

Design of gas turbine combustors for the reduction of nitrogen oxides emissions by heterogeneous catalysis

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Abstract

Flammable mixtures of most fuels are normally burned at relatively high temperatures, 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. Additionally, limiting such residence time makes it difficult to maintain stable combustion even after ignition. The present study relates to the design of gas turbine combustors for the reduction of nitrogen oxides emissions by heterogeneous catalysis. Steady-state simulations are performed using computational fluid dynamics. The fluid viscosity, specific heat, and thermal conductivity are calculated from a mass fraction weighted average of species properties, and the specific heat of each species is calculated using a piecewise polynomial fit of temperature. Natural parameter continuation is performed by moving from one stationary solution to another. Particular emphasis is placed upon the sustained combustion of at least a portion of fuel under essentially adiabatic conditions at a rate which surmounts the mass transfer limitation. The results indicate that 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. Flammable mixtures of carbonaceous fuels normally burn at relatively high temperatures, and substantial amounts of nitrogen oxides inevitably form if nitrogen is present. Complete catalytic combustion of a target species can only occur when oxygen gas is found in molar stoichiometric excess; a condition which is easily met when the target species is present in trace quantity in air. In combustion systems utilizing a catalyst, there is little or no nitrogen oxides formed in a system which burns the fuel at relatively low temperatures. In the mass transfer limited zone, the reaction rate cannot be increased by increasing the activity of the catalyst because catalytic activity is not determinative of the reaction rate. Among the unique advantages of the catalytically supported thermal combustion in the presence of a catalyst is the fact that mixtures of fuel and air which are too fuel-lean for ordinary thermal combustion can be burned efficiently.

Keywords: Heterogeneous catalysis; Nitrogen oxides; Gas turbines; Flammable mixtures; Thermodynamic properties; Combustion phenomena

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1. Introduction

Combustion is a chemical reaction between substances, usually including oxygen and usually accompanied by the generation of heat and light in the form of flame [1, 2]. The rate or speed at which the reactants combine is high, in part because of the nature of the chemical reaction itself and in part because more energy is generated than can escape into the surrounding medium, with the result that the temperature of the reactants is raised to accelerate the reaction even more [3, 4]. Combustion encompasses a great variety of phenomena with wide application in industry, the sciences, professions, and the home, and the application is based on knowledge of physics, chemistry, and mechanics; their interrelationship becomes particularly evident in treating flame propagation [5, 6]. Combustion, with rare exceptions, is a complex chemical process involving many steps that depend on the properties of the combustible substance. It is initiated by external factors such as heat, light, and sparks [7, 8]. The reaction sets in as the mixture of combustibles attains the ignition temperature. 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 [9, 10]. 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 complexity of the combustion reaction mechanism and the rapidly varying temperatures and concentrations in the mixture make it difficult and often impossible to derive an equation that would be useful for predicting combustion phenomena over wide temperature and concentration ranges. Instead, use is made of empirical expressions derived for specific reaction conditions [11, 12]. 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 [13, 14]. The latter are either measured or calculated in terms of the kinetic theory of gases. The process of diffusion is of great importance in combustion reactions, in flames, that is, in gaseous mixtures, and in solids or liquids. Diffusion heat transfer follows a law stating that the heat flux is proportional to the temperature gradient [15, 16]. The

coefficient of proportionality, called the thermal conductivity coefficient, is also measured or calculated in terms of the kinetic theory of gases, like the diffusion coefficient. Convective transport of mass and energy may be accounted for by buoyant forces, external forces, and turbulent and eddy motions. Convection is in the main responsible for the mixing of gases. Flame combustion is most prominent with fuels that have been premixed with an oxidant, either oxygen or a compound that provides oxygen, for the reaction [17, 18]. The temperature of flames with this mixture is often several thousand degrees. The chemical reaction in such flames occurs within a narrow zone several micrometers thick [19, 20]. This combustion zone is usually called the flame front. Dilution of the burning mixture with an inert gas, such as helium or nitrogen, lowers the temperature and, consequently, the reaction rate [21, 22]. Great amounts of inert gas extinguish the flame, and the same result is achieved when substances that remove any of the active species are added to the flame [23, 24]. Conditions must be such that the flame is fixed at the burner nozzle or in the combustion chamber [25, 26]. This positioning is required in many practical uses of combustion. Various devices, such as pilot flames and recirculation methods, are designed for this purpose.

A number of combustion systems promote partial conversion of a fuel followed by complete combustion of that fuel in a downstream combustion zone [27, 28]. These methods generally comprise introducing a fuel and air mixture into a combustion zone wherein a portion of the fuel has been partially reacted prior to entering the combustion zone. Such partial reaction may be promoted chemically, catalytically, or by any other conventional means depending upon each particular application [29, 30]. As the partially reacted fuel and air mixture are introduced into a region of the combustion zone, as with a dump, a flame is established to promote complete combustion of the fuel within the fuel and air mixture. Flame stabilization is a common problem in these combustion systems [31, 32]. A flame will propagate through a fuel-air mixture only when certain conditions prevail. Initially, a minimum percentage of fuel must be present within the fuel-air mixture to make the fuel-air mixture flammable, namely the lean flammability limit. Similarly, a maximum percentage of fuel must be present within the fuel-air mixture wherein greater than this percentage will prevent burning, namely the rich flammability limit. The flammability range of a fuel-air mixture is that range of the percentage of fuel within the fuel-air mixture between the lean flammability limit and the rich flammability limit [33, 34]. The stoichiometry of a fuel-air mixture contributes to its flammability range [35, 36]. A stoichiometric fuel-air mixture composition contains sufficient oxygen for complete combustion thereby releasing all the latent heat of combustion of the fuel [37, 38]. The strength of a fuel-air mixture composition typically is expressed in terms of its equivalence ratio. The equivalence ratio is the actual fuel-air ratio divided by stoichiometric fuel-air ratio. For example, an equivalence ratio of one represents a stoichiometric fuel-air mixture composition. An equivalence ratio less than one represents a lean mixture and an equivalence ratio less great than one represents a rich mixture.

Pressure and temperature contribute to the flammability range of fuel-air mixtures. Typically, with increases in pressure, the rich flammability limit is extended thereby extending the flammability range of the fuel-air mixture. Temperature, on the other hand, partially defines the flammability range of fuel-air mixtures [39, 40]. The lowest temperature at which a flammable fuel-air mixture can be formed, based upon the vapor pressure of the fuel at atmospheric pressure, is the flash point of that fuel-air mixture [41, 42]. Within the flammability range of a fuel-air mixture, at temperatures exceeding the flash point of the fuel-air mixture, auto-ignition of the fuel vapor occurs. Auto-ignition generally occurs at or slightly above the stoichiometric fuel-air mixture composition. The time interval between the mixing of the fuel-air mixture such that it is combustible and the auto-ignition of that fuel-air mixture is known as the auto-ignition delay time [43, 44]. One reason that flame stabilization is required in combustion systems is to prevent the flame front from moving upstream from the combustion zone toward the source of fuel, namely a flashback. During a flashback event, the heat of combustion moves upstream and may damage numerous structures within the fuel and air mixing region of the combustor. Flashback may occur due to auto-ignition of a fuel-air mixture caused by a residence time of the fuel-air mixture in a region upstream of the combustion zone that exceeds the auto-ignition delay time of that fuel-air mixture at the temperature and pressure of that region [45, 46]. Flame stabilization also is dependent upon speed of the fuel-air mixture entering the combustion zone where propagation of the flame is desired [47, 48]. A sufficiently low velocity must be retained in the region where

the flame is desired in order to sustain the flame. A region of low velocity in which a flame can be sustained can be achieved by causing recirculation of a portion of the fuel-air mixture already burned thereby providing a source of ignition to the fuel-air mixture entering the combustion zone. However, the fuel-air mixture flow pattern, including any recirculation, is critical to achieving flame stability.

