Heat Transfer Parameters of Cylindrical Catalyst Particles with Internal Voids in Fixed Bed Reactor Tubes

A Major Qualifying Project Report

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Abstract

This study measured and analyzed fixed bed heat transfer parameters for different size and shape packing materials that might be used for steam methane reforming. These parameters were compared to those found in the literature for similar packing types with fair agreement. The monoliths and 4-hole cylinders gave better radial heat transfer than the raschig rings; however the monoliths are too brittle for industrial processes.

Executive Summary

Steam methane reforming is a very important process for the industrial world and has many large scale applications. This process requires a very high operating temperature, high process flow rates, and also requires the presence of a catalyst. It is therefore necessary to optimize the process by finding a suitable catalyst shape and size that has a high surface area and a low pressure drop. The catalyst particle must be durable enough to withstand the process conditions, and it must also be able to transfer heat evenly and effectively to ensure uniformity throughout the process.

To more accurately study how different shaped particles transfer heat through fixed bed reactors, we employed a column with a heated air flow through the middle of the bed with a water cooled jacket to regulate temperature. The column was packed with different shaped packing materials and the heat dispersion was measured across the cross section of the column using an array of thermocouples that measured the temperature at several radial positions. The process was conducted using four different packing materials, each at four different bed depths and six different air flow rates. This process was conducted in a column with a diameter of 4 inches, and again in a column with a diameter of 2 inches.

Once the system was at steady state for each run, the temperature readings for each run were collected and analyzed. We used the Inlet Profile Plug Flow model to analyze the data and compared results for the different packing types. When fitted to the data, the IPPF model allowed us to determine which packing had the best heat transfer through the column. Overall, we found that the monoliths and 4-hole cylinders had better heat transfer characteristics than the raschig rings; however, we noted that the monoliths are too brittle to be considered for an industrial process as they are very prone to break and create dust (causing large problems in this type of setting).

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Introduction

In many cases, it is beneficial to convert natural gas into syngas (H_2 and CO) and then into a liquid through use of the Fischer-Tropsch process. The most common method used to convert natural gas into hydrogen is through conventional steam reforming (SR). Other methods similar to SR include Autothermal Reforming (AR) and Gas Heated Reforming (GHR). Commercial hydrogen production is also necessary for the production of ammonia, the production of methanol, as a reducing agent in metal ores, to increase the level of saturation in unsaturated fats and oils (such as margarine) and in hydrogen fuel cell research (New York Energy Research and Development Authority, 2009). The reaction taking place is shown below.

$$CH_4 + H_2O \leftrightarrow CO + 3H_2$$
 $\Delta H = +206 \text{ kJ/mol}$

At high temperatures, above 700 degrees Celsius and with the use of a nickel catalyst, steam reacts with methane to produce hydrogen and carbon monoxide in fixed bed reactors. The process is highly endothermic, therefore this process consumes a lot of energy as heat. Thus, large amounts of energy are required to heat the reactors; over half of the plant's operating cost will be used for the syngas generation section alone. (US DOE, 2009) Production of hydrogen from methane has an efficiency of about 70%, and is the most common and cheapest method to produce hydrogen.

Fixed bed reactor tubes can have very high radial temperature gradients, and it is very important to ensure that there is effective heat transfer from the wall to the packing so that the reactor does not overheat and fail. A steam reformer facility usually has several hundred of these fixed bed reactor tubes and they each usually cost about \$6,000-\$7,000 due to the high process temperature that they are designed for. It is therefore very important to fully understand how the packing shape affects the radial heat transfer to maximize the lifespan of the reactors. Since altering the wall temperature of one of these tubes by 20 K can shorten the lifespan of the tubes from ten years to less than five, this can save the company millions of dollars over time (Dixon, 2009).

A lot of effort is therefore spent researching the best methods to heat the reactor tubes, so that the tubes are heated evenly and efficiently. There are several different approaches to the design of burners, including top, side and terraced wall firing for the steam reformers. Hundreds of burners are used to ensure that the heating is done in a uniform manner through the reformer.

In order to improve the interaction between the steam and the methane within the fixed bed, the reactor is filled with catalyst coated packing (Smirnov, 2003). The shape of the packing determines the amount of interaction between the steam and methane, and also determines the heat flow through the reactor. In order to control the process most efficiently, it is necessary to analyze how the shape and size of the packing alters the flow of heat through the reactor. In addition, the packing must function practically within the tube. The strength and durability of the packing are serious concerns and the packing must also be shaped properly to avoid sharp edges on the packing that lead to particle deterioration and dust that would contaminate the equipment.

Another vital concern in the development of a steam reformer is ensuring that the catalysts are loaded into the reactor tubes in a uniform manner (Stitt, 2005). If any tubes are packed tighter or less densely than another, then the reactant flow to the tubes which are in parallel would tend to have higher flows in some of the tubes than others which lead to hot spots and eventually system failure. To overcome this challenge, technicians perform highly accurate tests to ensure that the pressure drop across each tube for a given flow rate is as uniform as possible to ensure that the reactant flow does not tend towards certain reactor tubes over others.

There is a difficult tradeoff when deciding what kind of catalyst packing to use in a packed bed reactor. Catalysts are more effective with higher surface area, which would indicate that smaller particles with a larger surface area would be beneficial. But it is also true that larger packing particles require a lower pressure drop across the column, which allows for higher flow rates through the column. In addition, the tube to particle diameter ratio also determines the efficiency of the radial heat transfer (Stitt, 2005).

Research in the past regarding heat transfer modeling in packed beds is lengthy. Some of the first significant research was conducted by Coberly and Marshall in 1951, who were the first

engineers to use h_w and k_r as prior work had utilized k_r and $k_r(r)$. In 1972 DeWasch and Froment were the first to question the perceived dependence of k_r and h_w to bed height. In 1975, Gunn and Khalid were the first to introduce the concept of axial thermal dispersion, represented by the term k_a . In 1978, Bauer and Schünder developed what is today the standard model for k_r . Also in 1978, Dixon and Creswell introduced the theory that k_r and h_w ware not height dependant. In 1985, Dixon showed that k_a had little physical basis, he also measured heat leaks through the calming section to eliminate height dependence. In 1988, Dixon reviewed Pe_r and Bi with a significant amount of heat transfer data. In 1993, Borkink, Borman and Westerterp introduced the Inlet Profile Plug Flow model to eliminate height dependence of the packing. In 1993, Martin and Niles found a relationship to describe Nu_w. In 1996, Landon, Hebert and Adams published results for different shaped packing materials. In 1997, Dixon looked at the effect that a low tube-to-particle ratio has on heat transfer for spheres.

There are many different shapes of packing used industrially for steam reforming, but there has been little research done exploring the heat transfer through fixed beds containing cylindrical packing with internal holes and domes. The main purpose of the team's research is to analyze the heat transfer across a fixed bed with different shaped packing. From these findings, optimal designs for catalyst particles can be suggested, which is a major step in the effort to develop more efficient steam reforming technology.

Background

Heat transfer is categorized into three types: conduction, convection, and radiation. For these experiments, radiation does not apply as the temperatures were relatively low. Through research done over the years, models have been created using Fourier's law where conduction and convection parameters are strictly based on packing size and shape as well as fluid flow. The effective radial thermal conductivity, k_r , represents all mechanisms of heat transfer in the center of the bed (unaffected by wall effects). The wall heat transfer coefficient, h_w , is a parameter representing the increased heat transfer resistance close to the wall. Both of these parameters can be found from data using models such as the Plug Flow (PF) model or the Inlet Profile Plug Flow (IPPF) model.

In order to model the heat transfer using the equipment and model used, the following assumptions were made:

- The system was in steady state
- No reaction took place
- There is no radiation
- The pressure throughout the bed was constant
- No radial variation of the superficial gas velocity (Reynolds number)
- The wall temperature was constant and steady for each run
- The physical properties of the gas and solid were independent of temperature

IPPF Model

The standard model used to analyze the temperature profile data acquired was the Inlet Profile Plug Flow (IPPF) model. The PF model uses the temperature of the inlet air as the inlet temperature; however, with the apparatus in the laboratory, it was impossible to determine the temperature of the air flowing into the packing at z = 0 due to heat loss in the calming section. The IPPF model uses the first height temperature profile as the inlet temperature profile, and compares it to the temperature profiles at consequent heights.

The IPPF model is derived from the following dimensional energy balance:

$$GC_p \frac{\partial T}{\partial z} = k_r \left(\frac{\partial^2 T}{\partial r^2} + \frac{1}{r} \frac{\partial T}{\partial r} \right)$$

With the following boundary conditions:

$$T = T_{z,o}(r) \text{ at } z = z_o$$
$$\frac{\partial T}{\partial r} = 0 \text{ at } r = 0$$
$$-k_r \frac{\partial T}{\partial r} = h_w (T - T_w) \text{ at } r = R$$

These equations are usually made dimensionless by defining the following:

$$\vartheta = \frac{(T - T_w)}{(T_o - T_w)}$$
$$y = \frac{r}{R}$$
$$x = z/R$$
$$Pe_R = \frac{GC_pR}{k_r}$$
$$Bi = \frac{h_wR}{k_r}$$

In this case, using the first measurement height at $z=z_{o}$ as the inlet temperature, we get:

$$\zeta = \frac{\frac{z}{L} - \frac{z_o}{L}}{1 - \frac{z_o}{L}}$$
$$\frac{\partial \zeta}{\partial z} = \frac{1}{L} \frac{1}{1 - \frac{z_o}{L}}$$

So:

$$\frac{\partial T}{\partial z} = \frac{\partial T}{\partial \zeta} \frac{\partial \zeta}{\partial z} = \frac{1}{L} \frac{1}{1 - \frac{z_o}{L}} \frac{\partial T}{\partial \zeta}$$

Then:

$$GC_p \frac{1}{L} \frac{1}{1 - \frac{z_o}{L}} \frac{\partial \vartheta}{\partial \zeta} = k_r \frac{1}{R^2} \left(\frac{\partial^2 \vartheta}{\partial y^2} + \frac{1}{y} \frac{\partial \vartheta}{\partial y} \right)$$

i.e.

$$\frac{\partial \vartheta}{\partial \zeta} = \frac{k_r L}{G C_p R^2} \left(1 - \frac{z_o}{L} \right) \left(\frac{\partial^2 \vartheta}{\partial y^2} + \frac{1}{y} \frac{\partial \vartheta}{\partial y} \right)$$

Define:

$$Bo_{h,r}^{\theta} = \frac{GC_p R^2}{k_r L}$$
$$\theta = \frac{\vartheta}{\vartheta_o}$$

Where:

$$\vartheta_o = \frac{(T|_{r=o} - T_w)}{(T_o - T_w)}$$

Gives:

$$\frac{\partial \theta}{\partial \zeta} = \frac{1}{Bo_{h,r}^{\theta}} \left(1 - \frac{z_o}{L} \right) \left(\frac{\partial^2 \theta}{\partial y^2} + \frac{1}{y} \frac{\partial \theta}{\partial y} \right)$$

With boundary condition:

$$-k_r \frac{\partial T}{\partial r} = h_w (T - T_w)$$

This can be made dimensionless into:

$$-k_r \frac{1}{R} \frac{\partial \vartheta}{\partial y} = h_w \vartheta$$

This may be reduced to the following:

$$\frac{\partial\vartheta}{\partial y} + Bi\vartheta = 0$$

And then:

$$\frac{\partial\theta}{\partial y} + Bi\theta = 0$$

Assume a parabolic profile at $z = z_o$:

$$T = T|_{r=0} + A'r^2$$

Where A'<0 for cooling.

$$\frac{(T-T_w)}{(T_o-T_w)} = \frac{(T|_{r=o} - T_w)}{(T_o - T_w)} + \frac{A'}{T_o - T_w} R^2 y^2$$
$$\vartheta = \vartheta_o + \frac{A'R^2}{T_o - T_w} y^2$$

So:

$$\theta = \frac{\vartheta}{\vartheta_o} = 1 + \frac{A'R^2}{(T_o - T_w)\vartheta_o}y^2 = 1 - Ay^2$$

Where $A = \frac{A'R^2}{(T_o - T_w)\vartheta_o} > 0$ always.

