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INVESTIGATION OF HEAT AND MASS TRANSFER FOR THE CATALYTIC COMBUSTION OF HYDROGEN

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With recent emphasis on environmentally friendly energy processes, and with increasingly strict requirements on NOₓ emissions and the release of unburned hydrocarbons from combustion processes, catalytic combustion has become increasingly more researched. By using a catalyst with combustion processes the same amount of energy can be released from fuels but at temperatures lower than the adiabatic flame temperatures. This reduces the amount of NOₓ emissions because NOₓ may be formed from the leftover products of combustion at the higher standard combustion temperatures. To date, however, not much research has been published about the heat transfer effects which occur during catalytic combustion. The focus of this thesis is to present research to determine the rates of heat transfer which occur during the catalytic combustion of hydrogen and determine the enhancement in heat flux from the catalyst surface reactions.
CHAPTER 1
INTRODUCTION

In this chapter catalyst structures and catalytic combustion is introduced. Classical convection heat transfer was discussed as well as heterogeneous heat transfer that occurs during a catalytic surface reaction. Some of the past works in the field of catalytic combustion and heat transfer effects are discussed. The use of a mass spectrometer as a tool for determining the chemical species concentrations and mass flow rates is introduced. Then at the end of this chapter the previous work with the annular catalytic combustion analysis is discussed and what was done different in the present study to build upon the previous understanding.

Catalytic Combustion

Catalyst structures have been widely explored for a use in combustion processes. One such area is in catalytic combustion to reduce pollutants such as CO, NO\textsubscript{x}, and unburned hydrocarbons, which are released into the atmosphere under normal combustion procedures. Very high temperatures are required to reach the flammability limits of fuels under the normal combustion procedures. At these high temperatures nitrous oxides are likely to combine in the products of combustion. It has been found that at combustion temperatures of 1800-2300 K nitrous oxides form readily in the combustion products [1]. Using a catalyst it is possible to work outside the flammability limits of a fuel. With reaction temperatures maintained below the flammability limit of the fuel, reaction conditions can usually be controlled more precisely. This ability to work with lower temperatures can minimize the production of nitrogen oxides.

Turbine operators have to comply with increasingly strict exhaust emissions regulations. The types of exhaust of the most concern are oxides of nitrogen (NO\textsubscript{x}).
Nitrous oxides are a major form of air pollutants that cause smog formation, acid rain and are a precursor to ozone [2, 3]. NO\textsubscript{x} emissions have been reduced in gas turbines using lean premixed combustion of fuels. Catalytic combustion has the potential to provide the needed reduction of NO\textsubscript{x} emissions to very low single digit levels in ppm [4]. Also allowing better temperature control and kinetic reaction rates, catalytic combustion can be used in power generating turbines, where the turbines are very sensitive to high temperatures which can damage machinery. The use of a catalytic reactor within the combustion system allows the combustor flame temperature to be maintained at lower levels than just using the premixed combustors.

Combustion temperatures greater than about 1200\degree C are required for gas-phase reactions to complete the burnout of fuel and CO in a reasonable residence time (on the order of 10 ms) [4]. Therefore a pre-burn is required where only a portion of the fuel can react with the catalyst. The challenge is to limit the reaction within the catalyst bed to keep excessive heat from damaging the catalyst or the support, while still releasing sufficient heat so that the downstream gas-phase combustion is stabilized. This creates ultra low NO\textsubscript{x} emission conditions. Understanding heat transfer in these reactions becomes very important to understand to maintain thermal stability. Catalytic combustion can be done using fuel lean or fuel rich conditions, under different processes. One way is to create a preburn using lean premixed fuel, so the degree of reaction can be limited by the chemical reaction rate upon the catalyst. Another is by channeling the fuel so that only a limited amount of fuel contacts the catalyst surface to ensure gas-phase reactions do not occur within the catalyst bed [4]. Fuel rich catalyst systems can also be used, where an insufficient amount of oxygen exists to fully oxidize all fuel in the
catalyst bed. In this scenario the extent of reaction is limited even if gas-phase reactions occur. Then additional air can be introduced downstream of the catalyst where a fuel lean complete combustion can occur. For a fuel lean mixture complete combustion occurs over the catalyst. However in a fuel rich mixture the reaction over the catalyst produces both total and partial oxidation products. These reaction products are then mixed with excess air and burned in a homogeneous process to form complete combustion products [3].

Steinfeld et al. [5] defines a catalyst as “a chemical substance which increases the rate of a chemical reaction without itself being consumed in the reaction”. In this paper heterogeneous catalysis is discussed and defined as “a catalytic process which occurs at the surface of a solid particle and is in contact with the gaseous or liquid solution” [5]. The catalyst rod structure consists of a support, wash coat, and a catalyst. The support can act to thermally stabilize the catalyst as well as provide the structure the catalyst is added to. The wash coat is a porous material used to create a rough irregular surface, increasing the amount of surface area and therefore the effectiveness of the catalyst. The thermal stability of the catalyst is strongly dependent on the support. Noble metal based catalysts such as PdO and Pt produce outstanding activity for methane combustion processes. However they are particularly sensitive to operation at temperatures higher than about 850°C [6]. So understanding the heat transfer in catalytic combustion reactions is important to maintain the appropriate catalyst surface temperatures.

**Heat Transfer Effects**

Uncontrolled catalytic activity can result in extremely high surface temperatures. When a catalyst surface temperature becomes too high there can be negative results.
Effects such as sintering or catalyst decomposition are of particular importance. One of the most researched effects of an overheated catalyst is with PdO catalysts and its decomposition to Pd, a less active form of the catalyst. Forzatti found that at temperatures above $800^\circ C$, PdO-Pd decomposition occurs and is complete at approximately $900^\circ C$ [8]. Palladium based catalysts are most widely used for catalytic methane combustion. Pd-based catalysts offer acceptable activity, light-off temperature, and resistance to volatilization for methane oxidation under fuel lean conditions. Sintering is another problem which can occur at elevated surface temperatures to greatly reduce the effectiveness of the catalyst surface and the duration of the catalyst life. Catalyst sintering degrades the catalyst performance because of the reduction of active surface area. The sintering of catalysts and their supports is a process which is complicated and generally unavoidable because it is accelerated at higher temperatures. Therefore the heat generated from catalytic combustion results in some sintering of the catalysts and their supports [1]. The thermal stability of the catalyst coating can be strongly dependent on the support. Thermal deactivation of a catalyst can result from sintering of the catalyst, support, or both. When the support sinters, a phase transformation can significantly reduce surface area as well as cause pore closure and encapsulation of active catalyst sites.

In non-reacting fluid flows heat transfer occurs through classical convection, which is conduction taking place in the thermal boundary layer. The thermal boundary layer is the region starting at the hot surface. There, due to the no slip condition the velocity is zero and therefore the fluid particles are at a thermal equilibrium with the surface. The thermal boundary layer continues radially from the hot surface until the fluid temperature
nears the temperature of the free stream flow [9]. The fluid velocity boundary layer is similar. Due to the no slip condition at the surface of the catalyst, which states the fluid velocity must near zero at a surface. This effect continues radially from the surface as the shear stress of the fluid slows the next layer of fluid. This trend continues until the velocity eventually reaches the free stream velocity, marking the end of the fluid velocity boundary layer. Due to the lack of fluid motion at the surface of the hot surface, the heat transfer at the surface is best predicted by Fourier’s law of conduction.

