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Prediction of Frictional Pressure Drop During Water Permeation Through Packed Beds of Granular Particulates

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Abstract

A methodology has been developed based on the Kozeny-Carman equation to predict frictional pressure drops during water permeation of packed columns containing essentially non-compressible, but highly irregular particles. The resulting model accurately predicts pressure drop as a function of liquid flow rate and resin particle size for this system. A total of five particle sieve cuts across the range -20 to +70 mesh were utilized for testing using deionized water as the mobile phase. The Rosin-Rammler equation was used to fit the raw particle size data (wet sieve analysis) for the as-received resin sample and generate a continuous cumulative distribution function based on weight percent passing through the sieve. Probability distribution functions were calculated from the cumulative distribution for each particle sieve cut tested. Nine particle diameter definitions (i.e., number mean, volume mean, etc.) were then selected from the distribution function for each sample to represent the average spherically-equivalent particle diameter as input to the Kozeny-Carman equation. Nonlinear least squares optimization of the normalized pressure drop residuals were performed by parameter estimation of particle shape factor and bed porosity for all samples simultaneously using a given average particle diameter definition. Good fits to the full experimental data set were obtained when utilizing the number mean and the number median diameters. However, the shape factor and porosity values of 0.88 and 0.40, respectively, obtained from fitting the data using the number mean diameter were more consistent with experimental observations.

Introduction

Particulate ion exchange materials are frequently utilized in packed column configurations for preparative-scale pharmaceutical applications [1] and for large-scale chemical removal processes [2]. High frictional pressure drops across the columns during processing of liquid feed streams can cause pressure limits for safe operation to be exceeded, lead to pump failure, or result in physical degradation of the ion exchange material. For this reason, a number of reports have described the development of models for the prediction of bed pressure drop as a function of liquid flow rate through packed columns [1, 3-4]. The models are typically used for design optimization of column processes utilizing these materials. Such reports often focus on columns containing non-rigid materials where pressure drop is influenced by bed compression and decreased permeability under dynamic flow conditions. We have developed a model for the prediction of bed pressure drop as a function of liquid flow rate and particle size for columns containing an essentially non-compressible, granular ion exchange material that is intended for use in large-scale (approximately 450 gallon bed) ion exchange columns. Although the hydraulic characteristics of packed beds of this material are simplified by low material compressibility, the large scale of the operation and the potential complexity of shape effects for these highly irregular particles upon bed permeability motivated testing and modeling efforts on this system.

Frictional pressure drop across packed beds of particulate materials depends upon several factors, including: fluid viscosity and flow rate, bed dimensions, material compressibility, and bed permeability (which is primarily determined by particle size distribution and shape, but is influenced by other factors as well). For the design process under study in this work, fluid

viscosity and flow rate were somewhat fixed by process throughput requirements. The geometry of the packed column was limited to an L/D near 1.0 to minimize bed compaction resulting from resin shrinking and swelling during acid elution and caustic regeneration cycles. Material compressibility effects were minimal, as preferred, and particle shape could not be easily varied without significant capital investment. The resin mesh size range could, however, be varied with minimal impact to cost and schedule. As a result, tests were conducted with various sieve cuts of the ion exchange material to correlate bed pressure drop and liquid flow rate for each size range.

Determination of the optimal particle size for ion exchange processes utilizing a column configuration involves balancing between ion exchange kinetics (favored by smaller particle sizes) and column pressure drop (minimized by larger particle sizes) effects. Since the ion exchange resin under study is utilized under mass transfer limited conditions where chemical performance is significantly impacted by exchange kinetics, it was necessary to develop predictive models for both chemical and hydraulic performance as a function of the resin mesh size range. A separate report will describe the chemical performance modeling. Herein, we report the development of a bed pressure drop model for this system that can be used to generate pressure drop versus flow rate profiles as a function of resin mesh size range. The theoretical approach involved the combination of the known Kozeny-Carmen relationship for packed beds, as well as the Rosin-Rammler equation for generation of continuous size distribution functions for these highly irregular particles.

