**Energy Efficient Thermal Management for Natural
Gas Engine Aftertreatment via Active Flow Control**

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Abstract

The project is focused on the development of an energy efficient aftertreatment system capable of reducing NO_x and methane by 90% from lean-burn natural gas engines by applying active exhaust flow control. Compared to conventional passive flow-through reactors, the proposed scheme cuts supplemental energy by 50%-70%. The system consists of a Lean NO_x Trap (LNT) system and an oxidation catalyst. Through alternating flow control, a major amount of engine exhaust flows through a large portion of the LNT system in the absorption mode, while a small amount of exhaust goes through a small portion of the LNT system in the regeneration or desulfurization mode. By periodically reversing the exhaust gas flow through the oxidation catalyst, a higher temperature profile is maintained in the catalyst bed resulting in greater efficiency of the oxidation catalyst at lower exhaust temperatures. The project involves conceptual design, theoretical analysis, computer simulation, prototype fabrication, and empirical studies.

This report details the progress during the first twelve months of the project. The primary activities have been to develop the bench flow reactor system, develop the computer simulation and modeling of the reverse-flow oxidation catalyst, install the engine into the test cell, and begin design of the LNT system.

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Introduction

Lean burn natural gas engines have high thermal efficiency and durability. However, lean burn engine exhaust has a wide temperature span and is rich in oxygen, which makes aftertreatment difficult. Lean burn NO_x levels can be much lower than stoichiometric burn levels, but are still too high to meet the 0.1 g/hp-hr target and a de- NO_x scheme has to be applied. The leading de- NO_x contenders for lean burn engines are Lean NO_x Catalyst (LNC), Selective Catalytic Reduction (SCR), and Lean NO_x Trap (LNT) [1].

A Lean NO_x Trap, or NO_x adsorber, is a flow-through catalyst-bed that temporarily stores exhaust NO_x during typical lean burn operations. Before the NO_x adsorbent becomes fully saturated, the exhaust gas composition must be adjusted to produce a fuel-rich exhaust. This adjustment may be accomplished by either by modifying engine fueling strategy or by introducing fuel into the exhaust stream. Under fuel rich conditions, the stored NO_x is released from the adsorbent and subsequently reduced to N_2 over precious metal sites. A desulfurization process must be conducted at high temperature and fuel rich conditions [3-8]. Such Lean NO_x Traps have demonstrated high NO_x conversion efficiencies; approaching 95%, for lean burn gasoline engines and diesel engines [4-8].

The LNT is the focus of this research. It should be pointed out that the LNT approach has to overcome the dilemma that while the oxidation of methane requires high temperatures, the NO_x absorption requires much lower temperatures. Significantly, de- NO_x systems normally involve hydrocarbon slip. In the case of compressed natural gas (CNG) fuel, the predominant hydrocarbon is methane, which is difficult to destroy in downstream catalytic treatment of the exhaust. Presently, LNT research has not been as successful for CNG applications as when applied to lean burn gasoline engines or diesel engines [4-11]. Energy efficient breakthroughs are needed for LNT regeneration, desulfurization, and methane oxidation. The objective of this project is to develop a system capable of improving the effectiveness of LNTs applied to CNG-fueled lean-burn engines.

This project is focused on the development of an energy efficient aftertreatment system capable of reducing NO_x and methane by 90% from lean-burn natural gas engines by applying active exhaust flow control. The system consists of a Lean NO_x Trap (LNT) system and an oxidation catalyst. Through alternating flow control, a major amount of engine exhaust flows through a large portion of the LNT system in the absorption mode, while a small amount of exhaust goes through a small portion of the LNT system in the regeneration or desulfurization mode. The restricted partial flow has a very low space velocity that reduces the requirement for secondary fuel. By periodically reversing the exhaust gas flow through the oxidation catalyst, a higher temperature profile is maintained in the catalyst bed resulting in greater efficiency of the oxidation catalyst at lower exhaust temperatures. The project involves conceptual design, theoretical analysis, computer simulation, prototype fabrication, and empirical studies.

An integrated LNT -oxidation embedment is being developed which will have no negative impacts on engine operation in terms of output, efficiency, durability, and maintenance. The flow reversal system is being designed with zero slip during switching. The results of this research will help to attain the long-term NO_x emission target of 0.1 g/hp-hr, energy efficiency of 50%, and cost reduction of 10%.

Experimental

Bench Flow Reactor System Description

Bench-Flow Reactor

The bench-flow reactor (BFR) was built to supply necessary kinetic data and system optimization information for the ARES natural gas (NG) stationary power-generating engine. The data collected will provide the ARES program with vital information pertaining to nitric oxides (NO_x) and total hydrocarbon (THC) conversion efficiency. Both NO_x and THC conversion are functions of the catalyst type and loading, space velocity, temperature, and exhaust gas mixture. Each of these parameters are controlled and examined to obtain the needed data in a laboratory setting.

Reverse-Flow Oxidation Catalyst Reactor

The main objective of the RFOCR is to increase the temperature profile over the catalyst surface and hence to increase the THC conversion efficiency. These are the main parameters that control the surface temperature of the catalyst. The exhaust gas flow rate defines the residence time, which is the time available for the exhaust gas to react with the catalyst. The exhaust gas temperature defines the rate of reaction on the surface. Switching time represents the time duration between the exhaust gas flowing in the forward direction to the reverse direction.

The goal of the BFR is to reduce total hydrocarbon (THC) in the simulated natural gas exhaust flow. THC reduction is investigated as a function of the directional duration of the exhaust gases through the catalyst. Finally, the addition of supplemental fuel in order to achieve optimal THC reduction at low inlet temperatures will be investigated.

THC reduction is achieved by means of a palladium-alumina catalyst (EmeraChem). The THC catalytic chemical reaction, at an elevated exhaust gas temperature, is an exothermic reaction. The reverse-flow configuration of the reactor has the capacity to take advantage of this increase in temperature due to the exothermic reaction. The switching of the inlet and outlet exhaust direction “traps” this increased temperature inside the catalyst reactor, increasing the overall temperature of the exhaust gas through the catalyst.

Supplemental fuel implementation to the reverse-flow oxidation catalyst reactor (RFOCR), in the form of hydrogen or carbon monoxide (H₂ and CO, respectively), is presently being investigated for low inlet temperature (engine idle or low load) conditions. Periodically injecting supplemental fuel into the RFOCR increases the temperature across the catalyst reactor allowing for increased THC conversion. Supplemental fuel is introduced into the BFR system via a piezoelectric valve (MaxTek).

It is important to control the time of forward flow and reverse flow for maximum temperature profile over the catalyst length. A prolonged flow in a particular direction will have the same effect as of that of a normal unidirectional flow. So it is necessary to identify the optimum switching time. A comparison between the symmetric and asymmetric switching can also be done. When there is an unequal forward flow and reverse flow periods, this is defined as asymmetric switching. The effect of asymmetric switching on the conversion of methane and the temperature on the surface of the catalyst can then be identified.