The IPPF model is incorrect as given by Borkink (1993). It may be made correct by defining:

$$Bo_{h,r}^{\theta}(corrected) = \frac{GC_{p}R^{2}}{k_{r}L} \frac{1}{1 - \frac{z_{o}}{L}} = Bo_{h,r}^{\theta} \frac{1}{1 - \frac{z_{o}}{L}}$$

Convert into present variables using corrected $Bo_{h,r}^{\theta}$:

$$Bo_{h,r}^{\theta} = Pe_R \frac{R^2}{Ld_p} \frac{1}{1 - \frac{z_o}{L}} = Pe_R \frac{R}{L} \frac{1}{1 - x_o R/L}$$

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$$\zeta = \frac{x \frac{R}{L} - x_o \frac{R}{L}}{1 - x_o \frac{R}{L}}$$
$$\frac{\partial \zeta}{\partial x} = \frac{R}{L} \frac{1}{1 - x_o \frac{R}{L}}$$
$$\vartheta = \frac{(T - T_w)}{(T_o - T_w)} / \frac{(T|_{r=o} - T_w)}{(T_o - T_w)}$$
$$= \left[\frac{(T_w - T_o)}{(T_w - T_o)} - \frac{(T - T_o)}{(T_w - T_o)}\right] / \left[\frac{(T_w - T_o)}{(T_w - T_o)} - \frac{(T|_{y=o} - T_o)}{(T_w - T_o)}\right]$$
$$= (1 - \vartheta) / (1 - \vartheta_o)$$

Equations become:

$$\frac{\partial \vartheta}{\partial x} = \frac{1}{Pe_R} \left(\frac{\partial^2 \vartheta}{\partial y^2} + \frac{1}{y} \frac{\partial \vartheta}{\partial y} \right)$$

With:

$$\vartheta = 1 - (1 - \vartheta_o)(1 - Ay^2) at x = x_o$$

 $\vartheta finite at y = 0.$
 $\frac{\partial \vartheta}{\partial y} + Bi(\vartheta - 1) = 0 at y = 1$

This reduces to the conventional PF model if $x_o = 0$, A = 0, and ϑ_o . The solution is:

$$\vartheta = 1 - 2(1 - \vartheta_o) \sum_{i=1}^{\infty} \{ \frac{\left[(Bi\lambda_i^2 + 4ABi - 2A\lambda_i^2 - ABi\lambda_i^2) J_o(\lambda_i y) \right]}{\lambda_i^2 [\lambda_i J_o(\lambda_i) + BiJ_i(\lambda_i)]} e^{\left[-\frac{\lambda_i^2}{Pe_R}(x - x_o) \right]} \}$$

$$=1-2(1-\vartheta_o)\sum_{i=1}^{\infty}\{\frac{(Bi+4ABi/\lambda_i^2-2A-ABi)}{[Bi^2+\lambda_i^2]}\frac{J_o(\lambda_i y)}{J_o(\lambda_i)}e^{\left[-\frac{\lambda_i^2}{Pe_R}(x-x_o)\right]}\}$$

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Dimensionless Parameters

The Reynolds number (Re) is a dimensionless fluid mechanics parameter that is directly proportional to the superficial velocity of the fluid. The equation for the Reynolds number of the air flowing through a fixed bed is:

$$Re = \frac{v_s \rho d_p}{\mu}$$

Where v_s is the superficial velocity of the air in ft/min, ρ is the density of the air, d_p is the diameter of the particle in inches, and μ is the dynamic viscosity of the air

The Peclet number (Pe_r) is given by the following equation:

$$Pe_r = \frac{Re * Pr}{k_r / k_f}$$

Assuming kr/kf is linearly related to Re, and Pr is assumed to be constant at 0.71 for air at 60 °C:

$$Pe_r = \frac{Re * Pr}{m * Re + b}$$

Where m and b are the slope and intercept respectively of the line fitting our k_r/k_f versus Re data. The Peclet number should increase rapidly for a low range of Reynolds numbers and stay relatively constant with respect to Reynolds number after reaching a certain velocity. This is because when Re is low, the intercept (b) has more of an effect on Pe_r .

The Biot Number (Bi) is a relationship between the wall heat transfer coefficient and the thermal conductivity of the packing. It is given by the following equation:

$$Bi = \frac{h_w R}{k_r} = N u_w \frac{R/d_p}{k_r/k_f}$$

Since k_r is assumed to be linearly proportional to the Reynolds number, and h_w increases at a slower rate with Reynolds number, we would assume the Biot number to decrease with respect to the Reynolds number. If Nu_w was linearly proportional to Re, the Biot number would decrease linearly, but theoretically Nu_w should fit the equation:

$$Nu_w = Nu_w^o + ARe^B Pr^C$$

Where A, B, and C are experimentally determined constants. With only six data points, it is impossible to create an accurate fit line with this model for Nu_w , so we just assume that the Biot number will decrease with respect to Reynolds number.

The dimensionless parameter N is a ratio of tube diameter to particle diameter.

$$N = \frac{2 * R}{d_p}$$

Several studies completed have given various parameters that must be met in order to receive accurate data when using models such as the IPPF. Many of these studies suggest that the first bed depth used should be at least 1.5 times the diameter of the column (Borkink & Westerterp, 1993). This being said, the optimum first height for the 4 inch column would be 6 inches. With the limited number of particles available, the first height chosen was 4 inches, which could have had an effect on the accuracy of our data.

Methodological Procedure

Equipment

The apparatus used in a cooling experiment with a fixed bed reactor tube is shown in Figure 1.





Columns

Each column used was constructed using two concentric brass tubes to form an annular space between the two (water jacket). The columns were attached to calming sections using four screws. Screws used in the 4 inch column were constructed from nylon, whereas the screws used in the 2 inch column were constructed from steel. The calming sections were constructed out of nylon and filled with ¹/₄ inch steel spheres. The steel spheres were separated from the test packing by a circular piece of steel mesh. The calming section allowed for the airflow to be evenly

dispersed over the cross-section of the column. The columns had internal diameters measuring 2 inches (50.8mm) and 4 inches (101.6mm).

Determining Reynolds number of inlet air flow

To measure the flow rate of the air entering the column, it was passed through a ¹/₂-27-G-10 rotameter tube with a ¹/₂-GSVT-48A float. Depending on the pressure of the air, different amounts of air would flow through the rotameter at 100% of the total flow. These values are listed in Table 1.

Gauge pressure	MAX	Gauge Pressure	MAX
0	44.9	26	2.94
1	4.73	27	2.91
2	4.6	28	2.88
3	4.46	29	2.84
4	4.34	30	2.8
5	4.23	31	2.78
6	4.13	32	2.75
7	4.03	33	2.72
8	3.94	34	2.69
9	3.86	35	2.67
10	3.78	36	2.64
11	3.7	37	2.61
12	3.64	38	2.59
13	3.57	39	2.56
14	3.51	40	2.54
15	3.45	41	2.52
16	3.39	42	2.49
17	3.34	43	2.47
18	3.28	44	2.45
19	3.23	45	2.43
20	3.19	46	2.41
21	3.15	47	2.39
22	3.1	48	2.37
23	3.06	49	2.35
24	3.02	50	2.34
25	2.98	51	2.32

Table 1: Flow data for a 1/2-27-G-10 rotameter tube with a 1/2-GSVT-48A float

The volumetric flowrate of air flowing through the column (SCFM) was found using the following equation:

$$SCFM = \% * Max * (\frac{14.7 + psig}{14.7})$$

Where % is the percentage read on the rotameter tube for air, Max is the maximum flow rate (at 100% on the rotameter, which can be found in Table 1) for the rotameter tube and float used, and psig is the pressure read off of the pressure gauge downstream from the rotameter. The percentage of airflow is read from the "knife-blade edge" of the float (which is at the widest diameter of solid on the float).

Since the column area, and the density and viscosity of air were assumed constant for air flowing though the column, the Reynolds number was then determined using the following equation derived for each column:

$$Re = C * SCFM * d_p$$

Where C is 380 for the 2 inch column and 95 for the 4 inch column.

Packings

Four different packing shapes were examined throughout the course of the project:

- $\frac{1}{2}$ inch ceramic spheres
- Raschig rings
- Johnson Matthey K-57 4-hole pellets (4-hole cylinders)
- Monoliths

The $\frac{1}{2}$ inch ceramic spheres measured 12.7mm in diameter and were porous in nature. There have been many studies completed using spheres with N=8 (in our case, the spheres were used in the 4 inch column only). By comparing our data to these studies, we could verify that our data is consistent with past experiments.

Raschig rings in this study are brittle non-porous ceramic cylinders with a single hole in the middle. The rings we used had heights and outer diameters equal to 13mm, with a wall thickness of 3mm. There have also been some studies with raschig rings with various values of N, which

allows us to do a comparison to verify our data is relatively consistent and will give values for the N we used.

The Johnson Matthey K-57 4-hole pellets were porous ceramic 4-hole cylinders. These were 17mm tall, with an outer diameter of 14mm and hole diameters of 4mm. Packings such as these are typical support for steam reforming.

The monoliths are ceramic cylinders, measuring 7mm tall with a diameter of 6mm and a hole density of 0.563 holes/mm² (56.3 holes/cm²). The monoliths used in this study are illustrated in Figure 2.



Figure 2: Monoliths (http://www.made-in-china.com/showroom/hisina88/product-detailKoXmSOuGZzVf/China-Automobile-Exhaust-Catalyst-Carrier.html)

The Inlet Profile Plug Flow model requires a particle diameter to complete calculations. This diameter is referred to as the equivalent spherical diameter. In other words, this is the diameter required in a sphere to obtain the same volume as the particle (without accounting for any internal voids). The three non-spherical packings were all cylindrical in shape, so the equivalent spherical diameter could be found using the following equation:

$$\pi \frac{d_c^2}{4}h = \frac{4}{3}\pi \frac{d_p^3}{8}$$

Where d_c is the diameter of the cylinder and d_p is the equivalent spherical diameter.

Table 2 lists the equivalent spherical diameters of the packings, as well as the ratio of each column to said diameters.