The Kozeny-Carmen equation is frequently utilized to predict pressure drops in packed beds during laminar fluid flow [3, 5]. For packed beds of granular materials, the determination of accurate bed porosity, particle size, and particle shape factor for equation input is experimentally difficult. For highly irregular and polydisperse particles, characterization of the distribution by a

single, universal mean particle diameter and standard deviation is not valid, since, even for an individual particle, no single diameter represents the whole. The properties of such materials can, in many cases, be correlated to some statistical parameter of the particle size distribution [6]. Different properties of a given material may correlate to different statistical parameters, although, due to the complexity of the particle size and shape distribution, it often is not understood why certain correlations exist. We have utilized the Rosin-Rammler equation to generate nine statistical representations of the average spherical equivalent diameter for five different particle size distributions of a granular ion exchange resin. The identification of a specific statistic showing strong correlation to the body of experimental pressure drop data generated for all samples using the Kozeny-Carmen relationship resulted in the generation of a predictive tool describing the hydraulic properties of this system.

Experimental

A granular, organic cation exchange material (SuperLig[®] 644 resin) was received from the manufacturer (IBC Advanced Technologies, Inc., American Fork, Utah) as a nominally -20 to +70 mesh (US Standard) sieve cut. The resin was pretreated prior to hydraulic testing by exposure to one acid/base (shrink/swell) cycle in order to remove chemical impurities and resin fines. All tests were conducted on the swollen sodium form of the resin. Five identical resin samples were carefully collected (as water slurries) for wet sieving using a vertical core sampling method based on ASTM-D-2676 [7]. The samples were then wet-sieved with deionized water using an automated sieve unit utilizing both water spray and vibration. 400 mL samples of the following mesh ranges were isolated: -20 to +70, -18 to +30, -20 to +40, -30 to +40, and -40 to

+70. These test samples were never fully dried during sieving and were stored under water after isolation. Duplicate sub-samples of resin were then collected from the as-received sample following the sampling methods described above and sieved with screen mesh sizes 18, 20, 30, 40, 50, 70, and 100 to provide a well resolved particle size distribution for the original material. In contrast to the resin sieve cuts isolated for testing, which were continually maintained in a damp state, these sub-samples were air dried by placing the mesh screens containing resin on the bench top. The material on each tray was subsequently removed, dried under vacuum, and weighed to provide a mass-based particle size distribution for the as-received resin sample.

Custom glass columns were prepared from borosilicate glass tubing for hydraulic testing. The resin containment reservoir had an inside diameter of 2 inches and was jacketed with larger diameter tubing for temperature control. Pressure taps were located at approximately 1 and 3 inches above the resin screen support and were attached to a manometer. During column operation the fluid in the manometer (Miriam Blue) was in direct contact with the water in the tubing connecting the manometer and column. Frictional pressure drop measurements were conducted under dynamic fluid flow conditions by determining the total fluid displacement in the manometer represented by the sum of the fluid height offset for the two sides of the u-tube. Manometer reading offsets, measured in millimeters of oil, were subsequently converted to psid. Each test sample was individually packed into the column by scooping wet resin with a large spatula directly into the water within the column. The height of water through which the resin fell in the column was maintained at a minimum (1-3 inches) during the packing process to avoid particle stratification during column loading. The beds were packed to a total height of approximately 6 inches. The top was placed on the column and the beds were hydraulically conditioned by exposure to repeated water flow cycles at superficial velocities of 0, 10, and 20

cm/min. This process was repeated until it was apparent, based on the measured frictional pressure drop and bed height, that the resin particles had migrated and reoriented within the bed to a stable packing configuration [8].