However in reacting flows Fourier’s law of conduction cannot be used to effectively predict the fluid particle heat transfer near the surface. In catalytic combustion a heterogeneous chemically reacting flow occurs, instead of the homogenous flow described above. For the case of catalytic combustion there is a bulk flow from the surface as well as a classical convective heat transfer between the hot catalytic surface and the fluid free stream. This bulk flow driven by heterogeneous chemistry may work to supplement the traditional convection heat transfer from the surface. In the early 60’s Rothenberg and Smith suggested that the heat transfer rate could be looked at as a linear combination of the classical convection plus the product of the reaction rate and the enthalpy of reaction [10]. In heterogeneous chemically reacting flow products adhere to the catalyst surface, with a lower enthalpy than the products released from the surface. This creates a bulk motion between particle adsorption and desorption at the catalyst surface. Due to this motion conduction at the surface is no longer a valid assumption as the only means of heat transfer.

During heterogeneous catalytic combustion there are several transport mechanisms as well as reaction mechanisms involved. To begin oxygen and fuel are
transported to the catalyst surface from the gas stream. The catalyst surface is porous allowing for gas diffusion into these pores and adsorption at the surface. A reaction occurs at the active catalyst sites, simultaneously generating heat. The products then desorb from the surface, transferred back into the gas stream via external mass transfer and pore diffusion. In the case where the catalyst is very active, the limiting mechanism is the mass transfer of reactants to the catalyst surface. However if the reaction is slow, reaction kinetics become the rate determining factor. Reaction rate is dependent on temperature, so reaction kinetics are typically more important earlier in the reaction before the catalyst temperature rises [12].

There is a general pattern for the catalytic combustion of hydrocarbons, related to the increase of temperature during the process, as mentioned above. To begin, as temperature increases as oxidation begins, at a temperature relative to the hydrocarbon fuel and the material makeup of the catalyst. In the early stages at low temperatures the rate of reaction is controlled through kinetics. As temperatures continue to increase there is a section of exponential increase in the rate of conversion, until it reaches a point where the heat generated by combustion becomes much greater than the heat supplied. This begins the next region where the reaction is controlled by mass transfer. Here heat transfer is also important. Eventually all the reactants are depleted and the final combustion products are realized. Therefore at low temperatures (kinetically controlled reaction) the heterogeneous reactions are dominant, while at higher temperatures the homogeneous reactions become more important [1].

**Previous Research**

Itaya et al. preformed reacting and non-reacting experiments using a heated cylinder, with and without a platinum catalyst surface, in a cross flow duct [13]. Oxygen
was used as the reference gas for the non-reacting experiments while SO2 and CH4 fuels were mixed with oxygen for the reacting experiments. The heat transfer coefficient was calculated by controlling the heat input to the cylinder and measuring the temperature difference between the inlet gas and the surface stagnation point temperature of the cylinder. It was found that the heat transfer coefficient increase was proportional to the reaction rate, where the heat transfer enhancement due to reaction was most significant where the reaction rate was at a maximum. Also, it was found that whenever the cylinder surface temperature was high enough for sustained catalytic activity, the experimental heat transfer coefficient would increase. From the results of the carried out experiments an overall heat transfer coefficient increase of 4-16% was reported, for the SO2 fuel reaction. The convective heat transfer coefficient enhancement that was observed was related to the magnitude of the enthalpy generated on the surface by the exothermic reaction, corresponded to the enthalpy due to diffusion of the reactants in the catalyst surface boundary layer.

Models of many different levels of complexity have been developed for monolith reactors. The most complex involving 3D models with the use of coupled Navier-Stokes equations [14]. However these complex models may still be unsolvable for some applications or require many hours of computer time, which can be computationally expensive. So two-dimensional and one dimensional models have also been proposed to provide quicker estimates. Di Benedetto et al. proposed a 2D mathematical model for the combustion of propane in laminar flow, coupling mass and energy fluxes [15]. They also provide a detailed discussion of other previous models and their failure to predict heat transfer correlations when applied to conditions with heterogeneous
chemistry. Their model represented a non-reacting entrance region followed by a reactive surface region, so that entrance effects could be eliminated before flows enter into the reactive region. They proposed a new definition of the Nusselt number, where the reference temperature is the gas bulk temperature and instead of the traditional wall temperature an adiabatic temperature corresponding to the inlet mixture is used. This relation is similarly proposed for the Sherwood number with respect to the reactant concentrations. Later work by Di. Benedetto et al. shows that the ignition with a heterogeneous reaction which is responsible for the significant enhancement of mass, momentum and heat transfer is comparable to the enhancement observed in the entrance effects of a duct inlet [16]. They propose that the ignition point in a heterogeneous flow may need to be considered as a new type of entrance effect. The accuracy of one-dimensional heterogeneous heat transfer models rely on accurate evaluations of mass and heat fluxes between the bulk flow and surface reactions. So with better knowledge of heat and mass transfer effects better reduced models can be used to estimate heterogeneous heat transfer effects in catalytic combustion.

**Mass Spectrometer**

A mass spectrometer can be used to determine species concentration. A probe within the combustion zone can determine the amount of each chemical species present in the reaction. Mass spectrometry is a technique that measures the mass-to-charge ratio of charged particles [17]. The mass spectrometer contains magnetic and electric fields. These fields exert forces on ions traveling through the analyzer part of the mass spectrometer. The electric field changes the speed of the ion while the magnetic field alters the ions direction. Based on Newton’s second law of motion the lighter ions are deflected more by the electromagnetic forces than the heavier ions. The
mass-to-charge ratio is determined by the magnitude of the deflection of an ion’s moving trajectory. A quadrupole ion trap mass spectrometer first ionizes the particles to form charged particles and the particles are subjected to RF forces while “trapped”. The mass spectrometer then detects and sorts each species to determine the species concentrations.

**Previous Work**

The apparatus used in the experiments described in this paper was very similar to and was modified from the apparatus used by Henry [18, 19]. In this work a concentric tube reactor was used to study heat and mass transfer under catalytic combustion of methane. Thermocouples were used to measure the reacting flow temperatures, initially before the start of the catalyst surface and after combustion before exiting the reactor. Hydrogen and methane concentrations were also measured at the end of the catalytic reaction using *in situ* Ramen spectroscopy. A Langmuir-Hinshelwood surface mechanism was assumed, and using determined local species as well as energy conservation the species of all the reactants and products were calculated along the axial distance of the catalyst surface. With known species concentrations the total heat release of the heterogeneous reaction at each axial location was determined, as well as the heat flux to the annular flow. A considerable (greater than a factor of two) enhancement in the heat transfer flux was found from the catalytically reactive surface compared to conventional solid surface non-reacting conditions. However a lot of approximations and calculations had to be made to get to these results. Experimental temperature measurements in the annular flow would allow for more accurate calculations of the heat fluxes. Also a mass spectrometer probe in the reacting annular flow would allow for more accurate species concentrations. This was done along the
length of the catalyst surface in the experiments described next in this paper is the current work.
CHAPTER 2
EXPERIMENTAL METHODS

In this chapter the specifications for the annular catalytic reactor used for analysis was described as well as the experimental procedures used with thermocouple and mass spectrometer data collection.