Table 2: Packing to column diameter ratios

Packing	Equivalent Spherical Diameter	N in the 2 inch column	N in the 4 inch column
Spheres	12.7		8.0
Raschig Rings	14.5	3.5	7.0
JM 4-hole cylinders	17.4	2.9	5.8
Monoliths	7.3	7.0	14.0

Micromega Heater

The heater used to heat the inlet air flow was controlled by an Omega Micromega CN77000 Series Controller. Complete instructions to use the controller may be found on Omega's website at http://www.omega.com/Manuals/manualpdf/M2491.pdf. The purpose of the heater in this experiment is to heat the inlet air to approximately 100 °C. There are two settings that may be altered to acquire this: the desired temperature and the rate at which it takes to get to the desired temperature (ramp and soak). Our desired temperature was set to 100 °C. The rate will affect how long it will take the system to reach equilibrium. Since it is impossible to get the temperature of the air to be exactly 100 °C at all times, the heater heats until the air is 100 °C and hinders heating until the flow is cooled below 100 °C, in which case it will start heating again. This creates an oscillation in temperature over time, and after infinite time the temperature will approach 100 °C. With a faster heating rate, the air flow will reach 100 °C sooner; however, larger oscillations will occur. Conversely, with a slower heating rate, the air flow will reach 100 °C slower, but smaller oscillations will occur. The ramp and soak configuration was disabled, which allows the heater to reach the desired setting as fast as possible.

Thermocouples

A diagram of the thermocouple cross can be seen in Figure 3.



Figure 3: Thermocouple Cross

The thermocouple cross consisted of two sets of four arms set 45 degrees apart from each other. Each arm had three thermocouples attached, measured at different radii. The radii for each thermocouple in millimeters are shown in Table 3.

Thermocouple	ermocouple 2 inch column		4 inch column	4 inch column
	(mm)	(dimensionless)	(mm)	(dimensionless)
А	0	0.00	0	0.00
В	8.5	0.33	9.5	0.19
С	12	0.47	19	0.37
D	15	0.59	28	0.55
Е	18	0.71	41	0.81
F	21.5	0.85	44	0.87
G	24	0.94	48	0.94

 Table 3: Radial position of each thermocouple represented in Figure 2 in both columns

Not counting the center thermocouple, data is collected by 24 thermocouples at six different radial positions. By rotating the thermocouple cross 45° and taking a second set or readings, 48 thermocouple readings are recorded (i.e. eight angular replicates of six radial positions). This allows for a more accurate measurement of the average temperature of each radial position across the cross-section of the column. The thermocouples were held in place by thin plastic tubing (like a small straw) connected to two nylon bars (each arm consisted of two bars) with holes drilled at the same radial position. A side view of the thermocouple cross (one of the eight arms) is shown in Figure 4.



Figure 4: Side view of one arm in the thermocouple cross

Consecutive thermocouples used in these experiments are placed at radii very similar to the ones before and after (within 3mm). These thermocouples must be verified before inserting the thermocouple cross into the column each time to ensure that they are not bent, as reading temperatures from 1 or 2 mm different than expected will yield faulty results.

Three thermocouples were placed in the cooling water jacket. These were placed 7.5, 24.5, and 37.5 mm from the bottom of the column. All three were averaged for each set of data in order to determine the wall temperature.

Temperature Profile Collection

The data acquisition device used was a Keithley Integra Series 2700 Datalogger. This device combines functions of DMM, switch system, and datalogger, and allows for the collection of

thermocouple data for up to 200 channels (35 of these channels were utilized in this experiment). The device worked in conjunction with ExceLINX (an add-in designed for Microsoft Excel) to collect data over time.

To install ExceLINX, the program file ExceLINX.xla must be placed in the Excel Add-ins folder. This can be found in explorer: C \rightarrow Documents and Settings \rightarrow All users \rightarrow Application Data \rightarrow Microsoft \rightarrow Add Ins. Then in Excel, go to Tools \rightarrow Add In. Click browse and select ExceLINX.xla from the menu. Now the ExceLINX tab should be available from the menu at the top of Excel.

The Keithley instrument must now be configured to use with the ExceLINX program. Start by turning on the Keithley instrument. Then, at the top of the Excel screen, select ExceLinx \rightarrow DMM Config and complete as shown in Figure 5.

1	A B C	D E	F G H	IJK	LMN		R S T U	V W X
1	Exce	VX Č			KEITH	ILEY A Grea	ater Measure of Conf	idence
4	Taski Configuro Soonning DMM Channels							
5	Tashi u	Name	DMM Config	13				
6		Description						
7		Created By	ashman					
8	Task	Company	Worcester Polytee	chnic Institute				
9		Date Created	6/1/2009					
10		Date Modified	10/12/2009					
11		Status/Cmds	Running					
12		Device	KE2700_COM1		2000			
13		Password						
14		Slot 1 Module	M7700					
15	Instrument	Slot 2 Module	M7700					
16	msuument	Slot 3 Module						
17		Slot 4 Module						
18		Slot 5 Module						
19		Front Panel Lockout	On					
20		Line Sync	Off					
21		Autozero	On					
22	Setun	Display Digits	61/2					
23	Jetup	DCV Input Divider	Off					
24		Open TC Detection	On					
25		Temp Scale	°C					
26		Digital Outputs	Off					
27	Distance in the	Pulse Output	Off					
28	Limits	Polarity	High					
29		Duration		60 sec				
30		Master Latch	Off		_			
31								
32	Observal		Castina	Channel	Scan List	D C#	contra 1	Ontines
35		measurement	Scaling	A NUMERA	Alarm Limits	Kep Filt	er sampling	
34	END LIST Tag	Function Range	Rel Math myret	D U ENT I			unt Rate AC BW Opt	Opt 2 Opt 3 Opt 4
20	UN 101-120,201-214	TEMP K	UTT None	UT	Uff	υm	SLOW INT	
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42								
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+5	N DUBL Com	MM Config De 157	Da 1570 h	De 1200	De 1000 h	000 / 0- 000	Do 676 / Do 67	
Fig	ure 5: DMM Scan D	g Screenshot	Re 15/0 b	ке 1209	ке 1209 b 📝 Re	e 909 📈 Re 909 t	0 / Ke 6/6 / Ke 6/ M	

Then click ExceLINX \rightarrow DMM Scan and complete as shown in Figure 6.

1	A	В	C	D		E	F	G	Н	1	J
1	Exce					KE	ITHU	EY A Grea	ter Measu	ire of Co	nfidence
2					-						
4		Task: Scan L	MM Channels		_						
5		Description	e DMM Scan								
7		Created B	ashman								
8	Task	Company	Worcester Polyt	echnic Institute							
9		Date Create	6/1/2009								
10		Date Modified	10/12/2009								
11		Status/Cmd	Task stopped s	uccessfully							
12	Configuration	Workshee	t DMM Config	274							
13		Mode	Scan								
14		Source	Immediate								
15		Delay	Auto	sec							
16	Trigger	Reading Coun	t INF								
17		Time	r	0.1 sec							
18		Monito	None								
19		Monitor Limit	None								
20		VVorkshee	t DMM Scan								
21		Starting Co									
22	Data	Organiza B	Powe								
20	Location	Autoincremen	t lise one table								
25	Location	Auto Wra	On								
26		Log File									
27		Forma	Delimited text (c	comma)							
28		Add Channel Tag	Yes	111-111							
29		Add Channels	Yes								
30		Add Units	No								
31	Data Display	Scroll Display	No								
32		Limit	None								
33		Timestam	None								
34		Update Interva	ſ	100 msec							
35					_						
36		Task	Data								
37					6						
38											
39											
40											
41											
42											
43											
44											
45		can DMM Coofie	Re 1570	Re 1570 h	a 1200	Re 11	no h	Re 000 / P	a 000 h	Re 676	Redit
Figu	re 6: DMM S	can Screenshot	NE 1.1/11			NE 17	17.1	NC 307 0		AC U/U	

To record data, select the dropdown menu "Status/Cmds", select "Start", and then press enter. Data will be recorded in the selected sheet under "Worksheet". To stop collecting, choose "Stop" from the "Status/Cmds" drop down menu and press enter very shortly after (almost instantaneously). Each column of data represents temperatures read at specific thermocouples

depending on their placement. In order to determine which thermocouple corresponded to each column, each thermocouple was tested by touching one by one with a finger-tip and checking ExceLINX to determine which column's temperature changed. The ambient temperature in the lab was usually close to 20 °C and after being touched by a finger-tip, the temperature reading for that thermocouple would read about 30 °C. There were five thermocouples removed from the column, but still connected to the Keithley instrument. Temperatures for these thermocouple connections read 9E31 on ExceLINX. After the channels on ExceLINX for these five thermocouples and the 25 thermocouples from the thermocouple cross were determined, the channels for the wall thermocouples and the inlet airflow needed to be determined. These were found by removing the connecting wires one by one and seeing which channel read 9E31.

Procedure

In order to ensure a good fit for our data, six different Reynolds numbers were measured during each day of column operation, and four bed depths were taken for the packings in each column. A table of all runs completed during the course of the project is shown in Table 4.

Table 4: Table of runs

Column Diameter	lumn Diameter Packing		Bed Depths (mm)	
(mm)				
101.6	¹ / ₂ " spheres	75, 145, 195, 260,	92, 150, 200, 225	
		335, 425		
101.6	Raschig Rings	85, 142, 190, 258,	100, 150, 200, 250	
		341, 432		
101.6	JM 4-hole cylinders	102, 198, 268, 358,	100, 150, 200, 250	
		460, 584		
101.6	Monoliths	43, 71, 95, 129, 171,	100, 160, 200, 260	
		216		
50.8	Raschig Rings	350, 490, 676, 909,	80, 150, 210, 255	
		1209, 1570		
50.8	JM 4-hole cylinders	409, 775, 1052,	80, 150, 200, 265	
		1412, 1822, 2275		
50.8	Monoliths	170, 323, 439, 589,	95, 155, 210, 260	
		760, 948		

It has been determined that the minimum packing depth should be 1.5 times the column diameter to let the parabolic temperature profile develop (Borkink & Westerterp, 1993). This must be

done so that the IPPF model would fit the data (requires parabolic temperature profiles). Although the minimum packing depth was determined to be 1.5 times the column diameter, this could not be done with some of the packings in the 4 inch column due to a lack of packing quantity. To ensure enough deviation in bed depths, and keeping consistency throughout the runs, runs in the 4 inch column started at a height close to 4 inches (only 1 times the column diameter).

Operating the Column

- 1. If running the tower for the first time, carefully unscrew and remove the top section of the tower.
- 2. Fill the calming section of the tower with metal spheres and cover the calming section with a wire mesh to keep it separate from the test packing.
- 3. Re-attach the top section of the tower.
- 4. Measure the inside height of the empty tower and record the value.
- 5. Add the desired amount of the packing you are working with to the tower. To do this properly you should:
 - a. Add the packing little by little and gently compact it from the top after each addition to assure for uniformity of packing.
 - Regularly measure the height of the empty space left in the column (space from packing to top) to ensure that you do not surpass your desired packing height.
 (Packing height=height of empty tower empty space left)
- 6. Set up the thermocouple cross so that once it is inside the tower the thermocouples are just above the packing (no more than 3-6 mm). To do this measure the distance from the cross bar that will support the cross to the bottom of the thermocouples and set it so that this distance is slightly less that the empty space remaining in the tower. Before lowering the thermocouple cross into the column each time, straighten all thermocouple tips and ensure that they are centered with the guide tubes.