Bed permeability tests were conducted with each packed column by measuring the frictional pressure drop during water permeation at various flow rates. The column temperature was maintained at 25 ± 1 °C throughout the testing by pumping water through the outer column jacket with a recirculator. Each sample was tested with the following sequence of target superficial velocities: 2, 4, 6, 8, 10, 15, 20, 15, 10, 8, 6, 4, and 2 cm/min. Each pressure drop data point was collected after full stabilization of the system (typically 10-15 minutes). The fluid flow rate through the column at each superficial velocity was confirmed by collecting and weighing duplicate 100-500 mL samples of water effluent over a known time period.

Modeling

The Kozeny-Carman equation was used to compute frictional pressure drops across the packed particulate beds. The functional form of the Kozeny-Carman equation used in this work is shown in Eq. 1.

$$\Delta p = \frac{C_{kc} \mu u_s (1 - \varepsilon_b)^2}{(\phi < d_p >)^2 \varepsilon_b^3} \Delta z \quad (\text{Eq. 1})$$

Δp	frictional pressure drop
C_{kc}	Kozeny-Carman coefficient (180)
μ	dynamic liquid viscosity
u_s	superficial velocity
ε_b	bed porosity
ϕ	particle shape factor

$\langle d_p \rangle$ average equivalent spherical particle diameter
 Δz pressure tap separation

The dynamic liquid viscosity, superficial velocity and pressure tap separation were known quantities. The Kozeny-Carman coefficient was set to the traditional value of 180. The bed porosity, particle shape factor, and average spherically-equivalent particle diameter required estimation. Measurement of bed porosity was not readily amenable to experimental determination. The shape factor was unknown due to the irregular particle shape. Computation of an average spherically-equivalent particle diameter required generation of a cumulative distribution function based on the Rosin-Rammler fit (Table 1) for weight percent passing versus sieve size. The Rosin-Rammler equation is one of the most commonly used theoretical equations for fitting measured cumulative particle size distributions of crushed minerals and blastpiles [9]. Once the cumulative distribution function (cdf) was determined for each test sample, probability distribution functions (pdf) on a weight and number basis were computed. Given the pdf of the particle distribution on a number basis, the average spherically-equivalent diameter was computed based on nine distinct average diameter definitions over the wet sieve interval of interest. Normalized residuals of the pressure drop data and values from the Kozeny-Carman equation were computed. A cost function based on the normalized pressure drop residuals was optimized in a nonlinear least squares fashion by parameter estimation of the bed porosity and particle shape factor for each particle diameter definition.

Nonlinear least squares optimization of the cost function computed from residuals of the wet sieve data and the Rosin-Rammler equation yielded characteristic size (x_c) and uniformity coefficients (m_n) of 632.38 and 3.8529, respectively. The as-received resin wet sieve data and the calculated Rosin-Rammler function are provided in Figure 1. The Powder Technology

Handbook [10] provides nine distinct definitions of mean or average particle diameters. Equations 2, 3 and 4 provide the definition and corresponding integral representations for the number mean, number median, and volume mean diameters (3 of the 9 distinct particle diameter definitions in reference 10) in terms of the number basis pdf. The number mean and number median statistical representations of the average diameter provided the best fit to the full set of experimentally determined pressure drop data. The range of integration covered the particle size or sieve size interval of interest. Spherically-equivalent particle diameters using these definitions were computed for each of the five samples tested. Results of these calculations are provided in Table 2.

$$\text{Number mean diameter, } D_1 = \int n(x)x dx / \int n(x) dx \quad (\text{Eq. 2})$$

$$\text{Number median diameter, } \text{NMD} = \exp\left(\int n(x) \ln x dx / \int n(x) dx\right) \quad (\text{Eq. 3})$$

$$\text{Volume mean diameter, } D_4 = \int n(x)x^4 dx / \int n(x)x^3 dx \quad (\text{Eq. 4})$$

