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#### Computational fluid dynamics studies of catalytically stabilized

## combustion of propane in flow tube reactors

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#### Abstract

The most efficient and stable combustion occurs in a catalytic reactor when the burning mixture is in contact with the catalyst for a sufficiently long period. When the contract period is too short, insufficient energy is generated adjacent to the catalyst surface to sustain combustion in the main or free stream. This study is focused mainly upon the essential combustion characteristics of propane-air mixtures in flow tube reactors with a heat-recirculating structure. Computational fluid dynamics simulations are performed to gain a greater understanding of the mechanisms of flame stabilization. The essential factors affecting flame stability and combustion characteristics are determined in order to obtain design insights. The results indicate that in order to meet the emission level requirements, for industrial low emission gas turbine engines, staged combustion is required in order to minimise the quantity of the oxides of nitrogen produced. The combustion catalyst has several desirable characteristics: they are capable of minimizing nitrogen oxides emission and improving the pattern factor. Operating the combustion process in a very lean condition, namely high excess air, is one of the simplest ways of achieving lower temperatures and hence lower nitrogen oxides emissions. The use of a catalytic combustor offers the advantage that all of the fuel can be oxidized therein, resulting in ultralow nitrogen oxides emissions and low carbon monoxide and unburned hydrocarbon levels. In mass transfer controlled catalytic reactions, one cannot distinguish between a more active catalyst and a less active catalyst because the intrinsic catalyst activity is not determinative of the rate of reaction. It is possible to achieve essentially adiabatic combustion in the presence of a catalyst at a reaction rate many times greater than the mass transfer limited rate. The maximum achievable velocity depends on flow conditions and catalyst parameters such as type, monolith cell size, and web thickness. **Keywords:** Emissions; Oxides; Mixtures; Pollutants; Temperatures; Energy

## **1. Introduction**

Various methods for the thermal reaction of carbonaceous fuels for powering conventional engines and power plants are known. One system which has been used in connection with stationary gas turbines is a catalytic combustor system [1, 2]. These systems operate based on a mechanism which has been called catalytically supported thermal combustion. The physical mechanisms allow sustained catalytic combustion to occur at high reaction rates. Basically, the catalytically supported thermal combustion state is achieved when there is a sufficiently intense catalytic combustion adjacent to the walls of the chamber containing the catalyst to maintain a high bulk temperature and to thereby support thermal combustion in the free stream of the fuel-air admixture. If the temperature is not sufficiently high, thermal combustion will be incomplete and substantial quantities of the fuel will not be burned. Accordingly, the two design parameters for steady state operations of such a catalytic reactor are the mixture inlet temperature and the fuel-air ratio [3, 4]. The latter parameter controls the temperature rise in the reactor.

Tests which have been performed have identified the minimum preheating temperatures that are required for lean catalytic operations [5, 6]. When the fuel is natural gas, values of about 350 degrees in the Fahrenheit scale for noble metal catalysts and 300 degrees in the Fahrenheit scale for base metal oxide catalysts are typical. For liquid fuels, values in the range of 300-600 degrees in the Fahrenheit scale have been demonstrated. In typical gas turbine systems, the compressor discharge temperatures are in the neighbourhood of 600 degrees in the Fahrenheit scale and therefore no additional preheating of the incoming air stream is necessary. In general, the maximum operating temperature of a catalytic combustor is limited by material capabilities to below 3000 degrees in the Fahrenheit scale and the minimum temperature is generally limited by the combustion stability of the fuel-air system to above 1800 degrees in the Fahrenheit scale. Since the current commercial stationary engines have turbine inlet temperatures of approximately 1850-2000 degrees in the Fahrenheit scale, they are well suited for use with catalytic combustors [7, 8]. During the steady state operation under load, the catalyst temperature is somewhat in excess of the turbine inlet temperature because of liner cooling and the dilution air which is added to the reactor combustion products.

Operation at about 2300-2400 degrees in the Fahrenheit scale, in fact, provides a substantial degree of flexibility in the selection of appropriate catalyst systems. However, since inlet temperatures are well below 1800 degrees in the Fahrenheit scale in combustors during the start-up sequence and during the loading sequence, and since air scheduling is not considered to be a desirable control technique, alternate means of start-up are required [9, 10]. It will be appreciated that the peak temperatures in the catalytic bed equals the adiabatic flame temperature of the entering fuel-air mixture [11, 12]. Accordingly, if the operating temperature is to be 2300-2400 degrees in the Fahrenheit scale, the maximum bed fuel-air ratio is limited. The most efficient and stable combustion occurs in a catalytic reactor when the burning mixture is in contact with the catalyst for a sufficiently long period. When the contract period is too short, insufficient energy is generated adjacent to the catalyst surface to sustain combustion in the main or free stream. While a number of analytical models of this complex have been developed, two parameters in particular, face velocity and nominal residence time of the mixture in the reactor, have been used to evaluate experimental performance in actual use [13, 14]. The maximum values of face velocity, namely the mean velocity of the mixture upstream of the catalyst, range for various catalysts [15, 16]. This condition also requires that the catalytic reactor have a minimum frontal area for the combustor air-fuel flow to traverse.

