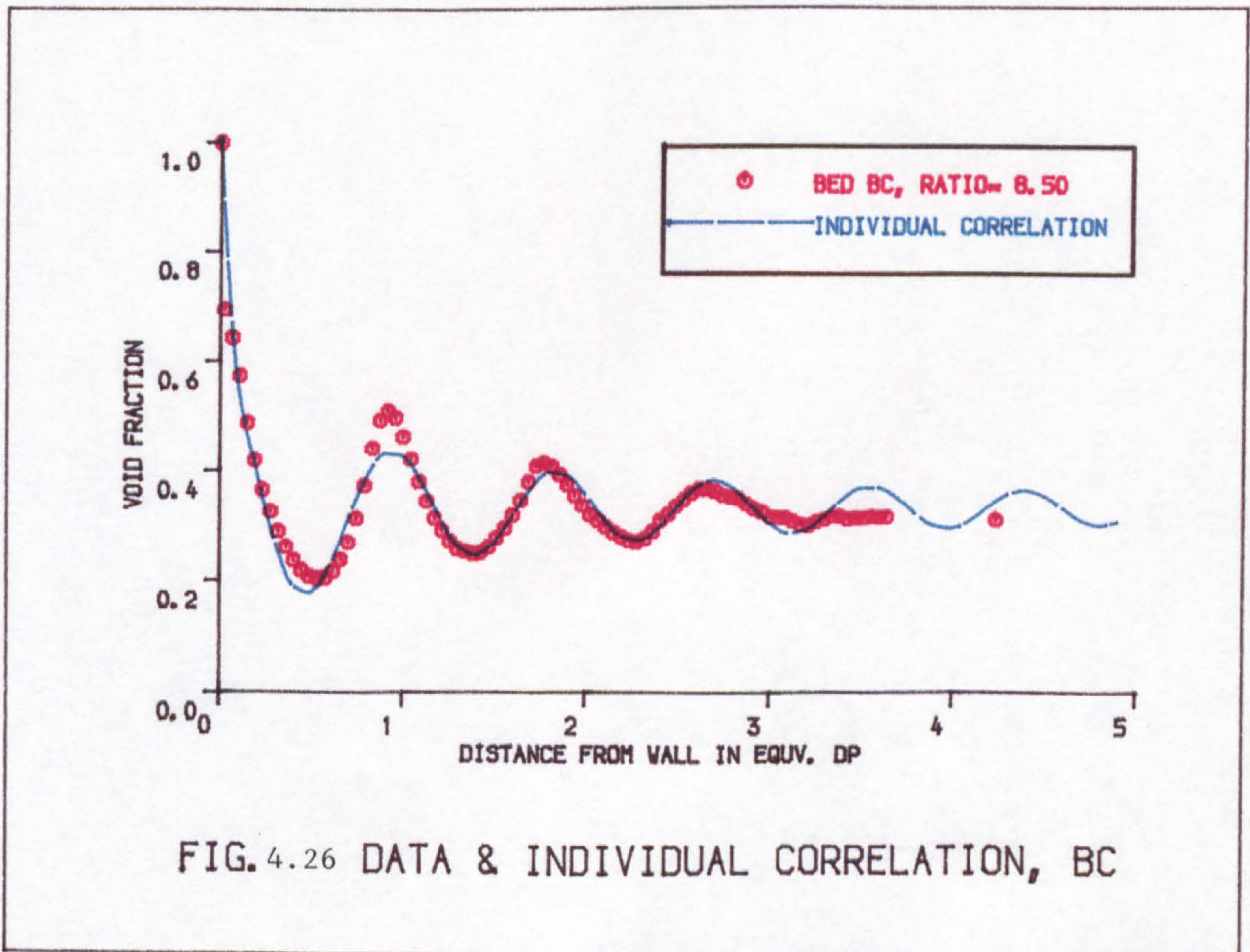
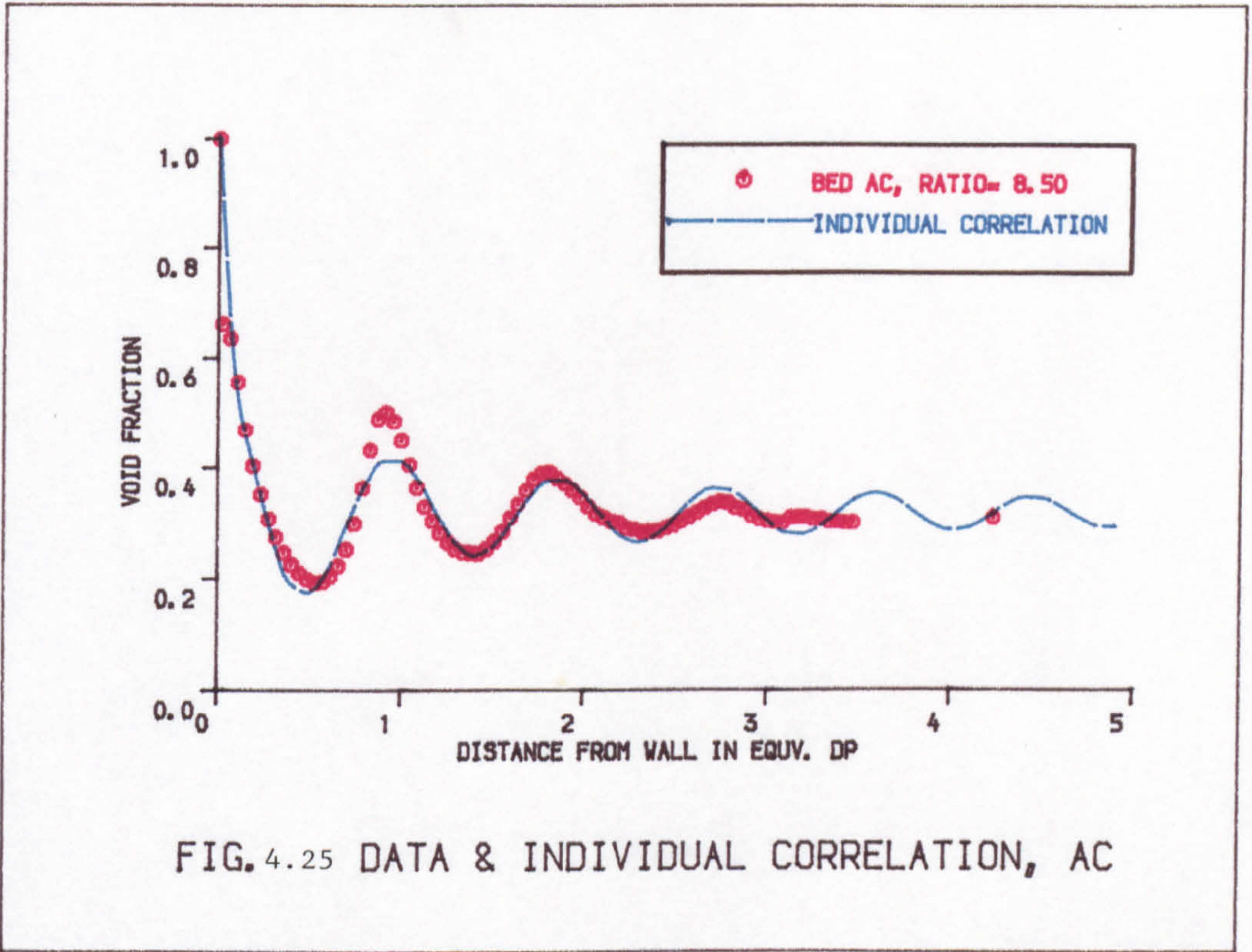
An individual correlation for each and every bed of specific d_t/d_{pe} was hence developed. Sum of squares of errors, SSE, and the standard error of estimates, SEE, were also calculated for each expression. Table 4.3 represents the evaluated correlation coefficients for each bed together with SSE and SEE values. The closeness of the predicted profiles with the obtained data are illustrated in Figures 4.25 to 4.34.

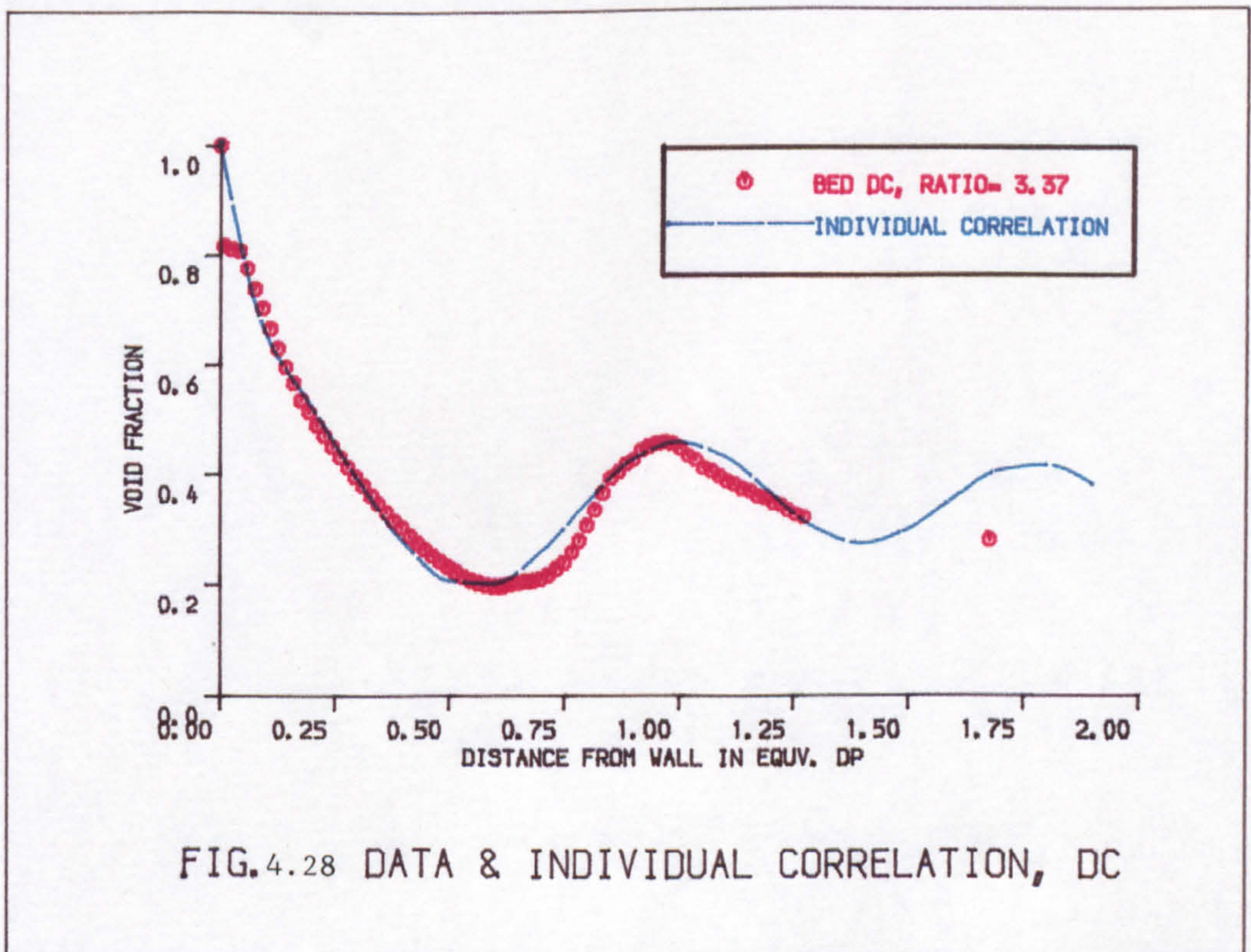
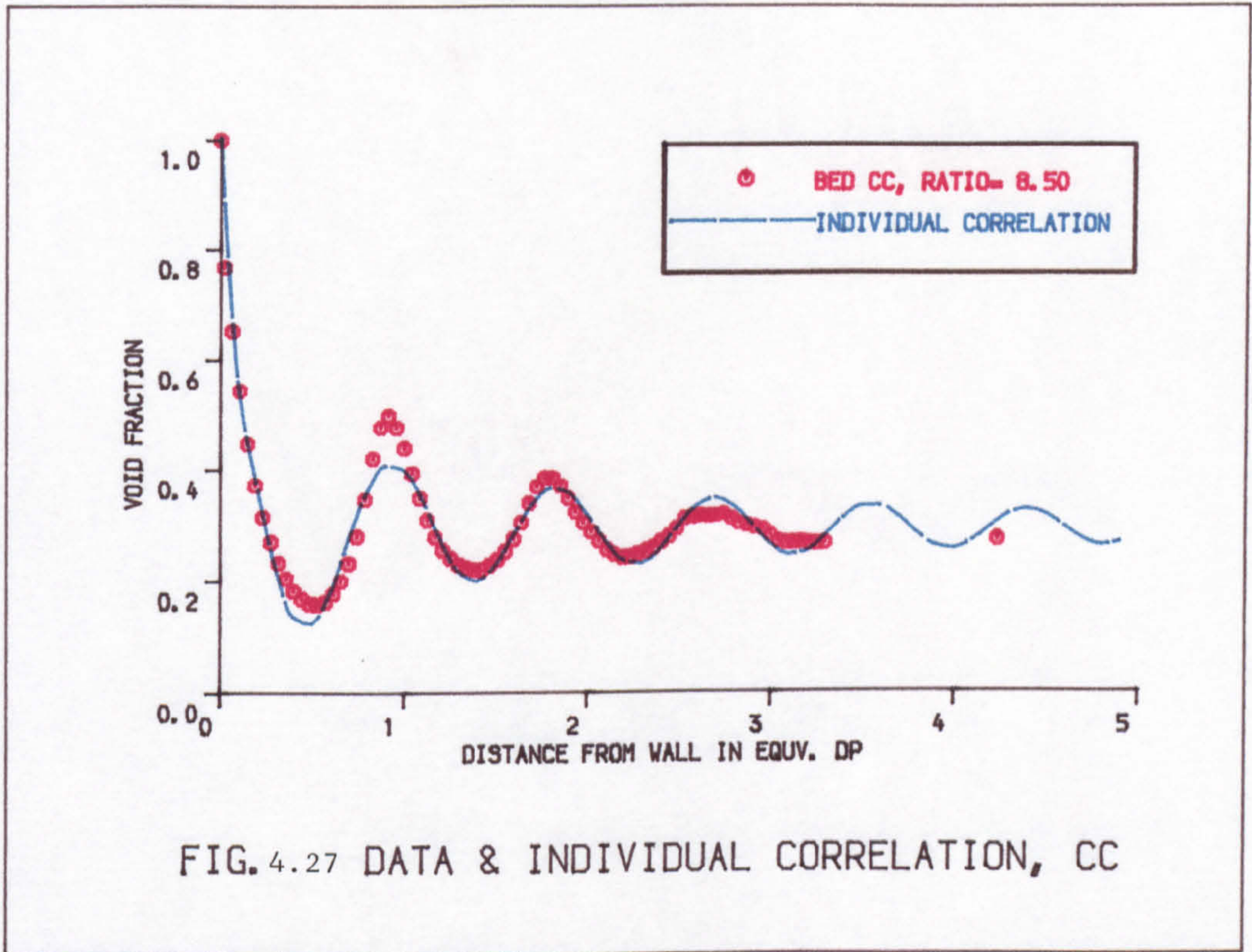
It is essential to characterise the corresponding correlation coefficients, α_s , of every individual expression to develop a generalized form of correlation. Several forms of expressions which were believed to be suitable for the prediction purposes

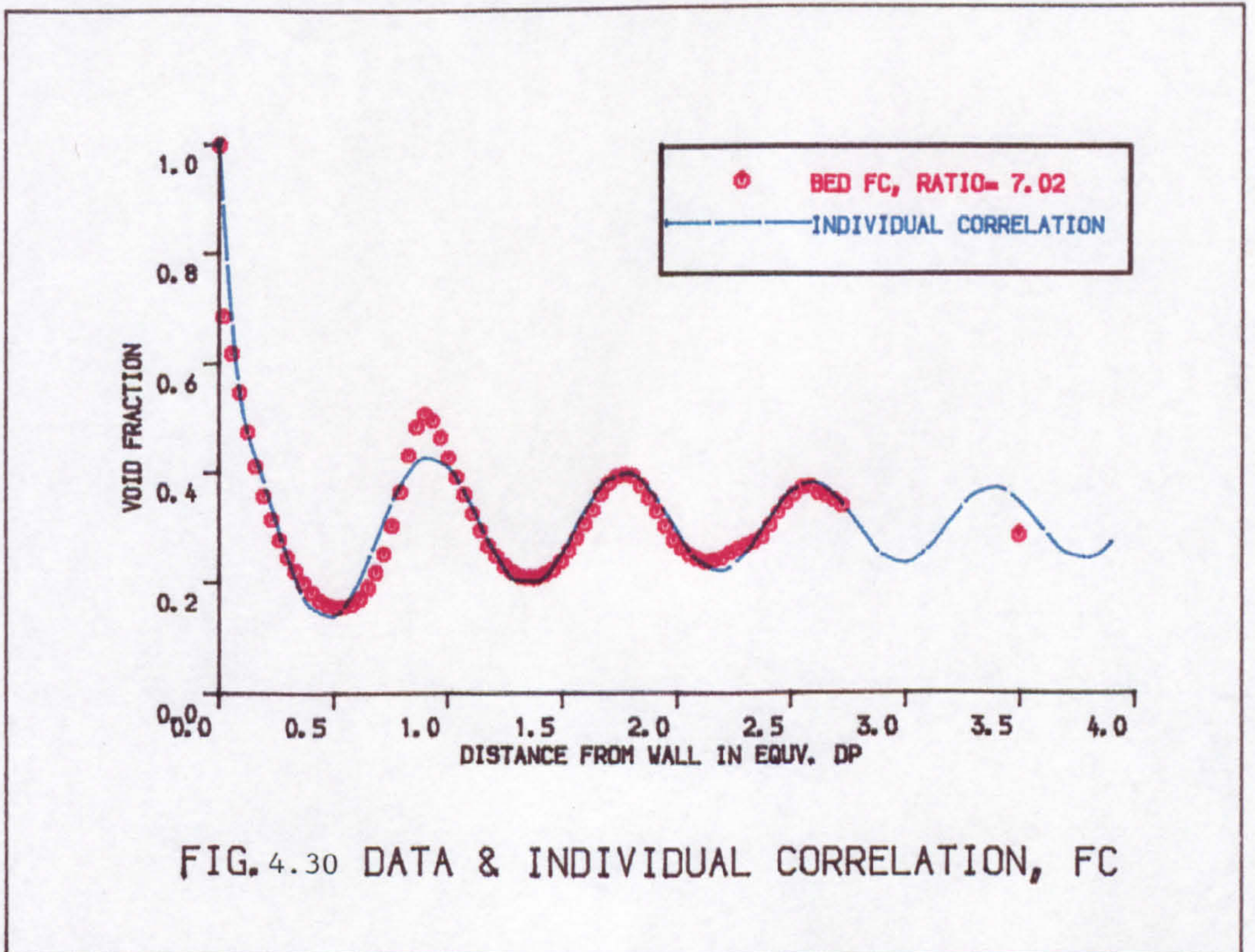
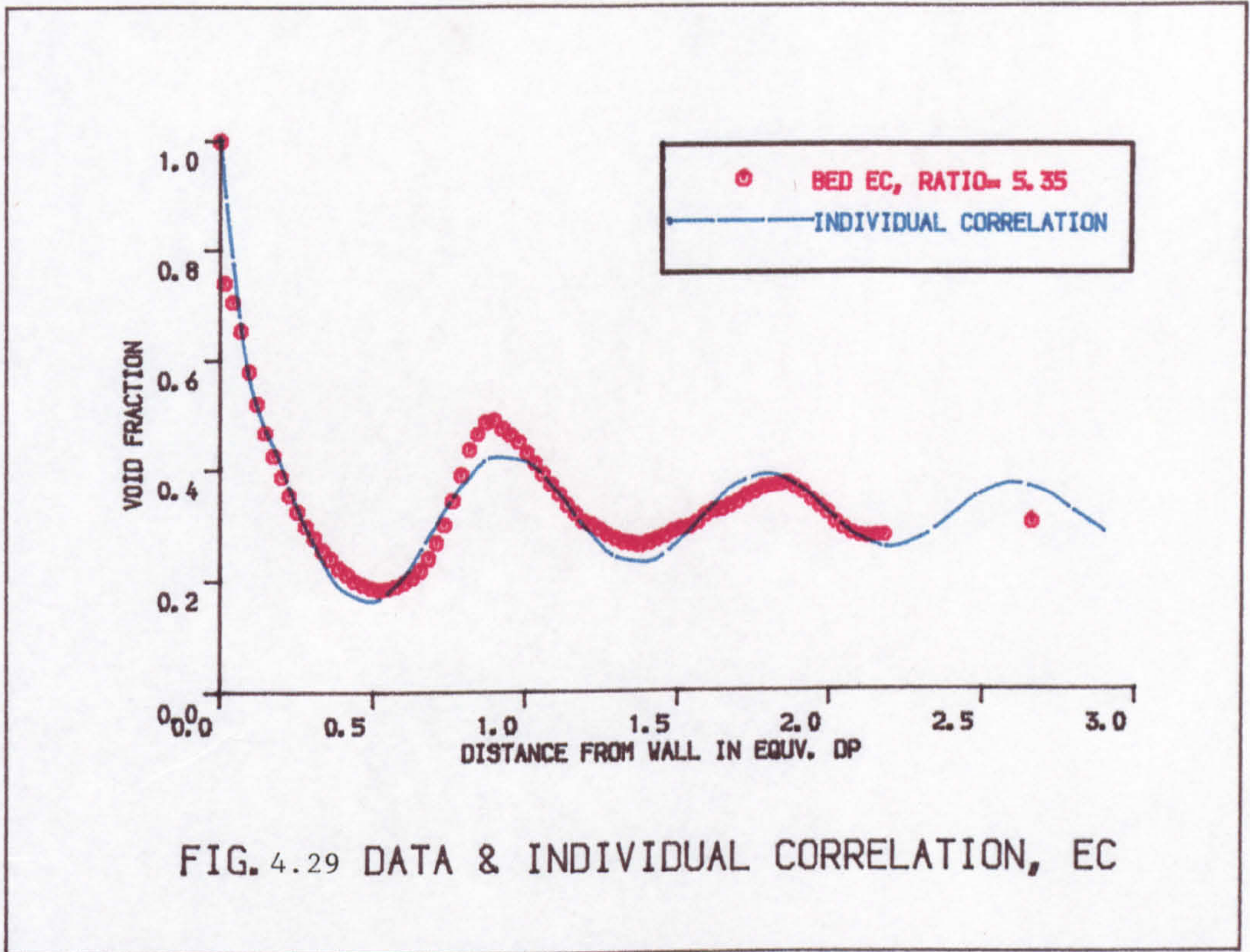
TABLE 4.3

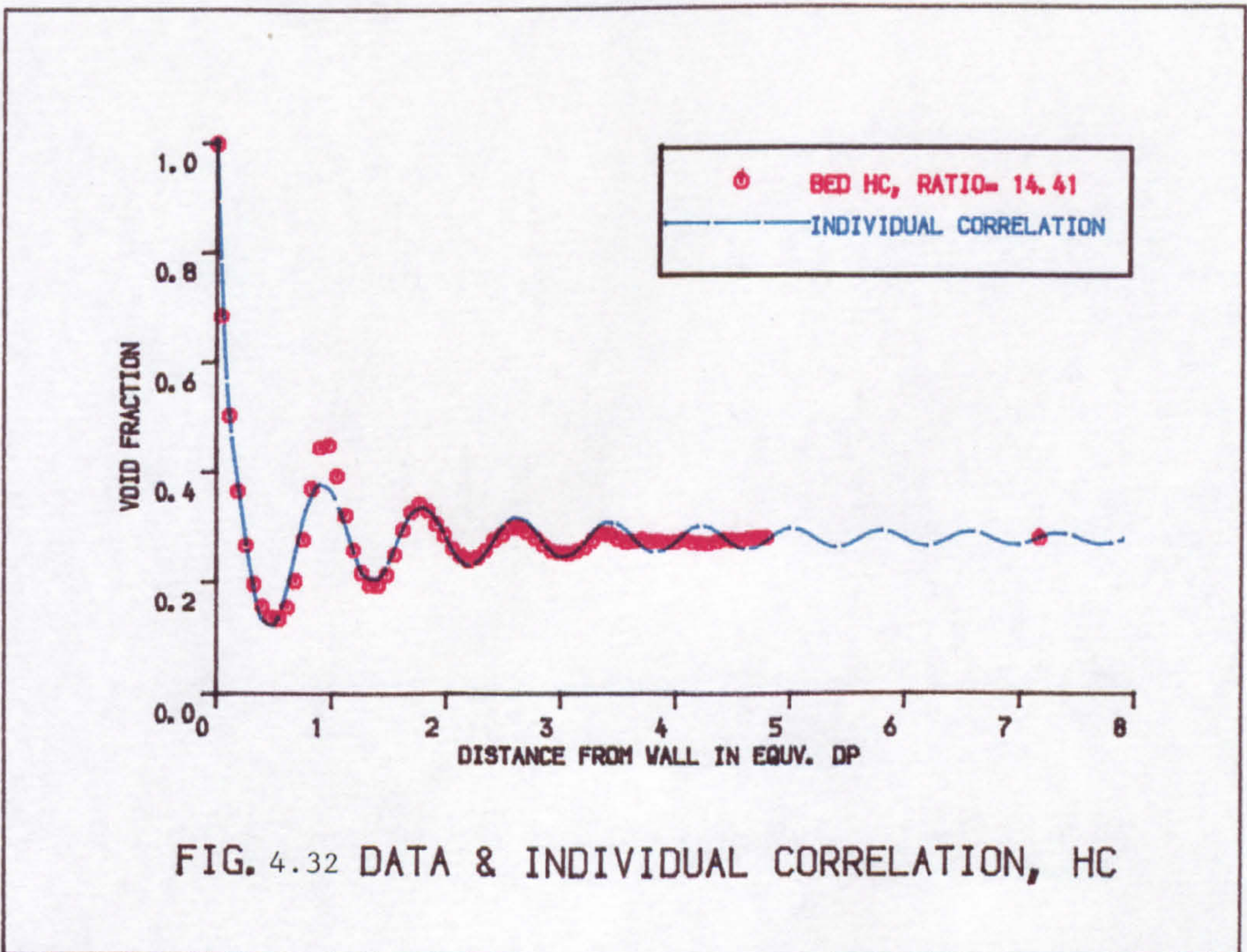
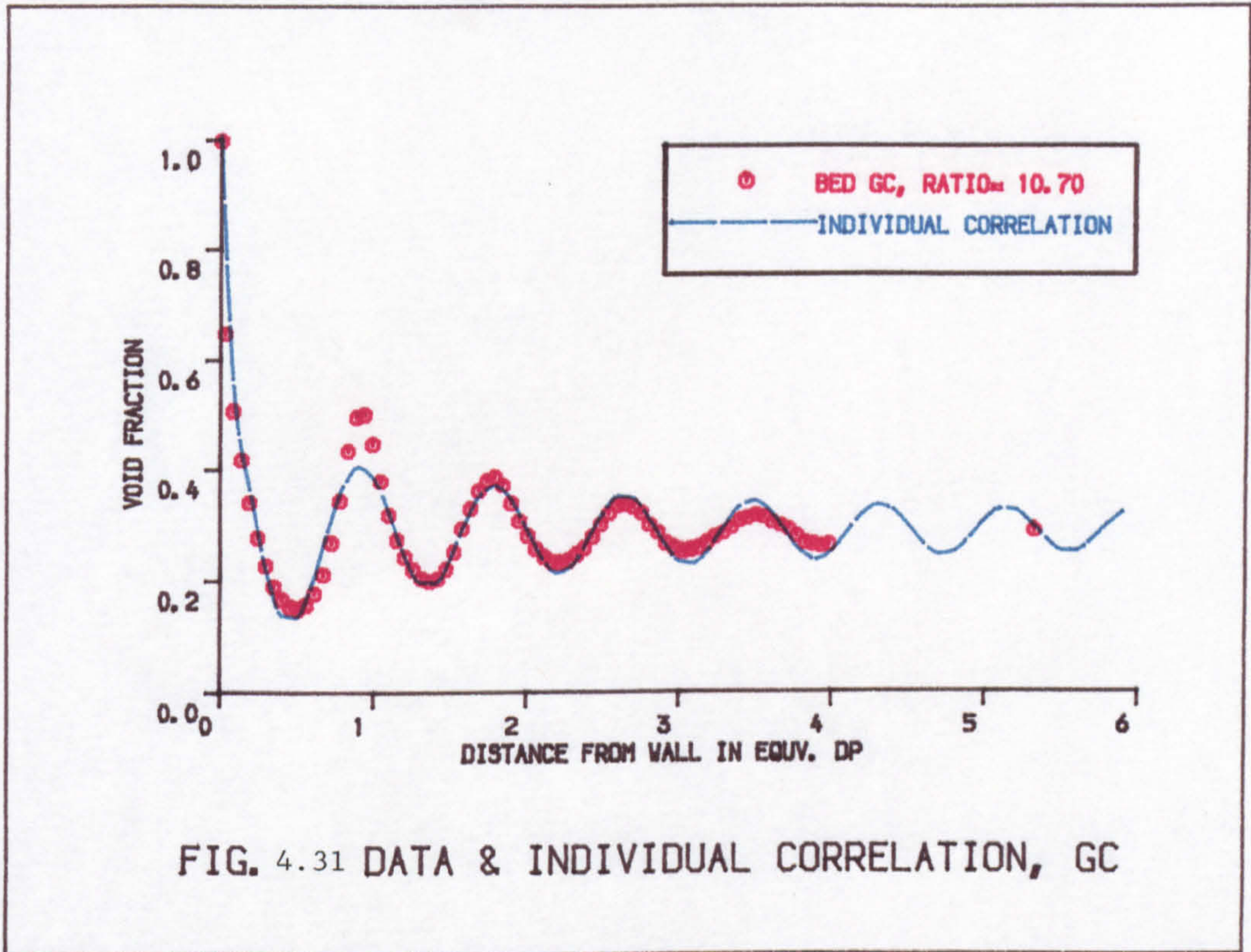
Estimates of the Individual Correlation Parameters for Radial Variation

ID	d_t/d_{pe}	α_1	α_2	α_3	α_4	α_5	Σe^2	SEE
AC	8.5	0.6800	1.9303	6.5317	0.3243	1.0524	514.2	2.6×10^{-2}
BC	8.5	0.6725	1.8558	6.5968	0.3206	1.0541	418.7	2.3×10^{-2}
CC	8.5	0.7070	1.8074	6.6489	0.3442	1.0480	460.7	2.5×10^{-2}
DC	3.37	0.6526	1.8022	6.1559	0.4219	1.2409	561.7	2.8×10^{-2}
EC	5.35	0.6823	1.8423	6.6596	0.3205	1.0849	498.4	2.5×10^{-2}
FC	7.02	0.7000	1.7187	6.7728	0.2514	1.0757	523.4	2.6×10^{-2}
GC	10.70	0.7093	1.8186	6.8420	0.2678	1.0422	452.3	2.5×10^{-2}
HC	14.41	0.7243	1.9940	6.7108	0.3785	1.0690	305.1	2.1×10^{-2}
YC	22.05	0.7141	1.7693	6.8194	0.3668	1.0358	279.6	2.0×10^{-2}
ZC	27.95	0.7116	1.7742	6.7197	0.4041	1.0602	201.0	1.7×10^{-2}









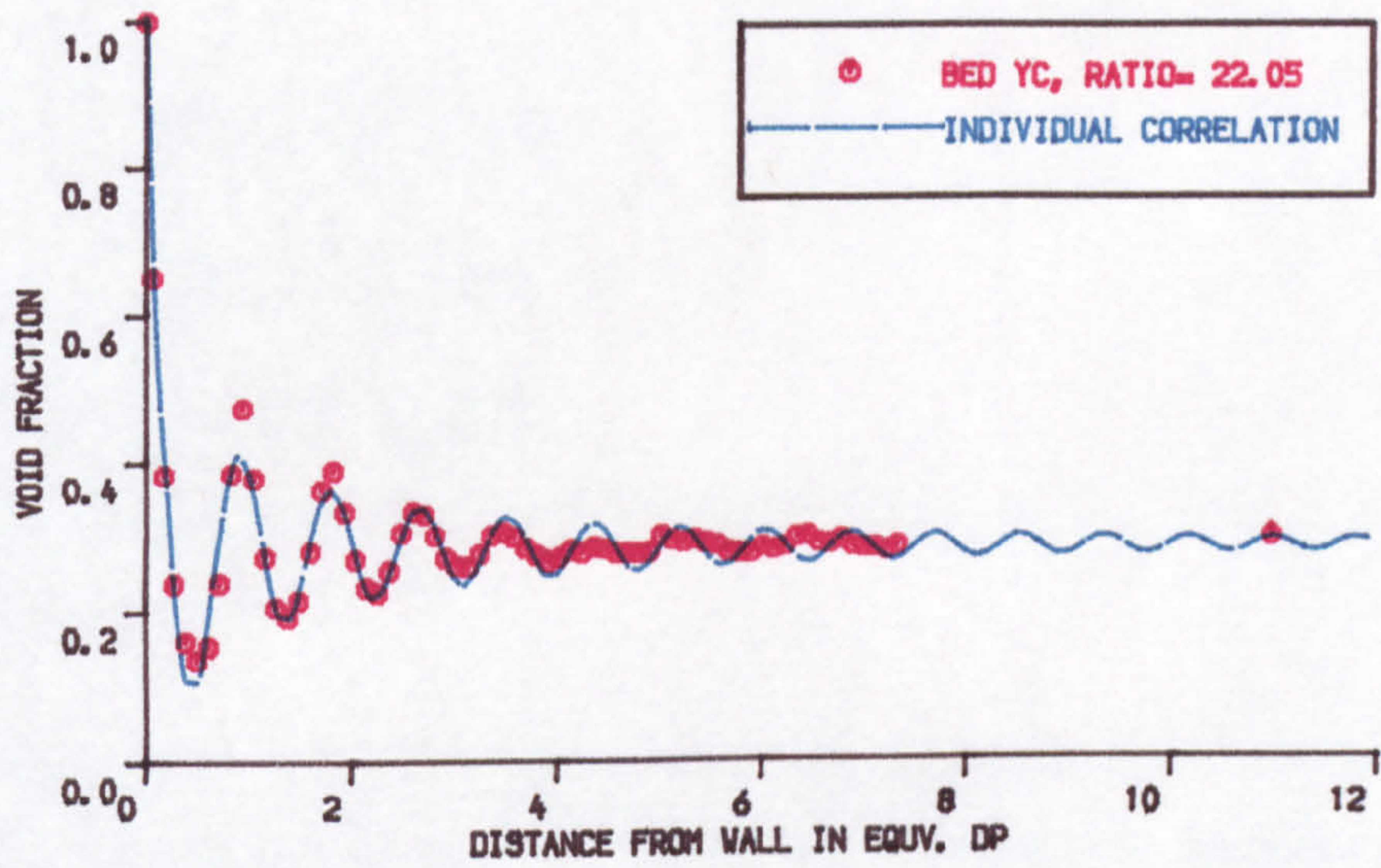


FIG. 4.33 DATA & INDIVIDUAL CORRELATION, YC

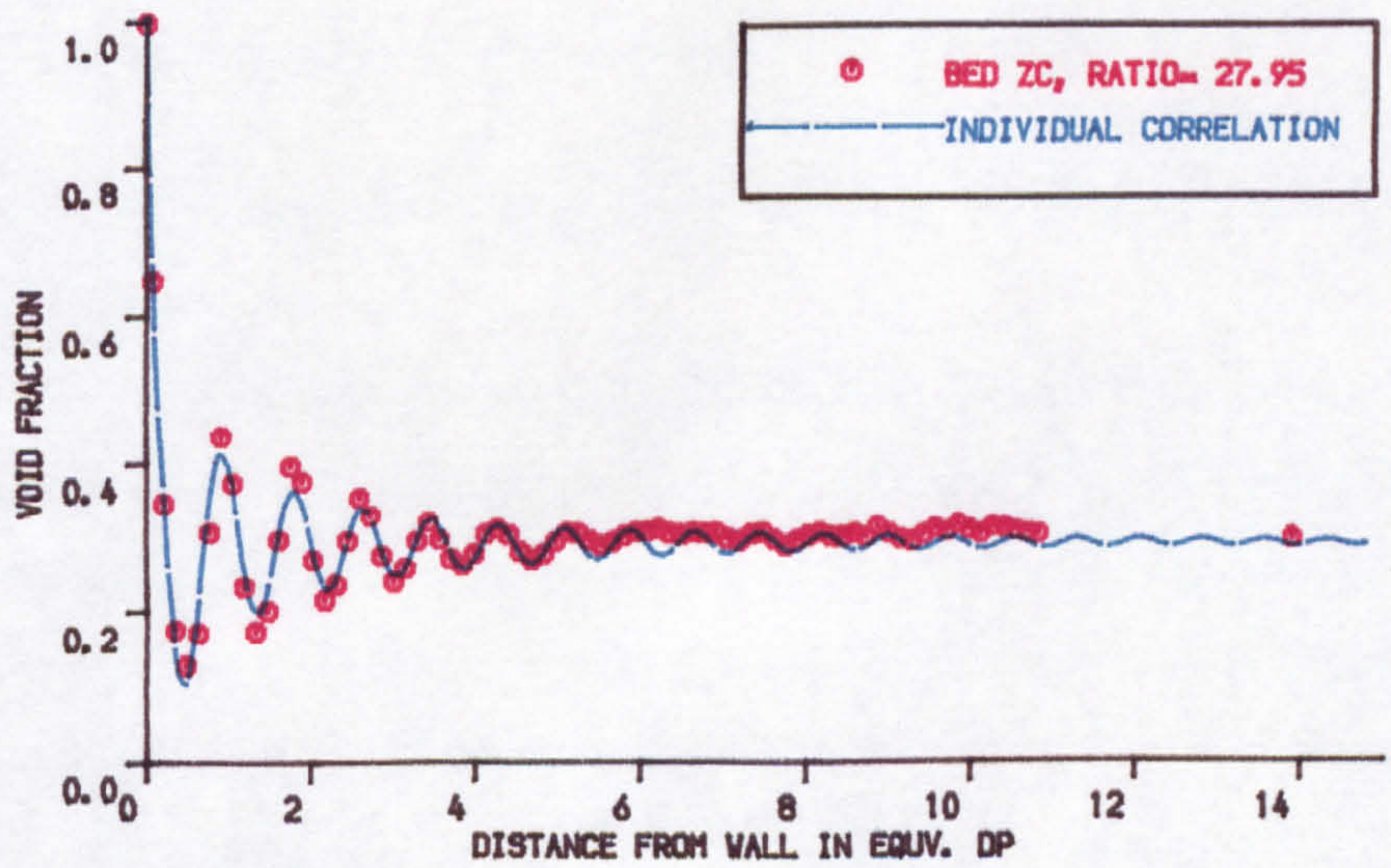


FIG. 4.34 DATA & INDIVIDUAL CORRELATION, ZC

have been considered. The values of SSE and SEE were a major factor in selecting the appropriate expressions. The resultant profiles are shown in Appendix B and the mathematical forms for the individual correlation of coefficients are as follows:

$$\alpha_1 = \frac{d_r}{1.375 d_r + 0.526} \quad (4.3)$$

$$\alpha_2 = \frac{d_r}{0.542 d_r + 0.032} \quad (4.4)$$

$$\alpha_3 = \frac{d_r}{0.145 d_r + 0.048} \quad (4.5)$$

$$\alpha_4 = 4.7 \times 10^{-4} d_r^2 - 0.012 d_r + 0.392 \quad (4.6)$$

$$\alpha_5 = 1.236 d_r^{-0.061} \quad (4.7)$$

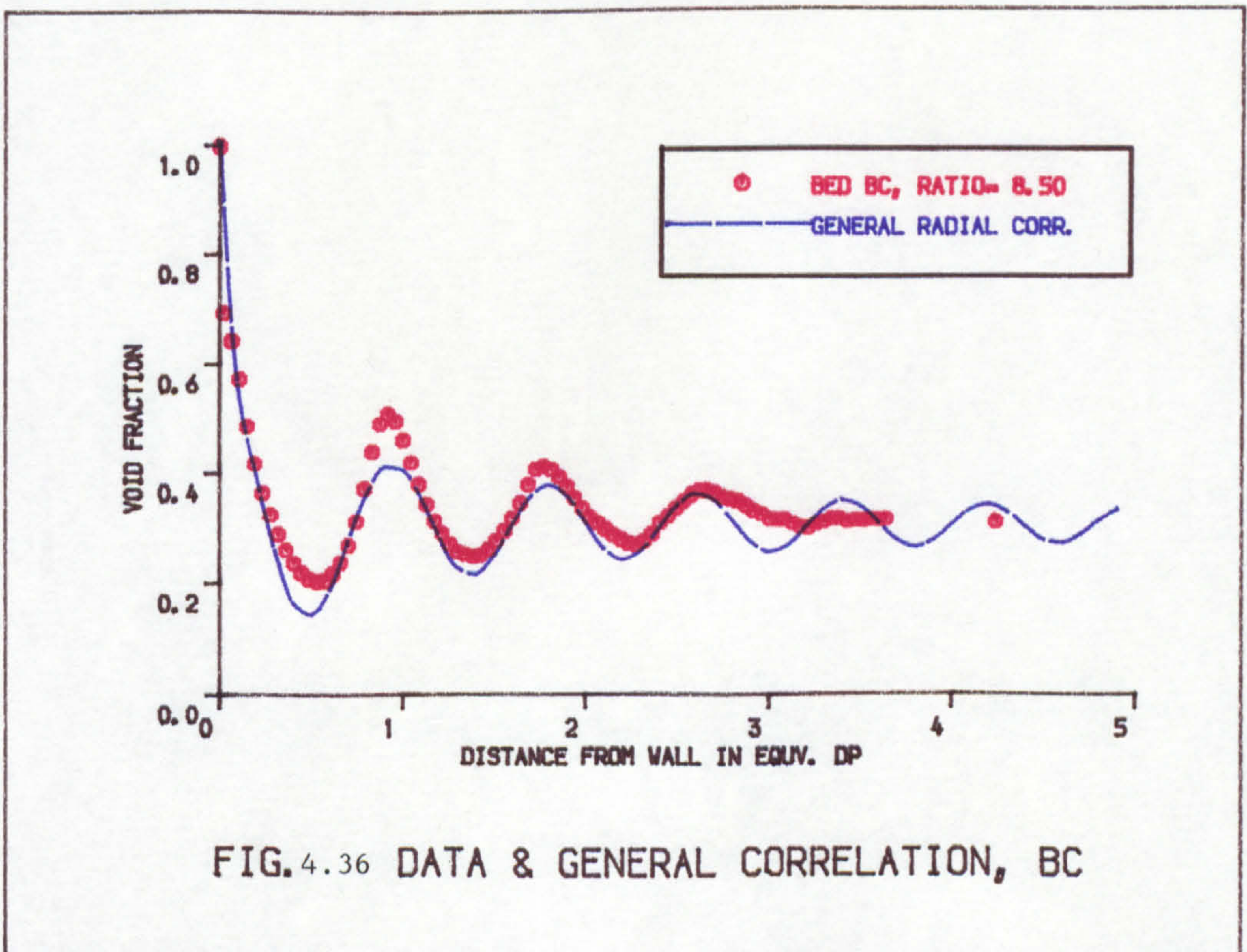
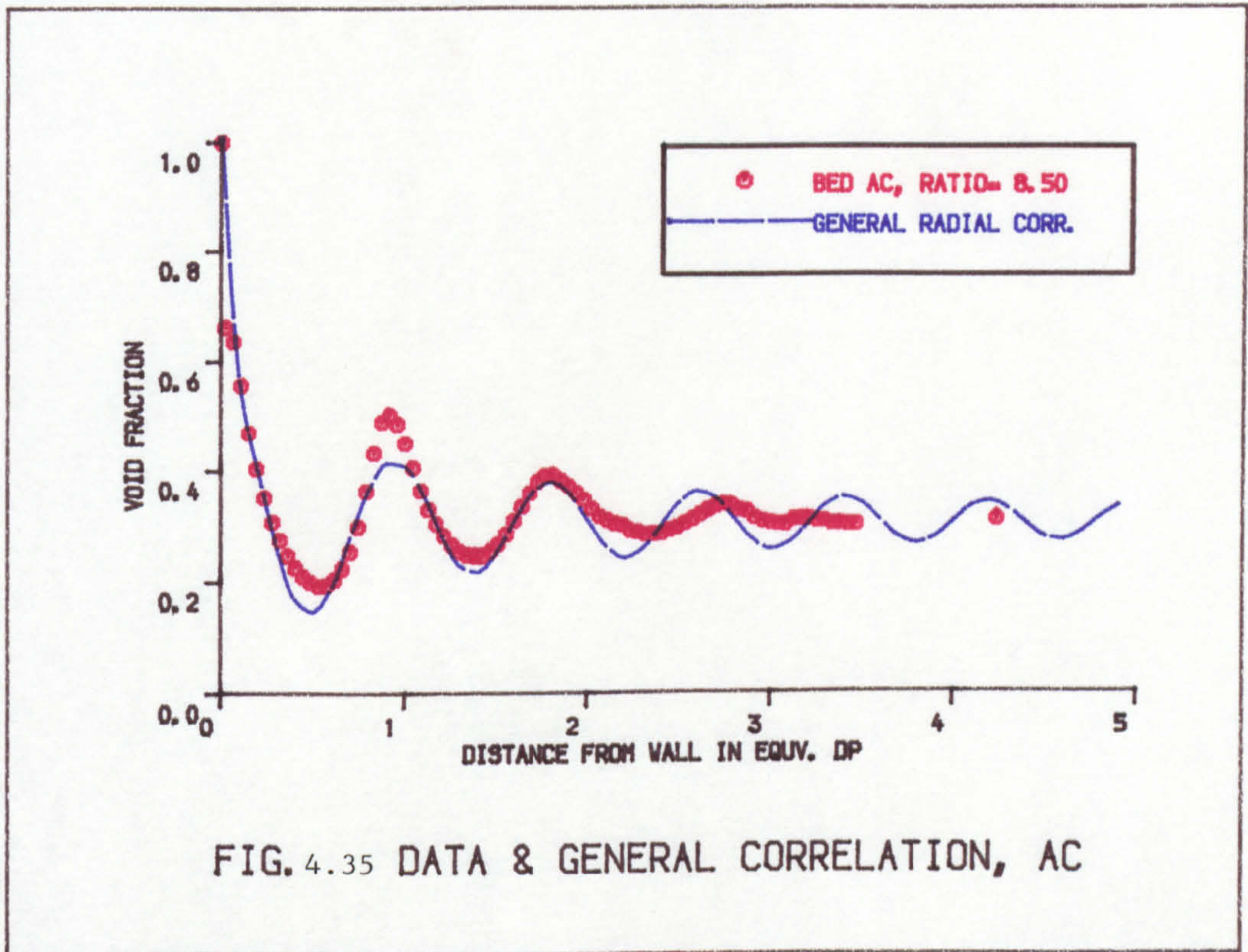
The values calculated for each α can then be substituted into Equation (4.2) to predict the radial distribution of voidage for that particular bed. Figures 4.35 to 4.44 show the closeness of this generalized correlation to the data obtained from each test bed.

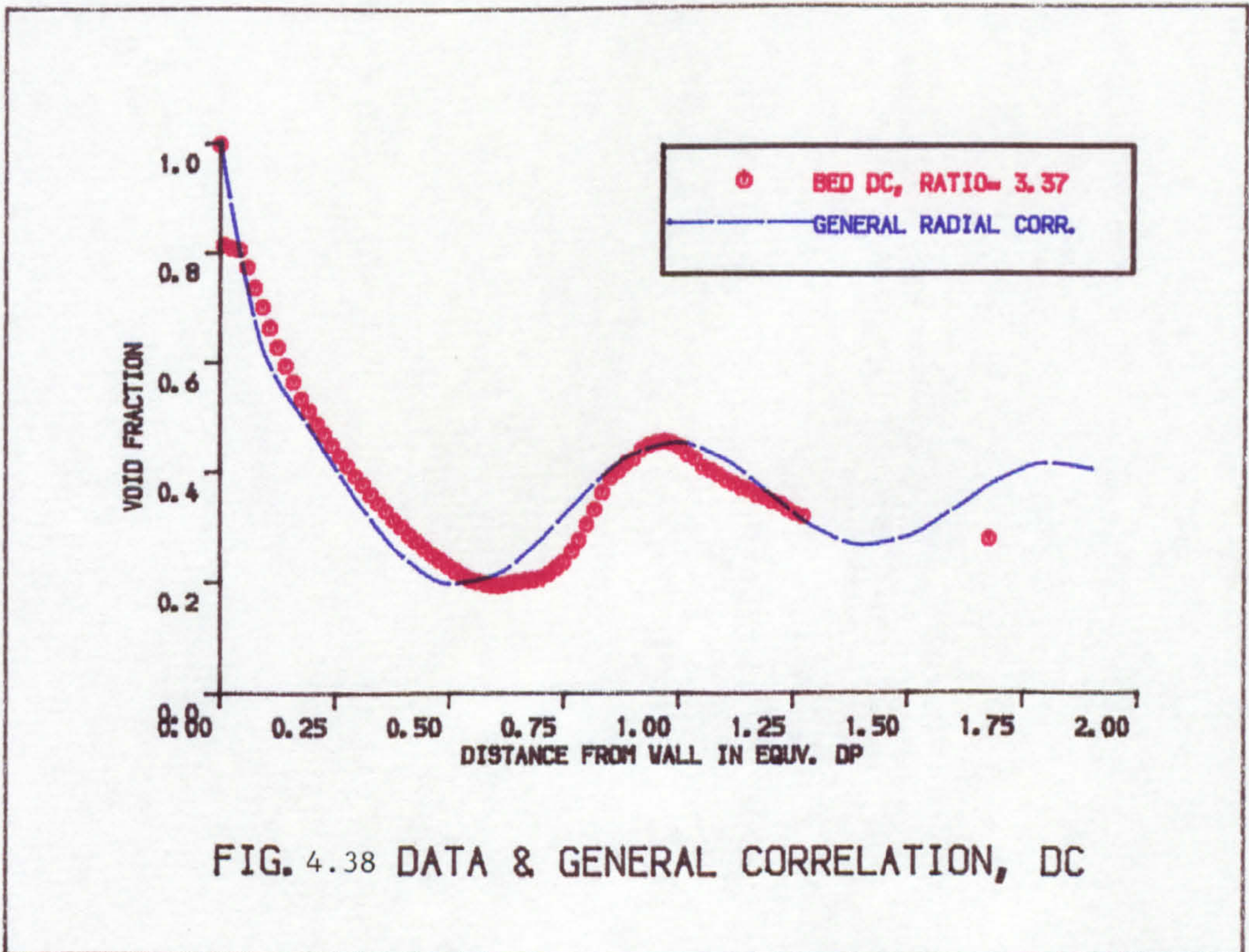
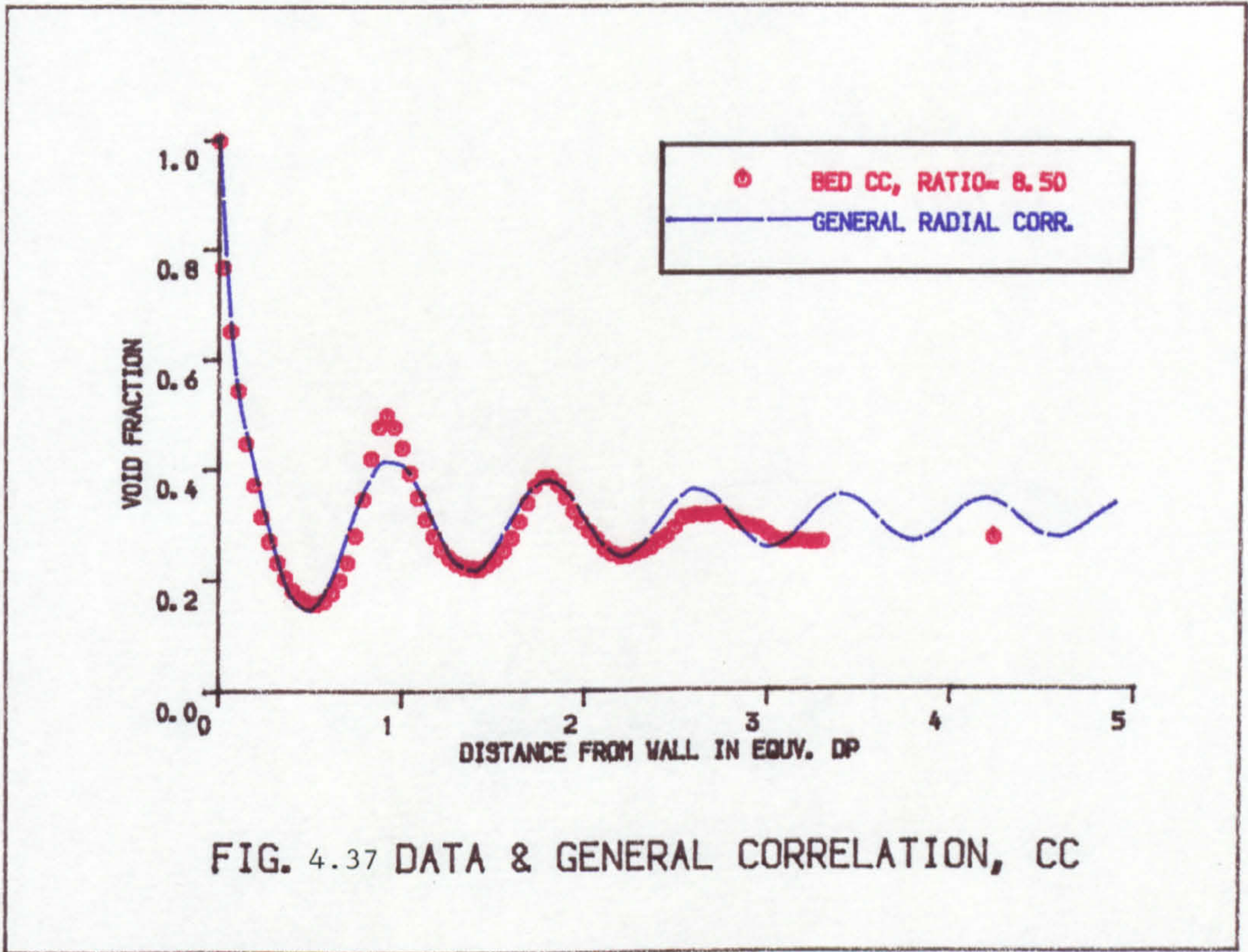
A simplified version of this generalized correlation was then considered when $\alpha = c$, where c is an optimised constant value:

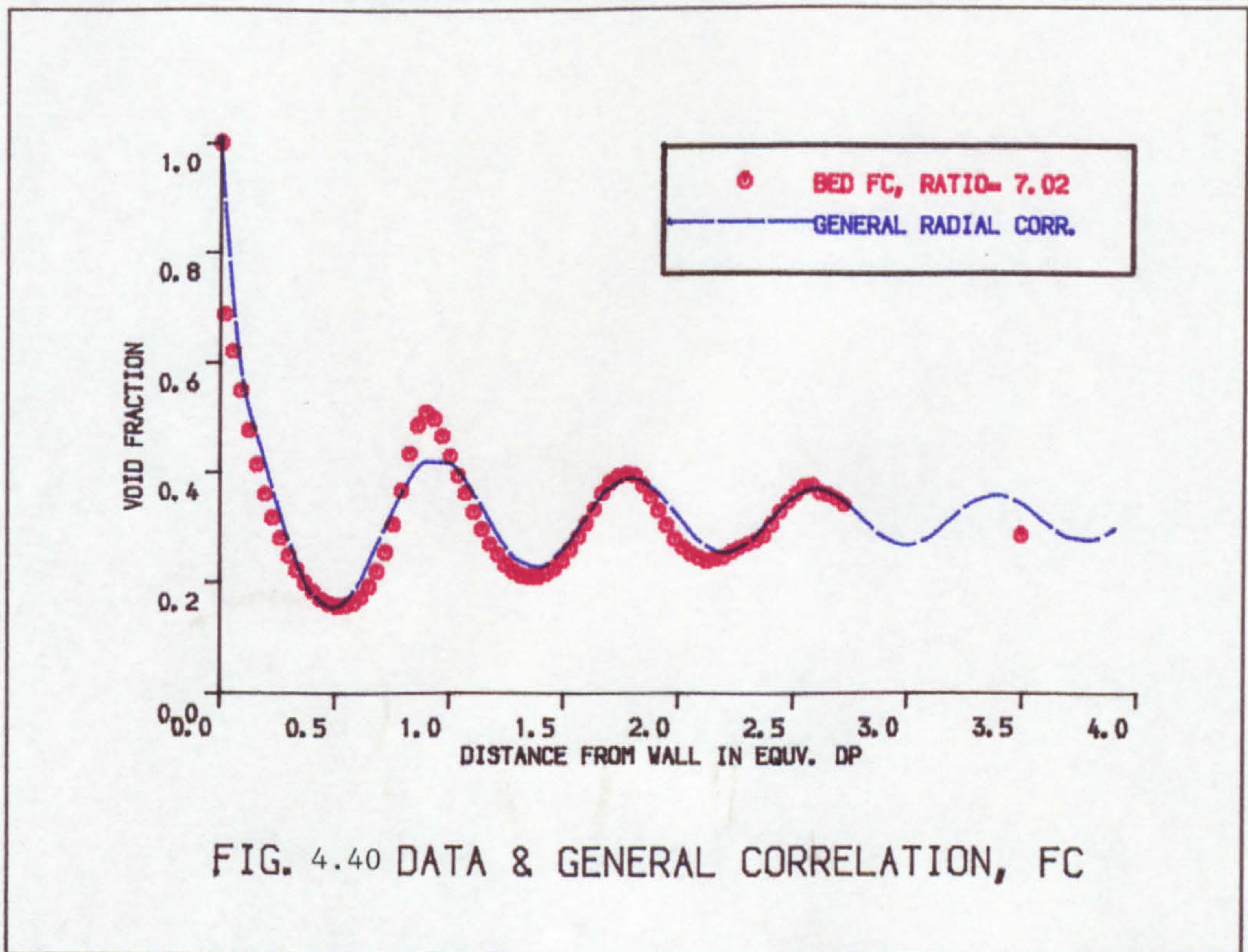
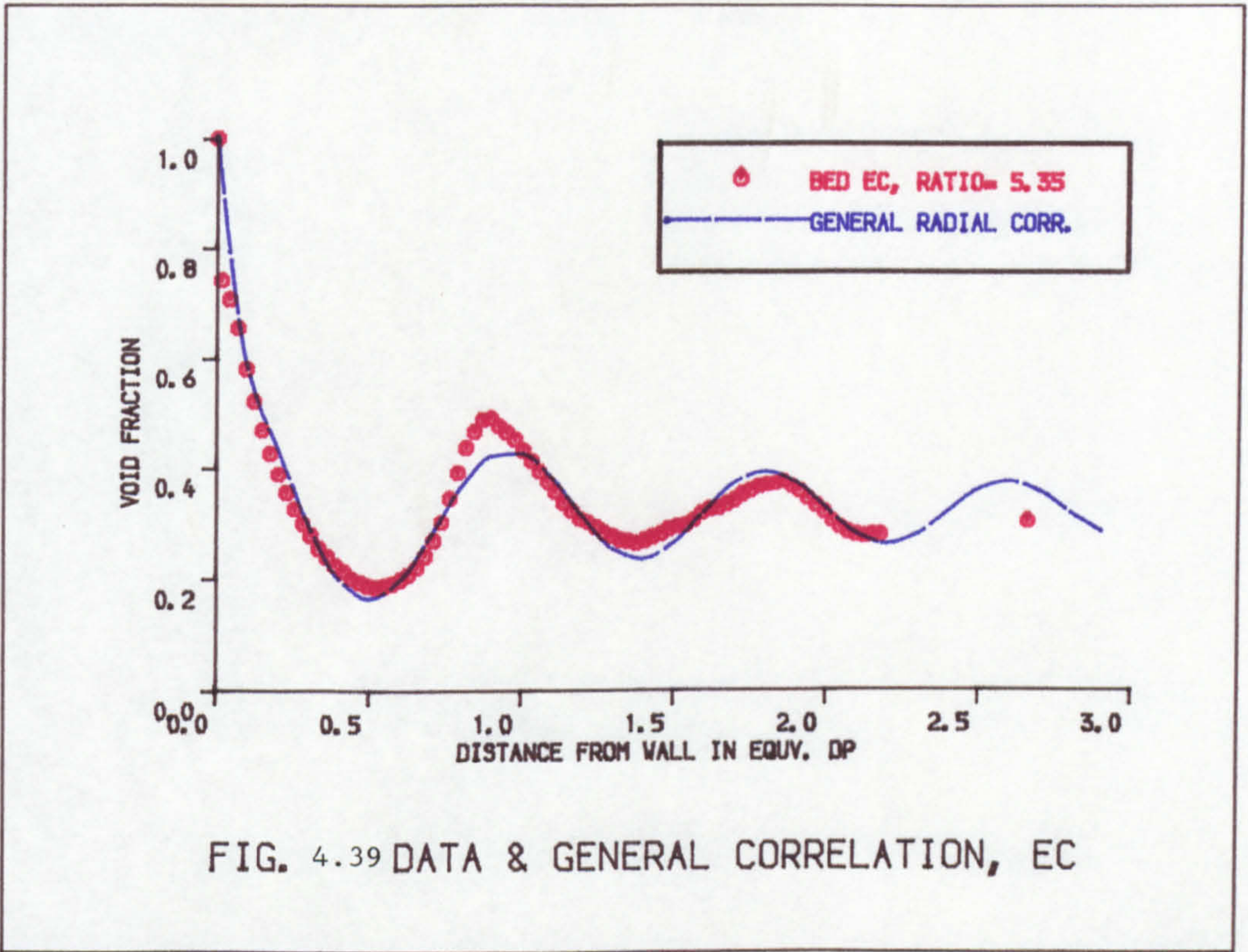
$$\epsilon(r) = 1.0 - 0.695 \{1.0 - \exp(-1.83 d_r^{0.34}) \cos(6.65 d_r^{1.08})\} \quad (4.8)$$

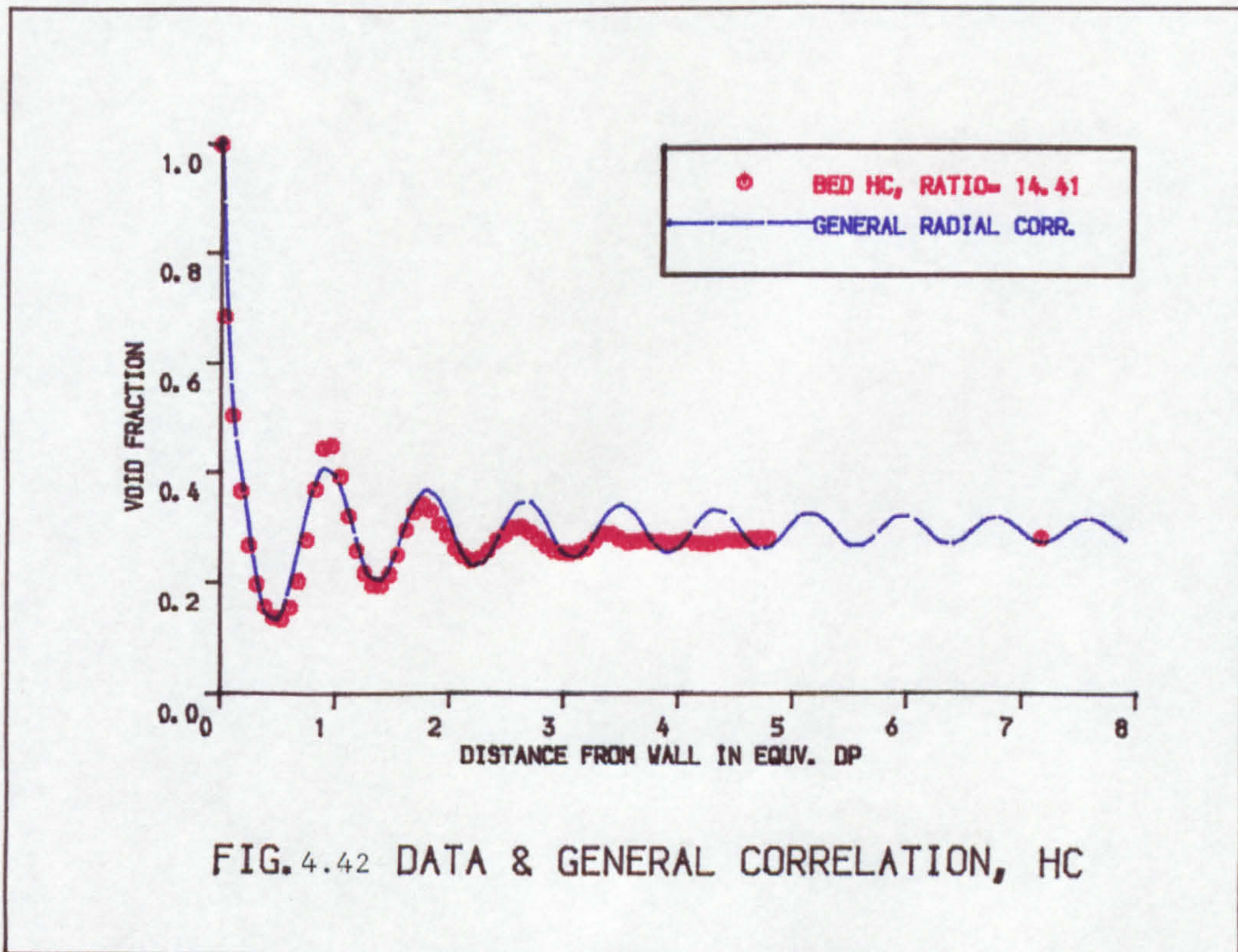
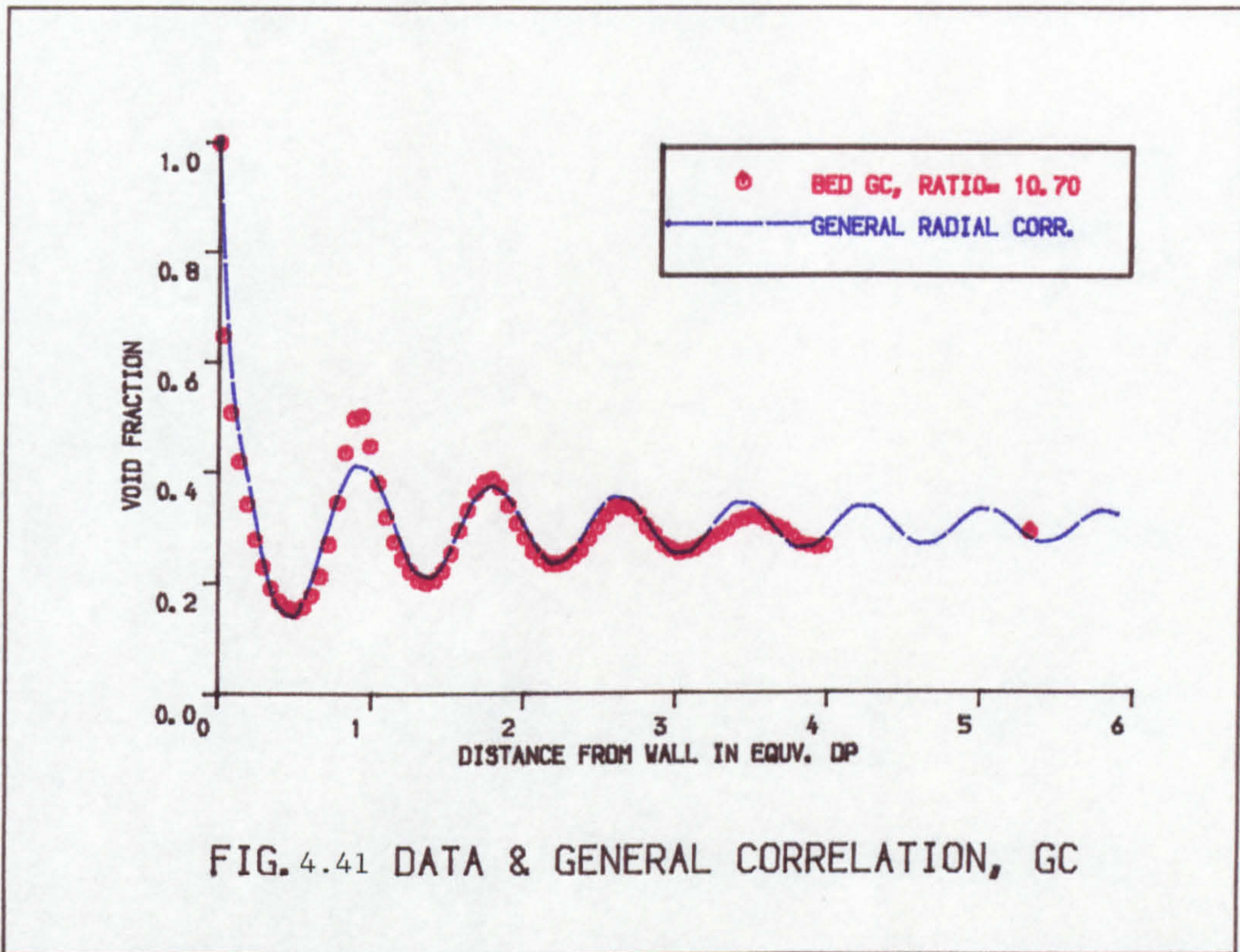
Appendix C contains the profiles of Equation (4.8) with the obtained data from each test bed.

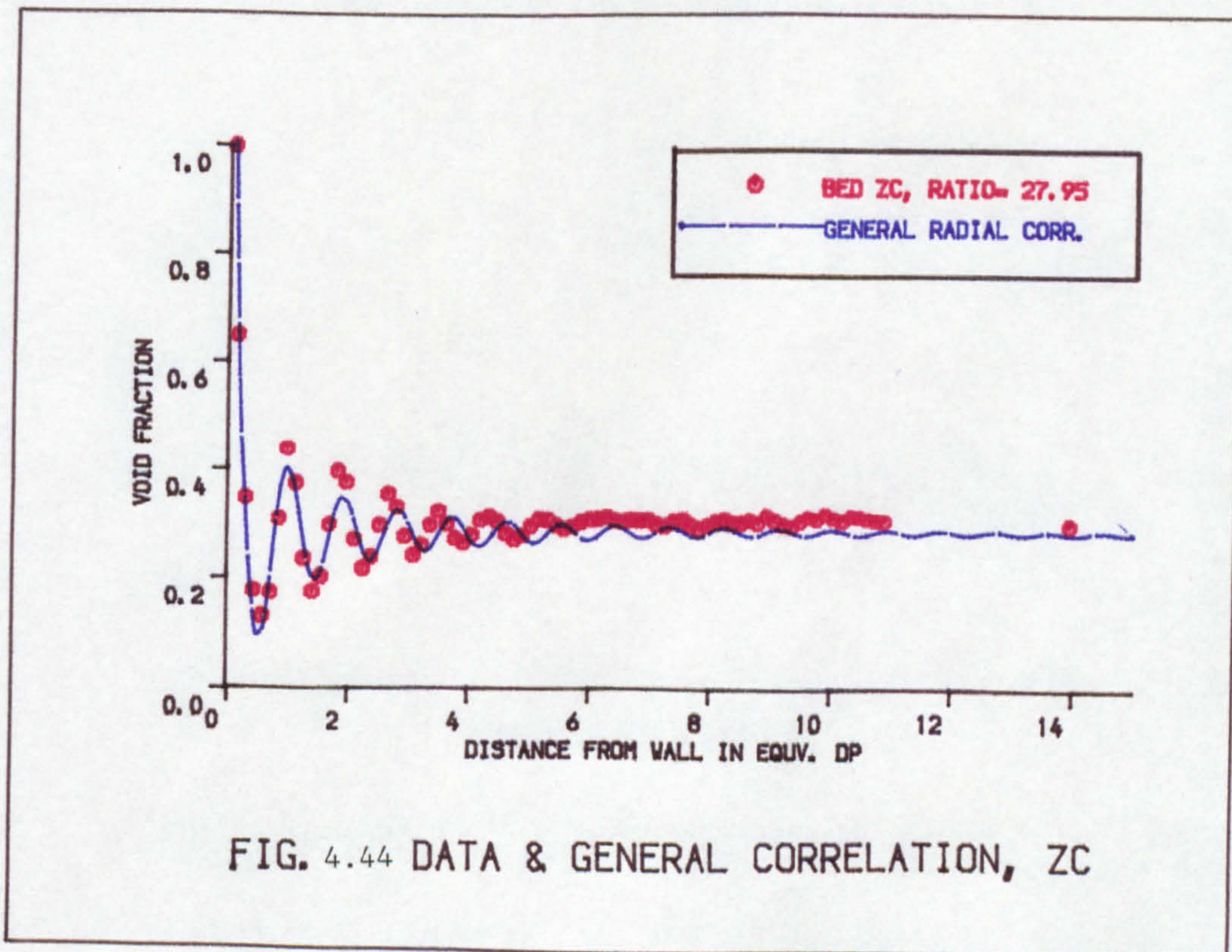
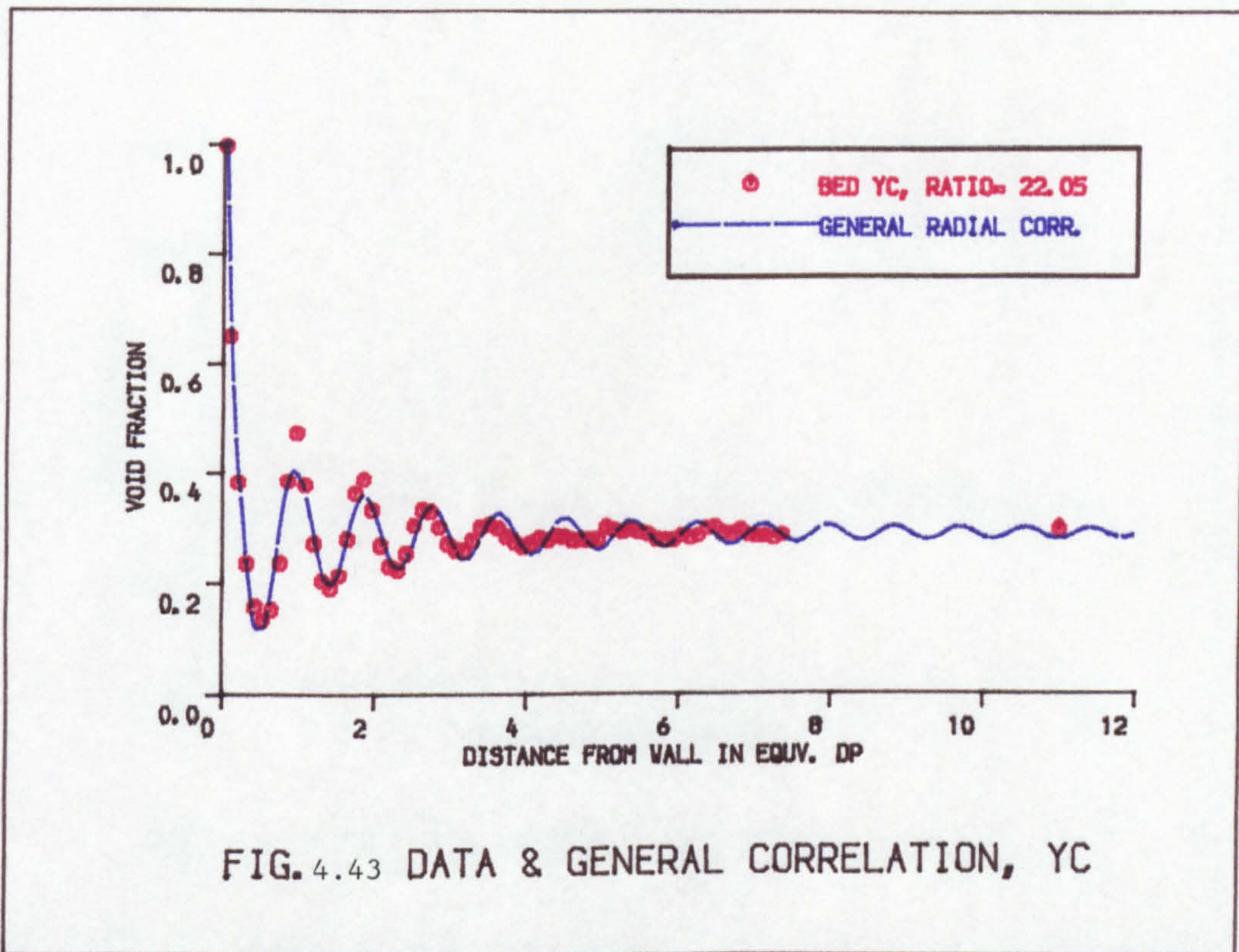
Since the variation of voidage in axial direction has a similar profile to that of the radial, the format of Equation (4.2) can be used for its characterization. Using the same procedure as described earlier, individual correlations were derived for each bed. Figures 4.45 to 4.54 compare the individual correlation profiles with the obtained experimental data. The correlation coefficients, β_s , which are associated with various characteristics of the pattern are evaluated for each bed together with SSE and SEE values are tabulated in Table 4.4.











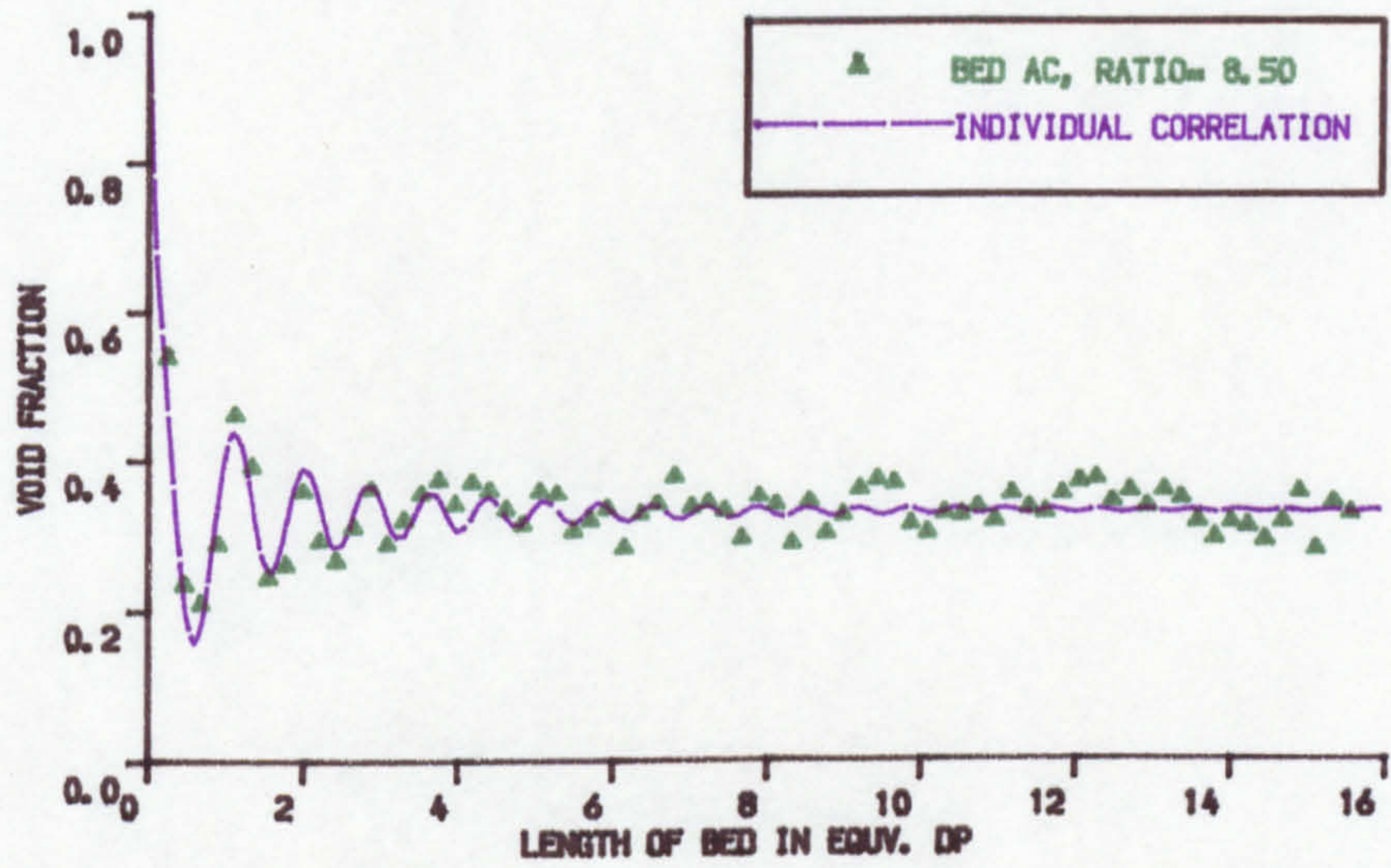


FIG. 4.45 DATA & INDIVIDUAL CORRELATION, AC

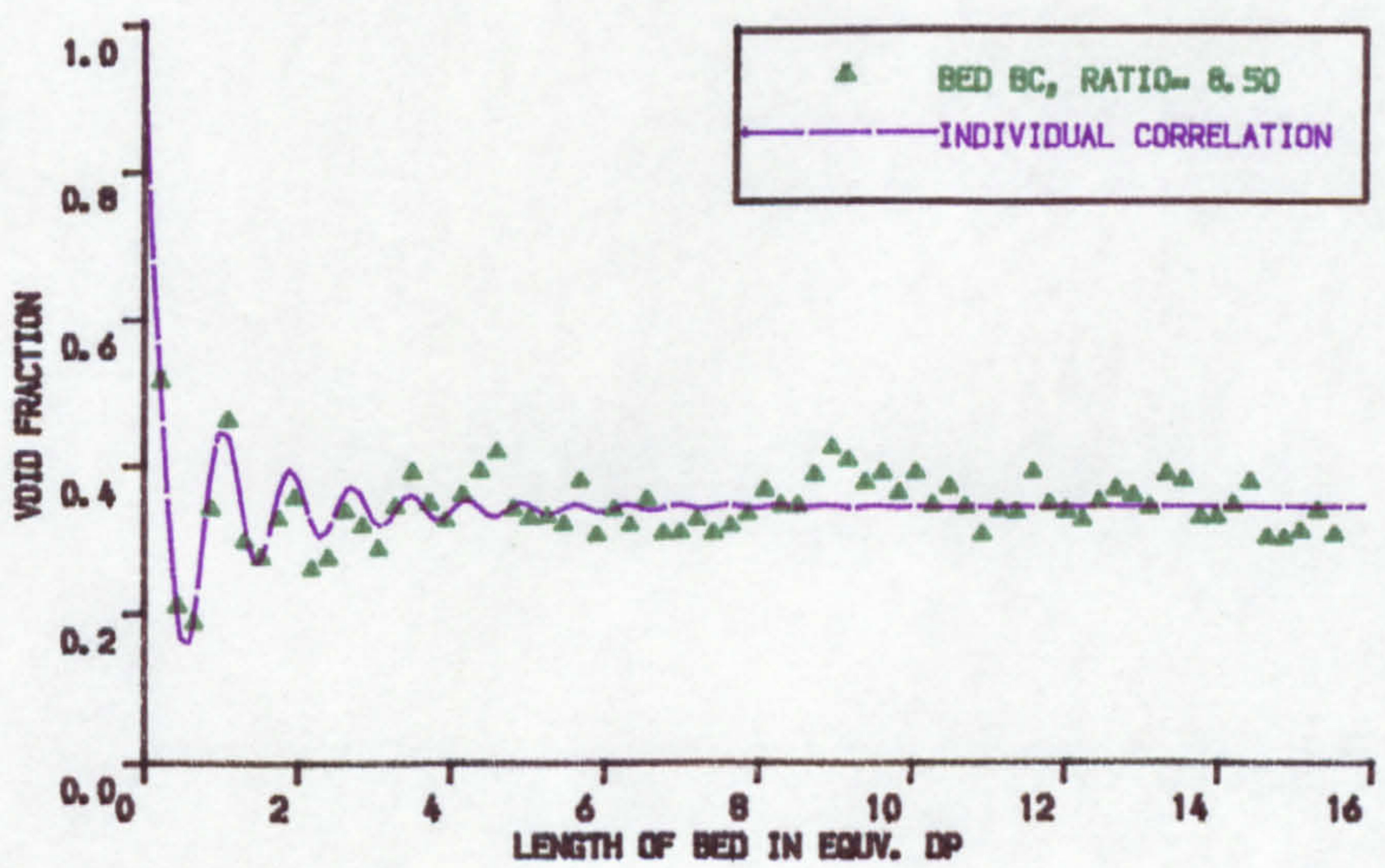
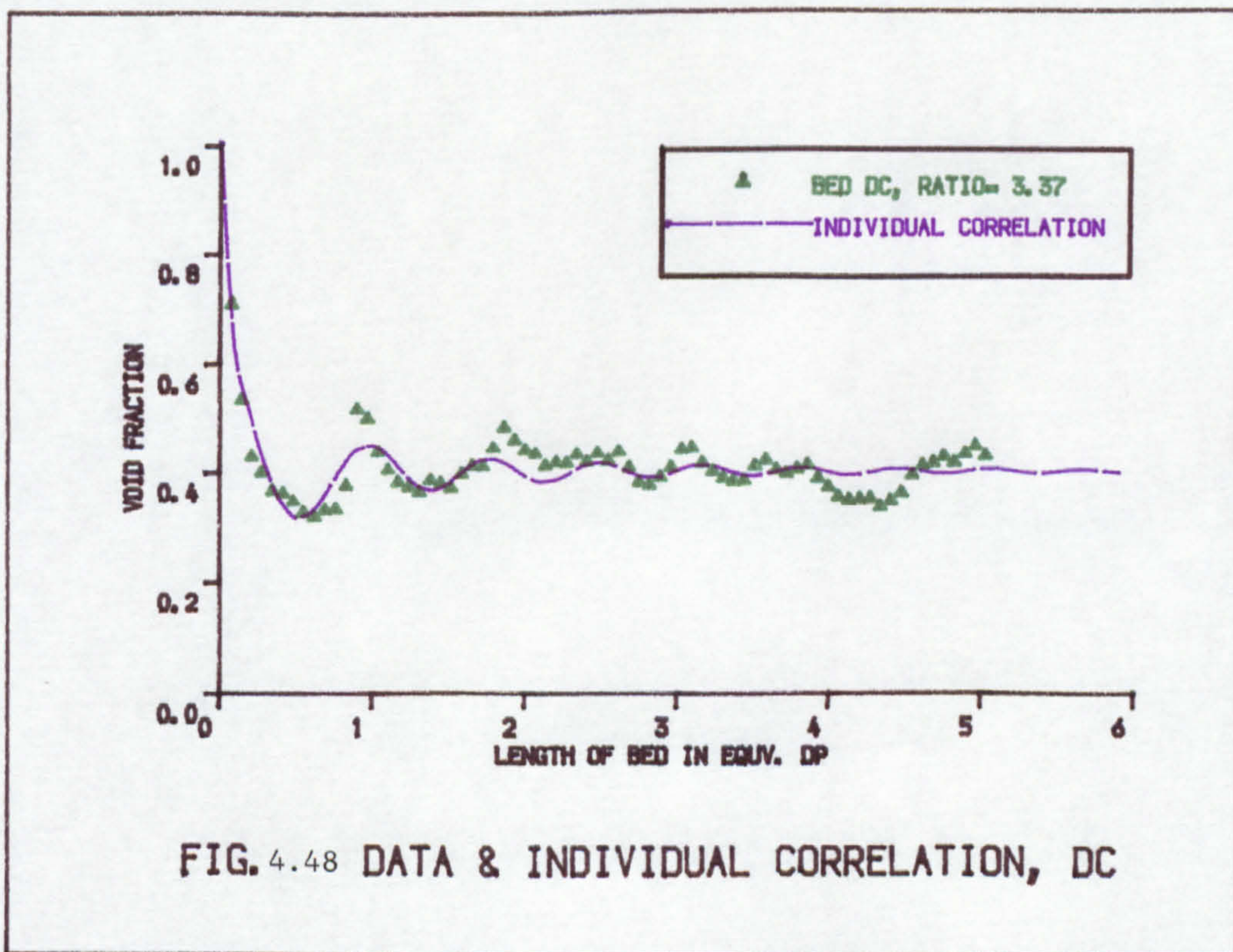
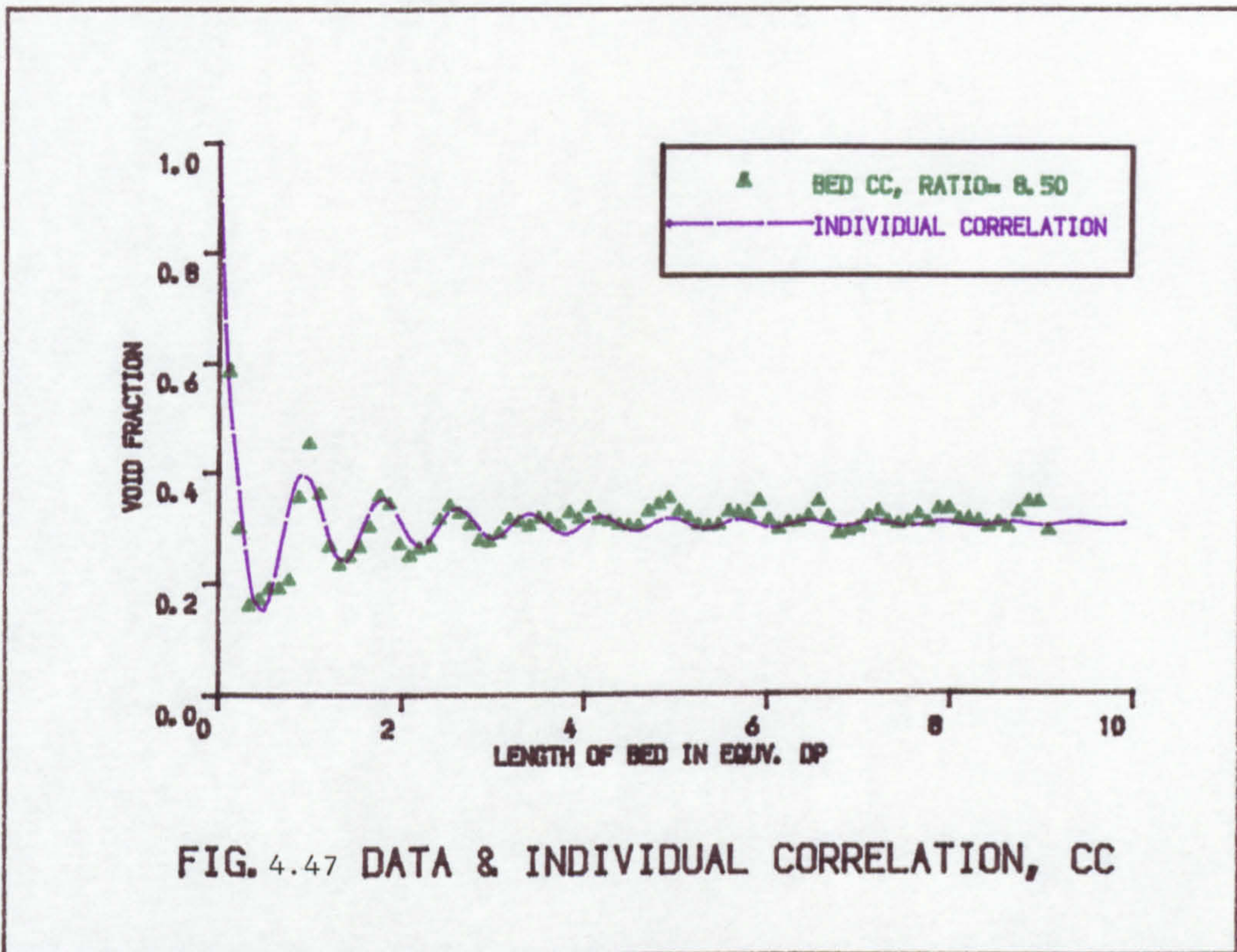


FIG. 4.46 DATA & INDIVIDUAL CORRELATION, BC



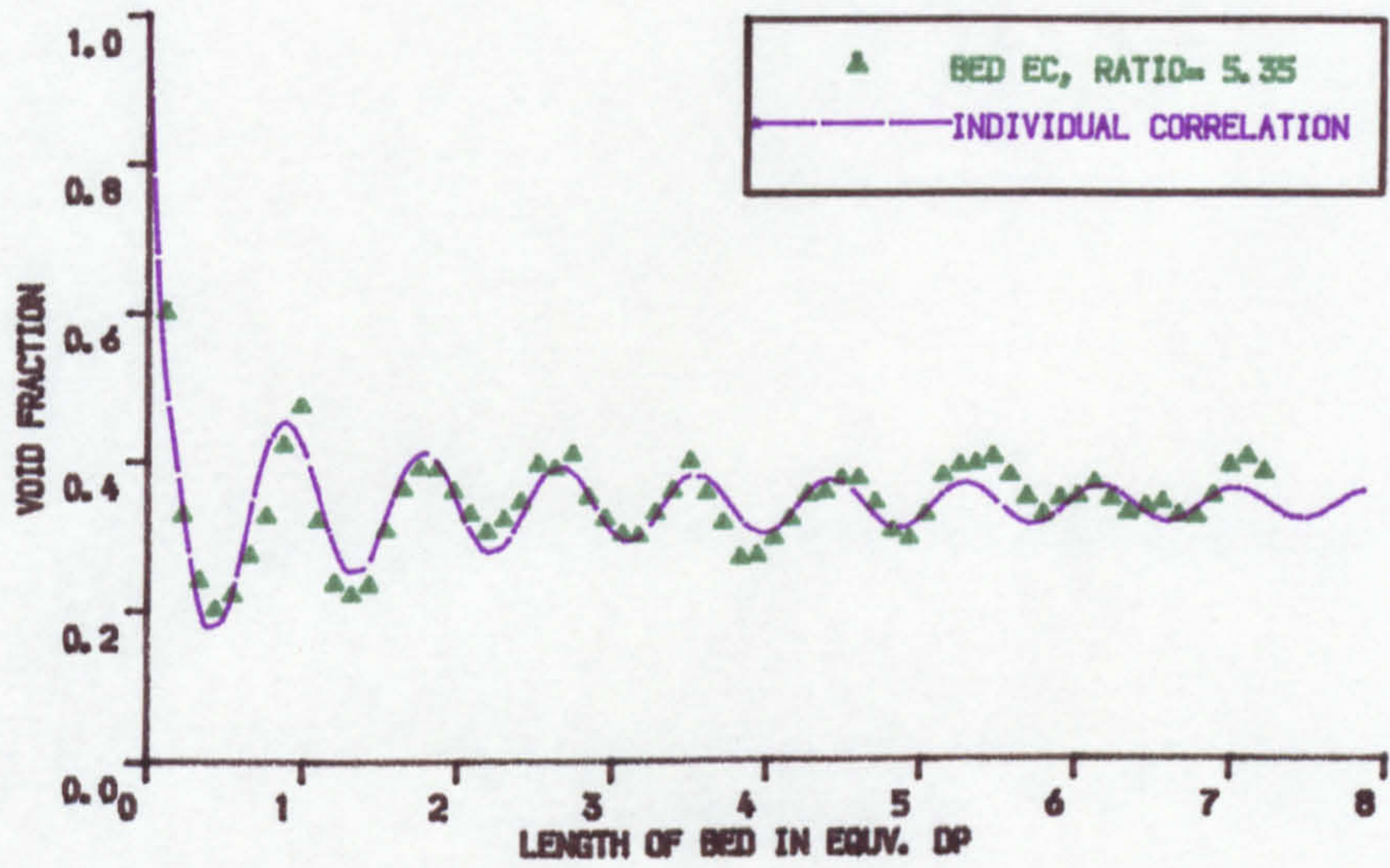


FIG. 4.49 DATA & INDIVIDUAL CORRELATION, EC

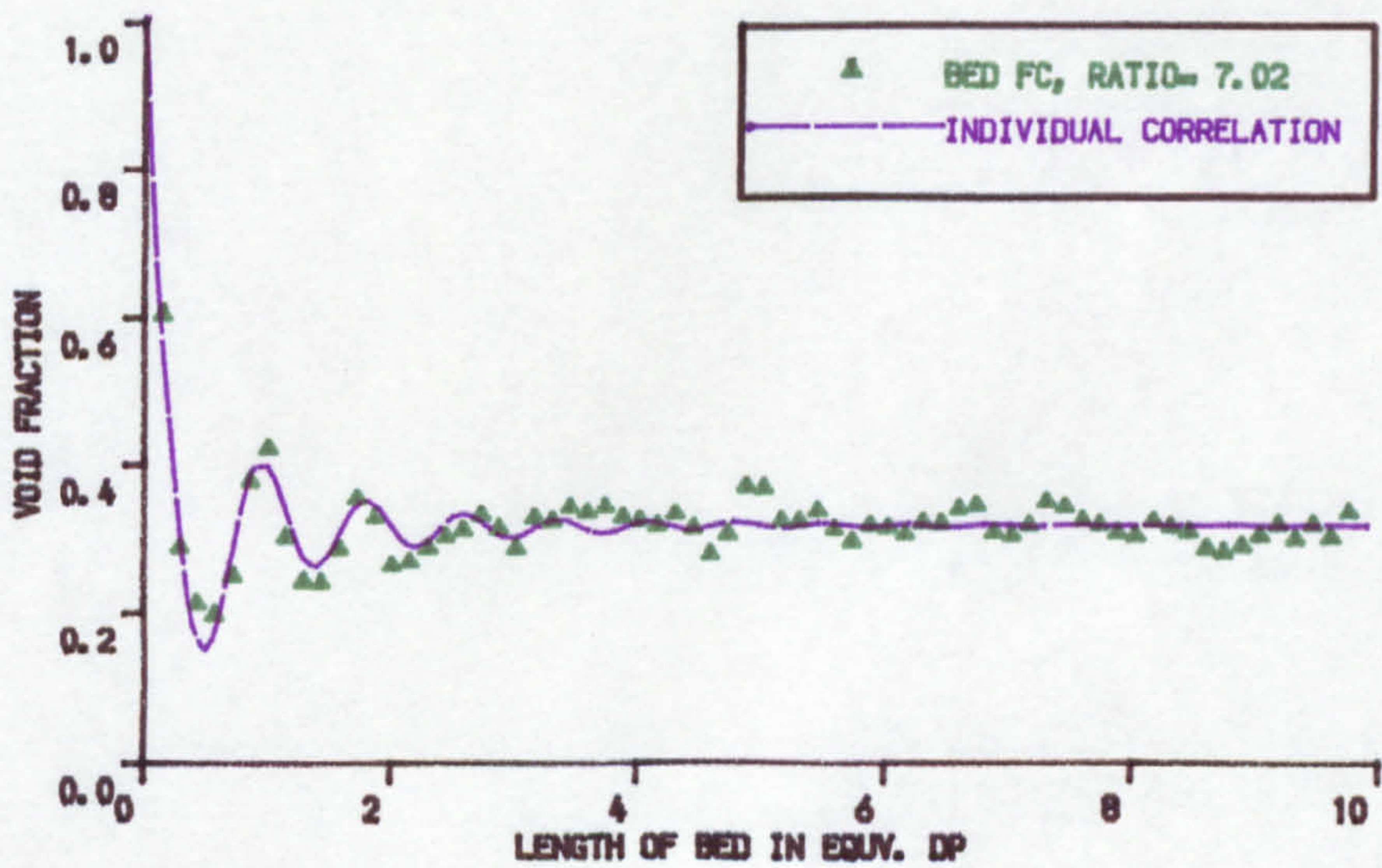
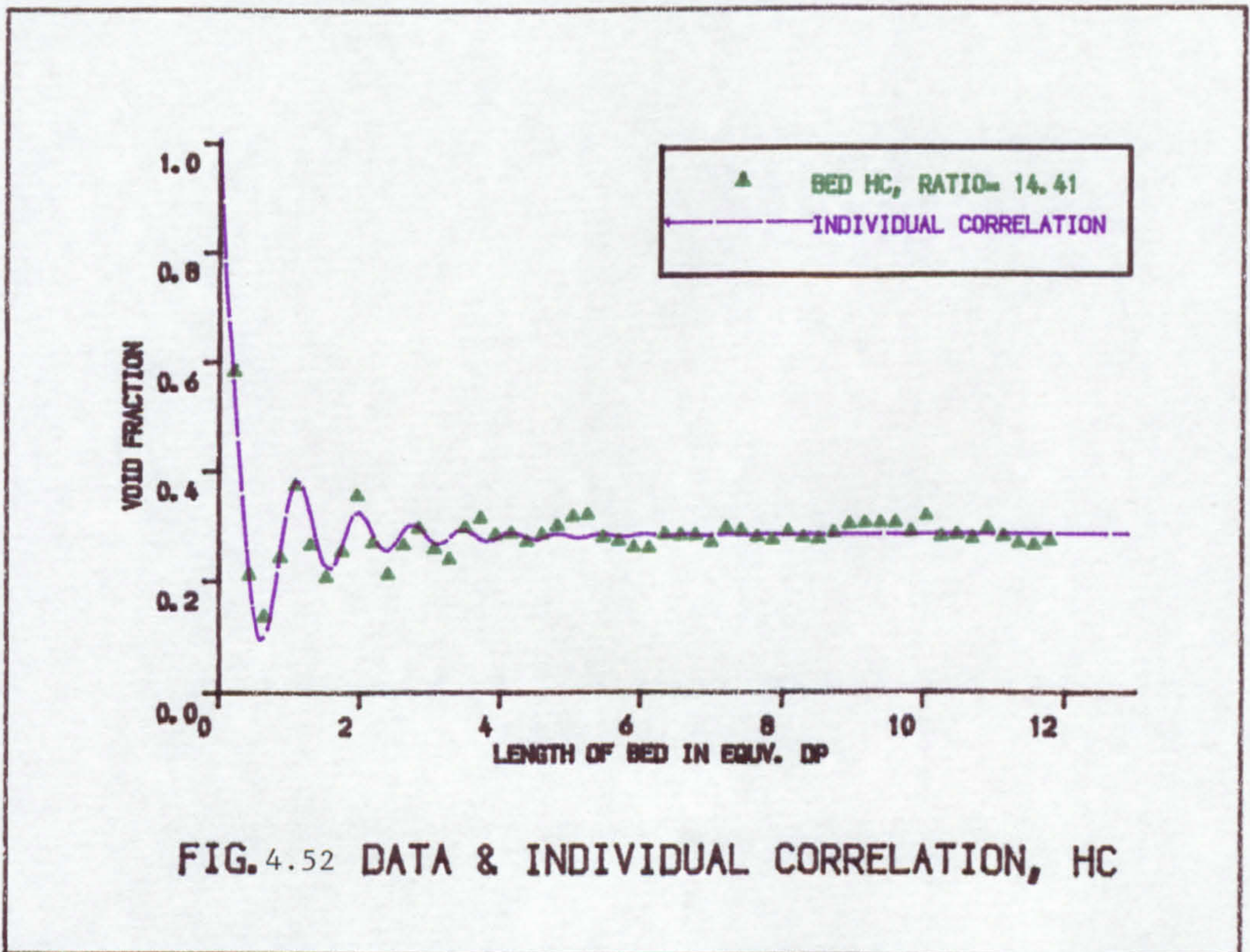
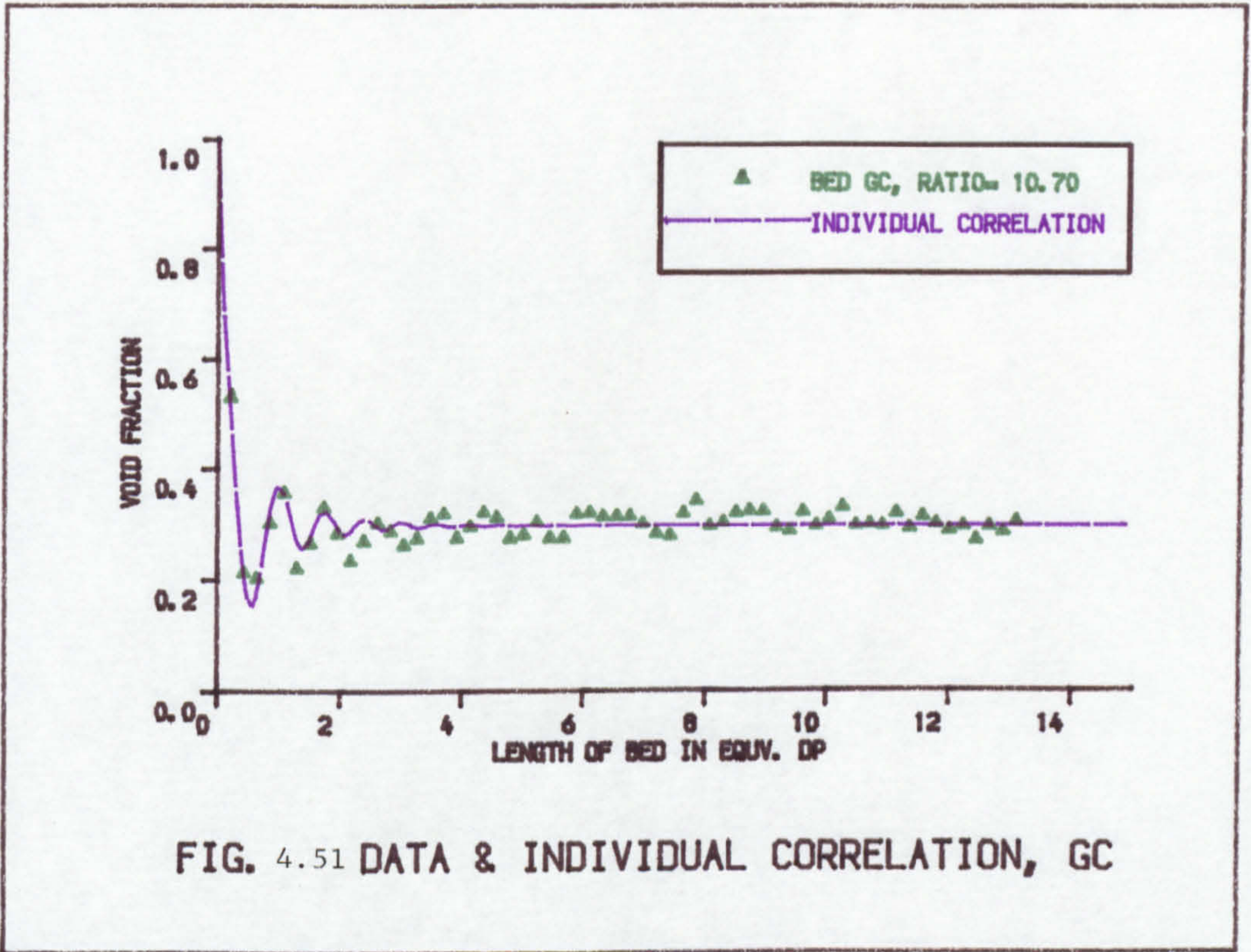


FIG. 4.50 DATA & INDIVIDUAL CORRELATION, FC



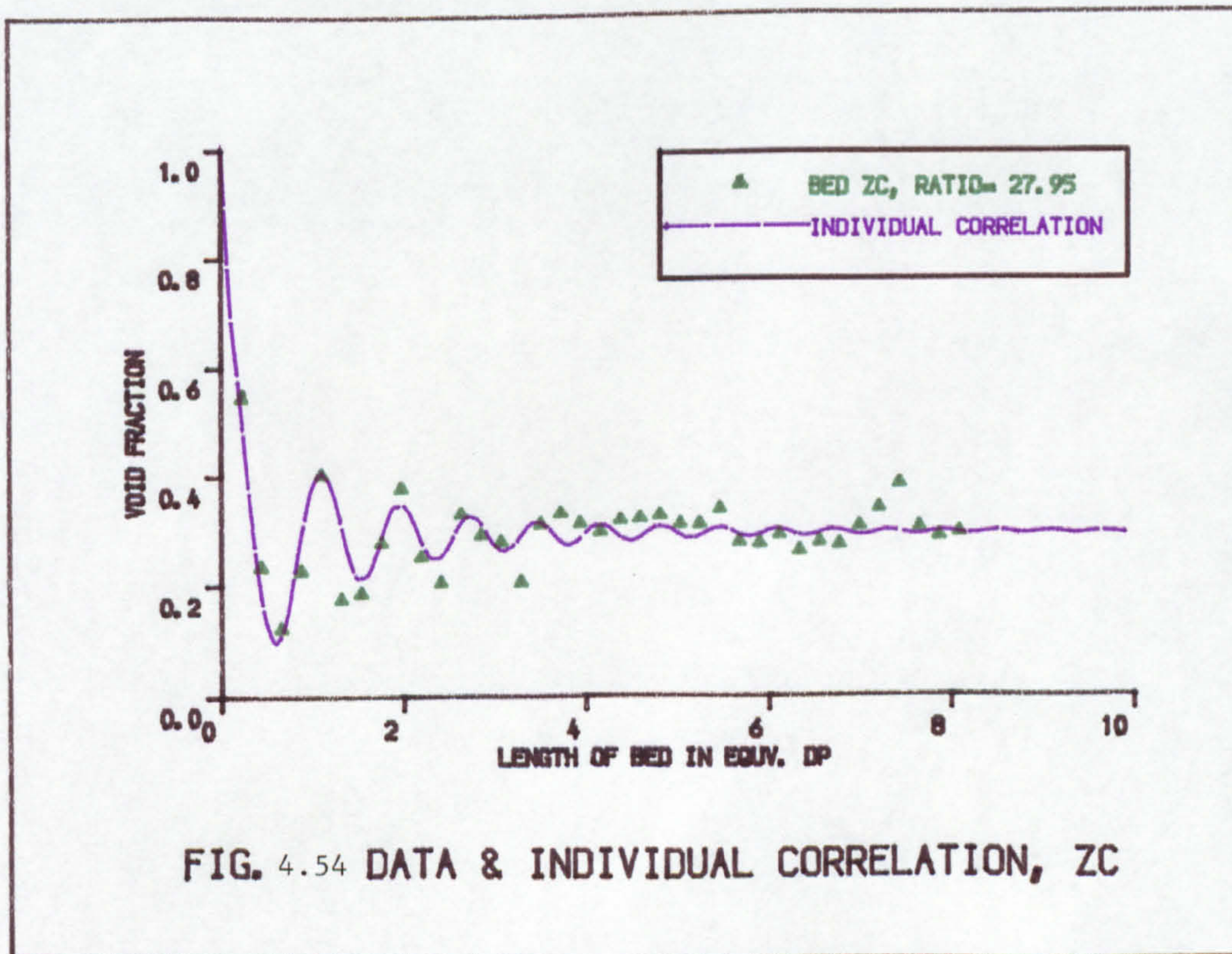
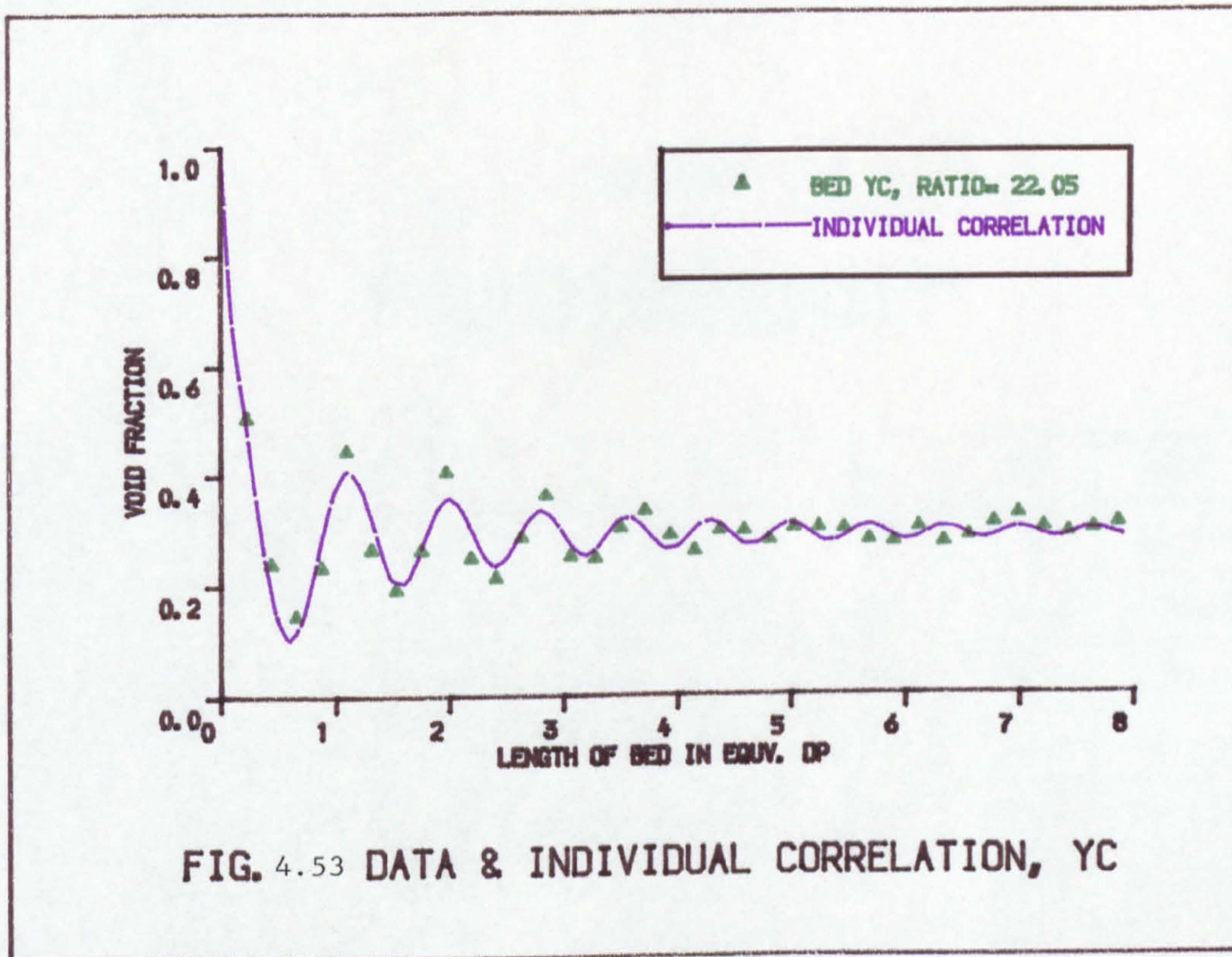


TABLE 4.4

**Estimates of the Individual Correlation
Parameters for Axial Variation**

ID	d_t/d_{pe}	β_1	β_2	β_3	β_4	β_5	Σe^2	SEE
AC	8.50	0.6702	1.7036	5.4103	0.4783	1.1881	476.4	2.7×10^{-2}
BC	8.50	0.6581	1.7294	5.8487	0.5744	1.1680	712.2	3.3×10^{-2}
CC	8.50	0.6969	2.0483	6.6503	0.4375	1.0848	602.3	2.8×10^{-2}
DC	3.37	0.6004	2.4854	6.3347	0.4141	1.1950	768.4	3.4×10^{-2}
EC	5.35	0.6635	1.8009	6.9467	0.3278	1.0144	524.9	2.9×10^{-2}
FC	7.02	0.6885	2.0823	6.4454	0.5656	1.1243	337.5	2.3×10^{-2}
GC	10.70	0.7045	2.2389	5.9210	0.6757	1.3021	253.4	2.1×10^{-2}
HC	14.41	0.7187	1.7905	5.2868	0.6670	1.2499	223.3	2.1×10^{-2}
YC	22.05	0.7114	1.6857	5.4501	0.4764	1.2018	270.7	3.0×10^{-2}
ZC	27.95	0.7104	1.6950	5.5056	0.5541	1.2225	584.6	4.3×10^{-2}

A generalized correlation was then developed based on the values of the coefficients estimated for the following expressions:

$$\beta_1 = \frac{d_r}{1.35 d_r + 0.96} \quad (4.9)$$

$$\beta_2 = \frac{d_r}{0.6 d_r - 0.625} \quad (4.10)$$

$$\beta_3 = 7.63 d_r^{-0.108} \quad (4.11)$$

$$\beta_4 = -1.2 \times 10^{-3} d_r^2 + 0.042 d_r + 0.25 \quad (4.12)$$

$$\beta_5 = 1.045 d_r^{0.05} \quad (4.13)$$

The profiles of the above expressions are shown in Appendix B. The data obtained for each bed are compared with this general expression and shown in Figures 4.55 to 4.64. Also a simplified general correlation based on a constant value is derived for the prediction of axial variation of voidage:

$$\epsilon(x) = 1.0 - 0.68 \{1.0 - \exp(-1.93 d_r^{0.52}) \cos(6.0 d_r^{1.175})\} \quad (4.14)$$

Appendix C also contains the profiles of Equation (4.14) with the experimental data obtained from each test bed.