Results and Discussion

Permeability tests were conducted on carefully packed beds of SuperLig[®] 644 resin while varying the flow of water through the beds. Frictional pressure drop was measured as a function of liquid flow rate across a 2-inch high portion of the bed using a manometer. The pressure taps were located at least one inch away from the ends of the bed to minimize end packing and flow distribution effects on the pressure drop data. Linear pressure drop increases with flow rate and consistent resin bed height during testing confirmed low material compressibility and Newtonian

fluid properties under the experimental conditions. Consistent and reproducible data was observed for each particle sieve cut. No hysteresis effects were observed as the superficial velocity was incrementally increased to the maximum of 20 cm/min and then decreased to the minimum of 2 cm/min. Trends in the pressure drop data between the various samples tested were as expected, with lower pressure drop being observed for samples with more coarse size distributions and higher pressure drop being observed for finer distributions.

Experimental pressure drop data and nonlinear least squares fits generated by Kozeny-Carman analysis are provided in Figures 2-4 for three of the nine average diameter definitions evaluated. Calculated bed porosity (ϵ_b) and particle shape (ϕ) factors for each particle diameter definition are also provided in the figures. The number mean and number median particle diameter statistics (D_1 and NMD) provided the best fit to the frictional pressure drop for the five sieve cuts used in this study. Strong data correlation to number-based statistics, indicates that the hydraulic properties of this system are primarily influenced by the fine particles in the distribution, as would be expected. The fit based on the volume mean diameter (D_4), which is weighted toward the coarse particles in the distribution (see Table 2), shows a poor fit to the full pressure drop data set as shown in Figure 4. Similarly poor fits to the experimental data set were observed for the remaining six particle diameter definitions evaluated. As a result, the least squares outputs for these statistics are not provided.

Iterative least squares optimization of the particle shape factor and bed porosity parameters using the D_1 and NMD spherically equivalent diameter statistics confirmed strong correlation to the trends observed in the full pressure drop data set. Low errors were observed in the fits generated using each of these statistics. Calculated bed porosity and particle shape factor values for this system were 0.396 and 0.88, respectively, for D_1 , and 0.377 and 0.997 for NMD. The bed

porosities calculated using the D_1 and NMD statistics were similar and were consistent with the expected value. The optimized particle shape factor of 0.997 calculated using the NMD statistic is more consistent with spherical particles than with granular particles of highly irregular shape. In contrast, the shape factor calculated using the D_1 statistic of 0.880 is consistent with significant deviation from sphericity. As a result, the D_1 statistic is considered to provide the most accurate and consistent correlation to the experimental data. The fact the good fits were obtained for the pressure drop data obtained for all five resin mesh sizes using common shape factor and porosity parameters indicates that consistent particle shape and bed packing characteristics exist across the particle size distribution.

Conclusions

Wet sieve analysis data of a complex particle distribution was shown to exhibit strong correlation to the hydraulic behavior of packed beds during dynamic liquid flow experiments. This was accomplished using established methods for the evaluation of the particle size of granular materials (Rosin-Rammler) and the hydraulic behavior of packed beds (Kozeny-Carmen). This analysis resulted in the generation of a predictive tool for pressure drop across packed beds of this material utilizing the number mean spherically equivalent particle diameter. Given a target liquid superficial velocity, the model could be used to select appropriate particle size ranges to operate within pressure drop limits for column operations. Although testing and modeling for this system were conducted using deionized water as the mobile phase, it is expected that the model could also accurately predict hydraulic performance for higher viscosity feeds. The

general methodology used in this work should also be useful for the prediction of frictional pressure drop for packed columns containing other granular materials.

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Table 1. Rosin-Rammler Cumulative and Probability Distribution Functions.