- 7. Carefully lower the thermocouples cross into the tower. If you feel it hit the top of the packing remove it from the tower, re-adjust the height of the cross bar, and straighten all thermocouple tips.
- 8. Open ExceLINX. If you have not yet set up ExceLINX as described in the ExceLINX section take the time to do so now.
- 9. Turn on and configure the Keithley 2700 Multimeter/Data Acquisition System.
- 10. Prior to starting any flows make sure that all the valves are in the fully shut position. For Water Valves (WV) 1&2 as well as Air Valves (AV) 1&3 this means that the valve is perpendicular to the tube on which it is located. For AV 2 this means that the valve is turned fully in the CLOCKWISE direction.
- 11. Slowly fully open WV 1.
- 12. Slowly open WV 2 until the rotameter reads a flow of 80%.
- 13. Slowly fully open AV 1. At this point if the pressure gauge following AV 1 is showing a non-zero pressure reading it means that AV 2 is not fully shut, in which case take the time to fully shut it now.
- 14. Slowly fully open AV 3.
- 15. If you are running the column for the first time adjust AV 4 so that at the desired setting it allows for a full range of desired Re values to be obtained using only AV 2 to adjust the flow. The following things should be noted:
 - a. Once this has been set AV 4 should not be touched in further runs to ensure for uniformity of values.
 - b. Never allow the rotameter to reach or exceed 100%.
 - c. The pressure gauge that should be used to calculate the Re values is the one that is right before entering the heater (i.e. downstream from the rotameter).

- 16. Turn on the heater. If you have not yet set up the Omega Micromega heating control unit take the time to do so now.
- 17. Set the flow to the desired amount by adjusting only AV 2.
- 18. Using ExceLINX start recording the data.
- 19. Once the system has reached steady state stop the data recording. This should take anywhere from 1 to 3+ hours depending on the flow rate, packing height, and if it's the first run of the day.
- 20. Carefully rotate the thermocouple cross 45 degrees. The top of the column has markings for 0 and 45° in which the cross is to be lined up with to accurately measure the 45° rotation.
- 21. Commence recording data in ExceLINX. Make sure to do this in a different Excel sheet then the one used for the previous data.
- 22. Once the system has once more reached steady state stop the data recording. This should only take about 5min after the angular rotation.
- 23. Repeat Steps 17-22 for all desired Re values.
- 24. Shut the system down in the reverse order that you started it in.

Safety Precautions

The spheres, raschig rings, and monoliths were all inert ceramic packings. However, the 4-hole cylinders contained a white powdered metal on the surface of each particle. Labels on the container provided by Johnson and Matthey warn that exposure may cause irritation. Although it has not been classified as hazardous, it is good practice to minimize exposure to this metal. In order to do so, gloves were worn at all times when handling the 4-hole cylinders. Also, in order to prevent inhalation, the first set of runs performed with this packing was done using the largest bed depth. Once packed for the first time, painter's masks and safety glasses were worn and then the air was set to the highest flowrate achievable. Visibly, no dust came from the column at the highest flowrate, so the air was left at a high flowrate overnight so that any particles that did get

blown up were not breathed in by anyone. No dust was visible anywhere in the lab the next day; a miniscule amount was actually loose enough to be blown off the particles.

The heater would reach temperatures of 115° C, and even though was insulated on all sides, could cause minor burns if touched. These surfaces were not touched for any reason, and if a particle or other object were to fall on top of the heater, it was immediately removed with pliers.

Data file format for model fitting

The IPPF model was fit to the temperature profile readings using an existing program written in FORTRAN. In order to run the program, it was required to create a text file in the correct format with data collected. A template of this format is shown in Table 5. In order to smooth the steady state thermocouple temperatures, the last five time values in our ExceLINX data collection sheet were averaged.

Table 5:	FORTRAN	input template
----------	---------	----------------

# of Profiles	# of Radial	# of wall	# of Angles		
	Positions	Temperature	_		
		Readings			
Column Diameter	Particle Diameter				
Radius 1	Radius 2	Radius 3	Radius 4	Radius 5	Radius 6
Reynolds Number	Bed Depth	Angle of Rotation			
Inlet Temperature			-		
Radius 1	Radius 1	Radius 1	Radius 1		
Thermocouple 1	Thermocouple 2	Thermocouple 3	Thermocouple 4		
Radius 2	Radius 2	Radius 2	Radius 2		
Thermocouple 1	Thermocouple 2	Thermocouple 3	Thermocouple 4		
Radius 3	Radius 3	Radius 3	Radius 3		
Thermocouple 1	Thermocouple 2	Thermocouple 3	Thermocouple 4		
Radius 4	Radius 4	Radius 4	Radius 4		
Thermocouple 1	Thermocouple 2	Thermocouple 3	Thermocouple 4		
Radius 5	Radius 5	Radius 5	Radius 5		
Thermocouple 1	Thermocouple 2	Thermocouple 3	Thermocouple 4		
Radius 6	Radius 6	Radius 6	Radius 6		
Thermocouple 1	Thermocouple 2	Thermocouple 3	Thermocouple 4		
Wall	Wall	Wall	-		
Thermocouple 1	Thermocouple 2	Thermocouple 3			

One text file could be made for all packing depths and Reynolds numbers for each packing in each column. Therefore, when using this template, the top three lines were only needed once at the top of the file. To indicate the end of the file, a "-1" was inserted on the last line to notify the program to stop. A sample text file is shown in Appendix A: Sample Fortran Input Text File (for JM 4-hole cylinders in the 2 inch column).

Fitting the IPPF model to collected data

In order to obtain a solution for the program in FORTRAN, the IPPF program uses iterations to minimize the sum of squares function given below:

$$\min(Pe_r, Bi) S = \sum (T_{calc} - T_{measured})^2$$

Where the measured temperatures are taken at each angular and radial position for the different bed depths. The temperatures are made dimensionless by:

$$\vartheta = \frac{(T - T_w)}{(T_o - T_w)}$$

The IPPF model can now be solved analytically. The program performs two sets of functions: first solving the partial differential equation, and the second minimizing the sum of squares shown above as a function of input parameters. The sequence in which this is performed is:

- 1. Guess Per and Bi
- 2. Solve partial differential equation
- 3. Form S
- 4. Is S at a minimum? If not, start back at 1 and reiterate

For the IPPF model, Pe_r and Bi vary for each packing and initial values are needed before running the program. The Biot number was assumed to be anywhere between 1 and 2, so was usually guessed to be 2 (unless iterations were not able to be completed). The Peclet number has been suggested from past research to be about 10 for spheres, and 6 for rings.

The function for the sum of squares is then solved using the guessed values for Pe_r and Bi. This sum is ended dependent on the user's desired accuracy.

The sum of squares function is in the category of target functions $S(\underline{x})$ with $\underline{x} = (x_1, x_2...x_n)$

$$S(\underline{x}) = \sum_{i=1}^{m} f_i(\underline{x})^2 = \underline{f^T f}$$

Where m is the number of data points and n is the number of parameters to be estimated. Then, a m*n Jacobian matrix is formed with the first partial derivatives:

$$J_{ij}\left(\underline{x}\right) = \left(\frac{\partial f_i}{\partial x_j}\right)_{\underline{x}}$$

Using this, the gradient of S is given by:

$$G = 2J^T J + 2\sum_{i=1}^m f_i K^i$$

Where K^i is the matrix of second derivatives of f_i (under some conditions, may be zero). S is minimized by the Levenberg-Marguardt algorithm. The accuracy of this solution is then determined by the accuracy of the radial temperature measurements used in the iterations. The approximate error (<u>E</u>) is then obtained by solving the following:

$$\left(J(\underline{x})^T J(\underline{x})\right) \underline{E} = -J(\underline{x})^T \underline{f}(\underline{x})$$

The second derivative of the target function must be available to approximate \underline{x} . The actual method for calculating \underline{E} depends on which of J, J^TJ , or $(J^TJ)^{-1}$ is available. The accuracy of x_i values is determined by observing how x_i changes over iterations. Values for x_i should not change over the last few iterations (Van Dongeren, 1994).

Although these iterations allow the determination of accurate values for Peclet and Biot numbers, it is important to determine confidence intervals for these parameters. As long as the hessian of S is available in some form, variances may be calculated. If J is known, $G=2J^{T}J$ can be evaluated. Letting H be the inverse of G and S be the sum of squares, then an estimate of the uncertainty in the ith parameter x_i is:

$$var(x_i) = \frac{S}{m-n}H_{ii}$$

Now that the variance is known, the values for a t-distribution with m-n degrees of freedom can be used to give $100(1-\beta)$ confidence intervals for <u>x</u>:

$$x_i - \sqrt{var(x_i)}t_{(\beta/2,m-n)} < x_i^* < x_i + \sqrt{var(x_i)}t_{(\beta/2,m-n)}$$
Where x_i^* is the true solution. For these calculations, a 95% confidence interval was used, meaning 2.5% is tolerated on each side (Van Dongeren, 1994).

F-test

For each flowrate observed, data collected at the same radial and axial positions, but different angular positions, are considered to be "replicates". Therefore, each data set for a given column, packing, and bed depth yields eight replicate sets of six measurements, if two angular positions are used. A first test of model adequacy is to perform an F-test to show adequacy of fit. The pure-error sum of squares is calculated by:

$$SSPE = \sum_{1}^{N} \left(\sum_{1}^{6} \sum_{1}^{8} \left(T_{observed} - T_{average} \right)^{2} \right)$$

Where N is the number of bed depths. Indices are omitted for clarity and averages are taken over each set of eight replicate measurements for each position. The mean square pure error is then calculated by:

$$MSPE = \frac{SSPE}{n_2}$$

Where n_2 =m-(number of averages found). The mean square pure error is an estimate of the error variance. Since this varies weakly over the bed depths, it must be estimated by averaging the error variance over the bed-depths for the "overall" analysis case. The mean lack-of-fit sum of squares is then calculated by:

$$MSLF = \frac{S - SSPE}{n_1}$$

Where $n_1 = m$ – number of parameters estimated – n_2 . If the model is linear in the parameters, the mean lack-of-fit sum of squares is an independent estimator of error variance. However, if the model is nonlinear, then it is a biased estimator, and a test of model inadequacy may be stated as:

$$F_c = \frac{MSLF}{MSPE} > F_{0.05}(n_1, n_2)$$

With values of $F_c > F_{0.05}$, either the model is a poor fit or the data has a low amount of variability. This indicated a lack of fit error is significant compared to inherent scatter in the data.

Results

In order to proceed *with* any results, it was necessary to first prove that the data collected was representative of what was expected compared to past experiments. This includes the following:

- Plots of θ vs. y should show a parabolic fit at the first bed depth
- On the Plots of θ vs. y for each Reynolds number, temperatures should be lower with a higher bed depth
- On a height to height basis, k_r/k_f and Nu_w should be height independent
- Peclet and Biot numbers are in line with previously found literature figures

Parabolic Fit

Over the vast amounts of experiments done with fixed bed reactor tubes, it has been found that temperature profiles in the tubes fit a parabolic curve of the form $T = A * r^2 + B$, or in dimensionless form, $\theta = A * y^2 + B$. Figure 7 shows an example of a parabolic fit for the first depth for raschig rings in the 4 inch column. As expected, the data follows a parabolic fit very closely with a coefficient of determination of 0.99. It would also be expected that $\theta_{y=0} = 1$ for the best fit, this is due to how the θ is calculated in terms of the center temperature and as so would only hold true if the center of the column had not cooled more than the surrounding locations. This example has an intercept (A) of 1.005, very close to 1. Some temperature profiles for other packings are not as parabolic as expected. Figure 8 is an example of a parabolic fit for the first height of JM 4-hole cylinders. This poor parabolic fit was uncommon however and only occurred in two instances, the raschig rings and the JM 4-hole cylinders in the 2 in column, both of which were instances with a particularly low N value. Graphs expressing the parabolic fit for first depth temperature profiles for each packing/column combination can be found in Appendix B: θ vs. y for First Heights.