In general individual correlations for each bed provide better predictions than the generalized ones which is due to a certain degree of approximation made in formulating the latter.

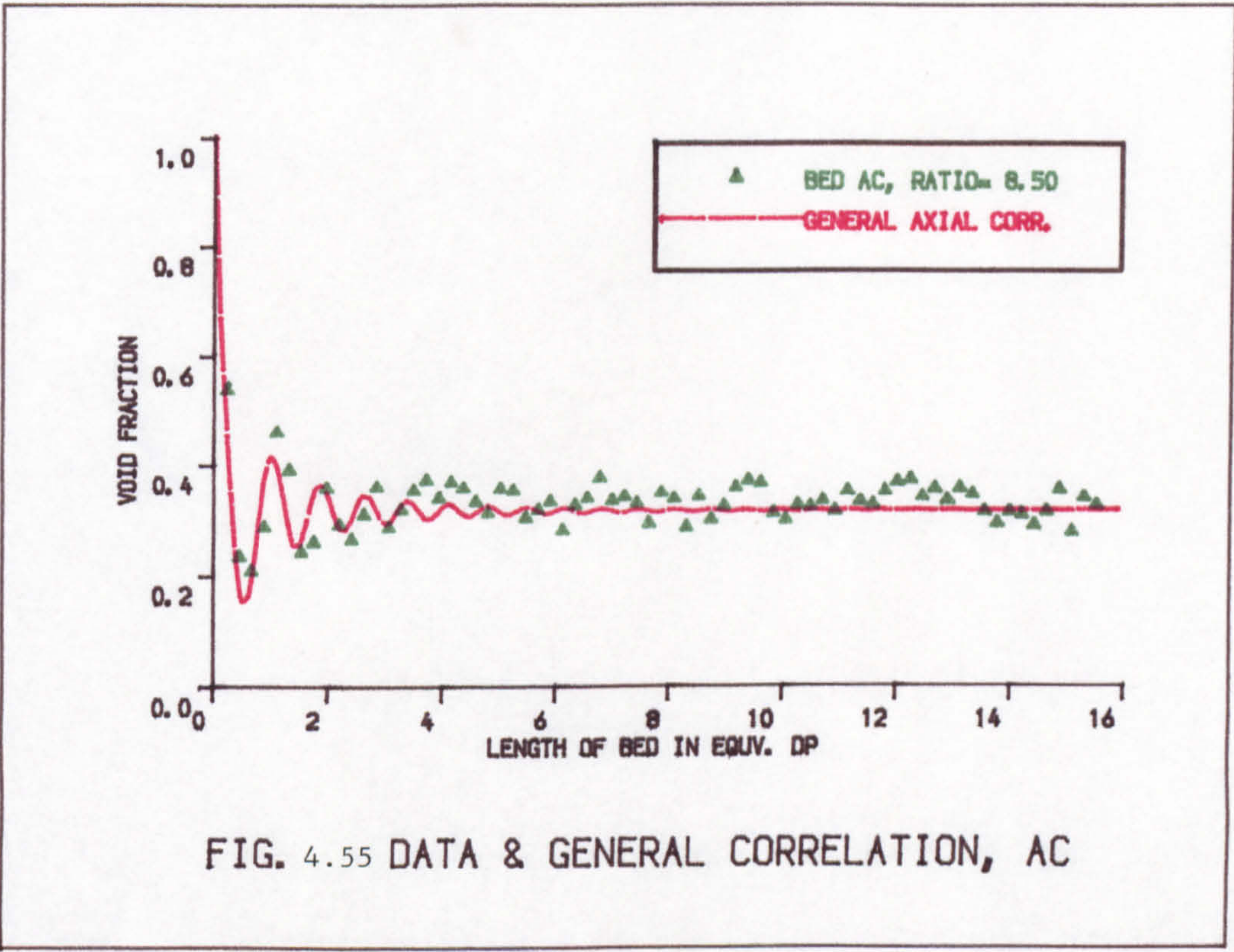


FIG. 4.55 DATA & GENERAL CORRELATION, AC

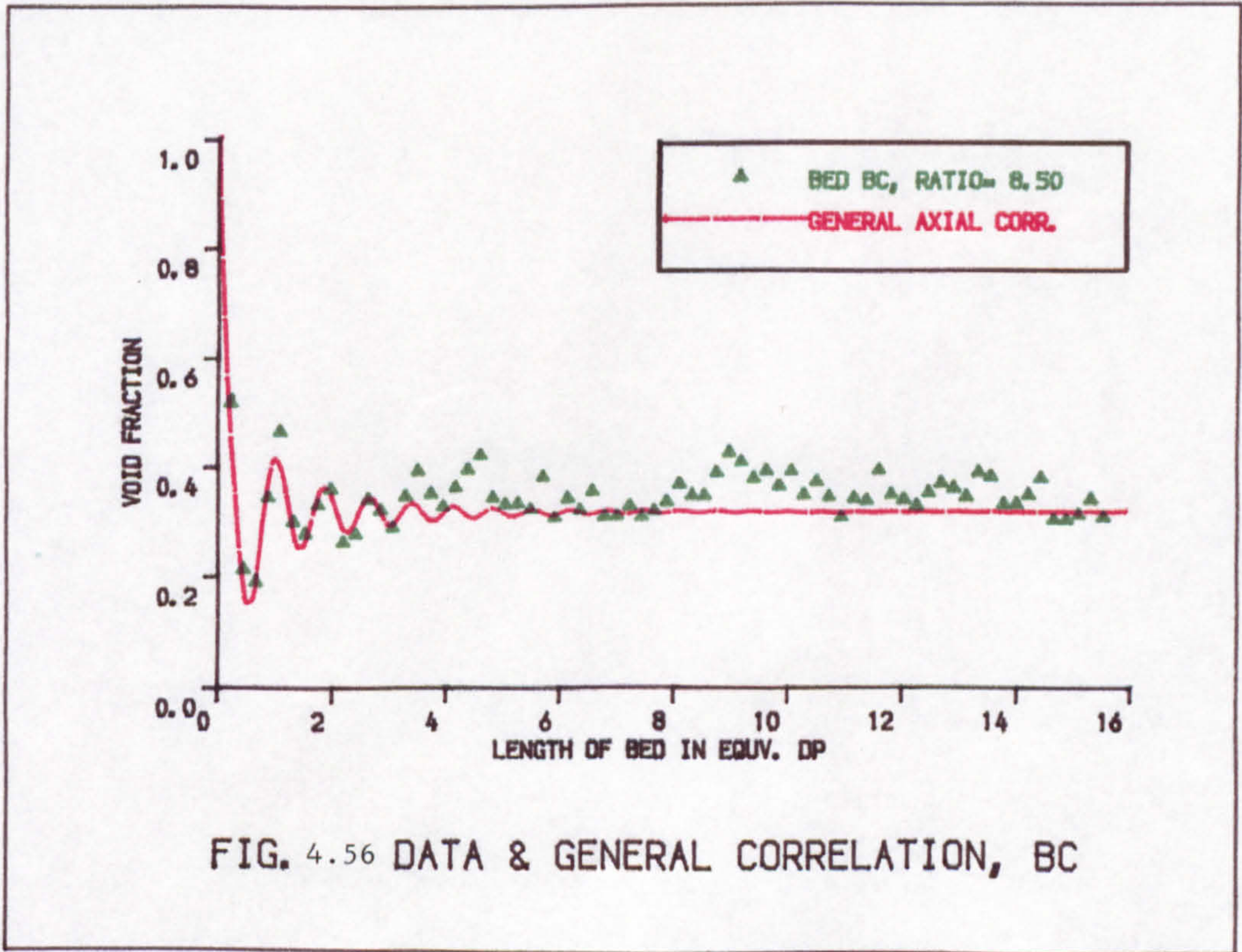
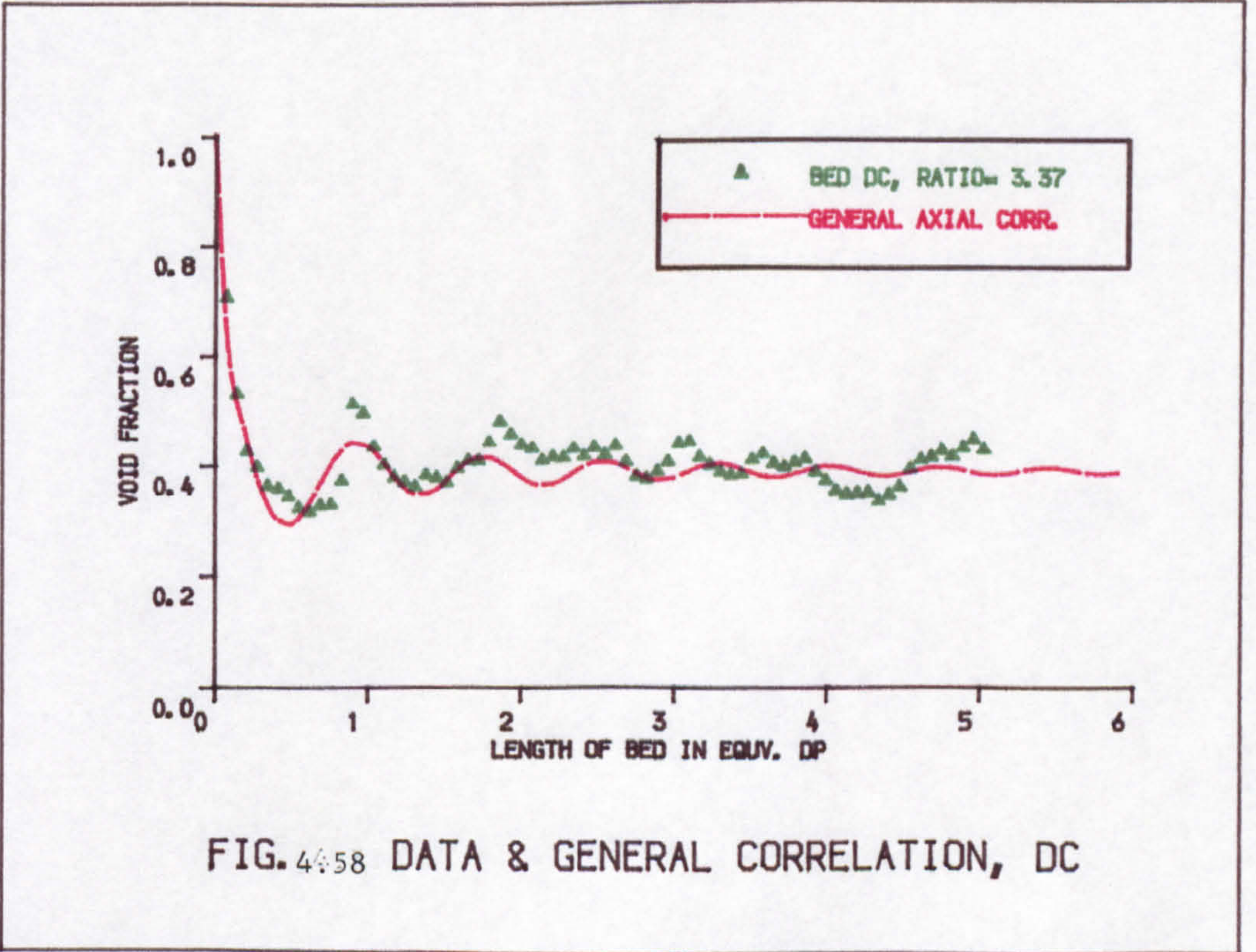
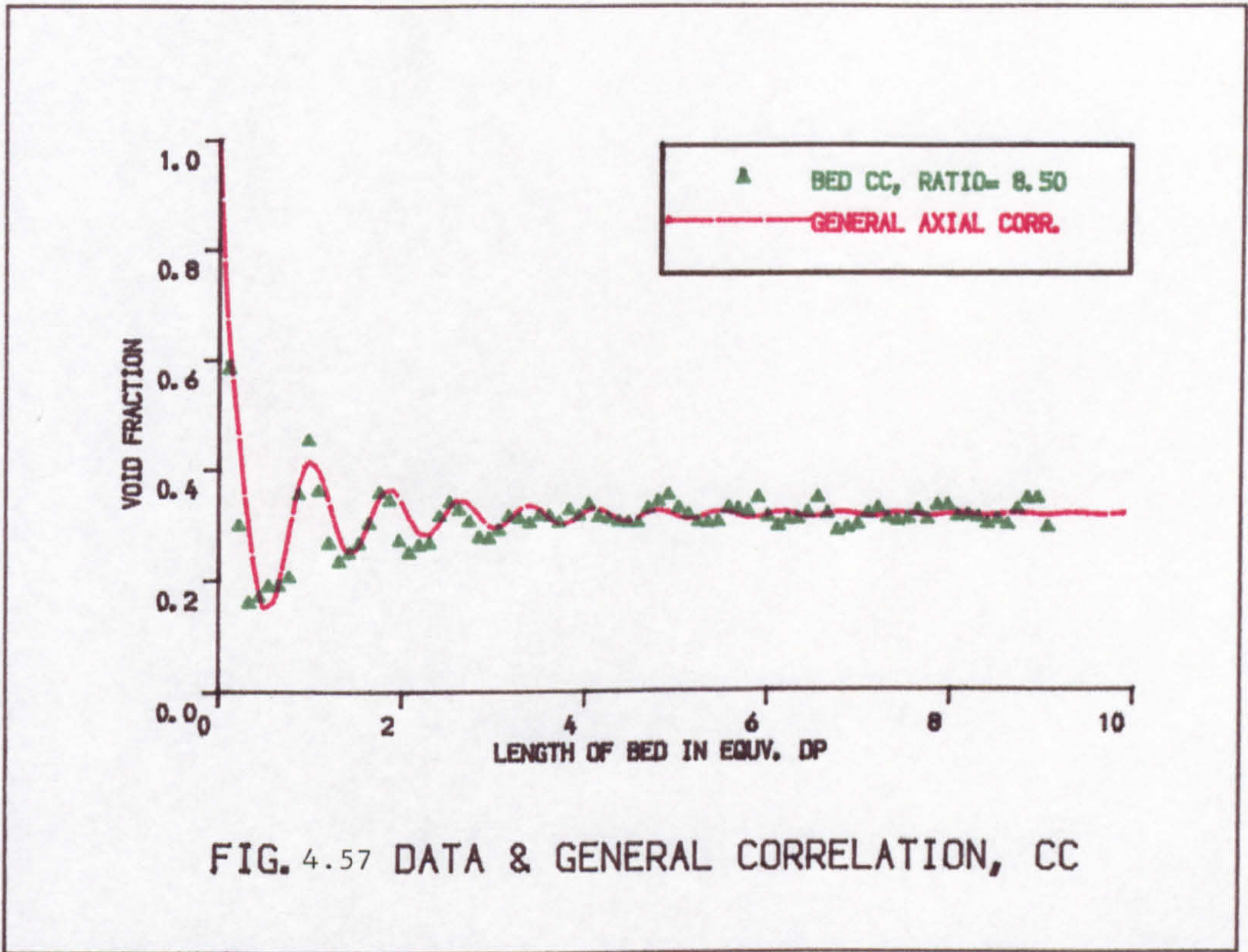
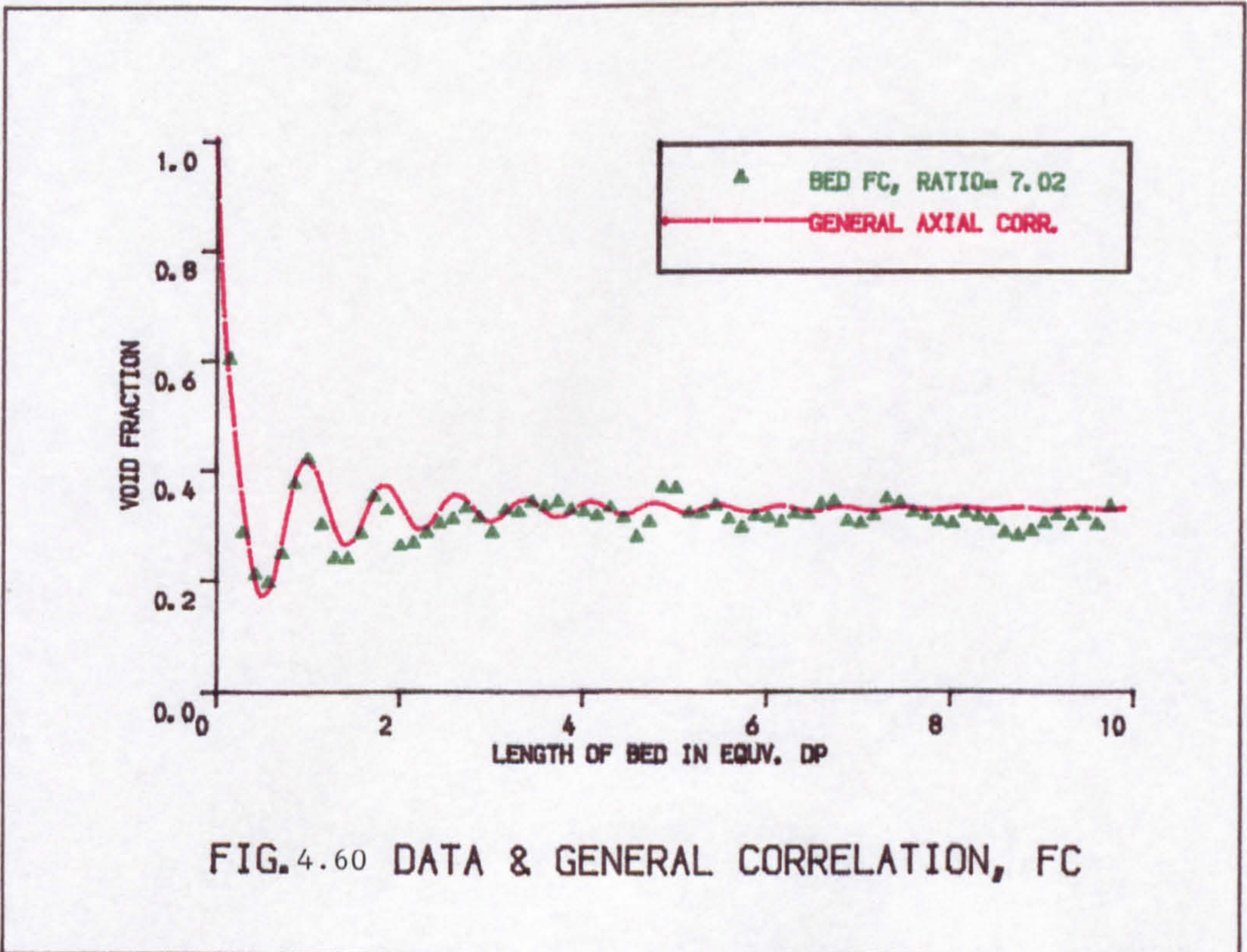
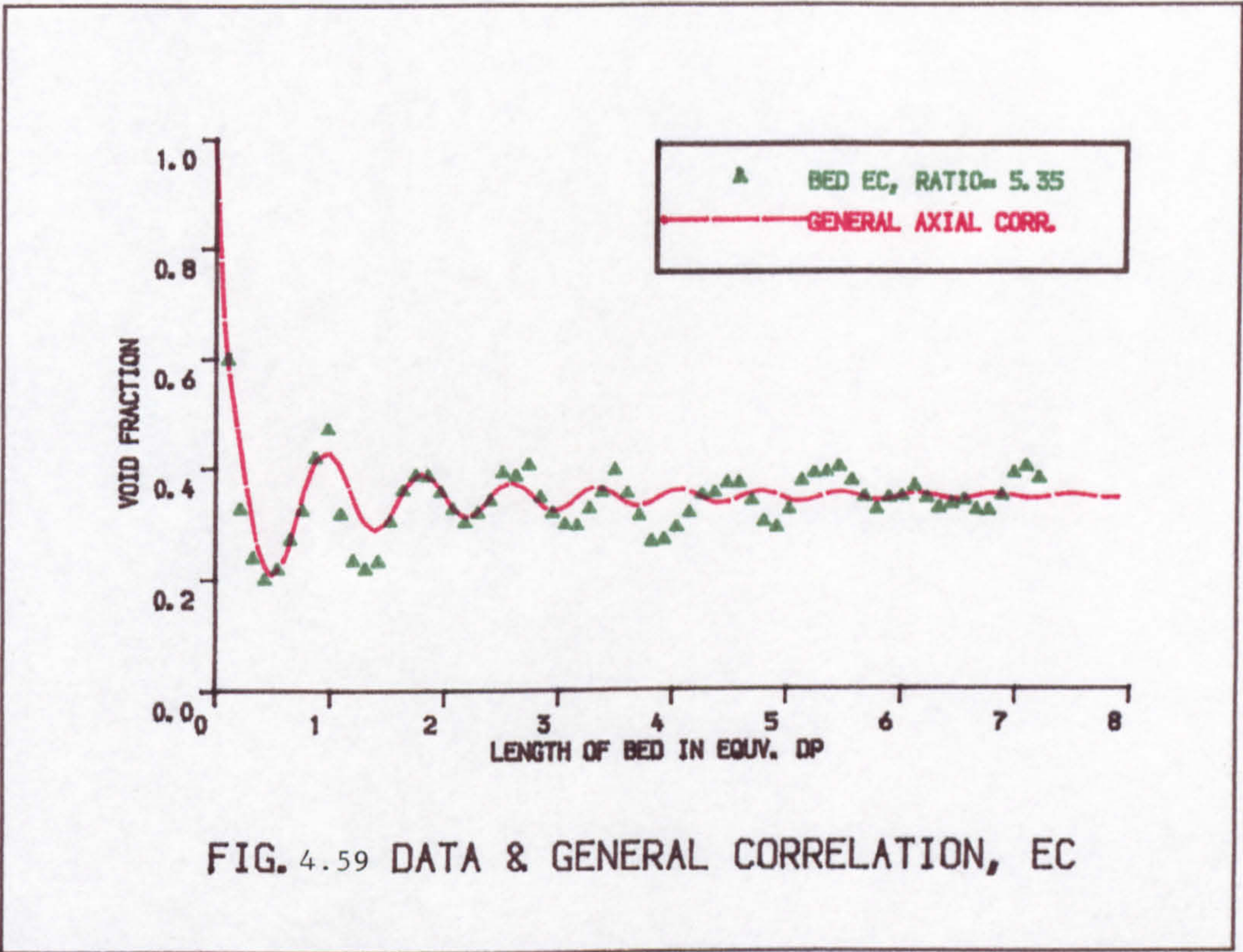


FIG. 4.56 DATA & GENERAL CORRELATION, BC





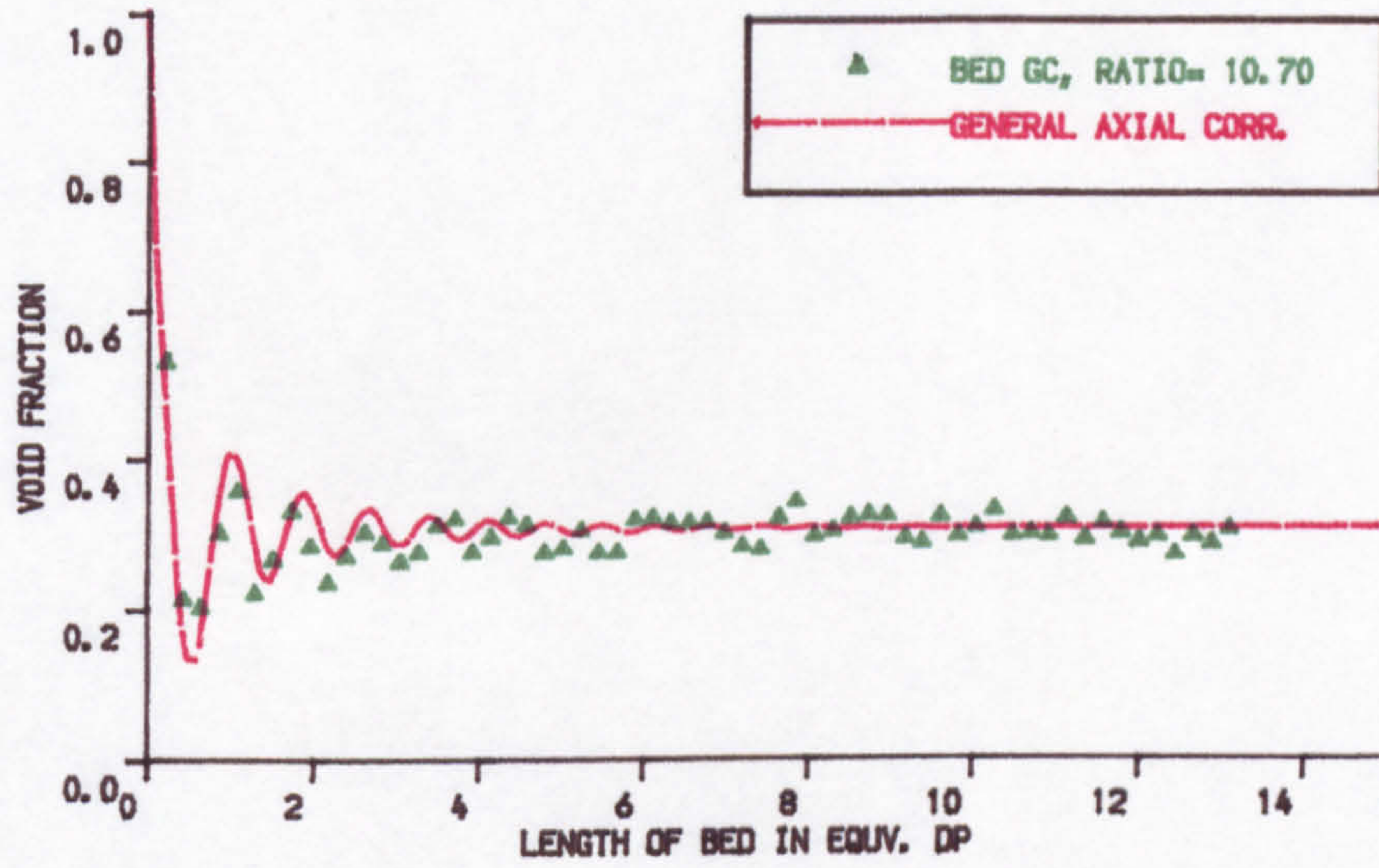


FIG. 4.61 DATA & GENERAL CORRELATION, GC

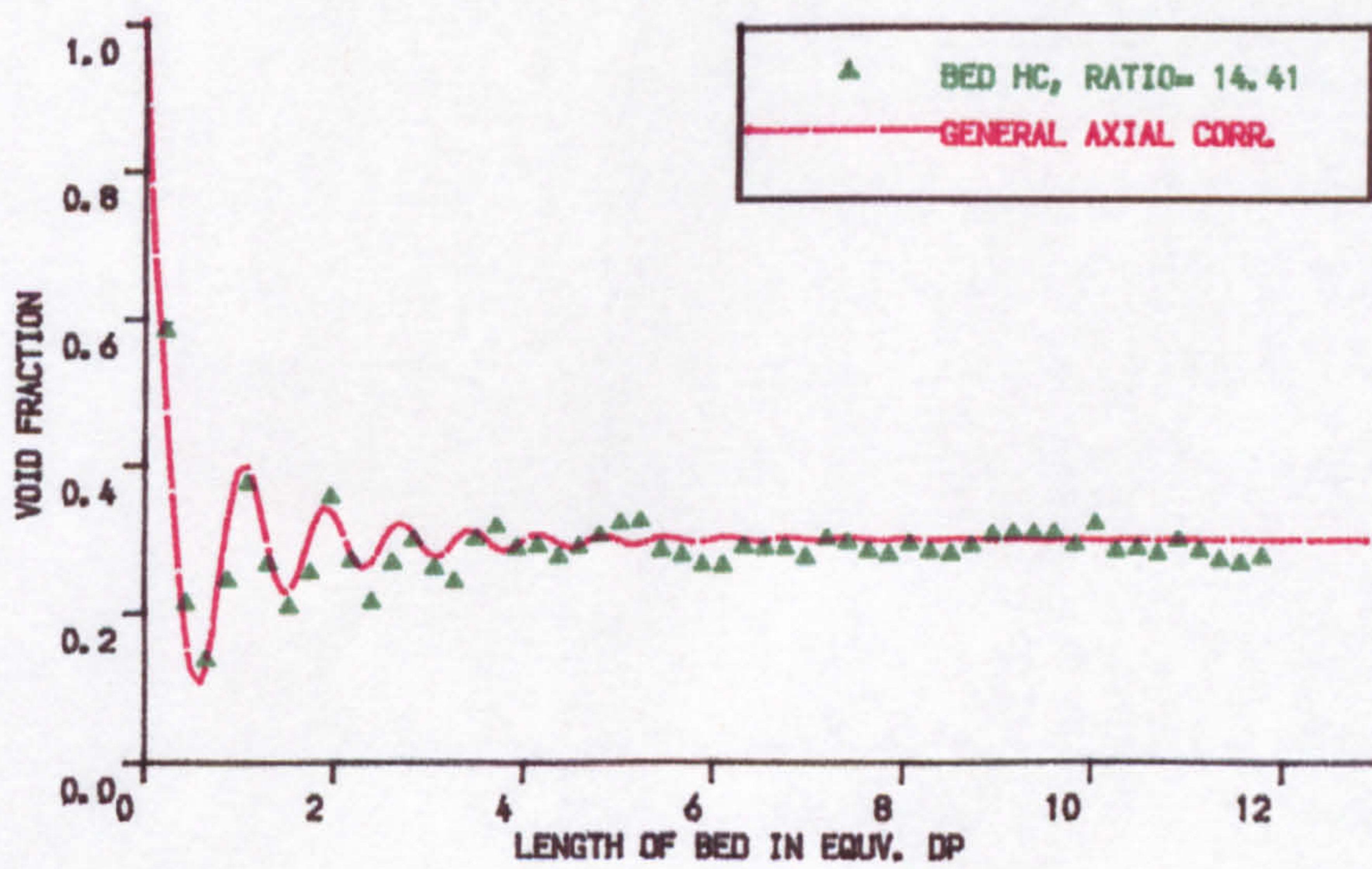


FIG. 4.62 DATA & GENERAL CORRELATION, HC

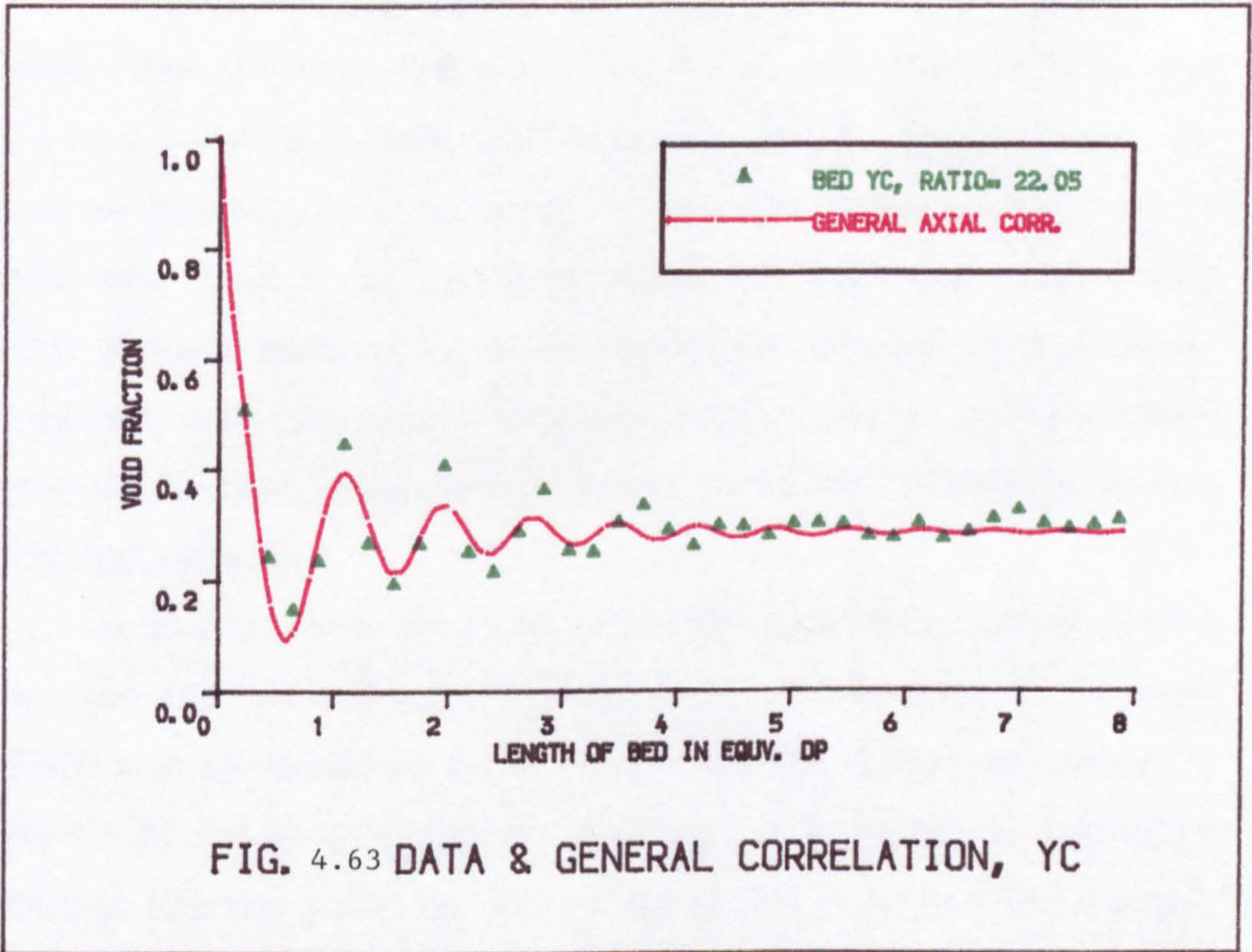


FIG. 4.63 DATA & GENERAL CORRELATION, YC

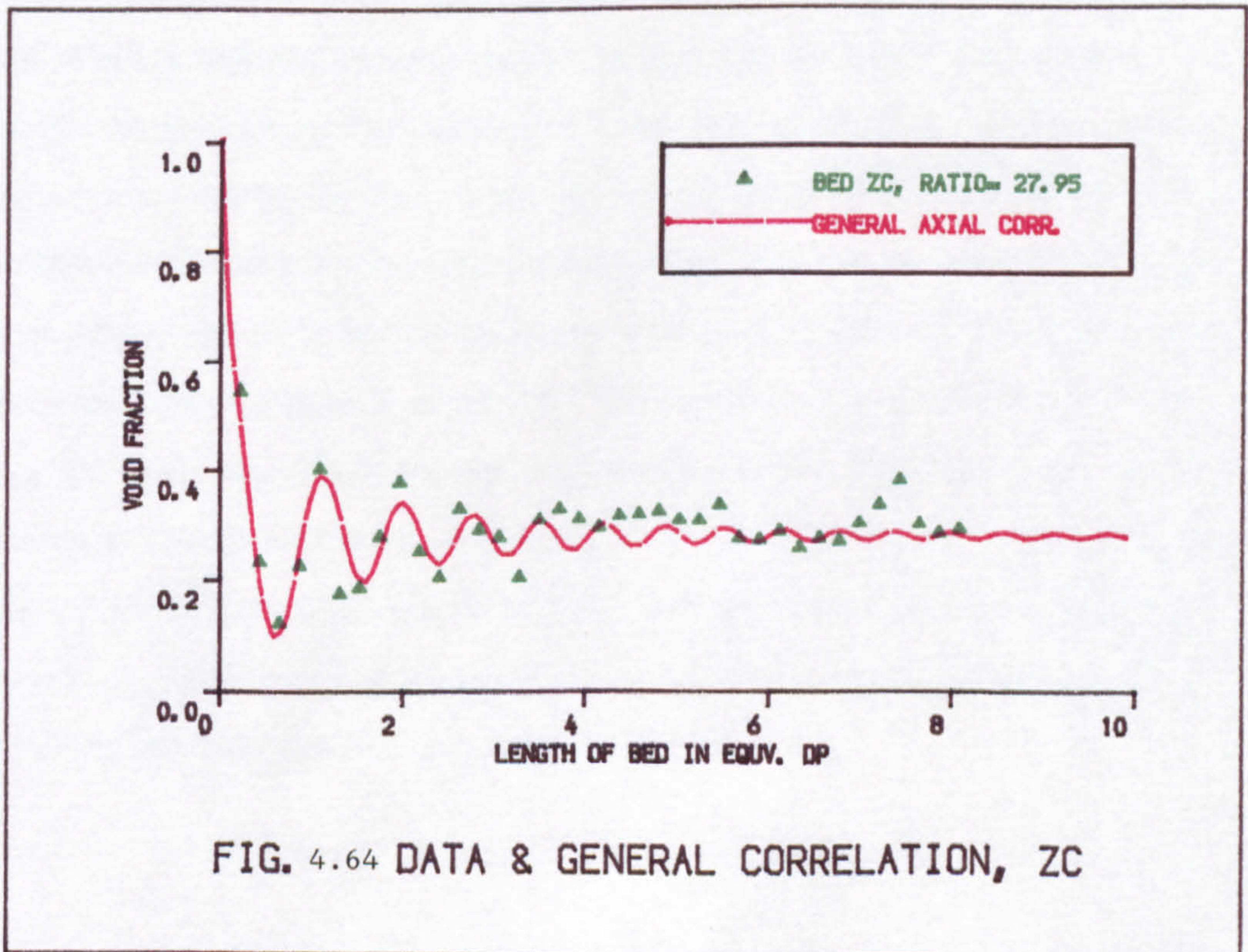


FIG. 4.64 DATA & GENERAL CORRELATION, ZC

4.5 Concluding Remarks

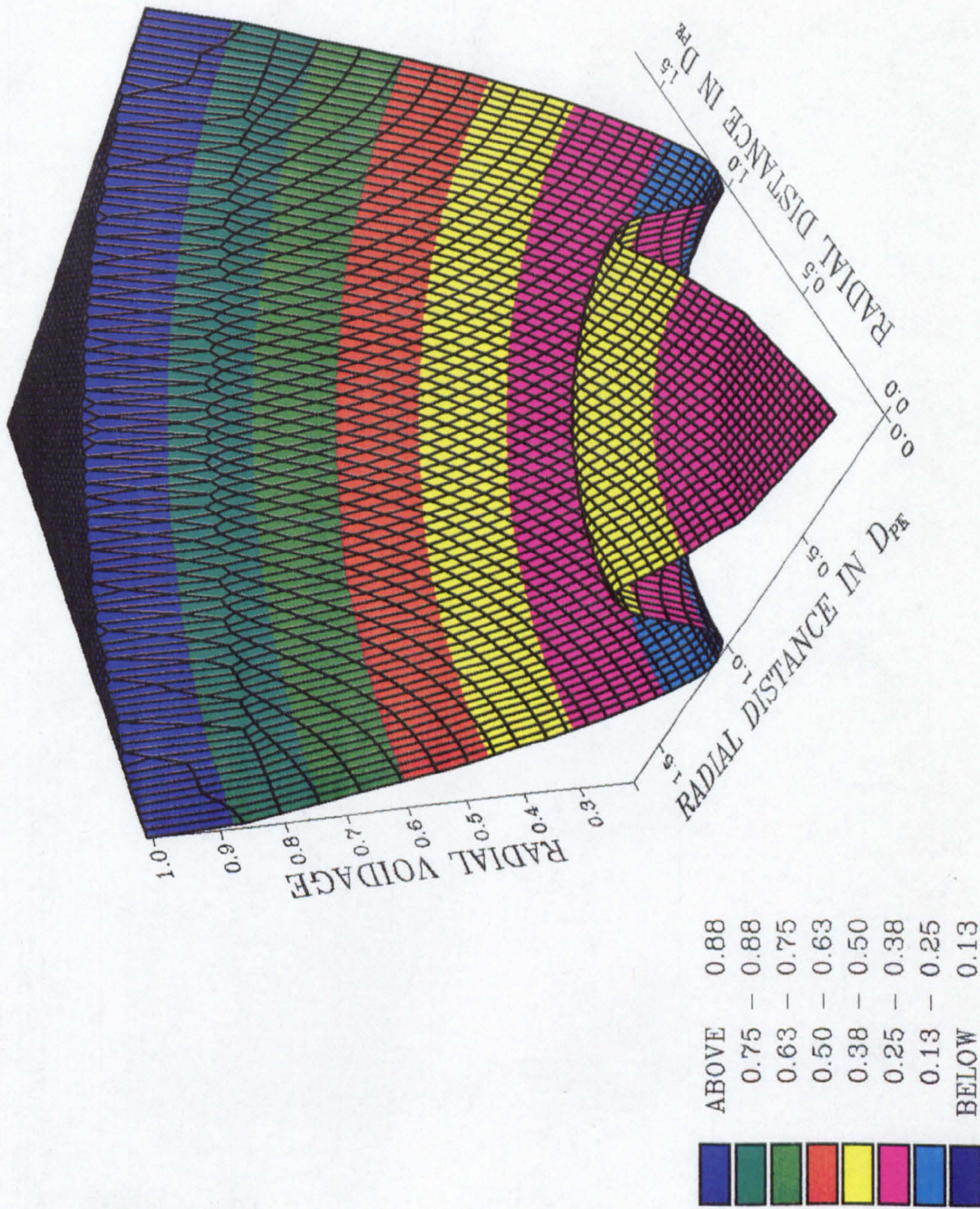
Experimental investigations into global properties of packed bed configurations of cylindrical particles indicate that the mean voidage of the geometry is only dependent upon the diameter ratio, d_t/d_{pe} , of the physical system. For relatively large beds with $d_t/d_{pe} > 10$, the mean voidage takes a constant value of about 30%. Based on the observed information, a generalized correlation is found which allows the mean voidage of beds comprised of full equilateral and different length-to-diameter cylindrical particles to be predicted reliably. Having a reliable value for the mean voidage of such beds is an essential pre-requisite for their modelling and design.

Local properties of the packed beds of full equilateral cylindrical particles were also examined with the aim of developing correlations capable of giving reliable structural information. A high resolution Image Analyser was employed to access local voidage data, $\epsilon(x,r,\theta)$. The variation of voidage in the angular direction, $\epsilon(\theta)$, was ignored for reasons discussed earlier and hence was averaged. The voidage profiles in the axial direction, $\epsilon(x)$, demonstrated the existence of an end effect pronounced up to 4 equivalent particle diameters away from the end after which the voidage became constant at about 30%. The profile is of oscillatory nature when compared with smooth exponential curve for that of spheres. The variation of the void fraction in the radial direction, $\epsilon(r)$, showed the presence of the wall effect, also in a damped oscillatory pattern which flattens in the core zone corresponding to a value of about 28%. The first minima and maxima occurs at $\frac{1}{2} d_{pe}$ and $1 d_{pe}$ away from the wall respectively with the wall zone extended to 3 equivalent particle diameters. It is concluded that the data obtained from these beds are reproducible, but there is a difference in the values of voidage when the beds are scaled up. Therefore more work needs to be done on the effect of scaling up/down, by examining beds with various scaling factors.

A suitable mathematical expression was thence chosen to characterize the pattern of the variation of voidage in axial and radial directions, as they both show the same features in their profiles. Generalized correlations derived from the individual ones were then developed for the prediction of $\epsilon(x)$ and $\epsilon(r)$ in packed beds of cylindrical particles of any d_t/d_{pe} ratio. In general all the established correlations have been found to be reliable and can provide accurate prediction of the structure of packed beds. In order to have a clearer picture of the local variations of voidage, the radial variations of all the test beds examined are presented in 3-dimensional form, Figures 4.65 to 4.74.