Function	Basis	Equation
CDF	Weight	$W(x) = 1 - \exp\{-(x/x_c)^{m_{rr}}\}$
PDF	Weight	$w(x) = \frac{dW(x)}{dx} = \left(\frac{m_{rr}}{x_c}\right) \left(\frac{x}{x_c}\right)^{m_{rr}-1} \exp\{-(x/x_c)^{m_{rr}}\}$
PDF	Number	$n(x) = w(x)/f(x)\sigma(x)x^3$

$W(x)$ = fraction by weight finer than a given sieve size or particle diameter

x = sieve size or particle diameter

x_c = Rosin-Rammler characteristic size, taken to be 63.2% passing size

m_{rr} = Rosin-Rammler uniformity coefficient

$f(x)$ = particle volume shape factor ($\pi/6$ for a sphere)

$\sigma(x)$ = particle density (assumed constant)

Table 2. Spherically-Equivalent Particle Diameters for Five Resin Mesh Cuts in Microns.

Mesh Size Range									
-18 to +30		-20 to +40		-30 to +40		-20 to +70		-40 to +70	
diam	value	diam	value	diam	value	diam	value	diam	value
D ₁	692.47	D ₁	558.20	D ₁	503.33	D ₁	419.12	D ₁	313.32
NMD	688.61	NMD	549.98	NMD	500.91	NMD	395.16	NMD	307.30
D ₄	719.44	D ₄	613.51	D ₄	517.94	D ₄	562.56	D ₄	346.72

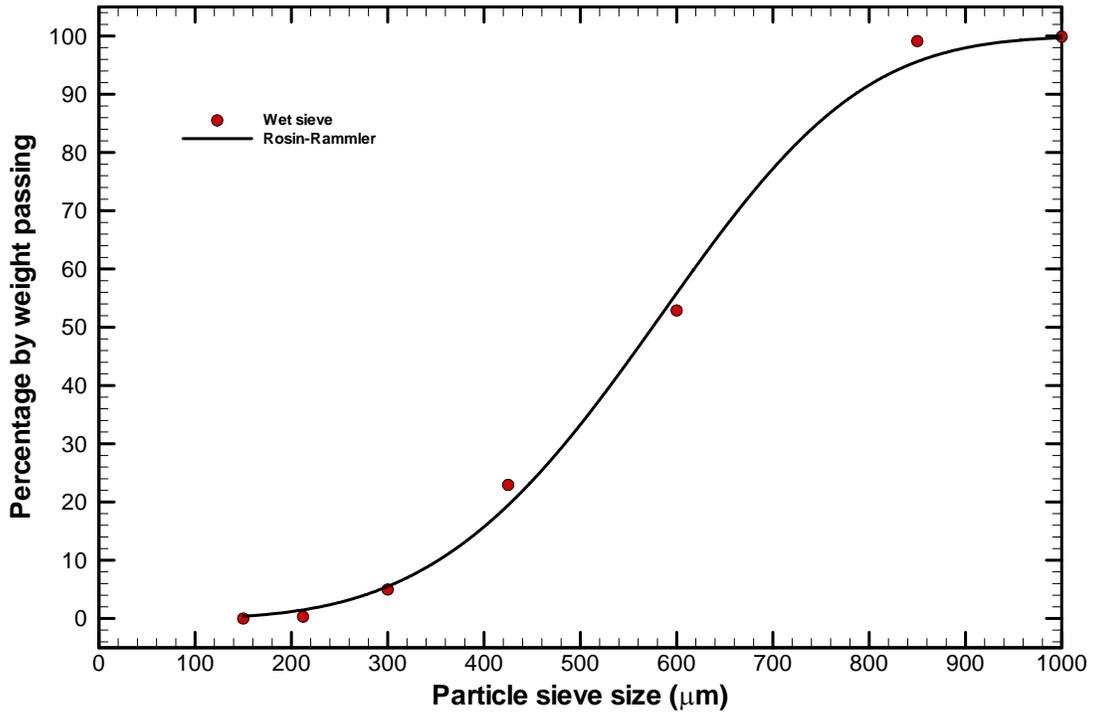


Figure 1. Rosin-Rammler Fit of Wet Sieve Data for the As-Received Resin.