Experimental Apparatus

A catalytic combustion reactor, shown schematically in Figure 2-1, was designed for catalytic combustion to take place in an annular region for the purpose of experimental data collection. The outside of the reactor was built from stainless steel housing with an inner diameter of 6.6 mm and insulated around the outside. Located concentrically inside the stainless steel housing was the catalyst rod, which had a hollowed inside and a catalyst surface on the outside. The catalyst rod had an inner and outer diameter of 4.24mm and 4.85mm, respectively, with a wall thickness of 304 µm. The catalyst rod was attached to a stainless steel holder by a solder joint. This holder was fitted into a 4-way-union Swagelok cross, which centered the catalyst rod in the stainless steel housing, creating the annular catalytic combustion region as well as the non-reacting cooling flow region through the inside of the catalyst rod. This region is shown in Figure 2-2 and Figure 2-3. The catalyst used for data collection experiments was a proprietary mixture of platinum and palladium coated onto a Haynes 230 alloy substrate. During ignition experiments, nitrogen flowed through the inside of the catalyst rod, providing a non-reacting cooling flow for the catalyst rod, while a mixture of hydrogen fuel and air flowed over the catalyst surface in the annular region. The catalytic reaction took place on the outer surface of the catalyst rod.
The air for the annular flow was preheated by a 200W T-type process heater regulated by an Omega CN132, PID temperature controller using thermocouple TC1, as shown in Figure 2-1. The hydrogen fuel for the reacting flow entered at 300 K (room temperature) and mixed with the preheated air before entering into the annular reactor region at the Swagelok cross. Once in the annular region, the temperature of the reacting mixture was measured by a type K thermocouple (TC3) probe sliding through the annular space and recorded using an Omega HH-26K digital thermometer.

A thin disk (6.35mm) was press fit inside the reactor housing 17 cm from the leading edge of the catalyst. This alignment disk had a hole in the center for the catalyst rod to fit through and remain centered in the housing as well as 13 small evenly spaced holes to allow the reacting flow to pass through, with one of the holes large enough for a thermocouple to slide through. This allowed for temperature measurements of the reacting flow along the length of the catalyst surface in the reactor.

The cooling flow for all experiments described in this paper was heated to represent the adiabatic compressed air stream of the actual combustion air in the application of catalytic combustion in gas turbine engines because this process is the most practical means for active cooling in these scenarios [18]. The cooling flow was preheated by a 700W T-type process heater, regulated by an Omega CN 132, PID temperature controller using thermocouple TC2, as shown in Figure 2-1. The cooling gas then flowed through a 25 cm section of packed (1/8 inch) brass beads to improve thermal inertia and provide stable temperature readings downstream. Then the cooling flow was directed inside the catalyst rod to provide cooling to the inner surface of the
catalyst substrate. The temperature of the cooling flow was measured by a type K thermocouple (TC4) probe sliding through the inside of the catalyst rod and recorded using an Omega HH-26K digital thermometer. A z shape was bent into the thermocouple just before the exposed tip to ensure the thermocouple tip was centered inside the catalyst rod. Thermocouple 5 (TC5 in Figure 2-1) was used to measure the exit temperature of the flow leaving the reactor. This flow was comprised of a mixture of the gasses from the inner and annular spaces. Pictures of the actual apparatus are shown in Figures 2-4 through 2-9.

The gas flow rates were all controlled with digital mass flow controllers having an accuracy of 1% of full scale. Each meter was appropriately sized for the range of flow rates for each of the gasses. The nitrogen cooling flow was metered to a volumetric flow rate of 30 lpm. The combustion air for the reacting flow was metered to 5 lpm, and the hydrogen fuel for the reacting flow was metered to 0.5 lpm. All the volumetric flow rates are were referenced to 1 atm pressure and 298 K temperature. For the no-fuel experiments, air was used as the cooling flow and also metered to 30 lpm. Then either air or argon flowed through the annular space. When air flowed through the annular space, the air was metered to 5 lpm or 10 lpm for different data collection experiments. When argon flowed through the annular space, the argon was metered to either 6.35 lpm or 10 lpm.

**Experimental Procedures**

**Thermocouple Measurement**

Before igniting the reactor, the heaters for both the inner and annular flows had to reach the steady state temperature of 673K. For this initial heating, 30 lpm of air flowed through the inner section, while 5 lpm of air flowed through the annular section.
Approximately 1.5 hours later, both air flows reached a steady state temperature of 673K. Once this happened, the inner flow was switched from air to nitrogen, still metered to 30 lpm. After about 15 to 20 minutes, the inner nitrogen cooling flow was back to a steady state temperature of 673K. Then 0.5 lpm of hydrogen fuel at room temperature (300K) was released into the annular section of the reactor, causing ignition in only a few seconds. At the start of the catalyst surface in the annular flow, the temperature jump immediately after ignition was around 45 degrees Celsius. Then about 20 minutes later, the annular flow reached thermal equilibrium and a steady state temperature that was approximately 75 degrees Celsius higher than the temperature of just the preheated air in the annular section. Once this steady state temperature was achieved, temperature data measurements could be recorded.

Once the temperatures reached steady state at 723K, thermocouple 4 was pushed all the way down the reactor almost to the solder joint 2.5 cm before the start of the catalyst surface. Temperature measurements were recorded every half cm for 19.5 cm up to 17 cm from the start of the catalyst surface. Then the exit temperature of the gasses leaving the reactor was recorded from Thermocouple 5. These measurements were taken twice. Then the thermocouple was rotated 180 degrees and pushed back down the inside of the catalyst rod. Once again, temperature measurements were recorded every 0.5 cm from –2.5 cm before the start of the catalyst surface to 17 cm away from the beginning of the catalyst surface. (The actual start of the catalyst coating on the rod was located at 0 cm and is presented that way in the data and results in the next section.) The exit temperature was recorded using TC5. This same procedure was followed for collecting the temperature measurements in the annular section, except
thermocouple 3 was used and pushed into the annular region instead of the inner region. Table 2-1 shows the flow rates of each of the gases used in ignition experiments.

**Mass Spectrometer**

The same ignition procedure was followed for the mass spec experiments as with the thermocouple experiments. Once both heaters reached a steady state temperature of 673K 30.0 lpm of nitrogen was turned on to flow through the inner section of the annulus, then 0.5 lpm of hydrogen was released into the annular combustion region of the reactor.