Lean NO_x Trap Catalyst Reactor

The objective of the lean NO_x trap catalyst reactor (LNT) is to reduce NO_x in the simulated natural gas exhaust stream. Several parameters are currently being investigated in the LNT experiment. These parameters include identifying the breakthrough time, identifying catalyst saturation, maximizing catalyst storage by shifting between a fuel lean phase mode and a fuel rich phase mode, and identifying the best reductant (CO, H₂, or CH₄) in the rich phase.

The term breakthrough time is defined as the onset of NO_x in the exhaust gas flow downstream of the LNT reactor. The saturation time is defined where NO_x conversion ceases. The lean phase and rich phase can be effectively operated to maximize NO_x storage in the catalyst. In the rich phase, a supplement fuel is injected in the exhaust gas stream for catalyst regeneration. The supplement fuel (CO, H₂, or CH₄) is introduced into the gas mixture separately in rich phase. By introducing the supplement fuels separately, the best reductant, that is the one that regenerates the catalyst the fastest, can be identified.

Data Acquisition System

The data acquisition system (DAC) consists of hardware components purchased commercially from National Instruments. Custom data acquisition and control software was developed, using LabVIEW™, for operating the BFR system and collecting and displaying experimental data. The purpose of the DAC, besides simply data logging, is to be a very flexible system and to allow for modifications of the BFR system as seen fit. LabVIEW has the capability for multiple component interfaces, allowing for all components integrated into the BFR system to be logged at a user defined sampling rate. Hardware components (thermocouples, pressure transducers, etc.) can be easily added to the BFR system and integrated.

Bench-Flow Reactor Components

Overall Description of the Bench-Flow Reactor

A schematic of the bench-flow (BFR) is shown in Figure 1. A picture of the BFR system is also shown in Figure 2. Simulated natural gas exhaust emissions are introduced into the bench-flow reactor system by means of mass flow controllers.

The simulated exhaust gases consist of CO₂, H₂, NO_x, CH₄, Air, N₂, and CO. An example of a particular gas cylinder concentration in a rich configuration is shown in Table 1. Water vapor is introduced into the system via the steam generator. Downstream of the steam generator the system is divided into two separate reactor systems, the Lean NO_x Trap Catalyst Reactor (LNT) or the Reverse-Flow Oxidation Catalyst Reactor (RFOCR). The exhaust gas is then sent to the Horiba analyzer bench. Experimental data is acquired through a Nation Instruments LabVIEW data acquisition system.

Figure 1 Schematic of bench-flow reactor system

Figure 2 Bench-flow reactor system

NO _x	1000 ppm
H ₂	750 ppm
CO	0.5%
CH ₄	1500 ppm
CO ₂	8%
H ₂ O	10%
O ₂	10%
N ₂	Balance

Table 1 Simulated Exhaust Composition

Reverse-Flow Oxidation Catalyst Reactor

The reverse-flow oxidation catalyst reactor (RFOCR) configuration is described below in detail and shown in Figure 3 and 4. The RF oxidation reactor system consists of stainless steel tubing, several valves, thermocouples, heating tape, insulation, a pressure transducer, and an oxidation catalyst. The purpose of these components is to obtain specified operating conditions for the bench-flow reactor.

Figure 3 Reverse-flow oxidation catalyst reactor

Figure 4 Reverse-flow oxidation catalyst reactor

The RFOCR consists of, as the entire BFR system, 1/4" stainless steel tubing (Swagelok). This is used to contain and direct the simulated natural gas exhaust gas. The tubing is arranged in a manner to minimize the dead volume in the RFOCR system, seen in Figure 4. Swagelok components are exclusively used in the construction of the BFR.

The RF oxidation reactor system consists of four high temperature pneumatically operated valves (Swagelok), dubbed "switching valves", seen in Figure 5. The operating air pressure of these valves is 60 psi. Two Peter Paul 24 VDC three-way solenoid valves pneumatically actuate the four switching valves, also seen in Figure 5. Solenoid mufflers (Alwitco) are added to each solenoid valve to minimize noise during operation, shown in Figure 5. A power supply in conjunction with solid-state relays controls the three-way solenoid valves via LabVIEW. The set-up of the power supply and solid-state relays can be seen in Figure 6. LabVIEW is then able to control the solenoid valves, and intern the switching valves, giving the operator the ability for precise control of the RFOCR. Switching valves one and four and switching valves two and three, shown in Figure 5, can be manipulated in such as way allowing the exhaust gas stream to flow in the forward direction or in the reverse direction through the oxidation reactor.

Figure 5 Reverse-flow schematic

Figure 6 Power Supplies with solid state relays

Adiabatic conditions are maintained in the RFOCR by a system of heating tapes, insulation, thermocouples, solid-state relay switches, and Labview. To avoid condensation and to maintain a constant gas temperature throughout the RF oxidation system, high temperature heating tape (Omega Engineering) and insulation (Zetex) are wrapped around the stainless steel tubing. Type-K thermocouples (Omega Engineering) are located between the preheater and RFOCR, before switching valves one and two, and five thermocouples located in the RF oxidation catalyst reactor. Figure 1 shows the exact location of each thermocouple in the BFR system. The heating tapes are wired to relay switches enabling the gas temperature to be controlled via Labview. High temperature Zetex insulation is wrapped around the outside of the heating tapes and tubing for adiabatic conditions as well as for safety purposes.

Figure 7 Pneumatic valve operation

Extensive examination of several technical papers concerning natural gas (NG) exhaust catalytic combustion concluded that noble metal-based catalysts (palladium, platinum, and rhodium) generally have higher activity and resistance to sulfur poisoning than metal/transition oxide-based catalysts [12]. Results show that CH_4 conversion efficiency varies by the following, palladium>platinum>rhodium, all on alumina substrate [13]. Based on these findings, a palladium-alumina catalyst was selected for the oxidation of CH_4 in the BFR system. All catalysts were obtained from Emerachem. Specifications of the two reactor catalysts, the LNT reactor and RFOCR, are shown in Figure 8. The catalyst sample is placed inside a 12" long quartz tube with an OD of 1". Cajon fittings (Swagelok) were used when connecting the quartz tubing to the stainless steel 1/4" tubing. Viton o-rings (Swagelok) were also used for the seal around the quartz tubing. To ensure that leakage around the catalyst once placed in the reactor was kept to a minimal FiberFrax was wound around the catalyst before inserting the sample into the quartz tube.

Figure 8 Physical parameters of catalyst

Lean NOx Trap Catalyst Reactor

The Lean NOx Trap System (LNT) consists mainly of three parts namely; the syringe pump, steam generator, and the LNT Reactor. The mass flow controllers (MFC) have two possible paths, one through the steam generator to the LNT inlet manifold and the other directly to the LNT manifold (steam generator bypass). All components are connected by quarter inch stainless steel tubing (Swagelok) and are insulated using one-inch (Zetex) strips.