First Height Parabolic Fit for Raschig Rings



Figure 7: Temperature Profile Parabolic Fit for Raschig Rings in the 4 inch column with Reynolds Number 432



Figure 8: Poor parabolic fit for temperature profile of JM 4-hole cylinders in the 2 inch column with Reynolds Number 1052

The dimensionless profiles should also make physical sense. Since this is a cooling experiment, higher bed depths should yield lower temperatures. Temperatures should also decrease farther from the center of the tubes. It would also make sense for temperature differences between successive bed depths to be lower at greater bed depths. As air travels through the tube, the temperature difference between the center of the tube and the walls is decreased, lowering the driving force of the heat transfer. This means that the rate of temperature decrease with increasing radial position should also decrease with increasing bed depths. A sample radial temperature profile is shown in Figure 9.



Figure 9: Temperature Profile for JM 4-hole cylinders in the 4 inch column for Reynolds Number 268

Figure 9 is representative of the majority of our temperature profiles, and shows the expected trends for radial profiles obtained during the experiment. It would also be expected for higher

Reynolds numbers to yield higher temperatures. Figure 10 is a temperature profile of the same packing/column as Figure 9 but with a higher Reynolds number.



Figure 10: Temperature Profile for JM 4-hole cylinders in the 4 inch column for Reynolds Number 358

Each data point on Figure 10 corresponding with the same bed depth and radius as points on Figure 9 have higher temperatures. Figure 11 displays a temperature profile for a given bed depth rather than Reynolds number.



Figure 11: Temperature Profile for JM 4-hole cylinders in the 4 inch column for Bed Depth 200mm

There were few temperature profiles that did not represent our expected findings. Some of the temperatures for higher bed depths would be higher than those for lower bed depths, as well as some temperatures at higher radii being higher than those closer to the center of the tube. This can be a result of bent thermocouples where even a slight deviation from their expected position would cause such a discrepancy. Furthermore since most of the uncharacteristic behavior was seen in the near wall region it could be the result of wall effects dominating this portion of the tube. Figure 12 is an example of these findings.



Figure 12: Temperature profile for Monoliths in the 4 inch column with Reynolds Number 129

It is also understood that θ should not exceed 1, as T_o should be the greatest temperature recorded; however, for some packings, θ values of greater than 1 were recorded. This also contradicts prior statements regarding the fit being parabolic with an intercept of 1. Larger than expected θ values were only recorded for raschig rings in the 2 inch column (N=3.5). An example of this is shown in Figure 13. This could be the result of the larger currents created by the larger packing. Because the larger packing takes up a much larger fractional area of the tube cross section it can in effect, if positioned in a certain manner, channel the hot air around the center thermocouple effectively creating a cool spot in this region and resulting in θ of over 1.



Figure 13: Example of θ exceeding 1; Raschig Rings in the 2 inch column with Reynolds number 1570

41 different sets of Reynolds numbers were examined over this course of this project (5 for the spheres, 6 for the other 6 packing/column combinations). Radial temperature profiles for all 41 sets of Reynolds numbers can be found in Appendix C: θ vs. y by Reynolds Number.

Depth independence on heat transfer parameters

The heat transfer coefficients k_r/k_f and Nu_w are strictly based on the packing size and shape, and airflow. It was once assumed that these parameters were both expected to decrease with bed depth because the models using to fit the parameters were not accounting for the axial dispersion of heat (DeWasch & Froment, 1972; Gunn & Khalid, 1975). However, it has been shown that bed depth should not be a factor in determining the heat transfer coefficients if the IPPF model is used. In order to show this, the data for the second, third, and fourth heights completed were

compared directly to the first height and values were found for the coefficients. Sample graphs of height by height analysis are shown in Figures 14 and 15, using IPPF model. The vertical error bars for all data collected in this study represent 95% confidence intervals.



Figure 14: Height by height analysis of kr/kf for JM 4-hole cylinders in the 4 inch column with Reynolds Number 584



Figure 15: Height by height analysis of Nuw for JM 4-hole cylinders in the 4 inch column with Reynolds Number 584

Most data obtained presents no strictly positive or negative trends. For respective bed depths, if k_r/k_f decreased, Nu_w would usually increase, and vice versa. Since the overall heat transfer coefficient, U, is dependent on both of these, increasing one and decreasing the other will not affect this parameter. Figure 16 represents how h_w and k_r/k_f are representative of this, showing that using the following equation for the overall heat transfer coefficient:

$$\frac{1}{U} = \frac{1}{h_w} + \frac{\alpha R}{k_r}$$

An increase in Nu_w and decrease in k_r/k_f will yield the same value for U. This has also been seen in the literature. Landon (1996) also showed this; this is shown in Figure 16.



Figure 16: Illustration of bed depth dependence for heat transfer parameters (Landon, 1996)

Peclet and Biot number analysis

Graphs of the Peclet and Biot numbers were also compared to expectations. All except for one, the graphs are representative of our expectations. As stated before, it was expected according to their definitions that the Peclet number would increase rapidly at low Reynolds numbers and level out at a certain value of the Reynolds number. Figure 17 presents the Peclet number over a range of Reynolds numbers for 4-hole cylinders in the 4 inch column.



Figure 17: Peclet number versus Reynolds Number for JM 4-hole cylinders in the 4 inch column

The data presented above shows that the Peclet number does not deviate with respect to Reynolds number. The data also shows that the Peclet number for Monoliths is approximately 4.5, which is believable compared to the expected 6 for ceramic particles. This lower value indicates superior heat transfer and as such would make the particle more appealing. Since lower Reynolds numbers were found not to yield reliable data, the values at which Peclet increases rapidly with respect to Reynolds number were not examined. One of the packing/column combinations yielded data unrepresentative of our expectations. This is shown in Figure 18.



Figure 18: Peclet Number versus Reynolds Number for JM 4-hole cylinders in the 2 inch column

It is very unlikely that Pe_r would behave in such a manner, and the reasoning for such behavior is uncertain. This might be due to the small N value not allowing the heat to disperse evenly across the cross section of the column, causing a discrepancy in parameters at different flowrates.

The Biot number is expected to decrease with respect to Reynolds numbers. All data of Biot number versus Reynolds number obtained in these experiments expressed this behavior. A sample of these graphs is shown in Figure 19.



Figure 19: Biot Number versus Reynolds Number for Raschig Rings in the 2 inch column

Most values for the Biot number fall in the range of 1-2, which is close to the expected value. All graphs of Peclet and Biot numbers plotted against Reynolds numbers can be found in Appendix E: kr/kf, Nuw, Bi, and Per for all Packings and Columns.

The Bauer/Schlunder Method

A study done by Smirnov (2002) showed an equation for finding k_r/k_f:

$$\frac{k_r}{k_f} = \frac{k_r}{k_f}$$
, bed + K * Re * Pr

Where Pr is the Prandtl number for air, which is assumed constant at 0.71. The parameter $k_r/k_{f,bed}$ is the effective radial thermal conductivity of the packing with stagnant air. This parameter must be determined experimentally by plotting k_r/k_f versus Reynolds number and finding the y intercept of the best fit line. In Smirnov's (2002) study, this was assumed to be 10 for ceramic particles (regardless of shape or size) and 20 for glass particles. Although this is much less significant than the slope of the line (considering steam methane reforming is undergone at very high Reynolds numbers), it is important to verify that this value is close to expected to ensure that the slope is accurate. This means that having a negative $k_r/k_{f,bed}$ value will yield a slope that cannot be assumed to be perfectly accurate, and therefore extrapolating will not be precise.

This leaves K, which may be determined experimentally or mathematically through the Bauer/Schlunder method. K varies for each packing, and depends on size (N), shape, and material. Experimentally, this would be the slope of the k_r/k_f versus Re plot divided by the Prandtl number. The Bauer/Schlunder method (Bauer & Schlunder, 1978) for determining K can be expressed by:

$$K_{Bauer} = \frac{X}{8 * \left\{ 2 - \left[1 - \left(\frac{1}{R/d_p} \right) \right]^2 \right\} * d_p}$$

Where X for cylindrical particles can be expressed as:

$$X = \frac{\varepsilon_{bed}}{\varepsilon_{hbed}} F_{cyl} d_p + \frac{\varepsilon_{hole} \left(1 - \varepsilon_{bed}\right)}{\varepsilon_{hbed}} F_{hole} d_{hole}$$

Where $F_{cyl} = 1.75$, $F_{hole} = 2.8$, $d_{hole} = \sqrt{2}l$, ε_{bed} can be assumed as 0.40 for cylindrical particles, ε_{hole} is the porosity of one particle, and $\varepsilon_{hbed} = \varepsilon_{bed} + \varepsilon_{hole} (1 - \varepsilon_{bed})$. For Spherical particles, X can be expressed as:

$$X = 1.15d_{p}$$

Experimental K values were compared with those found mathematically using the aforementioned equations. The Bauer/Schlunder equation does not correlate our results well. However, correcting the equation to read:

$$K_{Bauer,corrected} = \frac{X}{7 * \left\{ 2 - \left[1 - \left(\frac{1}{R/d_p} \right) \right]^2 \right\} * d_p}$$

By correcting the K_{Bauer} equation, it was found that the equation correlates our experimentally found K values nicely. This yields the following K values:

Table 6: Compa	rison of K _{Bauer} a	and K _{exp} to acco	ount for corrected	l equation
----------------	-------------------------------	------------------------------	--------------------	------------

Packing	K _{Bauer}	K _{Bauer,corrected}	K _{exp}	Ν
Raschig Ring (4 inch)	0.226	0.258	0.252	7.0
JM 4-hole cylinders (4 inch)	0.195	0.223	0.225	5.8
Raschig Ring (2 inch)	0.185	0.212	0.175	3.5
JM 4-hole cylinders (2 inch)	0.161	0.184	0.183	2.9

The value of 8 used in the denominator of the K_{Bauer} equation has been found for an ideal model. Since the experiment was not ideal, changing this value to 7 works similar to that of a correction factor to correct for the reality of this experiment.

A study completed by Borkink et al. (1993) compared Peclet number (Note that this is not the radial Peclet number, but instead Pe = Re * Pr) to the effective radial thermal conductivity for different packings including raschig rings with an equivalent spherical diameter of 6.2mm in a 49.9mm column (N=8). This was compared to data obtained in this study for raschig rings in the 4 inch column (N=7). To obtain the Peclet number, the Reynolds number was multiplied by the Prandtl number for air. This comparison is displayed in Figure 20.



Figure 20: Comparison of Borkink Raschig Ring effective radial thermal conductivity to values from this study

The slopes for both best fit lines are nearly identical, verifying that the value determined for K (which is the slope for the best fit lines above) using the corrected Bauer and Schlunder equation is accurate (according to this and Borkink et al.).

Comparison to past experiments using spheres

The data from this experiment was also compared to various other studies which used ¹/₂ inch porous ceramic sphere packing in the 4 inch column. These studies were performed by Hans Van Dongeren (1994) and Darryl Pollica (Dixon, Personal Communication). The two studies were both performed using a heating method rather than cooling; cool air entered the column at the inlet and steam in the jacket heated the air up. Although these were not cooling experiments as this study was, the two are comparable as the IPPF model adjusts all variables to be dimensionless.



