University of Alberta

Statistical Techniques for Correlating the Shape and Performance For New Random Packings

by

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Abstract

Separation processes are the heart of the chemical industry and are used for product purification. Distillation, absorption and stripping are some of the basic types of separation processes encountered in the industry. The operations are carried out in columns containing trays or packings. Packed columns have become popular in the industry because of the advantages associated with them. There are many types of packings available in the market and research is on going to find better packings. In the present study, we deal exclusively with random packings.

The existing models calculate the mass transfer efficiency and other performance parameters depending on the properties of the fluids being handled, the geometrical area, porosity and the shape of the random packing. The shape of the packing is generally characterized using a packing specific shape constant. Though these models cover a wide variety of random packings, one cannot predict the performance of newly developed packing elements without experimentation. In this work, we investigate the relation between the performance and the shape of the random packings.

Since geometric parameters like surface area and porosity do not define the shape of the packing uniquely, we attempt to define the shape of the packing using the porosity distribution characteristics. The porosity distribution was computed using an algorithm developed for packing random packings. Higher order statistics were used to define the porosity distribution and Partial Least Squares (PLS) modeling was used to develop a linear relationship between the shape defining parameters and the performance

characteristics of random packings. New packings were developed by selecting various shapes available in geometry and the mass transfer efficiency and pressure drop were predicted for these packings using the model.

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Chapter 1

Introduction

Distillation, absorption and stripping are the basic unit operations used in the chemical industry for clean up or separation of a wide variety of process streams. Product purification after a unit process is generally achieved in distillation columns and separation of lighter and harmful vapors and gases is usually accomplished in absorbers and strippers in most chemical industries. There are two types of separation columns used widely in the chemical industry, viz., tray columns and packed columns. Each type of column has its own advantages and replacement of one with the other is of great interest to many researchers. It cannot be concluded without detailed study that the performance of one type of column is better than the other in all cases. Tray towers are still widely used and not replaced with packed towers for some services. The comparison between trays and columns can be detailed as follows:

- Packing has a significant inherent advantage when it comes to pressure drop, because the interfacial area for packed columns is generated through liquid spreading on the packing surface - this is a low pressure drop phenomenon compared to the mechanism required to generate high mass transfer efficiency within a tray column.
- Packing has the disadvantage of requiring re-distributors if the height of the column required is large.
- Packed columns often can be designed with greater stable operating range than sieve trays. Some type of trays such as valve and bubble cap trays can have a

stable operating range equal to or even greater than that packed columns. The design of internals, such as distributors, for a stable operating capacity variation of more than two or three is much more difficult for packings than for trays.

In spite of the advantages and disadvantages of packed columns, there are certain applications which demand packed columns for separation. Packings can be divided into two different types – structured and random. Structured packing is of a definite shape designed to enhance contact between the vapor and liquid phases and random packing is of any shape, generally ranging from a nominal size of 1 inch to 3 inches. These packing elements come in different shapes and are dumped randomly into a cylindrical column thus locating themselves in different random orientations in the packed bed. Though there are packings like the Cascade Mini Ring that have a preferential orientation when dumped into a column, other packings have an aspect ratio (defined as the ratio of the diameter of the packing to the height) of one and can be in any possible orientation. This leads to overall randomness in the bed and this property aids in better distribution of the liquid and gaseous phases. Structured packings are manufactured in blocks of different sizes and generally have sheets of corrugated metal of different configurations and are arranged in particular geometries. Currently, packings are available in numerous shapes and sizes and are applied to different types of separation process. The efficiency of mass transfer depends on the geometry, size and texture of the packing. Two important geometric parameters or properties that influence the mass transfer intensity and pressure drop are the geometric surface area and the overall porosity (or void volume). Porosity is a measure of the empty space in the column when filled with different kinds of packing elements. Figure 1.1 shows random packings and structured packing assembled in different sections of a separation column. The top portion of the packing contains random packings of two different types and the bottom portion consists of structured packing. This arrangement of mixing the types packing is generally employed for better performance.

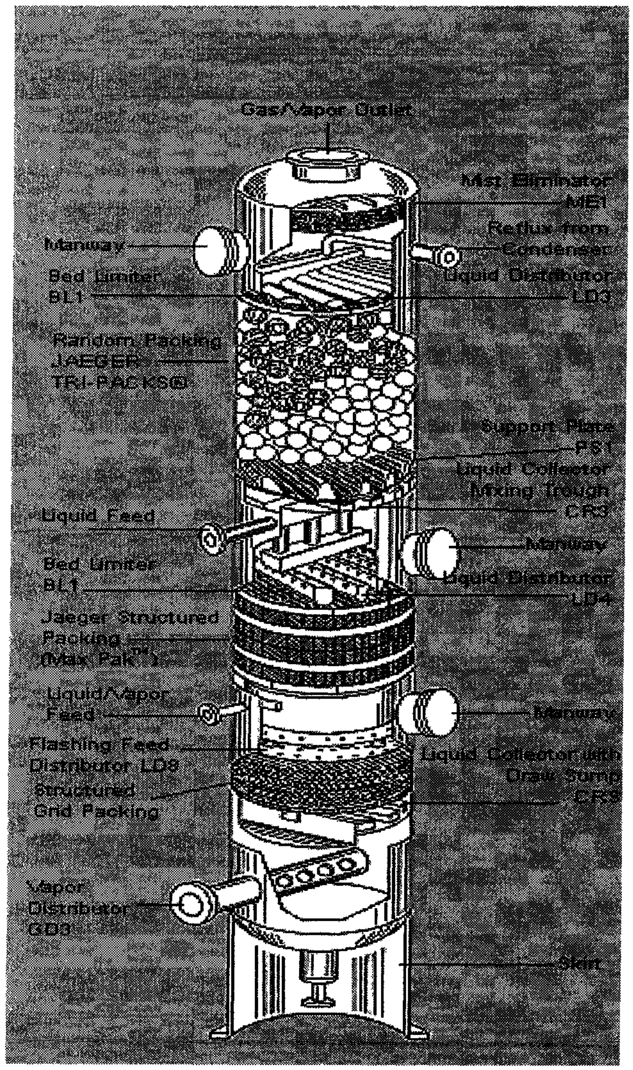


Figure 1.1: Separation column packed with Random packings in the top section and structured packing in the bottom sections

A greater surface area almost always gives a better mass transfer intensity but the pressure drop across the bed also increases due to decreased porosity. These parameters are bulk parameters and there can be two different types of packings having the same surface area and porosity but entirely different mass transfer and hydraulic properties. The porosity and the geometrical area of a packed bed give qualitative information about the mass transfer efficiency, capacity and pressure drop. Other parameters are required to evaluate the performance and they should allow for the geometry and texture of the packing material. One type of packing does not give an optimum performance in each and every application which explains the numerous designs available in the market.

In spite of the progress made in understanding the performance of both random and structured packings in distillation, it is quite difficult to predict from first principles, the efficiency, capacity and pressure drop of a tower packing using thermodynamic and thermo-physical properties of the chemical system. Selection of physical parameters of the packing to aid mass transfer is also based on empirical relationships and experience. The predictive methods that are available in literature have limited accuracy when applied to a wide variety of chemical systems or outside the database. The older models were based on a few packings available at that time and did not include the recently developed packings. Recent models are semi-empirical and are based on physically proven conditions of fluid dynamics in columns and include a wide variety of packings.

The number of stages required for a given separation is obtained from the application of equilibrium thermodynamics. The actual number of stages obtained from a packed tower either in a laboratory, pilot plant, or an industrial plant is divided by the equilibrium stages predicted by vapor-liquid equilibrium thermodynamics to obtain efficiency for the packed tower. Attempts have been made to generate semi-empirical correlations for packed tower efficiency from experimental data. The mass transfer capability of packing is typically expressed as Height Equivalent to a Theoretical Plate (HETP) or Height of Transfer Unit (HTU) etc., which are rate-dependent quantities. Attempts to derive generalized predictive methods for the mass transfer efficiency of packings using the two-film theory and dimensionless groups, and for the pressure drop and capacity using mechanistic models, have met with varying degrees of success.

Published results of these attempts are the works of Cornell *et al.* (1960), Onda *et al.* (1968), Bravo and Fair (1982), Bolles and Fair (1982), Billet and Schultes (1991; 1993 a, b; 1995; 1999) and Bornhütter and Mersmann (1993). Barring the models of Cornell *et al.* and Onda *et al.*, the models used in these predictive methods were checked against many sources of pilot plant data, especially those made by Fractionation Research, Inc. (FRI) and the Separation Research Program (SRP) of the University of Texas at Austin. Most of the recent models include some sort of empirical constant that accounts for the shape of the packing element.

The drawback of all the above-mentioned models is that they cannot be used to predict the properties for a new shape of packing element under development. Experiments are generally required to ascertain the performance of the packing. If direct or indirect theoretical methods were available to predict the performance of packing based on the shape, they would give an insight into these properties much before the packing is put to the experimentation stage. It would also allow one to correlate the shape of the packing with the performance. The term correlation has been used in a very loose sense in this work except when mentioned in the subsection describing the Partial Least Squares modeling theory. Correlation is used at times to describe the regression equation that equates the dependent and independent variables.

1.1 Objectives

The objectives of the thesis are two fold. First, to investigate and select characteristics that define the shape of random packings. And for existing packings, develop a model that describes the relationship between the shape defining and geometric characteristics and performance parameters like pressure drop and mass transfer. Second, develop new packings and use the model developed from existing packings to predict the performance of new packings using their shape-defining and geometric characteristics.

The following steps show the approach to the problem:

1. Geometric surface area, mean porosity and statistical parameters describing the porosity distribution are used to define the shape
2. Packing algorithm developed by Nandakumar et al. (1999) is used to extract the porosity distribution across the volume of the bed for existing random packings.
3. Performance parameters, viz., mass transfer coefficient and pressure drop for a common operating system (carbon dioxide-air-water) are calculated using Billet and Schultes model (1999) for the existing random packings

4. Partial Least Squares (PLS) modeling is used to develop a regression equation in the predictive form between the performance parameters and the shape defining characteristics
5. New packings are developed from various geometrical shapes available in literature.
6. Model for relating surface area of the new packings as a function of size is developed using Design of Experiments and Response Surface Methodology. This is done since the area for new packings in a packed bed is a function of the number of elements packing and this is can be ascertained only after packing the elements.
7. Area and porosity are selected to be within the range of the PLS model developed for he purpose. The size of the packing is then selected from the least squares model mentioned in Step 6.
8. The porosity distribution across the volume of the bed for these new packings is extracted using the Packing Algorithm.
9. The PLS model mentioned in Step 4 is used to predict the performance of the new packings.
10. Best possible shape and future directions for research are then concluded from the study.

1.2 Overview of the thesis

The main focus of this work is to establish a relation between the performance characteristics of the system such as mass transfer and pressure drop and geometric parameters such as surface area, porosity and the shape of the packing element. A relationship of this sort would help one in determining the optimal shape for suiting the performance characteristics. An attempt to relate these characteristics with the shape has been made in this thesis. For this purpose, the packing program developed by Nandakumar *et al.* (1999) has been used. The shape of the packing element within a packed bed can be distinguished using the distribution of the empty space within the bed. Statistical parameters were used to define the distribution and the relationship between the performance and these statistical parameters was developed using a regression methods. The subsequent paragraphs describe the overview of each chapter presented in the thesis.

Chapter Two discusses the previous work done in this area to ascertain and select the models available for mass transfer and fluid dynamics. These were used to relate the transport behavior to the shape of the packing element and to develop new types of elements. Different shape analysis techniques available in literature and the characteristic properties of a non-normal distribution using higher order moments are also discussed.

Chapter Three describes the algorithm developed by Nandakumar *et al.* (1999) and how it is used in the present work to develop a model for a packed bed with new random packings designed for the purpose. It discusses the requisite inputs for the program to simulate the packing process and the capability of the program to extract geometric parameters such as the porosity distribution. The chapter also describes the basis of sample size selection for measuring the porosity distribution and methods for testing the normality of a distribution using statistical tests. If the distribution is found to deviate from normality, it defines statistical parameters that can be used to characterize the non-normal distribution. The methods for estimating these parameters are also discussed here.

Chapter Four describes the new packing shapes that were developed for this study and reasons why these specific shapes were looked into. The properties of these shapes and the mathematical models that define them are also discussed in the chapter. This chapter explores the modeling methods used for predicting the area and porosity as a function of the size for each of the shapes. Techniques like Design of Experiments and Response Surface Methodology used to select the size based on the area are described in this chapter. The best possible fits with the minimum number of experiments required and models relating the area to the dimensional aspects are discussed.

Chapter Five describes the mass transfer and pressure drop model developed by Billet and Schultes (1999) to predict the properties of a large number of random packings that exist in the market and explains reasons why this model was preferred over other models in the literature. The system used to arrive at the mass transfer coefficients and pressure drop is also outlined in the chapter. The theory of Partial Least Squares (PLS) and the use of this modeling technique to develop a regression equation relating these parameters with shape defining characteristics are also discussed here. The application of the relations developed for the new packings are outlined.

Chapter Six summarizes the contributions of this thesis with respect to modeling of random packings and relationship developed between the shape of the packing and the performance using statistical techniques. The potential of this study as a tool for the development of new shapes is discussed as an area of future work in Chapter Seven.

Chapter 2

Literature Review

2.1 Models for predicting mass transfer and pressure drop

Prediction of the mass transfer efficiency of beds packed with random packings is not straightforward because of the complex two-phase flow that exists in these packings. For random packings, four methods have been developed in the past that merit serious consideration. The method of Cornell *et al.* (1960) utilizes the two-film model and empirical parameters specific to each packing type and size. The data set considered for this study was small and did not include the modern through-flow type packings. Onda *et al.* (1968) developed a random packing model based on a data set using the two-film model that included traditional packings like ceramic rings and saddles. Bolles and Fair (1982) expanded the Cornell *et al.* database and adapted the model to new experimental results. They included only the Pall ring as the through-flow packing. Bravo and Fair (1982) used the Onda model and the Bolles-Fair database for correlating the packing type and the effective interfacial area.

2.2 Mass transfer model by Wagner et al.

A new model was developed by Wagner *et al.* (1997) for correlating the mass transfer rates in distillation columns packed with newer random packings where emphasis is placed on the newer through-flow packings. In their work, the pressure drop and hold up model developed by Stichlmair *et al.* (1989) is used as it includes the determination of liquid hold up above and below the loading point. In building the model, experimental data from the laboratories of Fractionation Research, Inc., and the Separations Research Program (SRP) at the University of Texas at Austin was used.

In their model, the liquid hold up in the column is defined as the volume of liquid held under operating conditions per volume of packed bed. The hold up can be divided into two components, the static and the dynamic hold up. The static holdup consists of the liquid present in the voids of the packing. The dynamic portion of the hold up flows through the column and is a strong function of the liquid load. The hold up relation of Stichlmair *et al.* (1989) does not account for the physical properties and has been validated for air-water system only, but the equation is applicable for viscosities up to $5*10^{-3}$ Pa.s.

Above the loading point, the correlation for liquid hold up of Stichlmair *et al.* (1989) is dependent upon the irrigated pressure drop. Since the hold up is required for the calculation of irrigated pressure drop, iteration is necessary.

In this model a packing parameter specific to the packing called the "packing characteristic" is needed for the correlation for mass transfer. This parameter has been retrofitted from the available data for all types of packings. The model has been found to fit 95% of the 326 experimental values and the height equivalent to theoretical plate (HETP) to within ± 25%.