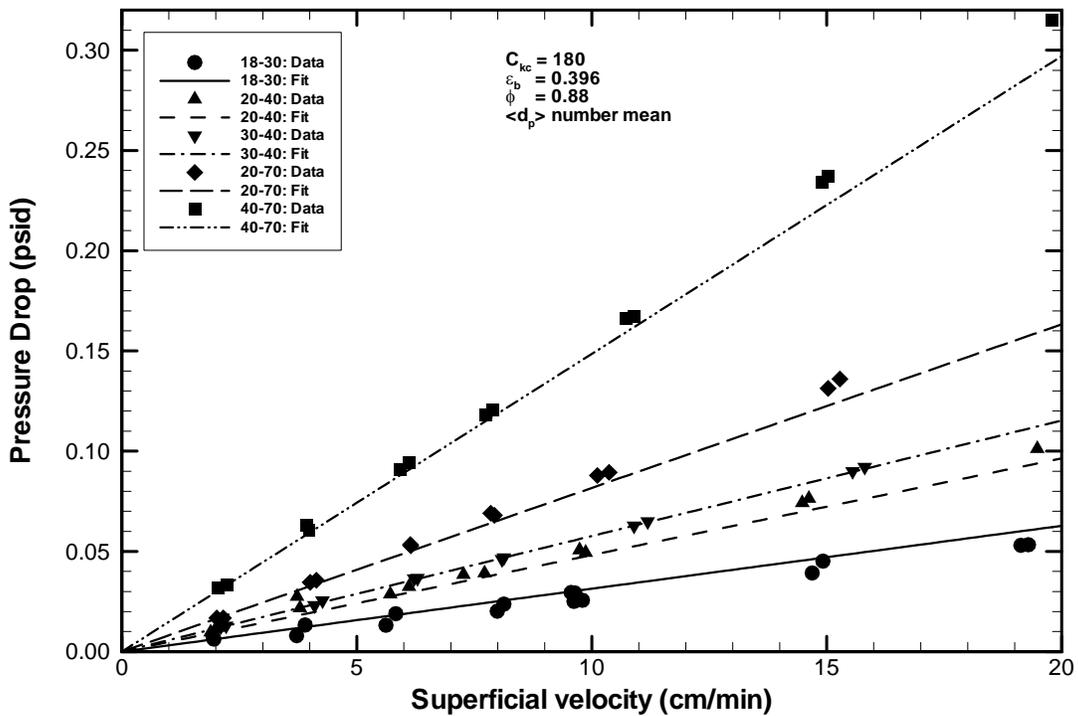


Figure 2. Fit of Frictional Pressure Drop Data using the Number Mean Diameter, D_1 .