Figure 21: Comparison of k_r/k_f for spheres



Figure 22: Comparison of Nu_w for spheres

The k_r/k_f comparison illustrates that this study, Pollica, and van Dongeren all determined that the intercept should lie somewhere around 15, however, none of the slopes strongly correlate from one study to another. Although it appears that van Dongeren has a stronger correlation with more data points than the other two studies, a slope (where slope = K*Pr) of 0.044 is far less than the expected 0.07 for a spherical packing with N=8 (Bauer & Schlunder, 1978).

Although Nu_w is not linear in nature with respect to Reynolds number, it can be compared using linear trends at low flowrates. The intercept collected in this study deviates greatly from the intercepts obtained by Pollica and van Dongeren (which are both about 16). However, disregarding the first data point (at a Reynolds number less than 100), the intercept is less than 20. The slope in this study is identical to that of Pollica, which shows that both our study and Pollica's will yield similar Nu_w values at very high flowrates.

Asymptotic radial Peclet relationship to k_r/k_f

Von Landon (1996) stated that at high Reynolds numbers, one may disregard $k_r/k_{f,bed}$ in calculating k_r/k_f by using the equation:

$$\frac{k_r}{k_f} = \frac{Re * Pr}{Pe_{r,f}^\infty}$$

Where $Pe_{r,f}^{\infty}$ is the radial asymptotic Peclet number at high Reynolds numbers. This equation states that $Pe_{r,f}^{\infty} = \frac{1}{K}$. There are empirically derived equations for $Pe_{r,f}^{\infty}$, such as Bauer and Schlunder, but when using different shaped particles, these equations cannot be assumed accurate. To determine this value, these experiments need to be run at high Reynolds numbers. However, with varied data for Pe_r at higher Reynolds numbers, this calculation cannot be assumed perfect. With the data taken in this study, $Pe_{r,f}^{\infty}$ is the approximate average of Pe_r values over the last 4 Reynolds numbers in which data was recorded. For example, for 4-hole cylinders in the 4 inch column (Figure 17), values for Pe_r average 4.5.

Packing	K _{exp}	$Pe_{r,f}^{\infty}$	$1/Pe_{r,f}^{\infty}$	N
Raschig Ring (4 inch)	0.252	4	0.25	7.0
JM 4-hole cylinders (4 inch)	0.225	4.5	0.22	5.8
Monoliths (4 inch)	0.225	3.5	0.29	14.0
Raschig Ring (2 inch)	0.175	6	0.17	3.5
JM 4-hole cylinders (2 inch)	0.183	6	0.17	2.9
Monoliths (2 inch)	0.225	4.5	0.22	7.0

Table 7: Correlation between experimentally found slopes and radial Peclet numbers

The only packing that greatly deviates from K_{exp} is the monoliths in the 4 inch column. The correlation between Peclet and Reynolds numbers is shown in Figure 23.



Figure 23: Peclet versus Reynolds number for monoliths in the 4 inch column

As shown in Figure 23, the Peclet number is increasing over Reynolds numbers instead of remaining constant. This is most likely due to the flowrates being too low for the Peclet number to asymptote, and therefore $Pe_{r,f}^{\infty}$ is not definable for this packing. According to the experimental K value determined by the slope, $Pe_{r,f}^{\infty}$ should be 4.5, and very well might be. Comparing $1/Pe_{r,f}^{\infty}$ and slopes of k_r/k_f to Reynolds number is also useful to confirm that the data has been collected at high enough flowrates where the flow is fully developed. The lower the value for $Pe_{r,f}^{\infty}$, the higher k_r/k_f is, meaning that the heat transfer would be overestimated for cases such as this.

Analytically determining a Nusselt wall value

Martin and Nilles (1993) proposed that Nu_w may be determined analytically by using the following equation:

$$Nu_w = \left(1.3 + \frac{5}{N}\right) \frac{k_r}{k_f}, bed + 0.19Re^{0.75}Pr^{0.33}$$

In a simpler form, this equation is:

$$Nu_w = A + 0.1697 Re^{0.75}$$

Where A is the intercept based on the packing's N and $k_r/k_{f,bed}$ value. There are equations to determine theoretical values for $k_r/k_{f,bed}$, however, this study used the values of the intercepts from graphs of k_r/k_f versus Reynolds number. Since Prandtl is assumed constant for air at 60°C to be 0.71, the second term is solely dependent on Reynolds number. The data collected in this study does not support the accuracy of this equation. An example of this equation compared to this study is shown in Figure 24.



Figure 24: Nu_w versus Re for raschig rings in the 4 inch column

In order to determine correct values for Nu_w , the intercept of k_r/k_f versus Re graphs must be accurate. Some of the data in this study yielded intercepts far lower than expected (some negative) which causes the Martin and Nilles comparison to be far off from the collected Nu_w data. The Martin and Nilles equation compared to this study's data for 4-hole cylinders in the 4 inch column is shown in Figure 25.



Figure 25: Nu_w versus Re for 4-hole cylinders in the 4 inch column

It can be seen that the series created by the Martin and Nilles is relatively parallel to the data obtained in this study; this deviation of the equation to collected data is solely dependent on the intercept ($k_r/k_{f,bed}$). After correcting the $k_r/k_{f,bed}$ value to be 8 instead of the obtained 0.2835 for the 4-hole cylinders in the 4 inch column, a much better correlation was found. This is shown in Figure 26.



Figure 26: Nuw versus Re for 4-hole cylinders in the 4 inch column with corrected kr/kf bed

N Dependence

Through the course of the experiment the value of N ranged from 2.9 to 14 depending on the packing and tube, the full list can be found in Table 2. Typically N can be expected to fall in the range of 5 to 15 (Von Landon, 1996) and it is generally accepted that this range will produce the desired heat dispersion profiles. As the N drops significantly lower than this range you start to encounter data that does not fit the predicted profiles. This is largely due to the wall effects which dominate the region within a particle diameter of the tube wall (Smirnov, 2002).

Therefore, for any N \leq 2, the entire tube region would be affected entirely by wall effects; N must be large enough so that the majority of thermocouple readings are not affected by this phenomena. This can be observed by looking at the parabolic fits for the runs where the Raschig Rings and the JM 4-hole cylinders in the 2in column, N=3.5 and N=2.9 respectively (figures 5 and 6), by far produced the least parabolic curves at the first bed heights. Further evidence of poor model prediction in this region comes from values of θ larger than one found for the Raschig Rings in the 2 inch column as well as an uncharacteristic Peclet versus Reynolds correlation for the JM 4-hole cylinders in the 2 inch column. These findings come in contrast to a study done by Dixon (1997) which concluded that an N of as low as 2 for non-sphere particles will still produce the desired results.

Particle Durability

All the particles in this study were composed of ceramics however they varied in both shape and size. It was found that the monolith packing, being of smallest size and having the thinnest walls was far more brittle than the other packings. While the packing heights in this study remained below 300mm in all instances it was still found that many monoliths at the bottom of the column, whether as a result of weight or agitation cracked and crumbled. The raschig rings were far more durable than the monoliths however even these resulted in a few broken particles most likely during the initial packing of the tower. The JM 4-hole cylinders and the spheres were by far the most resilient of the particles as none of these showed cracking or breaking.

Analysis of Variance

In order to ensure that the data is accurate, the F-ratio of F/F_{95} should be less than 1. However, it was found that for packings in the two inch column, values larger than 1 were very common. It was also found that values larger than 1 were often found for monoliths in the 4 inch column. For the raschig rings and 4-hole cylinders in the 2 inch column, this was most likely due to the low N value, causing less uniform heat dispersion, causing the sum of squares to be very large. However, since this is not the case with the monoliths, the discrepancy may be attributed to the large voidage within the particle. A table with all F-ratio values can be found in Appendix F: F-test values.

Conclusion

The IPPF model requires that the lowest height of packing used should provide a z/R ratio of 3. It has been stated that a value below this results in insufficient development of the profile at the inlet height resulting in failure of the model. However, data collected in this study for z/R ratios of 2 showed parabolic profiles. In future studies, if this is found not to be the case, a new model would need to be utilized that does not require a parabolic fit of the temperature profile.

The value of N displays a positive correlation to Nu_w resulting in a distinctive rise in the latter when N was double by moving to the smaller tube. Furthermore an N \leq 3.5 is dominated by wall effects and results in data that does not fit the model while \geq **S**.8 fits the model well. Further studies would need to be conducted in the range of $3\leq$ N \leq 5.8 to determine at which point the wall effects start to dominate a sufficiently large portion of the tube to negate the model.

Analyzing our data for the packings in the 4 inch column, we were able to not only get nearly identical findings for raschig rings, but were able to determine analytical models for determining heat transfer parameters based on shape and size. The Bauer/Schlunder equation, when corrected, gave parameters identical to those collected experimentally. It was also found that Von Landon's $Pe_{r,f}^{\infty}$ correlation to k_r/k_f is a fair estimation. It was however found that the Martin and Nilles equation for determining Nu_w cannot be relied on for accurate measurements as it only depends weakly on particle shape.

The JM 4-hole cylinders and Monoliths studied provided the best heat transfer parameters. However, since the Monoliths are brittle and would be subjected to high forces during steam methane formation, these should not be considered for industrial processes. Further studies should compare the differences between similar shapes such as 3-hole, 4-hole, and 5-hole cylinders.

Nomenclature

Bi	Biot Number $(Nu_w \frac{R/d_p}{k_r/k_f})$
$Bo_{h,r}^{\theta}$	Bodenstein Number $\binom{GC_p R^2}{k}$
Cn	Heat capacity
d_{n}	Particle diameter
d_{hole}	Characteristic length of the particle $(\sqrt{2}l)$
dt	Tube diameter
G	Mass flowrate of the gas
$\mathbf{h}_{\mathbf{w}}$	Wall heat transfer coefficient
k _r	Effective radial thermal conductivity
$\mathbf{k}_{\mathbf{f}}$	Effective thermal conductivity of the fluid
F	Variance
L	Largest bed length analyzed
Ν	Tube to particle diameter ratio (d_t/d_p)
Nu_w	Wall Nusselt number $\left(\frac{n_w a_p}{k_f}\right)$
Pe _r	Radial Peclet number $\left(\frac{R_{e*Pr}}{k_r/k_f}\right)$
$Pe_{r,f}^{\infty}$	Asymptotic radial Peclet number $\left(\frac{Re*Pr}{k_r/k_f}\right)$ at high Re values)
Pr	Prandtl number (0.71 for air at 60 \degree C)
r	Radial position
R	Radius of the tube
Re	Reynolds number $\left(\frac{v_s \rho d_p}{\mu}\right)$
Т	Temperature
To	Center temperature of the first bed depth
T_w	Wall temperature
U	Overall heat transfer coefficient
Vs	Superficial velocity
Х	Dimensionless bed depth (z/R)
У	Dimensionless radial position (r/R)
Z	Bed depth
α	Experimentally determined constant
E _{bed}	Overall had porosity
E _{hbed}	Deresity of one netticle
Ehole	Effective redial thermal conductivity (some as k)
λ.	Viscosity
μ	Density
Р Д	Dimensionless temperature $\binom{(T-T_w)}{v}$
σ	Dimensionless temperature $\left(\frac{w}{(T_0 - T_W)}\right)$
ζ	Dimensionless bed depth $\left(\frac{x^{n}/L - x_{o}^{R}/L}{1 - x_{o}^{R}/L}\right)$

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Appendices

Appendix A: Sample Fortran Input Text File (for JM 4-hole cylinders in the 2 inch column)