2.3 Models for predicting interfacial area

The amount of the existing effective interfacial area in a packed column is the basic factor that affects the mass transfer. Since all the available area is not utilized for mass transfer, the effective area is less than the total area except for high liquid loads when the area becomes equal to or higher than the geometrical area. The effective interfacial area is also reduced because of a shielding effect (Hanley *et al.*, 1994). This is because of the gas maldistribution caused by liquid filled voids which block vapor flow. A large part of the effective interfacial area also results from the drops (Bornhűtter, 1993).

The interfacial area in a packed column is closely related to the hydrodynamics. The fluid conditions are described through irrigated pressure drop and hold up. The amount of vapor-liquid surface area is directly linked to the operating hold up (Bravo and Fair, 1982). Relationships between actual and and the effective interfacial area have been developed by a number of researchers (Onda *et al.*, 1968; Puranik and Vogelpohl, 1974; Billet and Schultes, 1993; Bravo and Fair, 1982). These models serve as a tool for developing correlations for mass transfer for open packings but cannot be applied outside the database.

2.4 Model for prediction of mass transfer coefficients for new packings

While developing new packings, it becomes necessary to predict the mass transfer coefficient and pressure drop for the new shapes based on their geometrical properties.

The "cylinder model" by Bornhűtter and Mersmann (1993) allows one to predict the mass transfer coefficient based on an empirical model.

This model takes into account two different types of flow behavior for random packings. The first type describes the flow of liquid exclusively in the form of films and rivulets. This behavior is more pronounced in case of small packing elements, good wetting characteristic of the packing, low surface tension and very small openings in the surface of the packing elements. Onda *et al.* (1968) and Puranik and Vogelpohl (1974) based their equations on this type of liquid flow. The second type of liquid flow, in the form of drops, is more predominant in large packings with lattice structure, liquid of high surface tension and poor wetting conditions.

A decrease in the specific packing surface area is always accompanied by a decrease in the volumetric mass transfer coefficient. It has been shown in the work that there is no decrease in the mass transfer coefficient for large Hi-flow ring packing elements. It has been shown that the 50 mm and 90-0 mm packing elements have essentially the same mass transfer coefficients at all liquid velocities. But the mass transfer coefficient of 90-6 mm element is lower than both the 50 mm and the 90-0 mm packings. Since the only difference between the 90-0 mm element and the 90-6 element is the presence of pins at the edges of the element it was concluded that the pins are the reason for better liquid dispersion. This increases the drop-forming tendency which in turn increases the mass transfer coefficient.

An empirical model was built by Bornhütter and Mersmann (1993), which has size of the element among other physical and geometrical properties. The geometrical properties included the area, porosity and the number of elements per unit volume while the physical properties were the surface tension, viscosity and density.

Constants have been included to take care of the neglected influences such as impact, liquid jets and the texture of the packing (plastic or metal). The texture is important since the wetting tendencies and contact angles differ based on the construction material of the packing. The cylinder model can be used for prediction of mass transfer coefficients for existing packings and due to the general structure; the equations can be applied to small compact elements as well as new packings to be developed in the future.

The shortcomings of this model are that the mass transfer characteristics are correlated to geometrical properties such as area, porosity and the number of elements per unit volume. These variables are not sufficient to describe the shape of the packing. It is quite possible that two packings having different shape can be identical in these characteristics. The effect of the shape is not taken into account in the model. Since the present study concentrates on developing a relationship that takes shape into account, the "cylinder model" was not used in the present work for predicting the properties of new packings.

2.5 Combined particle-pipe model for predicting pressure drop

Stein (1998) used a combined particle-pipe model for predicting the pressure drop of gas flow in dumped and structured sphere packings. To calculate the pressure drop of any packing, the theoretically developed particle-pipe model for sphere packings is adjusted with a parameter called the tortuosity factor. It is defined as the ratio of the mean length of the streaming way (actual path followed by the liquid or gas) to the shortest length of the streaming way (i.e., the height of the packing). The maximum tortuosity factor of 2.0 was found for the Raschig ring and the value diminishes for increasingly open packing like the Pall rings. The lowest tortuosity factor among random packings was found to be for the Raschig Super ring.

2.6 Model by Billet and Schultes

The approach by Billet and Schultes (1999) is semi-empirical and is based on physically proven conditions of fluid dynamics. This model takes into account the kinetic laws of mass transfer in packed columns with dumped or arranged packings.

The Billet and Schultes model is a physically proven model for calculation of the performance parameters of columns with dumped or arranged packings. It is possible to determine the mass transfer efficiency, the pressure drop, the column hold up and the load limits based on a uniform theory. The results agree with one of the largest databases compiled under the supervision of Billet and includes over 3500 measured data points, more than 50 test systems and is based on measurements of over 70 types of dumped and arranged packings.

The basis of their approach is a model, which assumes that the empty spaces between the packed elements can be replaced for theoretical purposes by vertical flow channels, through which the liquid trickles down. The deviation from this vertical flow channels is expressed by a packing specific shape constant. Since the geometrical shape cannot be expressed in terms of surface area and porosity alone, this packing specific accounts for the shape of the element.

The model takes into account the thermodynamic properties of the system while calculating the mass transfer efficiency. The loading capacity of the dumped packing determines the column diameter and thus the flow velocity and residence time of the phases in the mass transfer apparatus.

The geometric surface area of the column packing and its ability to create a turbulent flow of phases also decides the effectiveness of mass transfer between phases. Since this is directly dependent on the shape of the packing, the model accounts for the shape in the form of a packing specific shape constant.

The volumetric mass transfer coefficients are dependent on the densities of phases, viscosities and the surface tension, the phase flow velocities, the specific surface area of the packing and the void fraction as well as the packing specific constants. Different correlations are used for conditions above and below the loading conditions and take into account the interfacial area above the loading point and near the flooding point.

On the basis of the theoretical model, the dry pressure drop across the packing can also be calculated. It is dependent on the geometric surface and the void volume of the column packings, the gas load factor and the wall factor that takes into account the increased void fraction at the column wall. For dry pressure drop, the resistance coefficient is dependent upon the Reynolds number of the gas stream.

When packings are irrigated, the free cross section for the gas flow is reduced by the column holdup, and the surface structure is changed. The column holdup is taken into account while calculating the irrigated pressure drop and the resistance coefficient of the two-phase flow is dependent on the Froude number of the liquid flow.

In an earlier model by Billet and Schultes (1991), the dependence of the resistance coefficient on the liquid load was expressed as a function of the Reynolds number. In the present model, it has been suggested that the Froude number describes the dependency of pressure drop on the liquid load better at high pressures and low liquid viscosities.

The model also allows the calculation of the load limits and the flooding conditions. The basis of the calculation is that below the loading point the liquid film trickles from top to bottom without the influenced of the gas counter flow. The column holdup is then a function of merely the liquid load. With increasing gas velocity, the shearing stress at the surface of the liquid increases so that the velocity at the phase boundary drops to zero. Again allowance is made for the packing type using a packing specific shape constant.

The average relative deviation for calculating the loading and flooding points in this model is 5% and for the column hold up it is 6.7%. The deviations for the pressure drop and the mass transfer efficiency are 9.1% and 12.4% respectively.

Because of the small deviations between actual and predicted data, this model was selected for calculating the mass transfer and pressure drop for existing packings

2.7 Defining phase distribution using properties of packed beds

The porosity distribution in the packings is dependent upon the shape and size of the packing. Recently models have been developed for prediction of pressure drop based on the porosity distribution rather than the porosity itself. Giese *et al.* (1998) obtained the superficial velocity profiles for liquid flow for monodisperse packings of spheres, deformed spheres, cylinders and Raschig rings by averaging the axial flow components within a cross section. Porosity distribution functions along the radial direction were also evaluated experimentally.

Giese *et al.* (1998) then extended the modified Brinkman equation by replacing the mean porosity with the porosity distribution function along the radial direction. This model was then used to predict the pressure drop and the average velocity profiles across a cross section of the bed.

Crine *et al.* (1992) introduced the concept of statistical hydrodynamics, in which all the local hydrodynamic quantities were considered as random variables (such as packing properties like porosity distribution). It is suggested in the work that the global hydrodynamics could be described by the local hydrodynamic parameters through a proper probability density function. The probability density function takes into account the structure of the bed which is defined using porosity distribution

The porosity and its distribution in a packed bed are important parameters that determine the flow distribution. To achieve a quantitative understanding of the porosity distribution in packed beds, efforts have been made in the past and correlations to this effect have been developed. It has been found that the mean porosity and the porosity distribution are determined largely by particle size, shape and particle surface properties (i.e., roughness and hardness) as well as the method of packing the bed.

To implement the complex geometry of the packed bed in the flow equations earlier researchers used the mean porosity or the longitudinally averaged porosity profile or empirical quantities. The exact porosity structure is completely changed with re-packing even with the same packing elements and the same packing method. However, one could obtain similar statistical quantities of the bed porosity distribution even after re-packing. One could generate a porosity distribution with the same statistical characteristics under certain constraints and use such a distribution for further calculations.

Gamma ray tomography experiments have been conducted in the recent past to provide the 3-D structure of packed beds and to detect any spatial patterns (Wang *et al.*, 2001). It was found that the type of porosity distribution depends on the size of the voxel. The porosity distribution is Gaussian if the voxel size is larger than the particle diameter.

In the present thesis, the porosity distribution for different types of packings was found using the Sutherland-Hodgman polygon clipper algorithm applied in the work by

Nandakumar *et al.* (1999). Computer simulations are used for the prediction of geometrical properties.

Jiang *et al.* (2001) used the statistical nature of the porosity distribution in which the beds have been characterized by a psuedo-Gaussian function and provided a correlation between the standard deviation of the liquid hold-up and the standard deviation of the porosity distribution. Extensive use has been made of the statistical properties such as skewness and kurtosis (for defining the distribution), which are different for different type of packings. It was also shown in this work that the skewness and kurtosis are different for packed beds having the same mean porosity but having different shape.

2.8 Statistical quantities

Since the porosity distribution in a packed bed is random or pseudorandom in nature statistical quantities can be used to determine the distribution. As pointed out by Jiang *et al.* (2001) four parameters can be used for defining a distribution. The probability density function has been characterized by moments such as mean, variance, skewness and kurtosis. The definitions of these quantities are available in literature. Richmond (1964) defines these moments in a physical sense. In mechanics, the moment represents a torque computed by summing the product of rotation producing force times their respective distances from the fulcrum. In statistics, the distances correspond to the deviations from mean and the forces to the weights or frequencies.

Mean and variance are well-defined quantities. Skewness is defined as the shape or lean of the distribution towards the negative or positive side of the mean. A negative value of skewness means that the distribution is skewed towards the left hand side and a positive value indicates that the distribution is skewed towards the right. The fourth moment, kurtosis, is a measure of the "peakedness" of the distribution. Peakedness refers to the shape of a curve and a measure of peakedness describes the degree to which the curve tends to be pointed or peaked. It has a high value when the curve is peaked and a low value when it is flat topped. Thus it has a high value when the there is a large concentration of observations in the center and a low value when the curve is more spread out. Peakedness and variation are closely related but since the measure of peakedness is a relative measure, i.e., normalized over variance, kurtosis defines how peaked a curve is for a given amount of variation.

2.9 Other shape defining techniques

Shape description refers to methods that result in a numeric descriptor of the shape. A shape description method generates a shape description vector. The goal of the description is to uniquely characterize the shape using its description vector. The required properties of the scheme are invariance to rotation, translation and scale. Shape descriptions have been done using Fourier transforms and are based on the well-developed theory of Fourier analysis. Stochastic methods and autoregressive models have also been used in shape description and for representing the boundary of a two dimensional shape. These techniques can be used for describing and modeling the individual shape. However, most of these shape-defining techniques have been confined to two-dimensional shapes. These techniques have been discussed in the review paper by Loncaric (1998).

In the current thesis, parameters like the statistical distribution functions discussed earlier have been used for prediction of properties for new elements. These parameters describe a bed of number of elements that have been packed randomly and contain information that can be repeated even after repacking the bed.

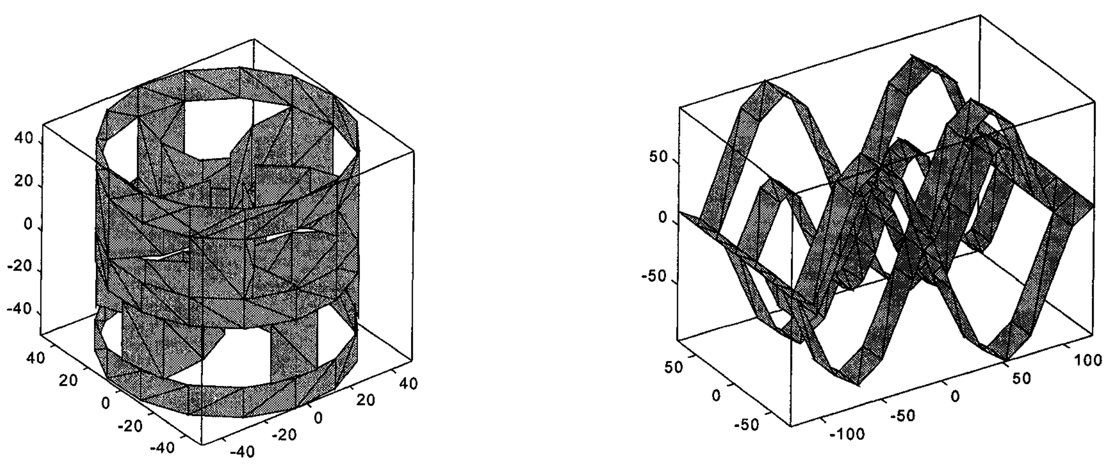
Chapter 3

Packing Algorithm and the Extraction of Geometrical and Statistical Properties

3.1 Packing Algorithm

3.1.1 Features of the packing algorithm

Packings are designed with the objective of maximizing the effective area and minimizing the resistance offered to the flow. The packing objects are then dumped into a column in a random manner and the mean properties of packed bed such as number of pieces per unit volume, specific surface area and the mean porosity are measured. These quantities are then used in prediction of the transport properties using empirical correlations.



Pall Ring

Raschig Super Ring

Figure 3.1: Triangulation of packing shapes for input into the packing program

The algorithm developed by Nandakumar et al (1999) allows one to pack elements of any arbitrary shape into a container. The surface of the packing object is converted to a three-dimensional polygonal object presented by a set of triangular surfaces (Figure 3.1). The dynamics of the element falling into the container are not captured but the final equilibrium position of each element is made available.

The algorithm places the elements such that the center of gravity of the elements is at its lowest and avoids collision between objects and yet places it close to its neighbors. The orientation of the object is selected in a random manner and this becomes the local co-ordinate system for the element.

The element is then lowered into the global co-ordinate system and the lowest position in the z direction of the global co-ordinate system is found. This objective of finding the lowest position z for a given initial position and orientation (x, y, α, β) is accomplished by the binary search algorithm and finding the overall lowest position possible (x, y, α, β) is accomplished with the modified conjugate-gradient method. The details are shown in Figure 3.2.

Since only z is varied in the search step for a randomly selected (x, y, α, β), it does not guarantee closeness of packing objects in the (x, y) plane. The single objective function cannot lead to a closest packing object set in the container. This problem is addressed using a multiple-objective optimization algorithm. It is accomplished by starting with N packing elements with random but conflict free initial positions and orientations and finding the lowest position. After this set of packing elements is created, the points with greatest value of r (where $r^2 = x^2 + y^2$) are retained. This procedure packs the packing elements at a given layer from the container wall to the center.