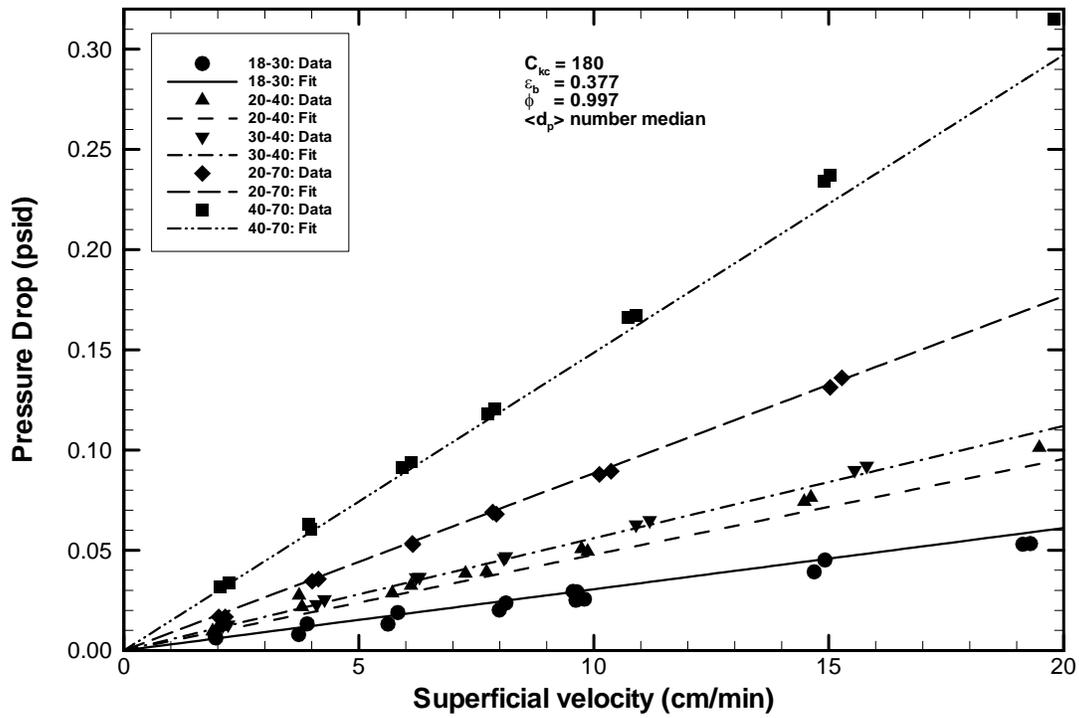


Figure 3. Fit of Frictional Pressure Drop Data using the Number Median Diameter, NMD.

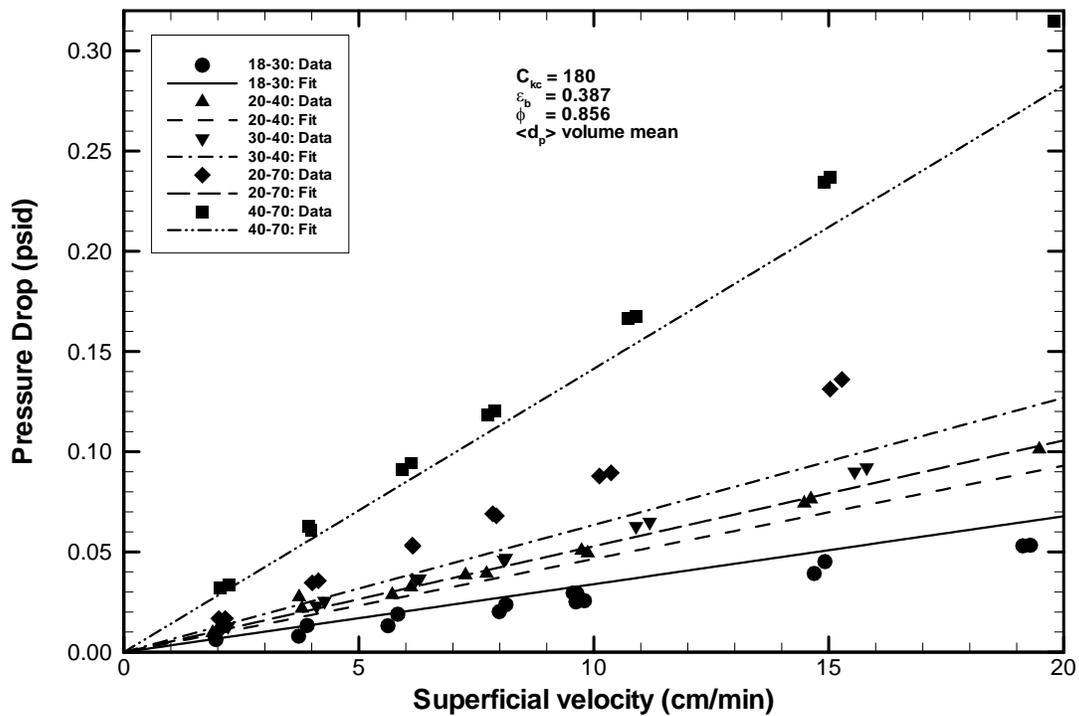


Figure 4. Fit of Frictional Pressure Drop Data using the Volume Mean Diameter, D_4 .