4	6	3	2			
50.8	17.4244					
8.5	12	15	18	21.5	24	
2275	80	0				
98 69934	515					
73 51792	2602	78 67514	038	77 5713	257	77 86281738
77 32102	2002	80 / 5686	5036	76.0621	673A	73 9961/106
76 00073	200	74 60718	2007	76 7808	3646	73.77014100
60 05260	000 077	70.06601	560	70.7000	7697	69 00951226
60 2140/	1012 157	(19.9000)	51	(0.7112	7062	71 04101106
09.21404	+37	09.03041	2009	70 5207	(4/4	/1.04101100
03.02440	506	60.36583	9998 195	12.5381	0342	60.9682289
19.60180	0308	21.43957	25	25.9464	1496	
2275	80	45				
98.77065	5276					
73.27800)448	75.00024	414	79.9264	8164	76.18695678
73.44379) 732	77.16934	51	75.3780	2274	78.39521024
70.07826	5082	76.28060)302	78.6166	8702	74.13243866
69.04063	3262	73.66384	276	73.6929	1838	71.69318848
72.88054	4352	68.13284	76	71.9868	3624	72.6760376
63.12102	2432	61.88073	3728	69.8736	2978	65.75786592
19.51357	7994	21.36859	955	25.9989	2502	
1822	80	0				
99.00328	337					
72.00116	5424	73.03620	758	78.0530	1664	74.74094236
71 51310)426	75 45122	2678	74 2185	2876	77 48835146
67 20661	1316	74 16232	908	78 2910	904	73 26902616
69 28578	R644	71 16252)612	73 7652	176	71 61493072
70 35703	3/28	71.04037	1012	68 3129	68/6	69 6669754
60 /158/	102	60 70260	010 1866	72 3522	7066	65 32000304
20.20622	+75	22 0019200	1000	72.3322	1022	03.32909394
20.29053	9002 90	22.00160	1892	23.9550	1952	
1022	80 2000	43				
98.91923	5066		226	70 5450	0770	75.07102526
77.16016	5844	70.76894	226	73.5458	8//8	75.0/192536
68.48989) 41	74.84447	022	77.0072	0824	76.84577026
79.58320)314	72.13587	802	74.3673	2786	74.35321196
76.23908	8386	61.07149	9504	69.6418	7774	76.7695099
72.09566	5194	70.35791	016	62.0732	8492	72.56248322
50.56270)676	64.38652	2344	57.6177	574	60.06700898
20.44216	5992	22.02308	3922	25.8932	8118	
1412	80	0				
98.76869	9048					
73.56212	277	67.04054	26	70.0010	3302	71.15197754
65.63481	1446	71.73593	3292	73.1474	1362	73.5072876
76.63531	195	68.17412	2874	70.9686	1876	70.14902648
72.67462	2006	57.41319	732	66.3239	807	73.62355348
67.15747	7682	66.80137	133	58,5466	9038	68.66848298
47 83555	5144	60 07205	5964	53 9432	6248	57 07460404
19 71891	1554	21 07030	0492	24 4406	624	57.07 100 101
1/12	80	A5		24.4400	024	
00 01114	50 570	45				
72 2/10	279 7029	60 24677	070	72 0000	0006	60 62702200
13.34181	1720 7211	U7.240/1	210	75 1 600	0070	07.02/02200
07.89277	1344	/3.18043	0524	75.1600	0/0	10.93996/06
70.12002	2104	/1.0/35:	021	/1.2335	5406	10.35523376
14.25852	2206	65.93112	2/94	/1.3691	0858	65.408847/04
64.26127	778	60.37468	3032	71.2966	324	61.95517502
52.64456	5712	64.01000)594	64.4524	0936	51.4372383

19.72838284	21.09444772	24.4977024	
1052 80	0		
98.8001999			
68.29434206	65.19507292	66.60065306	66.83636778
64.68190612	68.25741732	69.82284546	70.71866304
63.92250366	67.90347598	68.50434264	68.65388334
69.14316408	63.49986954	65.06321718	68.72596892
62.48444292	56,15174638	67.8101944	52.43913954
51.44942398	64.33299254	53.92048034	54.6673523
20.2851219	21.36883354	24.19305116	0 1100 / 00 20
1052 80	45		
98 89687198	10		
59 91440584	58 86440276	59 0906105	59 24870224
64 17294388	56 07076494	62 93479616	58 85851134
60 8/1555/3/	54 73794404	60 27732468	63 80253372
53 721001/6	63 26296768	59 98911668	55 78862226
<i>A</i> 7 9710197 <i>A</i>	58 2/19/3086	58 64743574	59 78501664
48 59130326	50.051342	56 233/3276	18 8/608/6
20 27505568	21 15076448	23 02082404	+0.0+000+0
20.27505508 775 80	0	23.92002404	
00 13381701	0		
50 15823716	61 688/680	62 73048478	62 46504518
65 0518/78/	61.0686/318	65 21030846	58 <i>4</i> 137367 <i>4</i>
50 70376434	57 51/10678	64 41526184	63 01205608
50.00545746	63 00750508	58 552787	53 02500182
51 70502546	61 2021370	56.01201406	58 56207108
<i>A</i> 7 50085186	55 72848282	53 45356826	14 60525436
20 21828842	21 1652122	23 46450046	44.00525450
20.31828842 775 80	21.1055122 45	23.40439040	
00 5616104	45		
68 68361206	63 5823050	61 86362016	62 27270254
62 52201472	65 00607784	61.00472102	62 42222518
64.0026122	64 20008626	61.09475192	65 22100006
04.0920132 57 55045020	64.59996020	60 02675782	64 28205602
57.55845808	04.03/30/32	00.02073782	04.38393092
00.25572408 56.00006926	59.1007772	02.8010829	39.52434004
56.02996826	50.4055042	55.21190646	48.15922166
20.31330402	21.18019424	25.48/11/58	
409 80	0		
118.6779892	52 45747606	51 051 640 46	52 74296059
01.39330008	52.45/4/000	51.95104940	55.74280958
52.38008300	55.49029696	50.82851026	55.43695144
35.15008468	53.41965256	50.87927324	55./39/484
48.3984268	52./3624648	48.4462166	50.59808884
40.3808/44	48.52955476	51.00205214	40.51519552
45.83642578	37.79503402	43.69383314	32.8810745
20.18028944	20.63690606	22.03336868	
409 80	45		
115.86/2944	10 202 (2502	10 1005150	17 00001 500
46.89469604	40./0/6/592	40.493/1/2	47.82091598
44.55/39288	37.52248078	44.38219528	4/.3962/154
49.52509384	34.64694902	40.84518738	46.16904524
41.36538162	42.24956284	45.1345604	44.68399504
30.60537682	42.41700286	43.01247252	45.32846682
32.8019684	38.80463332	42.5204437	50.85472978
20.1/482872	20.52550964	21.59088936	
2275 150	U		

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2027 72958 00244 59544 56608 71468 76772 26404 91338 9471 02538
2027 72958 00244 59544 56608 71468 76772 26404 91338 9471 02538 58202
2027 72958 00244 59544 59544 56608 71468 76772 26404 91338 9471 02538 58202
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2027 72958 00244 59544 56608 71468 76772 26404 91338 9471 02538 58202 84662 70826
2027 72958 00244 59544 56608 71468 76772 26404 91338 9471 02538 58202 34662 70826 421
2027 72958 00244 59544 56608 71468
2027 72958 00244 59544 56608 71468
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2027 72958 00244 59544 59544 59544 59544 59544 59544 50608 71468
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2027 72958 00244 59544 56608 71468 76772 26404 91338 9471 02538 58202 34662 70826 421 50302 01564 57564

41.19297028	49.95391008	50.35421524	43.21306688
38.8451248	47.79931642	49.53569186	47.91036528
51.21306152	43.73466644	52.38024904	45.05809172
47.18281328	50.41306534	46.15266038	36.77309798
33.3092529	48.60114438	50.73813096	36.52398222
19.16157456	20.50416106	23.67472992	
1052 150	45		
98 62762146			
46 55139084	50 37760542	48 8636734	41 72287138
45 81043778	50.49033814	48 18919678	40 85421524
46 73076936	50.27729416	51 40846788	43 54044876
51 33971708	47 48421098	48 51408158	38 6225525
50 43963316	52 52350996	45 03630522	42 02005766
38 76716766	<i>4</i> 9 8013588	12.09050922	31 79/61822
10 2223526	20 58517052	23 8/02817	51.79401022
775 150	0	23.0402017	
100 /860/02	0		
15 82597198	18 88180924	48 64862902	13 629951/18
45 46760638	49 50203702	48 50621186	42 5876/3//
46 45537264	49.30203702	40.50021100	45 5058670
40.45557204	49.10773392	49.0059912	40.000079
49.3028244	45.025275	47.20775512	12 73/80836
47.500525	49.20344370	40.74000362	42.73489830
10 61/28872	20 74641036	40.74909302 23.3406680	52.50404094
775 150	20.74041030	23.3490089	
07 / 851503	45		
/8 8/131/7	/9 661/138	15 1838379	46 24140396
48 72106704	48 00358522	43 23610688	45 0/310012
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47.4505007	46.55472710	40.49301912	46 35172424
40.4001447	43.473304	45.57594250 36.64361114	40.33172424
10 61/37088	20 76703708	23 27057 <i>1</i> 22	52.95051194
19.01437988	20.70703798	23.37937422	
108 5680758	0		
26 21 22 9 26	26 00975951	27 71766916	22 0001000
36 42000266	33.04670334	30.76653078	33.9221222
35 80542754	36 14840626	30.70033978	34.41045554
21 25526106	27 22017542	20.22703204	22 74912216
24 72056100	37.33917342	29.37197228	33.74812310 22.4170845
22 75461652	24.12473432	32.94790344	35.4170643
52.75401052 10.10100756	29.23907438	20.09002202	20.30184174
19.19199/50	19.052/851	21.40013428	
409 150	43		
26 70264756	26 4160065	22 58012774	24 5112465
26 55220592	24 16927616	21 1742697	24.0112403
27 12656172	34.1003/010	31.1/4208/	24.80330/14
3/.120301/2	30.32278898	30.07872924	34.82304308
24 00120416	37.33473206 25.24252044	29.40720340	25 00122766
32 65042026	23.34233044 28.8647785	22.03740304 26.1224550	25 78507009
32.03042030 10.15055592	20.004//0J	20.1234338	23.10391908
17.13733382	19.0201311	21.4020009	
2213 200 00 16752000	U		
56 80071019	56 5553677	55 35040804	55 66502022
56 48750004	55 8770172	51 6775700	55.00593932
55 06776856	55 50765220	53 07120220	51 80701426
55.00270850	55.50705220	55.7/100520	51.00721430