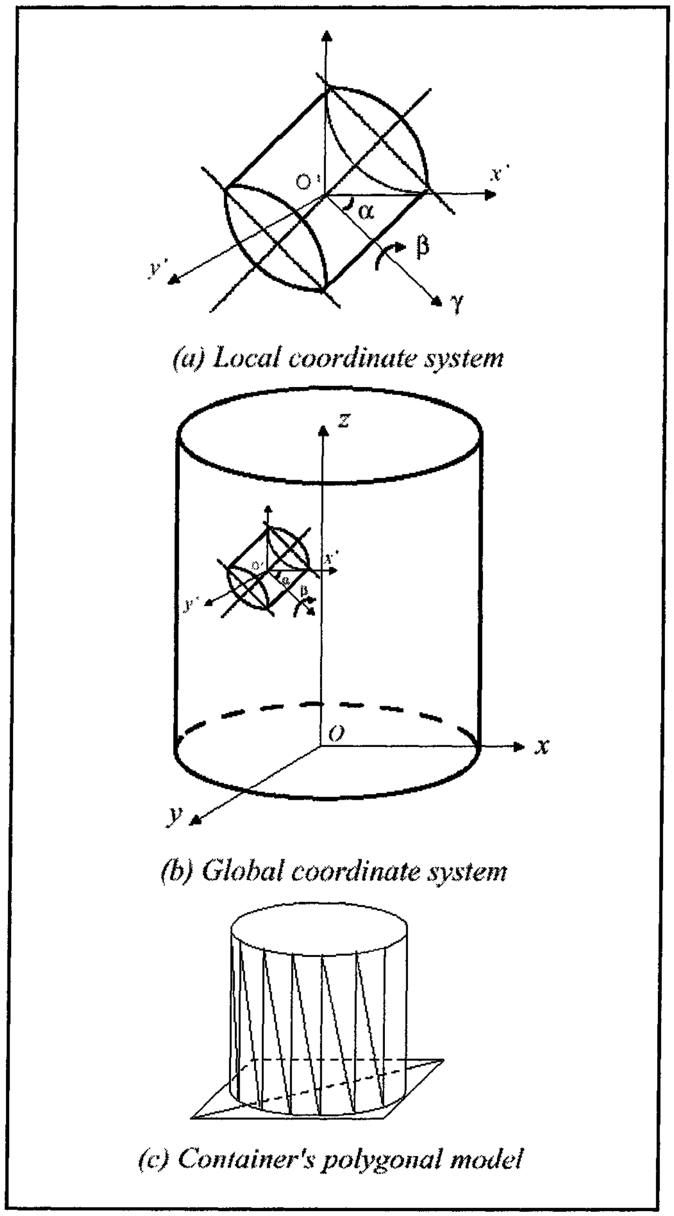


Figure 3.2: Local and global co-ordinate systems for packing element

To eliminate the combinatorial explosion of the possible positions only the top two layers are considered for conflict resolution and packing the elements close to one another. The outcome of the simulation set is the elements' final orientation and position which can be used in subsequent analysis such as porosity (distribution), surface area and other geometrical and statistical properties.

3.1.2 Calculation of geometrical quantities

Porosity is defined as the ratio of the void volume inside a three dimensional measurement space to the total volume of the space. If the measurement space is taken as the total volume occupied by the packing, it gives the mean porosity of the bed. This along with the number of packing elements per unit volume and the specific surface area (surface area per unit volume) are the volume-averaged quantities generally reported.

Since the packing algorithm generates the details on the location and orientation of each element from the simulation, the variations can be reconstructed by choosing the measurement volume in the form of a cuboid as depicted in Figure 3.3. This requires

a) defining the location (r, θ, z) and the size δr, $\delta\theta$, δz of the measurement space
b) locating and clipping the packing elements that lie within the measurement space
c) measuring the solid volume and the surface area of the packing elements that lie within the volume.

Since the cube shape is not well suited for porosity measurement in a cylindrical container it will introduce an error, especially towards the wall of the container. In experimental measurements, the sample space is typically circular rings, which provide circumferentially averaged values of geometrical properties. The center point location of this sample volume (r, θ, z) is varied over $r \in [r_i, r_f]$, $\theta \in [\theta_i, \theta_f]$, $z \in [z_i, z_f]$. The measurement subspace is still approximated by a cuboid and thus it is essential to have a fine resolution in the θ direction.

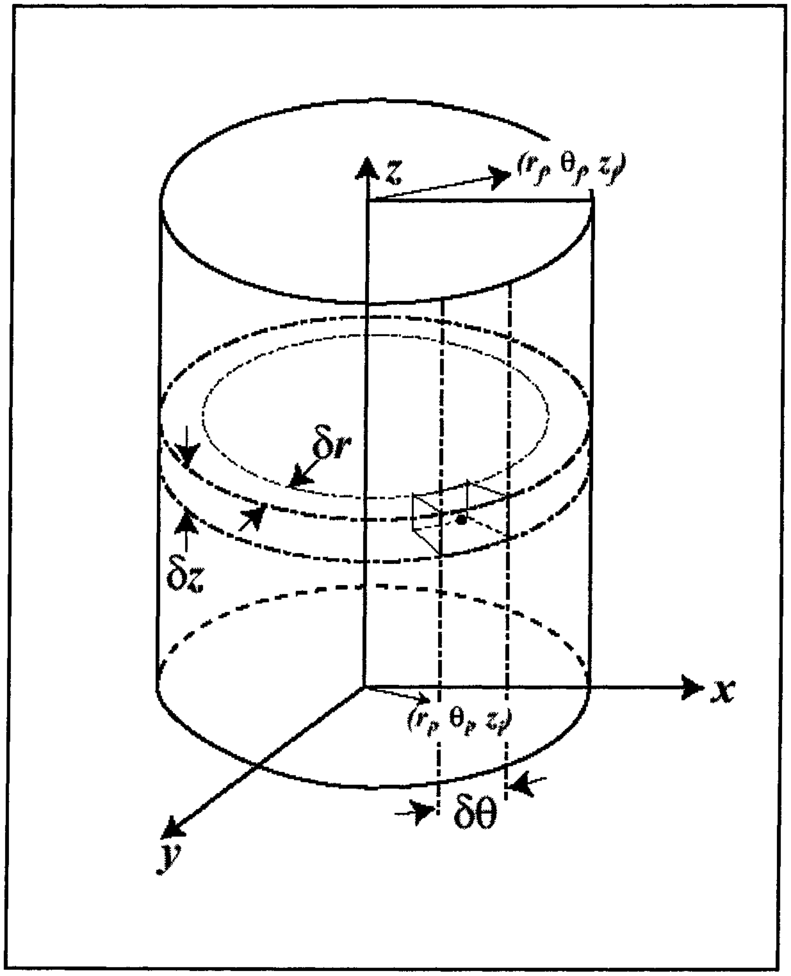


Figure 3.3: Measurement volume in the form of a cuboid for porosity distribution

The volume of material inside the sample volume is estimated by identifying the elements or parts of elements that are inside the measurement volume. The parts of the element that lie outside the measurement subspace are clipped (or cut off).

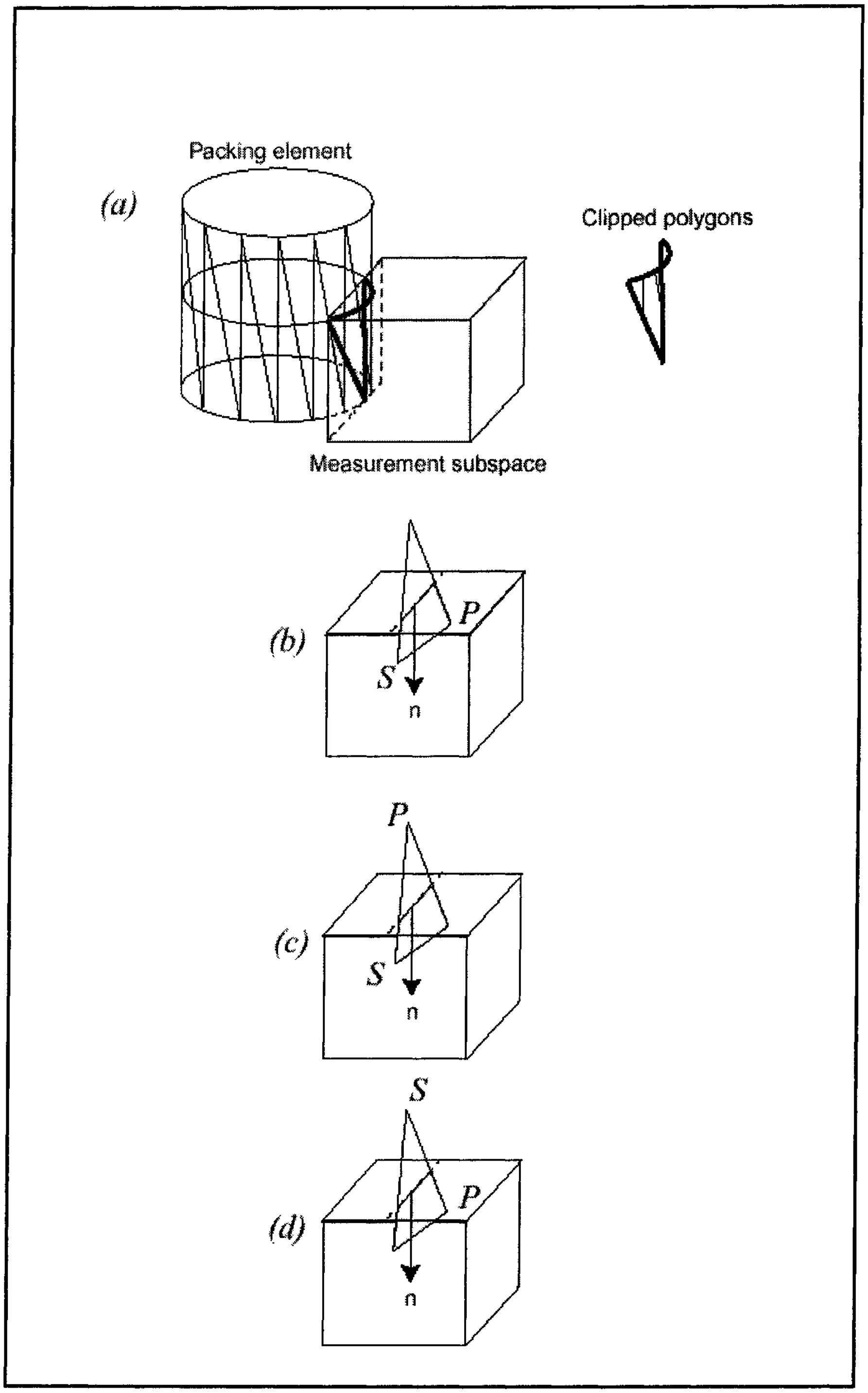


Figure 3.4: Clipping Algorithm to measure the amount of solid volume in the measurement sub-space

Since the general geometrical problem of clipping curved surfaces in three-dimensional space is difficult, the triangulated data structure of the packing elements comes in handy to construct the polygonal model of the element/s in the measurement subspace. The clipping methodology is shown in Figure 3.4a.

The Sutherland-Hodgman Polygon-Clipper Algorithm, which is a three-dimensional algorithm that clips a general convex polygon with any number of planes, is used for the purpose of generating the solid volume inside a measurement space. From the algorithm, the most important step in porosity calculation is the determination of total solid space of packing elements inside the porosity measurement subspace. The Clipper Algorithm can be represented as follows:

For each edge of a triangular plane, ***S*** is the starting point of the edge and ***P*** is the end point of the edge –

- If ***S*** is inside and ***P*** is inside append the edge ***SP*** for computing the solid volume inside the measurement subspace (Figure 3.4b)
- If ***S*** is inside and ***P*** is outside compute and append the intersection point of the edge ***SP*** with the clipping plane for computing the solid volume inside the measurement subspace (Figure 3.4c)
- If ***S*** is outside and ***P*** is inside compute and append the intersection point of the edge ***SP*** with the clipping plane, then append the second node ***P*** for measurement (Figure 3.4d)

The clipped polygons are decomposed to sets of triangles and the surface area is computed. Since most of the modern packings and the new packings dealt with here are open type packings, thickness of the element needs to be provided for the estimation of the solid volume.

3.2 Basis of selection of the sample size

To extract the variations in the structural changes in these properties within the bed on a scale comparable to the pore scale, measurement volumes of the same order of magnitude as the packing object size should be considered. Thus for each of the packing element, the sample size was selected to be same as the major dimension of the packing which would ensure two things. First, within the same element shape, it would allow one to extract the variations in porosity distribution for different sizes. Second, any porosity variation in the radial, axial and circumferential directions can be attributed to the characteristics other than the just the size within the shape. As pointed out by Bornhutter and Mersmann (1993), the mass transfer efficiency is not the function of the geometrical area alone. There are other factors that influence the effective area available for mass transfer, one of them being the number and orientation of pins on the packing element. It is expected that the defining parameters of the porosity distribution would capture this aspect.

The sample size has been selected to be of the same order of magnitude as the packing element size but the increment in the (r, θ, z) directions is the same in all the cases. As the sample volume moves in these directions, it would encounter different solid volumes for difference in packing element sizes. This ensures that the distribution is different even if the shape of the packing element is same. This would in essence capture the differences in the distribution that would influence the liquid and vapor flow behavior.

The effect of sampling at different packing element sizes is shown in the Figures 3.5, 3.6, 3.7 and 3.8. The figures show the distribution in the radial direction for Raschig Rings, Pall Rings of nominal size 1" for two different sample sizes. The variation in porosity for a smaller sample size is wave-like since the sample volume will encounter empty spaces between packing elements because the volume of sampling element is small when compared to the packing size. This is not the case when the sample volume is equal to the size of the element. The sample volume would always encounter some solid volume for all increments in the three directions.