The mass spectrometer probe was inserted into thin stainless steel tubing. This tubing was inserted into the reactor in the annular combustion region similarly to TC3. The end of this tubing was pushed into the reactor 2.5 cm before the start of the catalyst surface, just before the Swagelok cross solder joint in the reactor, as TC3 was for the temperature experiments. Once the output from the mass spectrometer showed a steady state concentration of hydrogen, nitrogen, and oxygen the data collection procedure began. The mass spec was given one minute to collect data at every half cm data point. After that minute the tubing was pulled a half cm out of the reactor. One minute of time passed to allow the mass spec to adjust to the new spot in the reactor then one more minute of time for the mass spec to collect data. This was repeated for every half cm down the reactor until the end of the tubing where the mass spec probe was pulling samples from was 17 cm beyond the start of the catalyst surface. This entire mass spectrometer ignition procedure was repeated on five different days to ensure accurate data collection.
Table 2-1. Ignition experiment flow rates for temperature data collection and mass spectrometer data collection.

<table>
<thead>
<tr>
<th>Gas</th>
<th>Flow rate (lpm)</th>
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<tbody>
<tr>
<td>Inner air/nitrogen</td>
<td>30.0</td>
</tr>
<tr>
<td>Annular air</td>
<td>5.0</td>
</tr>
<tr>
<td>Annular hydrogen</td>
<td>0.5</td>
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Figure 2-7. Picture of Omega CN 132, PID temperature controller.
Figure 2-8. Close-Up picture of thermocouple entered into the reactor. The tubing for the mass spectrometer probe entered the same way.
Figure 2-9. Flow control to switch between air (red) to nitrogen (green) for the inner “cooling” flow during the ignition experiments.
CHAPTER 3
RESULTS AND DISCUSSION

In this chapter the results from the experimental data collection are shown and presented in an analysis of the different heat transfer which occurred during the catalytic reaction. The implications of this heat transfer and temperature profiles is discussed. A model is presented to predict the increase in heat flux expected from the catalytic combustion. Then conclusions were drawn and suggestions for future work are made.

**Inner Nitrogen Cooling Flow**

The nitrogen inner bulk flow temperatures were measured by thermocouple 4 every 0.5 cm, beginning 2.5 cm before the corresponding catalyst surface on the other side of the catalyst tube until 19.5 cm downstream. This temperature data was recorded while catalytic combustion of hydrogen took place in the annular region. Temperature data was collected from the thermocouple in two orientations ensuring the most accurate measurements of the inner flow temperature, as described in the previous section. The temperatures from both sides of the thermocouple averaged after all the different experimental runs, as well as the exit temperature are presented in Figure 3-1, including error bars which represent the variance in the data collected between each different experimental run. In the Figure, 0 cm represents the start of the catalyst coating on the annular side of the catalyst rod in the reacting flow. Once the reacting flow reaches the start of the catalyst surface the inner flow begins pulling more heat from the annular region. A 3rd order polynomial was used to interpolate the temperature data averaged from all data sets of both thermocouple orientations to 1-mm intervals from the beginning of the catalyst surface (0 cm) to 17 cm downstream, shown in Figure 3-2. These bulk flow temperatures represent the average temperature
on each 1 mm interval and were used for the rest of the calculations and analysis to follow.

The heat flux to the inner nitrogen flow could then be calculated using classical heat transfer correlations. The use of the conservation of energy in the form of Equation 3-1 below with the known temperature differences for each 1 mm interval the heat flux into the inner flow was calculated.

\[ q''(P\Delta x) = \dot{m}c_p\Delta T \]  

In the above form of the conservation of energy P represents the inner perimeter of the catalyst tube, \( \Delta x \) is the 1 mm axial interval, \( \dot{m} \) and \( c_p \) represent the mass flow rate and specific heat, respectively, of the nitrogen flow, and \( \Delta T \) is the temperature difference in the 1 mm axial interval calculated from the 3rd order polynomial shown in Figure 3-2. The mass flow rate was calculated based on the experimentally metered volumetric flow rate and the density of nitrogen at room temperature (i.e. the reference state). The temperature dependent specific heats were interpolated from tabulated data [9]. The calculated heat flux to the inner cooling nitrogen flow was plotted as a function of axial distance shown in Figure 3-3. As was expected the heat flux begins high as heat is pulled at a fast rate from the annular flow during reaction and then becomes low when the temperature becomes steady toward the end of the reactor.

The outer surface temperature of the catalyst surface was determined with the known heat flux into the inner flow. There are no surface reactions in the inner flow, so classical convection heat transfer models can be used to relate the heat flux data to the inner catalyst tube surface temperature. Then from the steady state heat conduction model the temperature of the actual reactive catalyst surface can be determined. First
The Reynolds number describing the inner nitrogen flow was calculated to be 5800, which for internal flow represents a transition flow between laminar and turbulent conditions [9]. To determine the Nusselt number for the cooling flow the Gnielinski correlation [20] was used:

\[ Nu_d = \frac{(f/8)(Re_d-1000)Pr}{1+12.7(f/8)^{1/2}(Pr^{2/3}-1)} \] (3-2)

In the above Nusselt correlation \( f \) is the friction factor and calculated from the correlation by Petukhov [21]:

\[ f = (0.790 \ln Re_d - 1.64)^{-2} \] (3-3)

Petukhov assumes a smooth inner pipe, which was deemed an appropriate assumption for the catalyst tube inner surface, having a clean and polished surface appearance. While the above Nusselt correlation was initially developed for turbulent flow it has also been found to be valid for transitioning flow, similar to the inner nitrogen flow. With a Prandlt number of 0.7 [9], the calculated Nusselt number was 18. Inlet regions are neglected because the inner flow travels further than ten diameters before reaching the region where the flow was analyzed and fully developed. Then from the relation below the heat transfer coefficient \( h \) was determined:

\[ h = \frac{Nu_d k}{D_{in}} \] (3-4)

Where \( D_{in} \) represents the inside diameter of the catalyst tube, and \( k \) represents the thermal conductivity of the nitrogen flow. However thermal conductivity values are temperature dependent and the bulk temperature of the nitrogen flow was continually increasing, so \( k \) values were interpolated from tabular data at each 1-mm temperature increment. Once the heat transfer coefficient was calculated for each temperature
interval the inner surface temperature of the catalyst tube was calculated using Newton’s law of cooling:

\[ T_{\text{sur}} = \frac{q^*}{h_{\text{conv}}} + T_{\infty} \]  

(3-5)

In the above equation the reference temperature \( T_{\infty} \) was the interpolated nitrogen bulk flow temperature from Figure 3-2. Once the inner temperature of the catalyst support was known the outer catalyst surface temperature (reacting surface temperature in annular region) could be calculated based on the conservation of energy from the steady state conduction heat transfer equation across a cylindrical shell at each axial location:

\[ \Delta T = \frac{q^* \ln(r_i/r_o) \delta}{k} \]  

(3-6)

To plug into the above equation, the heat flux was known (Figure 3-3 data, via conservation of energy), \( r_i \) and \( r_o \) were the measured values of the inner and outer radii, respectively, of the catalyst tube, with \( \delta \) the calculated thickness of the catalyst support (304 µm) and \( k \) the average thermal conductivity of Haynes 230 equal to 26 (W/m K). There was not much difference in temperature as was expected between the inner catalyst support temperature and the outer catalyst temperature. Subtracting delta \( T \) from the inner surface temperature the catalyst surface temperature was determined and plotted for each 1-mm axial location along the reactor in Figure3- 4. The catalyst surface temperature closely follows the trends of the heat flux into the inner flow seen in Figure 3-3. The hottest the catalyst surface heats up to is in the beginning when the hydrogen reaction began but the inner flow had not yet pulled much of the heat out of the annular region and into the inner flow area. At almost 900K the catalyst surface is not hot enough to undergo significant sintering.
Annular Reacting Flow

At 2.5 cm before the start of the catalyst coating and 17 cm after along the length of the catalyst surface at every half cm one minute of raw data was collected from the mass spectrometer. Concentrations of hydrogen, oxygen, nitrogen and water vapor were collected. When the mass spec ionizes the particles the water can get split into separate hydrogen and oxygen signals. To account for this water fractionation the following equation is used to determine the actual amount of hydrogen present neglecting any hydrogen that may have been introduced from the water vapor.

\[
(hydrogen) - (0.045 \times water) = actualhydrogen
\]  

(3-7)

The water signal was already so low that when the above correction was made there was negligible difference to the hydrogen concentration raw data.