In gas turbines, compressed air enters the combustion chamber where it mixes with the fuel. The expanding combustion products impart their energy to the turbine blades. However, flammable mixtures of most fuels are normally burned at relatively high temperatures, 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. Additionally, limiting such residence time makes it difficult to maintain stable combustion even after ignition. The present study relates to the design of gas turbine combustors for the reduction of nitrogen oxides emissions by heterogeneous catalysis. More particularly, the present study is directed to an improved method for more efficiently operating a catalytically supported thermal combustion gas turbine system, and at the same time provide low emissions of unburned hydrocarbons, carbon monoxide, and nitrogen oxides. The present study is focused primarily upon at least a portion of the thermal combustion of the fuel takes place in the expansion zone of the gas turbine to counteract the cooling effect of the expansion of the gases. Natural parameter continuation is performed by moving from one stationary solution to another. A critical point is denoted as the solution to the problem when a turning point is reached. Knowledge of critical parameters gains a fundamental understanding of the essential factors affecting the stability of the catalytically supported thermal combustion process. The critical parameters are useful as the design guides associated with the gas turbine combustor. The principal quantitative characteristic of a flame is its normal, or fundamental, burning velocity, which depends on the chemical and thermodynamic properties of the mixture and on pressure and temperature, under given conditions of heat loss. The burning velocity value ranges from several centimeters to even tens of meters per second. The dependence of the burning velocity on molecular structure, which is responsible for fuel reactivity, is known for a great many fuel-air mixtures. Steady-state simulations are performed using computational fluid dynamics. The present study aims to provide an improved method for operating a gas turbine combustor by catalytically supported thermal combustion of carbonaceous fuel. Particular emphasis is placed upon the sustained combustion of at least a portion of fuel under essentially adiabatic conditions at a rate which surmounts the mass transfer limitation.

2. Methods

Computational fluid dynamics is an approach to solving fluid flow problems by solving models that include numerical methods and algorithms used to represent fluids. The methods and algorithms are generally solved by computers [49, 50]. The solution of the models can provide a simulation of an interaction of fluids with a structure defined by surfaces, each of which can be defined by boundary conditions within the model. The results, or solution, can be used to validate and improve designs, for example. Computational fluid dynamics is the analysis and prediction of fluid flows and heat transfer using a computer model. Computational fluid dynamics uses mathematical methods to solve problems that include fluid flow [51, 52]. It may be used to predict the flows of fluids through a heat exchanger, or through a valve or a mixing vessel for example [53, 54]. 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 perform the computational fluid dynamics modeling 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 [55, 56]. 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 [57, 58]. However, the very sophistication that enables the software to cope with any fluid flow problem, makes it expensive and also complex.

The gas turbine combustor configured to combust propane is represented physically in Figure 1 for the reduction of nitrogen oxides emissions by heterogeneous catalysis. The gas turbine combustor comprises a concentric annular channel, wherein the concentric annular channel further comprises an inner annular channel and an outer annular channel. A platinum catalyst is deposited only upon the interior surface of the inner channel, and the wall of the outer channel is chemically inert and catalytically inactive. The reactant stream flows through the catalytically-coated inner channel and the product stream flows out of the outer non-catalytic channel. Fuel is present for combustion in both the catalytic and non-catalytic channels. The concentrically arranged annular channel is 5.0 millimeters in inner channel length, 5.6 millimeters in outer channel length, 0.8 millimeters in innermost diameter, 2.6 millimeters in outermost diameter, 0.1 millimeters in catalyst layer thickness, and 0.2 millimeters in wall thickness, unless otherwise stated. The gas turbine combustor can have any dimension unless restricted by design requirements. All the walls have the same thickness. The spacing between the inner channel and the outer channel is 0.4 millimeters and remains constant. One of the potential problems associated with the gas turbine combustor, as with all micro-scale combustion systems, continues to be combustion stability. The scale of the gas turbine combustor is on the order of sub-millimeters, which is much smaller than the quenching distance of the combustible mixture in the absence of a catalyst. The quenching distance defines a critical dimension under which propagation of the propane flame is not possible. The quenching distance is approximately 2.5 millimeters, at which combustion cannot be sustained within the gas turbine combustor in the absence of a catalyst.

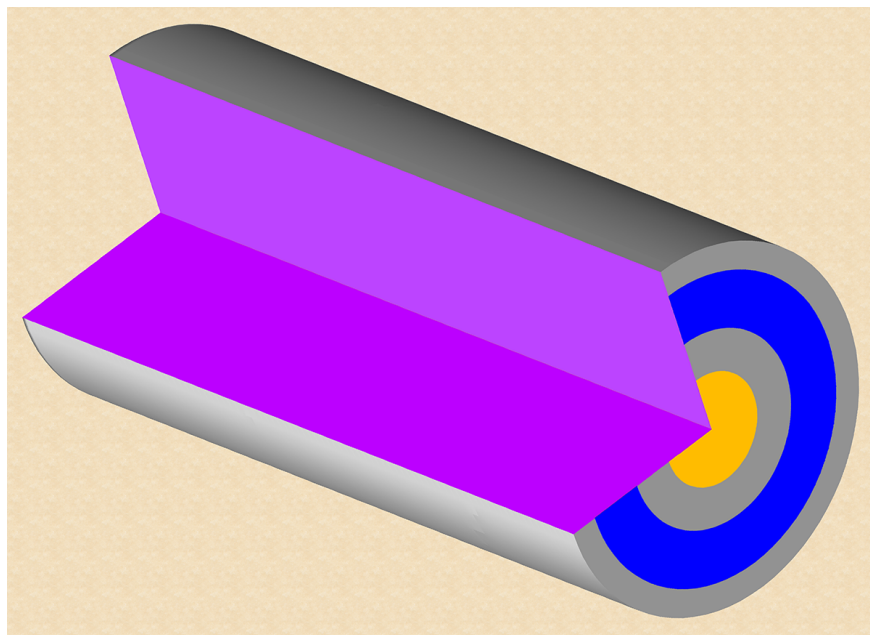


Figure 1. Physical representation of the gas turbine combustor configured to combust propane for the reduction of nitrogen oxides emissions by heterogeneous catalysis.

Complex modeling methods and algorithms are required for the gas turbine combustor due not only to the complex geometry but also the complex physicochemical processes involved. It is therefore essential to reduce the complexity of the model through use of certain simplifying assumptions. Steady-state analyses are performed, variations in reactor pressure and temperature are determined in accordance with the ideal gas law. The model is implemented in computational fluid dynamics software to obtain the solution of the problem. Computational fluid dynamics is used to describe a broader range of calculations for a wide variety of scientific and engineering applications. Thermodynamics is an important consideration in many of these applications. It relates internal energy to temperature, which affects the flow of heat. Further sources of heat include thermal radiation and chemical reactions, in particular combustion. Heat transfer

may involve conduction in solid materials, coupled with the fluid flow, known as conjugate heat transfer. A modern definition of computational fluid dynamics would be the prediction of fluid motion and forces by computation using numerical analysis, generally extended to include heat, thermodynamics, chemistry and solids. Numerical analysis provides many methods and algorithms that are suitable for computational fluid dynamics. The methods include finite volume, finite element and finite difference, which calculate the distributions of properties, for instance, pressure, velocity and temperature, over regions of space which are usually fixed. Alternative methods attribute properties to particles represented by points in space, whose motions are calculated. To perform the calculation first requires a description of the problem by the domain occupied by the fluid, equations that represent the fluid behavior in terms of properties such as pressure and velocity, and conditions at the boundary of the fluid domain and initially within the domain for the fluid properties.

The typical computational mesh for the fluid and solid of the gas turbine combustor configured to combust propane is illustrated schematically in Figure 2 for the reduction of nitrogen oxides emissions by heterogeneous catalysis. A computational fluid dynamics simulation begins with a solution domain which specifies a region of space of a particular geometric shape, in which fluid dynamics equations are solved. The process of mesh generation subdivides the solution domain into a mesh of small volumes. The computational mesh consists of about 600,000 nodes in total for the fluid and solid of the gas turbine combustor. A computational fluid dynamics analysis is carried out on the mesh. A mesh independence test is performed to assure independence of the solution to the problem. The specification of boundary conditions is one of the most challenging tasks in setting up a computational fluid dynamics simulation. The range of possible boundary conditions is endless, to cover all of the potential applications and physics. Velocity inlet boundary conditions are used to define the velocity properties of the flow at the inlet boundary of the fluid region. A uniform velocity profile is specified at the flow inlet. The temperature of the mixture is prespecified at the flow inlet. Under-relaxation is a general method used to improve solution convergence by limiting the amount that a variable changes during a solution step. The computational fluid dynamics calculations may take days in order to arrive at a reasonably accurate solution, using fine grids of the gas turbine combustor, due to the time-consuming nature of the model. Natural parameter continuation is performed by moving from one stationary solution to another. A critical point is denoted as the solution to the problem when a turning point is reached. Knowledge of critical parameters gains a fundamental understanding of the essential factors affecting the stability of the catalytically supported thermal combustion process. The critical parameters are useful as the design guides associated with the gas turbine combustor.