This study relates to the essential characteristics of catalytically stabilized combustion in a microscale system with a heat-recirculating structure. Numerical simulations are conducted to gain insights into system performance and to determine what changes can be made to improve its robustness and stability. Detailed kinetics are used for modelling the system in computational fluid dynamics. Methods of applying a heat-recirculating structure to the channel walls are employed, which may be utilized with presently existing designs of micro-scale combustion systems. The factors affecting combustion stability are determined for the system. The objective of this study is to investigate the essential characteristics of catalytically stabilized combustion in a micro-scale heat-recirculating system so as to gain a greater understanding of the mechanisms of flame stabilization. Particular focus is placed on determining essential factors for design considerations of the system with improved flame stability and combustion characteristics so that the system operates more efficiently.

#### 2. Computational details

Computational fluid dynamics is the analysis and prediction of fluid flows and heat transfer using a computer model [17, 18]. It may be used to predict the flows of fluids through a heat exchanger, or into the cylinder head of a diesel engine, or through a valve or a mixing vessel for example [19, 20]. The first stage involves constructing a numerical model of the structure around or through which the flows are occurring, this being similar to the process of computer aided design. It is also necessary to provide to the numerical model the nature of the fluid flow as it enters the structure. The second stage is to a perform the computational fluid dynamics modelling for that structure and that input flow, this typically being performed in an iterative manner. The final stage is to convert the resulting flow information into an output form, for example a graphical representation showing the flow paths. Highly sophisticated software is now available for performing these activities, which enables a skilled user to model fluid flows and heat transfer in or around any conceivable structure.

To gain a better understanding of the system, computational fluid dynamics simulations for propane-air combustion over a catalyst are performed. The insights gained from homogeneous combustion are not necessarily applicable to the catalytic systems studied in this work. For example, temperatures in gas-phase combustion are too high in comparison to those in catalytic combustion [21, 22]. Furthermore, while gaseous radicals are typically quenched on walls, causing flame extinction, catalysts serve mainly to form radicals that drive surface chemistry [23, 24]. Heat transfer is also different. For gaseous micro-burners, heat transfer of the heat generated in the gas phase to the walls can be slow in comparison to the chemistry time scale [25, 26]. On the other hand, the heat is liberated on the wall in catalytic microreactors, accelerating heat transfer within the walls [27, 28]. As a result of such differences, a model incorporating catalytic combustion is developed.

The specific heat, viscosity, and thermal conductivity are calculated using a mass fraction weighted average of the species properties. The species-specific heats are computed using a piecewise polynomial fit of the temperature. The species viscosities and thermal conductivity are determined from kinetic theory. Multicomponent diffusion is considered in this system, where the binary diffusion coefficients are determined from kinetic theory. A one-step, irreversible, catalytic reaction mechanism is used to model the propane-air catalytic combustion chemistry. Nonuniform node spacing is employed in this work, with more nodes in the reaction zone. The number of nodes varies depending on dimensions. Meshes in excess of 200,000 nodes are utilized for the largest dimensions. Typical wall node spacing is 50 microns in the axial direction and 20 microns in the transverse direction, where the temperature does not vary. Only a few nodes are placed in the transverse direction within the wall. However, as the mesh is nonuniform, these are just representative values.

The conservation equations were solved implicitly with a two-dimensional steady-state segregated solver using an under-relaxation method. The segregated solver first solves the momentum equation, then the continuity equation, and then updates the pressure and mass flow rate. The pressure is discretized using a Standard method. The pressure-velocity coupling is discretized using the SIMPLE method. The momentum, species, and energy equations are discretized using a two-order upwind approximation. The conservation equations are then checked for convergence. Convergence is determined from the residuals of the conservation equations as well as the difference between subsequent iterations of the solution. When parallel processing is used, the message passing interface is used to transmit information between nodes. In order to achieve convergence as well as compute extinction points, natural parameter continuation is implemented. The calculation time of each

computational fluid dynamics simulation varies between several hours and several days, depending on the difficulty of the problem.