Ten monolith catalysts for the LNT were obtained from EmeraChem. The catalyst has a cordierite substrate and alumina washcoat with Barium Oxide. The cell density of the catalyst was 300 cells/inch² and had 100g/ft³ of platinum. The catalyst samples are 3" long and 7/8" in diameter. The catalyst was placed in a quartz tube, which had a 1" OD and 7/8" ID. The catalyst is wrapped with glass wool (FiberFrax) to prevent the gas from flowing through the edges of the catalyst. The inlet side of the tube is fixed with 5mm beads (Pyrex) to maintain a uniform flow through the catalyst. Temperature probes are placed at the inlet and the exit of the catalyst to identify the temperature change across the catalyst. The probes are fixed as close as 0.2" from the catalyst edges and are made sure that the probes do not touch the catalyst surface. The tube furnace (Lindberg, Model TF55035A-1) can reach a maximum temperature of 1100°C. The exit of the reactor is led to the analyzer for analyzing the exhaust gases. The tubing from exit is wrapped with silicone rubber heating tape (Omega) and insulated using insulating strips (Zetex) to ensure that the water vapor does not condense before reaching the analyzer.

Instrument Panel

The instrument panel houses three major components of the Bench Flow Reactor System, shown in Figure 9. Mass flow controllers (MFC) precisely control the exhaust gas mixture that is introduced to the BFR. The data acquisition system (DAC) interfaces the analyzer output signal with LabVIEW. The instrument panel also houses a number of displays. These displays represent the temperature and pressure of the exhaust gas at specific points across the BFR system. The data is also displayed in LabVIEW for easy data storage.

Figure 9 Instrument panel (front)

Mass Flow Controller

The Mass Flow Controllers are used to control the flow of all the exhaust gases through the bench flow reactor. In our system, MFC's manufactured by Omega (Model FMA 5400/5500) are exclusively used. A MFC is used for each of the gas species used, which include Carbon Monoxide (CO), Oxides of Nitrogen (NO_x), Methane (CH₄), Hydrogen (H₂), Air, Nitrogen (N₂), and Carbon Dioxide (CO₂). MFC's require a 15 V DC power supply and are controlled by the LabVIEW software. Varying input voltages controls the MFC's.

Figure 10 Wiring of displays and mass flow controllers inside the cabinet

There are several different ranges for the MFC. The operating range for CO is 0 – 1 L/min. The operating range for NO_x, CH₄ and H₂ is 0 – 5 L/min and for that of N₂ and CO₂ is 0 – 10 L/min. The operating range for air is the maximum in the BFR system, which is 0 – 20 L/min. All the Mass Flow Controllers are in a linear correspondence with a voltage range of 0 – 5 V. All the MFC's are calibrated by the manufacturer in terms of N₂ and so it is necessary to have a correction factor, called the K-factor, for the other gas species flowing through them. The concentrations of different gases that need to flow through the MFC are calculated keeping in mind the K-factor correction.

The MFC has a feedback system, which measures the difference between the user input and the actual output and hence controls the input flow rates for accuracy purposes. The output from the MFC's for H₂, CH₄, NO_x, CO and air are combined into one flow manifold, which is controlled by a solenoid valve connected to Labview. The output from the solenoid valve is sent either to the inlet of the LNT or directly to the exhaust gas analyzer (BFR system bypass). N₂ and CO₂ are combine into another manifold and is controlled by another solenoid valve. These two gases are then sent to the steam generator to sweep out water vapor or to the BFR system bypass line, directly to the gas analyzer

Pressure and Temperature Displays

The system includes four pressure transducers (Cole Parmer, Model 68072 – 06) and fourteen temperature thermocouples (Omega). These are connected to four pressure displays (Cole Parmer, Model 94785 – 00) and nine temperature displays (Omega, Model DP 18-KC1) that are housed in the cabinet. The other five temperature output can be displayed using a single temperature display using a temperature scanner (Omega, Model MDP – 18). The analog output

from the pressure transducer is 4 to 20 mA and this is in linear correspondence with a pressure range of 0 to 50 psi. The temperature displays have a range of 0 – 1250°C. The four pressure transducers are placed at the following positions in the system:

- i. inlet of the steam generator
- ii. outlet of the oxidation catalyst
- iii. inlet of the Lean NOx Trap
- iv. outlet of the Lean NOx Trap

Out of the fourteen thermocouples for measuring the temperature, the Reverse-Flow Oxidation Catalyst System is using seven thermocouples. The remaining seven are positioned as described below:

- i. inlet of the steam generator
- ii. inlet of the steam generator bypass
- iii. inlet manifold of the LNT
- iv. near the entrance of the catalyst inside the LNT furnace
- v. near the exit of the catalyst inside the LNT furnace
- vi. inlet of the preheater
- vii. outlet of the preheater

Heating Tape Controller

There are two heating tape controllers (Model – XT16) mounted on the front side of the cabinet. This is used to control the two heating tapes (Omegalux, heavily insulated Samox) that are wrapped around the inlet of the steam generator up to the three-way valve, shown in Figure 15, and to the LNT furnace inlet manifold. The purpose of the heating tape is to maintain the temperature from dropping due to heat loss to the environment from the stainless steel tubing. The heating tape has a maximum range of 1400°F.

Manually Operated Switches

There are four manual switches in the front of the cabinet to control the following:

1. analyzer bypass
2. manual reactor selection switch (Ox reactor or LNT)
3. heating tape between steam generator and three-way valve
4. heating tape between three-way valve and LNT inlet manifold

Syringe Pump

Figure 11 Harvard syringe pump

To simulate an actual NG exhaust gas mixture, it is important to include water vapor into the BFR system. Water is introduced by means of a syringe pump, shown in Figure 11. The syringe pump injects a near exact amount of water that is required in the exhaust gas. The syringe pump (Harvard, Model 11 plus) has a 50 ml syringe and a range of 0.0028 ml/min to 46.4 ml/min, with a precision of 0.5% error or less.

Steam Generator

Figure 12 Steam generator

After the syringe pump injects the required amount of water into the BFR system, it is forced into the steam generator and is swept out by N_2 and CO_2 . Both the inlet and outlet of the steam generator are sealed with copper gasket and stainless steel flange (1.75" ID and 2 $\frac{3}{16}$ " OD). The flanges are assembled using a torque wrench to ensure there are no leaks in the steam generator portion of the BFR system. The steam generator has a wick strip inside to help the water from pooling in the tube. The stainless steel tubing can be heated up to 1850°F using the steam generator (Lindberg heavy duty tube furnace), shown in figure 12. The outlet of the steam generator connects with the steam generator bypass line and then on to the three-way valve, shown in Figure 14, which allows the exhaust flow to enter either the RFOCR or the LNT reactor.