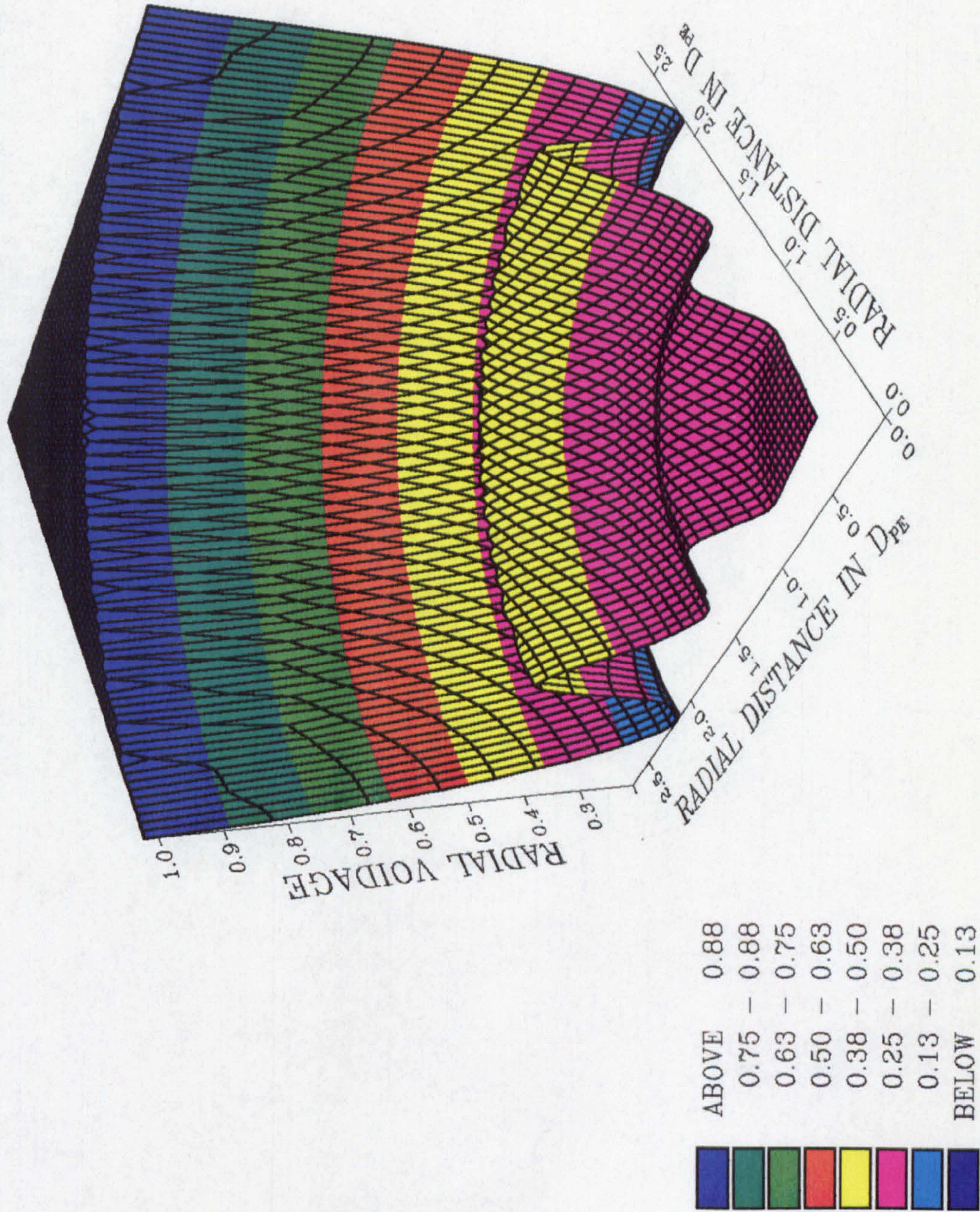
BED CHARACTERISTICS
 $D_T / D_{PE} = 3.37$

FIGURE 4.65



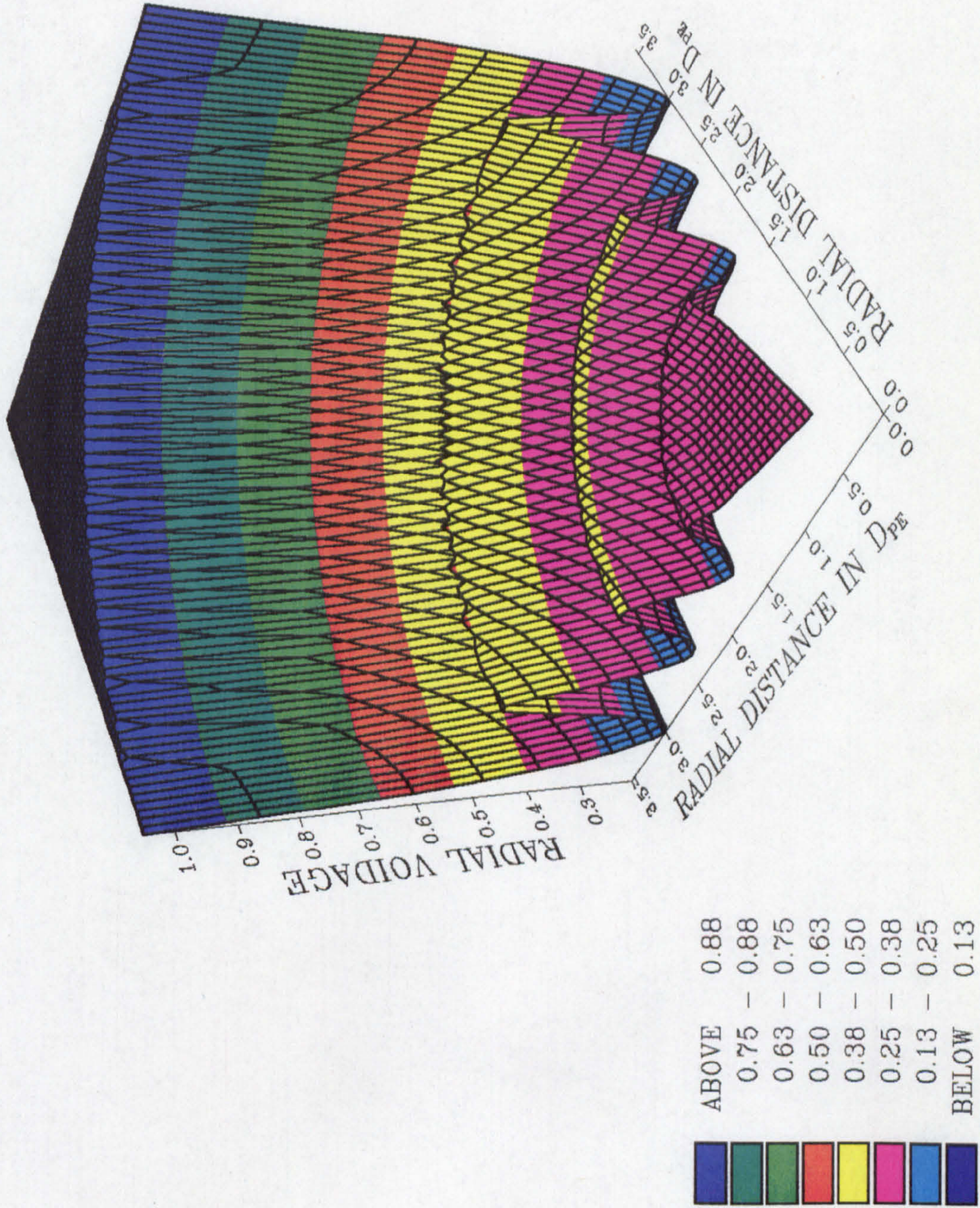
BED CHARACTERISTICS
 $D_T/D_{PE} = 5.35$

FIGURE 4.66



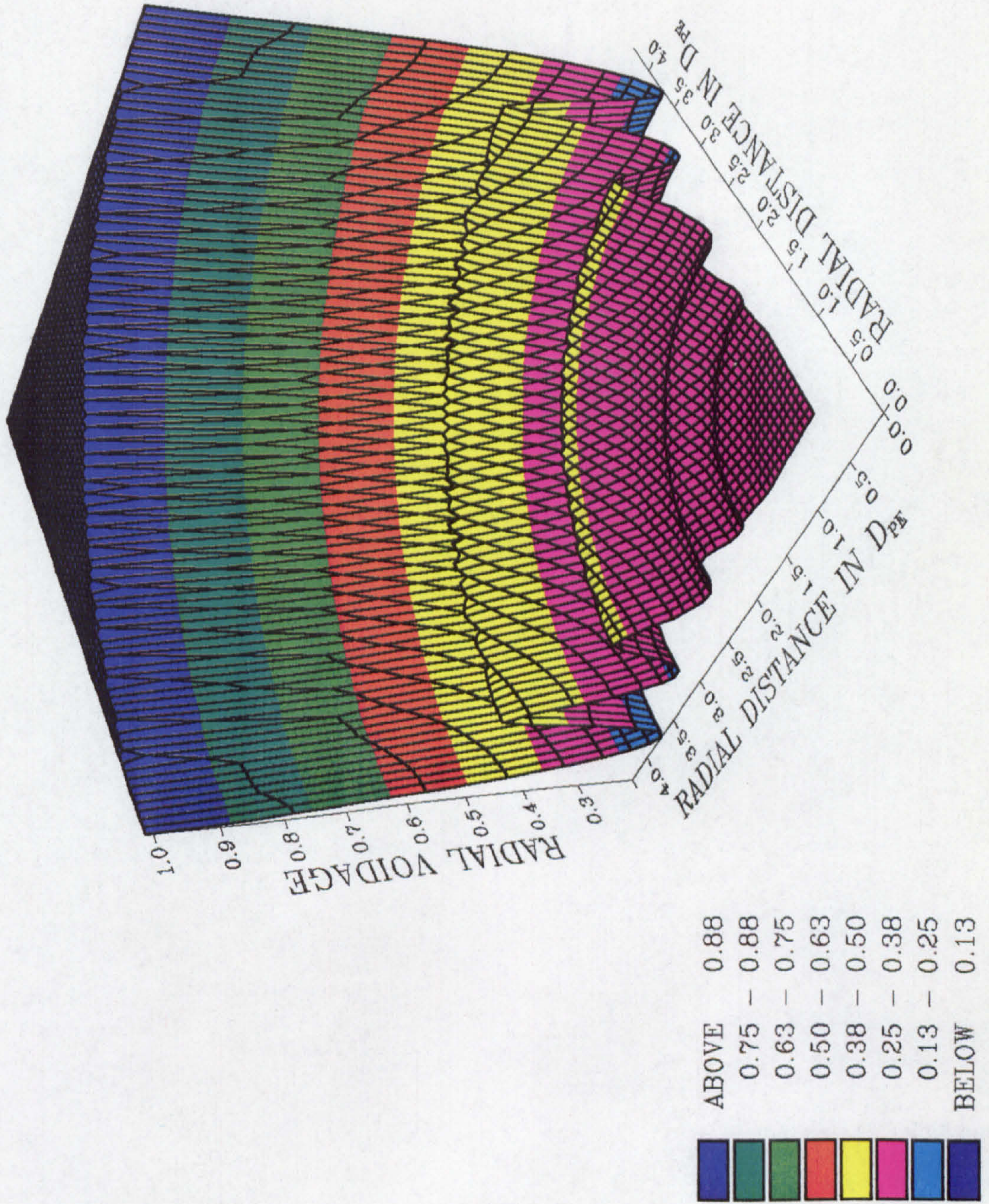
BED CHARACTERISTICS
 $D_T/D_{PE} = 7.02$

FIGURE 4.67



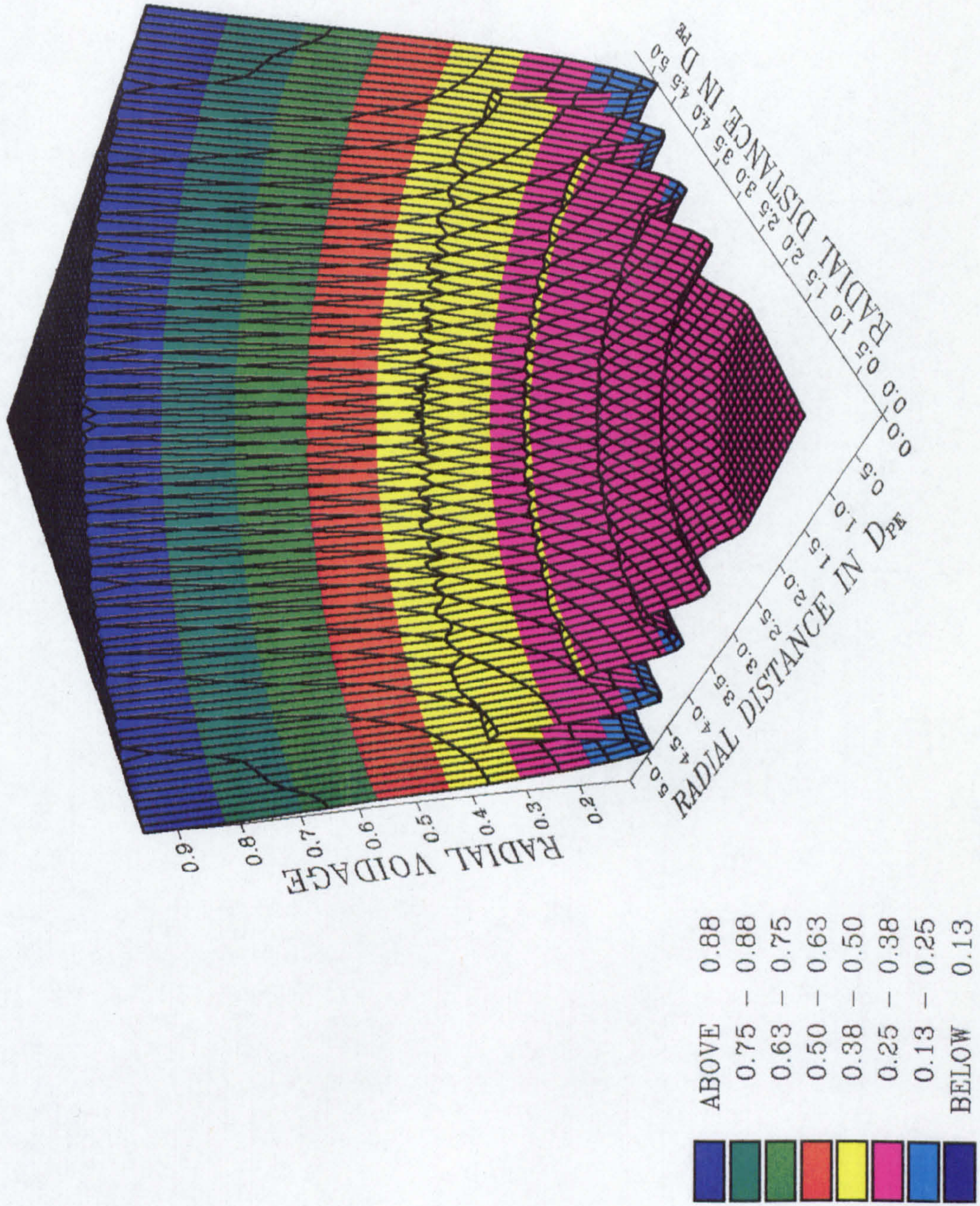
BED CHARACTERISTICS
 $D_T / D_{PE} = 8.50$

FIGURE 4.68



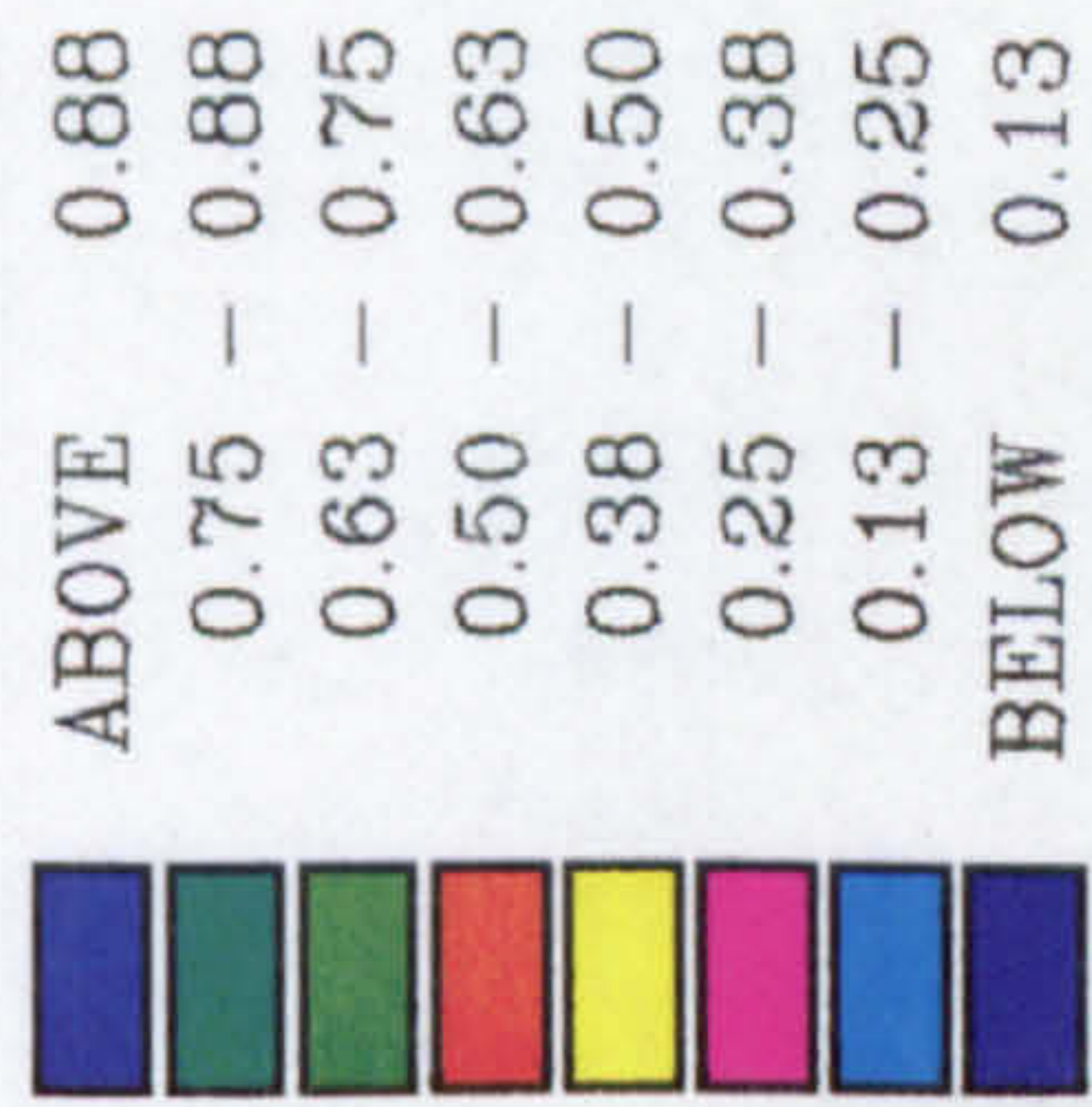
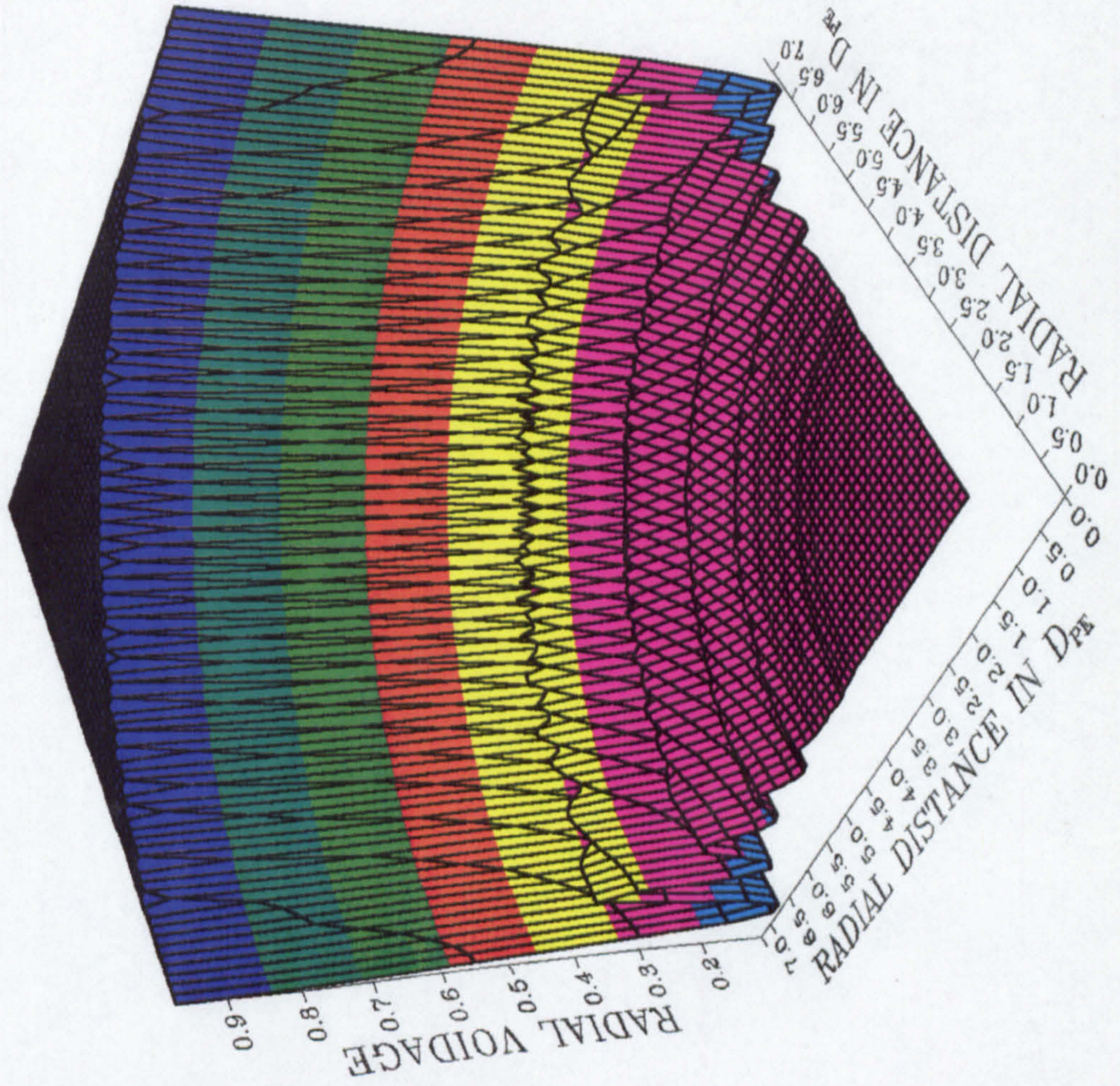
BED CHARACTERISTICS
 $D_T / D_{PE} = 10.7$

FIGURE 4.69



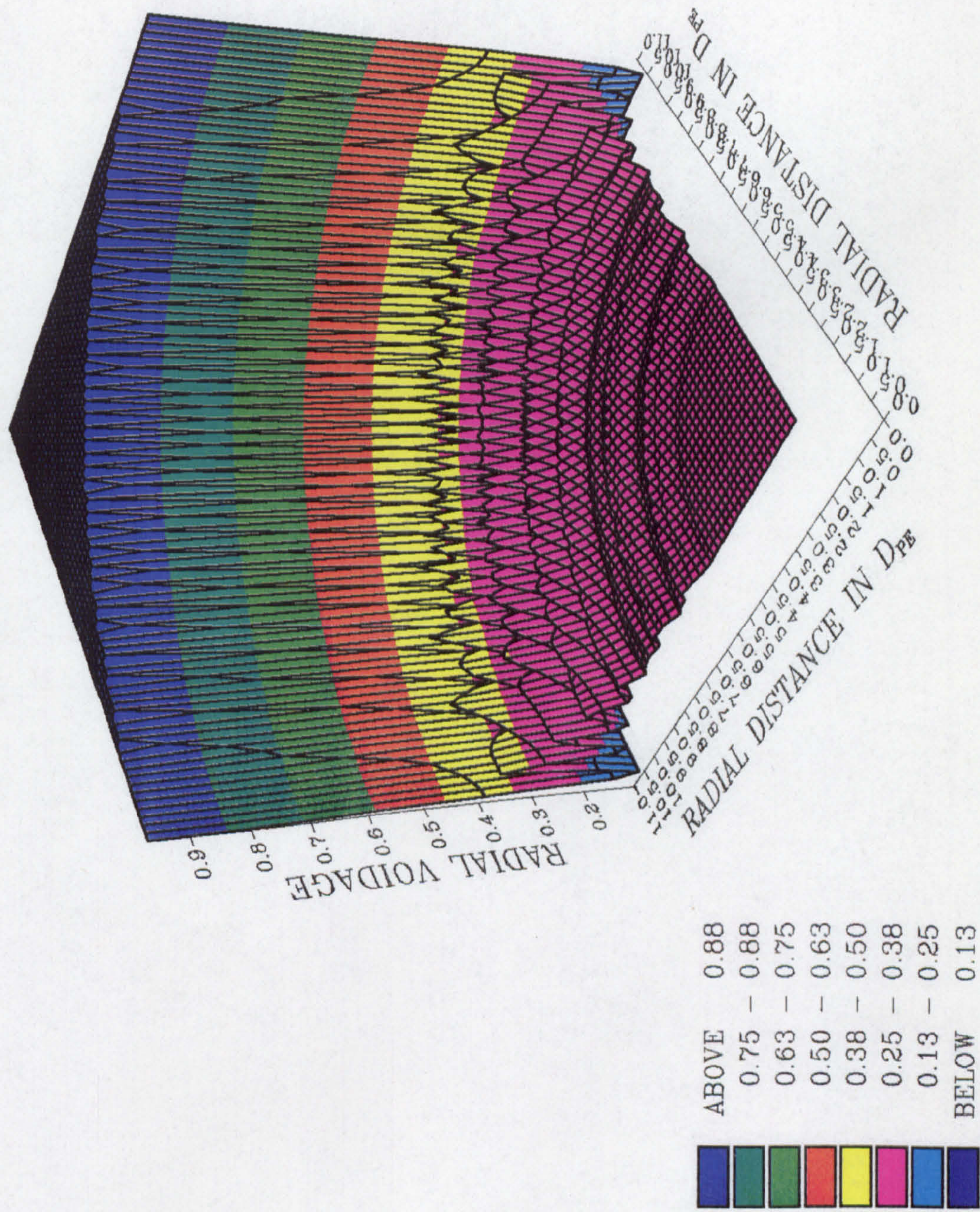
BED CHARACTERISTICS
 $D_T/D_{PE} = 14.41$

FIGURE 4.70



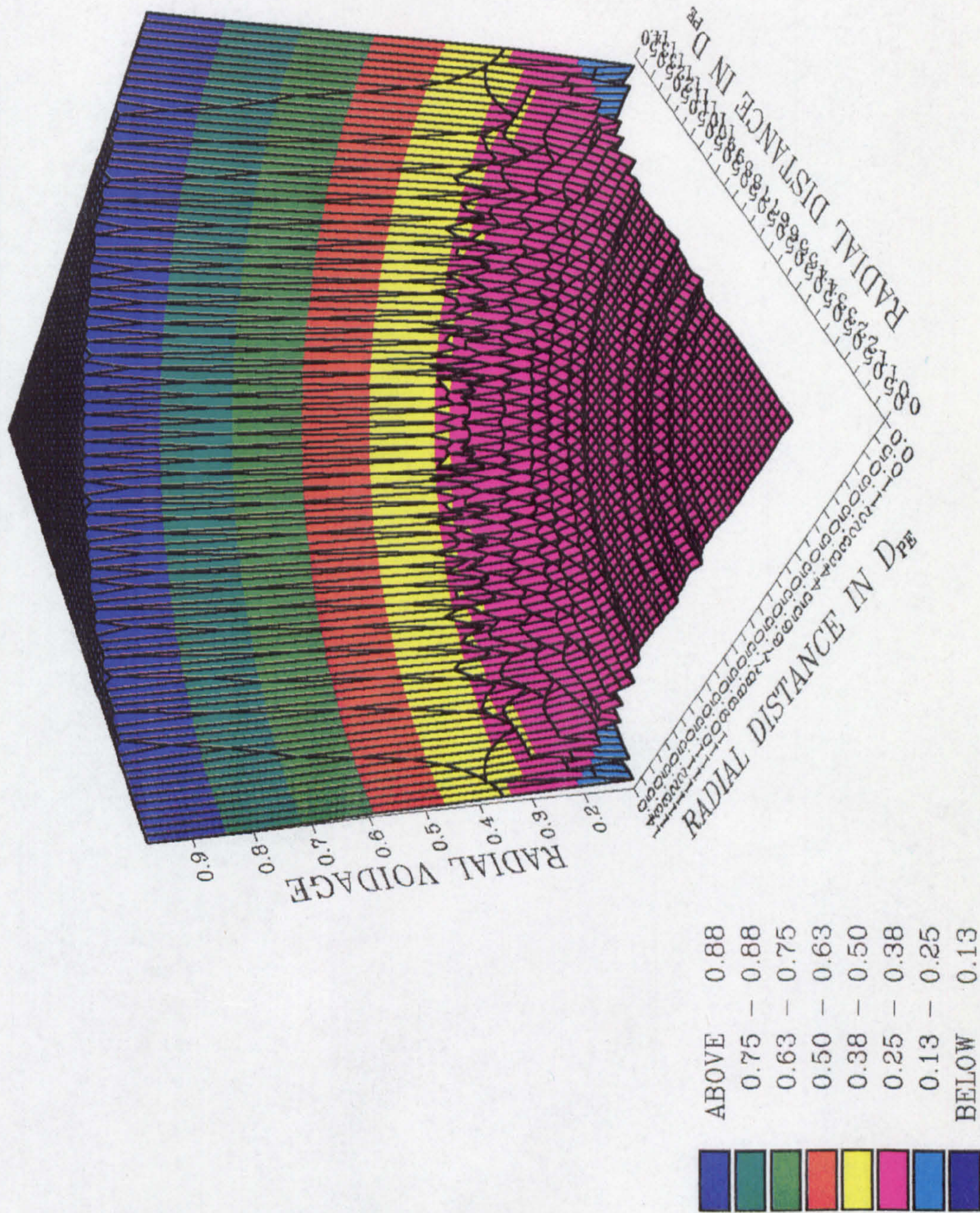
BED CHARACTERISTICS
 $D_T / D_{PE} = 22.05$

FIGURE 4.71



BED CHARACTERISTICS
 $D_T / D_{PE} = 27.95$

FIGURE 4.72



CHAPTER 5

**COMPARISON OF PRESENT AND
PUBLISHED CORRELATIONS**

CHAPTER 5

COMPARISON OF PRESENT AND PUBLISHED CORRELATIONS

5.1 Introduction

In order to validate the accuracy of the obtained data for both the global and local structural properties of packed beds of cylindrical particles, the correlations derived from these data as well as the data itself are compared with the ones available in the literature. Although the literature is very poor in this respect, the trend of the few correlations available in various directions are examined. This chapter shows the similarities and argues the discrepancies between the present and published data and expressions.

5.2 Mean Voidage Correlation

The experimental data obtained by Roblee, et al., (1958); showed that the mean voidage of packed beds of full cylindrical particles was about 0.25. More recently McGreavy, et al., (1986); and Moallemi, (1989); demonstrated a functional relationship between the mean voidage, ϵ_{mean} , in particulate beds and the associated dimensional parameters represented by the tube to particle diameter ratio. There have been attempts to develop correlations for the mean voidage by Carman, (1938); Haughey and Beveridge, (1966); but these do not take into account the importance of dimensional parameters. The only expressions that do include the diameter ratio, are the ones proposed by Dixon, (1988) and Foumeny and Roshani, (1990). The two most common methods of measuring bulk mean voidage are water displacement, and weighing a known packing volume. The former method has been used for the present study, whereas the latter method is employed by Dixon, which is extremely tedious for large volumes of packing. However, for his study at low

diameter ratios, $d_t/d_p < 10$, the volumes of packings were not large and the method was feasible. The correlation derived from the present data is therefore compared with Dixon's correlation for full equilateral cylinders. Figure 5.1 shows that the general trend between the two correlations appear to be similar, but nevertheless a noticeable difference exists. This could be due to a number of factors such as size variation, method of packing, and experimental error. In general, all the published correlations for the prediction of mean voidage in particulate beds tend to have limited applicability, particularly in terms of the range of diameter ratios they cover. However, the correlation proposed here is applicable to a wide range of diameter ratios being employed in practice and also can be used for cylindrical particles with different length-to-diameter ratios. The proposed correlation is therefore the most reliable and comprehensive of its kind, since it is based on accurate data acquisition and powerful computational optimisation.

To compare the mean voidage values of packed beds of spheres with equilateral cylinders, the correlation derived by Moallemi, (1989), for the prediction of ϵ_{mean} in beds of unisized spheres was chosen. Figure 5.2 illustrates the difference between the two profiles which have the same overall features. The mean voidage decreases exponentially with increasing diameter ratio in both cases and then becomes constant at about 40% and 30% for spheres and cylinders, respectively. This indicates that beds comprised of cylindrical particles produce a more compact geometry and this is because of different orientations possible with these particles. It is interesting to note that the criterion for constant mean voidage appears to be the same for spheres and cylinders at a diameter ratio of 10.

The expression proposed by Aerov (1951) for the prediction of mean voidage in packed beds of spheres is also compared with the present proposed correlation for equilateral cylinders. Figure 5.3 shows the trend of the two profiles, which resemble having the same general pattern but differ in indicating lower values of mean voidage for large diameter ratios of beds of cylinders. For large beds of

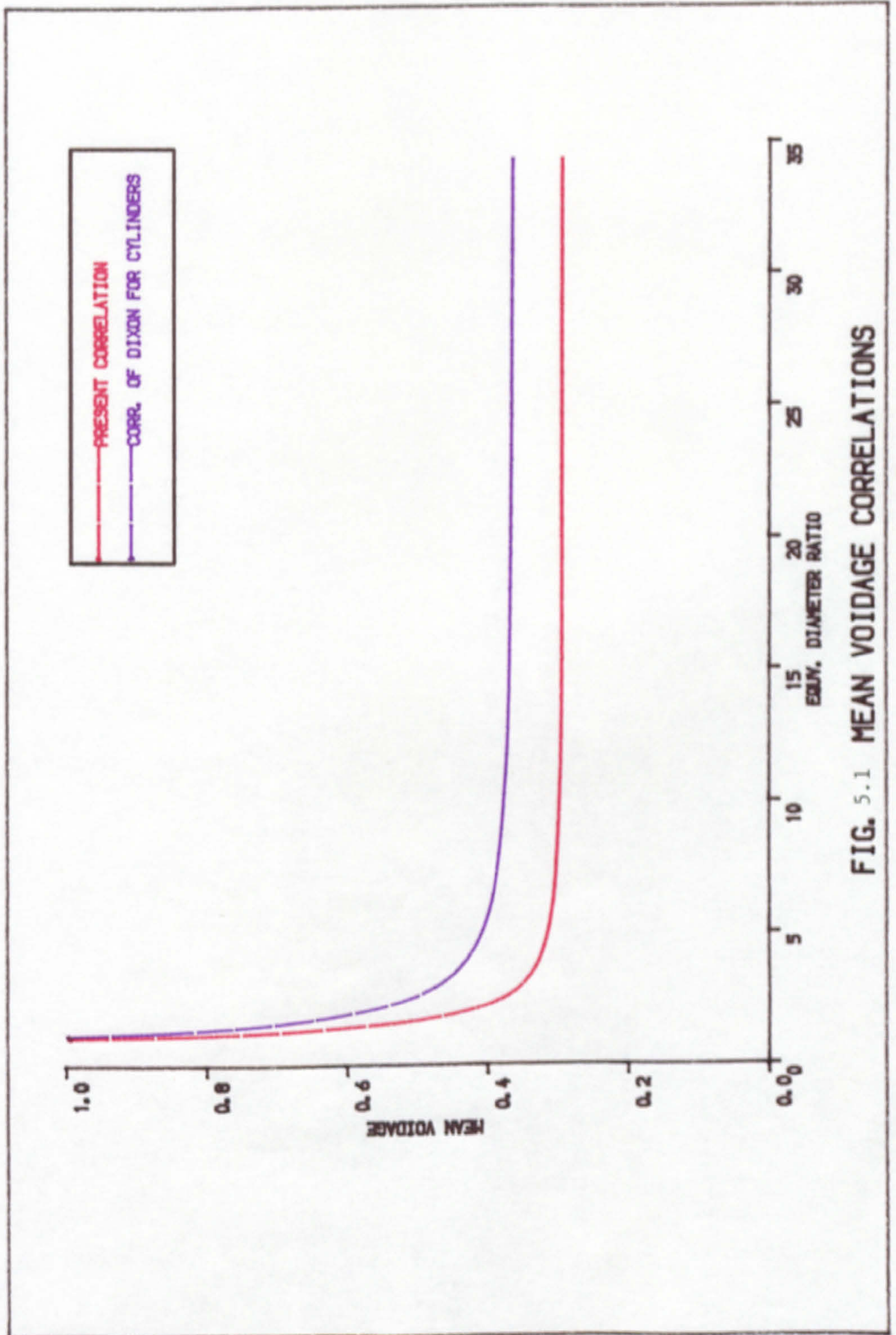


FIG. 5.1 MEAN VOIDAGE CORRELATIONS

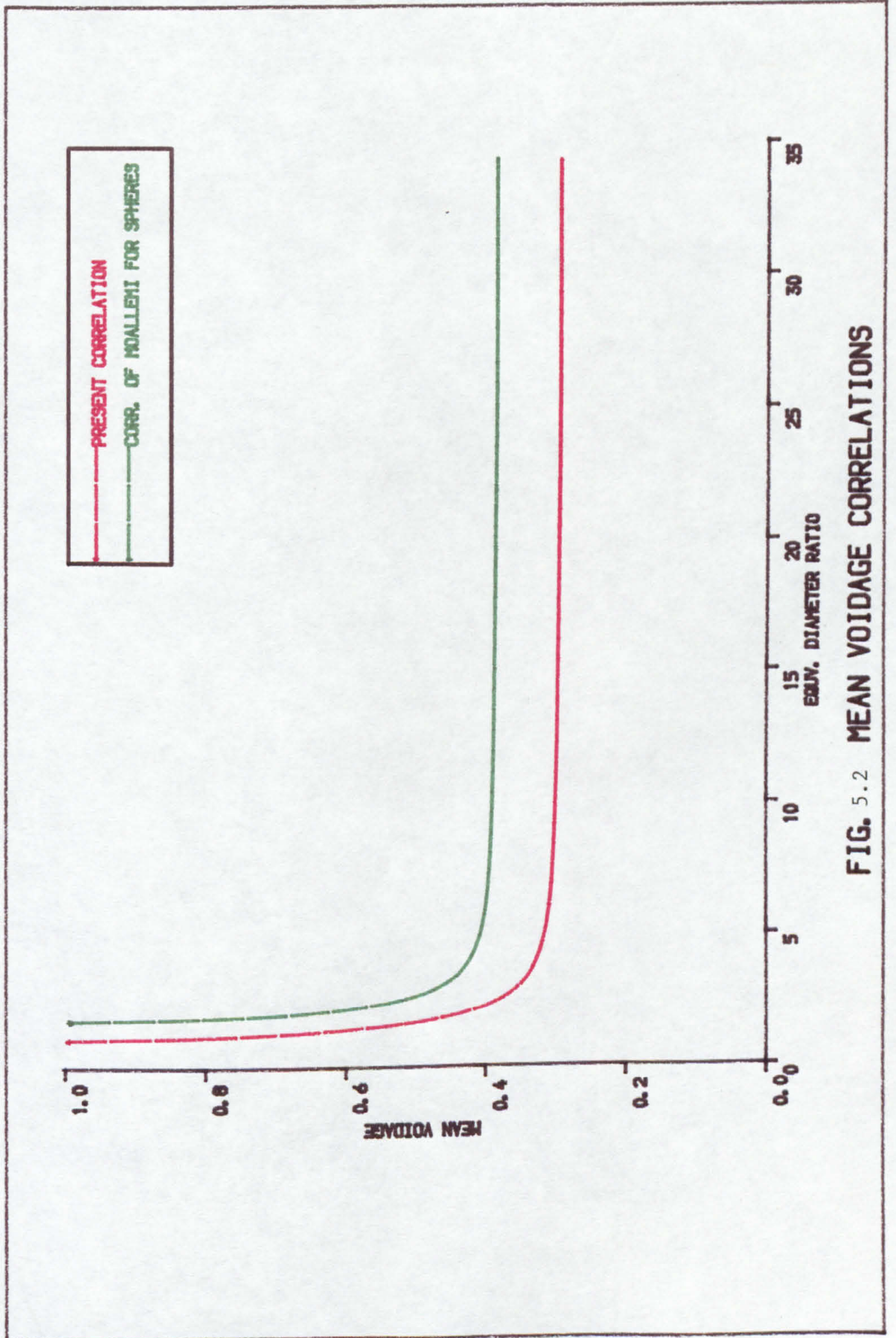


FIG. 5.2 MEAN VOIDAGE CORRELATIONS

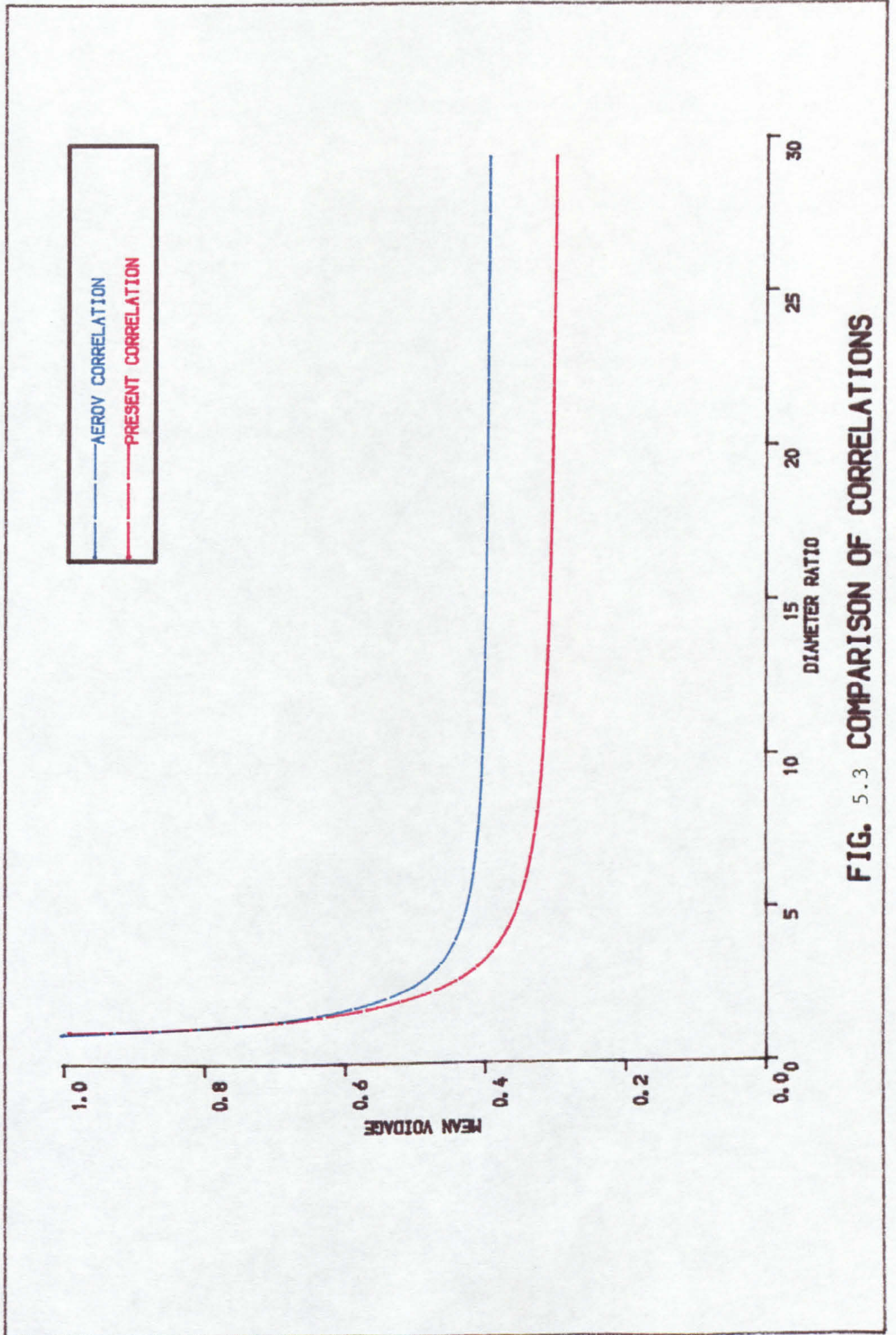


FIG. 5.3 COMPARISON OF CORRELATIONS

spheres, $d_t/d_p \geq 10$, the bulk mean voidage has a value of about 0.4 according to Aerov. The criterion for constant ϵ_{mean} is also at a diameter ratio of 10.

The correlation derived by Ayer and Soppett, (1964), for beds of unisized spheres is compared with the present correlation for cylinders in Figure 5.4. A constant value of ϵ_{mean} is indicated after a diameter ratio of 10, which is about 0.36. According to their expression the highest value of mean voidage possible is 0.58 when d_t/d_p reaches 0.

Beavers, et al. (1973) correlation for beds of spheres, shown in Figure 5.5, indicates lower values of ϵ_{mean} for diameter ratios between 1 and 3 compared to present expression. However, a constant mean voidage value of about 0.4 is reached at $d_t/d_p \geq 10$ which again is the same criterion for cylinders.

Griffiths (1986) correlation is different to all the other expressions mentioned earlier, in the sense that no information can be derived for diameter ratios less than 3.5. Also a constant mean voidage value of about 0.41 is reached at a diameter ratio of 5 for packed beds of unisized spheres. Figure 5.6 illustrates the comparison between his correlation and the present one.

5.3 Local Voidage Correlations

As it was mentioned in previous chapters, review of the literature to find an expression for the prediction of packed beds of cylindrical particles structure proved unsuccessful. The nearest that one can get is the correlation proposed by Kondilik et al., (1968), which predicts the distance of the first minimum local void fraction from the wall for various diameter ratios of packed beds of cylinders. They found that the minimum local voidage occurs at 0.5 to 0.75 d_p away from the wall and it is influenced by the d_t/d_p ratio. The value of this minimal void fraction is reported to be between 0.1 and 0.2, about one half of the void fraction found in beds of equal spheres. The range of the diameter ratio of the beds investigated were between 2 and 8. Figure 5.7 compares their correlation with the findings of the present study.

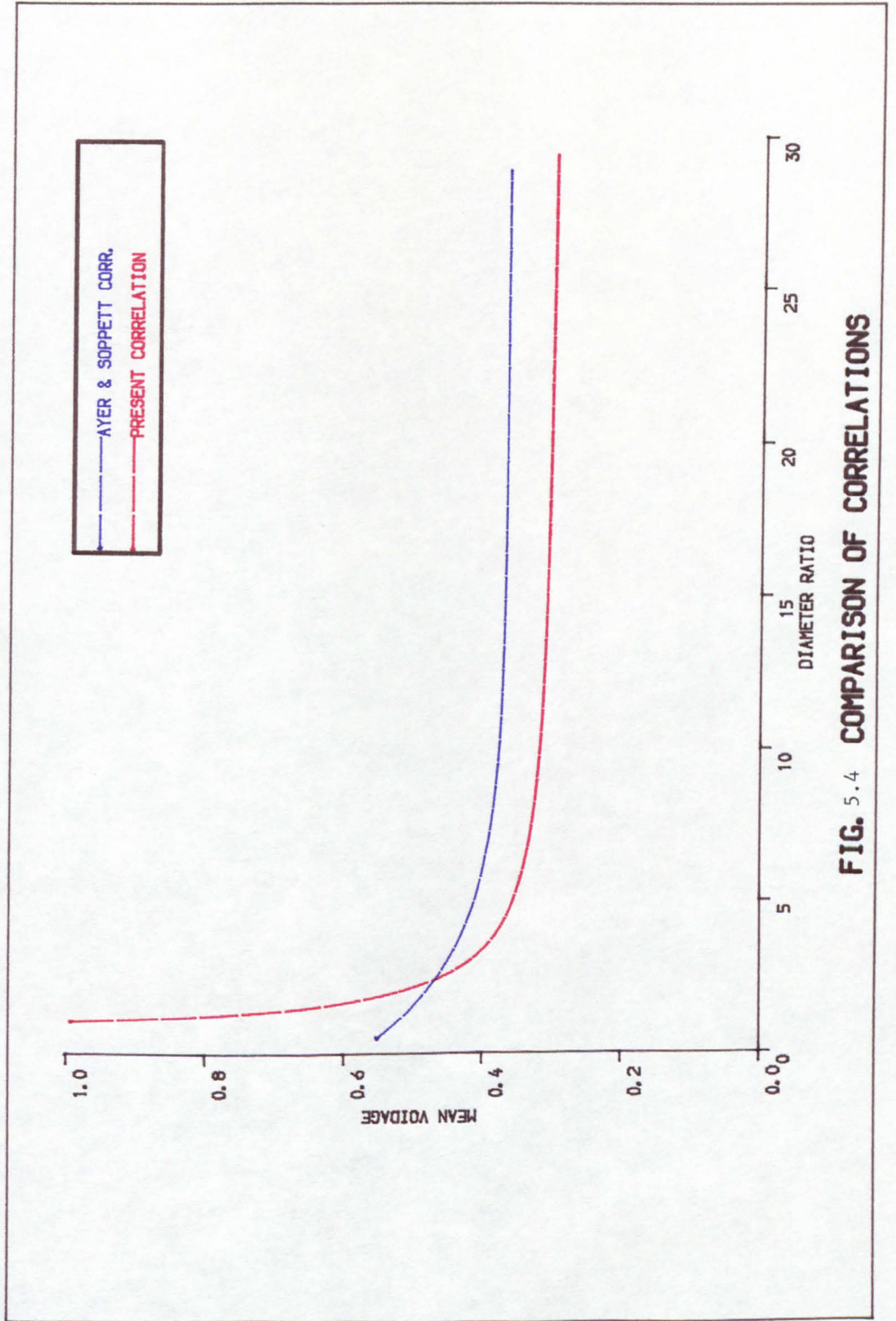


FIG. 5.4 COMPARISON OF CORRELATIONS

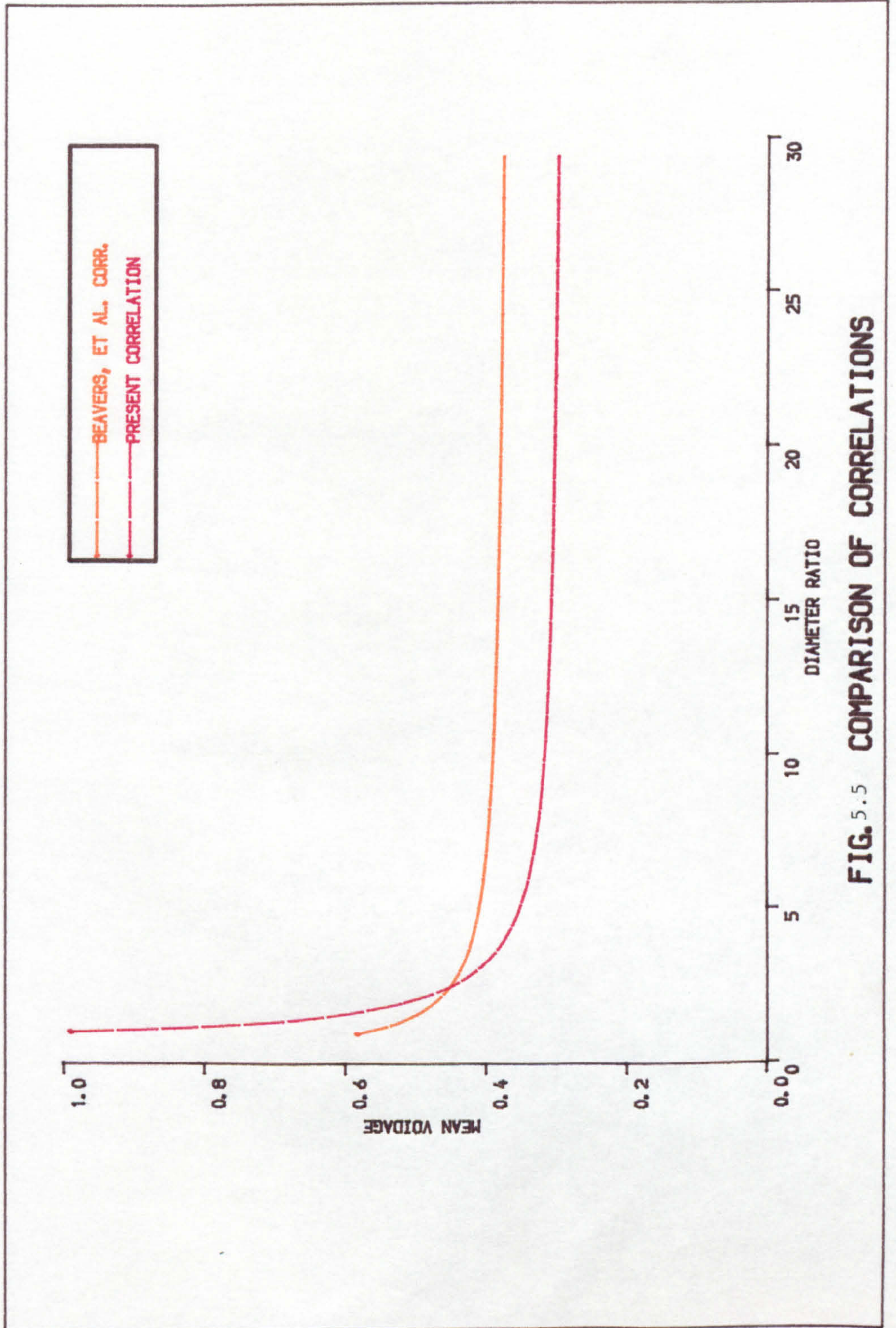


FIG. 5.5 COMPARISON OF CORRELATIONS

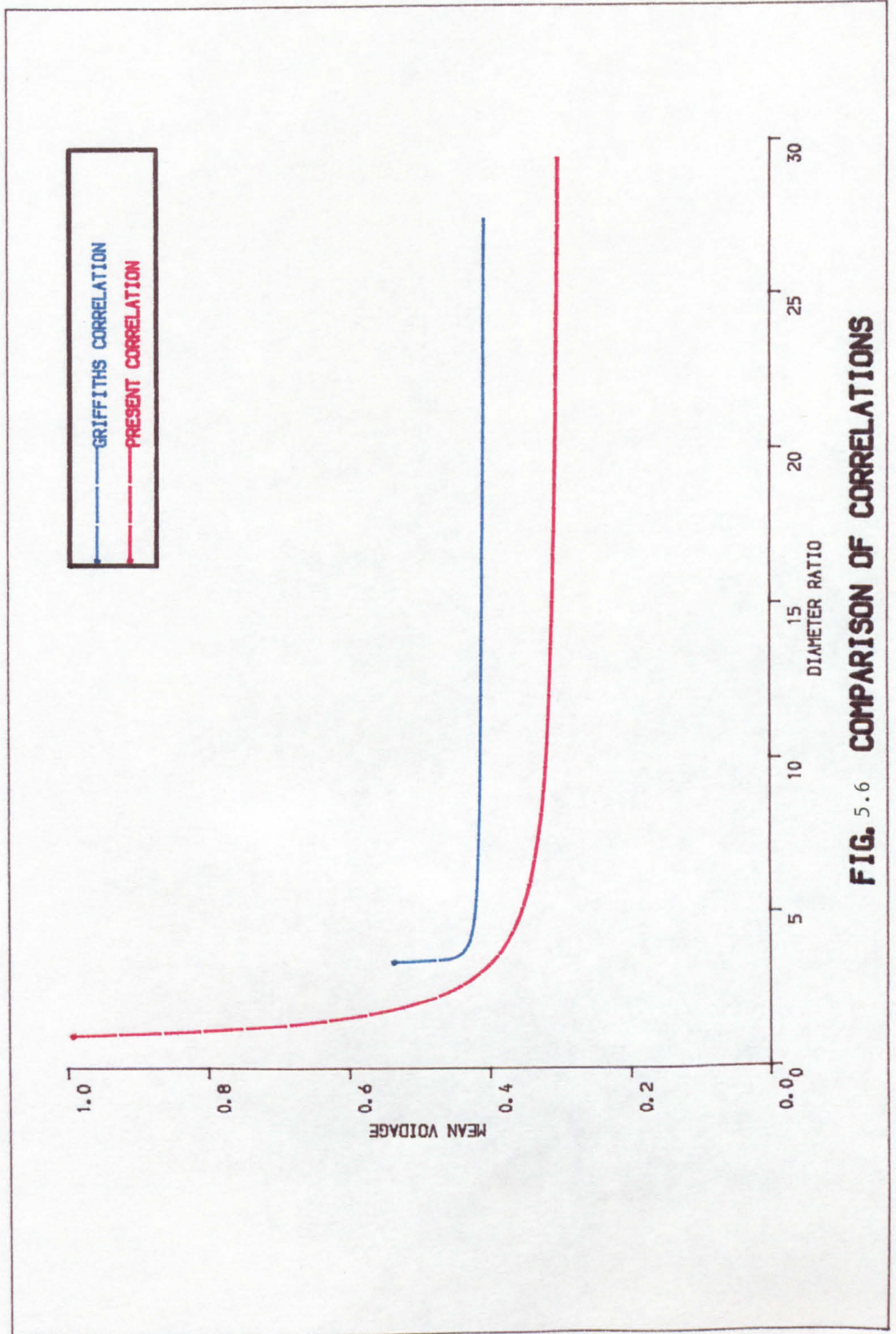


FIG. 5.6 COMPARISON OF CORRELATIONS

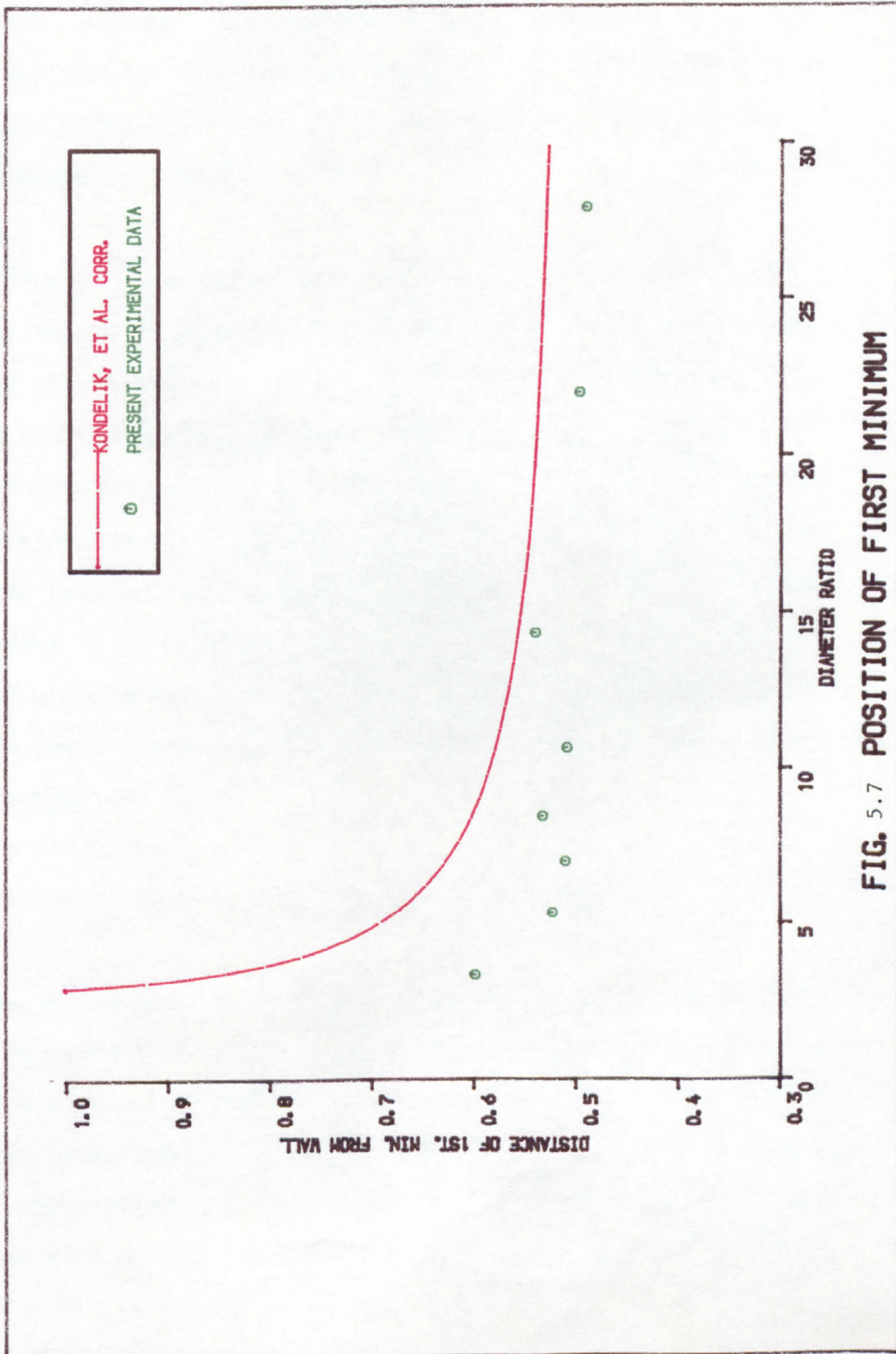


FIG. 5.7 POSITION OF FIRST MINIMUM

It is clear that Kondilik, et al., overpredicts the position of the first minimum from the wall. This discrepancy could be due to inadequate data points which their expression is based upon, and also the accuracy of the data points themselves. Table 5.1 indicates the values of the minimal void fraction obtained in this study for various beds together with the position at which it occurs, and also the predicted position by Kondilik et al. correlation. The values of the minimal void fractions for the beds examined in this study are between 0.1 and 0.2 which agreed with their findings.