Boundary conditions consist of flow inlets and exit boundaries, wall, repeating, and pole boundaries, and internal face boundaries. ANSYS FLUENT has a wide range of boundary conditions that permit flow to enter and exit the solution domain. Recommendations for determining inlet values of the turbulence parameters are provided. ANSYS FLUENT provides ten types of boundary zone types for the specification of flow inlets and exits: velocity inlet, pressure inlet, mass flow inlet, pressure outlet, pressure far-field, outflow, inlet vent, intake fan, outlet vent, and exhaust fan. The inlet and exit boundary condition options in ANSYS FLUENT are as provided follows:

- Velocity inlet boundary conditions are used to define the velocity and scalar properties of the flow at inlet boundaries.
- Pressure inlet boundary conditions are used to define the total pressure and other scalar quantities at flow inlets.
- Mass flow inlet boundary conditions are used in compressible flows to prescribe a mass flow rate at an inlet. It is not necessary to use mass flow inlets in incompressible flows because when density is constant, velocity inlet boundary conditions will fix the mass flow. Like pressure and velocity inlets, other inlet scalars are also prescribed.
- Pressure outlet boundary conditions are used to define the static pressure at flow outlets and also other scalar variables, in case of backflow. The use of a pressure outlet boundary condition instead of an outflow condition often results in a better rate of convergence when backflow occurs during iteration.
- Pressure far-field boundary conditions are used to model a free-stream compressible flow at infinity, with free-stream Mach number and static conditions specified. This boundary type is available only for compressible flows.
- Outflow boundary conditions are used to model flow exits where the details of the flow velocity and pressure are not known prior to solution of the flow problem. They are appropriate where the exit flow is close to a fully developed condition, as the outflow boundary condition assumes a zero streamwise gradient for all flow variables except pressure. They are not appropriate for compressible flow calculations.
- Inlet vent boundary conditions are used to model an inlet vent with a specified loss coefficient, flow direction, and ambient total pressure and temperature.
- Intake fan boundary conditions are used to model an external intake fan with a specified pressure jump, flow direction, and ambient total pressure and temperature.
- Outlet vent boundary conditions are used to model an outlet vent with a specified loss coefficient and ambient static pressure and temperature.
- Exhaust fan boundary conditions are used to model an external exhaust fan with a specified pressure jump and ambient static pressure.

Pressure inlet boundary conditions are used to define the fluid pressure at flow inlets, along with all other scalar properties of the flow. They are suitable for both incompressible and compressible flow calculations. Pressure inlet boundary conditions can be used when the inlet pressure is known but the flow rate and velocity is not known. This situation may arise in many practical situations, including buoyancy-driven flows. Pressure inlet boundary conditions can also be used to define a "free" boundary in an external or unconfined flow.

Velocity inlet boundary conditions are used to define the flow velocity, along with all relevant scalar properties of the flow, at flow inlets. In this case, the total or stagnation pressure is not fixed but will rise in response to the computed static pressure to whatever value is necessary to provide the prescribed velocity distribution. This boundary condition is intended for incompressible flows, and its

use in compressible flows will lead to a nonphysical result because it allows stagnation conditions to float to any level. In special instances, a velocity inlet may be used in ANSYS FLUENT to define the flow velocity at flow exits. In such cases, overall continuity is maintained in the domain.

Mass flow boundary conditions can be used in ANSYS FLUENT to provide a prescribed mass flow rate or mass flux distribution at an inlet. As with a velocity inlet, specifying the mass flux permits the total pressure to vary in response to the interior solution. This is in contrast to the pressure inlet boundary condition, where the total pressure is fixed while the mass flux varies. However, unlike a velocity inlet, the mass flow inlet is equally applicable to incompressible and compressible flows. A mass flow inlet is often used when it is more important to match a prescribed mass flow rate than to match the total pressure of the inflow stream. A mass flow inlet boundary condition can also be used as an outflow by specifying the flow direction away from the solution domain.

Wall boundary conditions are used to bound fluid and solid regions. In viscous flows, the no-slip boundary condition is enforced at walls by default, but a tangential velocity component can be specified in terms of the translational or rotational motion of the wall boundary or a "slip" wall can be modelled by specifying shear. The shear stress and heat transfer between the fluid and wall are computed based on the flow details in the local flow field. Wall boundaries can be either stationary or moving. The stationary boundary condition specifies a fixed wall, whereas the moving boundary condition can be used to specify the translational or rotational velocity of the wall, or the velocity components.

Pressure outlet boundary conditions require the specification of a static pressure at the outlet boundary. The value of the specified static pressure is used only while the flow is subsonic. Should the flow become locally supersonic, the specified pressure will no longer be used; pressure will be extrapolated from the flow in the interior. All other flow quantities are extrapolated from the interior. A set of "backflows" conditions is also specified should the flow reverse direction at the pressure outlet boundary during the solution process. Convergence difficulties will be minimized if realistic values are specified for the backflow quantities. Several options in ANSYS FLUENT exist, where a radial equilibrium outlet boundary condition can be used, and a target mass flow rate for pressure outlets can be specified. One of the options that may be used at pressure outlets is non-reflecting boundary conditions. This option is only available when the density-based solver and ideal gas law are used.