Preheater

Figure 2 shows the location of the gas preheater relative to the other components in the BFR system. Because of the high "light-off" temperature of methane (CH_4), the simulated NG exhaust gas is heated to a specified temperature and uniformly maintained in the preheater furnace (Lindberg). Pyrex beads (5mm diameter), held in place by stainless steel mesh disks (McMaster), and are placed inside a 22" long stainless steel pipe with a 1" ID. Due to the high temperature (~500°C) of the preheater, Pyrex was selected for its specific physical properties and working temperature of the glass beads. The purpose of the Pyrex beads is to assist in a uniform

temperature rise of the exhaust gas. The end fittings of the preheater pipe, dubbed “Christmas tree fitting,” are shown in Figure 13. The Christmas tree fittings have several ports, two being the inlet and outlet of the preheater. These ports allow the thermocouples and a RFOCR bypass line to be introduced in the BFR system. A type-K thermocouple is placed inside of the preheater to accurately determine the gas temperature inside the preheater. The RFOCR bypass line is open and shut via a manual high temperature stem valve (Swagelok).

Figure 13 Preheater “Christmas Tree” fitting

Pressure transducers

Three pressure transducers are positioned in the BFR system to ensure there is no pressure loss or backpressure in the system, shown in Figure 1. Cole Parmer pressure transducers are exclusively used in the BFR system. Pressure transducers are connected at the steam generator bypass line and downstream of the NO_x absorbing and RF oxidation catalyst reactors. The purpose of positioning a pressure transducer inline of the steam generator bypass line is to assure that there is not a backpressure at the high temperature pneumatic valve, shown in Figure 14. A pressure difference between the exhaust gases flowing through the steam generator and the steam generator bypass line runs the possibility of one of the exhaust gas lines to cease to flow. Pressure transducers located downstream of the two reactors are position to allow a pressure loss to be detected through the respective catalyst reactors.

Figure 14 High temperature three-way pneumatic valve

Emission analyzers

Exhaust emission conversion efficiency is of primary concern for the BFR system, namely methane (CH_4) and nitric oxide (NO_x) conversion. In order to maximize conversion efficiency the exhaust gas must be analyzed for the species mixture concentration. This is accomplished via the Horiba analyzer bench. This bench consists of four analyzers, CO (Low)/CO₂ and CO (High) (AIA-220), Hydrocarbon (FIA-210), and NO_x (CLA-220), as shown in Figure 15. All analyzers are calibrated and utilized at standard range settings. Calibration and spanning procedures for each analyzer is strictly followed per protocol of Horiba Instruments.

Figure 15 Front panel of Horiba exhaust gas analyzer

Prior to each experimental run of the BFR, each analyzer is spanned for accuracy using a gas divider. Both the span gas and the inert gas (N_2) are fed through the gas divider and to the analyzer at various percentages of the span gas concentration. Full calibration of the analyzers is performed monthly, as per Horiba instruction.

Data Acquisition

Data acquisition of the BFR system is accomplished through National Instruments hardware and software components. The hardware components of the data acquisition system consist of the following: a shielded rack mounted BNC adapter chassis (BNC-2090), PXI-1002, shielded rack mounted terminal block (TC-2095), and a 4-slot chassis (NI SCXI-1000), as shown in Figure 16. National Instruments LabVIEW 6.1 software is also used in the data acquisition system.

Figure 16 Data acquisition system – interface board

Mathematical Modeling of Reverse Flow Oxidation Catalyst

The most important objective of modeling the BFR is to simulate the forward-flow and reverse-flow of the exhaust gas through the RFOCR. The flow of the exhaust gas species through the catalyst has to be modeled with the help of a computer simulation. A one-dimensional model is developed and programmed with the help of softwares such as visual FORTRAN. The complex energy and mass balance equations, which are non-linear partial differential equations, have to be simplified to algebraic equations. This is achieved by the use of the backward difference method. By breaking down the equations to a simpler form, program code can be kept compact and uncomplicated.

It is also important to predict the effect of the following parameters on the temperature profile and methane (CH₄) conversion:

- a. exhaust gas flow rate
- b. exhaust gas temperature
- c. switching time

The modeling requires the kinetics of the global reaction over palladium. By operating the system in the differential mode with a ½” palladium based catalyst, the kinetics of the global reaction can be extracted. The purpose of maintaining a small catalyst length is to maintain an isothermal condition over the length of the catalyst.

Introduction to modeling

Computer modeling and simulation can give a valuable insight of the performance of a process. In many situations, computer modeling can yield results faster and cheaper than a actual

experiment. Many of the parameters for the actual design of the experiment can be obtained from the simulation. It is not feasible to change all the physical properties to obtain the optimum design for a actual experiment. This might make the experiment costly and waste valuable time if the data obtained is not useful. A computer simulation can overcome all these problems.

Flow through a catalyst can be modeled in different ways depending on the considerations and assumptions. Considering the changes in the physical properties along with one, two, or three spatial dimensions can create an accurate model. This mainly depends on the symmetry of the object to be modeled. A simple model can be obtained by modeling in a single spatial dimension, where complicated models require two or three dimensions. A basic insight of the model is given in the appendix section.

Modeling criteria

1. 1 – Dimensional

Our model is considered to be one-dimensional. The variations of the physical properties are along the direction of flow, and there is no change along the radial direction. This has been considered after seeing past results. It has been found that there is not much difference in the results obtained from a one-dimensional model and two or three-dimensional model. The later adds unwanted complication to the model considered.

2. Plug flow model.

The model is assumed to be of plug flow type. In a plug flow model there is no diffusion in the axial direction of flow.

3. Transient

This model is a transient model. The variation of the physical properties like temperature and concentration of the species is a function of time.

4. Pseudo homogeneous gas phase

The specific heat of solid is much more than that of the gas. The change in the gas phase temperature is much faster than the solid phase and hence with respect to the gas phase temperature, the solid phase temperature is assumed to be a constant. This condition is known as pseudo homogenous.

5. Heterogeneous solid phase

The gas-solid interface at the wall is assumed to be a discontinuity and separate mole and energy balances are written for the solid phase. These equations are coupled to the fluid phase equations through mass and heat transfer coefficients. This type of model is known as heterogeneous model.

Assumptions while modeling

Some of the assumptions that are made in modeling the reverse flow oxidation catalyst are listed below:

1. Neglect radial variations of gas phase temperature, concentration and velocity within the individual channels. These variables are interpreted as cross sectional averages.
2. Negligible temperature gradients in solid phase in the transverse direction.
3. Negligible axial diffusion of mass and heat in gas phase.
4. Number of active sites is a constant.

5. It is assumed that through out the process, the number of palladium sites that involves itself in the reaction is a constant. This means that the entire palladium site on the surface of the catalyst involves itself in the surface reactions. Deactivation is not considered in our model.
6. Chemical reactions occur only on the surface of the catalytic surface.
7. The model we have considered is a heterogeneous model and so the gas phase reactions are small and are neglected. Only the surface reactions are considered.
8. Thermo physical properties are assumed to be a constant.
9. All channels behave similarly.
10. Since all the channels in the catalyst have the same dimensions and properties, it is assumed to behave similarly. Because of this assumption, it is enough if we model for a single channel and the same model can be extended for all the channels.