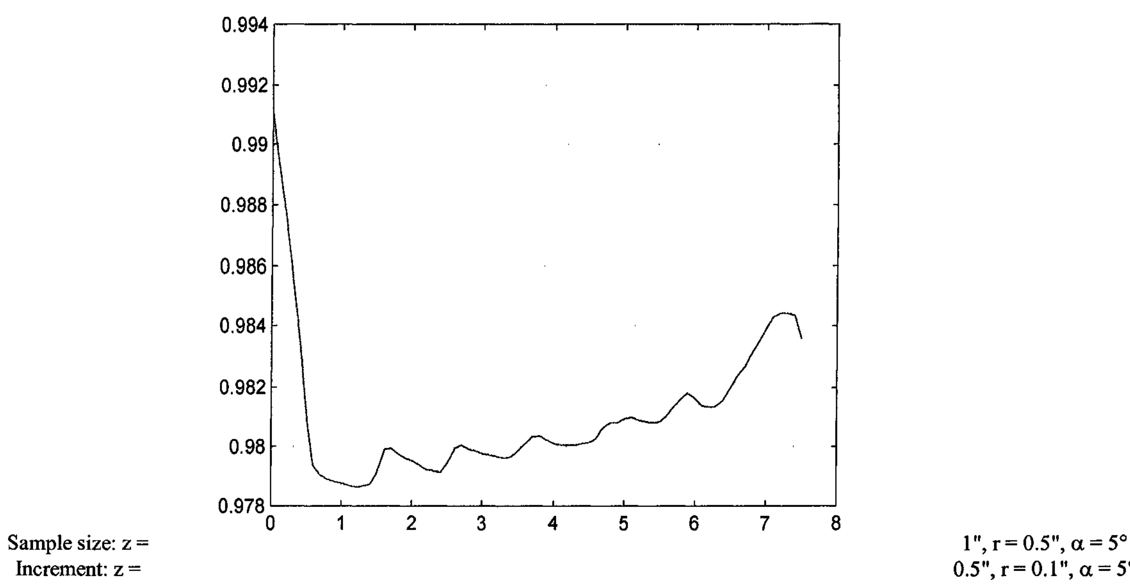
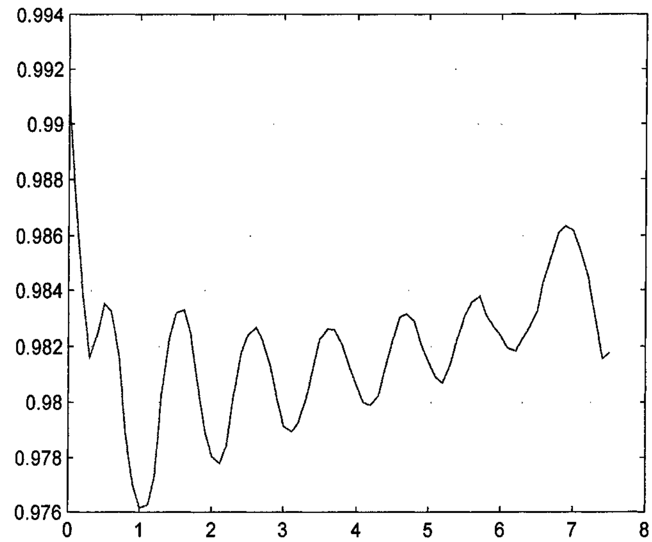
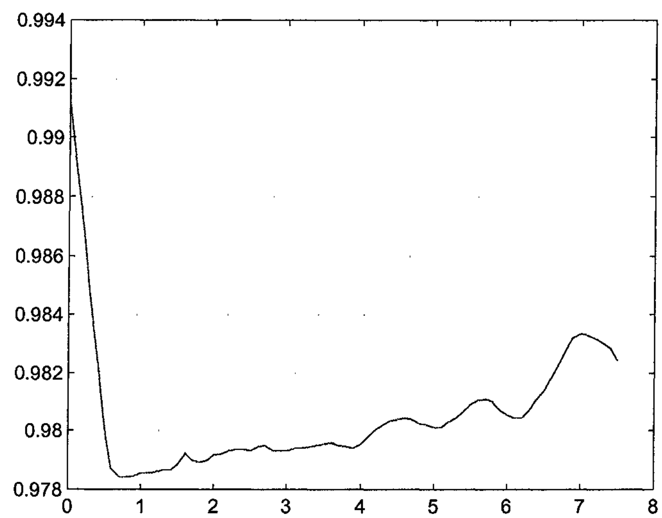


Figure 3.5: Porosity distribution for 1" Raschig Ring in the radial direction



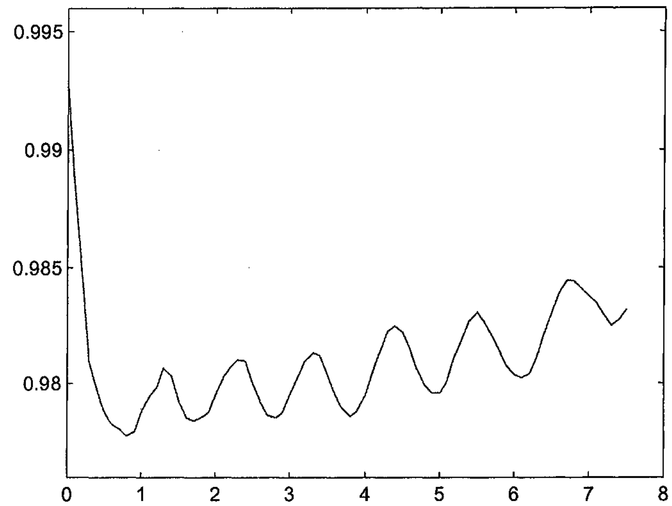
Sample size: z = 0.5", r = 0.25", α = 5°
Increment: z = 0.5", r = 0.1", α = 5°

Figure 3.6: Porosity distribution for 1" Raschig Ring in the radial direction



Sample size: z = 1", r = 0.5", α = 5°
Increment: z = 0.5", r = 0.1", α = 5°

Figure 3.7: Porosity distribution for 1" Pall Ring in the radial direction



Sample size: z = 0.5", r = 0.25", α = 5°
Increment: z = 0.5", r = 0.1", α = 5°

Figure 3.8: Porosity distribution for 1" Pall Ring in the radial direction

3.3 Porosity distribution and the properties of the statistical distribution

Random variation of porosity is an important feature of the random packed column. The porosity distribution has a significant effect on the liquid flow distribution within a packed column. Non-uniform distribution of porosity results in liquid flow maldistribution and a reduced separation efficiency. Since the flow and porosity are random in nature in packed bed, one can use statistical quantities to characterize such randomness or lack there-of of the system. A fundamental task in statistical analysis is to characterize the location and variability of a distribution. A further characterization of the data includes higher order statistical measures such as skewness and kurtosis.

3.3.1 Statistical quantities

The relevant statistical quantities can be described by the probability density function characterized by its moments such as mean (μ), variance (σ), skewness (γ_1) and kurtosis (γ_2). The higher order moments, skewness and kurtosis, would indicate the changes caused by a non-Gaussian probability density function.

The word moment is used in this application because of the similarity to the moments used in mechanics. In mechanics, a moment which represents a torque is computed by summing up the rotation producing force times the distance from the fulcrum or center of gravity. In this application in statistics, the distances correspond to the deviations from the mean, and the forces to the 'weights' or frequencies. Thus the first moment about the arithmetic mean measures the net rotational torque about the mean. Since, the arithmetic mean may be regarded as the balance point, this 'net moment' must be equal to zero. The first moment is always zero because the deviations about the arithmetic mean always cancel. The second moment, the variance, can never be zero (except when all the observations are identical) because of the squaring process, which eliminates all the negative signs. The third moment preserves the signs by using the cubes of the deviations; and is equal to zero when the distribution is symmetrical. In the cubing process the longer tail of the distribution is heavily weighted because although it has fewer elements in it, the deviations are large, and because they are cubed they have more importance than the same total deviation made up of a larger number of smaller deviations, as in the shorter tail.

The third moment is an absolute measure; and to reduce it to a relative measure, as is desirable for a measure of skewness, it is divided by the cube of the standard deviation. Skewness is a measure of the lack of symmetry. A distribution is symmetric if it is distributed equally on the both sides of the mean. Peakedness refers to the shape of the curve, and a measure of peakedness describes the degree to which the curve tends to be pointed or peaked. It has a high value when the curve is peaked and a low value when it is flat-topped. It might seem that the peakedness and variation are closely related but since the measure of peakedness is made a relative measure by using standard deviation as the divisor, the measure of peakedness really describes the shape of the curve. It tells how peaked the curve is for a given amount of variation. The common measure of peakedness is called kurtosis. Data sets with high kurtosis tend to have a distinct peak near the mean, decline rapidly and have heavy tails. Data sets with low kurtosis tend to have a flat top near the mean. A distribution which is considered to be neutral with respect to the peakedness is referred to as mesokurtic, while a more peaked curve is called leptokurtic and a flatter curve is called platykurtic.

Statistical distribution measures for Pall rings of nominal sizes 1" and 2" are shown in Table 3.1 and in Figures 3.9 and 3.10. The distributions are peaked when compared to a normal distribution. It is evident that the 1" Pall ring has a heavy tail towards the right hand side and is skewed towards the right and has a higher value of skewness than the Pall ring of size 2".

Nominal Size	Skewness	Kurtosis
1"	0.87	7.76
2"	0.83	3.48

Table 3.1: Higher order cumulants for the porosity distribution of Pall Ring

Since the distribution for the 1” Pall ring is more peaked than the 2” ring, the value of kurtosis for the 1” Pall ring is much higher.

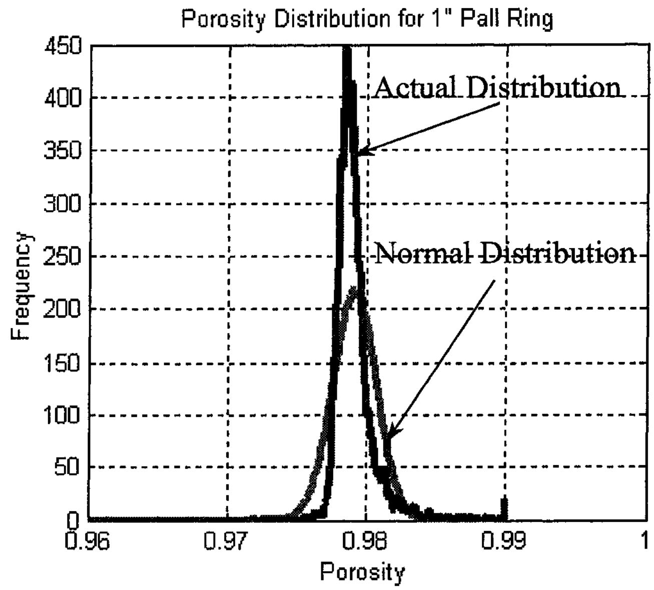


Figure 3.9: Porosity distribution for 1” Pall Ring (overall bed volume)

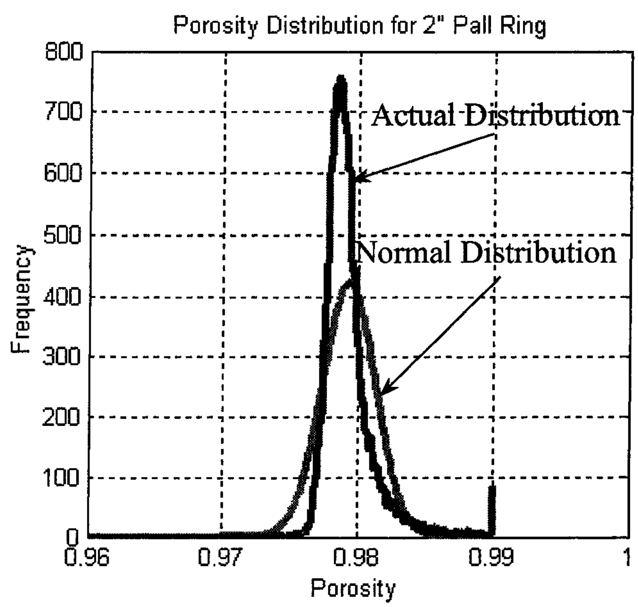


Figure 3.10: Porosity distribution for 2” Pall Ring (overall bed volume)

Nominal Size	Skewness	Kurtosis
Casc. Ring (1")	-0.09	0.51
Super Ring (2")	-0.19	5.43

Table 3.2: Higher order cumulants for the porosity distribution of Cascade Ring and Super Ring

Statistical distribution measures for the 1" Cascade Ring and 2" Super Ring are shown in Table 3.2 and in Figures 3.11 and 3.12. The distribution for the Cascade Ring is close to normal as shown in the figure which explains the values of skewness and kurtosis. But the 2" Super Ring is peaked when compared to a normal distribution. This explains the higher value of kurtosis. The figures for the distributions are shown in the following page.

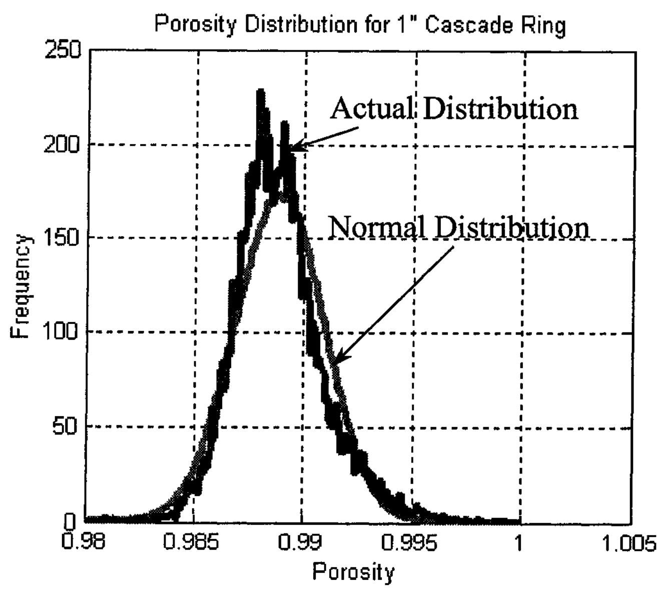


Figure 3.11: Porosity distribution for 1" Cascade Ring (overall bed volume)

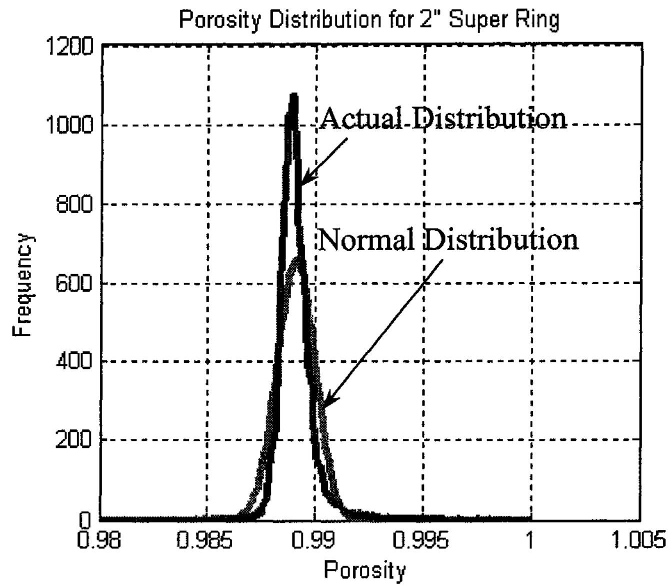


Figure 3.12: Porosity distribution for 2" Super Ring (overall bed volume)

3.3.2 Estimation of statistical quantities

The first order cumulant of a stationary process is the mean,

$$C_{1x} = E\{x(n)\}$$

The second, third and fourth order cumulants are defined as

$$C_{2x} = E\{x(n)\ x(n+k)\}$$

$$C_{3x} = E\{x(n)\ x(n+k)\ x(n+l)\}$$

$$C_{4x} = E\{x(n)\ x(n+k)\ x(n+l)\ x(n+m)\} - C_{2x}(k)\,C_{2x}(l-m)$$
$$C_{2x}(l)\,C_{2x}(k-m)\,M_{2x}(m)\,M_{2x}(k-l)$$

where $\qquad M_{2x} = E\{x(n)\ x(n+m)\}$

The zero lag second, third and fourth order cumulants are called the variance, skewness and kurtosis. Skewness and kurtosis are normalized over standard deviation and are both shift and scale invariant. For univariate data $x_1, x_2 \ldots x_n$, the unbiased estimate of skewness is expressed as

$$\gamma_1 = \frac{\sum_{i=1}^{n}(x_i - \bar{x})^3}{(n-1)\sigma^3}$$

where $\bar{x}$ is the mean, σ is the standard deviation and n is the number of data points. The skewness for a normal distribution is zero and any symmetric data would have skewness near zero. Negative values for skewness indicate data that are skewed left and positive values indicate that the data are skewed right. The expression for the unbiased estimate of kurtosis is

$$\gamma_2 = \frac{\sum_{i=1}^{n}(x_i - \bar{x})^4}{(n-1)\sigma^4}$$

The fourth moment about an arithmetic mean is a measure of peakedness because if two distributions have the same variance, the distribution that is more peaked must have longer tails to compensate for this concentration around the mean and equalize the variances. Thus the more peaked distribution will have more large deviations and also more small deviations (and fewer intermediate deviations) than the less peaked curve.

When these deviations are raised to the fourth power, the larger deviations assume more importance. Therefore the more peaked curve has a higher fourth order moment than the flatter curve. Thus it is the longer tails rather than the actual concentration around the mean which make the kurtosis a measure of peakedness.

If the distribution of x(n) is symmetrically distributed, its skewness is zero (but not vice-versa) and if the distribution x(n) is Gaussian distributed, its kurtosis is zero (but not vice-versa).