There are also a few reported correlations concerned with packed beds of monosize spherical particles, which are either too simplified (Chandrasekhara and Vortmeyer, (1979); Pillai, (1977); Kubie, (1974); Vortmeyer and Schuster, (1983)), or based on inaccurate data (Ridgway and Tarbuck, (1966); Cohen and Metzner, (1981); Martin, (1978); Johnson, (1970)). More recently, Moallemi, (1989); also proposed a correlation for prediction of local voidage in the radial direction of beds of spheres based on very accurate data acquisition which is by far the most representative of the structure of such beds. His derived expressions for the prediction of coefficients of the general radial correlation were substituted with an optimised constant value, as described in Section 4.4.2, which resulted in the following expression:

$$\epsilon(r) = 1.0 - 0.6 \{1.0 - \exp(-1.7 d_{re}^{0.52}) \cos(5.57 d_{re}^{1.25})\} \quad (5.1)$$

The above expression is compared with its analogous expression, Equation (4.8), derived for equilateral cylinders. Figure 5.8 shows the difference in the profiles of the two equations and also indicates the resemblance of their features. They both start at the voidage value of 1.0 at the wall and decrease in a damped oscillatory manner. The amplitude of the oscillations is different and the voidage at the core zone is about 0.4 and 0.3 for spheres and cylinders, respectively. The first minima

TABLE 5.1
Comparison of the Present Experimental Data
with Kondilik et al., Correlation

Diameter Ratio	r	r Predicted	minimal Void Fraction
3.37	0.598	0.876	0.193
5.35	0.522	0.680	0.181
7.02	0.509	0.627	0.154
8.50	0.532	0.601	0.155
10.70	0.508	0.578	0.149
14.41	0.540	0.556	0.133
22.05	0.496	0.536	0.134
27.95	0.489	0.528	0.129

r = distance from the wall in diameter of particles unit.

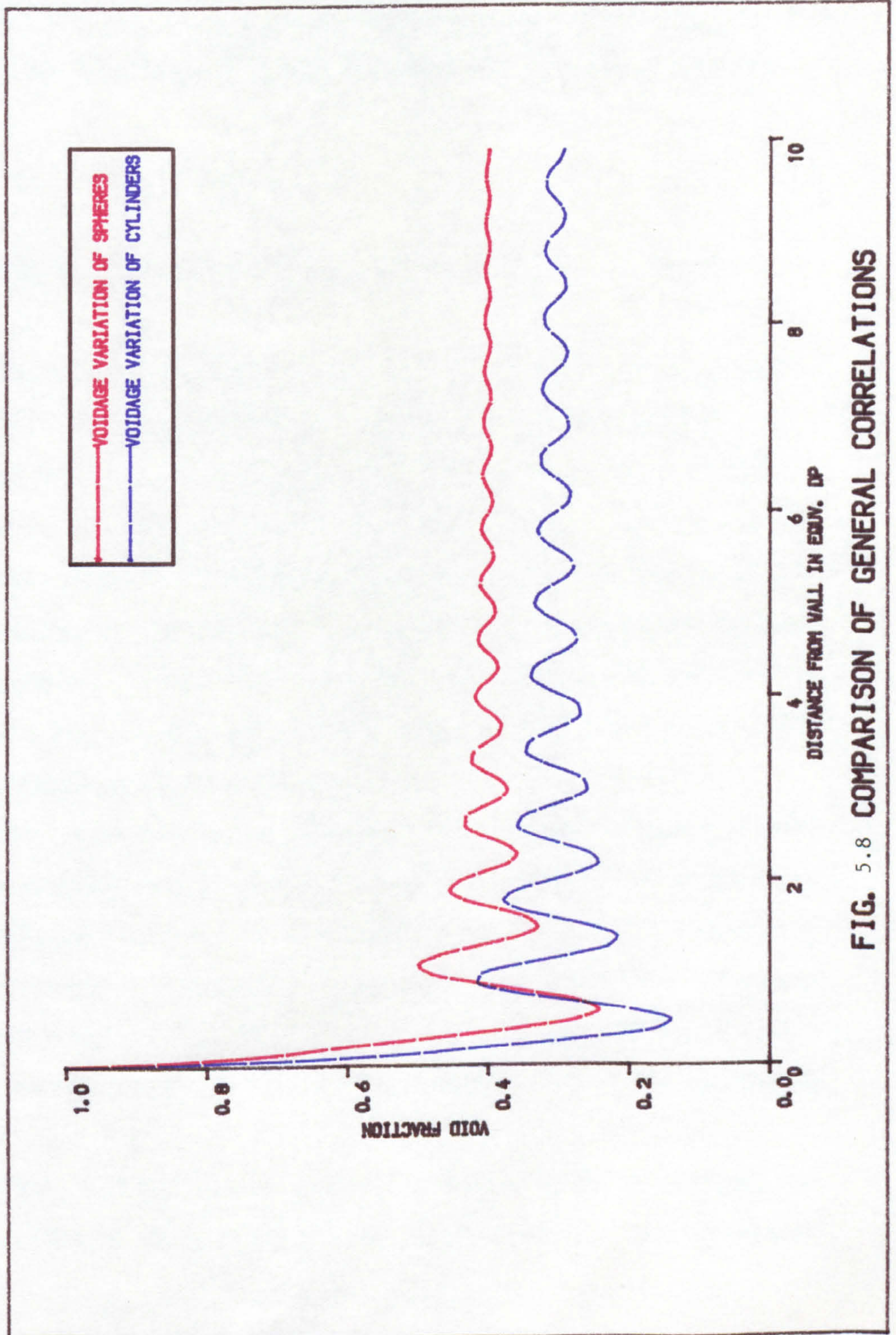


FIG. 5.8 COMPARISON OF GENERAL CORRELATIONS

and maxima occur at $\frac{1}{2} d_p$ and $1 d_p$ away from the container wall in both profiles. Because of the presence of such similarities in the profiles of the two packing materials, an attempt was then made to relate the two by means of sphericity, Φ , which is defined as the ratio of the surface areas of the sphere with any other particle shape, (cylinders in this case). The following relationship was concluded:

$$\alpha_{\text{cylinders}} = \alpha_{\text{spheres}} \Phi^n \quad (5.2)$$

where n is a constant value, and α is the coefficient of the radial correlation.

The value of sphericity, Φ , for the same volume of cylinders is calculated to be 0.8736. The values of n for each coefficient of the radial correlation, α , are optimized to be - 1.1, - 0.5, - 1.3, 3.1 and 1.1, respectively. Figures 5.9 and 5.10 compare the data obtained from the beds with $d_{re} = 5.35$ and 7.02 with the correlation profile based on sphericity. Equations 5.1 and 5.2 could then be used together for the prediction of variations of voidage in the radial direction in packed beds of spheres as well as equilateral cylinders. Appendix D reveals how the values of Φ and n are calculated together with the data points of each bed compared with the correlation profile based on sphericity.

The generalised correlation derived for the prediction of voidage in radial direction of packed beds of equilateral cylinders is compared with the correlation proposed by Johnson (1970) for unisized spheres. Figure 5.11 shows the profiles of the two expressions. The present correlation generally exhibits lower minima and maxima at $\frac{1}{2} d_{pe}$ and $1 d_{pe}$ away from the container wall, respectively. The value of void fraction at the core zone of beds of spheres is about 0.4 according to Johnson.

Figure 5.12 compares the exponential decaying profile of Vortmeyer and Schuster (1983) for beds of spheres with the present correlation. They also indicate

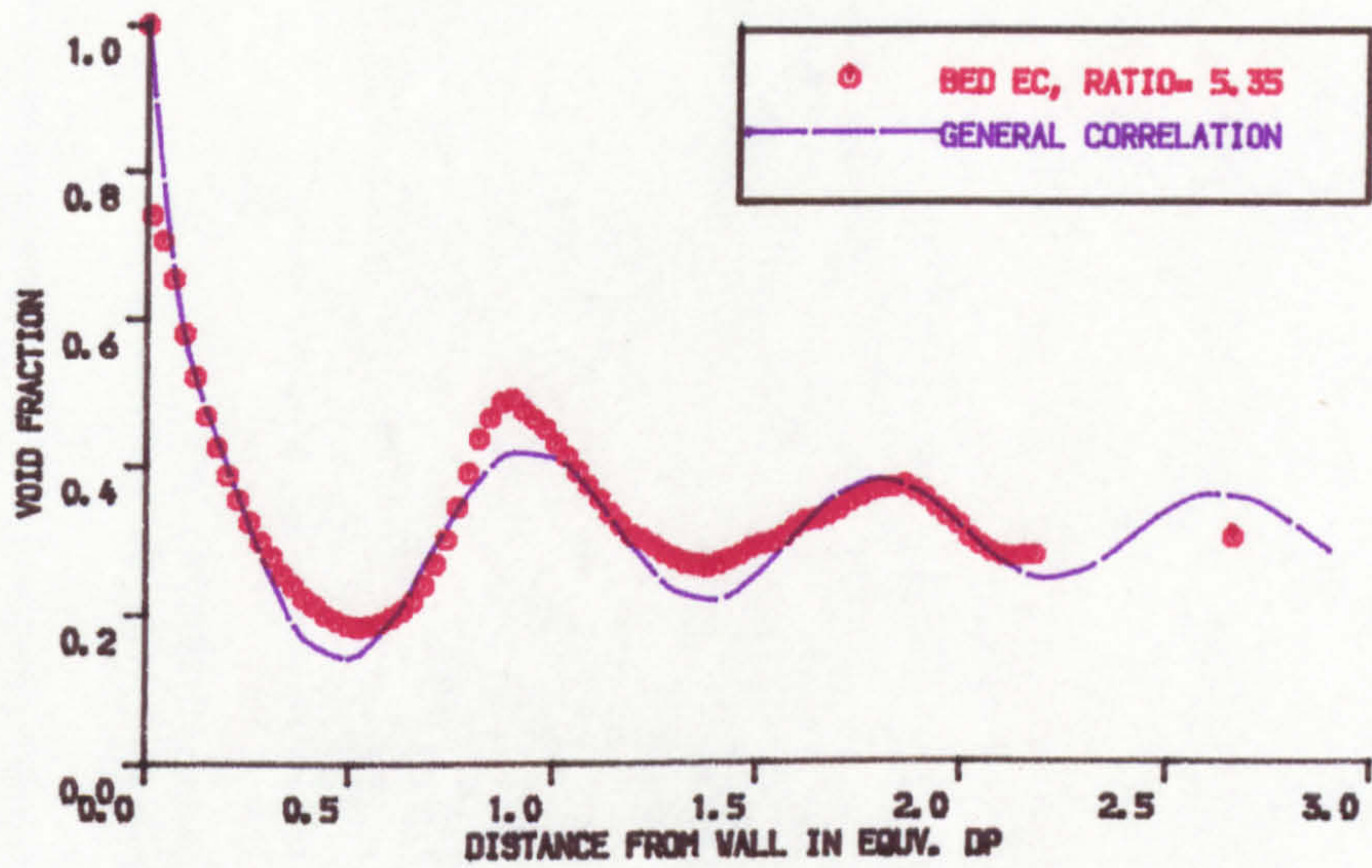


FIG. 5.9 SPHERICITY BASED CORRELATION, EC

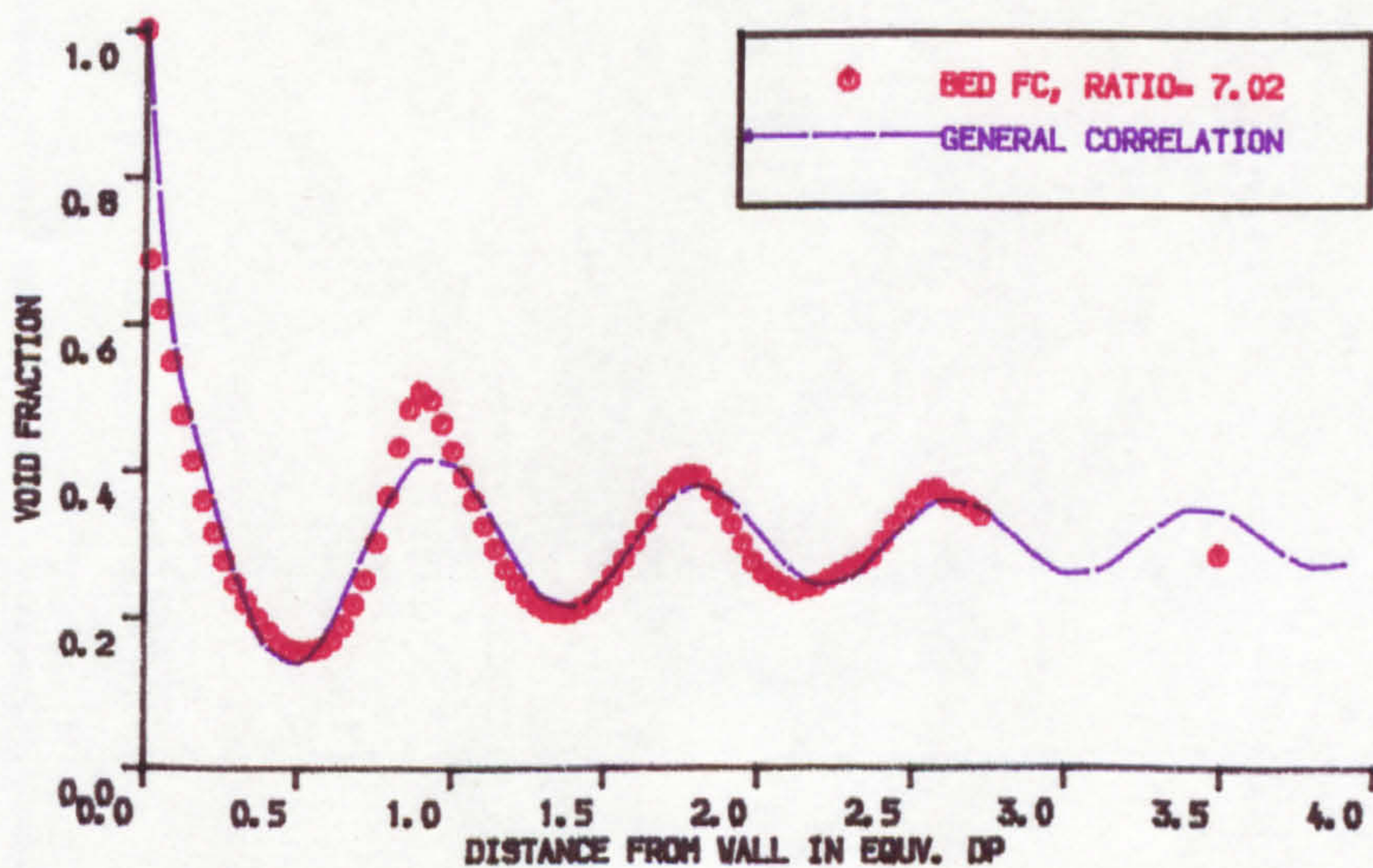


FIG. 5.10 SPHERICITY BASED CORRELATION, FC

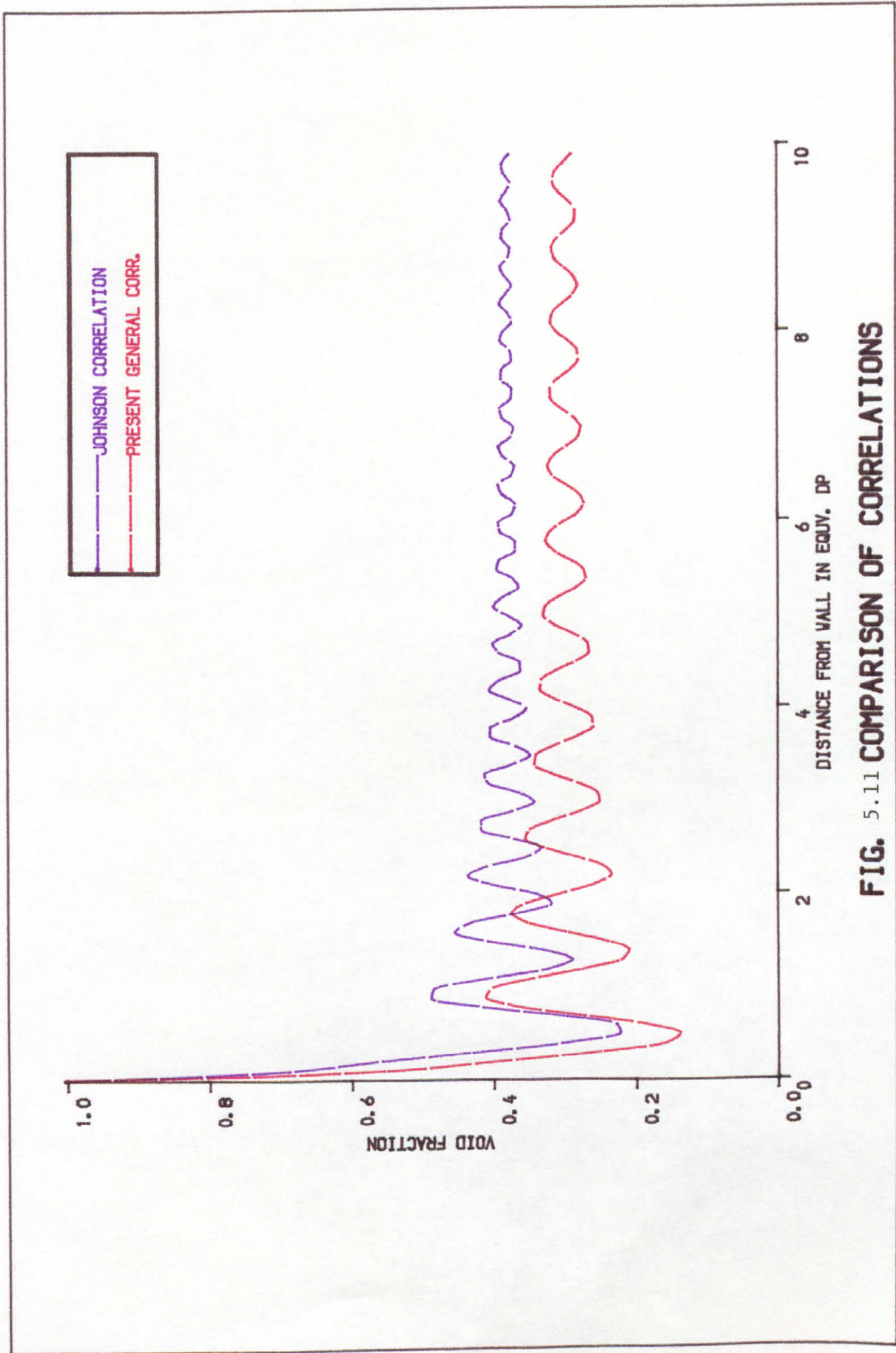


FIG. 5.11 COMPARISON OF CORRELATIONS

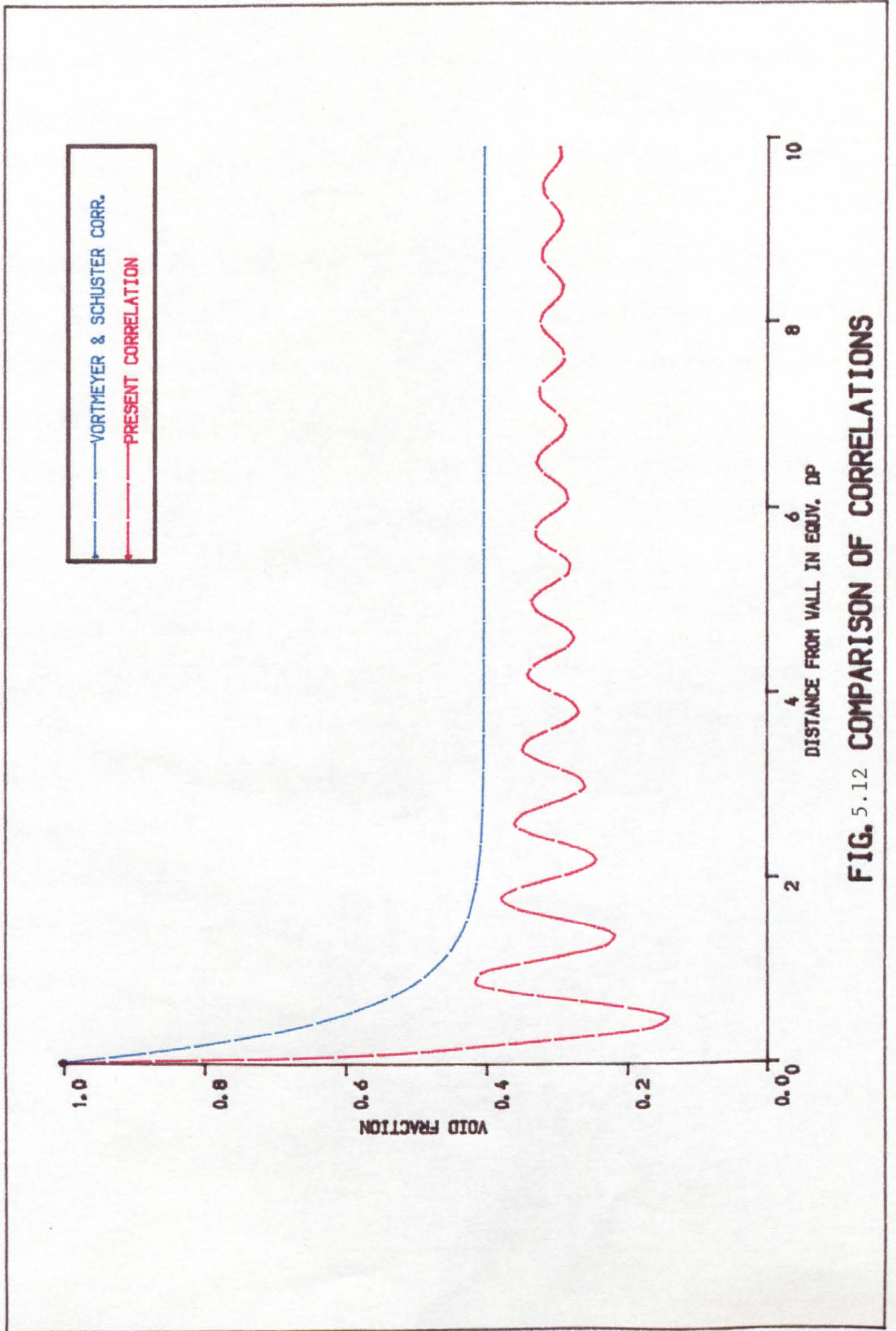


FIG. 5.12 COMPARISON OF CORRELATIONS

a value of 0.4 for the void fraction at the core zone. Kufner and Hofmann (1990), improved on the Vortmeyer and Schuster correlation by introduction of a cosine term into their expression. Figure 5.13 compares the profile of the resultant expression with the derived correlation of this study. The first minimum occurs at $\frac{1}{2} d_{pe}$ away from the wall, but has a lower value of void fraction when compared with the present correlation. Apart from this their profile generally exhibits higher minima and maxima, reaching a constant value of 0.4 after 4 particle diameters from the wall.

Generally speaking, the profiles of the proposed correlations for packed beds of monosized spheres show higher minima and maxima values of void fraction than the profile of general correlation for equilateral cylinders. Also the value of voidage at the core zone is higher in beds of spheres.

The experimental data of Roblee, et al., (1958); and Benenati and Brosilow, (1962); for beds of equilateral cylinders were also compared with the obtained data of this work. They both considered only a couple of beds, of which the bed with equivalent diameter ratio of 11.7 is chosen to be compared with the data obtained from bed GC with $d_{re} = 10.7$. Figures 5.14 and 5.15 show the oscillatory behaviour of both sets of data with lower maxima points, with the first minimum at $\frac{1}{2} d_{pe}$ away from the wall. It is apparent that both sets lack sufficient data points due to their poor techniques in acquiring them, as discussed in Chapter 2, and hence valuable information concerned with the structure of the bed is lost.

As far as the variation of voidage in the axial direction is concerned the correlation proposed in Chapter 4, Equation (4.14), is unique and therefore cannot be compared with any published correlations.

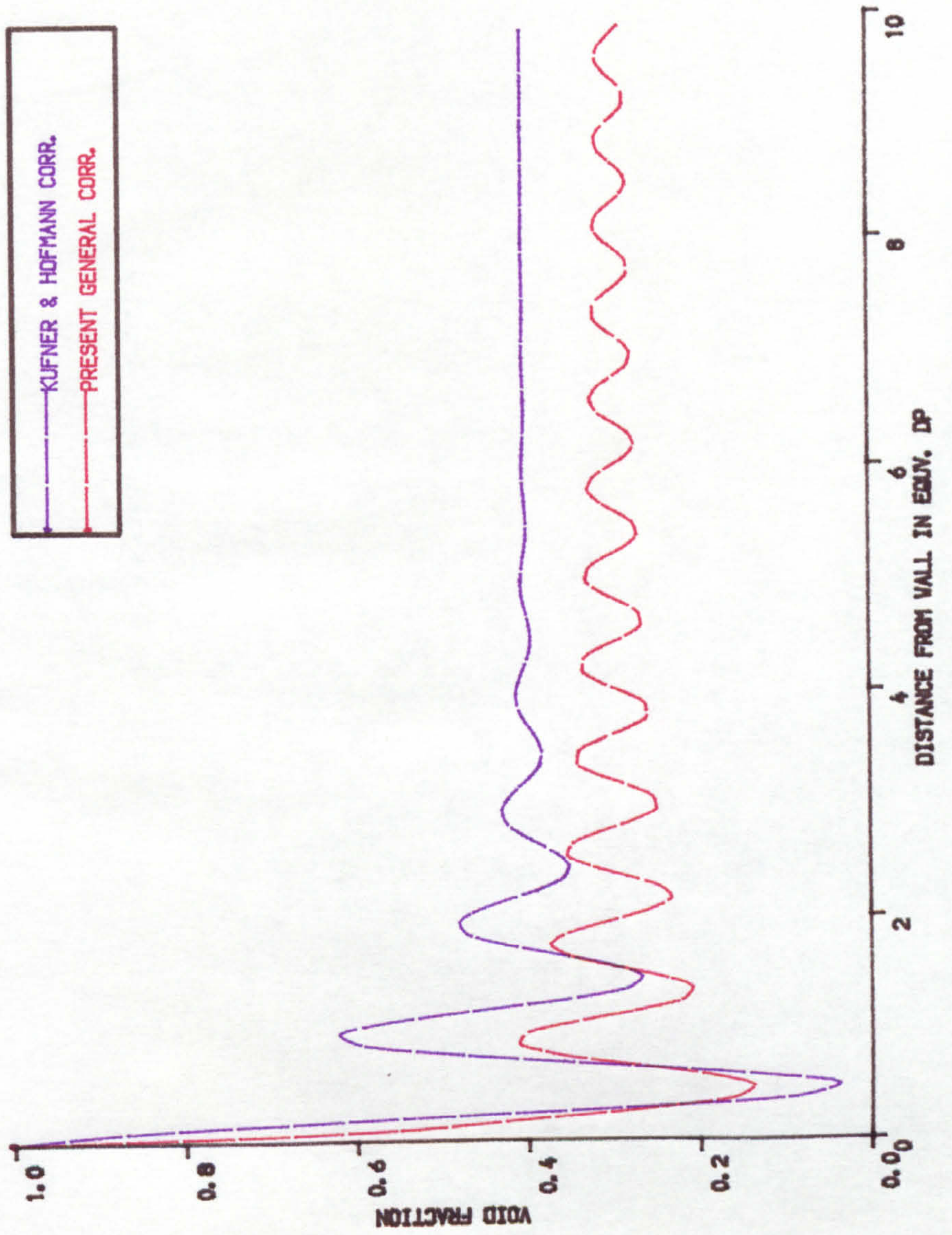


FIG. 5.13 COMPARISON OF CORRELATIONS

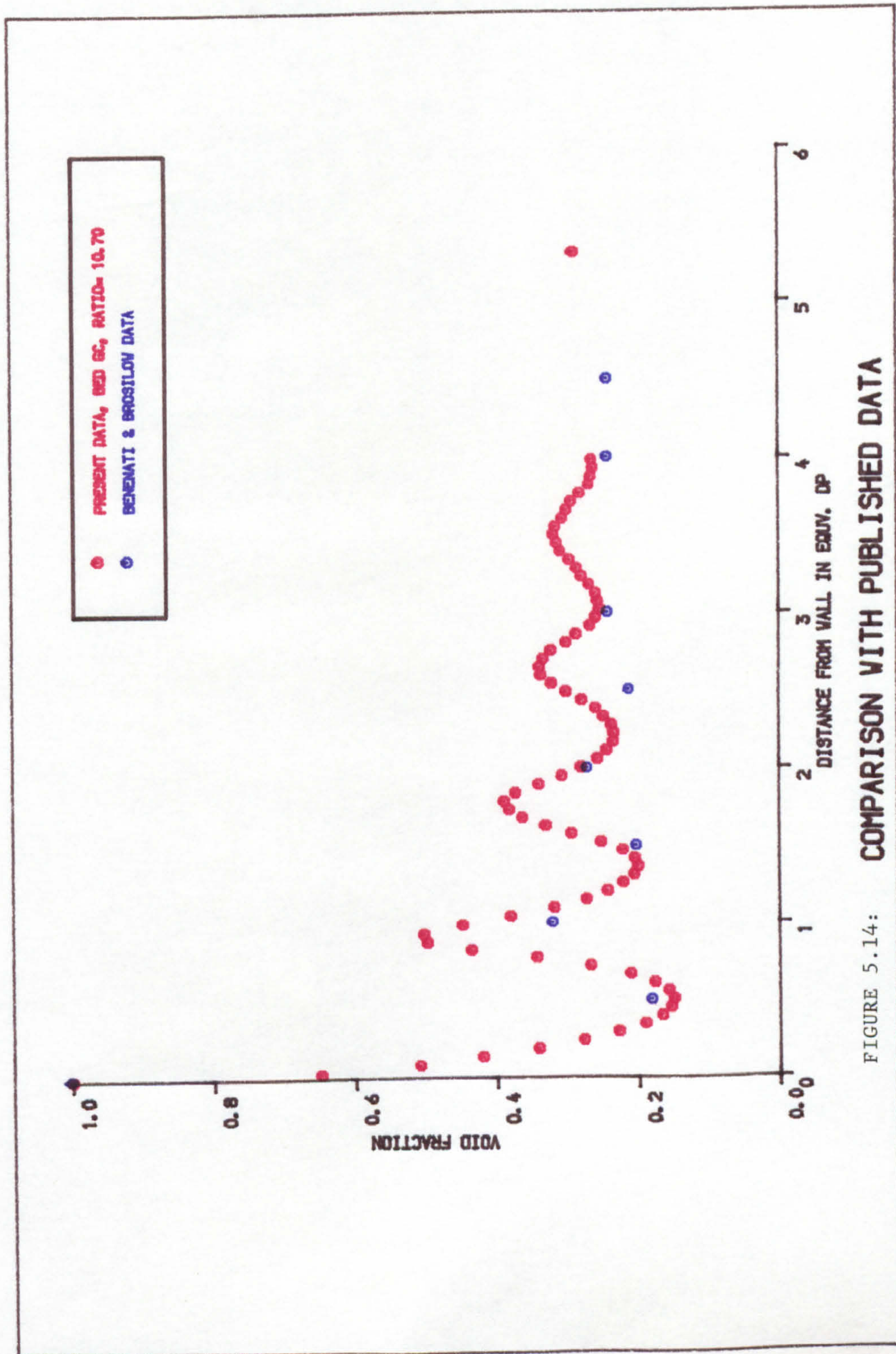


FIGURE 5.14: COMPARISON WITH PUBLISHED DATA

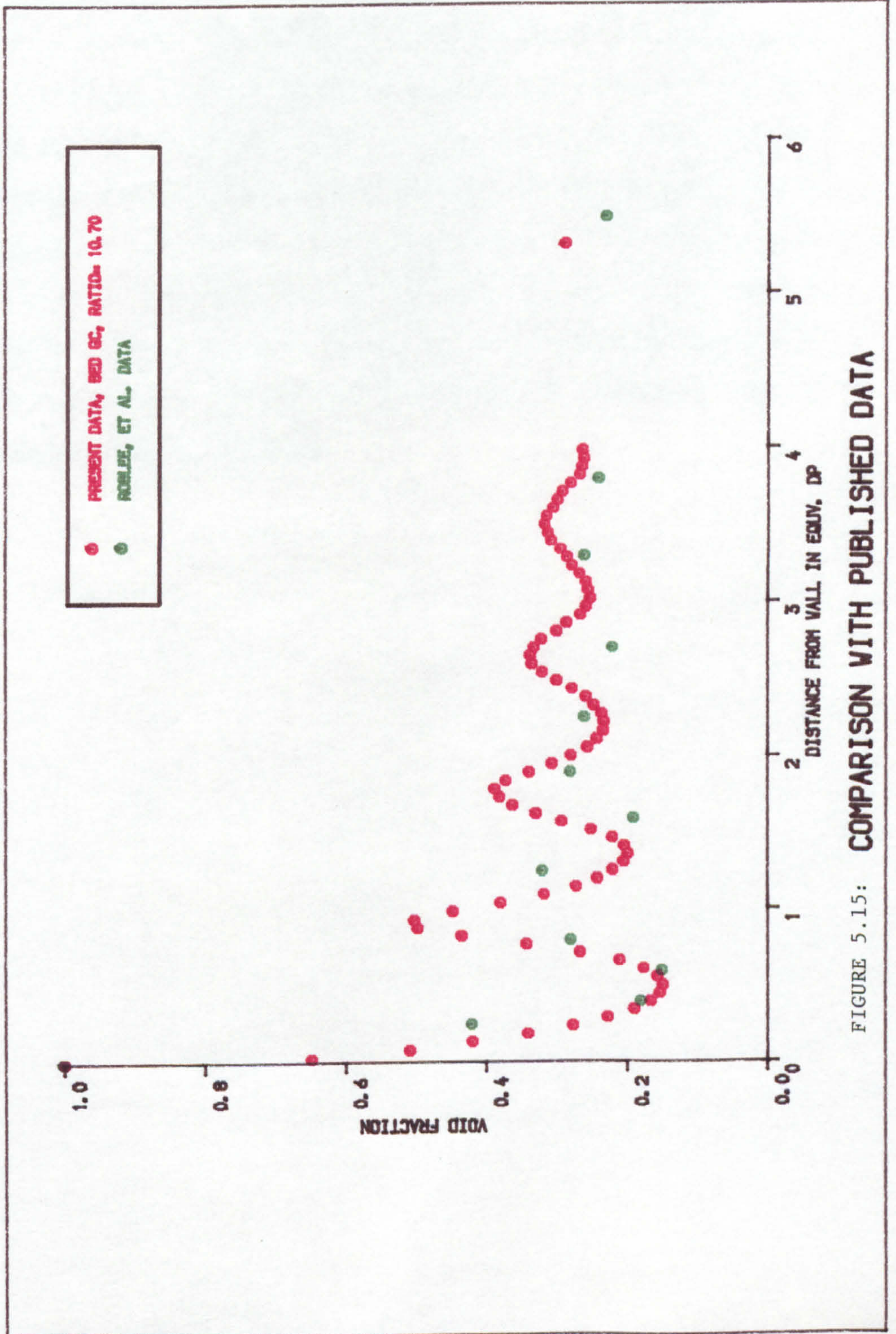


FIGURE 5.15: COMPARISON WITH PUBLISHED DATA

5.4 Concluding Remarks

The mean voidage correlation of the present work shows a functional relationship between the mean voidage, ϵ_{mean} , of the particulate bed and the tube to particle diameter ratio. The comparison of this expression with an appropriate published correlation revealed reasonable agreement. The expression was also compared with a few proposed for spheres. The correlation for the prediction of local voidage in the radial direction was manipulated to be used for packed beds of equilateral cylinders as well as monosize spherical particles. This general expression was then compared with sample data which illustrated a close fit. The generalised correlation for cylinders was also compared with a few correlations published for packed beds of spheres. Also the experimental data of this study were compared with the ones available in the literature.

CHAPTER 6

CONCLUSIONS AND RECOMMENDATIONS

CHAPTER 6

CONCLUSIONS AND RECOMMENDATIONS

6.1 Introduction

This chapter recalls the principal themes of this research activity and gives a brief summary of the main achievements and contributions to the field of voidage distribution in packed beds. Issues which require further investigation to resolve are also highlighted in this chapter, together with some suggestions in order to tackle them. These suggestions are offered in the form of recommendations for future activities in this field.

6.2 Conclusions of the Present Study

A comprehensive investigation into the global structural properties of the packed beds of cylindrical particles was conducted. The magnitude of the bulk mean voidage index which is the quantity most frequently used to characterise the overall space available for flow in such beds is concluded to be a function of the tube to particle diameter ratio. A constant mean voidage value of about 0.3 is obtained for beds of large d_t/d_{pe} , i.e. $d_t/d_{pe} \geq 10$. The end effect on the ϵ_{mean} value of packed beds of cylinders with $L/d_t > 3.0$ is found to be very little. The enhanced voidage region formed in beds with low tube to particle diameter ratio is due to the noticeable presence of the container wall which becomes less pronounced as the diameter ratio increases. Based on the observed experimental data, a generalised correlation is derived to allow the bulk mean voidage of beds comprised of full equilateral and different length-to-diameter cylindrical particles to be predicted reliably.

Succeeding the above activities, local properties of the structure of packed beds of equilateral cylinders were thoroughly examined in order to develop a general correlation capable of offering reliable structural knowledge. In order to obtain a 3-dimensional history of the local voidage, $\epsilon(x, r, \theta)$, a sophisticated, high resolution image analyser is employed. The system allowed the microstructural details within packed beds to be examined. All together 10 test beds were prepared, 3 of which were made to study the effects of scaling up the physical dimensions of the bed and particles, and also the effects of reproduction. The results showed that the data obtained from these beds are reproducible, but there is a difference in the values of voidage when the beds are scaled up. The other 7 beds with variable physical dimensions were constructed in such a way that covers the practical range used in industry. The diameter ratio of these beds ranges from 3 to 30 which adequately covers the above specification. The accurate data obtained which represents the magnitude of the radial non-uniformities of voidage in the packing matrix, could now be used as a strong and reliable pillar to base more descriptive and representative models of the physical system upon. These models would certainly facilitate better predictions of the transport coefficients and flow patterns of the fluid through the system, so that their values can subsequently be presented in the form of design correlations.

The stochastic nature of the variation of voidage in the angular direction meant that no recognisable pattern could be identified for the characterisation purposes and hence the variations were averaged in this direction. Inspection of the void fraction profiles in the axial direction, $\epsilon(x)$, revealed the existence of an end effect pronounced up to 4 equivalent particle diameters away from the bed end, after which the voidage becomes constant at about 0.3. One of the interesting findings of the present study is that the nature of the variation of voidage in the axial direction is oscillatory with the first minima and maxima occurring at $\frac{1}{2} d_{pe}$ and $1 d_{pe}$ away from the bed end, respectively. The study of variation of void

fraction in the radial direction, $\epsilon(r)$, revealed that the presence of the wall effect is pronounced up to 3 equivalent particles away from the confining surface, and it also shows a damped oscillatory pattern which flattens in the core zone of the bed corresponding to a value of about 0.28. The first minima and maxima also occur at $\frac{1}{2} d_{pe}$ and $1 d_{pe}$ away from the wall, respectively. The range of the void fraction values for the first minima and maxima are between 0.1 - 0.2 and 0.4 - 0.5, respectively. In general, analysis of the obtained data from the packed beds of equilateral cylinders revealed valuable information on the nature of the global and local voidage patterns, end effect, wall effect, locations of first minima and maxima, reproducibility, and scaling effects. Reliable prediction of the structure of packed beds can be expected based only on such information. A suitable mathematical expression was chosen to characterise the pattern of the variation of voidage in axial and radial directions, as the profiles of both directions exhibit the same features. The best estimates of the related coefficients were calculated by a powerful optimization technique. Statistical analysis indicated a good agreement between the experimental data and the fitted profile. Generalized correlations derived from the individual expressions were then developed by relating the coefficients of the expressions to the diameter ratio, d_t/d_{pe} , of the bed. These expressions allow the variations of voidage in the axial and radial directions to be reliably predicted for beds of cylinders with any value of d_t/d_{pe} .

The generalised correlations developed provide the designers of any packed bed system with an accurate and invaluable knowledge on the structure of the packing matrix. Such information could then be used to understand the fluid mechanics of the system in more detail. Also, development of more realistic and representative mathematical models of heat and mass transfer in such beds are now possible, since the value of mean voidage and interstitial mean velocity in plug flow models could be substituted with the values of local voidage and velocity. The refined models together with appropriate experimental data can be utilised to

extract the best estimate of the transport coefficients which subsequently would be used for correlation purposes. As far as the performance prediction is concerned, the transport parameters, effective radial and axial thermal conductivities, K_{er} , K_{ea} , wall heat transfer coefficient, h_w , and radial and axial dispersion coefficients, D_R and D_L , based on the local flow information can be used to simulate the behavioural characteristics of the system in question.

6.3 Recommendations for Future Work

The present research activity was concentrated on cylindrical assemblies based on equilateral cylindrical particles. The complex nature of this subject both in theory and practice necessitates even more research to be carried out, so that the structural properties of the packed bed configurations could be characterised as comprehensively as possible. As cylindrical particles with different length-to-diameter ratios are also employed in industrial packings, the structure of beds comprised of such particles are recommended to be looked at. Also the geometry of the tube could be changed, say to a square assembly, and then examine the effect and influence of the flat surface boundaries on the packing structure of beds comprised of spheres, equilateral and non-equilateral cylinders.

As mentioned earlier, the scaling up problem of the physical dimensions of the packed beds need to be examined in more detail. So that the functional dependency of the relationship between the variation of void fraction and the physical dimensions of the configuration can be scrutinised in more detail. Therefore, it is recommended to prepare a few beds with different scaling factors and hence study the local variations of voidage in them.

Regarding the improvement to the accuracy of the experimental data, a more sophisticated and accurate cutter is recommended for cutting lead rods into cylinders. The cost and time consumption could be a decisive factor in choosing a more suitable option.

As the profiles of the variation of voidage in axial and radial directions show similar features in packed beds of equilateral cylinders, it is recommended to derive a mathematical expression and calculate the corresponding coefficients, so that the variations of voidage in both directions could be predicted at the same time. Such information can then be used in appropriate heat and mass transfer models for better representation of the system.

The present well-founded data which is an essential pre-requisite for accurate design of any packed bed system could now be utilised in appropriate mathematical models, pseudohomogeneous or heterogeneous, to provide valuable information not only in terms of performance prediction but also parameter estimation. More representative models of the physical system can therefore be formulated and solved based upon the available data on the bed structure.

The findings of this research work indicate that refined mathematical models incorporating the desired non-uniformities can be employed to provide the behavioural characteristics of packed beds. As an example a typical two-dimensional, pseudohomogeneous model is given below where the heat transfer is described by a number of equations, Equations (6.1) to (6.5). In this specific example only the variations of properties in the radial co-ordinates of the cylindrical geometry are considered.

$$\left[\epsilon(r) \rho_f c_f + (1 - \epsilon(r)) \rho_s c_s \right] \frac{\partial T}{\partial t} = k_{ea} \frac{\partial^2 T}{\partial z^2} + \frac{1}{r} k_{er}(r) \frac{\partial}{\partial r} \left[r \frac{\partial T}{\partial r} \right] - u(r) \rho_f c_f \frac{\partial T}{\partial z} \quad (6.1)$$

with the initial conditions:

$$t = 0 \quad 0 \leq z \leq L \quad T = \text{known}$$

$$0 \leq r \leq R$$

with the boundary conditions:

$$z = 0 \quad 0 \leq r \leq R \quad \rho_f c_f u_i (T - T_i) = K_{ea} \frac{\partial T}{\partial z} \quad (6.2)$$

$$z = L \quad 0 \leq r \leq R \quad \frac{\partial^2 T}{\partial z^2} = 0 \quad (6.3)$$

$$r = 0 \quad 0 \leq z \leq L \quad \frac{\partial T}{\partial r} = 0 \quad (6.4)$$

$$r = R \quad 0 \leq z \leq L \quad -K_{er} \frac{\partial T}{\partial r} = h_w (T - T_i) \quad (6.5)$$

where,

c_s, c_f specific heat capacities of solid and fluid,

h_w wall heat transfer coefficient based on local information,

K_{ea} effective axial thermal conductivity coefficient,

K_{er} effective radial thermal conductivity coefficient,

L length of the bed,

r radial co-ordinate,

R radius of the tube,

T temperature,

t time,

u interstitial velocity,

z axial co-ordinate,

ϵ point void fraction,

ρ_s, ρ_f densities of solid and fluid,

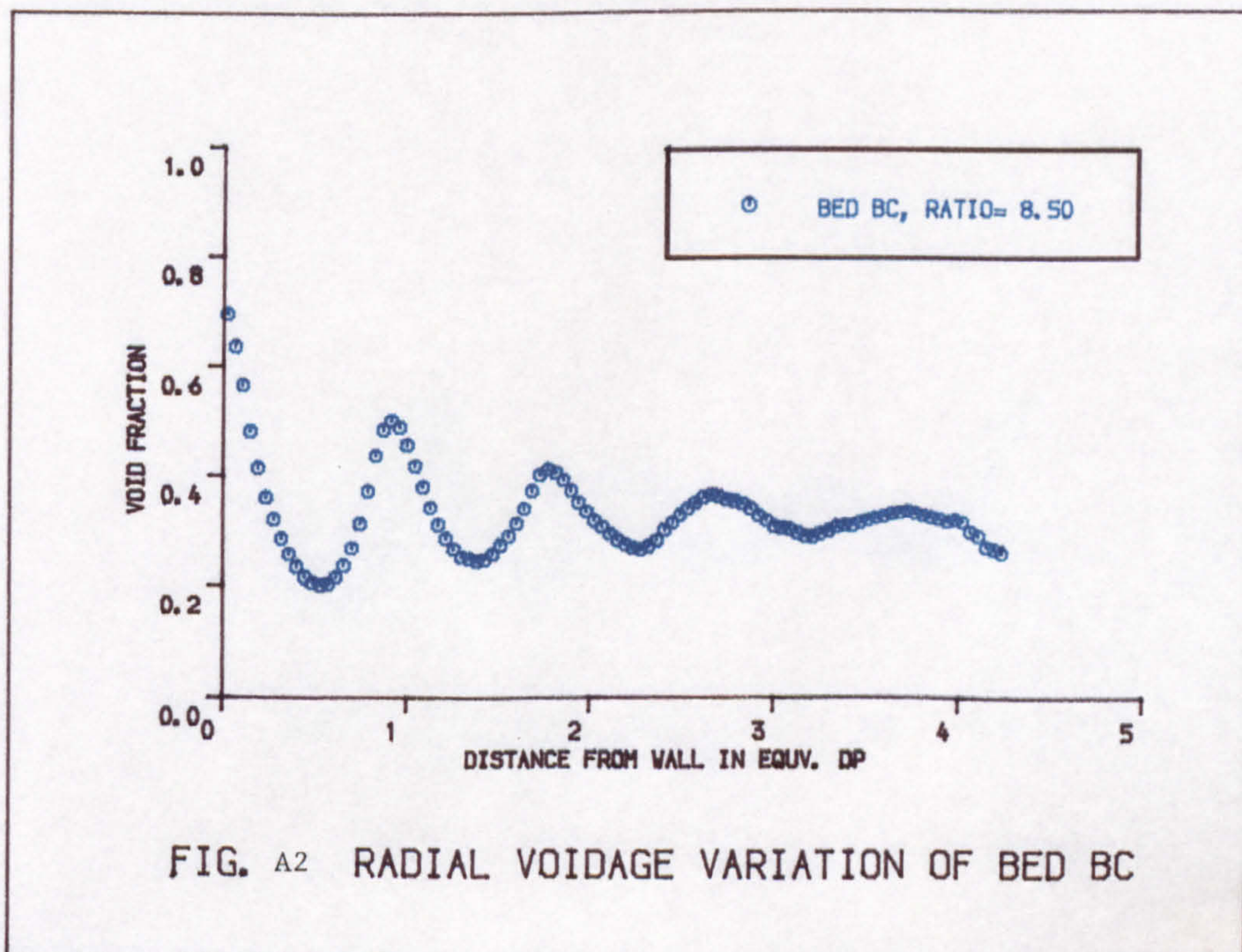
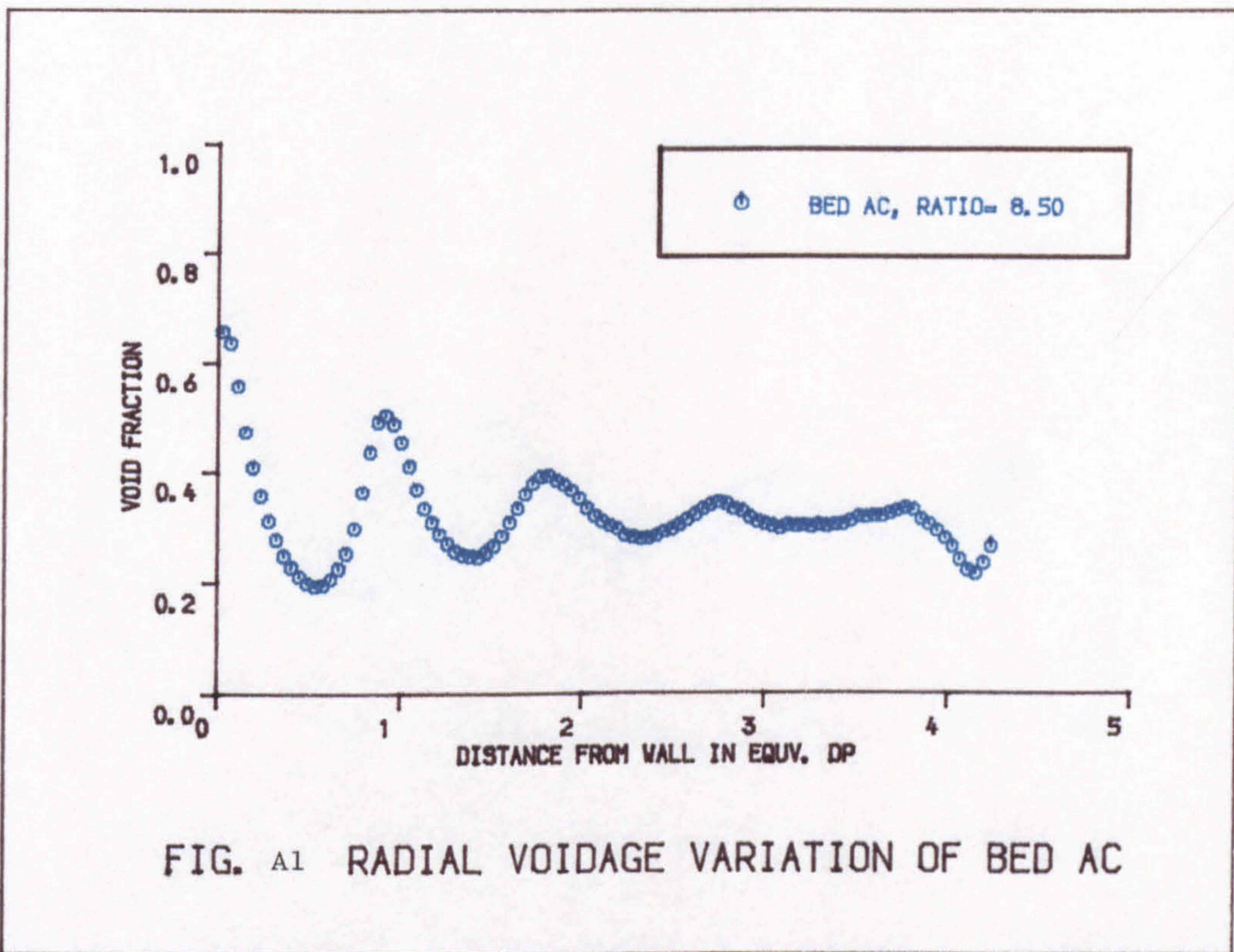
i subscript, denotes condition at inlet.