Derivation of governing equations for the model

Consider a monolith catalyst channel that has a square cross section. To develop the species conservation equation and the energy equation, we consider a small incremental volume ΔV as shown in the figure 17 below. This volume element considers both the gas phase and the solid phase (washcoat and the substrate of the catalyst). In the derivation of these equations we consider the case of a single chemical reaction that proceeds catalytically on the wall surface and homogeneously in the gas phase.

We have 4 equations, two species conservation equations, one for solid phase and one for gas phase, and two energy equations, one for solid phase and one for gas phase.

Energy balance equation is solid phase

Accumulation of energy =

Change in conduction + Convective heat loss + Generation due to the
in Z direction chemical reaction

Figure 17 Control volume considered for a channel

Change in conduction along the Z direction

$$= \text{Conduction at } Z - \text{Conduction at } Z + \Delta Z$$

$$= \left(-K_z A_z \frac{\partial T_s}{\partial Z} \right)_Z - \left(-K_z A_z \frac{\partial T_s}{\partial Z} \right)_{Z+\Delta Z}$$

$$A_z = 2\pi r \Delta r$$

Let the fraction of gas in the total volume = ϕ

Therefore the fraction of the solid = $1 - \phi$

The effective thermal conductivity $K_z = (1 - \phi) K_w$

$$Z \text{ direction} = - \left((1-f) K_w 2\pi r \Delta r \frac{\partial T_s}{\partial Z} \right)_Z - \left((1-f) K_w 2\pi r \Delta r \frac{\partial T_s}{\partial Z} \right)_{Z+\Delta Z}$$

Expanding the above equation using Taylor's theorem and neglecting the higher order terms, we get

$$Z \text{ direction} = \Delta \left((1-f) K_w 2\pi r \Delta r \frac{\partial T_s}{\partial Z} \right)$$

Energy due to the chemical reactions

$$Q_{\text{generated}} = \eta [\Delta H * \text{Sum of Reaction Rate of all species} * \text{Incremental surface area}]$$

$$= \eta [\Delta H_R \sum (-RA)_{\text{surface}} \Delta S]$$

η Is the effectiveness factor i.e. the fraction of the surface that is available for the reaction.

Convective heat transfer

$$Q_{\text{conv}} = \text{heat transfer coefficient} * \text{Incremental surface area} * \text{temperature difference}$$

$$= h * \Delta S * (T_s - T_{\text{gas}})$$

Where ΔS is the incremental surface area.

$$Q_{\text{conv}} = h \Delta S (T_s - T_{\text{gas}})$$

$$\text{Accumulation of energy} = m C_p \Delta T$$

$$= \rho \Delta V (1-f) C_p \frac{\partial T_s}{\partial t}$$

Therefore the energy balance equation in the solid phase is

$$\Delta \left((1-f) K_w 2\pi r \Delta r \frac{\partial T_s}{\partial Z} \right) - \eta [\Delta H_R \Sigma (-RA)_{\text{surface}} \Delta S] - h \Delta S (T_s - T_{\text{gas}}) =$$

$$r \Delta V (1-f) C_p \frac{\partial T_s}{\partial t}$$

$$\text{Where } \Delta V = 2 \pi \Delta r \Delta Z$$

$$\left((1-f) \frac{\partial}{\partial Z} K_w \frac{\partial T_s}{\partial Z} \right) - \eta A_v \Delta H_R \Sigma (-RA)_{\text{surface}} - h A_v (T_s - T_{\text{gas}}) =$$

$$r \Delta V (1-f) C_p \frac{\partial T_s}{\partial t}$$

$$\text{Where } \Delta S / \Delta V = A_v$$

Therefore the energy balance equation becomes

$$\left((1-f) \frac{\partial}{\partial Z} K_w \frac{\partial T_s}{\partial Z} \right) - \eta A_v \Delta H_R \Sigma (-RA)_{\text{surface}} - h A_v (T_s - T_{\text{gas}}) =$$

$$r \Delta V (1-f) C_p \frac{\partial T_s}{\partial t}$$

Mole Balance equation for Solid State

The mole balance equation is obtained by equating the number of moles that is transported to the catalyst surface to the reaction rate.

Moles transported to the catalyst surface

= Mass transfer coef. * Bulk conc. * Change in concentration * Incremental volume

$$= k_m C_b (Y_{\text{surface}} - Y_{\text{gas}}) f \Delta V$$

$$\text{Reaction Rate} = h (-R_A)_{\text{Surface}} f \Delta V$$

Dividing the equation by $f \Delta V$

Mole Balance equation for Solid Phase

$$k_m C_b (Y_{surface} - Y_{gas}) = h (-R_A)_{Surface}$$

Energy balance equation in gas phase

Energy accumulated in gas phase =

Heat added to gas from surface + Heat generated in gas phase due to reactions

Energy accumulated in gas phase

$$\begin{aligned} &= \overset{o}{m} C_p \Delta T_{gas} \\ &= Q_V \mathbf{r} C_p \Delta T_{gas} \end{aligned}$$

Heat added to the gas phase from the surface due to convection

$$= h A_s (T_s - T_{gas})$$

Heat generated due to the chemical reactions in the gas phase

$$= -\Delta V \mathbf{f}(-R_A)_H \Delta H_R$$

Therefore the energy balance in gas phase is

$$Q_V \mathbf{r} C_p \Delta T_{gas} = h A_s (T_s - T_{gas}) - \Delta V \mathbf{f}(-R_A)_H \Delta H_R$$

Dividing the above equation by ΔV

$$\frac{Q_V}{A_s} \mathbf{r} C_p \frac{\partial T_{gas}}{\partial Z} = h A_v (T_s - T_{gas}) - \mathbf{f}(-R_A)_H \Delta H_R$$

$$V_s \mathbf{r} C_p \frac{\partial T_{gas}}{\partial Z} = h A_v (T_s - T_{gas}) - \mathbf{f}(-R_A)_H \Delta H_R$$

A_v = Surface Area / Volume

V_s = Superficial Velocity

Therefore the energy balance equation in gas phase becomes

$$V_s \mathbf{r} C_p \frac{\partial T_{gas}}{\partial Z} = h A_v (T_s - T_{gas}) - \mathbf{f}(-R_A)_H \Delta H_R$$

Bringing them to one side

$$h A_V (T_s - T_{gas}) - f (-R_A)_H \Delta H_R - r C_P V_S \frac{\partial T_{gas}}{\partial Z} = 0$$

Mole Balance Equation in gas phase

$$\text{Moles in} - \text{Moles out} - \text{Moles reacted in} - \text{Moles transported} = 0$$

homogenous reaction to catalyst surface

$$\text{Moles in} - \text{Moles out} = (F_A)_Z - (F_A)_{Z+\Delta Z} = -\Delta F_A$$

$$\text{Moles reacted in homogenous reaction} = (-R_A)_H f \Delta V$$

$$\text{Moles transported to the catalytic surface} = k_m C_b (Y_{A,B} - Y_{A,S}) A_S$$