3.4 Testing for Normality

Normality of the porosity distribution has been tested using the Jarque-Bera statistic. The Jarque-Bera test evaluates the hypothesis that an input data vector has a normal distribution with unspecified mean and variance, against the alternative that it does not have a normal distribution. The test is based on the sample skewness and kurtosis of a distribution. For a true normal distribution, the sample skewness should be near 0 and the sample kurtosis should be near 3. The Jarque-Bera test determines whether the sample skewness and kurtosis are unusually different than their expected values, as measured by a chi-square statistic.

The test calculates the skewness coefficient to determine if the distribution is symmetric and then measures the kurtosis of the distribution. The Jarque-Bera statistic (JB) is estimated as follows

$$JB = n\left[\frac{\gamma_1^2}{6} + \frac{(\gamma_2 - 3)^2}{24}\right]$$

where n is the number of observations in the sample, γ_1 is the skewness and γ_2 is the kurtosis of the distribution. This statistic has a Chi-squared distribution under the null hypothesis of normality. A symmetric distribution has a skewness coefficient of zero and the kurtosis of a normal distribution is three.

This test is implemented in Matlab in the routine *jbtest.* The routine performs the Jarque-Bera test on the input data vector and returns H, the result of the hypothesis test. The result is: $H = 1$ if the null hypothesis, that the data set has a normal distribution, can be rejected. If the statistic $H=0$, then null hypothesis cannot be rejected and the data tends to have a normal distribution. The hypothesis is rejected if the test is significant at the 5% level. The Jarque-Bera test is an asymptotic test, and should not be used with small samples. For checking the non-normality of the porosity distribution, the packed column was sampled with a sample size in the same range as the packing element size. Since the increment in the θ direction was small for improving the resolution, the data were resampled and the normality test was applied on the filtered data. Filtering was essential as the data tends to have a normal distribution in the θ direction and masks the existing non-normal behavior in the z and r directions. The H-value is shown in Table 3.3 for eight different types of packings, where a value of 1 indicates that the normality assumption can be rejected. This statistic suggests that the porosity distribution for all the types of packings is non-normal.

Strictly speaking the non-normality test is not essential as the normal distribution also has values for skewness and kurtosis. The value of skewness for a normal distribution is 0 and for the kurtosis it is 3. Since the shape of the distribution is affected by the type of packing which in turn affects the flow behavior of the gas and liquid, the test was conducted to see if the data deviated significantly from normality. Very high values of skewness and kurtosis may indicate that the liquid and vapor phases might not be distributed uniformly thus affecting performance. The Jarque-Bera test was used for checking non-normality since it is an asymptotic test and can be used for a large sample size. The sample size in this case was over 5000 data points.

Type of packing	Nominal size	H-Value	Mean porosity
	inch		m^3/m^3
Super Ring	0.3	1.00	0.960
Super Ring	0.5	1.00	0.975
Super Ring	1.0	1.00	0.980
Super Ring	2.0	1.00	0.985
Super Ring	3.0	1.00	0.982
Pall Ring	1.0	1.00	0.954
Pall Ring	1.5	1.00	0.965
Pall Ring	2.0	1.00	0.951
Hiflow Ring	1.0	1.00	0.962
Cascade Ring	1.0	1.00	0.971

Table 3.3: The hypothesis testing for normality for all of the above packings suggests that the distribution is not normal

The skewness and the kurtosis for the existing random packings are given in Table 3.4. The skewness and kurtosis are different for different packings even though the values of the geometrical area are close to each other (for example 1" Cascade and 1" Pall rings).

Hence these characteristic properties can be exploited to develop a relationship between the mass transfer and pressure drop and shape of the packing.

Type of packing	**Nominal size**	**Geometrical area**	**Mean porosity**	**Standard Deviation**	**Skewness**	**Kurtosis**
	Inch	m^2/m^3	m^3/m^3	(*100)		
Super Ring	0.3	315	0.960	0.55	-0.19	5.43
Super Ring	0.5	250	0.975	0.30	0.26	2.08
Super Ring	1.0	160	0.980	0.25	0.21	1.27
Super Ring	2.0	98	0.985	0.18	0.47	2.99
Super Ring	3.0	80	0.982	0.22	0.30	2.05
Pall Ring	1.0	224	0.954	0.28	0.87	7.76
Pall Ring	1.5	139	0.965	0.29	0.94	3.97
Pall Ring	2.0	113	0.951	0.35	0.84	3.48
Hiflow Ring	1.0	203	0.962	0.38	0.99	3.09
Cascade Ring	1.0	233	0.971	0.37	-0.09	0.51

Table 3.4: Higher order cumulants for various random packings

For developing a relationship between the performance parameters and the statistical quantities, the first 9 packings were considered. The Cascade Ring was excluded from building the model because the packing constant for mass transfer was not listed in the

Billet and Schultes (1993) work. Pressure drop for the Cascade Ring was computed and was later used as a validation data point for the Partial Least Squares model developed for the purpose.

Chapter 4

Geometric Properties for New Packings and Response Surface Modeling

4.1 New Packings and mathematical models

Different shapes were considered for the development of new types of packings. The relationship developed between the performance parameters and the shape-based properties was done using all carbon steel packings. The interfacial area available for mass transfer changes with the type of material because wettability characteristics for different materials are different. Contact angles for the liquid are a strong function of the type of the material along with the surface tension of the liquid. The affinity of liquid is different for different types of materials. For this reason, only steel packings were considered so that the model is not affected by this aspect. A general statistical model including the type of material would be too far fetched in this case because there would be an extra factor (material of construction of the packing) to be taken into account. The type of relationship would have to include logical regression parameters. It would make more sense to develop the model for the same type of material than try to generalize every aspect with one regression model.

When developing the new packings, one of the main aspects that was taken into account is the ease of manufacturing these new packings. The existing packings are manufactured from sheet metal (mainly carbon steel) by punching the projections in a specific pattern and then rolling it into the cylindrical shape. All the new packings with the exception of the helicoid can be manufactured in the same way.

Variations in the cylindrical shape of the Pall ring were considered to generate new shapes. Three different kinds of packings were studied under this category. The two-dimensional curves were projected in the *z*-direction to give three-dimensional shapes.

4.1.1 Rhodonea shaped element

The shape shown in Figure 4.1 is the projection of the curve called rhodonea in the z-direction.

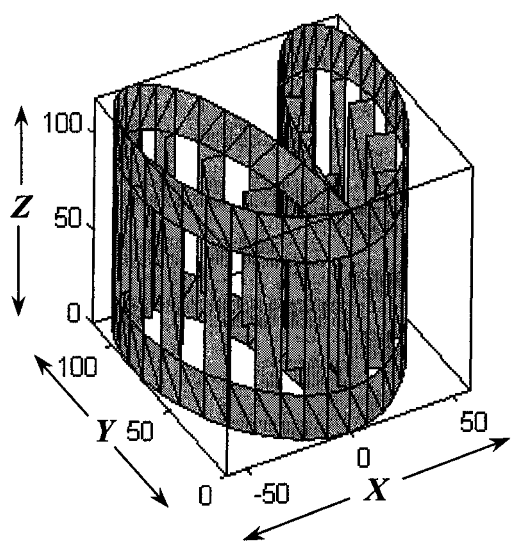


Figure 4.1: Rhodonea shape open type packing

The mathematical representation of the curve in two-dimensions is given by:

$$
\begin{aligned}
r &= \sin(2\theta) \\
x &= ar\sin(\theta) \\
y &= br\cos(\theta) \\
z &= c \\
0 &\le \theta \le \pi
\end{aligned}
$$

The curve was projected in the z direction and the figure was discretized in the form of triangles and the structure made open. The quantities *a*, *b* and *c* can be varied to obtain different sizes for the packing element. Triangulation was required for input into the packing algorithm and the open structure would ensure lower pressure drop which is a feature in all modern packings.

4.1.2 Eight shaped element

The eight-shaped curve shown in Figure 4.2, is a projection of the eight shaped curve in the *z* direction.

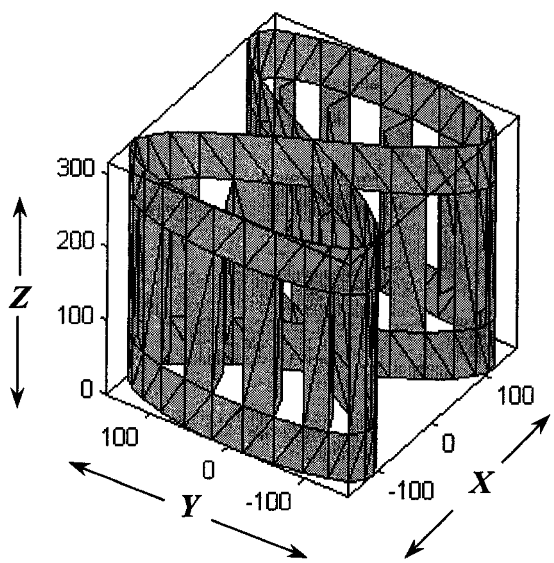


Figure 4.2: Eight shaped open type packing

The mathematical representation of the curve is as follows

$$
\begin{aligned}
x &= b\,a\sin(\theta) \\
y &= c\,a\sin(\theta)\cos(\theta) \\
z &= d \\
-\pi &\le \theta \le \pi
\end{aligned}
$$

a, b and *c* would decide the size of the packing in the *x* and *y* directions and the projection in the *z*-direction, *d*, would decide the height.

4.1.3 Spiral shaped element

The final shape in these types of packings is the spiral shape shown in Figure 4.3. This is a spiral projected in the z direction to enclose a greater surface area than an open cylindrical shape.

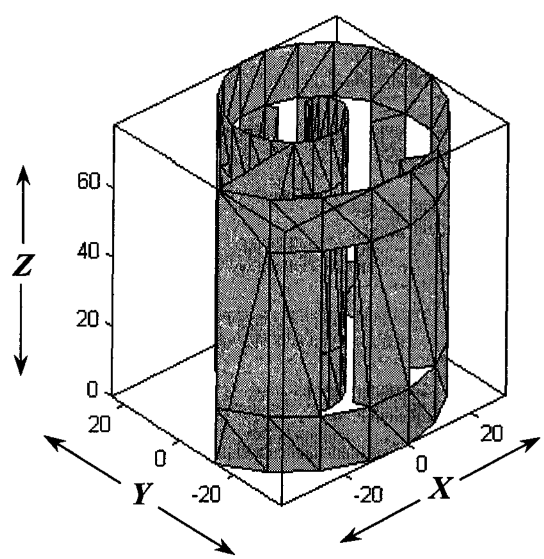


Figure 4.3: Spiral shaped open type packing

The mathematical representation of the shape is given as follows:

$$x = a\theta \sin(\theta)$$
$$y = b\theta \cos(\theta)$$
$$z = c$$
$$0 \leq \theta \leq 3\pi$$

a, b and c would decide the dimension of the element in the x, y and z directions respectively.

4.1.4 Mobius Strip

The packing being discussed as well as the one in the next section were selected because the shape would allow recurrent contact points within the element. The enhanced contact would result in an increased mass transfer coefficient. The effect of the shape on the pressure drop would be predicted from the model.

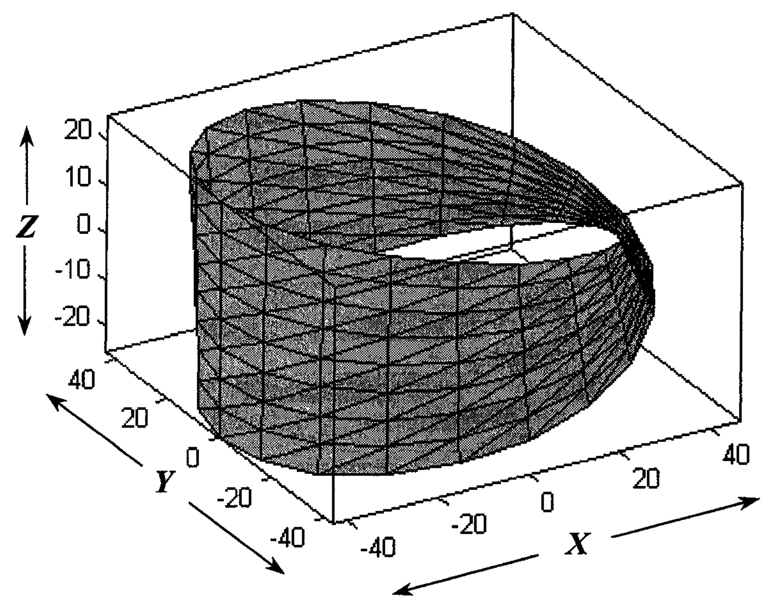


Figure 4.4: Mobius Strip

The shape shown in Figure 4.4 is known as the Mobius strip. It is very similar to a Raschig ring but one end is rotated by 90° before joining the other end.

The parametric equations for the Mobius surface are given by

$$x = a(\cos\theta + v\cos(\theta/2)\cos\theta)$$
$$y = b(\sin\theta + v\cos(\theta/2)\sin\theta)$$
$$z = c\ v\sin(\theta/2)$$
$$0 \le \theta \le 2\pi, -1 \le v < 1$$

a, b and c would decide the dimension of the element in the x, y and z directions respectively.

4.1.5 Helicoid

Another shape in this type of objects is the helicoid shape which is shown in Figure 4.5 below

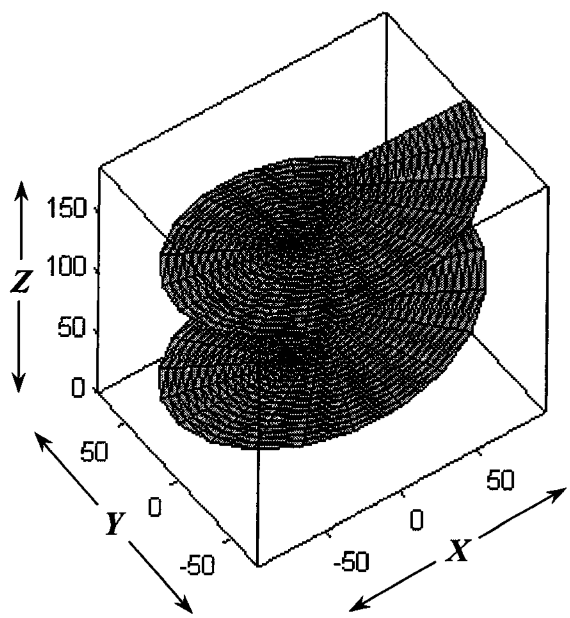


Figure 4.5: Helicoid

The parametric equations for the helicoid are given by

$$x = a\,v\cos\theta$$
$$y = a\,v\sin\theta$$
$$z = c\,\theta/\pi$$
$$0 \le \theta \le 4\pi, \ -1 \le v \le 1$$

Parameter a decides the dimension in the x and y directions and c would decide the dimension of the element in the z direction.

The last two packings would allow for the liquid rivulet to travel a greater distance on the surface when compared to a simple cylindrical shaped element. This may enhance the residence time and may increase the effective area available for mass transfer.

Different sizes of the packings mentioned in the previous sections were considered to arrive at a packing size equivalent to the existing ones used in the model. For selecting the size of the packing, different sizes of the new packing were considered together with the dependence of the surface area and porosity on the dimensions of the packing in cartesian co-ordinates. These studies were done using Response Surface Methodology (RSM) which is discussed in the subsequent sections.

4.2 Features of Response Surface Methodology

Response surface methodology (RSM) was developed by Box and Wilson to aid in the improvement of the manufacturing processes in the chemical industry. The response is a continuous variable (e.g., cost, yield and in this work area), and the mean response is a smooth but unknown function of the levels of factors (e.g., temperature, pressure and in the current work the dimensions of the packing). Factors are the independent variables that control the response (a dependent variable) and the levels are the high and low values of the factors which are varied to study the effect on the response. The mean response when plotted as a function of the treatment combinations (or various levels of factors) is called the response surface. Figure 4.6 shows the response surface of area of the rhodonea shaped packing versus the varying length and breadth of the packing.