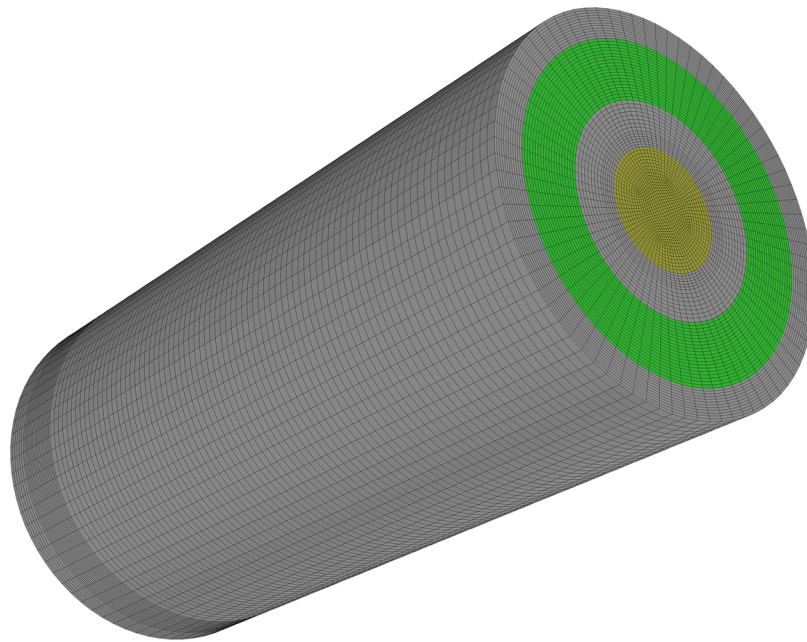


Figure 2. Schematic illustration of the typical computational mesh for the fluid and solid of the gas turbine combustor configured to combust propane for the reduction of nitrogen oxides emissions by heterogeneous catalysis.

Steady-steady simulations are performed, unless otherwise stated. The fluid density is calculated using the ideal gas law. The fluid viscosity, specific heat, and thermal conductivity are calculated from a mass fraction weighted average of species properties, and the specific heat of each species is calculated using a piecewise polynomial fit of temperature. The wall thermal conductivity and exterior convective heat loss coefficient are taken as independent parameters to understand how important thermal management is. The heat flux at the wall-fluid interface is computed using Fourier's law and continuity in temperature and heat flux links the fluid and solid phases. The left and right edges of the wall are assumed to be insulated. Newton's law of cooling is used at the outer edge of the wall. Computations are performed using meshes with varying nodal densities to determine the optimum node spacing and density that would give the desired accuracy and minimize computation time. As the mesh density increases, there is a convergence of the solution. The coarsest mesh fails to accurately capture the inflection point at the ignition point and the maximum temperature. Solutions obtained with meshes consisting of a few million nodes are reasonably accurate. The gas phase is assumed to be in quasi-steady-state. The walls are transiently modeled using energy diffusion and radiation. The gas phase is modeled using the forms of the momentum, energy, and species equations. To solve the conservation equations, a segregated solution solver with an under-relaxation method is used. The segregated solver first solves the momentum equations, then solves the continuity equation, and updates the pressure and mass flow rate. The energy and species equations are subsequently solved and convergence is checked. The latter is monitored through both the values of the residuals of the conservation equations and the difference between subsequent iterations of the solution. Numerical convergence is in general difficult because of the inherent stiffness of the chemistry as well as the disparity between the wall and the fluid heat conductivities. In order to assist convergence and compute extinction points, natural parameter continuation is implemented. The calculation time varies for these problems, depending on the initial guess as well as the parameter set.

3. Results and discussion

The propane mole fraction contour plots in the gas turbine combustor are illustrated in Figure 3 for the

reduction of nitrogen oxides emissions by heterogeneous catalysis. In order to meet the emission level requirements, for industrial low emission gas turbine engines, staged combustion is required in order to minimize the quantity of the oxides of nitrogen produced [59, 60]. 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 [61, 62]. The present design provides a gas turbine engine combustion chamber comprising a primary combustion zone and a secondary combustion zone downstream of the primary combustion zone. In this way, the temperatures in each stage can be reduced. Suitable temperature ranges in each stage are from 900 K to 1500 K, depending on the particular catalyst and support. An advantage of operating in this temperature range is that it is below the fixation temperature of nitrogen and consequently the combusted gases are free of nitrogen oxides. Additionally, catalytic combustion results in lower un-combusted fuel content. A further advantage of catalytic combustion is that it is possible to operate with minimum of air for combustion, namely excess oxygen in the combusted gases can be reduced almost to zero. The catalyst may be supported on a monolith. The preferred characteristics of the metallic monolith having a catalyst deposited thereon are that it presents low resistance to the passage of gases by virtue of its possession of a high ratio of open area to blocked area and that it has a high surface to volume ratio. Preferably, the metallic monolith is formed from one or more metals selected from the group comprising ruthenium, rhodium, palladium, iridium, and platinum. However, base metals may be used or base metal alloys which also contain a platinum group metal component may be used. Oxygen is the required element to support combustion. 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. In particular, if the operating temperature of the catalyst is increased substantially into the mass transfer limited region, the reaction rate again begins to increase rapidly with temperature. This is in apparent contradiction of the laws of mass transfer kinetics in catalytic reactions. The phenomenon may be explained by the fact that the temperature of the catalyst surface and the gas layer near the catalyst surface are above the instantaneous auto-ignition temperature of the mixture of fuel, air, and any inert gases and at a temperature at which thermal combustion occurs at a rate higher than the catalytic combustion rate. The fuel molecules entering this layer burn spontaneously without transport to the catalyst surface. As combustion progresses and the temperature increases, the layer in which thermal combustion occurs becomes deeper. Ultimately, substantially all of the gas in the catalytic region is raised to a temperature at which thermal combustion occurs in virtually the entire gas stream rather than just near the surface of the catalyst. Once this stage is reached within the catalyst, the thermal reaction appears to continue even without further contact of the gas with the catalyst.

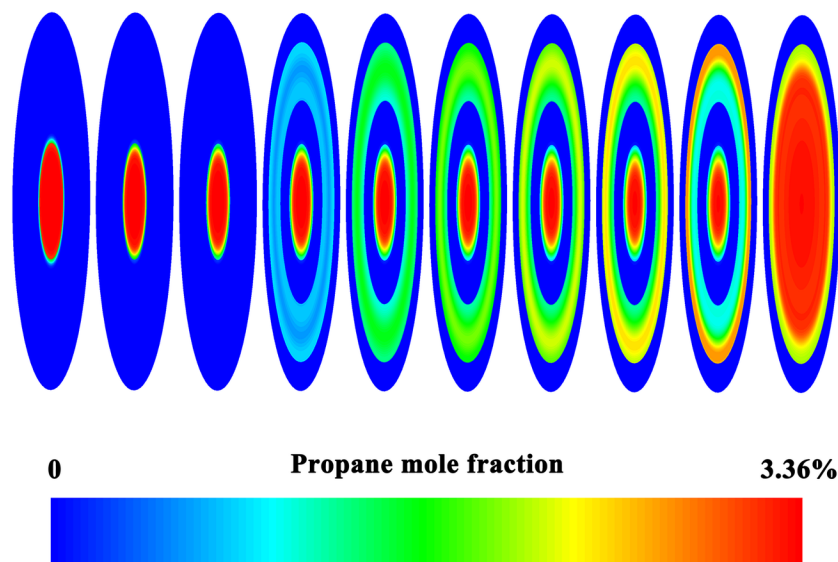


Figure 3. Propane mole fraction contour plots in the gas turbine combustor designed for the reduction of nitrogen oxides emissions by heterogeneous catalysis.