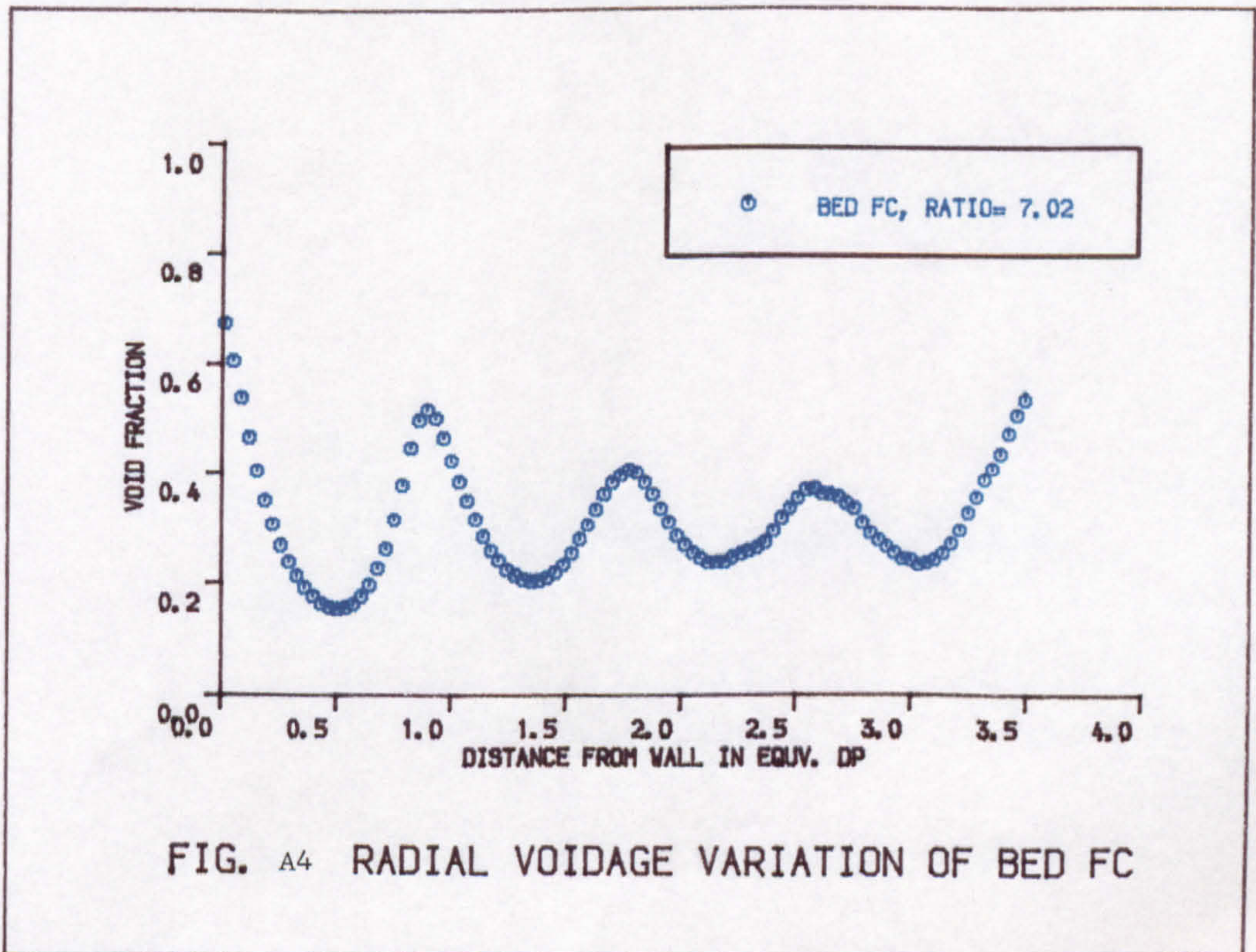
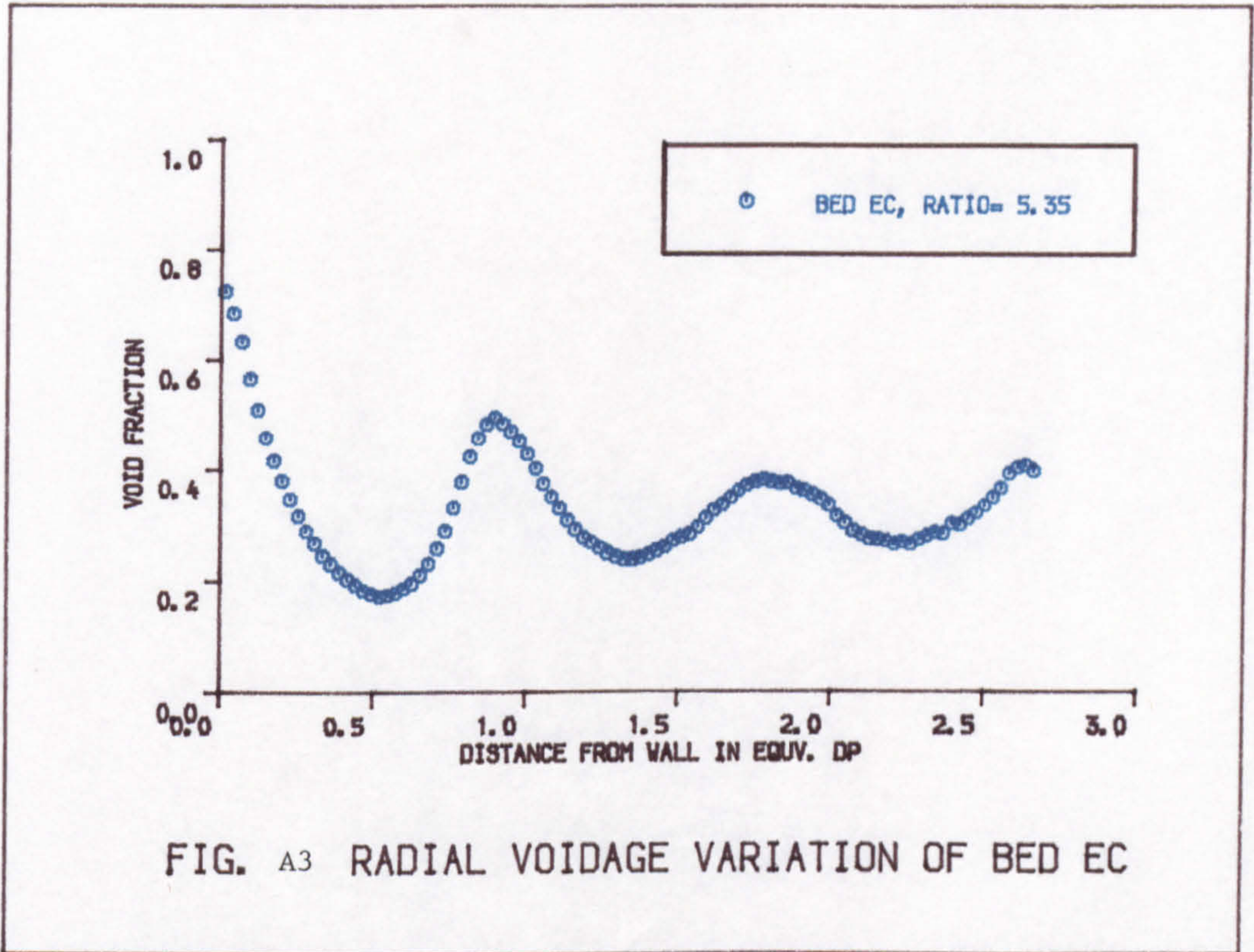
Finally, the results indicate that the diameter ratio is an important structural parameter in packed bed configurations, and that the mean voidage of the packing is a function of this ratio. Since the mean voidage affects the pressure drop-flow

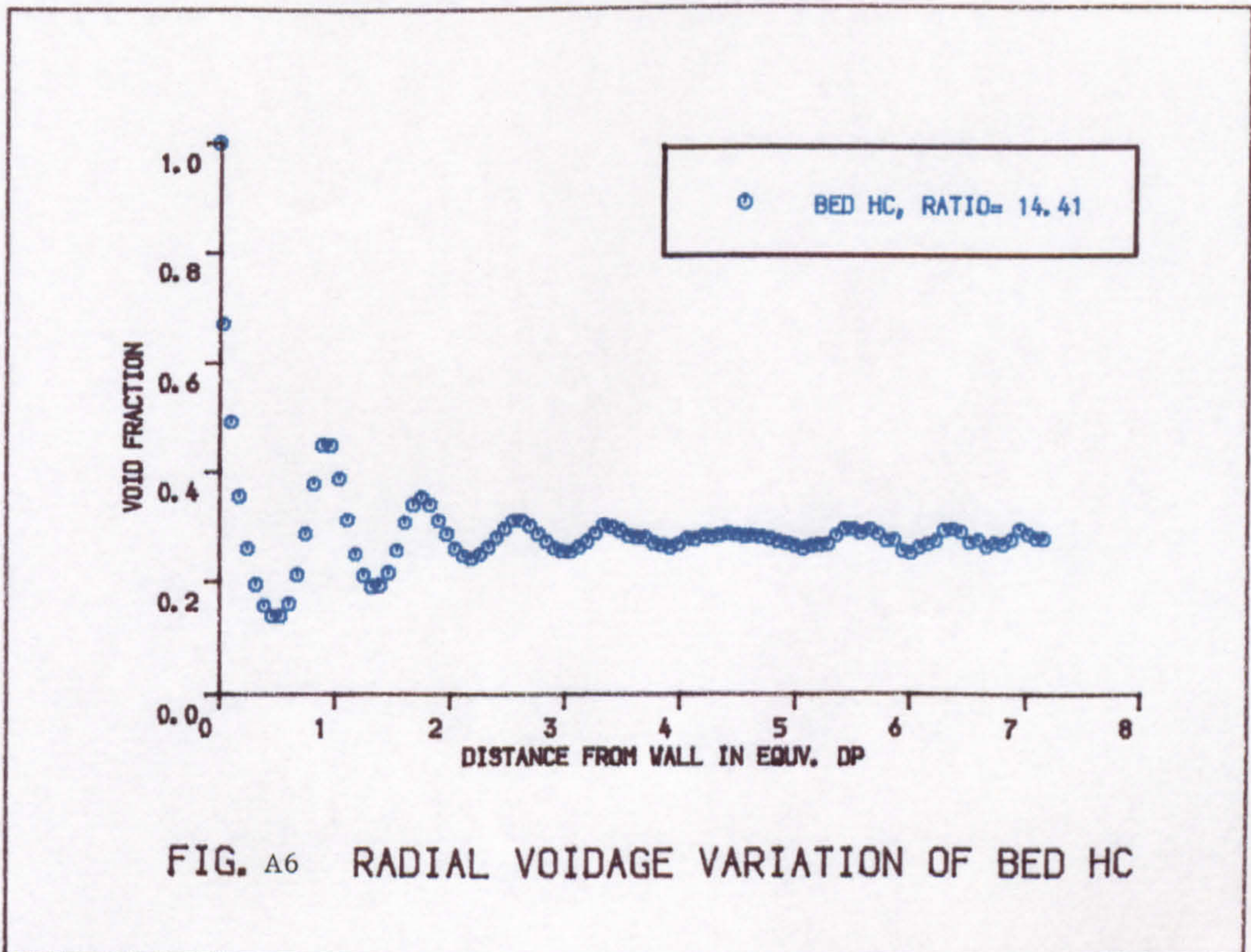
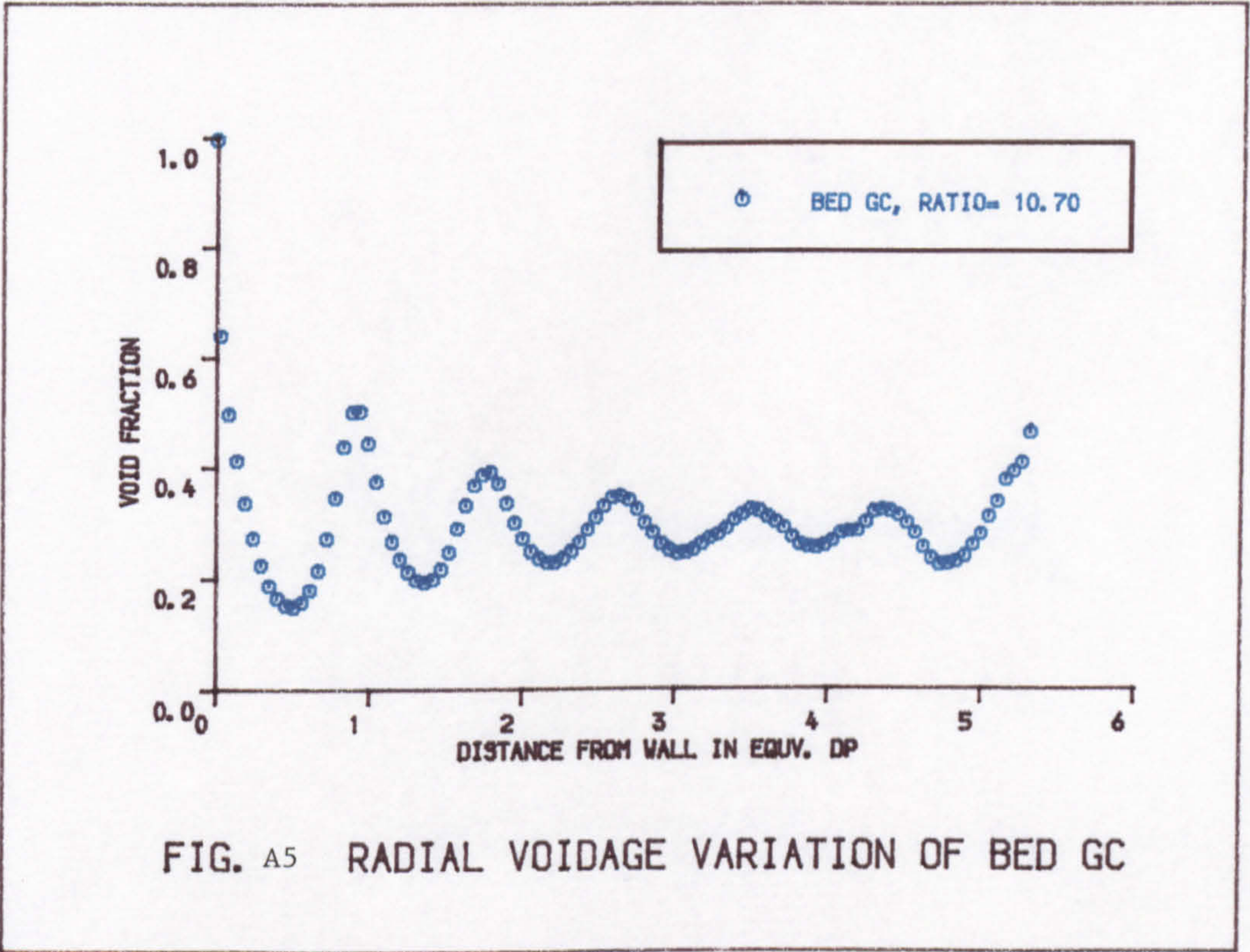
properties of packed beds, it is expected that the diameter ratio plays a role in any attempt to develop predictive pressure drop-flow correlations, especially in the wall zone of the system where channelling occurs at lower diameter ratios. The existing pressure drop correlations do not account adequately for the strong wall effect at very low diameter ratios where the wall zone occupies a significant proportion of the bed cross-section. Therefore, there is a need to re-examine the present correlations with regard to the global structural properties of packed bed systems.

APPENDIX A

Profiles of radial variation of all the test beds







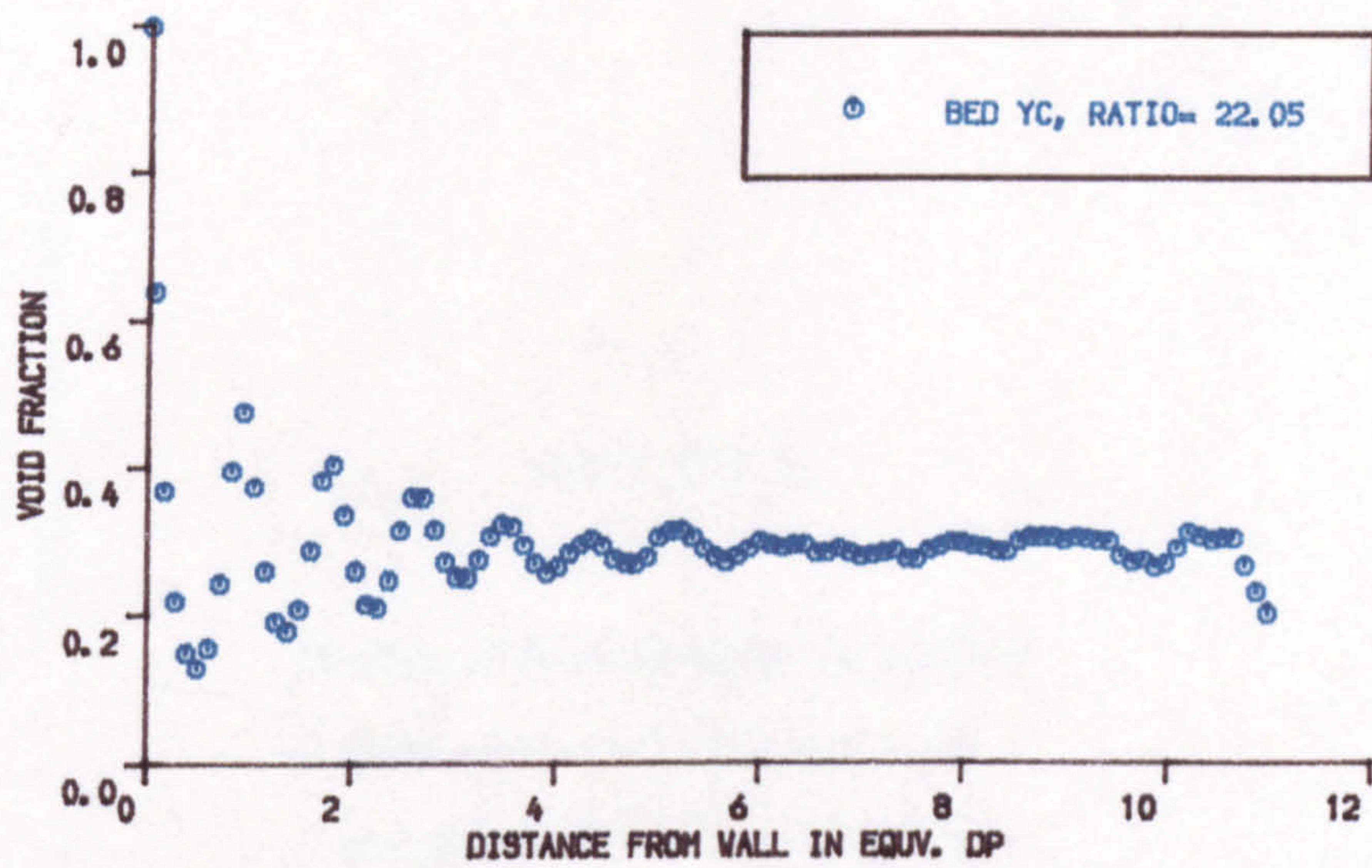


FIG. A7 RADIAL VOIDAGE VARIATION OF BED YC

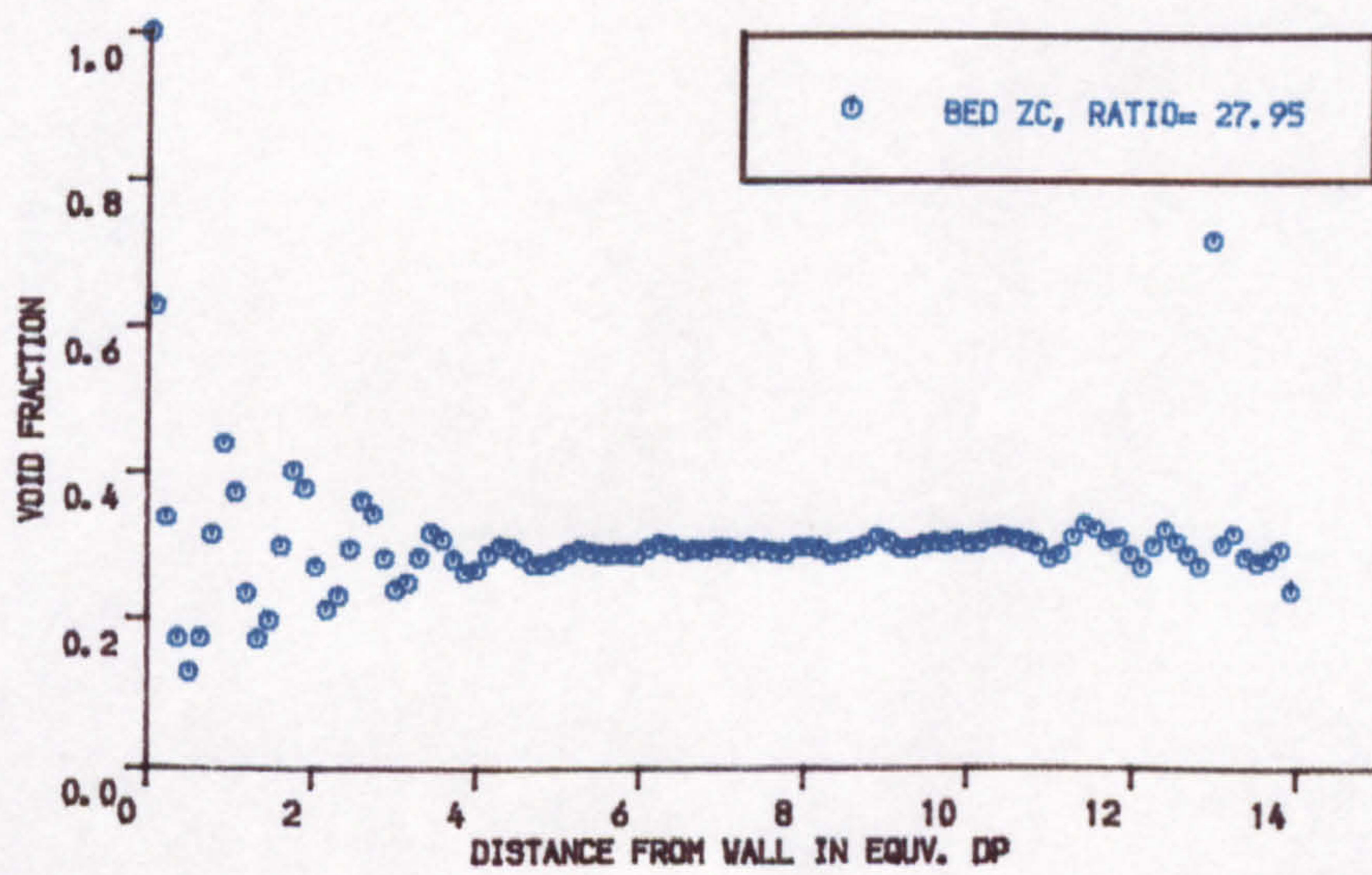
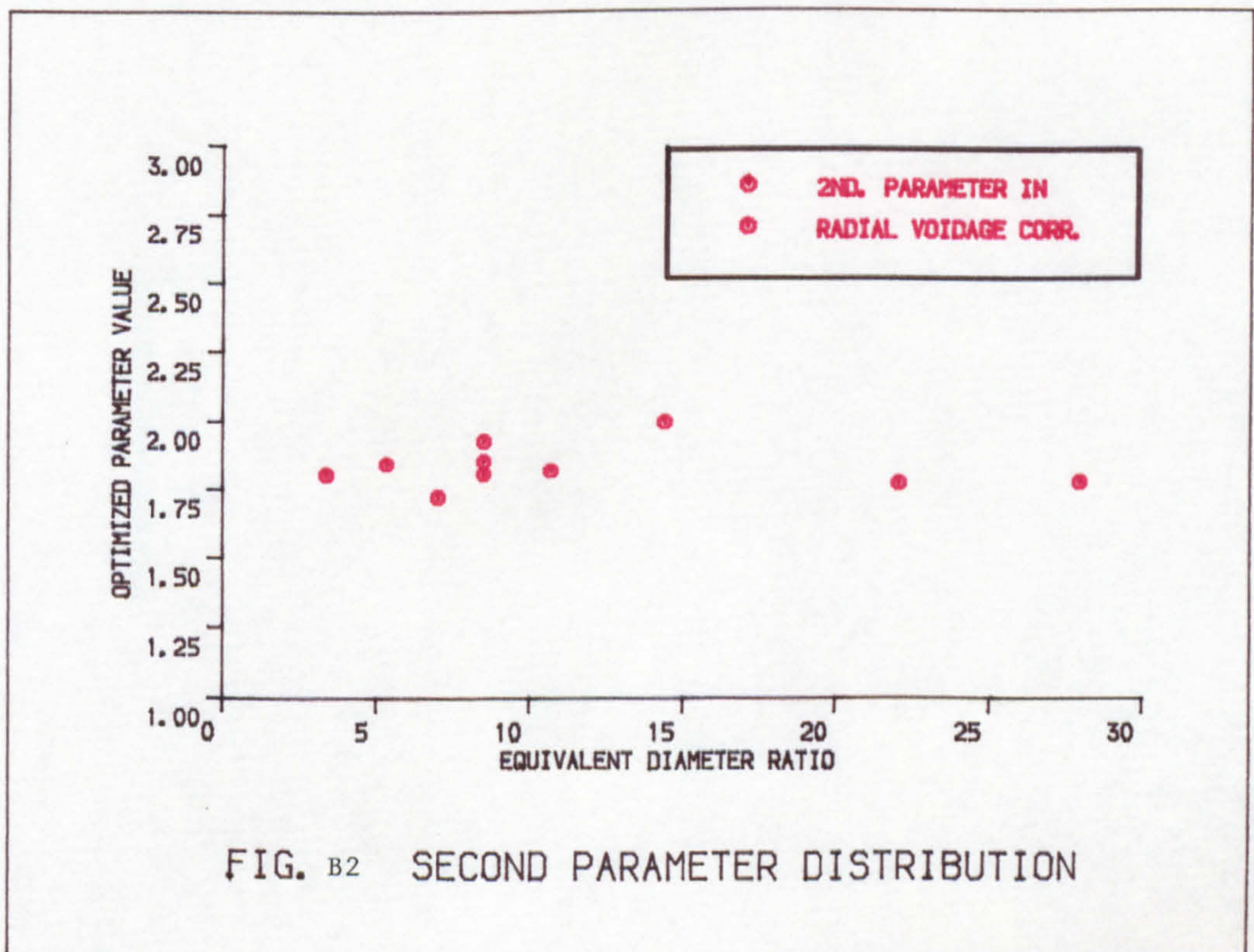
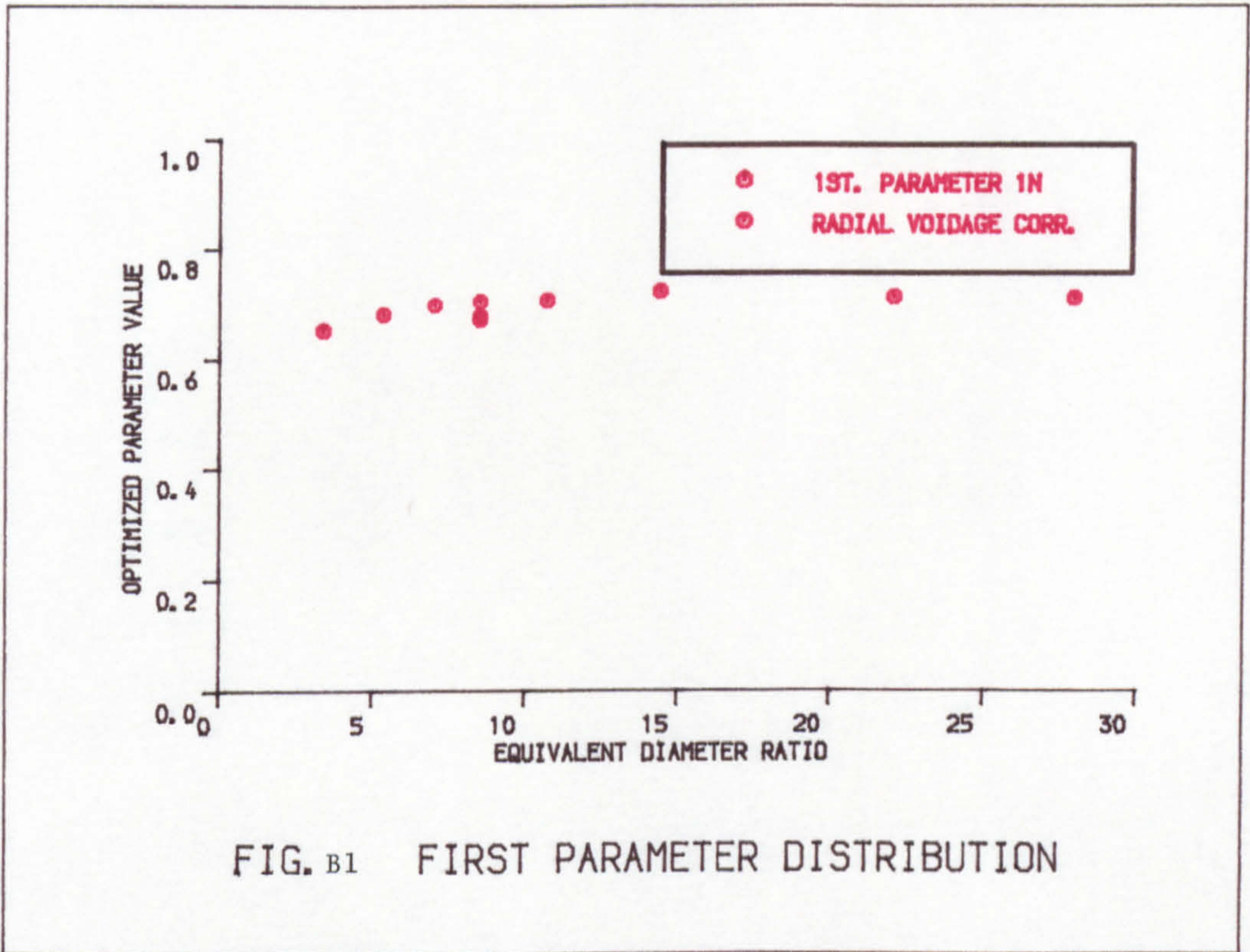


FIG. A8 RADIAL VOIDAGE VARIATION OF BED ZC

APPENDIX B

**Profiles of the individual correlation
parameters for radial and axial
variations of voidage against the
diameter of the packed beds**



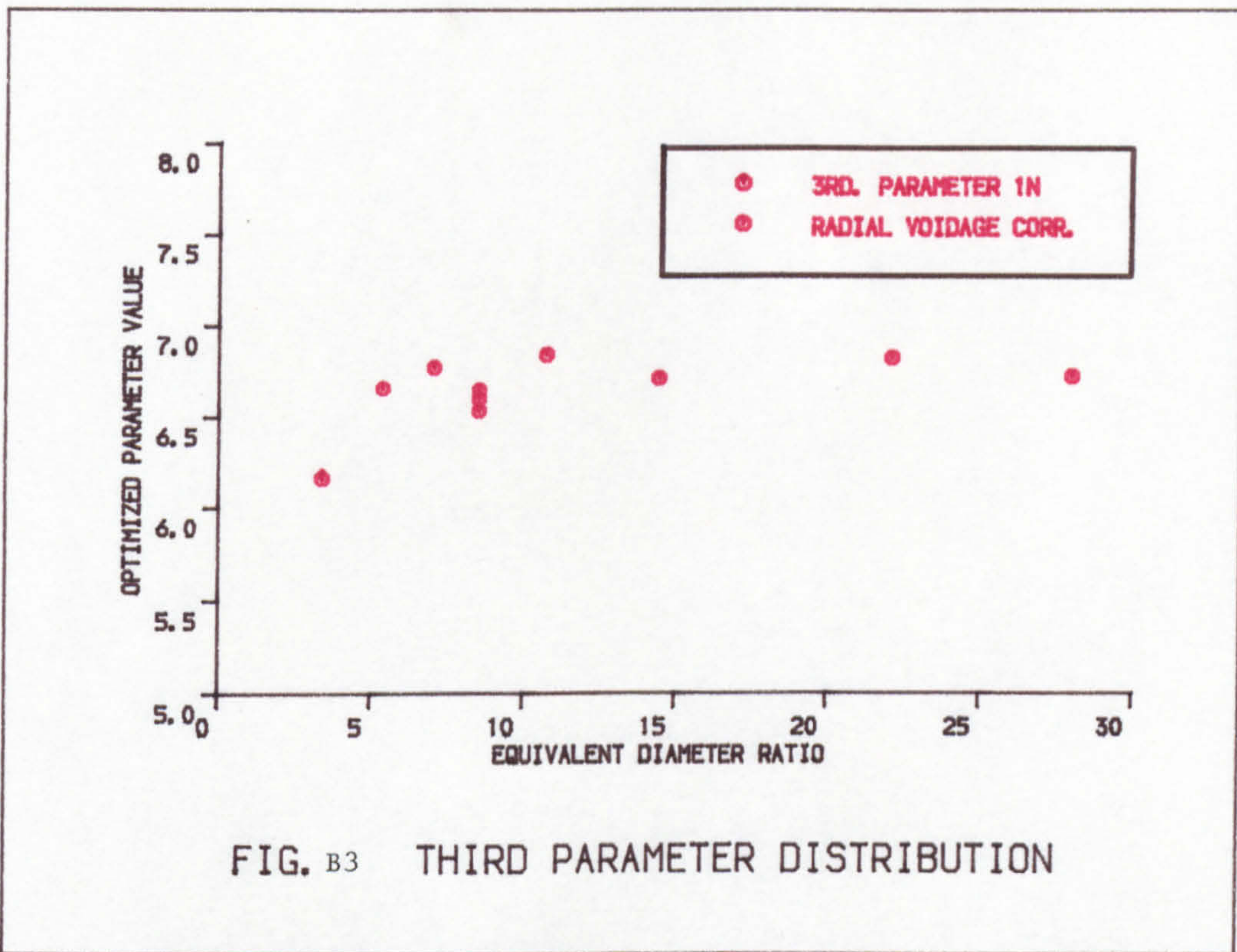


FIG. B3 THIRD PARAMETER DISTRIBUTION

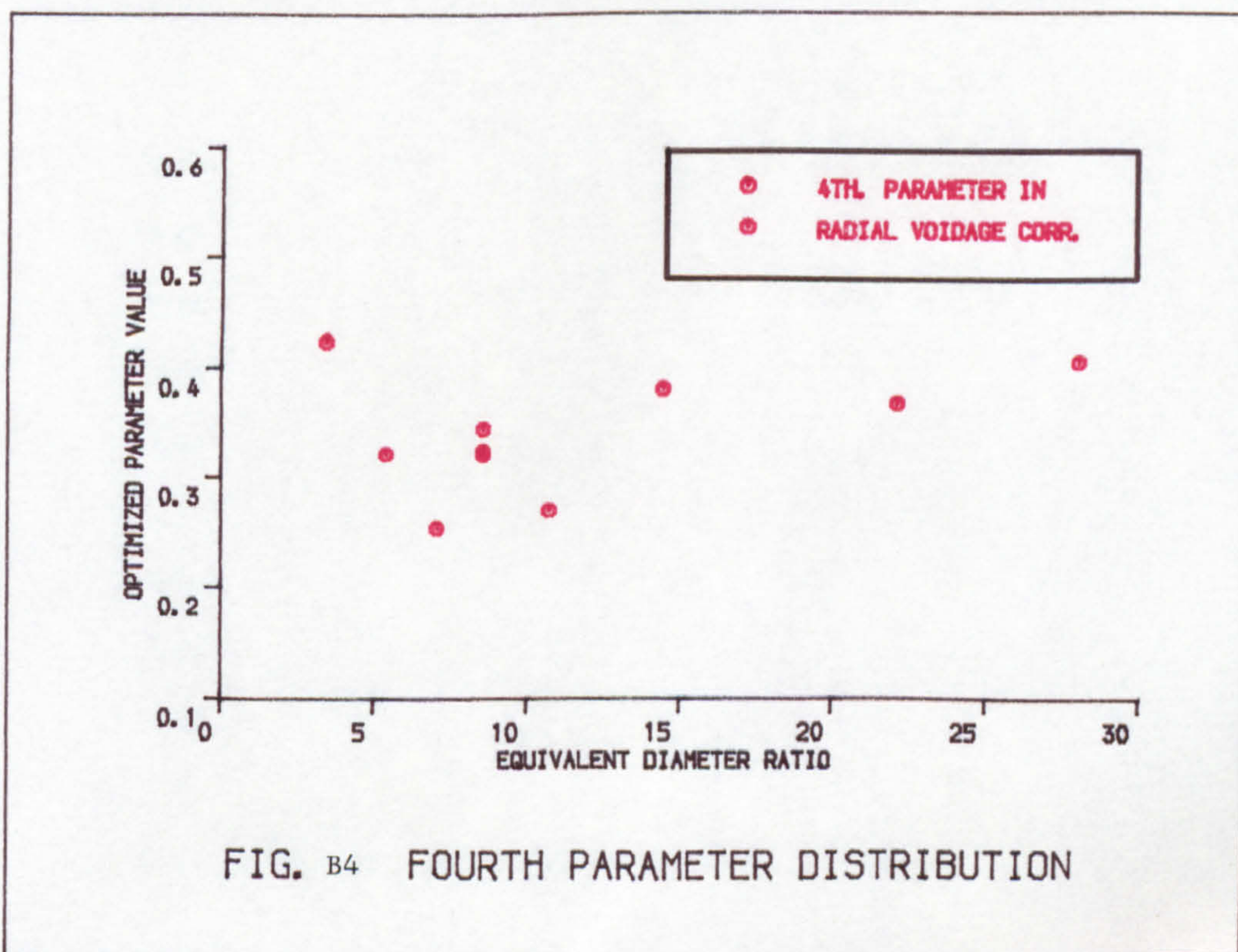
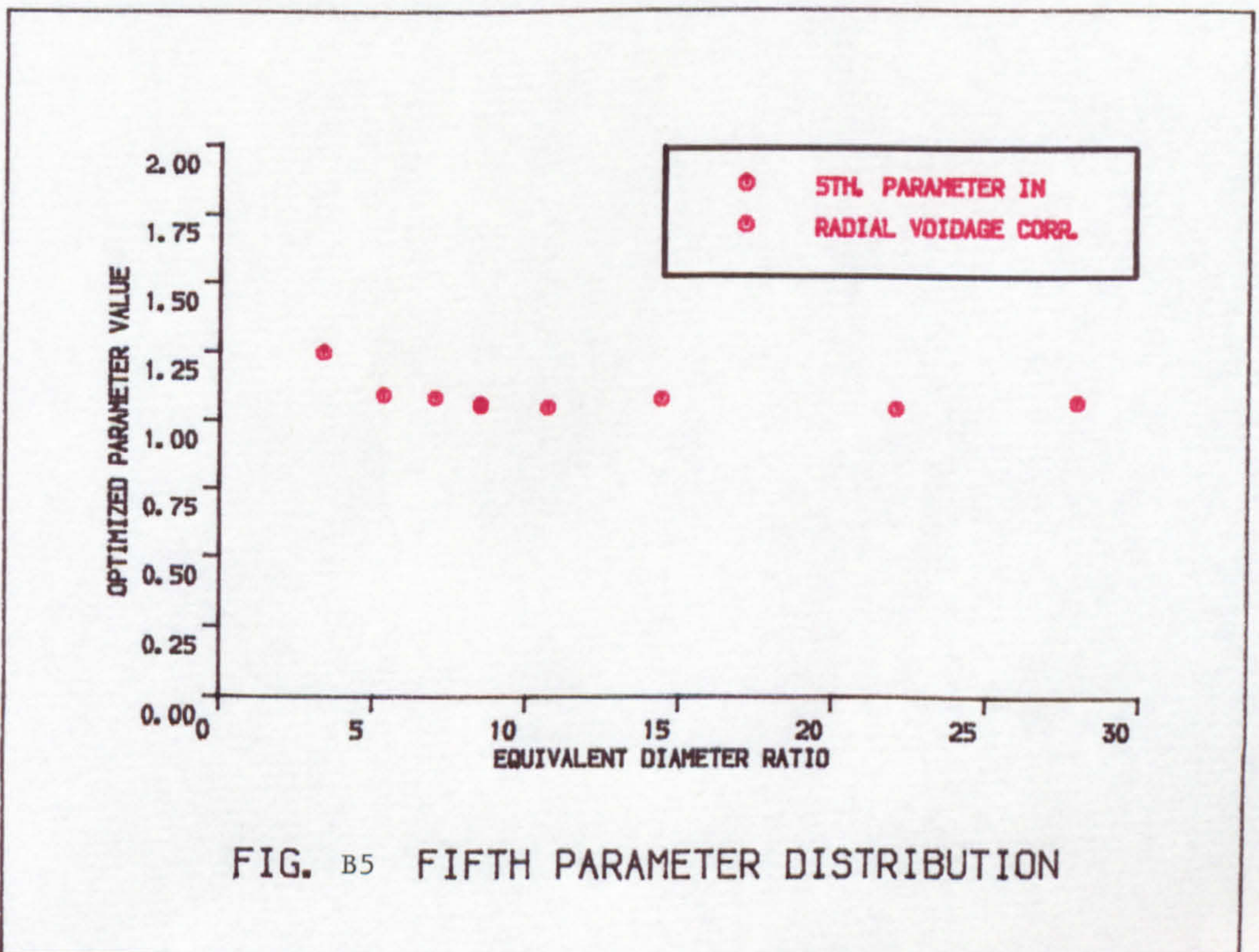


FIG. B4 FOURTH PARAMETER DISTRIBUTION



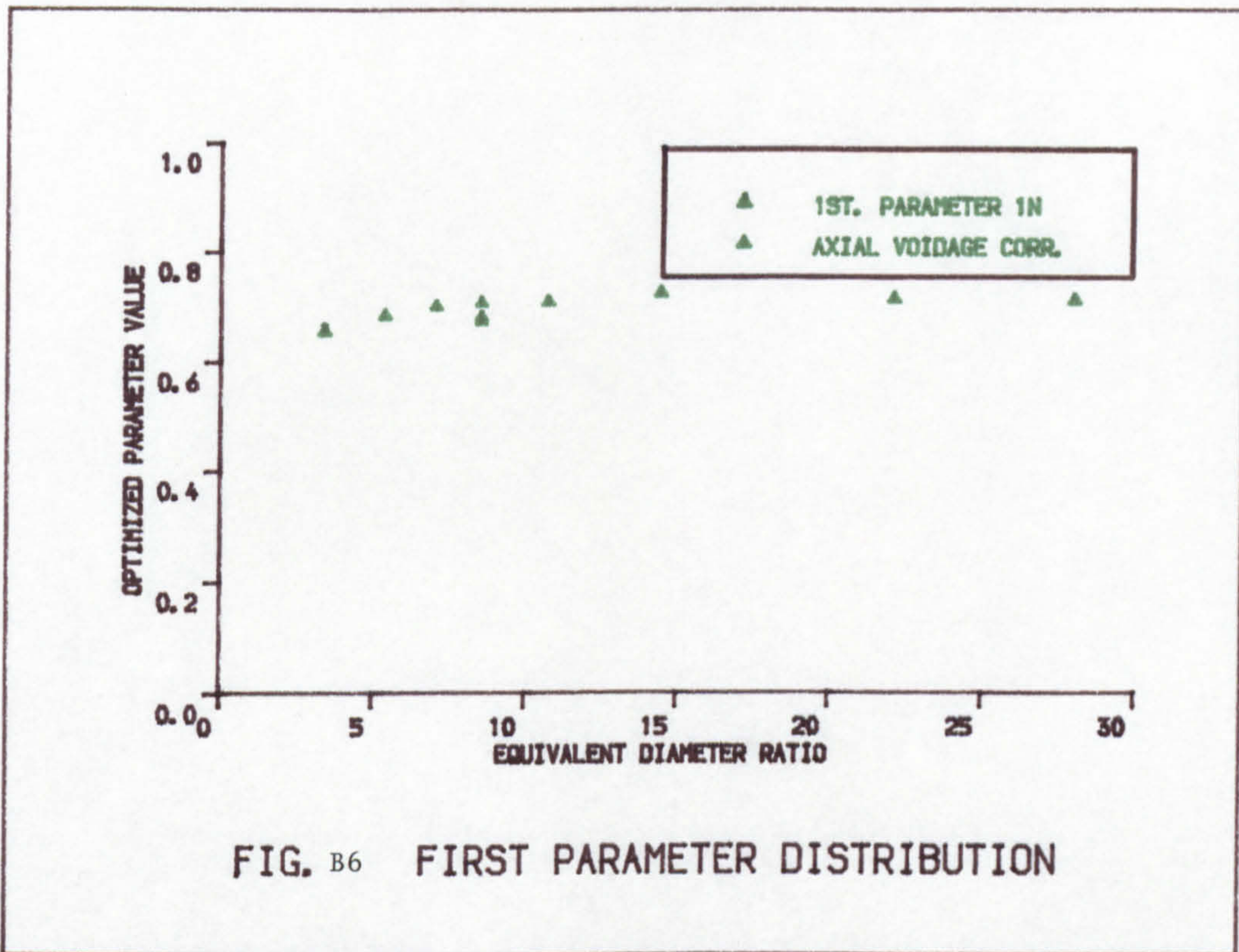


FIG. B6 FIRST PARAMETER DISTRIBUTION

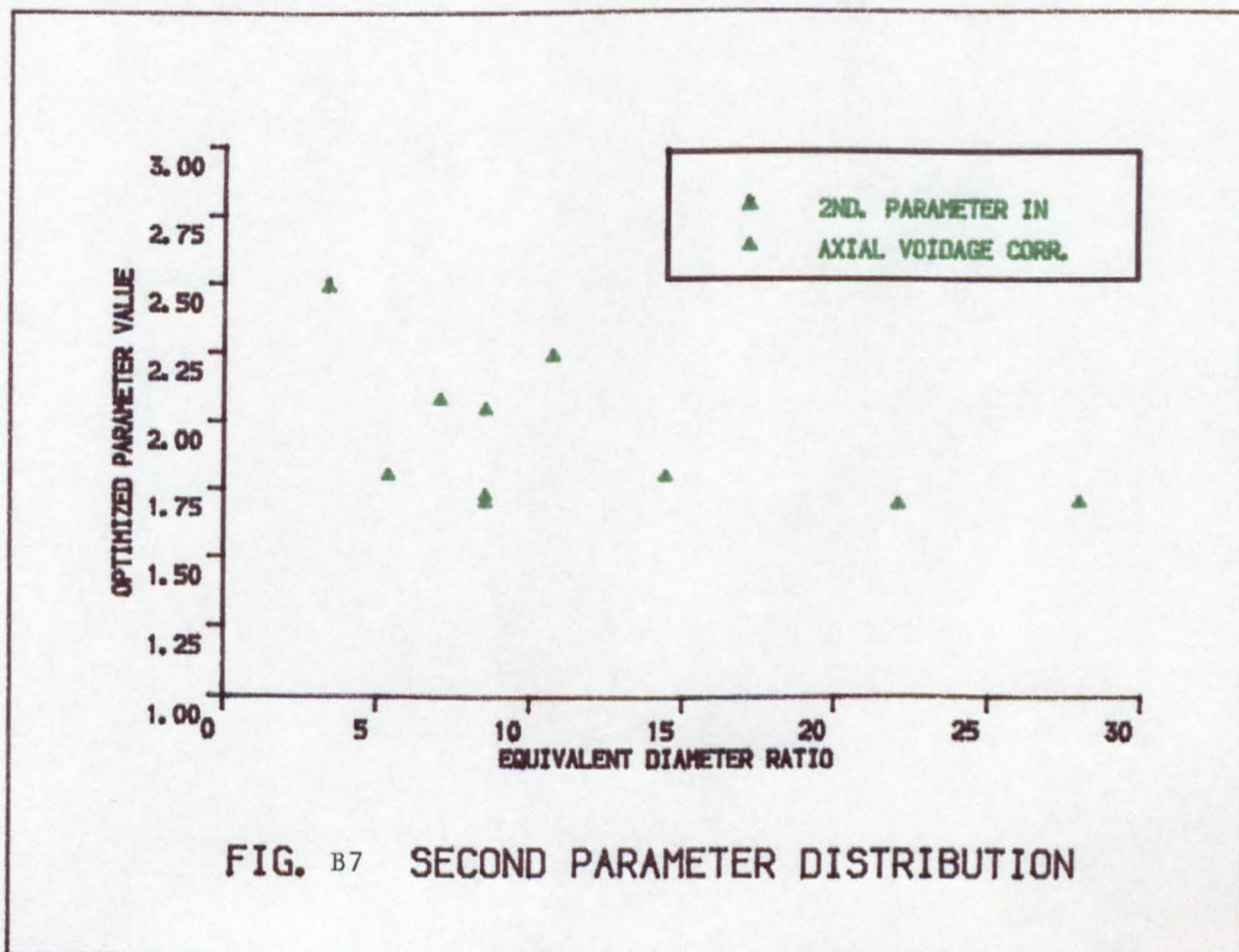


FIG. B7 SECOND PARAMETER DISTRIBUTION

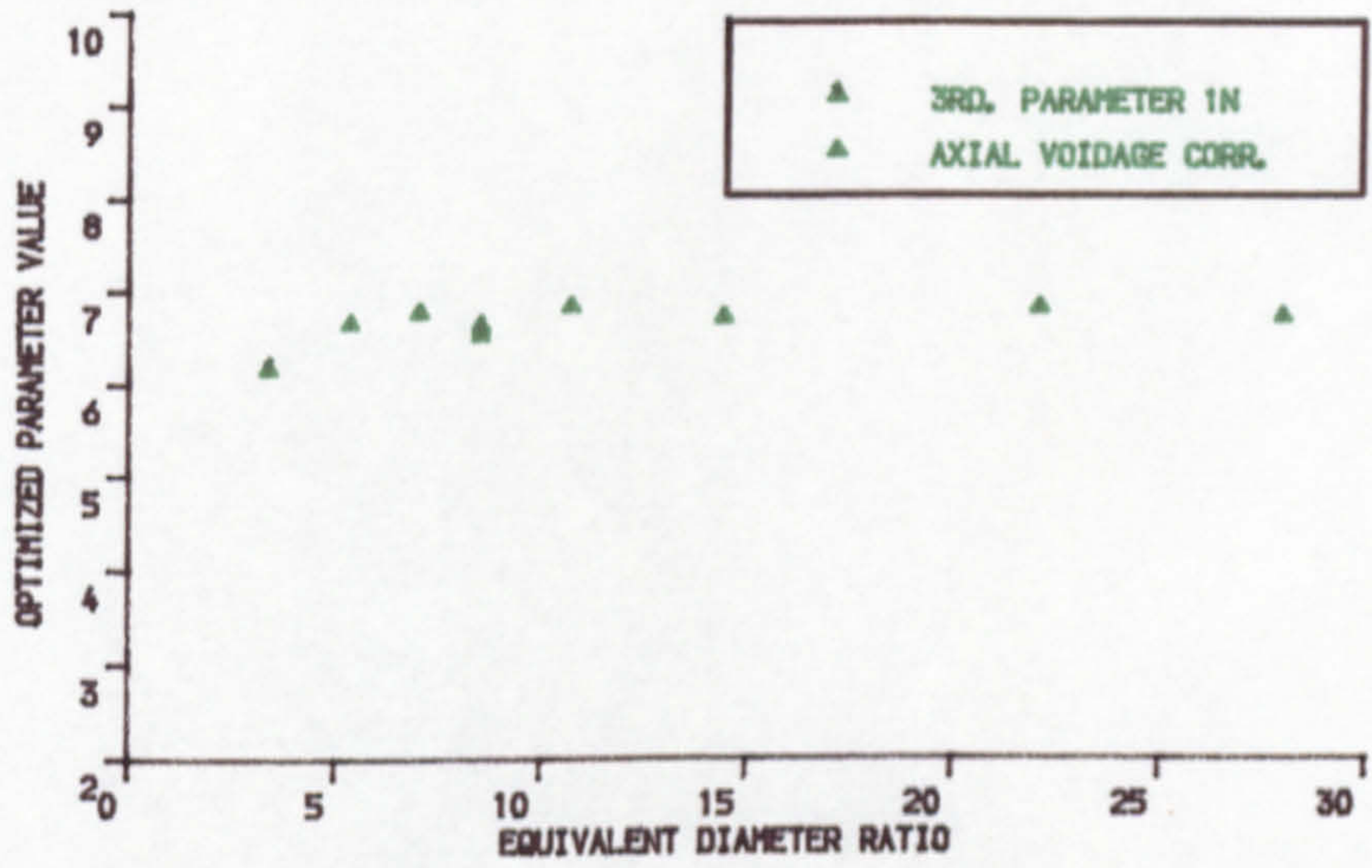


FIG. B8 THIRD PARAMETER DISTRIBUTION

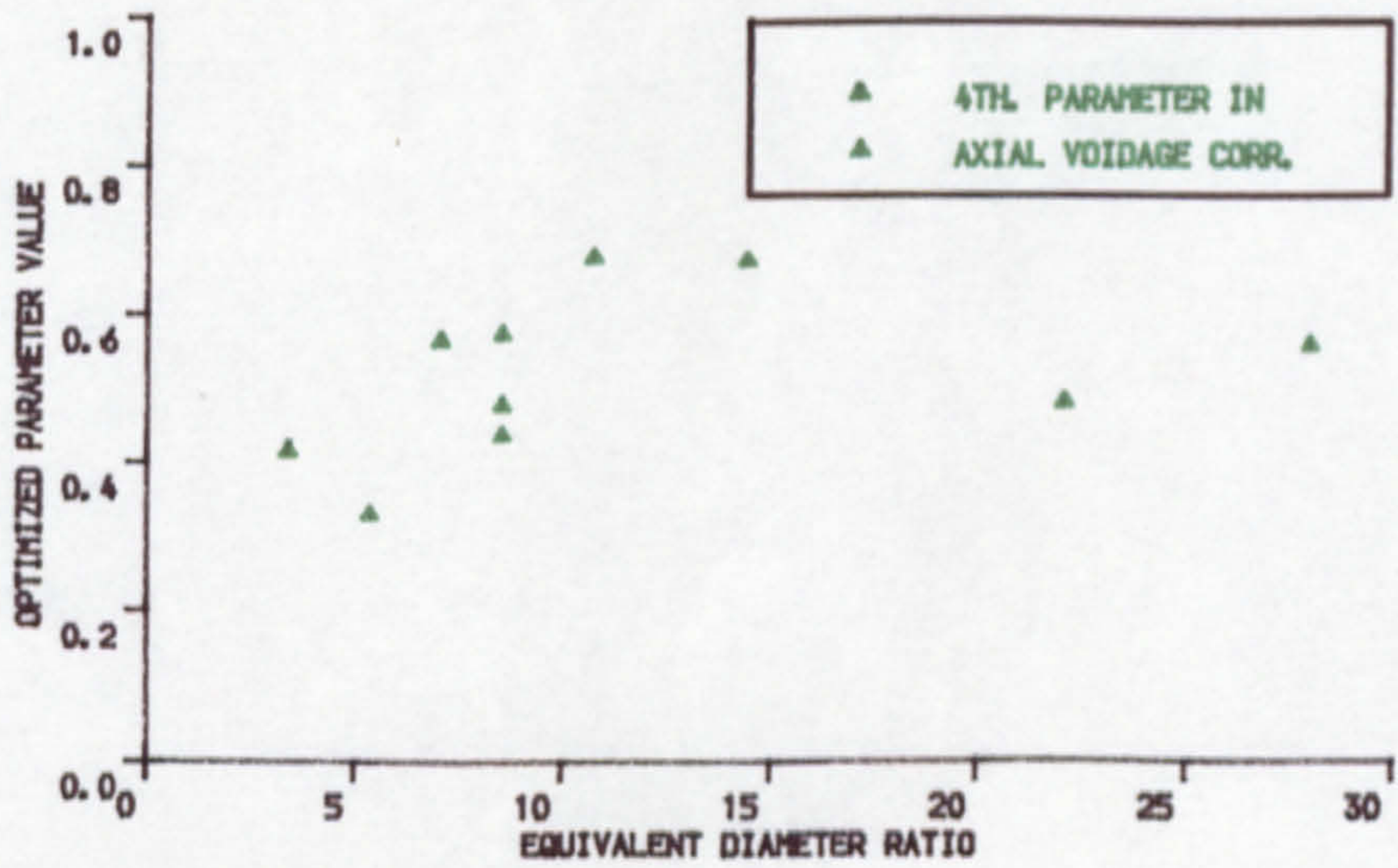


FIG. B9 FOURTH PARAMETER DISTRIBUTION

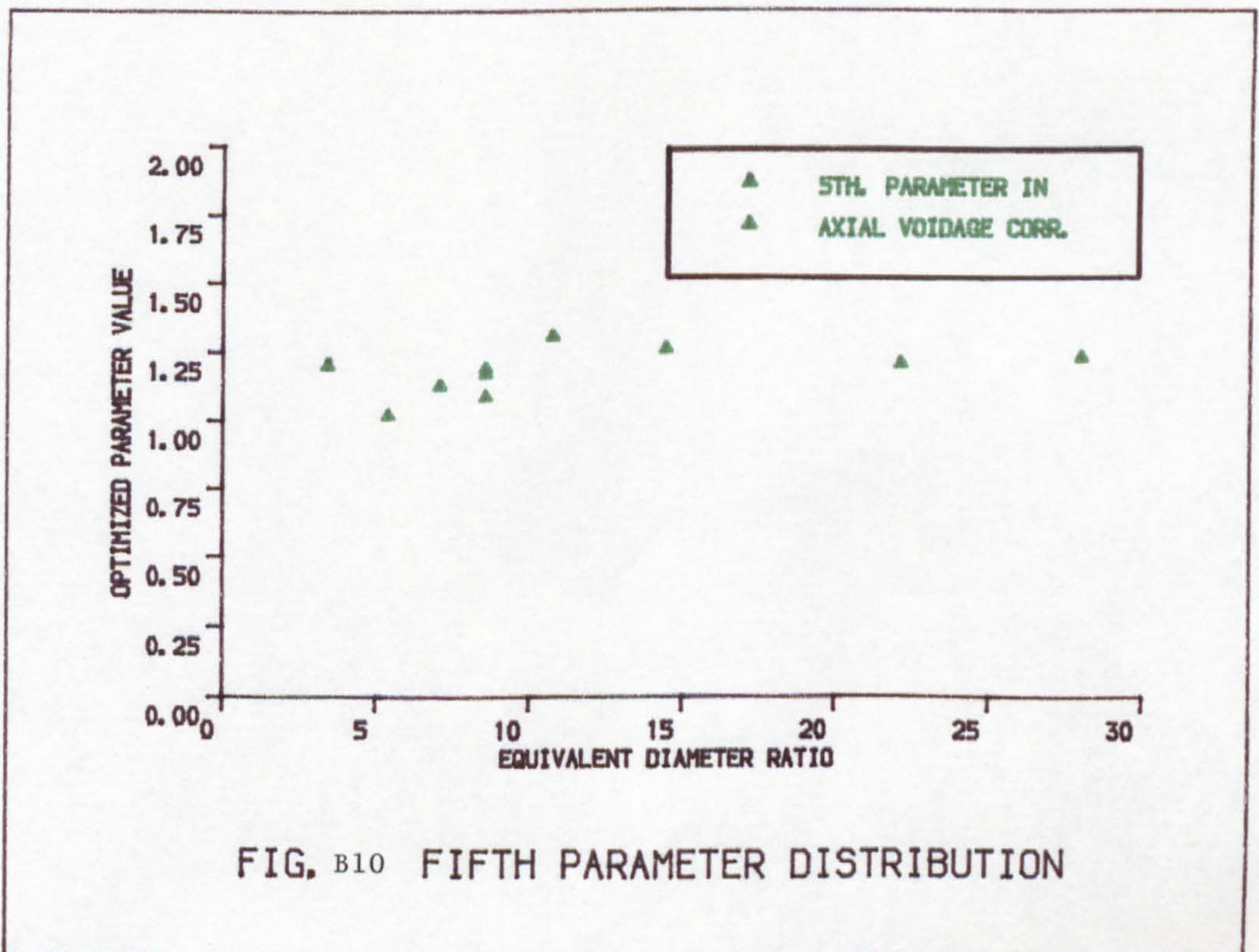
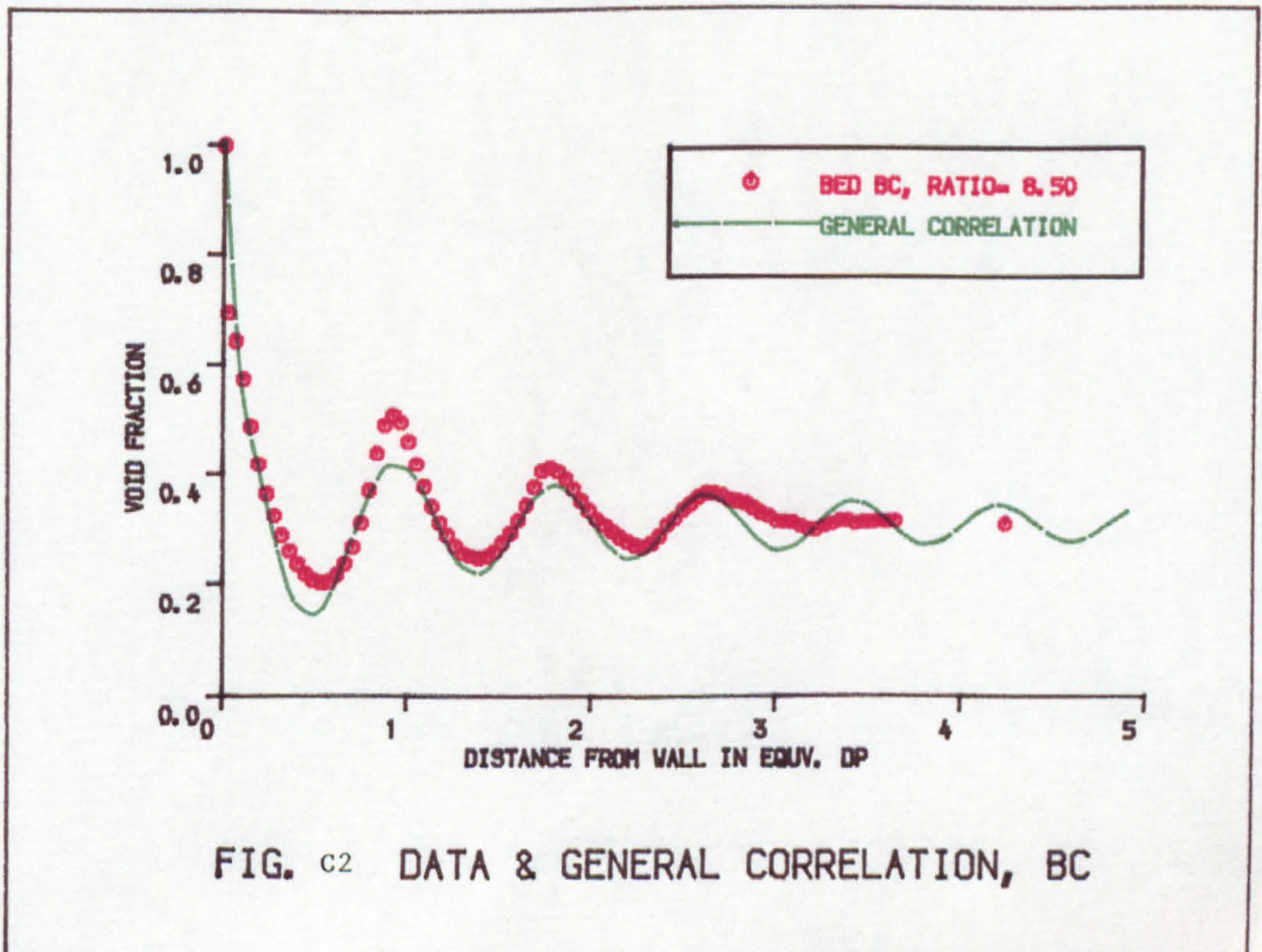
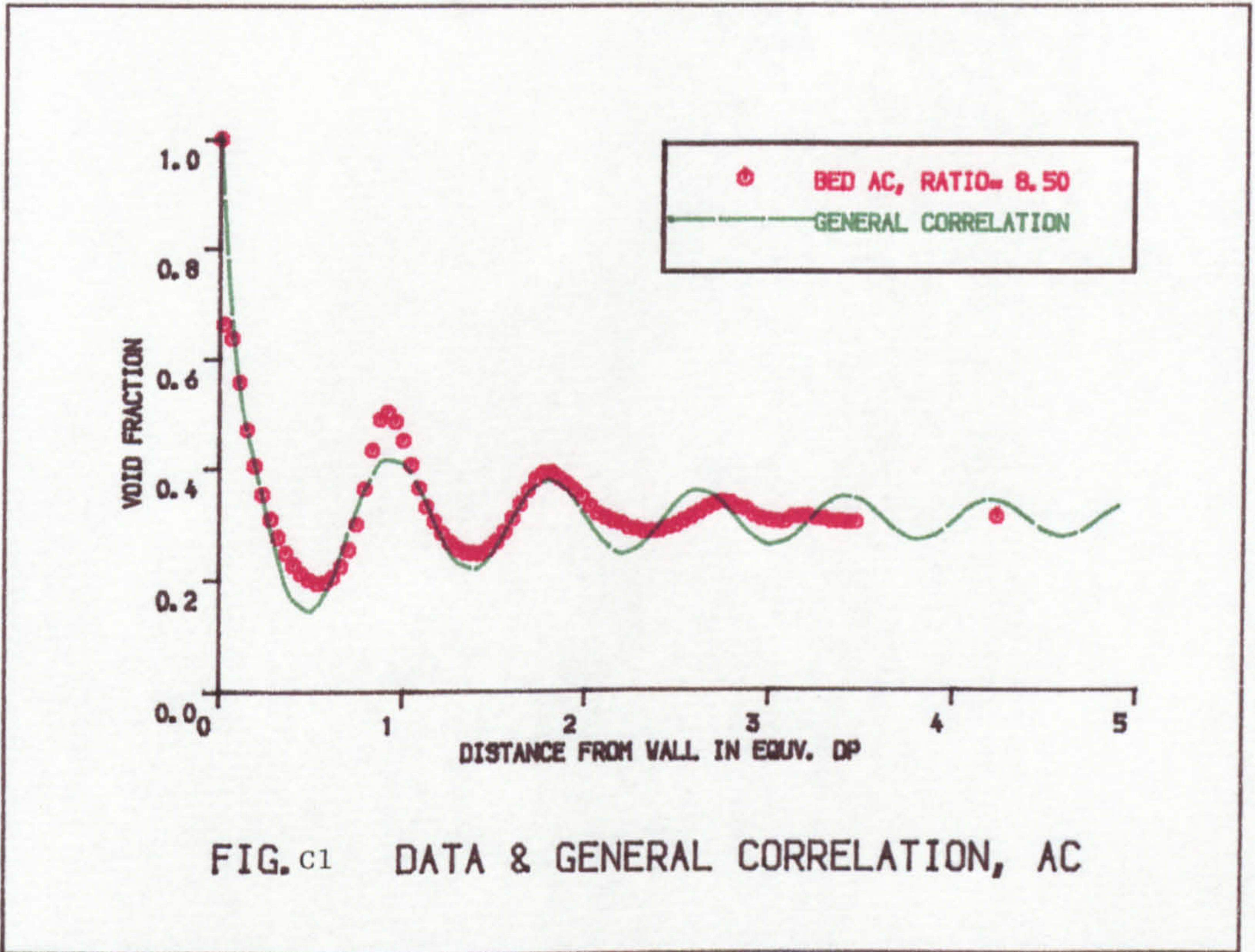