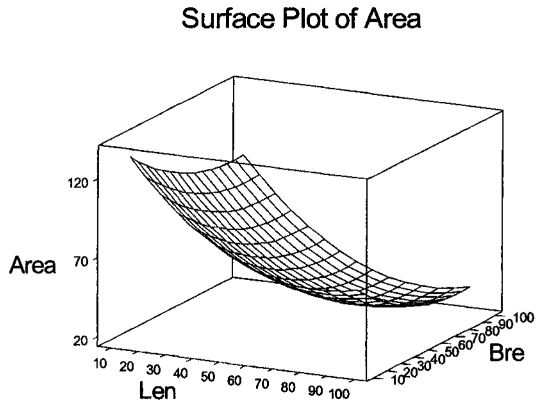


Figure 4.6: Effect of the dimensions on the area of the Rhodonea type packing

4.2.1 Designs for RSM

First order designs are used for fitting models linear in the independent variables and the models are called first order models. These designs consist of n_f factorial points and n_0 center points. The factorial points consist of the treatment combinations of a 2^p factorial experiment run as a completely randomized design. The center points are observations collected at the center of the local region under study. The observations at the center points are repeated to ascertain the experimental error (or pure error) and provide adequate power for a test of lack of fit of the model. The first order models can be represented as follows:

$$Y_u = \beta_0 + \beta_1 x_{u1} + K + \beta_k x_{uk} + \varepsilon_u$$

where Y_u denotes the observed response for the uth trail, x_{ui} represents the level of factor i at the uth trail, β_0 and β_i, $i = 1,2,\ldots,k$ are unknown parameters and the error variables ε_u are assumed to be independent with $N(0,\sigma^2)$ distribution. The parameter β_i is a measure of the local linear effect of the ith factor.

For higher order models, second order designs are required which can detect the curvature in the response surface. In the current work second order designs have been used exclusively to fit second order models. The response was expected to have curvature which necessitates second order models and thus second order designs. The standard second order model is represented as:

$$Y_u = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \beta_{ii} x_i^2 + \sum_{i=1}^{k} \beta_{ij} x_i x_j + \varepsilon_u$$

where Y_u denotes the observed response for the uth trail, x_{ui} represents the level of factor i at the uth trail, β_0 and β_i, $i = 1,2,\ldots,k$ are unknown parameters and the error variables ε_u are assumed to be independent with $N(0,\sigma^2)$ distribution. The parameter β_{ii} represents the quadratic effect of the ith factor and β_{ij} represents the interaction effect between the ith and jth factors.

The second order design should allow for efficient estimation of the response surface; allow for a test of lack of fit for the second order model and also allow for efficient estimation of all model parameters. Second order designs must have at least $(p+1)(p+2)/2$ distinct design points (p is the number of parameters) otherwise not all the parameters can be estimated.

Central composite designs consist of a standard first order design with n_f orthogonal factorial points and n_0 center points, augmented by n_a axial points. Axial points are points located at a specified distance α from the design center in each direction on each axis defined by the coded factor levels. This is pictorially represented in Figure 4.7. +1 represents the high level and –1 represents the low level.

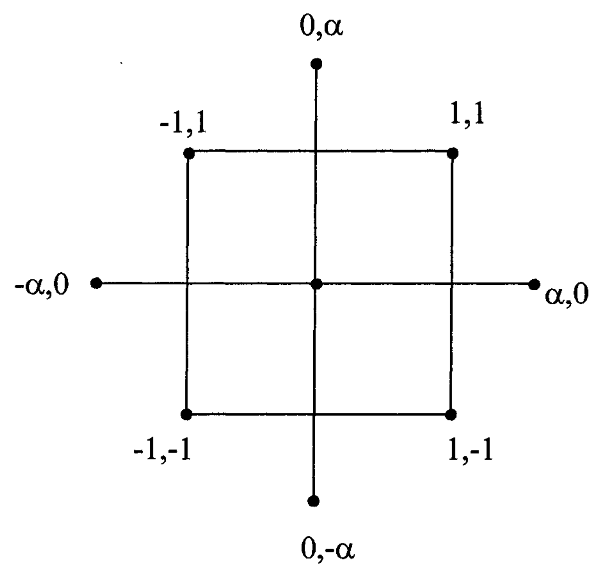


Figure 4.7: Experimental points for Central Composite Design

Two axial points are located at a specified distance α from the design center in each direction on each axis defined by the coded factor levels. The above figure shows four axial points for two factors. The value of α is chosen as

$$\alpha = F^{1/4}$$

where F is the number of points in the factorial region of design. In the above example the number of points in the factorial region is 4. So the value of α for two factors is $\sqrt{2}$. This value of α allows the design to be rotatable. The rotatability property of a design allows equal variance in all directions and provides the direction of subsequent experimentation. The central composite design (CCD) was used to generate experimental points.

4.2.2 Analysis of variance for a design

The total variation in a set of data is called the total sum of squares (*SST*). The quantity *SST* is computed by summing the squares of deviations of the observed Y_u's (the observed variables at different levels of factors) about their average value.

$$SST = \sum_{u=1}^{N} (Y_u - \bar{Y})^2$$

where $\bar{Y}$ is the mean.

The quantity *SST* is associated with *N-1* degrees of freedom where *N* is the total number of observations. It can be partitioned into two parts; the sum of squares due to regression (sum of squares accounted by the fitted model) and the sum of squares unaccounted by the fitted model. The sum of squares due to regression is computed by

$$SSR = \sum_{u=1}^{N} (\hat{Y}(x_u) - \bar{Y})^2$$

The deviation $(\hat{Y}(x_u) - \bar{Y})$ is the difference between the value predicted by the fitted model for the *u*th observation and the overall average. The degrees of freedom for *SSR* are *p-1* where *p* is the number of parameters.

The sum of squares unaccounted by the fitted model is given by

$$SSE = \sum_{u=1}^{N} (Y_u - \hat{Y}(x_u))^2$$

This quantity is called the sum of squares due to error (*SSE*) or the residual sum of squares. The degrees of freedom associated with it is *N-p*.

The residual sum of squares (*SSE*) is partitioned into the sum of squares due to pure error (SS_{PE}) and sum of squares due to lack of fit (SS_{LOF}).

The sum of squares due to pure error is represented as

$$SS_{PE} = \sum_{l=1}^{m} \sum_{u=1}^{n} (Y_{ul} - \bar{Y}_l)^2$$

where Y_{ul} is the *u*th observation at the *l*th design point, where $u = 1,2,\ldots,n$; $l = 1,2,\ldots,m$ and $\bar{Y}_l$ is the average of *n* observations at the *l*th design point.

The sum of squares due to lack of fit is found by subtraction

$$SS_{LOF} = SSE - SS_{PE}$$

The test of the null hypothesis on the adequacy of fit (or lack of fit is zero) involves calculating the *F*-ratio

$$F = \frac{SS_{LOF} / (n-p)}{SS_{PE} / (N-n)}$$

and comparing the value in the above equation with a table value of *F*. Lack of fit can be detected at the α level of significance if the value of *F* in the above equation exceeds the table value, $F_{\alpha,n-p,N-n}$, where the latter quantity is the upper 100α percentage point of the central F-distribution.. If the F-statistic in the above equation possesses a central F-distribution, then the model used is the true model.

4.2.3 Response surface methodology for decision on the size of new packings

Response surface methodology was used for fitting the geometrical area as a function of the dimensions of the packing. For each of the new elements, packing was carried out using the packing program for different aspect ratios. Aspect ratio is defined as the ratio of height to the diameter. The Central composite design was used to determine the high, low and axial point values for the dimensions (Details of this type of design are given in Section 4.2.1). Least squares fit was constructed from the data points with area as the response variable and the dimensions (diameter and height of the packing element) of the packing element as the factors. For each case, the probability of the lack of fit (LOF) statistic was checked and the factors were changed to increase the probability associated with the lack of fit statistic above the significance level. From the least squares fit an area was selected for each of the packing and the corresponding packing element dimension was calculated. The aspect ratio of the packings was maintained as 1. The linear-fit equation correlating the area to the dimensions of the new packings and the Analysis of Variance (ANOVA) table depicting the lack of fit are given in the subsequent paragraphs. The definitions for the 'sum of squares' statistic shown in Tables 4.1 to 4.5 have been described in Section 4.2.2. The ANOVA table and the Lack of Fit statistic aid in determining if the fitted equation is an adequate representation of the actual relation between the dependent and independent variables or not. The probability associated with the F-statistic for the Lack of Fit indicates the level of confidence one can have in the fitted equation.

4.2.3.1 Rhodonea shaped element

The fitted equation is of the form:

$$a = 308.397 - 4.469 * l - 0.832 * b + 0.026 * l^2$$

where a is the area and l is the dimension in the x-direction and b is the dimension in the y-direction as shown in Figure 4.1.

Source	DF	Seq SS	Adj SS	Adj MS	F	P
Regression	3	30230.6	30230.6	10076.9	39.72	0
Linear	2	26669.9	13912.2	6956.08	27.42	0
Square	1	3560.7	3560.7	3560.68	14.03	0.002
Residual Error	13	3298.4	3298.4	253.72		
Lack-of-Fit	5	2414.2	2414.2	482.84	4.37	0.032
Pure Error	8	884.2	884.2	110.52		
Total	16	33528.9				

Table 4.1: ANOVA table for fitting the geometric area of the Rhodonea element as a function of the dimensions

The ANOVA for the fit shown in Table 4.1 for the Rhodonea element shows that the lack of fit is significant. The fit has been used despite this fact because the predicted area does not deviate very much from the actual geometric area. For selecting the dimension of the element, an area of 180 m^2/m^3 was considered and for an aspect ratio of 1 the packing element dimension is 28.1 mm. For subsequent analysis a size of 30 mm was used.

4.2.3.2 Eight shaped element

The fitted equation is of the form:

$$a = 273.563 - 3.778 * l - 0.545 * b + 0.022 * l^2$$

where a is the area and l is the dimension in the x-direction and b is the dimension in the y-direction as shown in Figure 4.2.

Source	DF	Seq SS	Adj SS	Adj MS	F	P
Regression	3	20705.1	20705.1	6901.7	25.92	0
Linear	2	18538.4	7645.2	3822.6	14.36	0.001
Square	1	2166.7	2166.71	2166.71	8.14	0.016
Residual Error	11	2929	2929.03	266.28		
Lack-of-Fit	5	1555.3	1555.31	311.06	1.36	0.356
Pure Error	6	1373.7	1373.73	228.95		
Total	14	23634.1				

Table 4.2: ANOVA table for fitting the geometric area of Eight shaped element as a function of the dimensions

4.2.3.3 Spiral element

In Table 4.3 the Lack of Fit statistic is very significant. An appropriate fit could not be found for any combination of the variables. So tentatively a size of 30 mm was used for further calculations.

Source	DF	Seq SS	Adj SS	Adj MS	F	P
Regression	2	257.153	257.153	128.577	4.06	0.051
Linear	2	257.153	257.153	128.577	4.06	0.051
Residual Error	10	316.956	316.956	31.696		
Lack-of-Fit	6	311.061	311.061	51.844	35.18	0.002
Pure Error	4	5.895	5.895	1.474		
Total	12	574.11				

Table 4.3: ANOVA table for fitting the geometric area of the Spiral element as a function of the dimensions

4.2.3.4 Mobius Strip

The fitted equation is of the form:

$$a = 400.372 - 8.509 * d + 0.054 * d^2$$

where a is the area and d is the dimension in the x-direction as shown in Figure 4.4. The least squares fit predicts that the height is insignificant. The reason for this is that the geometric surface area is approximately the same even though the height of the packing is different. The lower surface area of a smaller element is compensated by the fact that more elements can be packed within a given volume and thus give the same overall surface area per unit volume. The surface area is relatively insensitive to the aspect ratio of the packing. The linear model is being used only to give an indication of the value of geometric surface area. For example if the selected size is 29 mm as in this case, the dimension is being rounded off to 30 mm (or the nearest multiple of 5). This is because the area would still be within the range that was used for building the predictive Partial Least Squares model (described in the next chapter) with the existing packings. ANOVA table (Table 4.4) shows that the equation predicts the relationship between the area and the dimensions well.

Source	DF	Seq SS	Adj SS	Adj MS	F	P
Regression	2	56697.7	56697.7	28348.8	31.36	0
Linear	1	47205.8	20071.5	20071.5	22.2	0.002
Square	1	9491.9	9491.9	9491.9	10.5	0.014
Residual Error	7	6327.5	6327.5	903.9		
Lack-of-Fit	2	1461.1	1461.1	730.6	0.75	0.519
Pure Error	5	4866.4	4866.4	973.3		
Total	9	63025.2				

Table 4.4: ANOVA table for fitting the geometric area of the Mobius Strip as a function of the dimensions

For a geometrical surface area of 200 m^2/m^3, keeping the aspect ratio as 1, the packing element size calculated was 29 mm. A packing element size of 30 mm was used for subsequent calculations.

4.2.3.5 Helicoid

The fitted equation is of the form:

$$a = 249.4 - 138.3 * h + 23.1 * h^2$$

where a is the area and h is the dimension in the z-direction as shown in Figure 4.5.

Source	DF	Seq SS	Adj SS	Adj MS	F	P
Regression	2	20719.6	20719.6	10359.8	107.65	0
Linear	1	16952.3	8019.45	8019.45	83.33	0
Square	1	3767.4	3767.37	3767.37	39.15	0
Residual Error	10	962.4	962.35	96.24		
Lack-of-Fit	2	379.5	379.52	189.76	2.6	0.135
Pure Error	8	582.8	582.83	72.85		
Total	12	21682				

Table 4.5: ANOVA table for fitting the geometric area of the Helicoid as a function of the dimensions

The least squares fit predicts that the diameter is insignificant. The reasoning is the same as given in Section 4.2.3.4. Elements with smaller diameter have a smaller surface area per element but the number of elements packed per unit volume is more. So the geometric surface area is approximately the same for different diameters. As mentioned before, an indication of the area is sufficient so that the packing size can be selected accordingly. The Lack of Fit statistic shows that the predicted equation depicts the relationship well. For a geometrical surface area of 150 m^2/m^3, keeping the aspect ratio as 1, the packing element size calculated was 21 mm. A packing element size of 25 mm was used for subsequent calculations.

Having an approximate fit to relate the area to the dimensions allows one to decide on the size of the packing that would give a specified area. Though the area of an individual packing element is known, the overall surface area when the elements are packed is difficult to determine because one cannot estimate the number of elements per unit volume. In such cases tools like Response surface methodology allow one to predict approximate relationship for interpolation. As shown in this chapter the use of statistical methods is a better alternative than choosing the size of the packing element randomly. A trial and error procedure is not advised when better methods are available.

Chapter 5

Correlating Performance to Geometric and Statistical Parameters

5.1 Mass Transfer and Pressure Drop model

Mass transfer models in the past years have been designed on the basis of empirical calculation approaches which were based on experimental data, but could not be traced back to any physical context. These relations always resulted in unproven design when extrapolated outside the experimental database. Recent calculations are based on physically proven conditions of fluid mechanics and mass transfer. One such model is by Billet and Schultes (1999) for the prediction of mass transfer efficiency, pressure drop, column hold-up and load limits. This model has been exclusively used in this work to predict the physical properties of existing packings (like Pall rings, Super ring, Cascade ring etc.). Physical properties are used to build a model based on the statistical properties of the porosity distribution. The model was built using Partial Least Squares which is discussed in the next section.