The temperature contour plots in the gas turbine combustor are illustrated in Figure 4 for the reduction of nitrogen oxides emissions by heterogeneous catalysis. While gas turbine engines employing purely thermal combustion have been used extensively as prime movers, especially in aircraft and stationary power plants, they have not been found to be commercially attractive for propelling land vehicles, such as trucks, buses and passenger cars [63, 64]. One reason for this is the inherent disadvantages of systems based purely on thermal combustion or conventional catalytic combustion [65, 66]. Catalysts are available for promoting catalytic combustion. These catalysts are useful in eliminating nitrogen oxides from gases which also contain some oxygen. In general, reaction of oxygen with combustible hydrocarbons or hydrogen provides sufficient heat to raise the temperature of the gas mixture so that nitrogen oxides present will decompose when the oxygen present is depleted by combustion. The reactions are all exothermic and provide considerable heat. Reactions between oxygen and hydrocarbon gases or between oxygen and hydrogen initiate at different temperatures. Plant efficiencies are markedly reduced and pollution limits for nitrogen oxides exceeded when catalysts do not function. There is sufficient heat in gases leaving the catalytic combustion catalyst to reform a mixture of natural gas or other hydrocarbon and steam. Use of this heat via indirect heat exchange for reforming converts the incoming natural gas or hydrocarbon into a stream of gas containing a high percentage of hydrogen. This hydrogen is more than sufficient to initiate the catalytic combustion and to raise temperatures in the catalyst to a point where the remaining combustibles in the gas stream will also react with oxygen, reducing the oxygen content and further raising the temperature so that nitrogen oxide will decompose. According to the catalytically-supported, thermal combustion method, carbonaceous fuels can be combusted very efficiently at temperatures between about 1,200 K and about 2,000 K, for example, without the formation of substantial amounts of carbon monoxide or nitrogen oxides by a process designated catalytically-supported, thermal combustion. In conventional thermal combustion of carbonaceous fuels, a flammable mixture of fuel and air or fuel, air, and inert gases is contacted with an ignition source to ignite the mixture. Once ignited, the mixture continues to burn without further support from the ignition source. Flammable mixtures of carbonaceous fuels normally burn at relatively high temperatures. At these temperatures, substantial amounts of nitrogen oxides inevitably form if nitrogen is present, as is always the case when air is the source of oxygen for the combustion reaction. Mixtures of fuel and air or fuel, air, and inert gases which would theoretically burn at temperatures below about 2,080 K are too fuel-lean to support a stable flame and therefore cannot be satisfactorily burned in a conventional thermal combustion system.

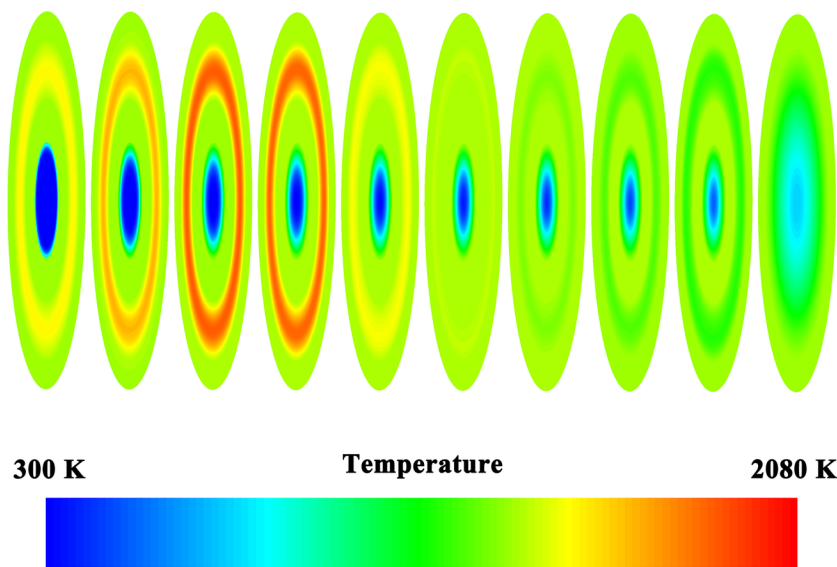


Figure 4. Temperature contour plots in the gas turbine combustor designed for the reduction of nitrogen oxides emissions by heterogeneous catalysis.

The propane mole fraction profiles along the fluid centerline of the gas turbine combustor are presented in Figure 5 under different fluid velocity conditions. In conventional catalytic combustion, the fuel is burned at relatively low temperatures [67, 68]. However, catalytic combustion is regarded as having limited value as a source of thermal energy [69, 70]. In the first place, conventional catalytic combustion proceeds relatively slowly so that impractically large amounts of catalyst would be required to produce enough combustion effluent gases to drive a turbine or to consume the large amounts of fuel required in most large furnace applications. In the second place, the reaction temperatures normally associated with conventional catalytic combustion are too low for efficient transfer of heat for many purposes. Typically, catalytic combustion is also relatively inefficient, so that significant amounts of fuel are incompletely combusted or left un-combusted unless low space velocities in the catalyst are employed. Catalytic combustion is a chemical process whereby a combustible species, in the gas phase, is reacted over a solid catalyst to completely oxidize the target molecules. For molecules containing only carbon and hydrogen or molecules containing carbon, hydrogen, and oxygen, the products of catalytic combustion are solely carbon dioxide and water. Catalytic combustion is frequently, but not exclusively, employed in applications where the concentration of the target species is below its lower ignition limit. When combustible gases are below their lower ignition limit then the mixture will not ignite when exposed to an ignition source. At such dilute concentrations, it is more efficient to use catalysts to react, convert, or combust, the target compounds because a catalyst can facilitate complete oxidation of the target species at a temperature significantly lower than the auto-ignition temperature of the molecule. It is generally desirable to develop catalysts that are capable of facilitating catalytic combustion at the lowest possible temperature in order to reduce the energy costs associated with operating a catalytic combustion system. Complete catalytic combustion of a target species can only occur when oxygen gas is found in molar stoichiometric excess; a condition which is easily met when the target species is present in trace quantity in air. Even in a reduced-oxygen environment, provided more moles of oxygen gas are present compared to moles of carbon atoms to be combusted then complete combustion of the target species can be realized. The required residence time of the gases in the space between the catalyst and the inlet of the turbine expansion zone is a function of the temperature of the gases exiting the catalyst. In any event, the gas residence time between the exit of the upstream oxidation zone and the inlet of the turbine gas expansion zone may be minimal and is such that at least a significant amount of the combustion takes place in the turbine gas expansion zone. If desirable, this residence time may be so small that at least a major portion of the total combustion occurring subsequent to the upstream catalyst zone is in the turbine expansion zone. The fuel is combusted in contact with free or molecular oxygen and free or molecular nitrogen. The fuel may occur or be obtained in admixture with components which are essentially inert in the oxidation system. The fuel has a relatively high energy content and is of a nature which permits the preparation of the oxidation feed streams.

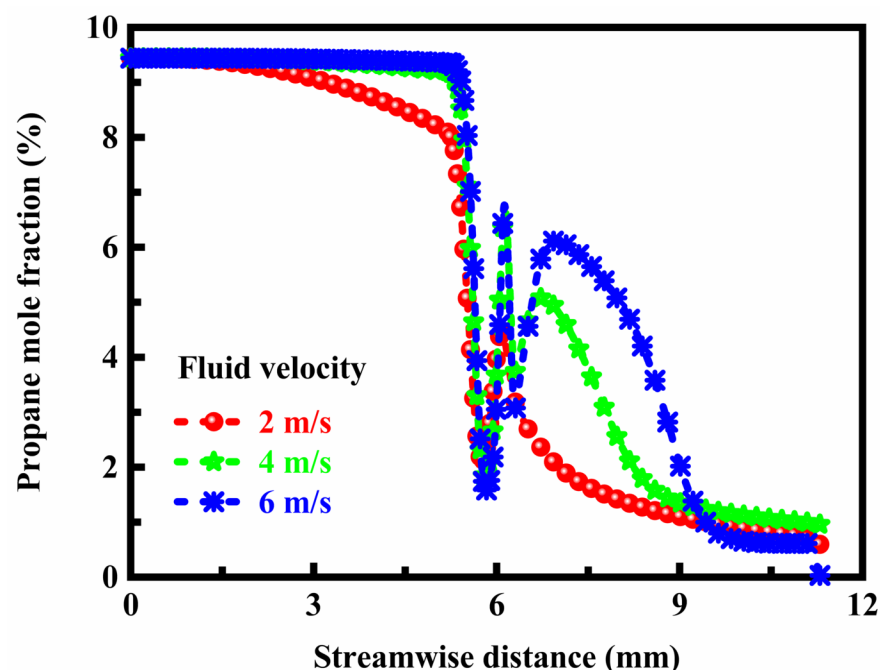


Figure 5. Propane mole fraction profiles along the fluid centerline of the gas turbine combustor under different fluid velocity conditions.