Every minute of data which corresponded to a half cm along the reactor was averaged. Then the data from all five different experimental runs were averaged together. Once the raw data was all averaged it was made relative to its initial concentration. Every concentration data point was divided by the initial concentration, so that the data was relative to itself and unitless. This is shown for all four species in Figure 3-5. The water vapor signal was not a smooth useful signal. This was possibly due to the water vapor cooling and condensing before it could be measured by the mass spectrometer. For the rest of the analysis of this data only the data from the hydrogen concentration was used. It is also noted that the Reynolds number for the reacting annular flow was approximated as 2300, and considered transition flow between laminar and turbulent flows.

The initial mass flow rate of hydrogen into the reactor before any reactions began was known. The hydrogen was metered to a volumetric flow of 0.5 lpm. This value was
converted into a mass flow rate and multiplied by the relative hydrogen concentration signal from the mass spectrometer data, shown in Figure 3-6. The error in Figure 3-6 shows the variance between different experimental data collection runs. This data was then fit to a 4th order polynomial trendline, shown in Figure 3-7. From the use of the 4th order polynomial values were interpolated on a 1mm interval.

The amount of energy released on the catalytic surface from the hydrogen reaction was called $Q_{released}$. It is calculated from the conservation of energy in the form of Equation 3-8.

$$Q_{released} = \Delta \dot{m}_{H_2} h_{fg}$$  \hspace{1cm} (3-8)

The change in mass flow rates was known from the mass spectrometer experimental data collection (Figure 3-7). The lower heating value for hydrogen was determined from tabulated data to be equal to 121 MJ/Kg, and represented as $h_{fg}$. $Q_{released}$ was plotted in Figure 3-8. It is shown in Figure 3-8 that the energy released decreases to zero around 12 cm, where there is no longer any hydrogen reaction because all the hydrogen has already been burned as evident from Figure 3-7.

From the amount of heat released from the hydrogen reaction on the catalytic surface heat should move into the annular flow, into the inner flow and axially conducted through the catalyst rod. The amount of axial conduction at each 1 mm interval was calculated from the classical conduction Equation 3-9.

$$Q_{conduction} = kA_c \frac{dT}{dx}$$  \hspace{1cm} (3-9)

The conduction at each 1 mm control volume along the length of the catalyst surface was determined. A diagram of the conduction through the control volume is shown in Figure 3-9. In Equation 3-9 $k$ is the thermal conductivity of platinum (75 W/m K) which
has the highest thermal conductivity between platinum and palladium (catalyst coating) and Haynes 230 alloy (catalyst support). Ac is the cross sectional area of the catalyst rod. The derivative term was determined using a 3rd order trendline equation (Equation 3-10) taken from the temperature profile of the catalyst surface (shown in Figure 3-4). The derivative of this trendline equation with respect to x was calculated at each 1 mm interval. Then this slope is used as the delta T term in Equation 3-9, with delta x as the 1 mm interval.

\[ y = 13881x^2 + 3461.x^2 - 1967.x + 894.4 \]  

(3-10)

The \( Q_{\text{conduction}} \) heat rate was plotted in Figure 3-10. However this heat rate is significantly smaller than the heat released on the surface by the catalytic hydrogen reaction, the heat rate transferred into the inner region \( Q_{\text{inner}} \) and the other transfers of heat that will be discussed. Therefore the rate of heat that is conducted through the catalyst rod is neglected for the rest of the heat analysis.

\( Q_{\text{annular}} \) was then calculated from the subtraction of \( Q_{\text{inner}} \) from \( Q_{\text{released}} \), and plotted in Figure 3-11.

\[ Q_{\text{annular}} = Q_{\text{released}} - Q_{\text{inner}} \]  

(3-11)

Where \( Q_{\text{inner}} \) represents the rate of heat that was pulled out of the annular flow region into the inner flow region by the nitrogen “cooling” flow. \( Q_{\text{annular}} \) then represents the total rate of heat energy from the hydrogen reaction left in the annular region of the reactor. However this term also includes the rate of heat lost from the reaction to the insulated outer annular wall and into the surrounding air. Therefore \( Q_{\text{annular}} \) can be represented as the sensible annular heat plus the heat lost.

\[ Q_{\text{annular}} = Q_{\text{ann,sensible}} + Q_{\text{lost}} \]  

(3-12)
The sensible heat, $Q_{\text{ann}, \text{sensible}}$, represents the rate of heat determined by the bulk flow temperature in the annular region as well as the mass flow rates and specific heats of each species in the annular flow. 

$$Q_{\text{ann}, \text{sensible}} = \sum_i m_i c_{p_i} \Delta T$$  \hspace{1cm} (3-13)

The four species in the annular region were hydrogen, oxygen, nitrogen, and water vapor. The mass flow rates of hydrogen were known from the measured values shown in Figure 3-7. The mass flow rates of oxygen and water vapor were determined from the known hydrogen mass flow rates and the stoichiometric chemical equation for complete hydrogen combustion.

$$2H_2 + O_2 \rightarrow 2H_2O$$  \hspace{1cm} (3-14)

From this equation knowing the change in mass of hydrogen the new mass flow rates for each other species in the reaction was determined by using ratios of species mass to mole concentrations from the above chemical equation. For the new mass flow rate of $O_2$ Equation 3-15 below was used.

$$\left( \Delta m_{H_2} \right) \left( \frac{1 \text{ mol } H_2}{2 \text{ grams } H_2} \right) \left( \frac{1 \text{ mol } O_2}{2 \text{ mol } H_2} \right) \left( \frac{32 \text{ grams } O_2}{1 \text{ mol } O_2} \right)$$  \hspace{1cm} (3-15)

So the new $O_2$ mass flow rate was equal to the change in the hydrogen mass flow rate multiplied by a factor of 8. The initial mass flow rate of $H_2O$ was zero. To determine the new mass flow rate of water vapor Equation 3-16 below was used.

$$\left( \Delta m_{H_2} \right) \left( \frac{1 \text{ mol } H_2}{2 \text{ grams } H_2} \right) \left( \frac{1 \text{ mol } H_2O}{1 \text{ mol } H_2} \right) \left( \frac{18 \text{ grams } H_2O}{1 \text{ mol } H_2O} \right)$$  \hspace{1cm} (3-16)

So the new $H_2O$ mass flow rate was equal to the change in the hydrogen mass flow rate multiplied by a factor of 9. The mass flow rates of the reacting species were plotted in Figure 3-12. The nitrogen mass flow rate was constant and equal to 73% of the initial
mass flow rate of air. Now all the new mass flow rates could be plugged into Equation 3-13.

\[ Q_{\text{ann,sensible}} = [(\dot{m}c_p)_{H_2} + (\dot{m}c_p)_{O_2} + (\dot{m}c_p)_{N_2} + (\dot{m}c_p)_{H_2O}]\Delta T \] (3-17)

The specific heat values were iterated from tabulated data to correspond with the annular bulk flow temperature at each 1 mm location.