53.6000389	56.02751234	55.77091596	54.49867782
53.37738264	50.68227464	48.56469346	52.22539214
50.68758774	47.39768144	53.3546509	42.7739067
19.9816288	24.27942428	27.3081413	
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99.32751464			
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56.35706176	57.1123329	55.01497806	55.8926445
56.32222672	54.2671028	56.18304598	54.26648866
55.97688828	55.09260254	52.54919816	52.98763428
52,77218934	52,95060806	54,98386228	52.06026304
48.62270968	54.22021406	50.13025282	47.67022018
19.96664236	24.4571537	27.47696648	
1822 200	0		
98.75254518	•		
52.77601622	52.2064331	52.1440544	51,74226988
52 39601898	52.94602584	50 73092422	51 81161346
52 09917146	49 93562622	51 71668168	49 73918764
50 47908556	50 70390778	48 12815628	48 78876494
47 19949416	46 8016281	50 37097092	48 32941208
<i>A</i> 3 98571776	40.0010201	<i>16</i> 1387558	40.52241200
19 47835502	22 96445274	25 7378071	44.31240724
1822 200	45	25.7570071	
08 00816742			
A6 73545458	12 03614576	15 30166628	18 28736572
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45.19461560	21.75469702	47.51058042	40.57550880
28 2220200	39.40233032	43.84931708	40.02294230
38.2229309 20.54504574	38.030700	44.00333870	44.44020092
39.54594574	43.10226364	46.//5444/8	36.17896652
32.50676192	32.80727462	38.04145966	41.05914232
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47.7934906	46.25/5569	48.8368439	47.38877412
45.2327606	47.04674378	48.03281784	46.26497802
44.3535927	40.13817138	47.12823716	47.4955345
44.86109696	35.17089234	44.22092666	48.9150421
39.61377488	44.59798128	44.9947319	38.58955536
30.74430542	42.20111084	37.01335526	43.82090456
19.7666401	22.48489492	25.22439576	
1412 200	45		
98.5345642			
46.32924804	48.3742035	48.14803698	47.5756569
46.00261536	48.96492462	46.95700226	46.0573738
42.17110368	46.27195664	48.52537918	44.81440962
46.5326851	38.79361878	45.09022902	41.37486496
43.4593918	44.30651016	45.34169312	34.4771507
37.65764236	44.62176666	43.78035812	34.187677
19.81744498	22.5064728	25.27298088	
1052 200	0		
98.90518646			
41.69141464	43.93356322	43.96338578	43.00163726
42.08356248	44.7333618	42.92609786	41.22968826
38.4459564	41.72648162	44.28848724	40.32664414
42.73218156	40.75215912	41.01621246	36.77588576
38.96191406	39.5943901	40.25822066	30.58843956

33.66010284	40.84319838	40.1227333	30.39145394
19.6530487	21.6501873	24.25802344	
1052 200	45		
98 94475404			
42 642659	40 93933562	43 49178466	42 71018754
40.83062362	40.999955502	42 83531266	41 70334852
30 34235308	35 26044732	41 42165374	42 3284668
40 17110006	26 20108656	41.42103374	43.3264006
40.1/119900	30.29108030	39.33080138	42.11/3003
33.40/955/0	39.31444244	41.40008244	34.30531998
28.95948334	35.948/8388	32.84912796	37.29596784
19.65926816	21.48394086	24.13/45194	
775 200	0		
98.44761658			
38.6051117	37.4156601	39.48222732	38.845166
37.08525314	38.17751694	39.177832	37.64824982
35.41690064	32.3655525	37.5056557	39.48755798
36.19804	32.72103272	35.99720536	38.1797447
30.77688676	35.59904098	37.54720688	31.55895578
26.36857068	31.97444456	29.73430366	33.805246
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775 200	45		
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37.01532516	35.3193451	37.41386568	37.83820574
29.9499901	32.99298934	37.02521744	38.69386292
29.44973108	35.1542488	34.28367692	38.43269804
28.45227546	26.36058654	34.58326036	34.8159691
19.44895478	20.87421608	23.1486824	
409 200	0		
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31.03306464	30.35135156	30.88413926	31,98917502
29 54854852	28 77385824	30.87115518	32,11796418
29 87023812	27 45694084	29 4767399	31 0357235
23.83875616	27.13691001	29.65249024	29 92760734
23.03075010	28.011090/18	29.05249024	30 6950/01/
23.40037130	20.01107040	20.00205050	27 32002602
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100 4164222	45		
20 67201578	76 11555876	29 50524269	20.02022416
29.0/2013/8	20.41555820	28.30324308	29.95025410
20.89048918	20.20057092	29.77409740	29.27575514
27.51098212	23.35940628	27.73017768	29.8/352526
23.93672906	23.09819526	27.94829558	29.11346052
22.03939018	26.90578842	28.60467338	25.22316246
21.42739336	21.35714682	25.24629404	25.95150148
19.3326126	19.96550522	21.64152298	
2275 265	0		
98.93516996			
51.52176742	49.21428452	50.76236496	49.38575362
48.88019104	49.23531266	50.61433334	49.36290664
50.31932298	46.08536074	51.42882308	49.4229492
50.92274398	45.73157654	52.48243558	48.54239732
42.85022354	50.2087555	48.30945514	46.24342498
38.42465594	47.09502638	47.69607544	41.99709398
19.15761566	22.83422852	25.63332102	

2275 265	45		
98.53293456			
49.338488	49.26488418	50.10499344	49.98358612
51.07362212	47.37760314	52.17178114	49.77518692
49.5806618	47.7857712	50.11046524	50.6676811
46.00337676	48.51928712	51,20819552	49.48350982
45 03485108	50 19804764	50 2034294	47 94532014
40 21110228	12 113/788/	/9 1099319/	13 3798/618
10 14161706	12.11347004	25 6095290	-5.5770-010
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45.66292114	44.4619667	46.02790068	47.06360014
41.8697502	45.01825792	47.16648484	45.427948
41.13500598	45.99850846	46.4579643	43.97706374
36.88848192	36.26695482	45.13731002	38.98748628
19.13657722	22.81351854	25.60926932	
1822 265	45		
98.54974824			
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40.36697006	42.1230606	40.6655464	43.69579698
39.12290878	41.32575532	39.48177186	42.29679184
40 51795654	38 9880913	38.06063768	43 35520782
41.0656563	39 8058/792	<i>AA</i> 13357772	37 30011594
41.0050505 32 17085114	36.0378525	34 38277056	40.00833436
10 16070799	20.9378323	25 6120021	40.00855450
19.100/9/00	22.79906216	25.0180051	
1412 205	0		
98.865/8064	10.00.000	11 010 51 50 5	20.2455025
38.85801544	40.3366/298	41.21251526	39.3657837
41.02693712	39.53769534	42.59932708	38.1136116
38.99116362	39.99701918	41.10022506	41.65617904
38.27532042	40.0788841	42.83440324	37.3191269
37.29112626	40.71051256	40.61609422	37.57031404
33.90524674	35.1454651	41.77336274	31.52881432
19.30854414	22.29714164	24.7952156	
1412 265	45		
98.84623718			
40.84026108	40.28384552	40.93802412	38.18530426
40.0435661	40.25686796	40.24929352	38.56129454
40.50852966	38.9292061	42.20601348	37.72995224
41 57413102	38 55087358	42 70633698	37 83195952
37 9129791/	12 59278488	36 / 5733796	38 7/069826
37.71277714	38 0/508132	38 72758042	32,00665246
10 24226056	22 20565499	30.727309 4 2 34.91909053	52.00005240
19.54550050	22.30303466	24.01000000	
1052 205	0		
98.09759826	00 1550 (100	0.6 40771 60.6	22 502 40 60 4
36.20451658	33.15526428	36.407/1636	33.58348694
32.71285326	35.2932205	36.85055848	33.38526384
34.7976776	33.23992082	36.67690506	34.27435608
35.24885482	28.84222374	40.1488388	33.02709502
31.88615686	37.90348968	33.2546242	30.61848222
25.40261496	32.41366576	37.13974838	29.28974496
19.3337341	21.68404466	23.84773864	
1052 265	45		
97.96114348			
35.79308546	32.31847306	35.40328292	35.34646988
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35.32076034	32.20355608	38.20213088	34.305616
33.40152894	30.65407182	35.30542906	36.50457916
32.60639648	32.333667	37.0570244	34.15605394
29.6860058	37.62654956	37.27133942	32.78776094
26.0327778	31.92216798	35.86511306	28.8260361
19.38240816	21.70701676	23.86766778	
775 265	0		
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31.82552604	28.38801652	30.5000683	31.62730332
31.06622048	27.57356606	33.3532692	30.88825838
29.44453966	26.32150536	30.79584962	32.26807864
27.96599694	28.06433906	32.06499098	30.6549839
25.32743416	32.47447206	33.15893628	28.53027764
23.201284	26.99083404	30.97368928	26.04771766
19.22278254	20.92047804	22.90697594	
775 265	45		
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31.92369768	28.91629524	31.81234208	30.4450676
27.01779098	30.30033952	31.98719826	31.88331568
28.97728652	27.63662642	34.2344177	30.6048332
32.50058058	26.23759192	31.85536194	29.03033294
27.94880486	31.50609702	30.267054	24.08791198
23.96117554	31.70822486	31.33988838	26.99343032
19.26017456	20.96857226	22.96351282	
409 265	0		
112.4658508			
24.88429414	23.86322706	24.40131724	23.95190508
22.69894904	24.37530212	24.39242288	24.7513981
23.15618592	22.77808152	25.97793732	24.04668348
25.1021969	21.79917872	24.2587055	23.52177584
22.74705086	24.2143673	23.62438622	21.05283624
20.57918664	24.04594078	24.72375946	21.10760954
18.73377798	19.5138748	21.00809098	
409 265	45		
109.804715			
24.10503808	25.07635842	24.41656036	23.32913706
24.43660392	24.4563286	24.6548073	22.89769706
23.58973198	26.3138546	23.8388481	22.91086046
24.27376138	24.76650392	23.57692644	22.27217254
25.59636114	23.14626388	21.51476516	22.94084472
23.4987427	22.99370384	22.18857652	20.10545082
18.7401596	19.53059462	21.00464324	
-1 -1	-1		

Appendix B: θ vs. y for First Heights

First Height Parabolic Fit for Raschig Rings

In the 4inch Column



First Height Parabolic Fit for 4-hole cylinders

In the 4inch Column



First Height Parabolic Fit for Monoliths

In the 4inch Column



First Height Parabolic Fit for Raschig Rings

In the 2inch Column



First Height Parabolic Fit for 4-hole cylinders

In the 2inch Column



First Height Parabolic Fit for Monoliths

In the 2inch Column



Appendix C: θ vs. y by Reynolds Number


















































































Appendix D: k_r/k_f and Nu_w Height by Height Analysis

























Appendix E: k_r/k_f, Nu_w, Bi, and Pe_r for all Packings and Columns

4 inch column Spheres













4 inch column Spheres (Van Dongeren)









4 inch column Spheres (Pollica)





4 inch column Raschig Rings







128







4 inch column JM 4-hole cylinders









4 inch column Monoliths













2 inch column Raschig Rings








2 inch column 4-hole cylinders









2 inch column Monoliths











Appendix F: F-test values

Column	Packing	Re	F/F95
4 Inch Column	Spheres	75	0.384578
		145	1.008367
		195	0.671051
		260	0.53057
		335	0.307174
		425	0.210865
	Raschig	85	0.42928
	Rings	142	0.338531
		190	0.377891
		258	0.331282
		341	0.386067
		432	0.424467
	4-hole	102	0.806354
	Cylinders	198	0.641854
		268	0.481158
		358	0.558235
		460	0.727119
		584	0.493383
	Monoliths	43	4.647282
		71	1.305055
		95	0.983003
		129	0.629038
		171	0.886185
		216	1.336643
2 Inch Column	Raschig Rings	350	3.912612
		490	2.507197
		676	3.358442
		909	5.43473
		1209	4.867726
		1570	3.886017
	4-hole	409	0.82748
	Cylinders	775	2.000006
		1052	0.668281
		1412	1.207582
		1822	1.15873
		2275	1.615775
	Monoliths	170	2.37015
		323	3.674283
		439	10.36014
		589	2.966539
		760	0.986552
		948	1.847035