The homogeneous reaction rate profiles along the fluid centerline of the gas turbine combustor are presented in Figure 6 under different fluid velocity conditions. 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 [71, 72]. 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 [73, 74]. The channels through the unitary body or skeletal structure can be of any shape and size consistent with the desired superficial surface and should be large enough to permit relatively free passage of the gas mixture. The channels may be parallel, or generally parallel, and extend through the support from one side to an opposite side, such channels being separated from one another by preferably thin walls. The channels may also be multi-directional and may even communicate with one or more adjacent channels. The channel inlet openings can be distributed across essentially the entire face or cross-section of the support subject to initial contact with the gas to be oxidized. The unitary, skeletal structure support type of oxidation catalyst can be characterized by having a plurality of flow channels or paths extending therethrough in the general direction of gas flow. The flow channels need not pass straight through the catalyst structure and may contain flow diverters or spoilers. The skeletal structure support is preferably constructed of a substantially chemically inert, rigid, solid material capable of maintaining its shape and strength at high temperatures. The support may have a low thermal coefficient of expansion and low thermal conductivity. Often, the skeletal support is porous but its surface may be relatively non-porous, and it may be desirable to roughen its surface so that it holds the catalyst coating better, especially if the support is relatively non-porous. The support may be metallic or ceramic in nature or a combination thereof. The combustion gases are exhausted to atmosphere after giving up some of their remaining heat to the incoming air provided from the compressor. Operating the combustion process in a very lean condition 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. In a catalytic combustor, fuel is burned at relatively low temperatures. While emissions can be

reduced by combustion at these temperatures, the utilization of catalytic combustion is 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. In conventional thermal combustion systems, a fuel and air in flammable proportions are contacted with an ignition source to ignite the mixture which will then continue to burn. Flammable mixtures of most fuels are normally burned at relatively high temperatures, 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 decreased 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 combustion systems utilizing a catalyst, there is little or no nitrogen oxides formed in a system which burns the fuel at relatively low temperatures. Such combustion heretofore has been generally regarded as having limited practicality in providing a source of power as a consequence of the need to employ amounts of catalyst so large as to make a system unduly large and cumbersome. Consequently, combustion utilizing a catalyst has been limited generally to such operations as treating tail gas streams of nitric acid plants. The catalytically supported thermal combustion involves essentially adiabatic combustion of a mixture of fuel and air or fuel, air, and inert gases in the presence of a solid oxidation catalyst operating at a temperature substantially above the instantaneous auto-ignition temperature of the mixture, but below a temperature which would result in any substantial formation of oxides of nitrogen under the conditions existing in the catalyst.

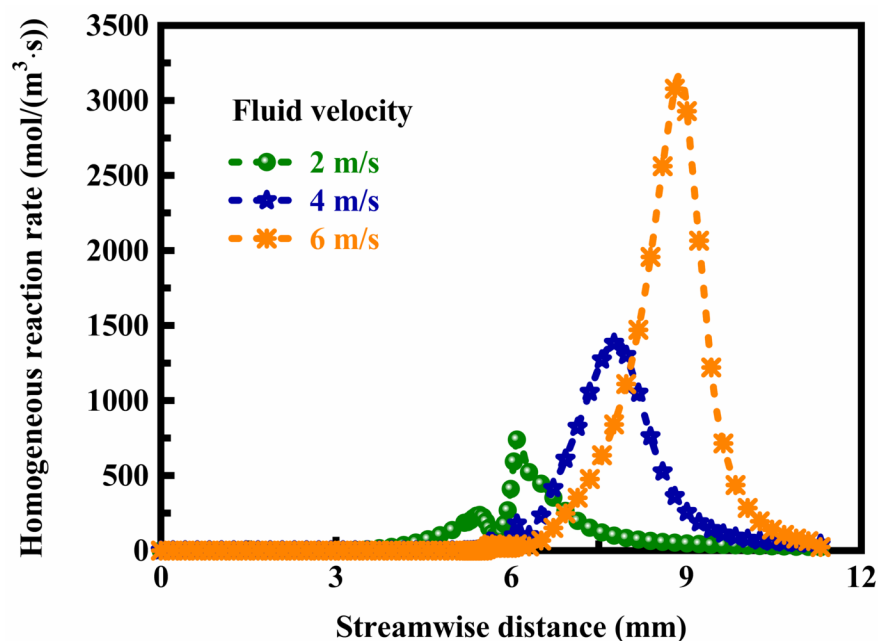


Figure 6. Homogeneous reaction rate profiles along the fluid centerline of the gas turbine combustor under different fluid velocity conditions.

The pressure contour plots in the gas turbine combustor are illustrated in Figure 7 for the reduction of nitrogen oxides emissions by heterogeneous catalysis. At relatively low temperatures, the catalytic reaction rate increases exponentially with temperature. As the temperature is raised further, the reaction rate enters a transition zone in which the rate at which the fuel and oxygen are being transferred to the catalytic surface begins to limit further increases in the reaction rate. As the temperature is raised still further, the reaction rate enters a so-called mass transfer limited zone in which the reactants cannot be transferred to the catalytic surface fast enough to keep up with the catalytic surface reaction and the reaction rate levels

off regardless of further temperature increases. In the mass transfer limited zone, the reaction rate cannot be increased by increasing the activity of the catalyst because catalytic activity is not determinative of the reaction rate. The only apparent way to increase the reaction rate in a mass transfer limited reaction is to increase mass transfer. However, this typically requires an increase in the pressure drop across the catalyst and consequently a substantial loss of energy. Sufficient pressure drop may not even be available to provide the desired reaction rate. Of course, more mass transfer can be affected, and hence more energy can always be produced by increasing the amount of catalyst surface. However, this results in catalyst configurations of such size and complexity that the cost is prohibitive and the body of the catalyst is unwieldy. For example, in the case of gas turbine engines, the catalytic reactor might very well be larger than the engine itself. In general, conventional adiabatic, thermal combustion systems operate at such high temperatures in the combustion zone that undesirable nitrogen oxides, 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. 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. Catalytic combustion systems, though, are capable of achieving ultra-low emissions [75, 76]. However, catalytic combustion systems are not able to offer the accuracy and controllability of the air staging system over a wide range of power levels, fuel properties and ambient operating conditions [77, 78]. The system may be operated in different manners to allow for low and high-power operation, as well as according to a controlled schedule that may be programmed. Under low power operation oxidation does not occur in the catalyst section. However, the mixing of the fuel and air in the fuel preparation and mixing section is enhanced by the presence of the catalyst. As the engine power level increases the compressor outlet air temperature will become high enough to activate the catalyst, and partial oxidation reactions will occur.

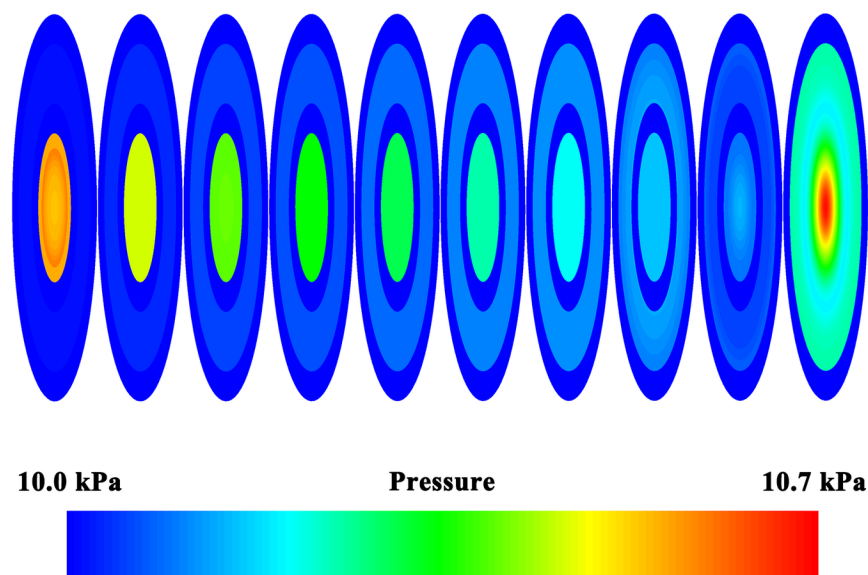


Figure 7. Pressure contour plots in the gas turbine combustor designed for the reduction of nitrogen oxides emissions by heterogeneous catalysis.