The annular bulk flow temperatures still need to be determined. There was a lot of error in the annular thermocouple temperature measurements, shown in Figure 3-13. The collected data shows that the inner “cooling” flow was hotter than the annular reactive flow, which is physically impossible. The heat should move from the annular flow into the inner flow. The thermocouple in the annular flow had a long length to travel where gravity must have pulled the exposed tip downward. This would cause the thermocouple to read lower temperatures further away from the reactive catalyst surface and possibly touching the outer insulated surface of the reactor. Due to the error in the measured temperatures of the annular flow they could not be used. However the annular bulk flow temperature was calculated through an iterative procedure described next.

The total amount of annular heat \( Q_{\text{annular}} \) is equal to the combination of the sensible heat, heat from radiation off of the hot catalyst rod, and the convective heat from the outer annular wall of the reactor.

\[ Q_{\text{annular}} = Q_{\text{ann,sensible}} + Q_{\text{radiation}} + Q_{\text{conv,outer}} \] (3-18)

A schematic of the different heat transfer rates were shown in Figure 3-14. Comparing Equation 3-18 with Equation 3-12, an equation to determine the radiation and outer convection terms was determined.
\[ Q_{\text{lost}} = Q_{\text{radiation}} + Q_{\text{conv,out}} \] (3-19)

The total heat rate lost in the reaction across the entire length of the catalyst reaction was determined, using experimental data from the previous work with the experimental reactor setup [19]. In the previous work the outer temperatures were measured and the rate of heat lost was calculated from a combination of free convection and radiation. The current experiments yielded similar temperatures to the previous experiments so a scale factor was determined comparing the differences between temperature and geometry, including differences in thickness of insulation. This scale factor multiplied by the previously calculated loss of 35 W predicted a rate of heat lost approximately equal to 20 Watts for the current experiments.

The \( Q_{\text{conv,out}} \) term was set equal to a thermal resistance represented as a constant multiplied by the delta T, corresponding to the temperature difference between the annular bulk flow temperature and the ambient air temperature outside of the reactor at 300 K. The thermal resistance circuit is shown in Figure 3-15, where the constant \( C \) below represents this thermal resistance.

\[ Q_{\text{conv,out}} = C(T_{\text{ann}} - 300) \] (3-20)

Initially annular temperatures needed to be guessed for this iterative method, so the experimentally measured values from Figure 3-13 were used as the initial guess. The thermal resistance values were unknown, so the constant in Equation 3-20 was a free parameter adjusted until the sum of the heat lost was equal to the calculated total heat rate lost of 20 W.
The convection heat loss out of the annular region could also be calculated from the convection heat transfer from the annular flow temperature to the outer annular surface.

\[ Q_{\text{conv,out}} = h \cdot A_{\text{out,sur}} (T_{\text{ann}} - T_{\text{out,ann,sur}}) \] (3-21)

The outer annular convective heat transfer coefficient was calculated from Equation 3-22. Nusselt number was determined to be 5.30 from correlations in table 8.3 [9] for fully developed laminar flow in a circular tube annulus with uniform heat flux maintained at both surfaces. The thermal conductivity (k) of the annular gas was determined as 0.05 (W/m K) from a mass average of the annular flow, which is dominated by nitrogen.

Then the hydraulic diameter for an annulus simplifies to the inner diameter of the annulus (catalyst rod) subtracted from the outer diameter of the annulus. The annular outer convective heat transfer coefficient was calculated as 112 W/m²K.

\[ h = \frac{N_u \cdot k}{D_h} \] (3-22)

With the known \(Q_{\text{conv,out}}\) from Equation 3-20, annular heat transfer coefficient above and a known area for the outer annular surface the temperature of the outer annular surface was calculated from Equation 3-21.

Next the annular heat loss from radiation was determined, where \(T_{\text{sur}}\) is the catalyst rod surface temperature from Figure 3-4, and \(T_{\text{out,ann,sur}}\) is the surface temperature of the outer annular wall, calculated from Equation 3-21.

\[ Q_{\text{radiation}} = C(T_{\text{sur}}^4 - T_{\text{out,ann,sur}}^4) \] (3-23)

The constant was determined by Equation 3-24, a correlation for radiation heat transfer between long concentric cylinders [9]. The emissivity (\(\varepsilon\)) was estimated to be 0.75 from tabulated data correlated to heavily oxidized stainless steel [9]. \(\sigma\) represents the Stefan-
Boltzmann constant, $A_I$ was the area of the catalyst surface, and the radii were the catalyst inner annular radius and outer annular radius.

$$C = \frac{\sigma A_I}{\frac{1}{2} \pi (r_I/r_O)} = 5.3E - 13$$  \hspace{1cm} (3-24)

From Equation 3-19 $Q_{lost}$ was now calculated for each 1 mm interval. Then by plugging Equation 3-17 into Equation 3-12 for the sensible annular heat, the delta $T$ for each 1 mm interval in the annular region was determined and the annular temperatures were calculated.

$$T_{ann} = \frac{Q_{\text{annular}} - Q_{\text{loss}}}{m c_p} = \frac{Q_{\text{ann,sensible}}}{m c_p}$$  \hspace{1cm} (3-25)

Then the annular temperatures from Equation 3-25 were used as the new initial guess annular temperature values and iterated through Equations (3-20, 3-22, 3-24, and 3-25) four times until the initial annular temperatures are the same as the annular temperatures calculated from Equation 3-25.

All of the rates of heat transfer shown in Figure 3-14 were now calculated. The radiation heat transfer loss was plotted in Figure 3-16. The annular convection heat loss from the outer annular surface was plotted in Figure 3-17. Then a plot of the radiation heat loss, convective heat loss, and the total heat loss along the length of the catalyst surface was plotted in Figure 3-18. The heat loss through radiation accounts for the majority of the heat loss until the annular flow temperature becomes hotter than the catalyst surface and the convection loss became the main contributor to overall heat loss. Also when the catalytic hydrogen reaction stops the heat loss begins to slowly increase. Figure 3-19 shows a plot with the rate of heat released by the hydrogen reaction on the catalyst surface as well as the rate of heat transferred into the inner and annular region and then the total heat lost from the annular region. Then a plot was
created to show the heat transfer in the annular region with the rates of heat transfer into the annular region, lost from the annular region and the annular sensible energy in Figure 3-20.