The Billet and Schultes model assumes that the empty space of the dumped packings can be replaced for theoretical considerations by vertical flow channels, through which the liquid trickles evenly downwards while the gas flows upwards. In reality the flow channels deviate from the vertical direction, and the actual direction is determined by the shape of the dumped packing. These flow channels are a function exclusively of the shape of the packing element. The shape and structure of a packing element cannot be defined by the geometrical surface area and void volume (or porosity) alone. To take this factor into account, the model assumes that the deviation of the real flow behavior of the phases from the vertical flow channels in a column can be expressed by a packing specific shape constant. The packing specific shape constants are different for different physical properties.

5.1.1 Mass transfer efficiency

For a given separation task, the height of a mass transfer column is calculated according to the HTU-NTU model, where HTU is the height of a transfer unit and NTU is the number of transfer units. The number of mass transfer units is defined by the concentrations of the incoming and outgoing flows of the material and by the phase equilibrium. It is therefore dependent only on the separation task and the thermodynamic properties. The height of a mass transfer unit is determined by the actual design of the column and the load of the internals. The geometric surface area of the packing decides the effective contact area between the phases and the ability of the packing to create a turbulent flow of the phases decides the effectiveness of mass transfer. If the transfer resistance is mainly situated in the liquid phase, the HTU of the liquid phase is the deciding factor in the mass transfer efficiency of the packing. In the present study, desorption of carbon dioxide from air with water was used for calculating the physical properties, and the mass transfer resistance is mainly in the liquid phase.

The height of a transfer unit (HTU) is a function of the volumetric mass transfer coefficient which is given as

$$\beta_L a_{Ph} = C_L 12^{1/6} \bar{u}_L^{1/2} \left(\frac{D_L}{d_h} \right)^{1/2} a \left(\frac{a_{Ph}}{a} \right)$$

$$\bar{u}_L = \frac{u_L}{h_L}$$

$$\frac{a_{Ph}}{a} = 1.5 (a d_h)^{-0.5} \left(\frac{u_L d_h}{\nu_L} \right)^{-0.2} \left(\frac{u_L^2 \rho_L d_h}{\sigma_L} \right)^{0.75} \left(\frac{u_L^2}{g d_h} \right)^{-0.45}$$

$$d_h = \frac{4\varepsilon}{a}$$

$$h_L = \left(12 \frac{1}{g} \frac{\eta_L}{\rho_L} u_L a^2 \right)^{1/3}$$

ρ_L is the density of the liquid phase, kg/m^3

ν_L is the kinematic viscosity, m^2/s

σ_L is the surface tension, kg/s^2

η_L is the dynamic viscosity, kg/ms

C_L is packing specific shape constant, dimensionless

D_L is the diffusion coefficient, m^2/s

$\bar{u}_L$ is the mean effective velocity, m/s

a is the geometrical surface area, m^2/m^3

ε is the porosity, m^3/m^3

a_{Ph} is the specific interface area between the phases

d_h is the hydraulic diameter, m

u_L is the velocity with reference to the free column cross section, m/s

g is the gravitational acceleration, m/s^2

h_L is the column holdup, m^3/m^3

5.1.2 Pressure Drop Model

The pressure drop in a packed column is dependent on the geometric surface, the void volume and the gas load factor F_V (given below). The model contains a wall factor (K) which takes into account the increased porosity at the column wall. The resistance coefficient for the packings is dependent on the Reynolds number (Re_V, defined in the equations given below). On the basis of the theoretical model, the pressure drop of dry packing can be calculated as follows

$$\frac{\Delta P_0}{H} = \psi_0 \frac{a}{\varepsilon^3} \frac{F_V^2}{2} \frac{1}{K}$$

$$F_V = u_V \sqrt{\rho_V}$$

$$\frac{1}{K} = 1 + \frac{2}{3} \frac{1}{(1-\varepsilon)} \frac{d_P}{d_S}$$

$$\psi_0 = C_{P0} \left(\frac{64}{\mathrm{Re}_V} + \frac{1.8}{\mathrm{Re}_V^{0.08}} \right)$$

$$\mathrm{Re}_V = \frac{u_V d_P}{(1-\varepsilon)\nu_V} K$$

$$d_P = 6 \frac{(1-\varepsilon)}{a}$$

For irrigated pressure drop, the dependency of the resistance coefficient (ψ_L) on the liquid load has been described by the Froude number. The model for the irrigated pressure drop is given as

$$\frac{\Delta P}{H} = \psi_L \frac{a}{(\varepsilon - h_L)^3} \frac{F_V^2}{2} \frac{1}{K}$$

$$\psi_L = C_{P0} \left(\frac{64}{\mathrm{Re}_V} + \frac{1.8}{\mathrm{Re}_V^{0.08}} \right) \left(\frac{\varepsilon - h_L}{\varepsilon} \right)^{1.5} \left(\frac{h_L}{h_{L,S}} \right)^{0.3} \exp\left(C_1 \sqrt{Fr_L}\right)$$

$$C_1 = \frac{13300}{a^{3/2}}$$

$$Fr_L = \frac{u_L^2 a}{g}$$

$\frac{\Delta P_0}{H}$ is the dry pressure drop in Pa per m of the height of the packing

$\frac{\Delta P}{H}$ is the irrigated pressure drop in Pa per m of the height of the packing

ψ_0 *and* ψ_L are the resistance coefficients for dry packing and irrigated packing respectively, dimensionless.

K is the wall factor, dimensionless

ν_V is the kinematic viscosity of the vapor, m^2/s

$C_{P,0}$ is packing specific shape constant, dimensionless

u_L is the mean effective velocity of liquid, m/s

d_P is the packing element diameter, m

u_V is the gas velocity with reference to the free column cross section, m/s

g is the gravitational acceleration, m/s^2

h_L is the column holdup, m^3/m^3

$h_{L,S}$ is the column holdup at loading point, m^3/m^3

Fr_L is the Froude number, dimensionless

The above relations were used to determine the volumetric mass transfer coefficient and the pressure drop. The system for calculating these parameters consisted of desorption of carbon dioxide from water with air at a temperature of 25°C in a column packed with various existing packings has been considered. The mass transfer resistance for this system exists mainly in the liquid phase. An air flow rate of 1500 m^3/h and the molar flow ratio of liquid and gas was fixed at 1.2 and the column was designed to operate at 80% of the capacity at flood point. The physical properties for the liquid and vapor phases were obtained from Billet and Schultes (1993).

Nine different kinds of packing were considered for developing a relation between the porosity distribution and geometric parameters and the performance. These packings were selected from the publication by Billet and Schultes (1999) which has more than 30 different types of packings with the relations to calculate the pressure drop and mass transfer efficiency.

The reason for considering only nine packings was due to the unavailability of the exact shape and size of the rest of the packings. Hence, this database can be improved by including the existing packings. The liquid side mass transfer coefficient and the pressure drop are tabulated in Table 5.1. The relationship of these parameters with the statistical variables is discussed in the next section.

Packing Type	Nominal Size	Mass Transfer Coefficient	Pressure Drop
	inch		Pa/m
Super Ring	0.3	0.0096	474.15
Super Ring	2.0	0.0079	134.10
Pall Ring	2.0	0.0055	115.72
Pall Ring	1.5	0.0064	165.22
Pall Ring	1.0	0.0084	368.96
Hiflow Ring	1.0	0.0099	312.38
Super Ring	0.5	0.0092	324.88
Super Ring	1.0	0.0077	178.08
Super Ring	3.0	0.0051	103.33

Table 5.1: Volumetric mass transfer coefficients and pressure drop for selected packings

5.2 Partial Least Squares (PLS) model for relating statistical parameters and performance characteristics

5.2.1 PLS Theory and Algorithm

Partial least squares regression is an extension of the multiple linear regression model. The linear model in its simple form specifies the linear relationship between a dependent (response) variable *Y*, and a set of predictor variables, *X*'s so that the relation can be expressed in the following form:

$$Y = b_0 + b_1 X_1 + b_2 X_2 + \mathrm{K} + b_P X_P$$

In the above equation b_0 is the regression coefficient for the intercept and the b_i values are the regression coefficients for the independent variables, 1 through p, computed from the data. The same approach has been used in the previous section for building models for packings using Response Surface Methodology. The linear regression model is useful when the predictor variables are uncorrelated. In case they are correlated, then $X^T X$ (the regressor matrix) will not be of full rank and as a result the variance of the regression parameter estimates will be high, indicating low confidence in these estimates.

The multiple linear regression model has been extended in a number of ways to address the more sophisticated data analysis problems. It serves as a model for multivariate methods such as discriminant analysis, principal components regression and canonical correlation. These methods have two important properties in common. They impose restrictions such that

1. factors underlying the Y and X variables are extracted from $Y'Y$ and $X'X$ matrices, respectively, and never from cross-product matrices involving both the Y and X variables
2. the number of the prediction functions can never exceed the minimum of the Y variables and the X variables

Partial least squares (PLS) regression extends the multiple linear regression without imposing the restrictions employed by the above methods. In PLS regression, prediction functions are represented by factors extracted from the $Y'XX'Y$ matrix. The number of such prediction functions that can be extracted exceed the maximum number of Y and X variables. PLS regression is the least restrictive of the various extensions of the linear regression model. It can be used in situations where the use of traditional multivariate methods is severely limited, such as when there are fewer observations than the predictor variables and when the predictor variables are correlated. The data set considered for the predictor variable has two columns which are known to be correlated, the geometric surface area and the porosity. The correlation between other variables is not known.

5.2.1.1 Basic Model

The main purpose of the partial least squares regression is to build a linear model

$$Y = XB + E$$

where Y is an n by m variables response matrix, X is an n by p variables predictor matrix, B is a p by m regression coefficient matrix and E is a noise term for the model which has the same dimensions as Y. The variables in X and Y are centered by subtracting their respective means and scaled by dividing by their respective standard deviations.

PLS regression produces factor scores as linear combinations of the original predictor variables, so that there is no correlation between the factor score variables used in predictive regression model. A regression using factor extraction for this type of data computes the factor score matrix, T, where,

$$T = XW$$

for an appropriate weight matrix W of dimension p by c. The columns of W are the weight vectors for the X columns producing the corresponding n by c factor score matrix T. These weights are computed so that each of them maximizes the covariance between the responses and the corresponding factor scores. Ordinary linear regression is performed to produce Q, such that

$$Y = TQ + E$$

where Q is a matrix of regression coefficients for T and E is the noise term. Once the loadings are computed the above regression model is equivalent to the linear regression model

$$Y = XB + E$$

where $B = WQ$. In this form the model can be used as a predictive regression model.

PLS regression produces the weight matrix W reflecting the covariance structure between the predictor and response variables. An additional matrix which is necessary for the complete description of the PLS regression procedures is the p by c factor loading matrix P which gives a factor model

$$X = TP + F$$

where F is the unexplained part of the X scores.

5.2.1.2 Algorithm

There are two types of algorithms for computing PLS regression components viz., nonlinear iterative partial least squares (NIPALS) and SIMPLS algorithm. The SIMPLS algorithm has been used for in the current work and the algorithm is given as follows:

For each h = 1,2,…,c, where $A_0 = X'Y$, $M_0 = X'X$, $C_0 = I$, and c given

1. compute q_h, the dominant eigenvector of $A_h{}'A_h$

2. $w_h = A_h q_h$, $c_h = w_h' M_h w_h$, $w_h = \frac{w_h}{\sqrt{c_h}}$, and store w_h into W as a column

3. $p_h = M_h w_h$, and store p_h into P as a column

4. $q_h = A_h' w_h$, and store q_h into Q as a column

5. $v_h = C_h' p_h$, $and\ v_h = \frac{v_h}{\|v_h\|}$

6. $C_{h+1} = C_h - v_h v_h'$ $and\ M_{h+1} = M_h - p_h p_h'$

7. $A_{h+1} = C_h A_h$

In the present work PLS has been used to develop a model relating the pressure drop to the statistical properties of the porosity distribution. The variables in the present work are the statistical properties of the porosity distribution like skewness and kurtosis which have been explained in Chapter 3, Section 3.3.1. These along with the area and the porosity are used to develop a relationship with the performance parameters. If one is faced with many variables and ill-understood relationships, multiple linear regression might be inappropriate (as in the case when predictor variables are correlated). In such a case, the approach of using empirical latent variable models is a good alternative to the classical multiple linear regression.

PLS provides a tool for the analysis of the relationship and is used for finding a regressive equation between the performance parameters like mass transfer coefficient and pressure drop and the variables mentioned earlier.

5.2.2 Partial Least squares model for existing packings

The type of packing, size and the independent variables, viz., geometric area, porosity and standard deviation, skewness and kurtosis of the porosity distribution are shown in Table 5.2. A relationship between the variables shown in Table 5.1 and the performance characteristics (mass transfer and pressure drop) was developed for 9 different type of packings. The size of the predictor block (*X*) is 9*5 and the of size response matrix is 9*2.

				Porosity Distribution		
Packing Type	**Size**	**Geometric area**	**Mean porosity**	**Std. Deviation**	**Skewness**	**Kurtosis**
	inch	**m^2/m^3**	**m^3/m^3**	(*100)		
Super Ring	0.3	315	0.960	0.55	-0.19	5.43
Super Ring	2.0	98	0.985	0.18	0.47	2.99
Pall Ring	2.0	113	0.951	0.35	0.84	3.48
Pall Ring	1.5	139	0.965	0.29	0.94	3.97
Pall Ring	1.0	224	0.954	0.28	0.87	7.76
Hiflow Ring	1.0	203	0.962	0.38	0.99	3.09
Super Ring	0.5	250	0.975	0.30	0.26	2.08
Super Ring	1.0	160	0.980	0.25	0.21	1.27
Super Ring	3.0	80	0.982	0.22	0.30	2.05

Table 5.2: Statistical parameters defining the porosity distribution and geometric quantities for existing packings

5.2.2.1 PLS model building

In this study, the X block or the predictor block contains the five variable mentioned above. To ensure that the results are invariant to the choice of measurement units, all variables are auto-scaled (zero mean and unit variance). Cross validation is used to determine the number of latent variables in the PLS model. The cumulative PRESS plot given in Figures 5.1 and 5.2 indicates that 4 latent variables is the ideal choice.

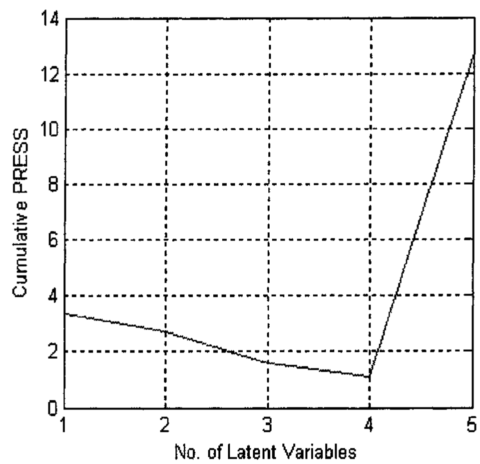


Figure 5.1: CUMPRESS Plot for pressure drop

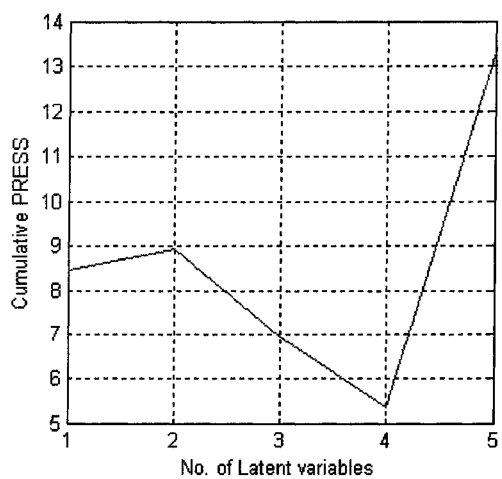


Figure 5.2: CUMPRESS Plot for mass transfer coefficient

	X Block		*Y* Block	
LV #	**This LV**	**Total**	**This LV**	**1.1.1 Total**
1	51.24	51.24	83.24	83.24
2	28.56	79.8	7.62	90.86
3	11.67	91.48	6.3	97.16
4	7.28	98.75	0.96	98.12

Table 5.3: Variance captured by Latent Variables in PLS Model for Pressure Drop

	X Block		*Y* Block	
LV #	**This LV**	**Total**	**This LV**	**1.1.2 Total**
1	48.53	48.53	46.3	46.30
2	28.17	76.70	18.06	64.36
3	12.75	89.45	18.99	83.34
4	8.80	98.24	1.16	84.50

Table 5.4: Variance captured by Latent Variables in PLS Model for Mass Transfer

Tables 5.3 and 5.4 list the variance explained by each latent variable. It can be seen that increase in the variance explained is minor when the number of latent variables increase from 3 to 4.