Pressure far-field conditions are used in ANSYS FLUENT to model a free-stream condition at infinity, with free-stream Mach number and static conditions being specified. The pressure far-field boundary condition is often called a characteristic boundary condition, since it uses characteristic information to determine the flow variables at the boundaries. Note the following limitations and restrictions when using pressure far-field boundary conditions:

- This boundary condition is applicable only when the density is calculated using the ideal-gas law. Using it for other flows is not permitted.
- It is incompatible with the multiphase models that are available with the pressure-based solver.
- It cannot be applied to flows that employ constant density, the real gas model, and the wet steam model, which are available in the density-based solver.

Outflow boundary conditions in ANSYS FLUENT are used to model flow exits where the details of the flow velocity and pressure are not known prior to solving the flow problem. Note that outflow boundaries cannot be used in the following cases:

- If a problem includes pressure inlet boundaries; use pressure outlet boundary conditions instead.
- Modelling compressible flow.
- Modelling unsteady flows with varying density, even if the flow is incompressible.
- With the multiphase models.

The zero-diffusion flux condition applied at outflow cells means that the conditions of the outflow plane are extrapolated from within the domain and have no impact on the upstream flow. The

extrapolation procedure used by ANSYS FLUENT updates the outflow velocity and pressure in a manner that is consistent with a fully-developed flow assumption, when there is no area change at the outflow boundary. The zero-diffusion flux condition applied by ANSYS FLUENT at outflow boundaries is approached physically in fully-developed flows. Fully-developed flows are flows in which the flow velocity profile and profiles of other properties such as temperature is unchanging in the flow direction. It is important to note that gradients in the cross-stream direction may exist at an outflow boundary. Only the diffusion fluxes in the direction normal to the exit plane are assumed to be zero. Note that convergence may be affected if there is recirculation through the outflow boundary at any point during the calculation, even if the final solution is not expected to have any flow re-entering the domain. This is particularly true of turbulent flow simulations.

Symmetry boundary conditions are used when the physical geometry of interest, and the expected pattern of the flow solution, have mirror symmetry. They can also be used to model zero-shear slip walls in viscous flows. ANSYS FLUENT assumes a zero flux of all quantities across a symmetry boundary. There is no convective flux across a symmetry plane: the normal velocity component at the symmetry plane is thus zero. There is no diffusion flux across a symmetry plane: the normal gradients of all flow variables are thus zero at the symmetry plane. The symmetry boundary condition can therefore be summarized as follows: zero normal velocity at a symmetry plane and zero normal gradients of all variables at a symmetry plane. As stated above, these conditions determine a zero flux across the symmetry plane, which is required by the definition of symmetry. Since the shear stress is zero at a symmetry boundary, it can also be interpreted as a "slip" wall when used in viscous flow calculations.

Periodic boundary conditions are used when the physical geometry of interest and the expected pattern of the flow solution have a periodically repeating nature. Two types of periodic conditions are available in ANSYS FLUENT. The first type does not allow a pressure drop across the periodic planes. The second type allows a pressure drop to occur across translationally periodic boundaries. ANSYS FLUENT treats the flow at a periodic boundary as though the opposing periodic plane is a direct neighbour to the cells adjacent to the first periodic boundary. Thus, when calculating the flow through the periodic boundary adjacent to a fluid cell, the flow conditions at the fluid cell adjacent to the opposite periodic plane are used.

An important step in the setup of the model is to define the materials and their physical properties. Properties may be temperature-dependent and composition-dependent, with temperature dependence based on a polynomial, piecewise-linear, or piecewise-polynomial function and individual component properties computed via kinetic theory. For solid materials, only density, thermal conductivity, and heat capacity are defined. Material properties can be defined as functions of temperature. When kinetic theory is applied to calculation of a fluid's thermal conductivity only, no inputs are required. For species transport calculations, there are two ways to model the diffusion of chemical species. For most applications the Fick's law approximation is adequate, but for some applications, the full multicomponent diffusion model is recommended. Mass diffusion coefficients are used to compute the diffusion flux of a chemical species in a laminar flow using Fick's law. A careful treatment of chemical species diffusion in the species transport and energy equations is important when details of the molecular transport processes are significant. As one of the laminar-flow diffusion models, ANSYS FLUENT has the ability to model full multicomponent species transport. For multicomponent systems it is not possible, in general, to derive relations for the diffusion fluxes containing the gradient of only one component. Here, the Maxwell-Stefan equations will be used to obtain the diffusive mass flux. This will lead to the definition of generalized Fick's law diffusion coefficients.