The annular flow temperature, inner flow cooling temperature, and catalyst surface temperature were plotted in Figure 3-21. In this Figure the temperature of the catalyst rod continually decreases, while the inner flow temperature continually increases. They both reach an almost steady value after 12 cm when no more heat was released on the catalytic surface due to hydrogen reaction. The annular temperature however increases quickly and becomes hotter than the catalyst surface temperature, before it begins to cool, but remains hotter than the catalyst surface temperature. There was an error associated with the radiation heat loss due to the chosen value for the emissivity. To prove that this uncertainty was small and does not change any temperature profile trends, the three temperatures in Figure 3-21 were plotted again in Figure 3-22, but this time with a chosen emissivity of 0.60. Figure 3-22 was very similar to Figure3-21, with a maximum difference of about 30 K, adding further confidence in the calculation for the amount of heat loss due to radiation, and the plots from Figure 3-21. Before looking further into the physical significance of Figure 3-21 the true flux in the annular region should first be considered.

The true annular convection flux was calculated from the total annular heat release minus the radiation heat release.

\[ q_{\text{ann,conv}} = \frac{Q_{\text{annular}} - Q_{\text{radiation}}}{A_I} \]  

(3-26)

Then the true annular heat flux accounts for both the sensible convection and the outer convection values, plotted in Figure 3-23. This heat flux becomes negative at 10.8 cm,
which is before the hydrogen reactions stop at 12 cm. The classical annular heat flux was also calculated, from Newton’s law of cooling using the classical heat transfer convection coefficient and the change in temperature between the catalyst surface temperature and the annular flow temperature. The classical annular heat flux represents what the convection from the hot catalyst surface into the cooler annular flow should be.

\[ q_{\text{ann,conv}}(\text{classic}) = h(T_{\text{sur}} - T_{\text{ann}}) \]  

(3-27)

The classical heat transfer coefficient was determined from a Nusselt number equal to 5.6 from a correlation in table 8.3 [9]. The value for \( h \) was calculated similarly from Equation 3-22 and was equal to 118 W/m²K, with an estimated 10-20% uncertainty. This became a very good approximation when it was compared to the convective heat transfer coefficient determined from the true experimental flux calculated from Equation 3-26. After 12 cm in the annular flow there were no longer any reactions, so classical convection takes over. Therefore the heat transfer coefficient from the reactive flux calculation from 12 cm-15cm should equal the classical convection coefficient. When those values were averaged over that distance they equaled 100 W/m²K, which is in excellent agreement with the above calculation of 118 W/m²K. This adds accuracy to the estimated classical heat transfer coefficient from [9]. Figure 3-24 shows a plot of the classical annular heat flux, compared with the actual reactive annular heat flux. This shows that there is an enhancement in the annular heat flux due to the catalytic reaction. This enhancement in heat flux was calculated from the subtraction of the classical heat flux from the actual annular heat flux and was then plotted in Figure 3-25.
Figure 3-26 shows a plot of the annular flow temperature, the catalyst surface temperature and the cooling inner flow temperature, separated into four different regions along the length of the catalyst surface. In all four regions the inner flow was heating up from the hotter catalyst surface. The first region was from the beginning of the catalyst surface, where reactions began up to about 6.5 cm where the annular bulk flow reaches the same temperature as the catalyst surface. In this region the annular flow temperature increases very quickly from the combination of classical convection heat transfer and the enhanced heat flux from the catalytic reaction. This creates a positive net heat flux into the annular region. The second region is between where the annular temperature increases hotter than the catalyst surface temperature and where the annular flux becomes zero, at 10.8 cm. In this region the classical convective heat flux became negative (Figure 3-24) because the annular flow was hotter than the catalyst surface. The classic heat flux here changes direction pulling heat back toward the catalyst rod and into the inner cooling flow. However the annular flow still continued to heat up and reached the max temperature in this region. The enhanced heat flux in this region is still positive and greater than the negative classical convective heat flux, which drives a net positive heat flux into the annular flow. Region 3 was a small distance along the length of the catalyst between where the net annular flux became negative and the hydrogen combustion reaction ended at approximately 12 cm. In this region the classical heat transfer was still negative because the annular flow was hotter than the catalyst surface temperature. The enhanced heat transfer was still positive because the catalytic combustion process was still occurring. However in this region the classical convective heat flux was greater than the enhanced heat flux, creating a
negative net heat flux into the inner cooling region of the reactor. The fourth and last region was where the catalytic hydrogen reaction stopped after complete combustion. In this region the annular flow follows classical convection correlations. The enhanced heat flux was zero. The classical convection heat flux was still negative because the annular flow remains hotter than the catalyst surface temperature, so there was a negative net heat flux in this region as well. The inner cooling nitrogen flow almost heats up to the temperature of the catalyst rod. The inner flow was purely classical convection heat transfer so it never became hotter than the catalyst rod and reached a steady temperature just below that of the catalyst rod.

**Model of Enhanced Heat Flux**

The enhanced heat flux into the annular region was likely due to molecules desorbing from the catalyst surface with energy that the molecules carried with them into the annular flow. Molecules from the annular flow with a certain enthalpy adhere onto small cavities in the catalyst coating where the hydrogen combustion occurs, generating heat and then the products desorb from the catalyst surface with a higher enthalpy. This movement of molecules creates another form of heat transfer that was seen from the experimental analysis as the enhanced heat flux into the annular flow. This form of heat transfer would explain how the annular flow temperature could continue to heat up beyond the temperature of the catalyst surface while the catalytic hydrogen combustion reaction was still occurring. A molar flux model is proposed next to approximate the enhanced heat flux, noting the very preliminary nature.

The energy from the reactant molecules is equal to the sum of the product of the molar flux and the annular temperature for hydrogen and oxygen as well as an undetermined constant.
\[ E_{in}^* = C_1(M_{H_2} \cdot T_{ann} + M_{O_2} \cdot T_{ann}) \] (3-28)

From the stoichiometric combustion equation (Equation 3-14) there was half a mole of oxygen for every mole of hydrogen, so the equation reduced to:

\[ E_{in}^* = M_{H_2} \cdot T_{ann} + \frac{1}{2} M_{O_2} \cdot T_{ann} = \frac{3}{2} M_{H_2} \cdot T_{ann} \] (3-29)

The molar flux of hydrogen was related to the known mass flow rate of hydrogen by a constant.

\[ \dot{M}_{H_2} = C \cdot \dot{m}_{H_2} \] (3-30)

This reduces the molar energy of the reactants in the annular flow represented in Equation 3-29.

\[ E_{in}^* = \frac{3}{2} C \cdot \dot{m}_{H_2} \cdot T_{ann} \] (3-31)

The energy released from the products off the catalyst surface were similarly calculated, using the catalyst surface temperature and water as the product from Equation 3-14 with a one to one molar ratio of hydrogen to water. The equation for this energy was reduced to Equation 3-32, below.

\[ E_{out}^* = C \cdot \dot{m}_{H_2} \cdot T_{sur} \] (3-32)

In the proposed model the enhanced heat flux was set equal to the energy in minus the energy out, and normalized to the max value plotted in Figure 3-27 along with the calculated enhanced heat flux.