The chemical enthalpy contour plots in the gas turbine combustor are illustrated in Figure 8 for the reduction of nitrogen oxides emissions by heterogeneous catalysis. Nitrogen oxides formation is thermodynamically favored at high temperatures. 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. The limits of the operating temperature are governed largely by residence time and pressure. The instantaneous auto-ignition temperature of the mixture is defined herein to mean the temperature at which the ignition lag of the mixture entering the catalyst is negligible relative to the residence time in the combustion zone of the mixture undergoing combustion. Essentially adiabatic combustion means in this case that the operating temperature of the catalyst does not differ by more than about 80 K, from the adiabatic flame temperature of the mixture due to heat losses from the catalyst. Among the unique advantages of the catalytically supported thermal combustion in the presence of a catalyst is the fact that mixtures of fuel and air which are too fuel-lean for ordinary thermal combustion can be burned efficiently. Since the temperature of combustion for a given fuel at any set of conditions is dependent largely on the proportions of fuel, of oxygen available for combustion, and of inert gases in the mixture to be burned, it becomes practical to burn mixtures which are characterized by much lower flame temperatures. In particular, carbonaceous fuels can be burned very efficiently and at thermal reaction rates at temperatures in the range from about 1,200 K to about 2,000 K. At these temperatures, very little if any nitrogen oxides are formed. In addition, because the catalytically supported thermal combustion is stable over a wide range of mixtures, it is possible to select or control reaction temperature over a correspondingly wide range by selecting or controlling the relative proportions of the gases in the mixture. The method may be carried out in various ways, including heating the catalyst body by electrical means, or by first thermally combusting a fuel and air mixture and applying the heat produced to the catalyst body. Once a catalyst temperature is reached at which the catalyst will function to sustain mass transfer limited operation, the combustion of fuel in the presence of the catalyst will bring it rapidly to the required operating temperature. Once operating temperature is reached, the catalyst will provide for sustained combustion of the fuel vapor. After the catalyst body reaches a temperature at which it will sustain mass transfer limited operation, the aforementioned application of heat to the catalyst body is no longer necessary and an admixture of unburned fuel and air is introduced into the system to establish the supported thermal combustion to provide a motive fluid for a turbine or heat to a furnace. No serious start-up problem normally is presented. The operation is substantially continuous and it is necessary to start the system only at infrequent intervals. Consequently, the substantial emissions of atmospheric pollutants which tend to occur in start-ups are not serious because of the small number of infrequent start-ups. While this pollution may be tolerated in stationary operations which are normally used continuously and for long periods of time, it cannot be tolerated in the vehicular type of installation where start-ups are frequent, due to intermittent operation [79, 80]. Also, the start-up must be rapid in order to be as efficient as in the conventional present-day automobile [81, 82]. The mixture of unburned fuel and air is not introduced to the catalyst body until it has reached a temperature at which it will sustain the desired rapid combustion. Such preferred procedure minimizes pollutant emissions during start-up. Once combustion in the zone containing the catalyst is achieved, the fuel-air admixture is passed to the catalyst at a gas velocity, prior to or at the inlet to the catalyst, in excess of the maximum flame propagating velocity. This avoids flash-back that causes the formation of nitrogen oxides [83, 84]. Various means have been proposed in the past to prevent such flash backs from traveling back to the gas supply source [85, 86]. Preferably, this velocity is maintained adjacent to the catalyst inlet [87, 88]. Suitable linear gas velocities are usually above about 8 meters per second, but considerably higher velocities may be required depending upon such factors as temperature, pressure, and composition.

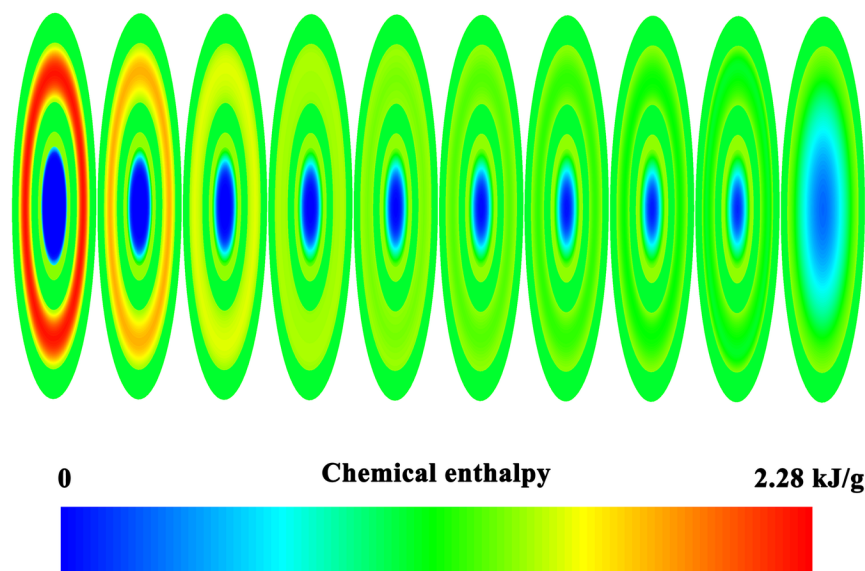


Figure 8. Chemical enthalpy contour plots in the gas turbine combustor designed for the reduction of nitrogen oxides emissions by heterogeneous catalysis.

4. Conclusions

Steady-steady simulations are performed using computational fluid dynamics. The fluid viscosity, specific heat, and thermal conductivity are calculated from a mass fraction weighted average of species properties, and the specific heat of each species is calculated using a piecewise polynomial fit of temperature. Natural parameter continuation is performed by moving from one stationary solution to another. Knowledge of critical parameters gains a fundamental understanding of the essential factors affecting the stability of the combustion process. Particular emphasis is placed upon the sustained combustion of at least a portion of fuel under essentially adiabatic conditions at a rate which surmounts the mass transfer limitation. The major conclusions are summarized as follows:

- 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.
- Flammable mixtures of carbonaceous fuels normally burn at relatively high temperatures, and substantial amounts of nitrogen oxides inevitably form if nitrogen is present.
- Complete catalytic combustion of a target species can only occur when oxygen gas is found in molar stoichiometric excess; a condition which is easily met when the target species is present in trace quantity in air.
- In combustion systems utilizing a catalyst, there is little or no nitrogen oxides formed in a system which burns the fuel at relatively low temperatures.
- In the mass transfer limited zone, the reaction rate cannot be increased by increasing the activity of the catalyst because catalytic activity is not determinative of the reaction rate.
- Among the unique advantages of the catalytically supported thermal combustion in the presence of a catalyst is the fact that mixtures of fuel and air which are too fuel-lean for ordinary thermal combustion can be burned efficiently.

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Design of gas turbine combustors for the reduction of nitrogen oxides emissions by heterogeneous catalysis

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Abstract

Flammable mixtures of most fuels are normally burned at relatively high temperatures, 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. Additionally, limiting such residence time makes it difficult to maintain stable combustion even after ignition. The present study relates to the design of gas turbine combustors for the reduction of nitrogen oxides emissions by heterogeneous catalysis. Steady-steady simulations are performed using computational fluid dynamics. The fluid viscosity, specific heat, and thermal conductivity are calculated from a mass fraction weighted average of species properties, and the specific heat of each species is calculated using a piecewise polynomial fit of temperature. Natural parameter continuation is performed by moving from one stationary solution to another. Particular emphasis is placed upon the sustained combustion of at least a portion of fuel under essentially adiabatic conditions at a rate which surmounts the mass transfer limitation. The results indicate that 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. Flammable mixtures of carbonaceous fuels normally burn at relatively high temperatures, and substantial amounts of nitrogen oxides inevitably form if nitrogen is present. Complete catalytic combustion of a target species can only occur when oxygen gas is found in molar stoichiometric excess; a condition which is easily met when the target species is present in trace quantity in air. In combustion systems utilizing a catalyst, there is little or no nitrogen oxides formed in a system which burns the fuel at relatively low temperatures. In the mass transfer limited zone, the reaction rate cannot be increased by increasing the activity of the catalyst because catalytic activity is not determinative of the reaction rate. Among the unique advantages of the catalytically supported thermal combustion in the presence of a catalyst is the fact that mixtures of fuel and air which are too fuel-lean for ordinary thermal combustion can be burned efficiently.