### 3. Results and discussion

The contour plots of propane mole fraction in the flow tube reactor are illustrated in Figure 1 in which sustained catalytically-supported thermal combustion occurs without formation of significant amounts of nitrogen oxides. Combustion encompasses a great variety of phenomena with wide application. Matrix stabilized porous burner technology is an advanced combustion method in which a mixture of fuel and oxidizer is burned within a solid porous medium, as opposed to open, burnerstabilized flames such as that on a Bunsen burner. The advantages of porous burners over that of burnerstabilized flames is mainly due to the thermal feedback effect provided by the solid medium through which heat conduction in the upstream direction results in preheating the incoming reactants. The process yields a flame temperature higher than the equilibrium adiabatic value achievable by the fueloxidizer mixture in the absence of a porous medium. This process is called super-adiabatic combustion. The heat transfer can be further enhanced by increasing the surface area to volume ratio of the porous medium and by the increased mixing due to turbulence generated in the porous structure. Therefore, porous burners help to sustain lean flames of fuel-oxidizer mixtures lower than the conventionally known lean flammability limit, and can also be used for burning low-calorific gases. Applications of porous burner technology include power generation via thermoelectric devices, small scale heating purposes, and combustion of low-calorific value landfill-seepage gases.





Figure 1. Contour plots of propane mole fraction in the flow tube reactor in which sustained catalytically-supported thermal combustion occurs without formation of significant amounts of nitrogen oxides.

The streamwise profiles of propane mole fraction in the flow tube reactor are presented in Figure 2 in which sustained catalytically-supported thermal combustion occurs without formation of significant amounts of nitrogen oxides. In a gas turbine engine, inlet air is continuously compressed, mixed with fuel in an inflammable proportion, and then contacted with an ignition source to ignite the mixture which will then continue to burn. The heat energy thus released then flows in the combustion gases to a turbine where it is converted to rotary energy for driving equipment such as an electrical generator. The combustion gases are then exhausted to atmosphere after giving up some of their remaining heat to the incoming air provided from the compressor. In order to meet the emission level requirements, for industrial low emission gas turbine engines, staged combustion is required in order to minimise the quantity of the oxides of nitrogen produced. The fundamental way to reduce emissions of nitrogen

oxides is to reduce the combustion reaction temperature and this requires premixing of the fuel and all the combustion air before combustion takes place. It is known to provide gas turbine engine combustion chambers which have staged combustion to minimise nitrous oxide emissions. It is also known to provide gas turbine engine combustion chambers which have a plurality of catalytic combustion zones arranged in series to minimise nitrous oxide emissions. In the operation of a conventional combustion turbine, intake air from the atmosphere is compressed and heated by rotary action of a multi-vaned compressor component and caused to flow to a plurality of combustor components where fuel is mixed with the compressed air and the mixture ignited and burned. The heat energy thus released then flows in the combustion gases to the turbine component where it is converted into rotary energy for driving equipment, such as for generating electrical power or for running industrial processes. The combustion gases are finally exhausted from the turbine component back to the atmosphere.



Figure 2. Streamwise profiles of propane mole fraction in the flow tube reactor in which sustained catalytically-supported thermal combustion occurs without formation of significant amounts of nitrogen oxides.

The contour plots of temperature in the flow tube reactor are illustrated in Figure 3 in which sustained catalytically-supported thermal combustion occurs without formation of significant amounts of nitrogen oxides. Various schemes have been explored to adapt combustion turbines for the aforementioned uses without exceeding the limits of nitrogen oxides emission [29, 30]. The use of catalytic combustion is a promising approach because it can occur at about 2300 to 2500 degrees in the Fahrenheit scale to produce a high turbine inlet temperature for turbine operating efficiency without any significant side effect nitrogen oxides generation from reactions between nitrogen and oxygen which occurs at temperatures over 3000 degrees in the Fahrenheit scale. In contrast, conventional flame combustion at about 4500 degrees in the Fahrenheit scale results in the generation of nitrogen oxides which typically exceeds the limits set in more restrictive areas [31, 32]. The combustion catalyst has several desirable characteristics: they are capable of minimizing nitrogen oxides emission and improving the pattern factor. However, one of their limitations is that their maximum operating temperature tends to be only marginally acceptable as a turbine inlet temperature. This limitation is inherent in the way the typical catalytic combustor operates. Catalysts initiate the combustion reaction at their surfaces and at temperatures lower than normal ignition temperature.

is initiated, it continues in the gas stream and persists beyond the catalyst in the form of afterburning. Simultaneously, the catalyst substrate temperature increases, resulting in an accelerated reaction which moves the reaction zone further upstream in the catalyst. The result may be damage of the catalyst and catalyst substrate if the fuel-air ratio is such as to give an excessive catalyst outlet temperature. Presently available catalysts have the capability of extended operation at about 2289 degrees in the Fahrenheit scale. However, a gas turbine inlet temperature of around 2500 degrees in the Fahrenheit scale is desired. Thus, given the aforementioned current catalyst temperature limits, the catalyst is clearly incapable of providing such a temperature at the gas turbine inlet.