The coefficient vector for using the PLS model in the predictive form can be found by superimposing the weight vectors (used for projecting the regressor matrix into the latent space) on the matrix of regression coefficients in the latent subspace.

Packing Type	Nominal Size	Pressure Drop (From the Model)	Pressure Drop (predicted using PLS)	% Error
	inch	Pa/m	Pa/m	
Super Ring	0.3	474	473	0.2
Super Ring	2.0	134	131	2.5
Pall Ring	2.0	116	122	5.8
Pall Ring	1.5	165	196	18.9
Pall Ring	1.0	369	363	1.6
Hiflow Ring	1.0	312	284	9.1
Super Ring	0.5	325	337	3.7
Super Ring	1.0	178	189	5.9
Super Ring	3.0	103	82	21.3

Table 5.5: Comparison between the actual values based on Billet Schultes model and predicted values by the PLS model for Pressure drop

For the pressure drop, the Billet and Schultes model has an error of 9% when compared to the experimental database for different kinds of random packings. The error predicted by the PLS model is within that range except for two data points. This discrepancy can be attributed to the smaller size of the model and the error in the Billet and Schultes model itself. The predictions would improve if the size of the data set is increased and by applying the experimental results directly to the system.

Packing Type	**Nominal Size**	**Mass Transfer Coefficient (from the model)**	**Mass Transfer Coefficient (predicted using PLS)**	**% Error**
	inch	(*100)	(*100)	
Super Ring	0.3	0.96	0.94	2.1
Super Ring	2.0	0.79	0.67	15.1
Pall Ring	2.0	0.55	0.54	1.8
Pall Ring	1.5	0.64	0.74	15.6
Pall Ring	1.0	0.84	0.84	0.0
Hiflow Ring	1.0	0.99	0.92	7.1
Super Ring	0.5	0.92	0.99	7.6
Super Ring	1.0	0.77	0.77	0.0
Super Ring	3.0	0.51	0.57	11.8

Table 5.6: Comparison between the actual values based on Billet Schultes model and predicted values by the PLS model for Mass Transfer Coefficient

For the mass transfer coefficient, the Billet and Schultes model has an error of 12% when compared to the experimental database for different kinds of random packings. The error predicted by the PLS model is very close to this range of error. As in the case of pressure drop prediction using the PLS model, this discrepancy can be attributed to the smaller size of the model and the error in the Billet and Schultes model itself. The predictions would improve if the size of the data set is increased and by applying the experimental results directly to the system. It must be kept in mind that the model has been developed from a semi-empirical fit of the mass transfer coefficients and pressure drops.

The relationships in the predictive form for mass transfer coefficient and pressure drop are given in the following equations.

$$m_c = -0.0752\sigma + 0.4377\gamma_1 + 0.0098\gamma_2 + 1.2415a + 0.5239\varepsilon$$

$$\Delta P_{IRR} = 0.036\sigma - 0.0155\gamma_1 + 0.3457\gamma_2 + 0.8305a + 0.1539\varepsilon$$

where m_c is the volumetric mass transfer coefficient, ΔP_{IRR} is the irrigated pressure drop, σ is the standard deviation of the porosity distribution, γ_1 is the skewness, γ_2 is the kurtosis, a is the area and ε is the mean porosity. The independent variables for Cascade Ring were not used in building the PLS model. It was used as a validation point for the model. The pressure drop predicted from the model is 262 Pa/m as against the value of 294 Pa/m obtained from the semi empirical equation of Billet-Schultes model. This translates to an error of 11%. Since the error between the experimental values and the Billet and Schultes model is 9%, the prediction by the PLS model is comparable.

5.3 Statistical properties for new packings

From the size selected from the least squares model developed using Response Surface Methodology, the elements were repacked and the porosity distribution was obtained for the five new shapes. For the new packings the packed bed was sampled with a sample size same as the packing element size. Since the increment in the θ direction was small for improving the resolution, the data were resampled and the normality test was applied on the filtered data. Filtering was essential as the data tends to have a normal distribution in the θ direction and masks the existing non-normal behavior in the *z* and *r* directions. Jarque-Bera normality test was applied on the porosity distribution. The results of the test are given in Table 5.7. As mentioned in Chapter 3, Section 3.4, an *H* value of 1 indicates that the distribution is not normal. Since the shape of the distribution is affected by the type of packing which in turn affects the flow behavior of the gas and liquid, the test was conducted to see if the data deviated significantly from normality. Very high values of skewness and kurtosis may indicate that the liquid and vapor phases might not be distributed uniformly thus affecting performance.

Type of packing	**Size**	**H-Value**	**Mean porosity**
	mm		m^3/m^3
Mobius	25	1.00	0.955
Eight shape	30	1.00	0.929
Rhodonea	30	1.00	0.923
Spiral	30	1.00	0.909
Helicoid	25	1.00	0.907

Table 5.7: The hypothesis testing for normality for new packings suggests that the distribution is not normal

The skewness and the kurtosis along with the variance and other geometrical properties are tabulated in Table 5.8.

Type of packing	Size	Geometrical area	Mean porosity	Standard Deviation	Skewness	Kurtosis
	mm	m^2/m^3	m^3/m^3	(*100)		
Mobius	25	112	0.955	0.19	1.16	7.12
Eight shape	30	179	0.929	0.47	0.27	1.94
Rhodonea	30	194	0.923	0.49	-0.05	0.10
Spiral	30	230	0.909	0.63	0.01	3.22
Helicoid	25	229	0.907	0.47	1.15	2.26

Table 5.8: Higher order cumulants for new packings

For all the new packings the skewness and the kurtosis are in the range developed using the regression equation given in Section 5.2.2.1 of this chapter.

5.4 Prediction of performance parameters for new packings

Using the PLS model in its predictive form, the mass transfer coefficients and pressure drop for the five new packings were predicted. As discussed in the PLS modeling theory, the data used for predicting the PLS model should be of zero mean and unit variance. When predicting the dependent variables from the independent ones, the data needs to be scaled. The mean and standard deviation of the new packings was used for scaling purpose since the mean and variance of the new packings differ significantly from the existing ones. The standardized values for the dependent variables were predicted using these scaled values.

Type of packing	Size	Mass Transfer Coefficient	Irrigated Pressure Drop
	mm		Pa/m
Mobius	25	0.006843	167.1487
Eight shape	30	0.007187	202.3062
Rhodonea	30	0.007184	214.9879
Spiral	30	0.008171	328.8627
Helicoid	25	0.009615	296.038

Table 5.9: Predicted volumetric mass transfer coefficients and pressure drop for new packings

The predicted values for mass transfer coefficient and pressure drop for new packings were rescaled with the mean and the variance used in building the PLS model. The data for the new packings is tabulated in Table 5.9.

The performance parameters for the new packings have been predicted from geometrical and statistical properties. This study provides a technique for selecting the type of packings on which further experimentation can be done.

Chapter 6

Summary and Conclusions

This study proposes a technique to relate the shape of a particle along with other geometric parameters to the performance of a column in terms of mass transfer coefficient and pressure drop. The shape here is defined by the porosity distribution of the bed packed with elements of any given shape. Simulation of existing packings to extract these geometrical properties was done using the packing algorithm. The algorithm perceives the shapes as a set of triangles and packs these elements so that the clearance is as minimal as possible. All the existing shapes and new shapes were presented as a set of triangles to the packing program. This capability, that allows the algorithm to pack an element of any arbitrary shape into a container, has been used to generate geometric characteristics for packings in a cylindrical bed.

Using the packing program, the porosity distribution for each type of packed bed was extracted. Since sample size plays an important part in defining the distribution, it was selected to be same as the size of the particle. However, the increment along each of the co-ordinate direction was the same in all the cases. This ensures that within a given shape, the effect of size is captured. It has been shown in this study that the sample size affects the porosity distribution. Finer resolution almost always gives a smooth wave-like function, which is explained by the fact that smaller volume encounters more void space and more solid metal as it is moved along the co-ordinate directions.

The increment in the azimuthal (θ) direction was selected to be small for improving the resolution. Filtering this set of data is essential because the data tends to have a normal distribution in the θ direction and masks the non-normal behavior in the z and r directions. This is very similar to the 'noise' that one encounters in data sets. The data was filtered so that the noise from the data set is minimized. For this reason the data were resampled to check for any non-normal behavior in the data set.

Jarque-Bera normality test was performed on the distribution for each type of packing and it was found that for all the packings considered in the study, the distribution is non-normal. Higher order moments like skewness and kurtosis were estimated and used to define the distribution. It was found that these statistical parameters vary by a factor of 2 to 3 for packings with similar area and porosity. From these observations it can be concluded with a reasonable degree of certainty that higher order statistical parameters such as skewness and kurtosis can be added to the geometrical area and porosity to define the packing shape.

Five different element shapes were developed in the present work. The material considered for these new packings is carbon steel. For developing the PLS model, all the packings used were steel packings. The interfacial area available for mass transfer changes with the type of material (since the contact angles for the liquid are a strong function of the type of the material). So to have a common denominator in this regard only steel packings were considered. Three different kinds of packing were developed as variations to the cylindrical shape of the Pall ring. The new two-dimensional curves were projected in the *z*-direction to give three-dimensional shapes. Rhodonea, Eight shape and Spiral are among these shapes. Two other shapes, the Mobius Strip and the Helicoid, were selected because the shapes would allow recurrent contact points within the element. The enhanced contact would result in an increased mass transfer coefficient. The effect of the shape on the pressure drop can predicted from the PLS model developed with existing packings.

The prediction of geometric surface area and porosity for each of the element as a function of the size, when in a packed bed, was estimated using Response Surface Methodology. Central Composite Design (CCD) was used to design the experiments for fitting the least squares model. CCD is a second order design which captures the curvature in the set of data. The data was predicted to be non-linear and hence the second order model was used.

Least squares fit was used to arrive at the best possible relation between the dimensional aspects of the packing element and its geometric properties (like surface area and porosity). The probability associated with the Lack of Fit statistic ranges from 0.002 to 0.519. For all the shapes other than the Spiral shape, the probability associated with the lack of fit suggests that the least squares model explains the variance in the data. The size of the packing was selected so that the independent variables such as area and porosity lie within the range of the data set used for developing the PLS model. Thus the size was selected from the least squares fit mentioned above. The selected size of the packing varied from 25 mm to 30mm, and the elements were packed to extract the geometrical and statistical properties.

For estimating the mass transfer coefficient and pressure drop of existing packings (Pall Rings, Hiflow Ring and Super Ring), the Billet and Schultes model was used. The reason for selecting this model is that the error in the estimated quantities is the least among all the available models for random packings. In the Billet and Schultes model, the error is 12-14% for mass transfer coefficient and 9.1% for pressure drop. The thermo-physical and fluid dynamic properties were estimated using the system of absorption of carbon dioxide from air with water at a temperature of 25°C. The mass transfer resistance for this system exists mainly in the liquid phase, and the liquid side mass transfer coefficient is sufficient for determining the overall mass transfer efficiency. The mass transfer coefficients for the existing packing elements ranged from 0.0051 to 0.0096 and the pressure drop ranged from 103 Pa/m to 474 Pa/m.

The Partial Least Squares (PLS) technique was used to develop a regression relation between the performance parameters and porosity distribution and geometric characteristics of the packed bed. The rationale for using the PLS technique can be explained by the fact that the relation between the independent variables is not very clear. Some of these variables are known to be collinear (like geometric area and porosity, generally higher surface area is associated with reduced porosity) and the relationship between the other variables is not clearly known. Also the PLS method gives a better prediction when compared to the ordinary least squares. Three latent variables were used for the model prediction in the latent variable subspace although the cumulative prediction sum of squares (PRESS) is the least for four variables. The increase in the variance explained by fourth variable is very minimal. For the given set of data, the amount of variance captured by the model is around 98% for the pressure drop model. This signifies that the model explains the correlation between the pressure drop and the packed bed parameters very well. For the mass transfer coefficient the variance captured by the model is 84% which is sufficient to say that the model captures the relationship very well.

The values predicted from the model were compared to the actual data for both the dependent variables. For the pressure drop the prediction error ranged from 0.2% to 9.1% except in two cases where the error was 18% for the 1.5" Pall ring and 21% for the 3" Super Ring. For the mass transfer coefficient the error ranged from 0% to 11% except in two cases where it was 15% for the 1.5" Pall ring and the 2" Super Ring.

For the Cascade Ring the pressure drop predicted from the model is 262 Pa/m as against the calculated value of 294 Pa/m from the Billet-Schultes model, which translates to an error of 11%. The Cascade Ring was not used in building the PLS model. It provides a validation point for the PLS model. The error suggests that the model is good enough although more data is needed for confirming the accuracy of the model.

For the new packings, porosity distribution for the packed beds was not confirming to normality. The mass transfer coefficient and pressure drop values for these new packings were estimated from the PLS model developed using existing packings. It was found that although the geometrical area was almost the same as in case of the 30 mm Spiral element and 25 mm Helicoid, the mass transfer coefficient is higher for the Helicoid. This suggests that the model captures the effect of the shape of the particle. In case of the predicted value of the pressure drop, a similar tendency has been observed.

In summary, it can be concluded that the proposed technique appears to be promising and warrants further study. If this technique can be established, it can be used for generating an optimized shape that would help in better overall performance characteristics for mass transfer operations.

Chapter 7

Recommendations

The present study has attempted to relate the shape defining parameters of random packings to the mass transfer and hydraulics in a column. The characteristics of a statistical distribution were used to define the empty spaces between packed beds. Since these spaces are caused by randomly packed elements, it is suggested that the effect of the shape is inherent in the distribution. Higher order moments can be used to define the distribution. The shape of the packing element along with the geometrical surface area and bulk porosity provide a comprehensive way to investigate the dependency of the packing shape on the performance. If this technique can be established with further studies, it would provide means for theoretical studies before actually manufacturing the packing.

For future studies it is suggested that the focus be along the following lines:

- The size of the database for building the model be increased to improve the accuracy of the model. There exist other types of random packings which have not been studied by Billet and Schultes. If these can be included in the future studies, one can have a better model for the prediction of the performance.
- In this study only the mass transfer coefficient and pressure drop have been estimated. Other parameters such as hold up and capacity can be included to

increase the prediction capabilities of the model.

- Most of the current models include packing constants that take into account the shape of the packing element. If this constant can be related to the shape defining parameters a direct relationship can be established. An effort in this direction was made but a proper fit was lacking.
- New packings have been developed as a part of this study. Further experimentation can be performed on these shapes to ascertain the theoretical findings.
- Other shapes can be studied by applying this model. The only drawback seems to be that one-to-one relation between the dependent variables (mass transfer coefficient and pressure drop) and some of the independent variables (higher order moments) is not easily apparent from the model.

The future thrust should mainly be in the direction of improving prediction capability of the model and experimental studies on the new shapes that have been developed as a part of this work. This technique would be very much useful in filtering out shapes and selecting those that have better performance characteristics.

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