FIG. B10 FIFTH PARAMETER DISTRIBUTION

APPENDIX C

**Profiles of the simplified versions
of the general radial and axial
voidage correlations of each test bed**



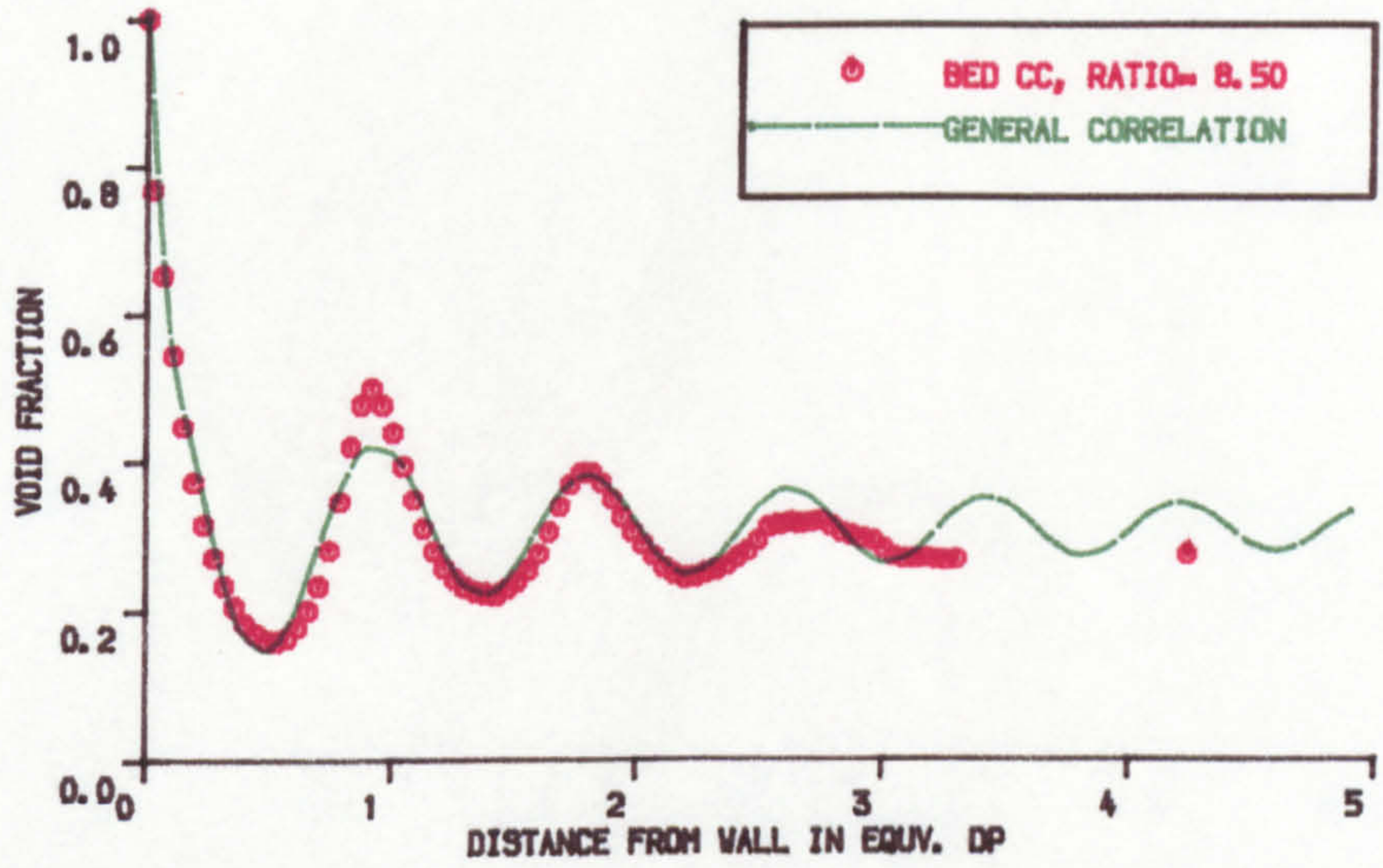


FIG. c3 DATA & GENERAL CORRELATION, CC

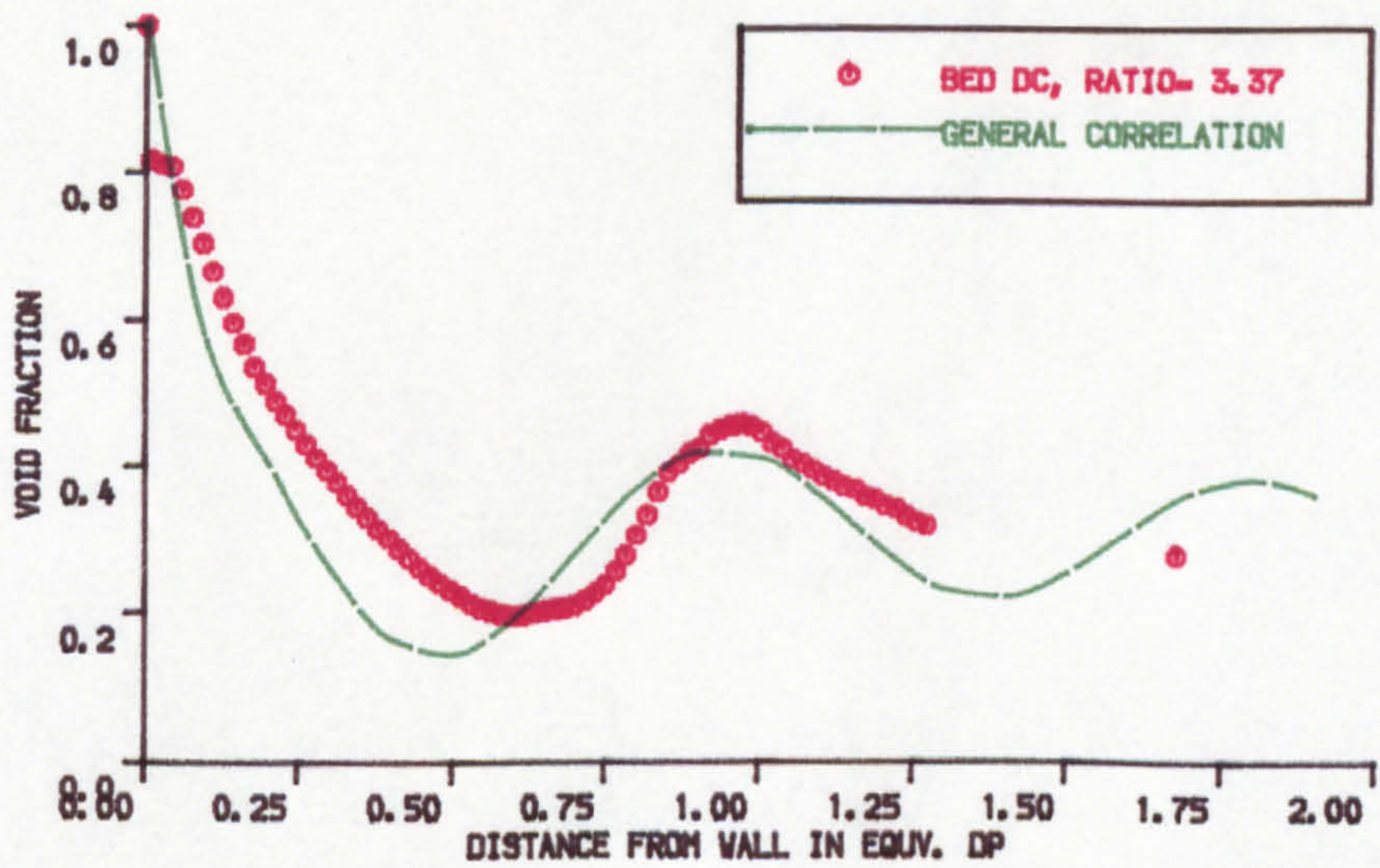


FIG. c4 DATA & GENERAL CORRELATION, DC

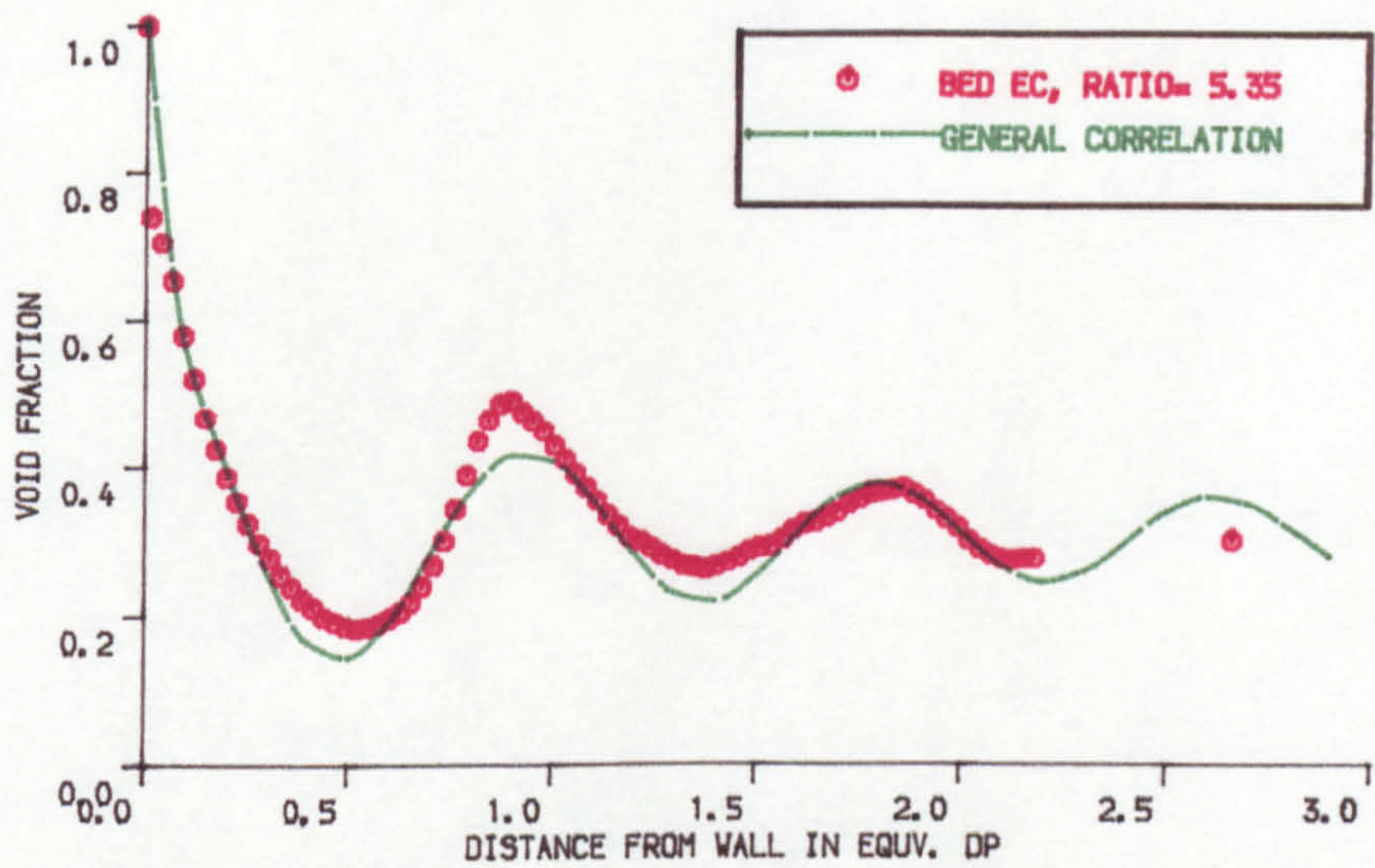


FIG. c5 DATA & GENERAL CORRELATION, EC

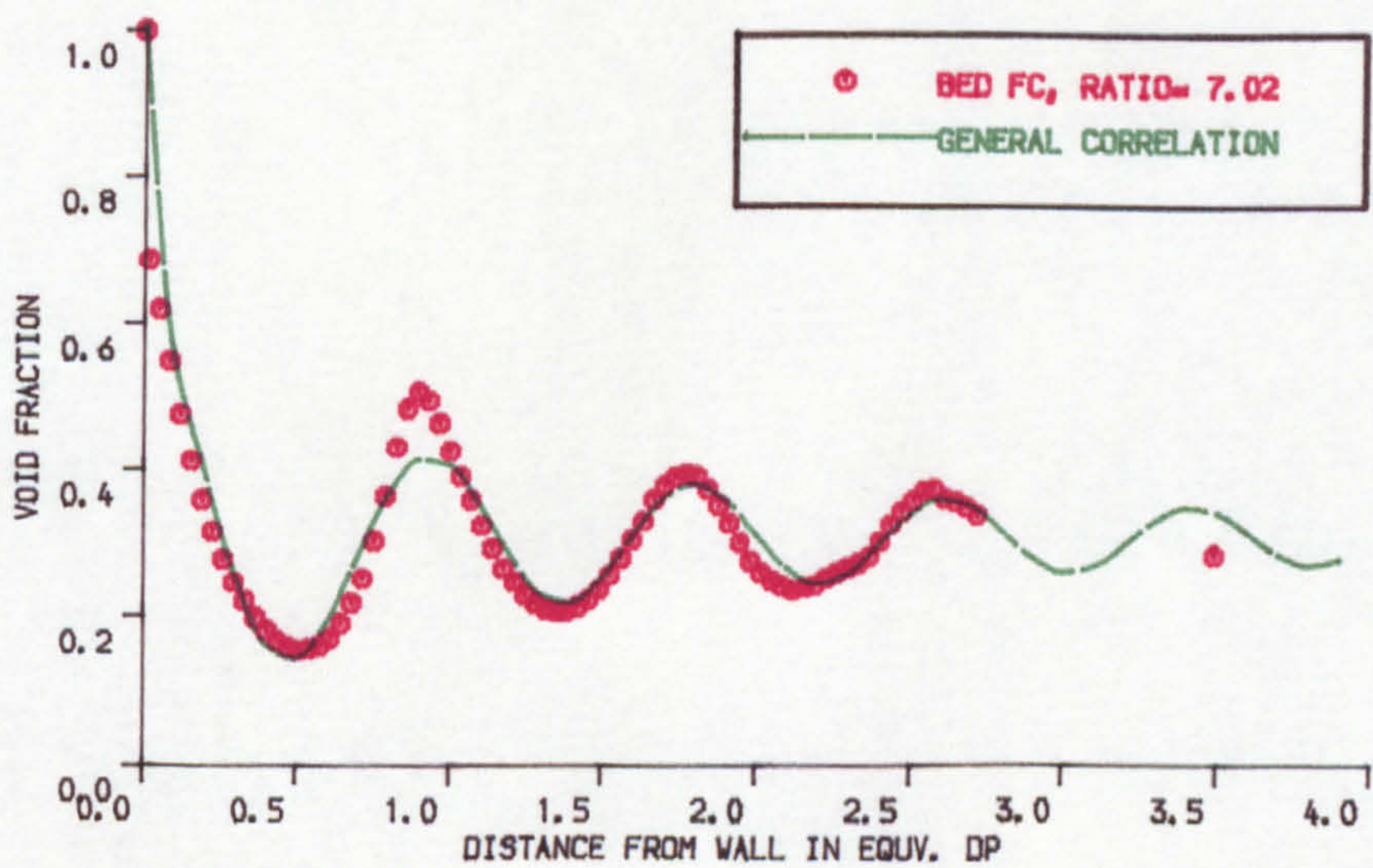


FIG. c6 DATA & GENERAL CORRELATION, FC

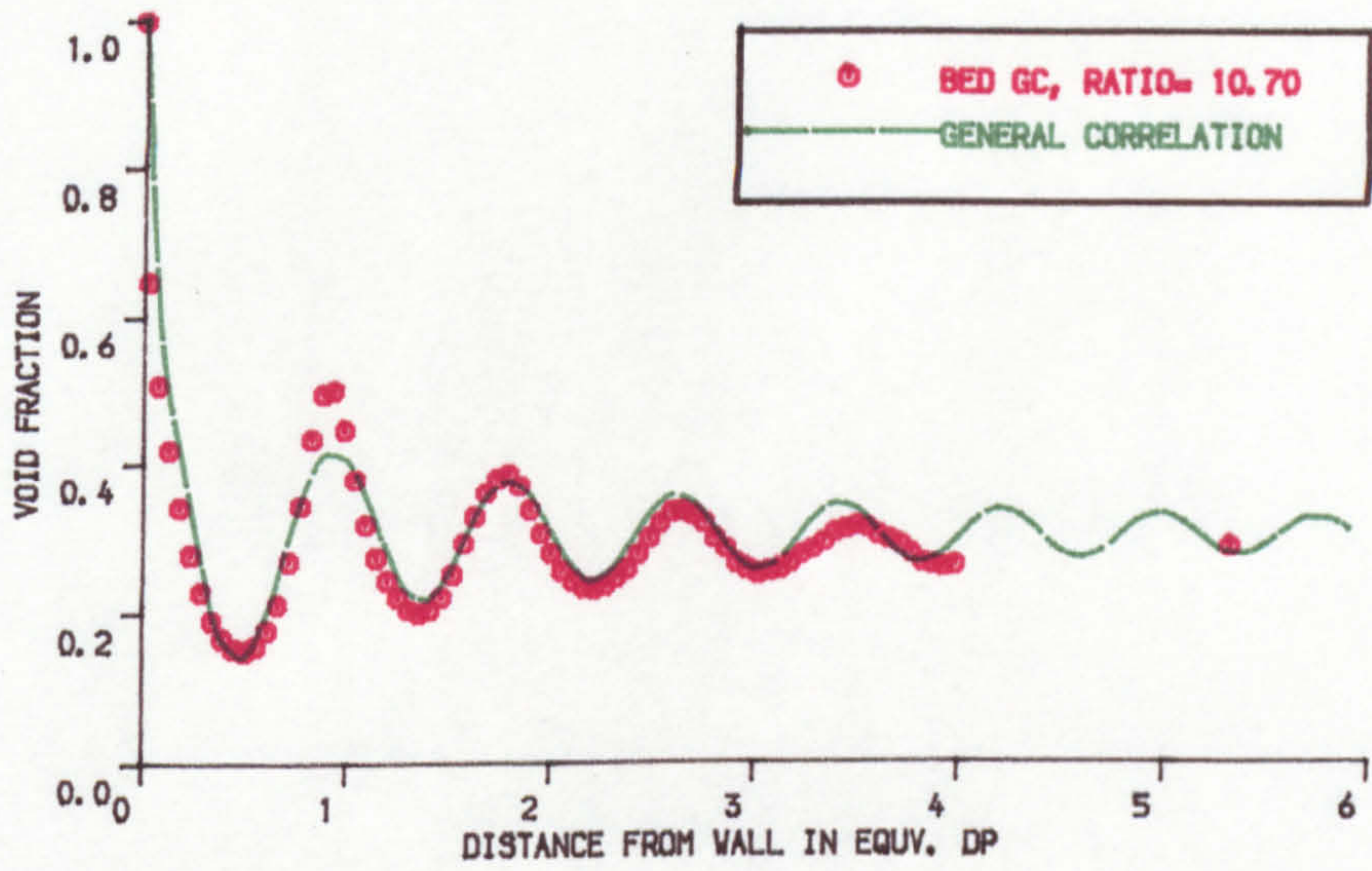


FIG. c7 DATA & GENERAL CORRELATION, GC

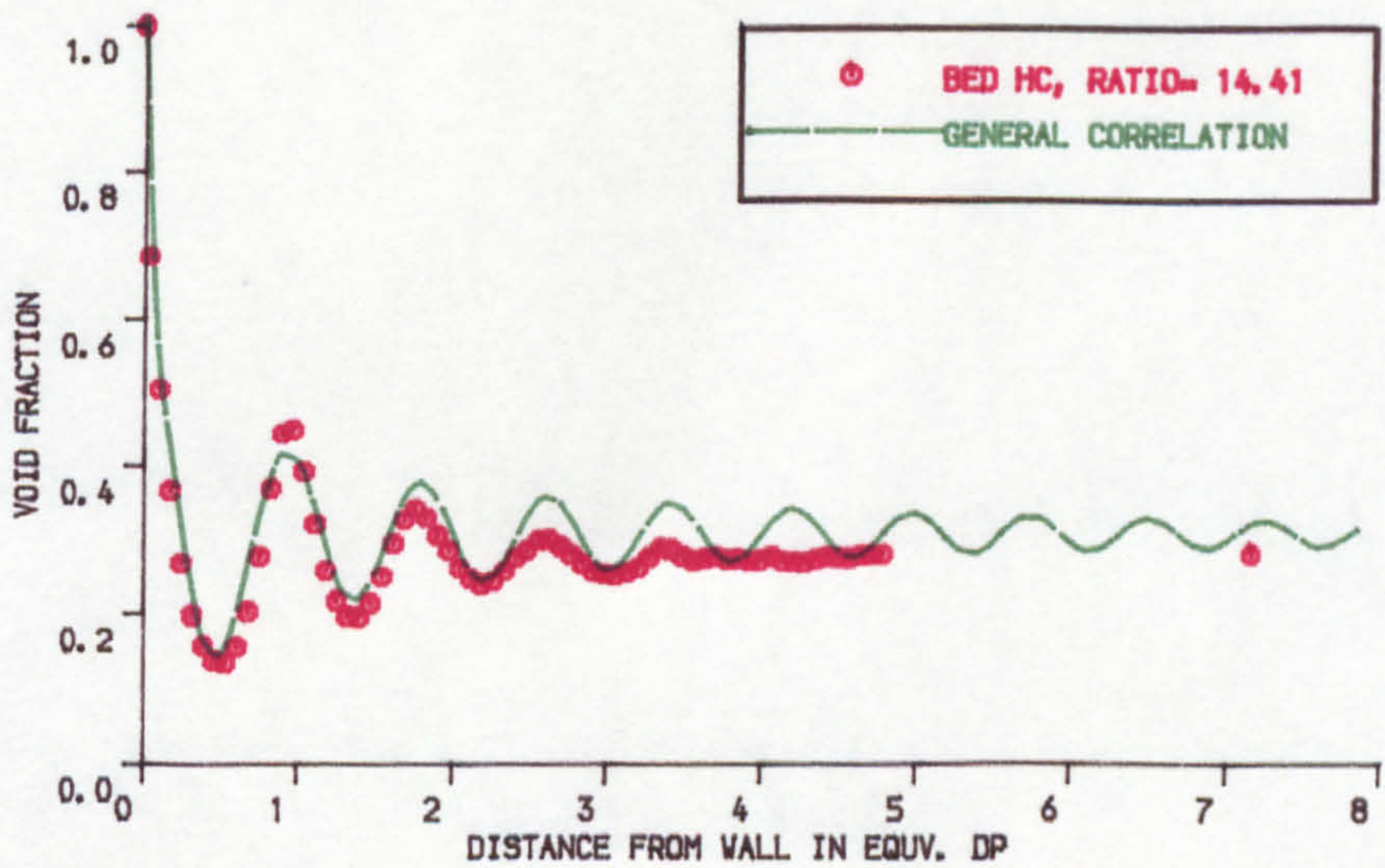
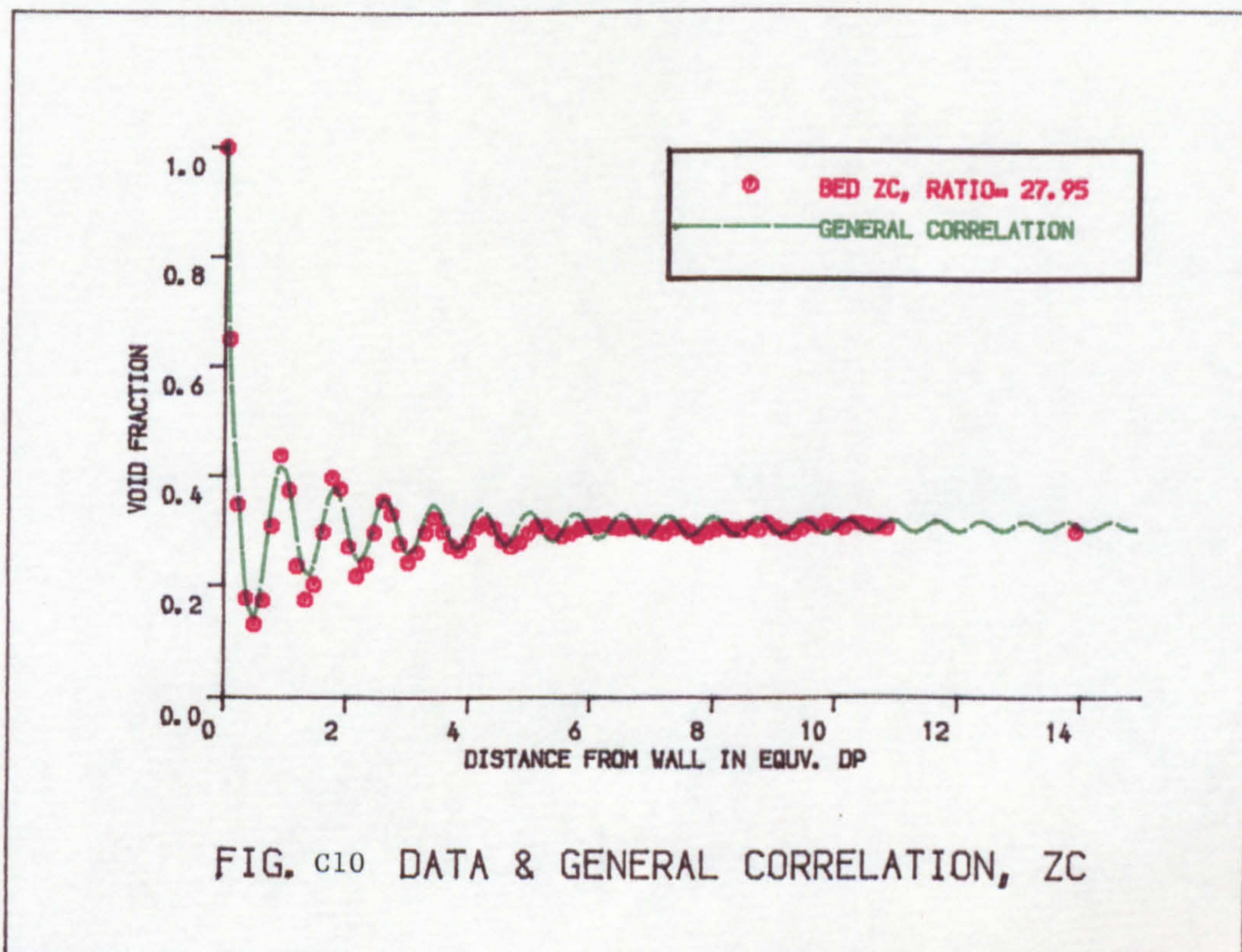
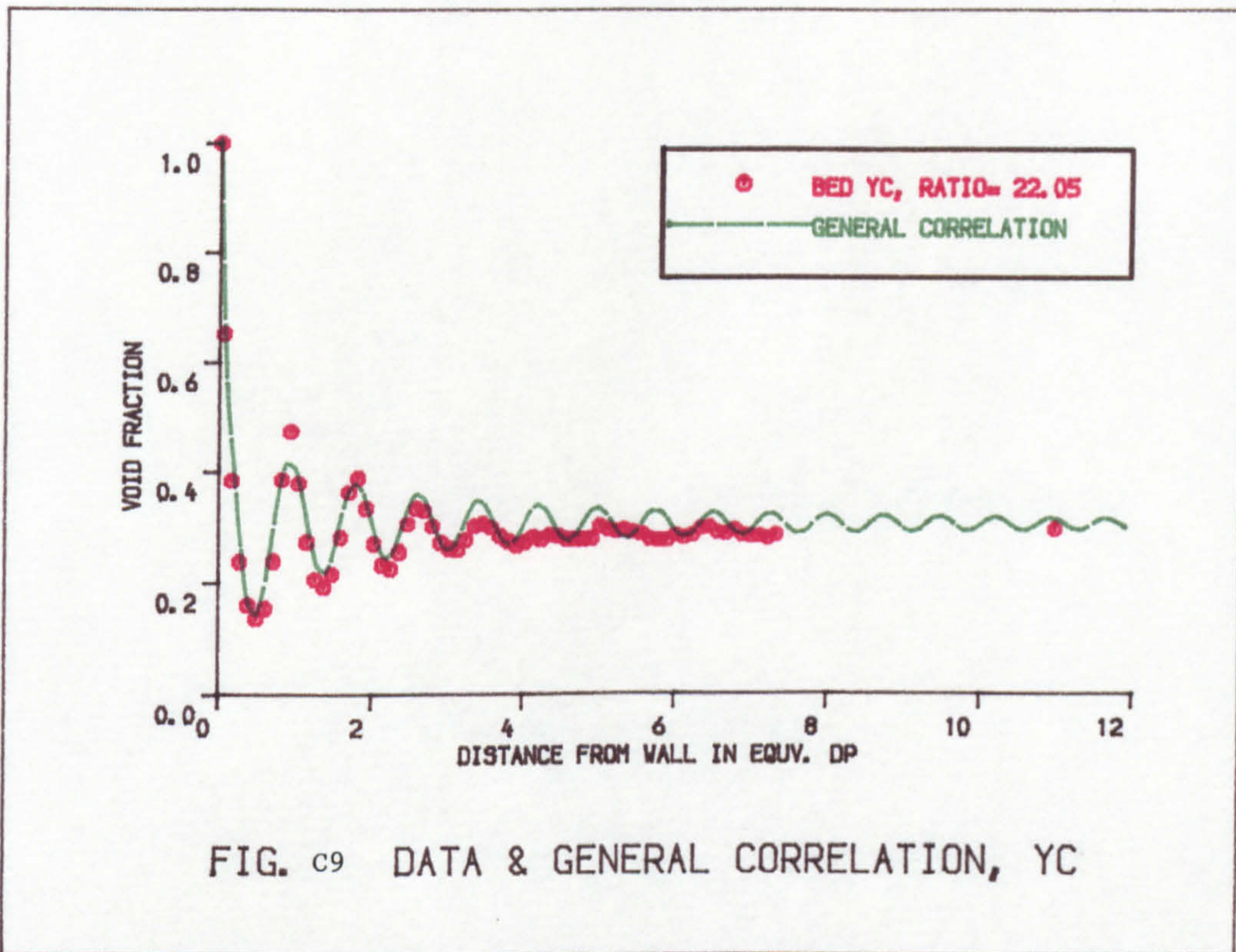
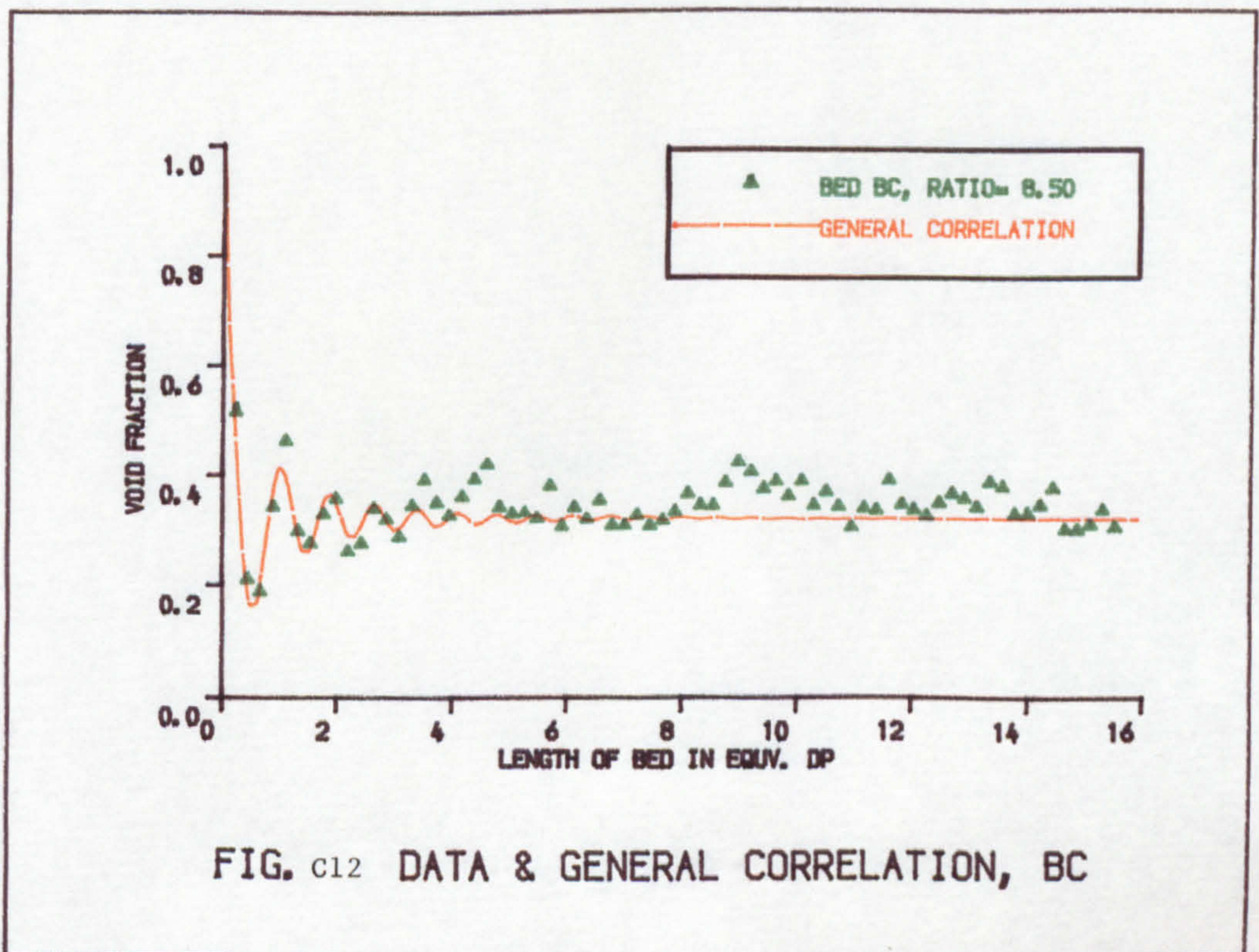
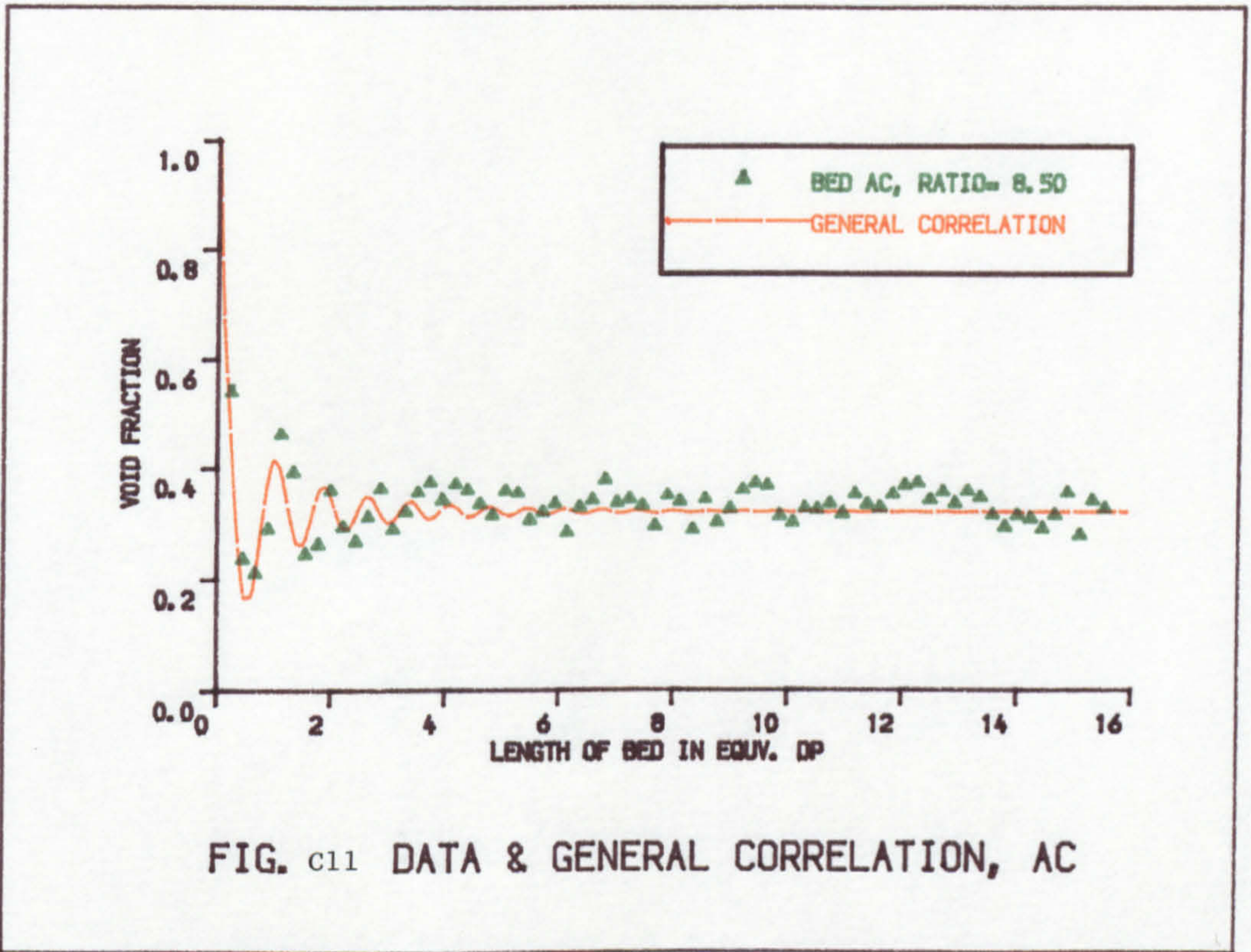