Thus the mole balance equation in gas phase becomes

$$-\Delta F_A - (-R_A)_H f \Delta V - k_m C_b (Y_{A,B} - Y_{A,S}) A_S = 0$$

Dividing the above equation by ΔV , $\Delta V = A_S \Delta Z$

Equation becomes

$$\frac{-\Delta F_A}{A_S \Delta Z} - (-R_A)_H f - k_m C_b (Y_{A,B} - Y_{A,S}) A_V = 0$$

$$F_A = Q_V C_{A,B} = V_M A_S C_{A,B} Y_{A,B}$$

Hence the mole balance equation becomes

$$-V_M C_{A,B} \frac{dY_{A,B}}{dZ} - (-R_A)_H f - k_m C_b A_V (Y_{A,B} - Y_{A,S}) = 0$$

Governing Equations For The Reverse Flow Oxidation Catalyst

Gas Phase

Energy Balance

$$h A_V (T_s - T_{gas}) - f (-R_A)_H \Delta H_R - r C_P V_S \frac{\partial T_{gas}}{\partial Z} = 0$$

Mole Balance

$$-V_M C_{A,B} \frac{dY_{A,B}}{dZ} - (-R_A)_H f - k_m C_b A_v (Y_{A,B} - Y_{A,S}) = 0$$

Solid Phase

Energy Balance

$$\left((1-f) \frac{\partial}{\partial Z} K_w \frac{\partial T_s}{\partial Z} \right) - \eta A_v \Delta H_R \Sigma (-RA)_{\text{surface}} - h A_v (T_s - T_{\text{gas}}) =$$

$$r \Delta V (1-f) C_p \frac{\partial T_s}{\partial t}$$

Mole Balance

$$k_m C_b (Y_{\text{surface}} - Y_{\text{gas}}) = h (-R_A)_{\text{Surface}}$$

Reaction mechanism

The reaction mechanism is another important factor in the model. It defines the rate at which the reaction takes place on the surface of the catalyst. In reality it is not possible to consider all the reactions of the mechanism. Previous research shows that there are approximately 177 elementary equations that are involved in the oxidation of methane and 50 species are involved. It is enough if we consider the most important of those equations that may have an influence on the overall reaction rate. Given below are the simplest reaction mechanism involving three equations. A more complex reaction mechanism can be considered after simulating a simple model.

Rate expressions for the above reactions are given by

$$\begin{aligned}\bar{R}_{CO} &= k_1 C_{CO} C_{O_2} / G & [mol\ CO / cm^2\ Pd.S] \\ \bar{R}_{CH_4} &= k_2 C_{CH_4} C_{O_2} / G & [mol\ CH_4 / cm^2\ Pd.S] \\ \bar{R}_{H_2} &= k_1 C_{H_2} C_{O_2} / G & [mol\ H_2 / cm^2\ Pd.S]\end{aligned}$$

These are the rate expressions that are used in the governing equations of the model. The literature review gives us the activation energy of these reactions, but not much has been said about the specific rate constants. Operating in a differential mode can help in extracting the kinetic data for the above reactions. A half-inch catalyst is placed in a reactor and at different temperatures the conversion is then analyzed. The reason for having a very small length of the catalyst is to try to maintain an isothermal condition. This can yield us the required specific rate constants needed while modeling.

Boundary conditions

It is very important to define the boundary conditions properly as they are the ones that define the problem. Initially we consider this model to be insulated at the sides and no radiation is considered. The model can be extended to radiation once we get the solution for the simpler case. The temperature is defined as the initial condition. Knowing the initial temperature and the concentration of all the gas species at the inlet define the initial boundary conditions.

$$T_s(X, 0) = T_{s0}(X)$$

The boundary conditions are defined below

$$C_{g,i}(0, t) = C_{g,i}^{in}$$

$$T_g(0, t) = T_g^{in}$$

$$\frac{\partial T_s}{\partial x}(0, t) = \frac{\partial T_s}{\partial x}(L, t) = 0$$

LNT Partial Flow Regeneration and Desulfurization Gas Divider Design

Schematics and calculation

Two options were under consideration to design a space – shared LNT system for partial flow regeneration and desulfurization:

- Design of the rotating catalytic absorber cartridge and
- Design of the rotating gas divider

In both concepts the total exhaust flow of combustion products is divided in two streams. Approximately 98-95% of the exhaust stream passes through the region of the absorber in storing NO_x mode, and 2-5% of the stream passes through the region of absorber in regeneration mode.

In the preliminary investigation of the rotating catalytic absorber cartridge, it became evident there would be a problem sealing the catalyst cartridge and supply manifold from the environment as it is rotated to divide the exhaust stream. Also, it would be difficult to control sealing internally between the two parts, as they rotate, to provide accurate division of the exhaust flow. For this reason the rotating gas divider concept was taken into further consideration.

Figure 18 Schematic of rotating gas divider

Schematically the gas divider (Fig. 18) consists of two concentric cylinders (1 and 2). The gas divider's outermost cylinder, (see Appendix B), houses annular channels that separate the combustion products into six sections as it enters the absorber (7). Each section transfers the combustion products to the absorber. Five sections (a, b, c, e, f - in the existing figure) transfer pure exhaust gases (19% -19.6% of exhaust gases per section) providing the operation of 5/6 of catalyst volume in absorption mode. The remaining section (d) transfers a mixture of 2-5% of exhaust gases with reach-burn combustion products providing the operation of 1/6 of the catalyst volume in the desorption mode. The main flow of combustion products is injected through the manifold (8) passing into the exhaust distribution piston (3) (see Appendix B) and finally it

moves through openings (9 & 10) into the appropriate channels. 95-98% of exhaust gases come through opening (9) into 5 annular channels and 2-5% of exhaust gases are directed into the remaining annular channel through opening (10). In this channel the exhaust gases are mixed with injected reach-burn combustion products. The exhaust distribution piston is rotated by electrical motor (4) in order to provide the operational mode switching from channel to channel in the sequence “d-e-f-a-b-c”.

Rich-burn combustion products provide the excess H_2 and CO concentration and mixture temperature of 350-550°C needed to accomplish the effective NO_x desorption.

Fuel injection system will be controlled to inject the rich-burn combustion products into the designated desorbing channel follow the distribution sequence of the exhaust distribution piston.

There are several strategies to achieve the desired conditions of the reach-burn combustion products. Since reach burn combustion products should have the temperature of 500-650°C a special gas generator could be used to produce them (Fig. 19).

Figure 19 Schematic of gas generator

Standard engine propellant - methane and air at approximately stoichiometric air/fuel ratio could be used to organize the combustion in the front part of combustion chamber (1), with the air/fuel mixture ignited by a regular spark plug (3). In order to decrease the temperature of combustion products, fuel is injected (2) in the rear zone of combustion chamber. An oxidation (reforming) catalyst (4) is used to reduce the methane concentration and provide the needed concentration of CO and H_2 for the following NO_x desorption.

Preliminary calculations of main parameters of the rotating gas divider have been done to design the divider for the CG-280 Engine.