Temperature contour plots

Figure 3. Contour plots of temperature in the flow tube reactor in which sustained catalyticallysupported thermal combustion occurs without formation of significant amounts of nitrogen oxides.

The streamwise profiles of temperature in the flow tube reactor are presented in Figure 4 in which sustained catalytically-supported thermal combustion occurs without formation of significant amounts of nitrogen oxides. Combustion, with rare exceptions, is a complex chemical process involving many steps that depend on the properties of the combustible substance. The mechanisms of combustion of hydrocarbons and of other organic compounds are known in general outline only. The overall mechanism of hydrocarbon combustion is complicated by the diversity of molecules and radicals involved. Quantities of air greatly in excess of stoichiometric amounts are normally compressed and utilized to keep the combustor liner cool and dilute the combustor exhaust gases so as to avoid damage to the turbine nozzle and blades. Generally, primary sections of the combustor are operated near stoichiometric conditions which produce combustor gas temperatures up to approximately four thousand degrees Fahrenheit [33, 34]. Further along the combustor, secondary air is admitted which raises the air-fuel ratio and lowers the gas temperatures so that the gases exiting the combustor are in the range of two thousand degrees Fahrenheit. The fuel injection pressure can vary and is typically six hundred pounds per square inch for full power and as low as sixty pounds per square inch to one hundred pounds per square inch for idle conditions. It is well established that nitrogen oxides formation is thermodynamically favoured at high temperatures [35, 36]. Since the nitrogen oxides formation reaction is so highly temperature dependent, decreasing the peak combustion temperature can provide an effective means of reducing nitrogen oxides emissions from gas turbine engines as can limiting the residence time of the combustion products in the combustion zone. Operating the combustion process in a very lean condition, namely high excess air, is one of the simplest ways of achieving lower

temperatures and hence lower nitrogen oxides emissions. Very lean ignition and combustion, however, inevitably result in incomplete combustion and the attendant emissions which result therefrom. In addition, combustion processes cannot be sustained at these extremely lean operating conditions. Lean ignition and incomplete combustion have also been encountered in internal combustion engines and catalysts have been utilized to promote and complete the combustion process. In a catalytic combustor, fuel is burned at relatively low temperatures in the range of from several hundred degrees Fahrenheit to approximately two thousand degrees Fahrenheit. While emissions can be reduced by combustion at these temperatures, the utilization of catalytic combustion has been limited by the amount of catalytic surface required to achieve the desired reaction and the attendant undesirable pressure drop across the catalytic surface. Also, the time to bring the catalytic combustor up to operating temperature continues to be of concern.



Figure 4. Streamwise profiles of temperature in the flow tube reactor in which sustained catalyticallysupported thermal combustion occurs without formation of significant amounts of nitrogen oxides.

The contour plots of reaction rate in the flow tube reactor are illustrated in Figure 5 in which sustained catalytically-supported thermal combustion occurs without formation of significant amounts of nitrogen oxides. The combustion spreads from the ignition source to the adjacent layer of gas mixture; in turn, each point of the burning layer serves as an ignition source for the next adjacent layer, and so on. Combustion terminates when equilibrium is achieved between the total heat energies of the reactants and the total heat energies of the products. Most reactions terminate when what is called thermal equilibrium has been attained, namely when the energy of the reactants equals the energy of the products. The use of a catalytic combustor offers the advantage that all of the fuel can be oxidized therein, resulting in ultra-low nitrogen oxides emissions and low carbon monoxide and unburned hydrocarbon levels. The conventional gas turbine combustor, as used in a gas turbine power generating system, requires a mixture of fuel and air which is ignited and combusted uniformly [37, 38]. Generally, the fuel injected from a fuel nozzle into the inner tube of the combustor is mixed with air for combustion, fed under pressure from the air duct, ignited by a spark plug and combusted. The gas that results is lowered to a predetermined turbine inlet temperature by the addition of cooling air and dilutant air, then injected through a turbine nozzle into a gas turbine. Exhaust gases produced by combusting hydrocarbon fuels can contribute to atmospheric pollution. This occurrence is attributed to

the development of localized high temperature zone. Exhaust gases typically contain many undesirable pollutants such as nitric oxide and nitrogen dioxide, which are frequently grouped together as nitrogen oxides, unburned hydrocarbons, and carbon monoxide. In conventional thermal combustion fuel and air in inflammable proportions are contacted with an ignition source, for example, a spark, to ignite the mixture which will then continue to burn [39, 40]. Flammable mixtures of most fuels are normally burned at relatively high temperatures, namely in the order of about 3300 degrees in the Fahrenheit scale and above, which inherently results in the formation of substantial emissions of nitrogen oxides. In the case of gas turbine combustors, the formation of nitrogen oxides can be greatly reduced by limiting the residence time of the combustion products in the combustion zone. However, even under these circumstances' undesirable quantities of nitrogen oxides are nevertheless produced. In addition, limiting such residence time makes it difficult to maintain stable combustion even after ignition.