FIG. c8 DATA & GENERAL CORRELATION, HC





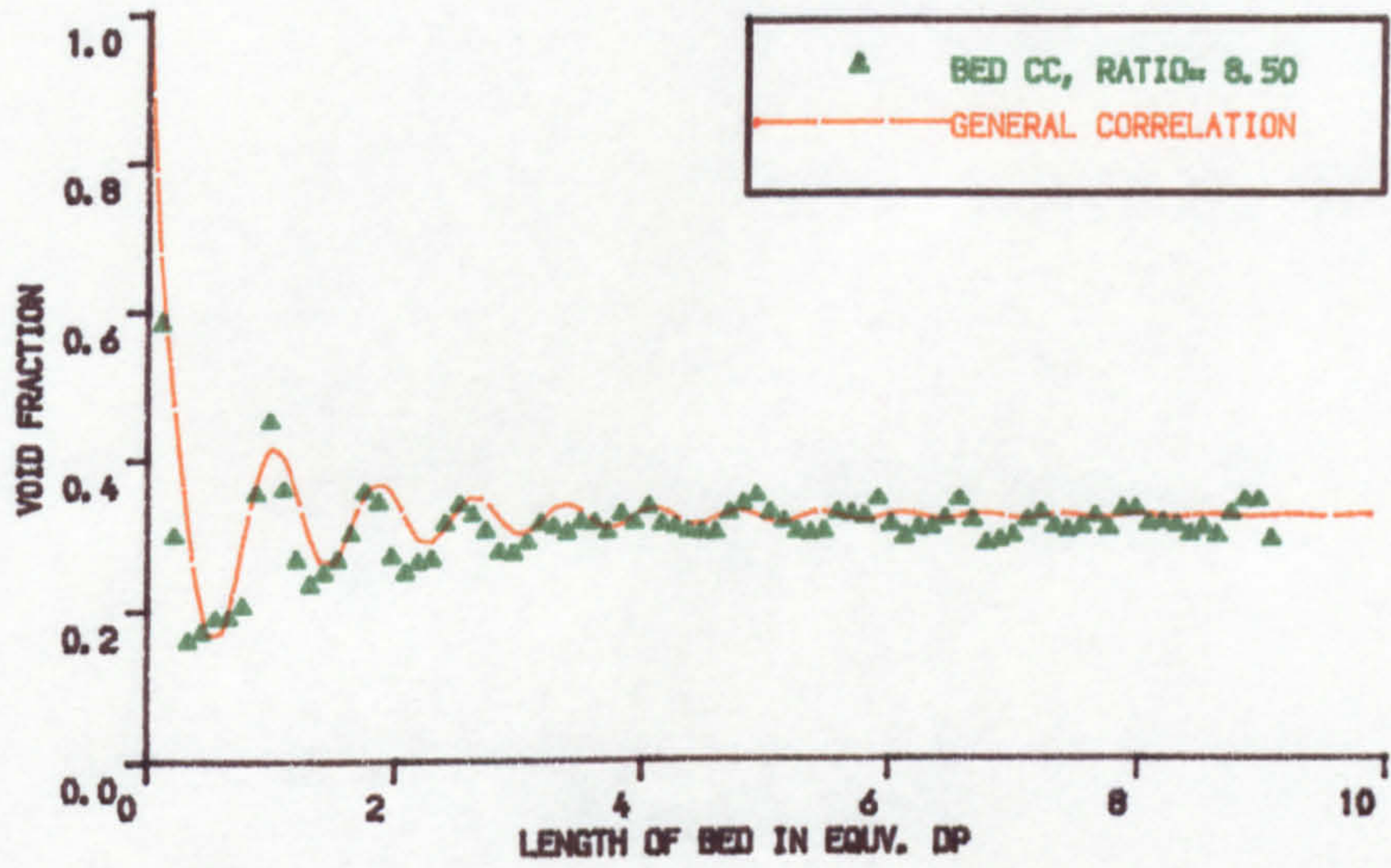


FIG. C13 DATA & GENERAL CORRELATION, CC

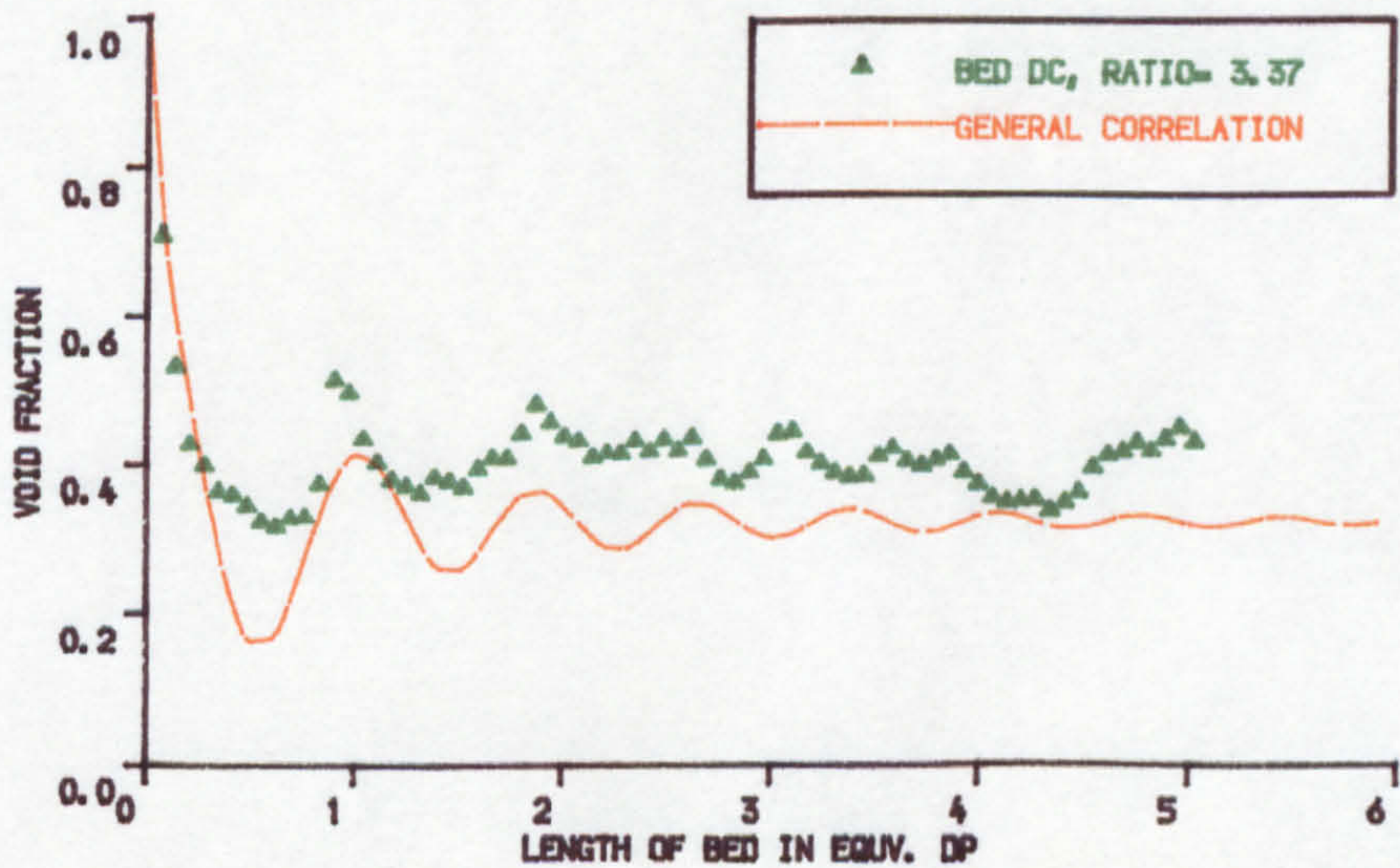


FIG. C14 DATA & GENERAL CORRELATION, DC

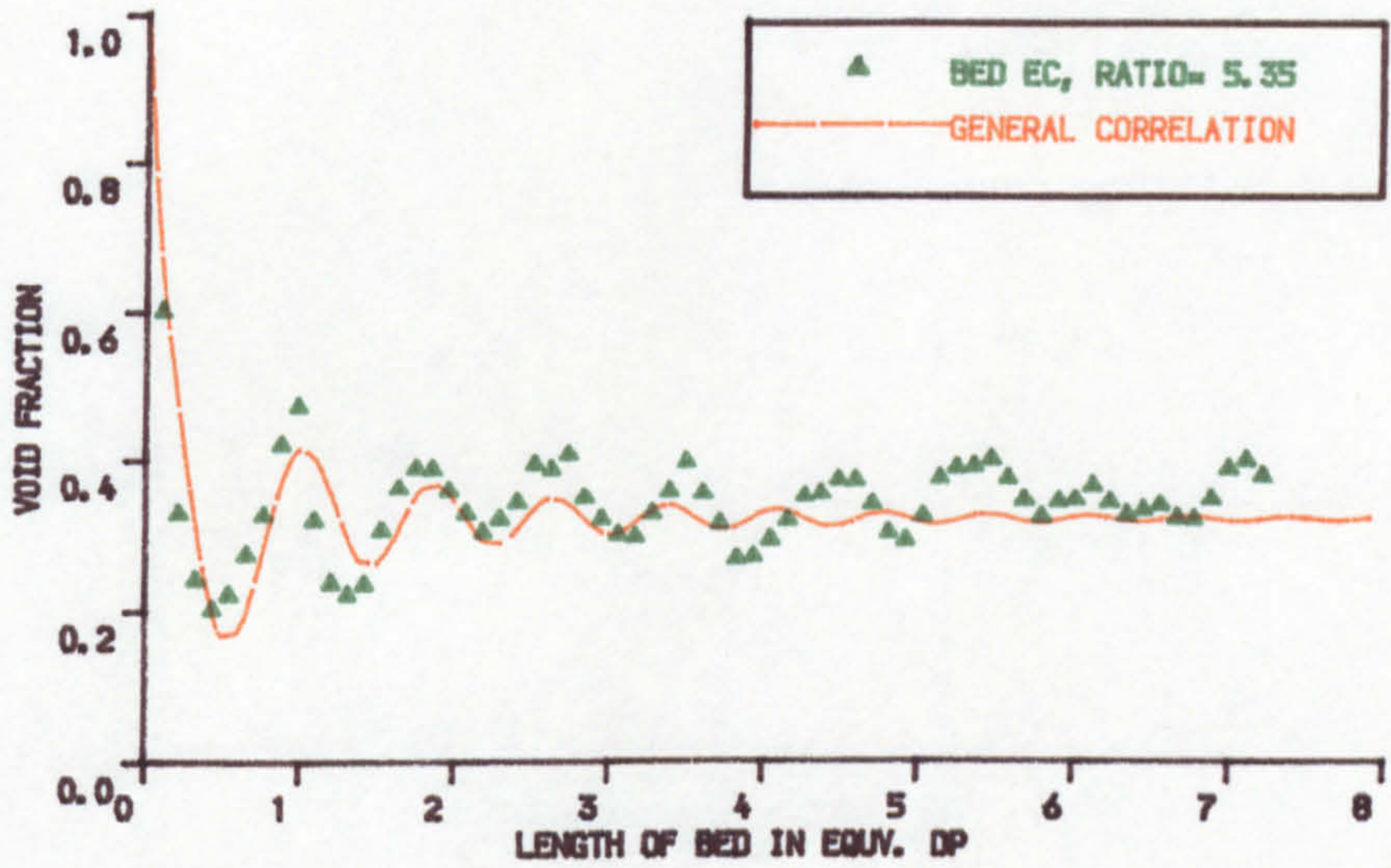


FIG. C15 DATA & GENERAL CORRELATION, EC

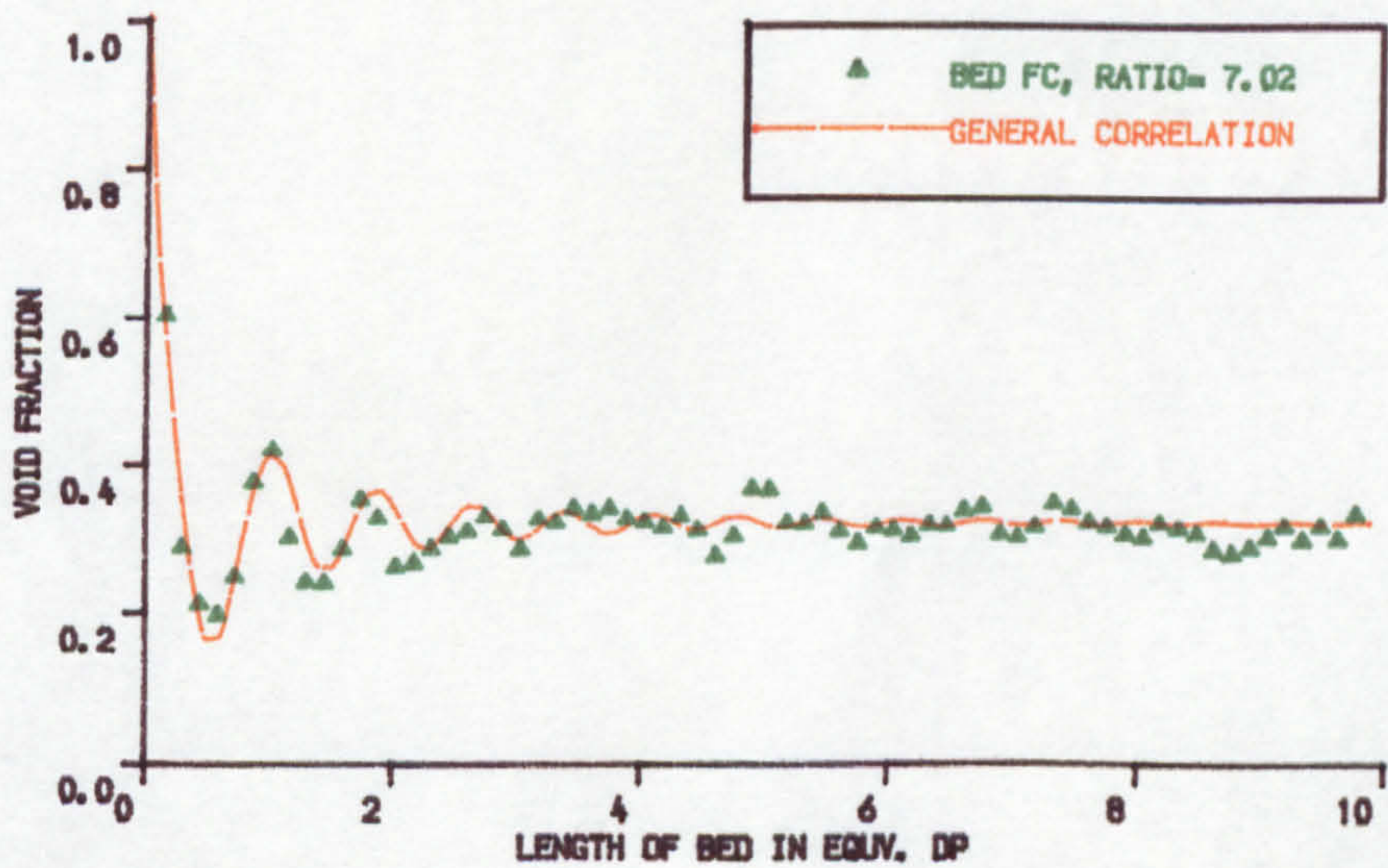


FIG. C16 DATA & GENERAL CORRELATION, FC

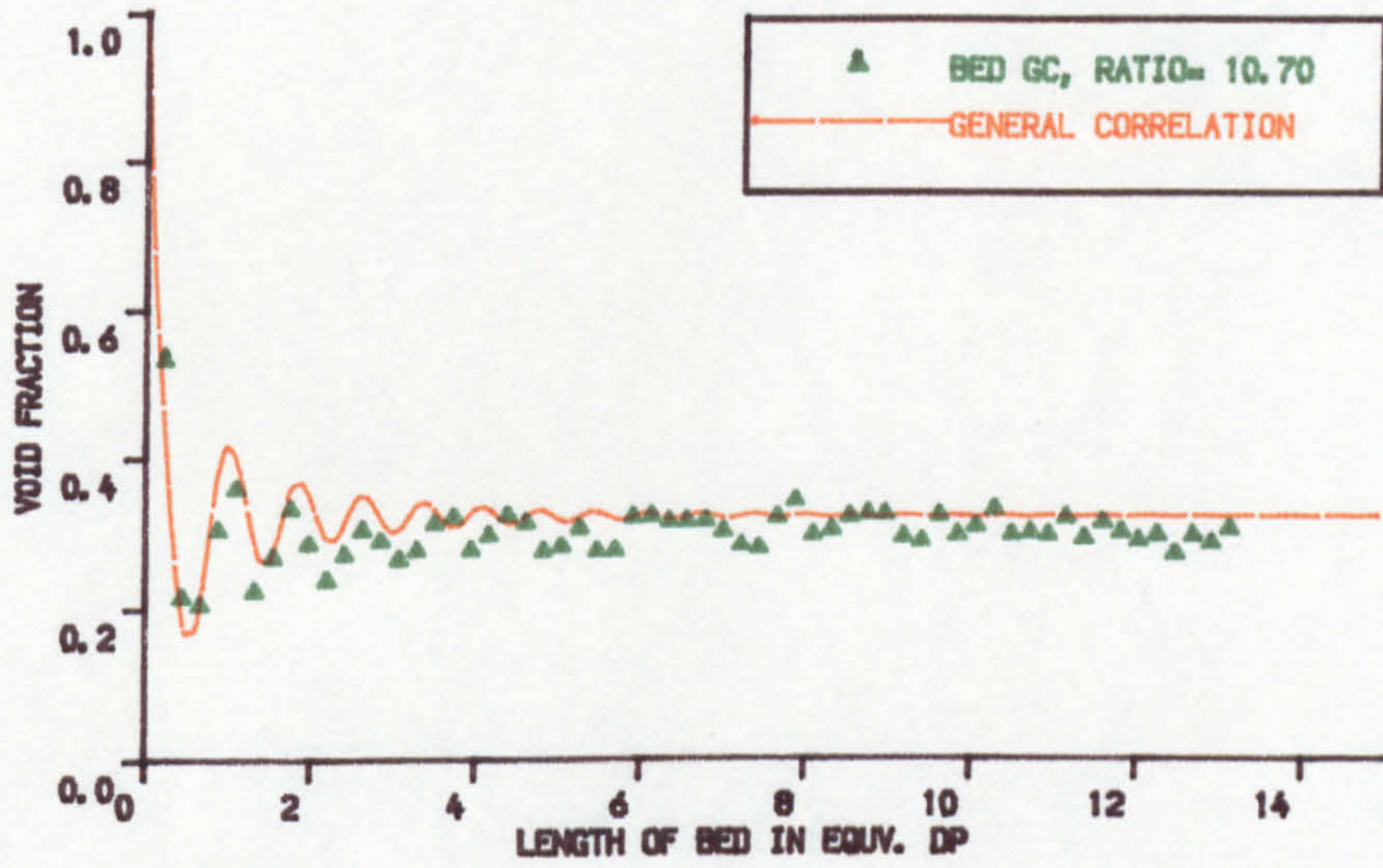


FIG. C17 DATA & GENERAL CORRELATION, GC

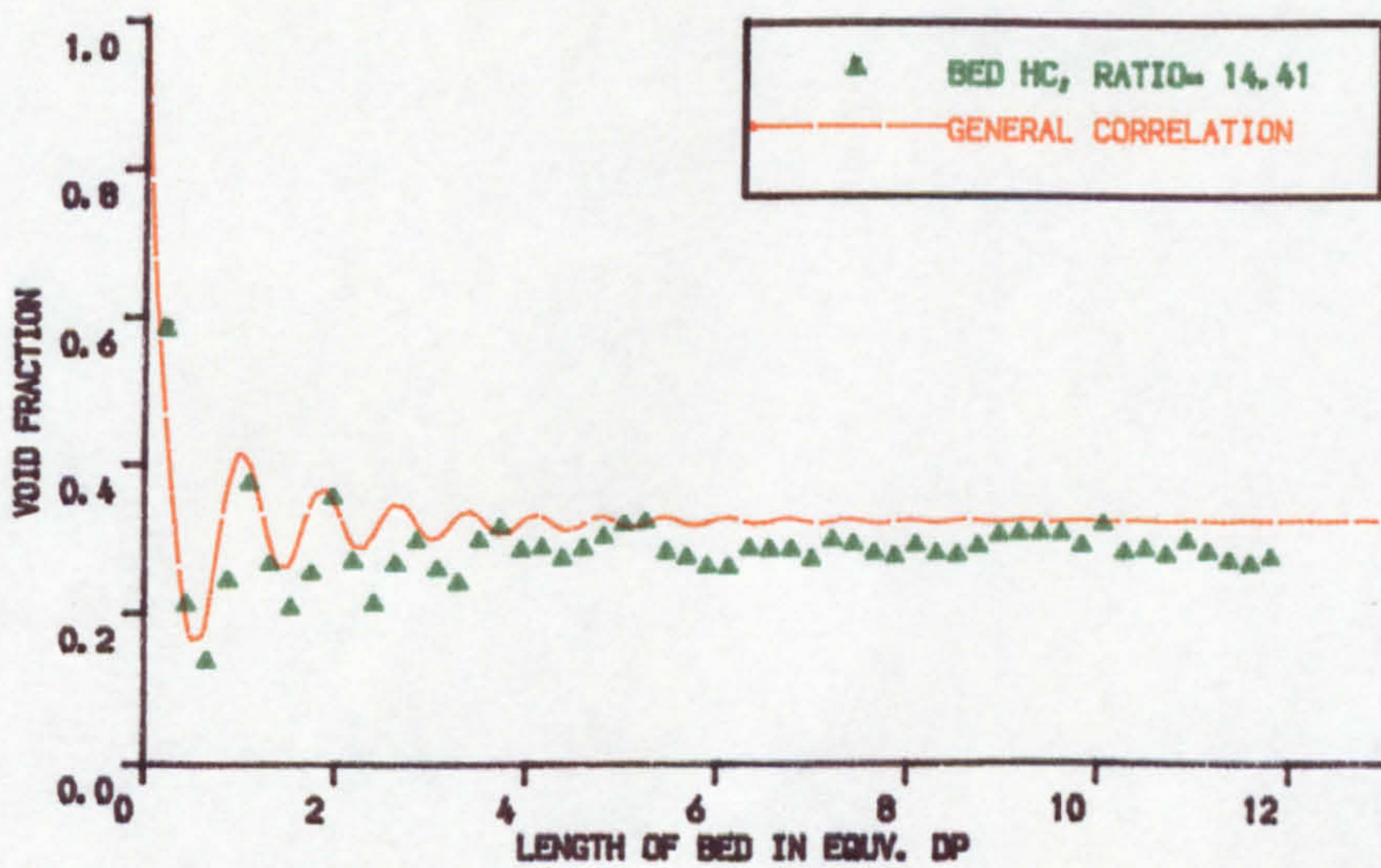
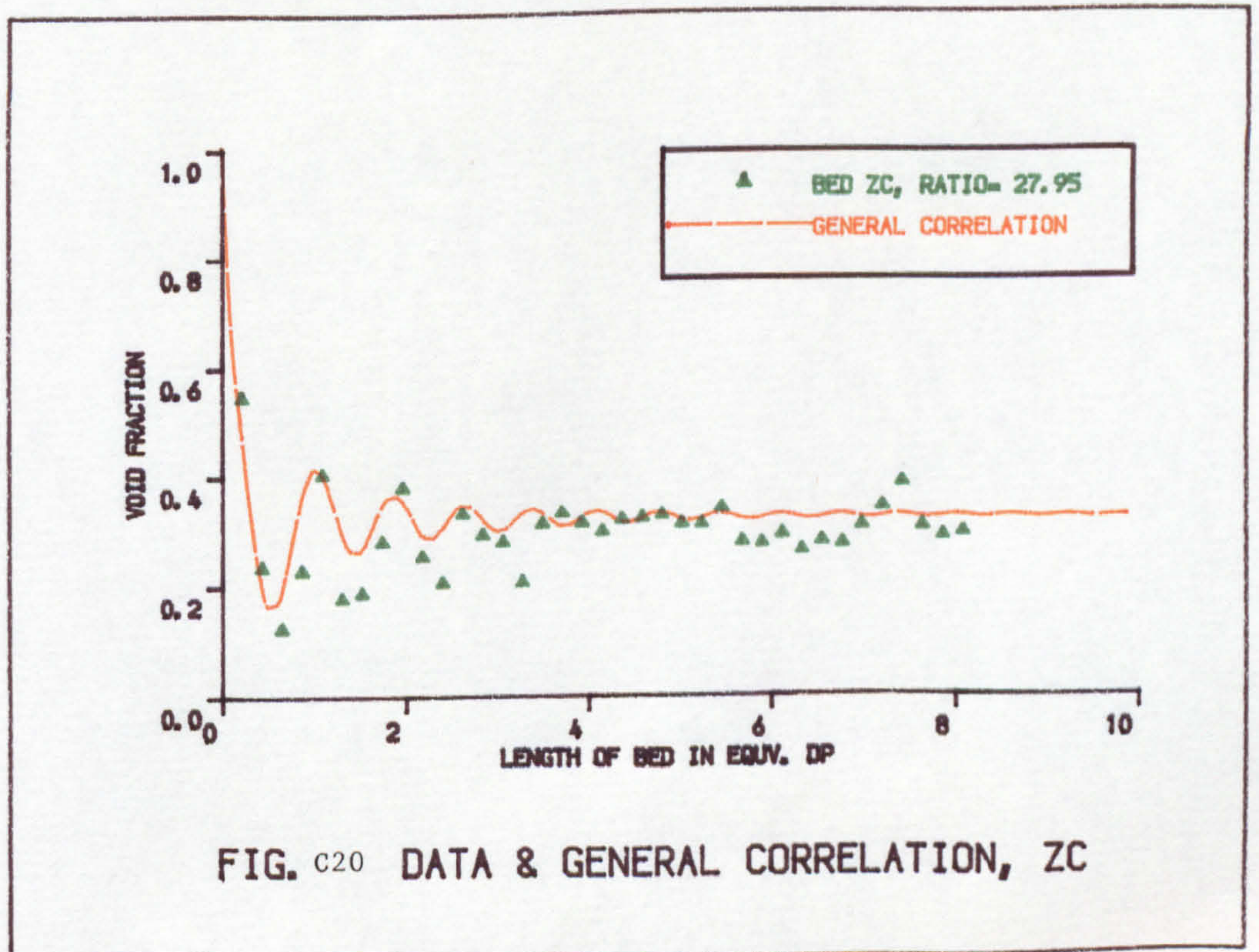
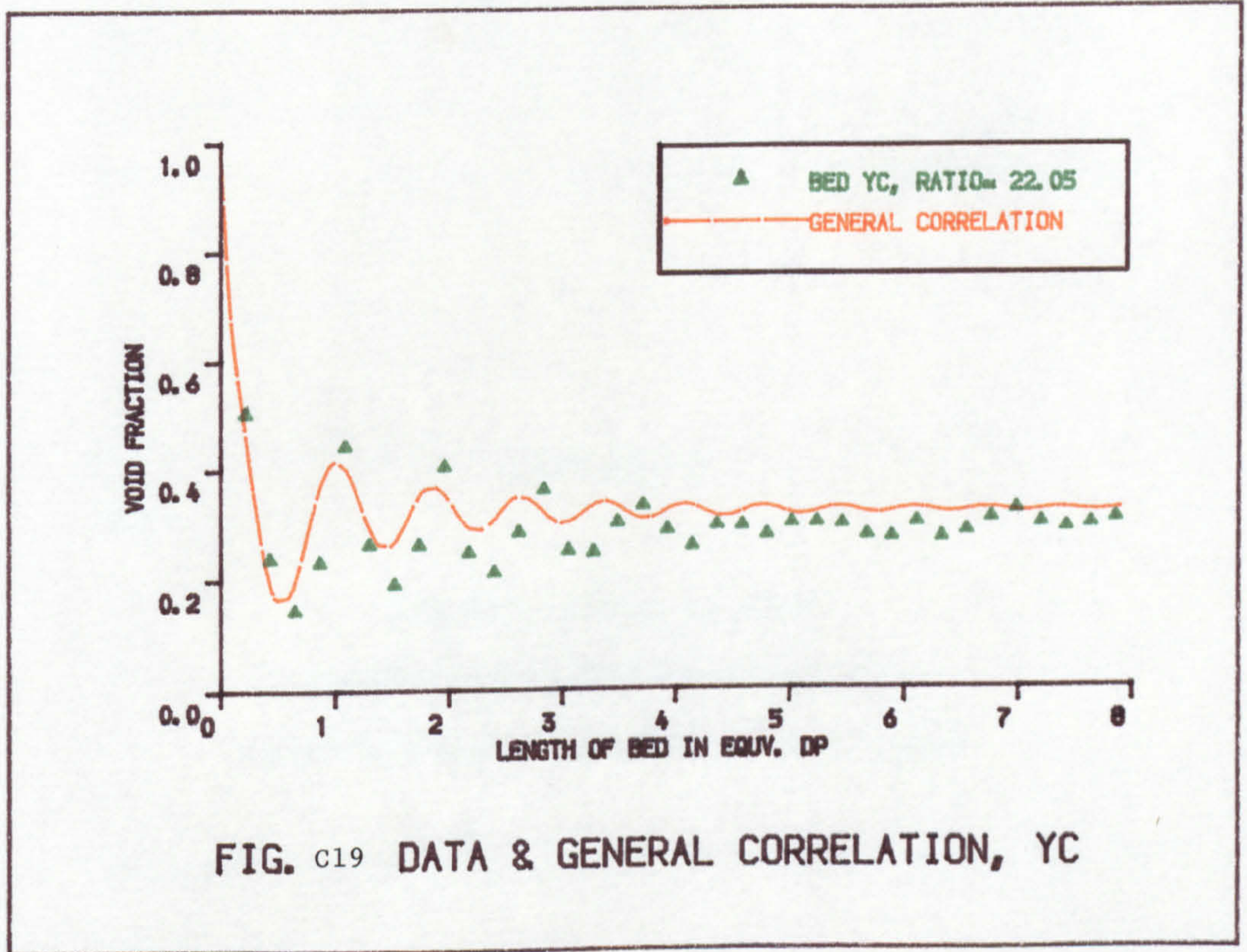


FIG. C18 DATA & GENERAL CORRELATION, HC



APPENDIX D

**Calculation of Φ and n values
together with profiles of radial voidage
correlation based on sphericity for each test bed**

APPENDIX D

1. Calculation of the value of sphericity for a cylinder

Considering a cylindrical particle with diameter, d , and length, L , the surface area of such particles can be obtained by the following relationship:-

$$A_{SC} = 2 \pi/4 d^2 + \pi dL \quad (D.1)$$

For an equilateral cylinder the above expression can be rewritten in terms of the diameter as:-

$$A_{SC} = \pi/2 d^2 + \pi d^2 \quad (D.2)$$
$$\therefore A_{SC} = 3/2 \pi d^2$$

The volume of an equilateral cylinder in terms of the diameter is given by:-

$$V_C = \pi/4 d^3 \quad (D.3)$$

The surface area of a spherical particle of diameter d_s is obtained by:

$$A_{SS} = \pi d_s^2 \quad (D.4)$$

and the volume by:

$$V_S = \pi/6 d_s^3 \quad (D.5)$$

Sphericity is defined as the ratio of surface area of a spherical particle to any other particle of the same volume. Therefore in the case of a cylindrical particle:

$$\Phi = \frac{A_{SS}}{A_{SC}} \quad (D.6)$$

Rearranging Equations (D.3) and (D.5) we obtain the following respectively:

$$d = 3 \sqrt[3]{\frac{4V_C}{\pi}} \quad (D.7)$$

and

$$d_S = 3 \sqrt[3]{\frac{6V_S}{\pi}} \quad (D.8)$$

Therefore when $V_C = V_S$, and substituting Equations (D.2) and (D.4) into Equation (D.6), sphericity can be written as:

$$\Phi = \frac{\pi \left[\frac{6 V_S}{\pi} \right]^{2/3}}{3/2 \pi \left[\frac{4 V_C}{\pi} \right]^{2/3}}$$

$$\therefore \underline{\Phi = 0.8736}$$

2. Calculation of the values of n in Equation (5.2)

The coefficients of the radial voidage correlation for packed beds of unisized spheres, based on Equation (5.1) are as follows:

$$\begin{aligned} \alpha_1 &= 0.6, \\ \alpha_2 &= 1.7, \\ \alpha_3 &= 5.57, \end{aligned}$$

$$\alpha_4 = 0.52,$$

$$\alpha_5 = 1.25.$$

According to Equation (5.2) which states:

$$\alpha_{\text{cylinders}} = \alpha_{\text{spheres}} \phi^n$$

and knowing the value of ϕ as 0.8736 and also the values of the coefficients of the radial voidage correlation for cylinders from Equation (4.8), the values of n can be calculated:

$$n_1 = -1.1$$

$$n_2 = -0.5$$

$$n_3 = -1.3$$

$$n_4 = 3.1$$

$$n_5 = 1.1$$

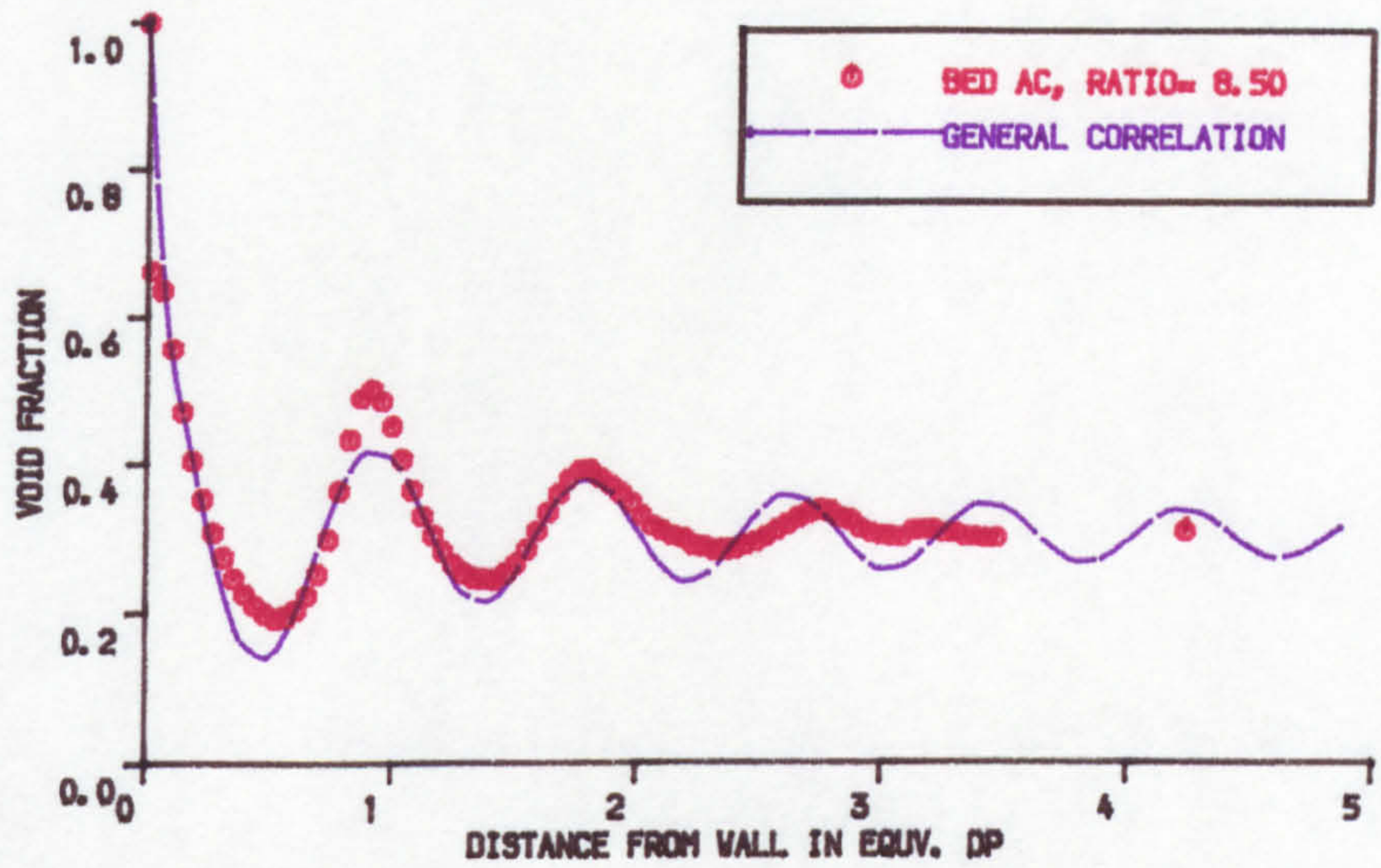


FIG. D1 SPHERICITY BASED CORRELATION, AC

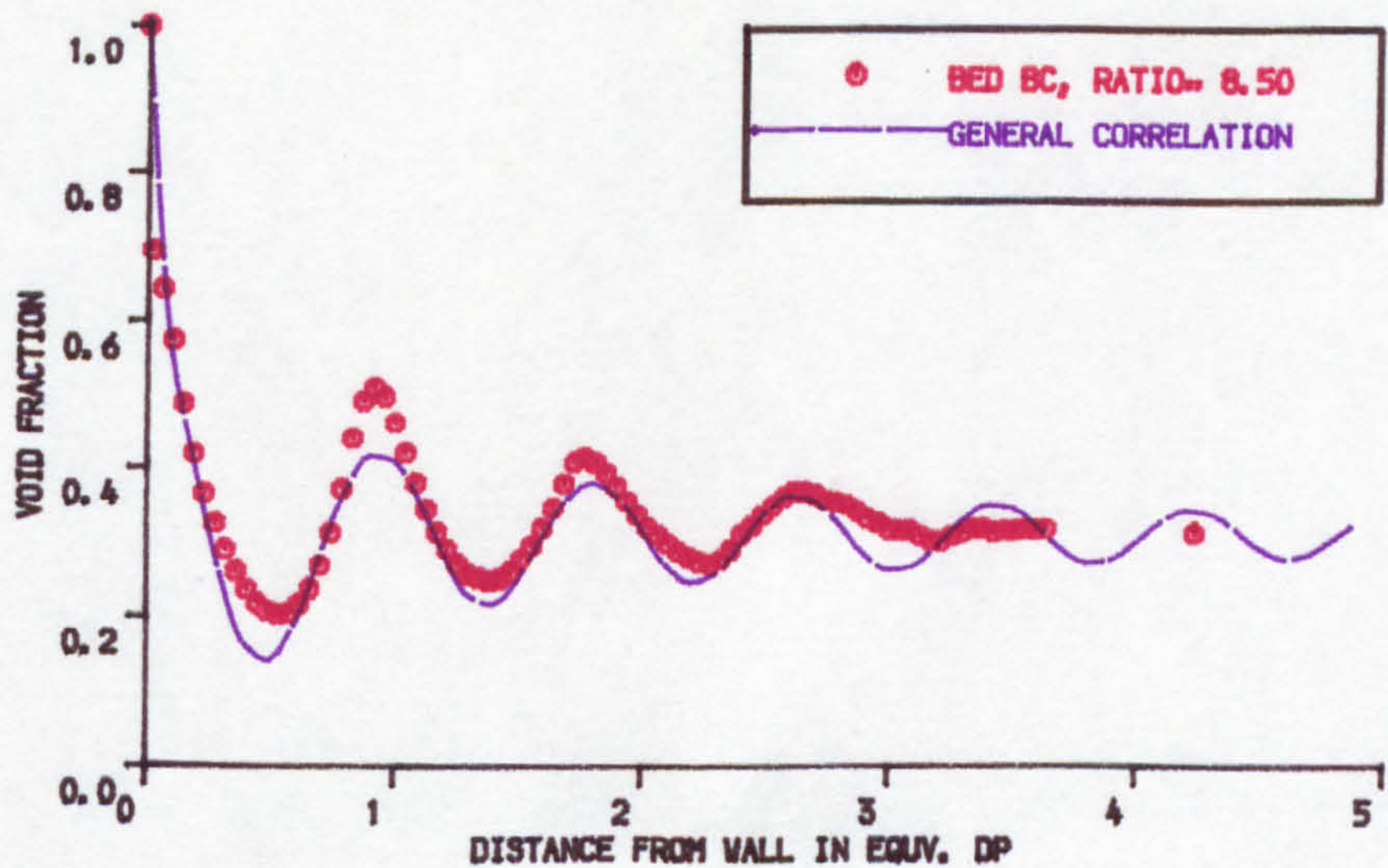
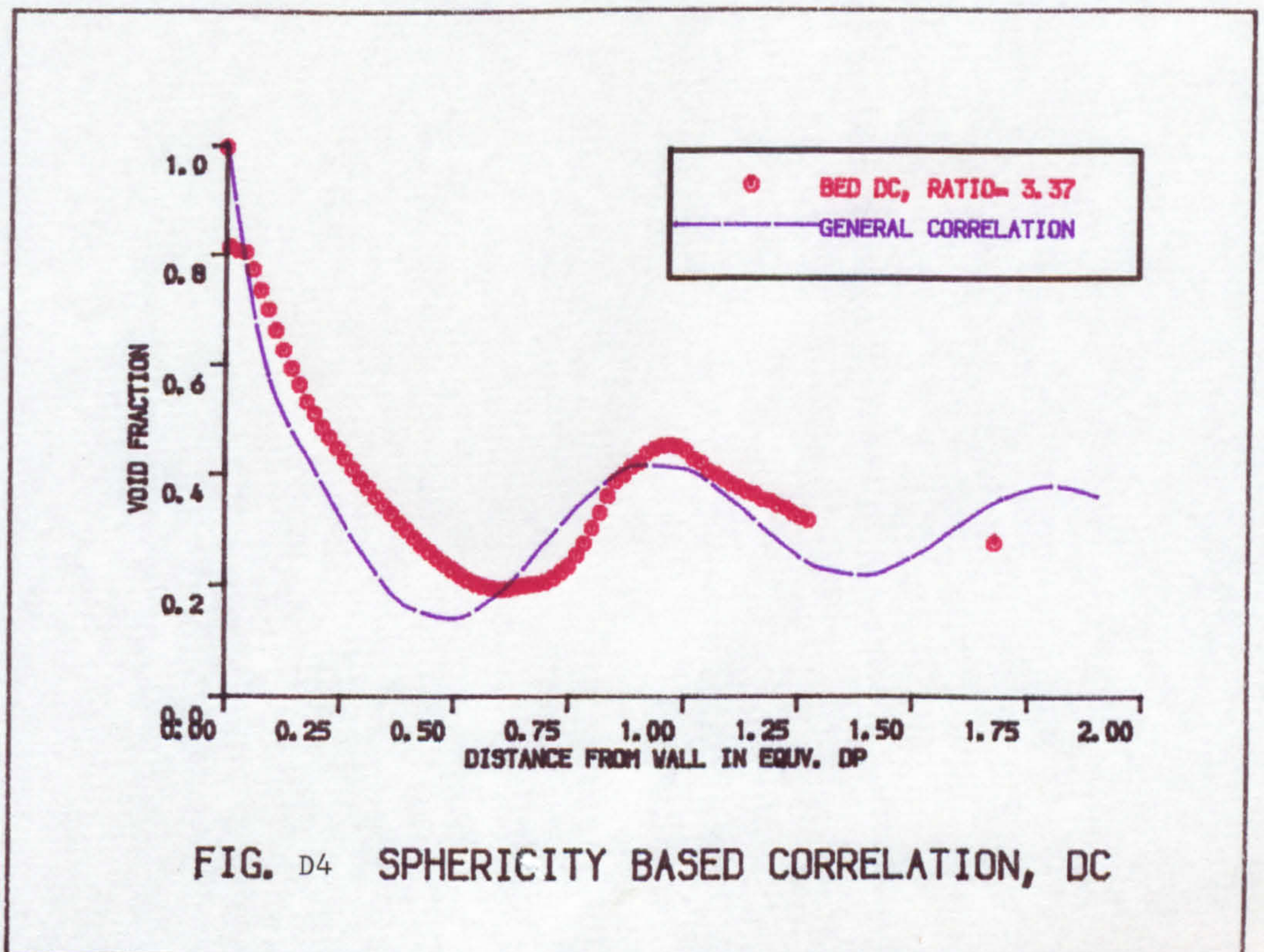
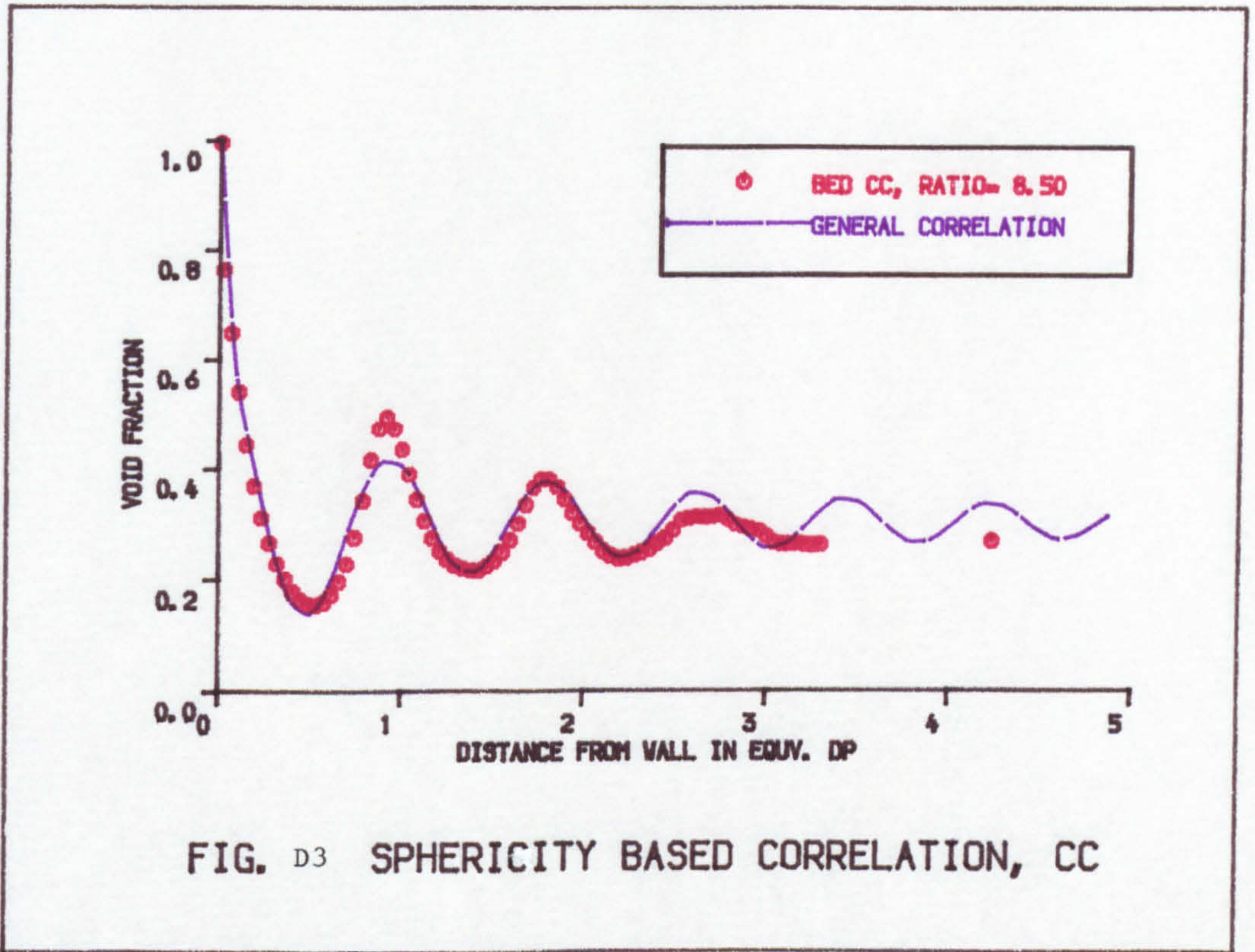


FIG. D2 SPHERICITY BASED CORRELATION, BC



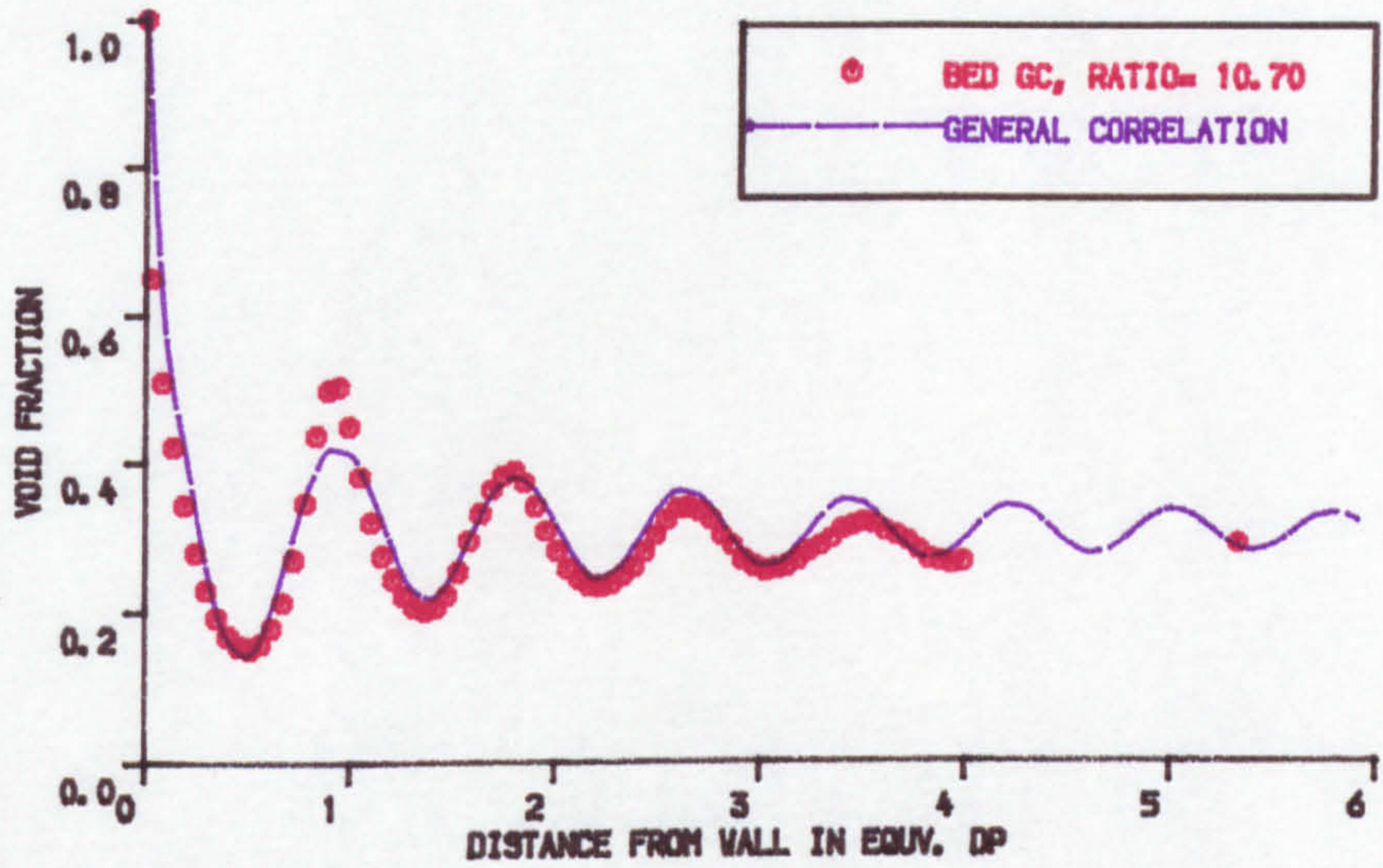


FIG. D5 SPHERICITY BASED CORRELATION, GC

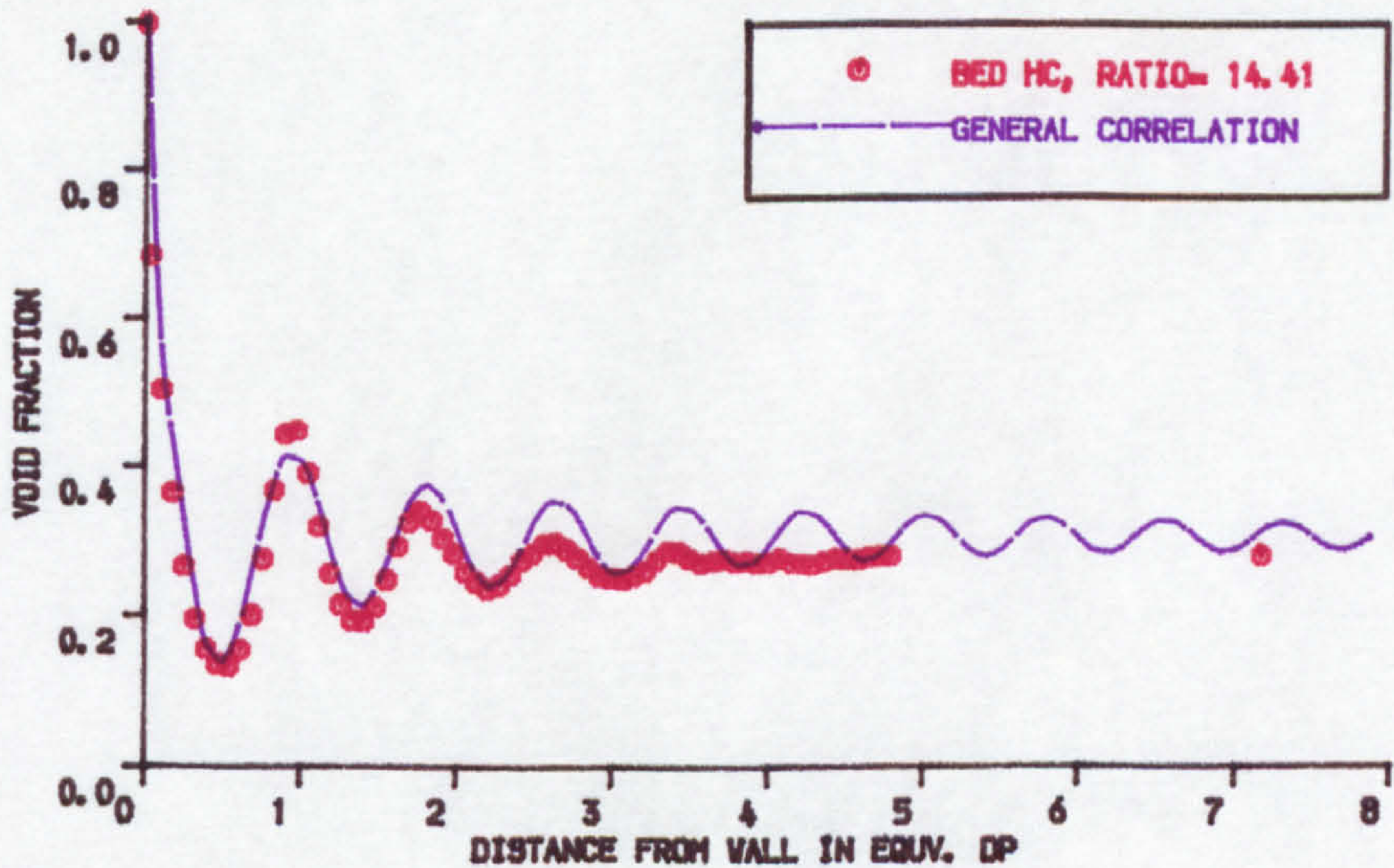


FIG. D6 SPHERICITY BASED CORRELATION, HC

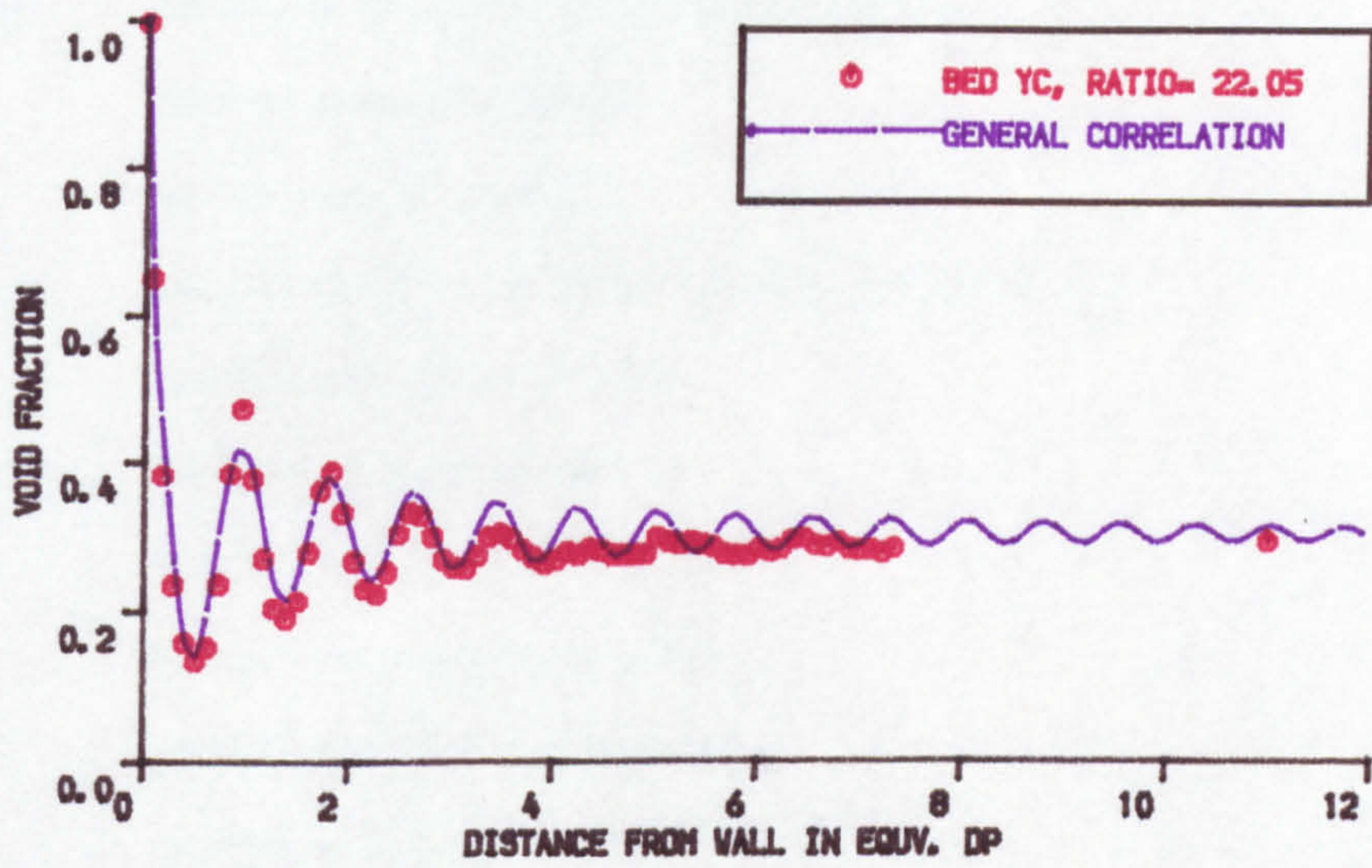


FIG. D7 SPHERICITY BASED CORRELATION, YC

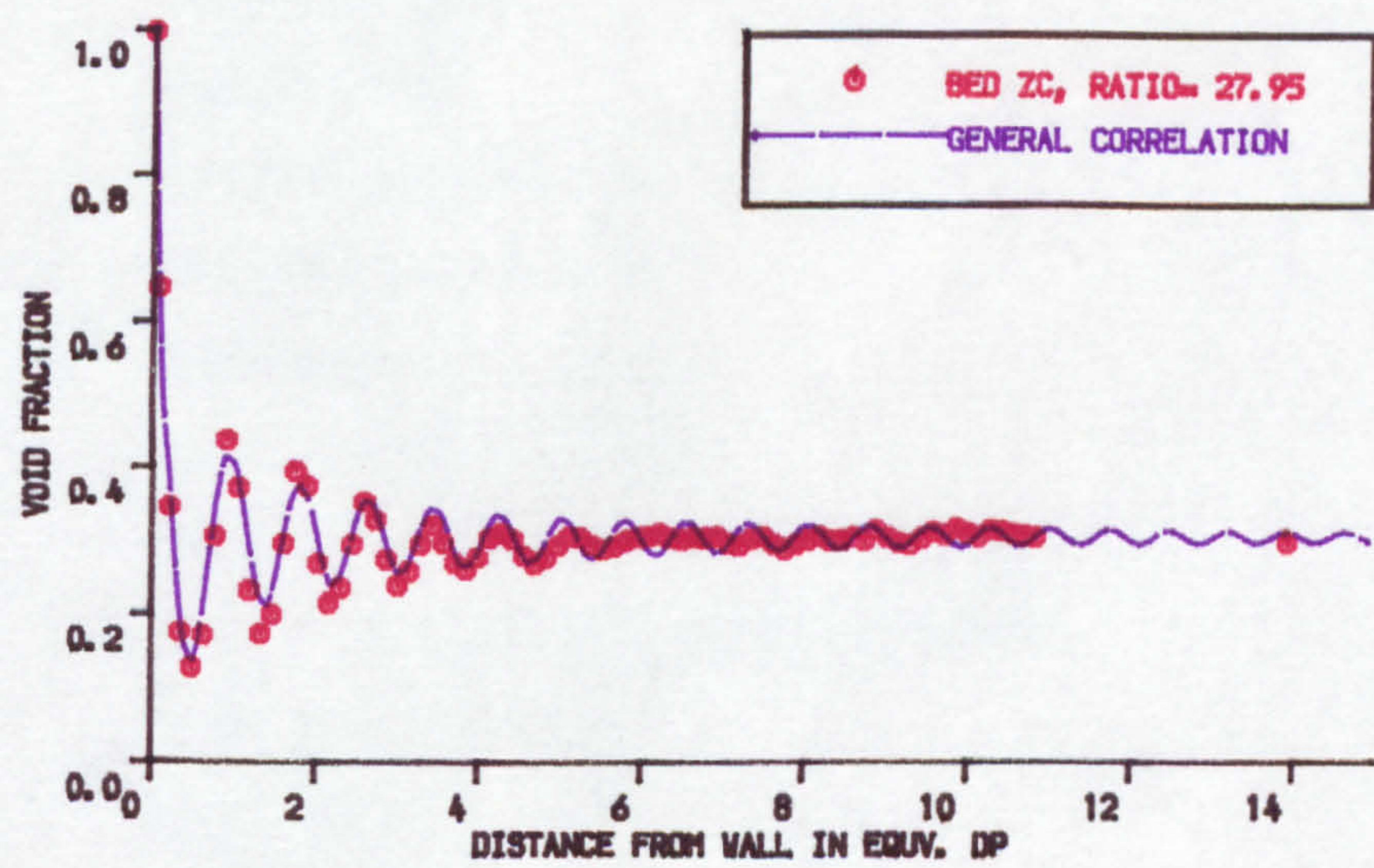


FIG. D8 SPHERICITY BASED CORRELATION, ZC

NOMENCLATURE

a	inside diameter of hollow cylinder, Equation (2.11)
a	aspect ratio, Equation (2.38)
a_1 to a_6	constants of Equations (2.23) and (2.33)
A_{sc}	surface area of a cylinder
A_{ss}	surface area of a sphere
b	outside diameter of hollow cylinder, Equation (2.11)
b	constant in Equation (2.30)
c	constant in Equation (2.30)
c_f	specific heat capacity of fluid
c_s	specific heat capacity of solid
d	particle diameter, Equation (2.5)
d_p	particle diameter
d_{pe}	equivalent particle diameter
d_{pv}	diameter of equivalent volume sphere, Equation (2.8)
d_r	diameter ratio, particle to tube
d_{re}	equivalent diameter ratio
d_s	diameter of sphere
d_t	diameter of tube
D_e	equivalent diameter of bed
D_L	axial dispersion coefficient
D_R	radial dispersion coefficient
h_w	wall heat transfer coefficient
K_{ea}	effective axial thermal conductivity coefficient
K_{er}	effective radial thermal conductivity coefficient
l_p	length of cylindrical particle
L	length of bed
LDA	Laser Doppler Anemometry

m	number of cylindrical concentric layers, Equation (2.35)
n	number of fractions, Equation (2.35)
n	constant of Equation (5.2)
N	total number of spheres in bed, Equation (2.35)
p	layer number, Equation (2.16)
Pr	Prandtl number
r	conversion rate, Equation (2.31)
r	radius of particle
R	radius of tube
Re	Reynolds number
SEE	Standard Error of Estimates
SSE	Sum of Square of Errors
t	time
T	temperature
u	point velocity at radius r, Equation (2.14)
u_0	point velocity at centre of pipe, Equation (2.14)
V_c	volume of a cylinder
V_s	volume of a sphere
x	distance from tube wall
y	distance from wall, Equation (2.28)

Greek Letters

α_1 to α_5	coefficients of Equation (4.2)
β_1 to β_5	coefficients of Equation (2.42)
β_1 to β_5	coefficients of general axial voidage correlation
γ	distance from wall, Equation (2.30)
ΔF_2	difference between cylindrical and flat wall boundary
Δr	thickness of a layer, Equation (2.39)

δ	void fraction at radius r , Equation 2.14)
δ_0	void fraction at centre of bed, Equation (2.14)
δ_w	width of wall layer, Equation (2.1)
ϵ	bulk mean voidage, Equation (2.1)
ϵ	mean voidage, Equation (2.31)
ϵ_b	bulk voidage, Equation (2.25)
ϵ_x	voidage at distance x from wall, Equation (2.16)
ϵ_w	voidage of wall layer, Equation (2.1)
ϵ_0	overall voidage, Equation (2.1)
ϵ_0	void fraction at centre of bed, Equation (2.30)
ϵ_∞	voidage at core zone
$\epsilon(r)$	radial variation of voidage
$\epsilon(x)$	axial variation of voidage
$\epsilon(\theta)$	angular variation of voidage
ϵ_{mean}	mean voidage of a packed bed
∂	distance of first minimum voidage value from the wall
ρ_f	density of fluid
ρ_s	density of solid
ψ	diameter ratio
Φ	sphericity
Σe^2	sum of square of errors

Subscripts

hc	hollow cylinders, Equation (2.11)
sc	solid full cylinders, Equation (2.11)

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