The standard Pt absorbing catalyst was chosen for the design. Because of high value of flow rate of combustion products (817 liter/sec for peak power) the catalyst brick has following dimensions: length – 6”, diameter – 9 ½”.

The needed space velocity this catalyst could provide was calculated as:

$$SV = (dV/dt) / V_c = 142 \text{ hr}^{-1},$$

where: dV/dt - volumetric flow rate of the exhaust gases; V_c – catalyst volume.

Since the space velocity of the mentioned standard catalyst brick is of 50,000 hr^{-1} , two catalyst bricks should be installed to provide the complete NO_x absorption. In this case $SV = 71,000 \text{ hr}^{-1}$, that less than $50,000 \times 2 = 100,000 \text{ hr}^{-1}$.

The cross section areas of the annular channel, exhaust piston, and the sum of cross section areas of 6 holes in the inner cylinder of the divider's housing were assumed equal in order to maintain constant velocity of combustion products in each cross section of the divider. The triangular shape of the holes in the inner cylinder (see Appendix B) of the divider housing maximizes the negative gradient of combustion products flow rate through the hole in the particular section of the annular channel during the transition from absorption mode to the regeneration mode (Figure 10). The cross section area of annular channel and cross section area of exhaust piston equal to 30.5286 in^2 . Cross section area of single triangle hole is equal to 5.087 in^2 . The area of the opening (10) in the exhaust piston is equal to 12 % from the area of single triangle hole. The area of the opening (10) can be varied to allow tuning of the exhaust flow in the regeneration channel during testing. Figure 20 below illustrates the predicted exhaust gas flow rate with the configuration described above.

Figure 20 Exhaust flow rate for AALNT

Description of Design

The designed gas divider can be divided into three parts. Their respective parts can be seen in Figures 21 and 22 below. The first is the exhaust product inlet and manipulation supply manifold. This portion consists of the gas inlet housing, the gas distribution piston, the gas divider manifold, and the fuel injection system. As described above, the exhaust gas will enter the supply manifold where it is then distributed by a rotating piston to its designated supply channel. From there it is then sent through the catalyst to regenerate or to be treated. The second part of the overall assembly is the catalyst chamber, catalyst chamber gaskets and catalyst housing force control rings. Sealing the flow of the six channels of exhaust products is important. If leaking between the channels occurs, the effectiveness of the catalyst regeneration can be reduced. Because the catalyst brick is brittle and the operating temperatures are high, the gasket material needs to be able to flex a small amount so as to not damage the brick but be durable enough to withstand the catalyst temperatures. A catalyst wrap material used to set catalysts in housings was used as the gasket. This will both withstand the high temperatures and provide the appropriate seal between the six supply channels. The copper force ring is used to control the force applied between the face of the exhaust channel dividers and the face of the catalyst brick.

The force can be adjusted by changing the thickness of the ring. The thickness of the force ring will be refined during testing. This ring is also used to seal the outer housing from leaking exhaust to the environment. The same type of gasket and seal is located on both ends of the catalyst chamber. The final section of the system is the test cell. This unit bolts to the exit side of the catalyst chamber and measures the temperature and concentration of the combustion products as they exit the catalyst.

Figure 21 Exploded view of partial flow LNT

Figure 22 Section view of partial flow LNT

Engine Test Cell

Test Bed Development

The test bed was developed to allow both The University of Tennessee and the Oak Ridge National Lab (ORNL) to evaluate their unique LNT systems, applied to a lean burn natural gas engine. The development of the test bed, including installation of the engine and setup of all controls and data acquisition for LNT management, has been completed. The engine was installed on a 500 hp DC dynamometer. DyneSystems hardware and software is used for all test bed controls and data acquisition. The Dyne-Loc IV, a digital dynamometer controller, and the DTC-1, a digital throttle control system, are used for dynamometer and engine throttle control. Cell Assistant for Windows, a PC-based data acquisition and controls software, is used for data display and collection. The emissions sampling system consists of two benches, one for measuring engine out and the other for catalyst out emissions. The benches consist of a set of California Analytical and Horiba instruments capable of measuring CO, CO₂, O₂, HC and NO_x.

C Gas Plus General Engine Data		
Name	C Gas Plus	
Model	CG-280	
Type	4 Cycle; In-Line 6 Cylinder	
Bore x Stroke (mm)	114 x 135	
Displacement (L)	8.3	
General Performance Data	Peak Power	Peak Torque
Engine Speed (rpm)	2400	1400
Engine Power (kW)	209	169
Engine Torque (N-m)	831	1153
Inlet Air Flow (L/sec)	293	205
Exhaust Gas Flow (L/sec)	817	539
Exhaust Gas Temperature (C)	643	587
Nominal Fuel Consumption (kg/hr)	47	34
Inlet Air Restriction (mm H ₂ O)	445	
Exhaust Restriction (mm Hg)	102	

Table 2 Engine specifications

The test bed will demonstrate NO_x reduction on a Cummins 8.3-liter lean burn natural gas engine. The engine produces 193 kW at 1800 rpm and a peak power of 209 kW at 2400 rpm. While this engine is not in the size range of interest to the ARES program it serves as a practical starting point for assessing the catalyst technology. Table 2 shows general performance data for the engine.

Currently the LNT system for ORNL is installed on the test bed to allow for an initial catalyst evaluation and development of regeneration parameters. This LNT system consists of a dual path NO_x adsorbing catalyst system shown below. One leg of the dual path system is regenerated while the other is in the adsorption stage. Methane is injected into the exhaust stream ahead of the catalyst to create the fuel rich environment necessary for regenerating the LNT, and a set of pneumatically actuated exhaust brake valves are used to control the space velocity. The catalyst system consists of an oxidation, reformer and LNT catalyst developed by EmeraChem.

Figure 23 Dual leg catalysts system schematic

Baseline Engine Testing

The baseline performance and emissions of the 8.3-liter natural gas engine have been characterized at a matrix of operating points. Some initial catalyst volume estimates have been made based on exhaust flow, NO_x level and temperature data. Baseline results will also be used to determine the validity of translating data from bench flow studies to full-scale engine data. This baseline data will also allow us to investigate the scalability of the 8.3-liter engine compared to the larger genset engines of interest to the ARES program. Figure 24 shows a matrix of engine out NO_x levels for the C Gas Plus.

Figure 24 NOx rating

Development of LNT Regeneration Strategy

Methane will be used as the reducing agent for the LNT. Before the storage sites of the catalyst become saturated methane will be injected into the exhaust directly upstream of the catalyst. Control over the injection rate will allow for control of the exhaust composition entering the catalyst. In addition, during regeneration the space velocity must be controlled across the catalyst to allow enough time for NO_x release and subsequent reduction. This will be done by throttling the exhaust flow with a set of exhaust brake valves. An algorithm will be developed to allow the operator to control space velocity by adjusting valve position and exhaust composition by adjusting methane injection rate. These two variables can then be used to optimize the NO_x conversion efficiency while minimizing the associated fuel penalty.