Reaction rate contour plots

Figure 5. Contour plots of reaction rate in the flow tube reactor in which sustained catalyticallysupported thermal combustion occurs without formation of significant amounts of nitrogen oxides.

The streamwise profiles of reaction rate in the flow tube reactor are presented in Figure 6 in which sustained catalytically-supported thermal combustion occurs without formation of significant amounts of nitrogen oxides. In purely catalytic combustion systems, there is little or no nitrogen oxides formed in a system which burns the fuel at relatively low temperatures [41, 42]. Catalytic combustion heretofore has been generally regarded as having limited practicality in providing a source of power as a consequence of the need to employ impractically large amounts of catalyst so as to make a system unduly large and cumbersome. Consequently, catalytic combustion has been limited generally to such operations as treating tail gas streams. For any given catalyst and set of reaction conditions, as the temperature in catalytic combustion is initially increased, the reaction rate is also increased. This rate of increase is exponential with temperature [43, 44]. As the temperature is raised further, the reaction rate then passes through a transition zone where the limiting parameters determining reaction rate shift from catalytic to mass transfer. When the catalytic rate increases to such an extent that the reactants cannot be transferred to the catalytic surface fast enough to keep up with the catalytic reaction rate, the reaction shifts to mass transfer control, and the catalytic reaction rate levels off regardless of further temperature increases. The reaction is mass transfer limited. In mass transfer controlled catalytic reactions, one cannot distinguish between a more active catalyst and a less active catalyst because the intrinsic catalyst

activity is not determinative of the rate of reaction. Regardless of any increase in catalytic activity above that required for mass transfer control, a greater catalytic conversion rate cannot be achieved for the same set of conditions. Because of this limitation, in order to increase the conversion rate for any given system, it appears essential either to increase the amount of catalyst surface or to increase the rate of mass transfer of reactants to the surface. The former, for practical combustion systems, will require either a catalyst size of such magnitude as to be unwieldy or a catalyst configuration which results in increased specific pressure drop and which would require unwieldy geometry to hold a total pressure drop constant. On the other hand, increasing the rate of mass transfer of reactants to the catalytic surfaces will result in increased pressure drop and consequently a substantial loss of energy; sufficient pressure drop may not even be available to provide the desired rate of reaction. Quite obviously, these approaches, while theoretically possible, are quite impractical.



Figure 6. Streamwise profiles of reaction rate in the flow tube reactor in which sustained catalyticallysupported thermal combustion occurs without formation of significant amounts of nitrogen oxides.

The contour plots of velocity in the flow tube reactor are illustrated in Figure 7 in which sustained catalytically-supported thermal combustion occurs without formation of significant amounts of nitrogen oxides. In addition to chemical reactions, physical processes that transfer mass and energy by diffusion or convection occur in gaseous combustion. In the absence of external forces, the rate of component diffusion depends upon the concentration of the constituents, pressure, and temperature changes, and on diffusion coefficients, a measure of the speed of diffusion. It is possible to achieve essentially adiabatic combustion in the presence of a catalyst at a reaction rate many times greater than the mass transfer limited rate. That is that catalytically-supported, thermal combustion surmounts the mass transfer limitation. If the operating temperature of the catalyst is increased substantially into the mass transfer limited region, the reaction rate again begins to increase exponentially with temperature. This is an apparent contradiction of catalytic technology and the laws of mass transfer kinetics [45, 46]. The phenomena may be explained by the fact that the catalyst surface and the gas layer near the catalyst surface are above a temperature at which thermal combustion occurs at a rate higher than the catalytic rate, and the temperature of the catalyst surface is above the instantaneous autoignition temperature of the fuel-air admixture [47, 48]. The fuel molecules entering this layer spontaneously burn without transport to the catalyst surface. As combustion progresses, it is believed that the layer becomes deeper.