Keywords: Heterogeneous catalysis; Nitrogen oxides; Gas turbines; Flammable mixtures; Thermodynamic properties; Combustion phenomena

1. Introduction

Combustion is a chemical reaction between substances, usually including oxygen and usually accompanied by the generation of heat and light in the form of flame [1, 2]. The rate or speed at which the reactants combine is high, in part because of the nature of the chemical reaction itself and in part because more energy is generated than can escape into the surrounding medium, with the result that the temperature of the reactants is raised to accelerate the reaction even more [3, 4]. Combustion encompasses a great variety of phenomena with wide application in industry, the sciences, professions,

and the home, and the application is based on knowledge of physics, chemistry, and mechanics; their interrelationship becomes particularly evident in treating flame propagation [5, 6]. Combustion, with rare exceptions, is a complex chemical process involving many steps that depend on the properties of the combustible substance. It is initiated by external factors such as heat, light, and sparks [7, 8]. The reaction sets in as the mixture of combustibles attains the ignition temperature. 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 [9, 10]. 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 complexity of the combustion reaction mechanism and the rapidly varying temperatures and concentrations in the mixture make it difficult and often impossible to derive an equation that would be useful for predicting combustion phenomena over wide temperature and concentration ranges. Instead, use is made of empirical expressions derived for specific reaction conditions [11, 12]. 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 [13, 14]. The latter are either measured or calculated in terms of the kinetic theory of gases. The process of diffusion is of great importance in combustion reactions, in flames, that is, in gaseous mixtures, and in solids or liquids. Diffusion heat transfer follows a law stating that the heat flux is proportional to the temperature gradient [15, 16]. The coefficient of proportionality, called the thermal conductivity coefficient, is also measured or calculated in terms of the kinetic theory of gases, like the diffusion coefficient. Convective transport of mass and energy may be accounted for by buoyant forces, external forces, and turbulent and eddy motions. Convection is in the main responsible for the mixing of gases. Flame combustion is most prominent with fuels that have been premixed with an oxidant, either oxygen or a compound that provides oxygen, for the reaction [17, 18]. The temperature of flames with this mixture is often several thousand degrees. The chemical reaction in such flames occurs within a narrow zone several micrometers thick [19, 20]. This combustion zone is usually called the flame front. Dilution of the burning mixture with an inert gas, such as helium or nitrogen, lowers the temperature and, consequently, the reaction rate [21, 22]. Great amounts of inert gas extinguish the flame, and the same result is achieved when substances that remove any of the active species are added to the flame [23, 24]. Conditions must be such that the flame is fixed at the burner nozzle or in the combustion chamber [25, 26]. This positioning is required in many practical uses of combustion. Various devices, such as pilot flames and recirculation methods, are designed for this purpose.

A number of combustion systems promote partial conversion of a fuel followed by complete combustion of that fuel in a downstream combustion zone [27, 28]. These methods generally comprise introducing a fuel and air mixture into a combustion zone wherein a portion of the fuel has been partially reacted prior to entering the combustion zone. Such partial reaction may be promoted chemically, catalytically, or by any other conventional means depending upon each particular application [29, 30]. As the partially reacted fuel and air mixture are introduced into a region of the combustion zone, as with a dump, a flame is established to promote complete combustion of the fuel within the fuel and air mixture. Flame stabilization is a common problem in these combustion systems [31, 32]. A flame will propagate through a fuel-air mixture only when certain conditions prevail. Initially, a minimum percentage of fuel must be present within the fuel-air mixture to make the fuel-air mixture flammable, namely the lean flammability limit. Similarly, a maximum percentage of fuel must be present within the fuel-air mixture wherein greater than this percentage will prevent burning, namely the rich flammability limit. The flammability range of a fuel-air mixture is that range of the percentage

of fuel within the fuel-air mixture between the lean flammability limit and the rich flammability limit [33, 34]. The stoichiometry of a fuel-air mixture contributes to its flammability range [35, 36]. A stoichiometric fuel-air mixture composition contains sufficient oxygen for complete combustion thereby releasing all the latent heat of combustion of the fuel [37, 38]. The strength of a fuel-air mixture composition typically is expressed in terms of its equivalence ratio. The equivalence ratio is the actual fuel-air ratio divided by stoichiometric fuel-air ratio. For example, an equivalence ratio of one represents a stoichiometric fuel-air mixture composition. An equivalence ratio less than one represents a lean mixture and an equivalence ratio less great than one represents a rich mixture.

Pressure and temperature contribute to the flammability range of fuel-air mixtures. Typically, with increases in pressure, the rich flammability limit is extended thereby extending the flammability range of the fuel-air mixture. Temperature, on the other hand, partially defines the flammability range of fuel-air mixtures [39, 40]. The lowest temperature at which a flammable fuel-air mixture can be formed, based upon the vapor pressure of the fuel at atmospheric pressure, is the flash point of that fuel-air mixture [41, 42]. Within the flammability range of a fuel-air mixture, at temperatures exceeding the flash point of the fuel-air mixture, auto-ignition of the fuel vapor occurs. Auto-ignition generally occurs at or slightly above the stoichiometric fuel-air mixture composition. The time interval between the mixing of the fuel-air mixture such that it is combustible and the auto-ignition of that fuel-air mixture is known as the auto-ignition delay time [43, 44]. One reason that flame stabilization is required in combustion systems is to prevent the flame front from moving upstream from the combustion zone toward the source of fuel, namely a flashback. During a flashback event, the heat of combustion moves upstream and may damage numerous structures within the fuel and air mixing region of the combustor. Flashback may occur due to auto-ignition of a fuel-air mixture caused by a residence time of the fuel-air mixture in a region upstream of the combustion zone that exceeds the auto-ignition delay time of that fuel-air mixture at the temperature and pressure of that region [45, 46]. Flame stabilization also is dependent upon speed of the fuel-air mixture entering the combustion zone where propagation of the flame is desired [47, 48]. A sufficiently low velocity must be retained in the region where the flame is desired in order to sustain the flame. A region of low velocity in which a flame can be sustained can be achieved by causing recirculation of a portion of the fuel-air mixture already burned thereby providing a source of ignition to the fuel-air mixture entering the combustion zone. However, the fuel-air mixture flow pattern, including any recirculation, is critical to achieving flame stability.

In gas turbines, compressed air enters the combustion chamber where it mixes with the fuel. The expanding combustion products impart their energy to the turbine blades. However, flammable mixtures of most fuels are normally burned at relatively high temperatures, 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. Additionally, limiting such residence time makes it difficult to maintain stable combustion even after ignition. The present study relates to the design of gas turbine combustors for the reduction of nitrogen oxides emissions by heterogeneous catalysis. More particularly, the present study is directed to an improved method for more efficiently operating a catalytically supported thermal combustion gas turbine system, and at the same time provide low emissions of unburned hydrocarbons, carbon monoxide, and nitrogen oxides. The present study is focused primarily upon at least a portion of the thermal combustion of the fuel takes place in the expansion zone of the gas turbine to counteract the cooling effect of the expansion of the gases. Natural parameter continuation is performed by moving from one stationary solution to another. A critical point is denoted as the solution to the problem when a turning point is reached. Knowledge of critical parameters gains a fundamental understanding of the essential factors affecting the stability of the

catalytically supported thermal combustion process. The critical parameters are useful as the design guides associated with the gas turbine combustor. The principal quantitative characteristic of a flame is its normal, or fundamental, burning velocity, which depends on the chemical and thermodynamic properties of the mixture and on pressure and temperature, under given conditions of heat loss. The burning velocity value ranges from several centimeters to even tens of meters per second. The dependence of the burning velocity on molecular structure, which is responsible for fuel reactivity, is known for a great many fuel-air mixtures. Steady-steady simulations are performed using computational fluid dynamics. The present study aims to provide an improved method for operating a gas turbine combustor by catalytically supported thermal combustion of carbonaceous fuel. Particular emphasis is placed upon the sustained combustion of at least a portion of fuel under essentially adiabatic conditions at a rate which surmounts the mass transfer limitation.

2. Methods

Computational fluid dynamics is an approach to solving fluid flow problems by solving models that include numerical methods and algorithms used to represent fluids. The methods and algorithms are generally solved by computers [49, 50]. The solution of the models can provide a simulation of an interaction of fluids with a structure defined by surfaces, each of which can be defined by boundary conditions within the model. The results, or solution, can be used to validate and improve designs, for example. Computational fluid dynamics is the analysis and prediction of fluid flows and heat transfer using a computer model. Computational fluid dynamics uses mathematical methods to solve problems that include fluid flow [51, 52]. It may be used to predict the flows of fluids through a heat exchanger, or through a valve or a mixing vessel for example [53, 54]. 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 perform the computational fluid dynamics modeling 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 [55, 56]. 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 [57, 58]. However, the very sophistication that enables the software to cope with any fluid flow problem, makes it expensive and also complex.