\section*{Conclusions}

From the experimentally measured inner flow temperatures and experimentally measured mass flow rates of the chemical species involved in the hydrogen reaction during the catalytic reaction in the annular flow, a physically accurate portrayal of the heat transfers was determined. Through the above analysis the rate of heat released
on the catalyst surface by the hydrogen combustion reaction was directly calculated from measured data. Then the amount of that heat transferred into the inner “cooling” flow was directly calculated from the measured temperature data. The amount of heat left to be transferred into the annular flow was then known. To determine how much heat was lost and how much remained in the annular region an iterative procedure was used to determine the temperature profile in the annular flow. From a physics based model the heat lost had to be a combination of radiation from the catalyst surface and convection heat losses from the outer annular surface. These values were determined from traditional heat transfer correlations combined with the known total quantity of heat lost from the reaction along the full length of the catalyst surface. With all the rates of heat transfer known an enhancement of heat flux into the annular region was shown that classical convection correlations could not account for. This enhancement in heat flux proves the products from the combustion reaction were leaving the catalyst surface back into the annular flow with a higher energy. This heat flux enhancement into the annular flow was then approximately modeled by a molar flux model based off of the stoichiometric hydrogen combustion.

**Future Work**

If this experiment is continued a new way to determine the annular reacting bulk flow temperatures is necessary for experimentally determined sensible heat calculations. With a thermocouple in the annular flow that could produce accurate temperature measurements the temperatures of the annular flow could be measured during non-reactive experiments without any fuel in the annular region. With this data the classical convective coefficient could be determined for the annular geometry with a
hot catalyst surface (non-reactive) heated by a hot inner flow. Then those values could also be compared with the reactive convective heat transfer coefficient and annular flux.

Also adding more insulation to the outer surface of the reactor to reduce the rate of heat lost to the ambient air in the room would be beneficial. Furthermore a way to keep the water vapor signal entering the mass spec from condensing on the way would allow better experimental data collection from the mass spec, to get experimental data from oxygen and water vapor. Then fewer calculations would have to be made to determine the species concentrations and mass flow rates, needed to determine the rates of heat transfer in the annular region.
Figure 3-1. Inner nitrogen bulk flow temperatures averaged from all data collection runs with error bars, representing the variance in data collection experiments. At 20 cm the exit temperature of the gases leaving the reactor is shown.
Figure 3-2. Inner nitrogen bulk flow temperatures averaged from all data collection runs with a third order trendline fit to the data.
Figure 3-3. Heat flux into the inner flow from the annular reacting region.
Figure 3-4. Surface temperature of the outer catalyst surface on the catalyst rod.
Figure 3-5. Data from the mass spectrometer averaged for each half cm along the length of the reactor and each species relative to itself.
Figure 3-6. The mass flow rate of hydrogen in the annular reacting flow along the length of the reactor is shown with error included. Zero cm corresponds to the beginning of the catalyst coating.
Figure 3-7. The hydrogen mass flow rate is shown along the length of the reactive catalyst surface, with the 4th order trendline.

\[ y = -6.34E-09x^4 - 1.09E-08x^3 + 7.51E-06x^2 - 1.34E-04x + 6.89E-04 \]
Figure 3-8. The rate of heat transfer of heat released by the hydrogen reaction along the length of the catalyst surface representative of the energy on each 1mm increment of area.

Figure 3-9. A diagram of the conduction through the control volume on the catalyst rod.
Figure 3-10. The $Q_{\text{conduction}}$ heat rate was plotted, representative of the energy on each 1mm increment of area. The values are an order of magnitude less than the rest of the contributing heat transfers, so the conduction heat transfer was neglected.

Figure 3-11. Rate of heat transfer for the energy on each 1mm increment of area, from the hydrogen reaction in the annular region minus the heat transferred into the inner nitrogen flow.
Figure 3-12. Species mass flow rates in the annular reacting region.

Figure 3-13. The bulk flow temperatures collected from the annular reacting region averaged over all data collection runs with error bars shown. These measured values for the annular region are less than the inner, which is physically impossible.
Figure 3-14. A representation of heat transfers in the reactor for each 1 mm interval along the catalyst surface. Heat is released from the hydrogen reaction on the catalyst surface. Then heat is conducted along the catalyst rod, (found to be negligible) and convection moves heat into the inner and annular flows. Then heat is lost from the annular region through radiation and convection out of the annular region.
Figure 3-15. The thermal resistance circuit is shown, equal to the constant multiplied by the temperature difference between the annular flow and the ambient air room temperature to calculate the outer convection heat transfer lost out of the annular region.

Figure 3-16. A plot of the rate of radiation heat loss from the catalyst surface to the outer annular surface, representative of the energy on each 1mm increment of area.
Figure 3-17. A plot of the rate of heat transfer on each 1mm increment of area, lost due to convection from the outer annular surface and into the ambient room air.
Figure 3-18. The rates of heat transfer on each 1mm increment of area, lost from the annular region of the reactor, with the total energy lost, plotted with the two types of heat loss which contribute to the total loss.
Figure 3-19. A plot of the total rates of heat transfer on each 1mm increment of area, from the heat released by the hydrogen reaction, the heat transferred into the inner and annular flows and the heat lost from the annular flow region.
Figure 3-20. A plot of the heat transferred into the annular region with the amount of heat transfer lost from the annular region and the sensible heat in the annular region also shown. The rates of heat transfer plotted were representative of the energy on each 1mm increment of area.
Figure 3-21. The annular flow temperature, inner flow cooling temperature, and catalyst surface temperature were plotted.
Figure 3-22. To prove that the uncertainty in the radiation heat loss calculation was small and does not change any temperature profile trends, the three temperatures in Figure 21 were plotted again in Figure 22, but this time with a chosen emissivity of 0.60.
Figure 3-23. The true annular heat flux accounts for both the sensible convection and the outer convection values.
Figure 3-24. A plot of the classical annular heat flux, compared with the actual reactive annular heat flux.

Figure 3-25. The amount of heat flux enhanced by the catalytic reaction.
Figure 3-26. The inner cooling temperature, catalyst surface temperature and the annular flow temperature were plotted and separated into regions along the length of the catalyst surface. R1 represents the region where the annular flow temperature was less than the catalyst surface temperature. R2 represents the region where the actual annular flux is positive. R3 represents the region where the annular flux is negative up to when the hydrogen reactions stop. R4 represents the region described by classical convection when there are no longer reactions occurring.
Figure 3-27. A plot of the enhanced heat flux into the annular flow and the molar flux model to predict the amount of $q^{"e}$enhanced.


BIOGRAPHICAL SKETCH

Julia Setlak was born in Melbourne, Florida on June 3rd 1987 to Dale Setlak and Kathleen Rettich. She graduated from Eau Gallie High School in Melbourne, Florida May 2005. Julia then began her undergraduate studies at the University of Florida, in Gainesville Florida. There she studied mechanical engineering. After completion of her B.S. degree in mechanical engineering she continued her education at the University of Florida in the graduate school for mechanical engineering specializing in thermal sciences and fluid dynamics. She began the program in 2010 under the direction of Professor David Hahn. The presented work is the culmination of her two years of research for a Master of Science degree in mechanical engineering.