Preliminary studies have been conducted to determine the exhaust composition as a function of the injection rate of the reducing agent. These studies were used to map out the stoichiometry of the exhaust at various injection rates for five engine operating points (100%, 75%, 50%, 25% and 10% load) at 1800 rpm. The goal was to achieve various rich exhaust conditions ($\lambda = 0.5, 0.7, 0.9$) by making adjustments to the duty cycle of the methane injectors. The following graph shows the injector duty cycles necessary to achieve the targeted rich exhaust conditions. The mass flow rate of the injected reducing agent was also measured using a coriolis mass flow meter. This will allow for predictions of the fuel penalty associated with regenerating the catalyst. The following graph shows the fuel penalty based on the following adsorption-regeneration cycle; 27 seconds lean exhaust (LNT adsorption), followed by 3 seconds rich exhaust (LNT regeneration).

Preliminary space velocity control studies have also been conducted at 1800 rpm. The goal of these studies was to determine the exhaust flow across the catalyst while the exhaust brake valve throttles the flow. This would be the exhaust flow condition during the regeneration phase. These results, which are shown below can be used to help estimate necessary catalyst volume and regeneration periods.

Figure 25 Injector duty cycle

Figure 26 Fuel penalty

Figure 27 Exhaust flow rate

Nomenclature

- $a(x)$ = Catalytic surface area per unit volume, cm^2/cm^3
 A_v = Frontal area of the monolith, cm^2
 $C_{g,i}$ = Concentration of species i in the bulk gas stream, mole fraction
 $C_{s,i}$ = Concentration of species i in the solid phase, mole fraction
 D_i = Diffusivity of the species i in the relative mixture, cm^2/s
 C_{pg} = Specific heat of gas, J/g.K
 C_{ps} = Specific heat of solid, J/g.K
 h = Heat transfer coefficient, $\text{J}/\text{cm}^2.\text{s.K}$
 K_{mi} = Mass transfer coefficient of the species i , cm/s
 R_h = Hydraulic radius of channel, cm
 R_g = Universal gas const.
 $(R_a)_H$ = Specific reaction rate in gas phase for species i , $\text{mol}/\text{cm}^2\text{Pd.s}$
 $(R_a)_{\text{surface}}$ = Specific surface reaction rate for species i , $\text{mol}/\text{cm}^2\text{Pd.s}$
 S = Geometric surface area per unit volume, cm^2/cm^3
 T_s = Solid Temperature, K
 T_{gas} = Gas Temperature, K
 v = Linear Superficial velocity of exhaust gas, cm/s
 W_g = Mass flow rate, g/s
 Y_{surface} = Mole fraction on the surface
 Y_{gas} = Mole fraction of species in gas phase

Symbols

- f = Void fraction of the monolith
 I_s = Thermal conductivity of the solid, $\text{J}/\text{cm.s.K}$
 r_g = Gas Density, g/cm^3
 r_s = Solid Density, g/cm^3

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Appendices

Appendix A- Computer Simulation of the Model

Software used

The software used for the computer simulation of flow of exhaust gases through reverse-flow oxidation catalyst is done using Visual Fortran version 6.0B. This software has exceptional mathematical abilities. FORTRAN stands for formula translator and as its name suggests, it's used for mathematical applications.

Main and Subroutines used in this program code

1. Driver

This is the main program which is executed first. This is where the variables are defined and the memory allocations are done. We can see that most of the variables are defined as common. This is to define it as a global variable so that its values are available for all the subroutines for further calculations. The main program is where we input all the values. This is the program that accepts the input values for the variables used. After accepting the input parameters, it divides the catalyst into 80 nodes for computing the concentration and the temperature along the length of the catalyst. All input boundary conditions are assigned to node zero. The main program is where all the subroutines are called from. After going through all subroutines, the results are displayed using a print statement.

2. c1solver

This subroutine is called from the main program. The purpose of this subroutine is to find the terms required for the mass balance equation. It calculates the mass transfer coefficients from Sherwood number and hydraulic radius. The Sherwood number is found to 3.608 in our case. From c1solver the control is transferred to node1 subroutine at node 1 and to nleqs subroutine for the remaining nodes.

3. node1

This is the subroutine where the parameters for the first node are found. Node1 subroutine computes the reaction rate, temperature and concentration of different species at the point. It uses a subroutine called Hybrd.

4. nleqs

This subroutine is called from c1solver and this is used for computing the values at the remaining 79 nodes. This solves fully coupled non linear algebraic equations. The complex species conservation and mass conservation equations are broken to non linear

algebraic equation using the finite difference method. The values that are obtained at the end of the subroutine is sent back to c1solver which in turn does calculation and send it back to the main program.

5. lsode

Lsode stands for Livermore solver for ordinary differential equation. LSODE is the basic solver of the collection, and solves explicitly given systems. It solves stiff and nonstiff systems of the form $dy/dt = f$, where y is the vector of dependent variables and t is the independent variable. In the stiff case, it treats the Jacobean matrix df/dy as either a dense (full) or a banded matrix, and as either user-supplied or internally approximated by difference quotients. It uses Adams methods (predictor-corrector) in the nonstiff case and Backward Differentiation Formula (BDF) methods (the Gear methods) in the stiff case. The linear systems that arise are solved by direct methods (LU factor/solve). LSODE supersedes the older GEAR and GEARB packages, and reflects a complete redesign of the user interface and internal organization, with some algorithmic improvements. This program is called from the main program.

When using LSODE, some important inputs are:

- Number of first-order ODE's
- Array of initial values
- Initial value of independent variable
- First point where output is desired
- Tolerance
- Name of subroutine for Jacobean matrix (optional)
- Method flag (can choose between BDF or Adams methods, user-supplied or internally generated Jacobean, banded or full Jacobean, etc.)

In addition, the expected output includes:

- The $y(t)$ vector of computed values
- Corresponding value of independent variable
- A state flag (indicates if program was successful or not and why)

6. HYBRD

The purpose of the entire HYBRD system of subroutines is to solve the nonlinear algebraic equations. The `hybrd1` subroutine is to find a zero of a system of N nonlinear functions in N variables by a modification of the Powell hybrid method. The user must provide a subroutine which calculates the functions. The Jacobean is then calculated by a forward difference approximation. It requires the physical variables to be renamed as mathematical variables and be used. In our model we have a total of 9 equations and 9 unknowns. Hybrd solves these 9 equations and it gives us the concentration of the various species and the catalyst temperature at all those nodal points. This subroutine is called from both `node1` and `nleqs`. It returns the main program with newly computed values at the nodal points.

Appendix B- LNT Drawings

Exhaust inlet housing

Gas Divider housing

Exhaust Distribution Piston

Piston Plate used to vary the percent of exhaust mixed with fuel during regeneration mode.

Gas Divider Manifold forward section

Expansion section of the Gas Divider Manifold, conic cross-section allows for maximum performance of catalyst by reducing unused catalyst surface area.

Catalyst can design

Flange design

Exit Exhaust sample collection system