The total gas is ultimately raised to a temperature at which thermal reactions occur in the entire gas stream rather than only near the surface of the catalyst. Once this stage is reached within the catalyst, the thermal reactions continue even without further contact of the gas with the catalyst as the gas passes through the combustion zone. The term "instantaneous auto-ignition temperature" for a fuel-air admixture as used herein is defined to mean that the temperature at which the ignition lag of the fuel-air mixture entering the catalyst is negligible relative to the residence time in the combustion zone of the mixture undergoing combustion. Catalytically-supported thermal combustion is achieved by contacting at least a portion of the carbonaceous fuel intimately admixed with air with a solid oxidation catalyst having an operating temperature substantially above the instantaneous auto-ignition temperature of the fuel-air admixture. At least a portion of the fuel is combusted under essentially adiabatic conditions. Combustion is characterized by the use of a fuel-air admixture having an adiabatic flame temperature substantially above the instantaneous auto-ignition temperature but below a temperature that would result in any substantial formation of oxides of nitrogen.



Velocity contour plots

Figure 7. Contour plots of velocity in the flow tube reactor in which sustained catalytically-supported thermal combustion occurs without formation of significant amounts of nitrogen oxides.

The streamwise profiles of temperature in the flow tube reactor are presented in Figure 8 with different diameters and at different velocities in which sustained catalytically-supported thermal combustion occurs without formation of significant amounts of nitrogen oxides. The adiabatic flame temperature is determined at catalyst inlet conditions. The resulting effluent is characterized by high thermal energy useful for generating power and by low amounts of atmospheric pollutants. Where desired, combustible fuel components, for example, un-combusted fuel or intermediate combustion products contained in the effluent from the catalytic zone, or fuel-air admixture which has not passed through a catalytic zone, may be combusted in a thermal zone following the catalytic zone. The maximum acceptable face velocity is established by the given values of the catalyst operating temperature, pressure, stoichiometry and preheating temperature. Further increases in velocity result in blowouts. Lower limits on the velocity of the mixture are set by the mixture flame speeds in order to avoid flash-back and are determined by proper selection of the combustor cross-sectional area. The maximum achievable velocity depends on flow conditions and catalyst parameters such as type, monolith cell size, and web thickness. The minimum residence time in the combustor is a function both of the face velocity and the reactor length. Since the velocity increases as the fuel burns and the temperature rises, the face velocity establishes a minimum velocity level just upstream of the catalyst

bed. The foregoing description has been concerned with a catalytic combustor operating at or near its design point. However, gas turbine operations require a much broader range of inlet temperatures and fuel-air ratios. In a typical situation, the fuel-air ratio limits are approximately 0.005-0.025 and the inlet temperature ranges from ambient at ignition to about 600 degrees in the Fahrenheit scale on the simple cycle machines. Catalyst ignition does not readily occur, however, at inlet temperatures which are less than about 1000 degrees in the Fahrenheit scale for most catalysts. In order to overcome this problem, the most common solution is to employ a pilot burner for ignition and acceleration and by waiting until the machinery is under part load to initiate catalytic burning. The conventional catalytic gas turbine combustor generally employs a pilot zone disposed upstream of the catalyst. This arrangement has two principal disadvantages. First, the catalyst bed is subjected to thermal shock when the pilot zone is ignited. Secondly, when the catalytic operation is initiated, the pilot burner must be extinguished. One method of extinguishing the pilot burner is to interrupt the fuel flow to the pilot. An alternative method is to reduce the fuel supply to the pilot zone while adding fuel to the catalytic main stage.



Figure 8. Streamwise profiles of temperature in the flow tube reactor with different diameters and at different velocities in which sustained catalytically-supported thermal combustion occurs without formation of significant amounts of nitrogen oxides.

#### 4. Conclusions

This study is focused mainly upon the essential combustion characteristics of propane-air mixtures in a micro-scale system with a heat-recirculating structure. Numerical simulations are performed to gain a greater understanding of the mechanisms of flame stabilization. The essential factors affecting flame stability and combustion characteristics are determined in order to obtain design insights. The main conclusions are summarized as follows:

- In order to meet the emission level requirements, for industrial low emission gas turbine engines, staged combustion is required in order to minimise the quantity of the oxides of nitrogen produced.
- The combustion catalyst has several desirable characteristics: they are capable of minimizing nitrogen oxides emission and improving the pattern factor.
- Operating the combustion process in a very lean condition, namely high excess air, is one of the

simplest ways of achieving lower temperatures and hence lower nitrogen oxides emissions.

- The use of a catalytic combustor offers the advantage that all of the fuel can be oxidized therein, resulting in ultra-low nitrogen oxides emissions and low carbon monoxide and unburned hydrocarbon levels.
- In mass transfer controlled catalytic reactions, one cannot distinguish between a more active catalyst and a less active catalyst because the intrinsic catalyst activity is not determinative of the rate of reaction.
- It is possible to achieve essentially adiabatic combustion in the presence of a catalyst at a reaction rate many times greater than the mass transfer limited rate.
- The maximum achievable velocity depends on flow conditions and catalyst parameters such as type, monolith cell size, and web thickness.

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