The gas turbine combustor configured to combust propane is represented physically in Figure 1 for the reduction of nitrogen oxides emissions by heterogeneous catalysis. The gas turbine combustor comprises a concentric annular channel, wherein the concentric annular channel further comprises an inner annular channel and an outer annular channel. A platinum catalyst is deposited only upon the interior surface of the inner channel, and the wall of the outer channel is chemically inert and catalytically inactive. The reactant stream flows through the catalytically-coated inner channel and the product stream flows out of the outer non-catalytic channel. Fuel is present for combustion in both the catalytic and non-catalytic channels. The concentrically arranged annular channel is 5.0 millimeters in inner channel length, 5.6 millimeters in outer channel length, 0.8 millimeters in innermost diameter, 2.6 millimeters in outermost diameter, 0.1 millimeters in catalyst layer thickness, and 0.2 millimeters in wall thickness, unless otherwise stated. The gas turbine combustor can have any dimension unless restricted by design requirements. All the walls have the same thickness. The spacing between the inner channel and the outer channel is 0.4 millimeters and remains constant. One of the potential problems associated with the gas turbine combustor, as with all micro-scale combustion systems, continues to be combustion stability. The scale of the gas turbine combustor is on the order of sub-millimeters, which is much smaller than the quenching distance of the combustible mixture in the absence of a catalyst.

The quenching distance defines a critical dimension under which propagation of the propane flame is not possible. The quenching distance is approximately 2.5 millimeters, at which combustion cannot be sustained within the gas turbine combustor in the absence of a catalyst.

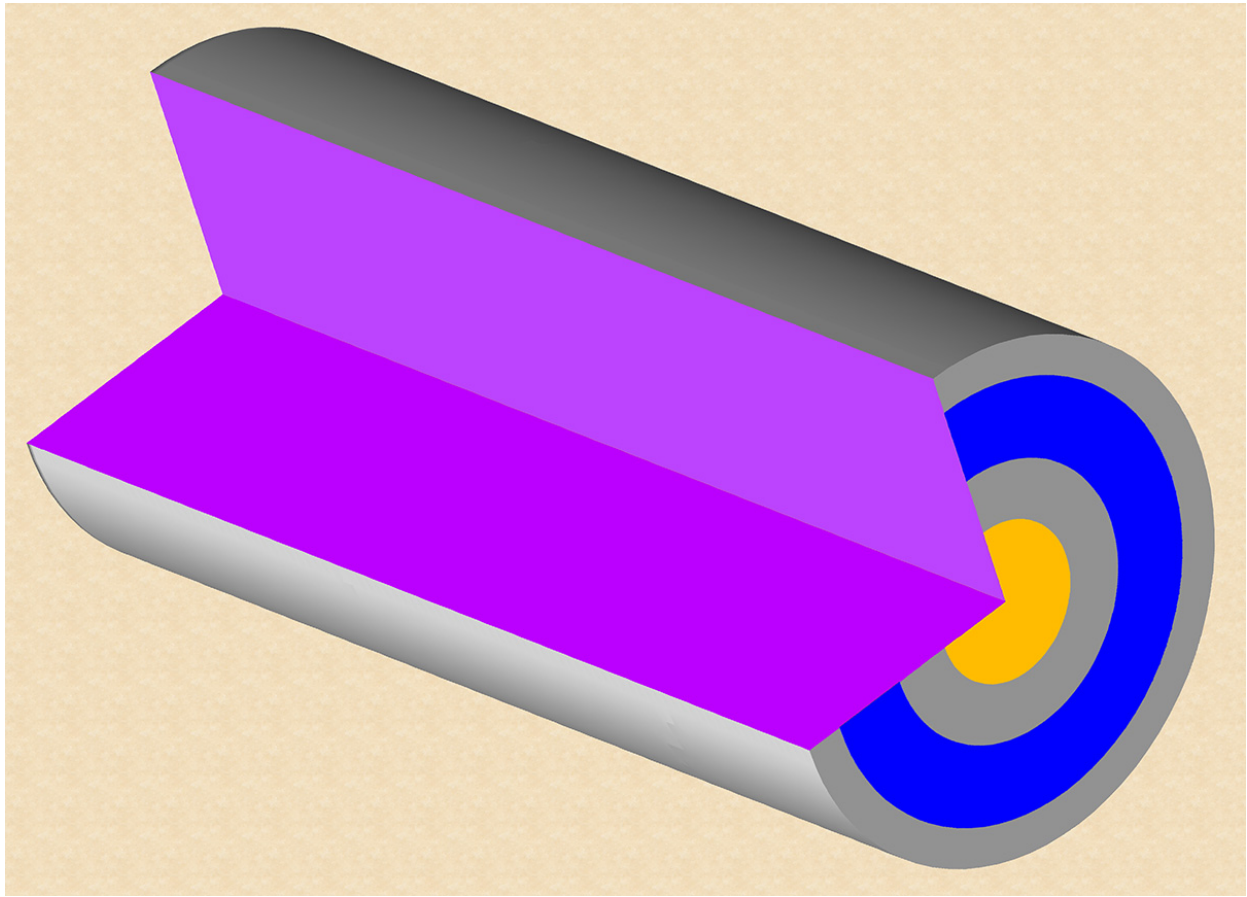


Figure 1. Physical representation of the gas turbine combustor configured to combust propane for the reduction of nitrogen oxides emissions by heterogeneous catalysis.

Complex modeling methods and algorithms are required for the gas turbine combustor due not only to the complex geometry but also the complex physicochemical processes involved. It is therefore essential to reduce the complexity of the model through use of certain simplifying assumptions. Steady-state analyses are performed, variations in reactor pressure and temperature are determined in accordance with the ideal gas law. The model is implemented in computational fluid dynamics software to obtain the solution of the problem. Computational fluid dynamics is used to describe a broader range of calculations for a wide variety of scientific and engineering applications. Thermodynamics is an important consideration in many of these applications. It relates internal energy to temperature, which affects the flow of heat. Further sources of heat include thermal radiation and chemical reactions, in particular combustion. Heat transfer may involve conduction in solid materials, coupled with the fluid flow, known as conjugate heat transfer. A modern definition of computational fluid dynamics would be the prediction of fluid motion and forces by computation using numerical analysis, generally extended to include heat, thermodynamics, chemistry and solids. Numerical analysis provides many methods and algorithms that are suitable for computational fluid dynamics. The methods include finite volume, finite element and finite difference, which calculate the distributions of properties, for instance, pressure, velocity and temperature, over regions of space which are usually fixed. Alternative methods attribute properties to particles represented by points in space, whose motions are calculated. To perform the calculation first requires a description of the problem by the domain occupied by the fluid, equations that represent the fluid behavior in terms of properties such as pressure and velocity, and conditions at the boundary of the fluid domain and initially within the domain for the fluid properties.

The typical computational mesh for the fluid and solid of the gas turbine combustor configured to combust propane is illustrated schematically in Figure 2 for the reduction of nitrogen oxides emissions by heterogeneous catalysis. A computational fluid dynamics simulation begins with a solution domain which specifies a region of space of a particular geometric shape, in which fluid dynamics equations are solved. The process of mesh generation subdivides the solution domain into a mesh of small volumes. The computational mesh consists of about 600,000 nodes in total for the fluid and solid of the gas turbine combustor. A computational fluid dynamics analysis is carried out on the mesh. A mesh independence test is performed to assure independence of the solution to the problem. The specification of boundary conditions is one of the most challenging tasks in setting up a computational fluid dynamics simulation. The range of possible boundary conditions is endless, to cover all of the potential applications and physics. Velocity inlet boundary conditions are used to define the velocity properties of the flow at the inlet boundary of the fluid region. A uniform velocity profile is specified at the flow inlet. The temperature of the mixture is prespecified at the flow inlet. Under-relaxation is a general method used to improve solution convergence by limiting the amount that a variable changes during a solution step. The computational fluid dynamics calculations may take days in order to arrive at a reasonably accurate solution, using fine grids of the gas turbine combustor, due to the time-consuming nature of the model. Natural parameter continuation is performed by moving from one stationary solution to another. A critical point is denoted as the solution to the problem when a turning point is reached. Knowledge of critical parameters gains a fundamental understanding of the essential factors affecting the stability of the catalytically supported thermal combustion process. The critical parameters are useful as the design guides associated with the gas turbine combustor.

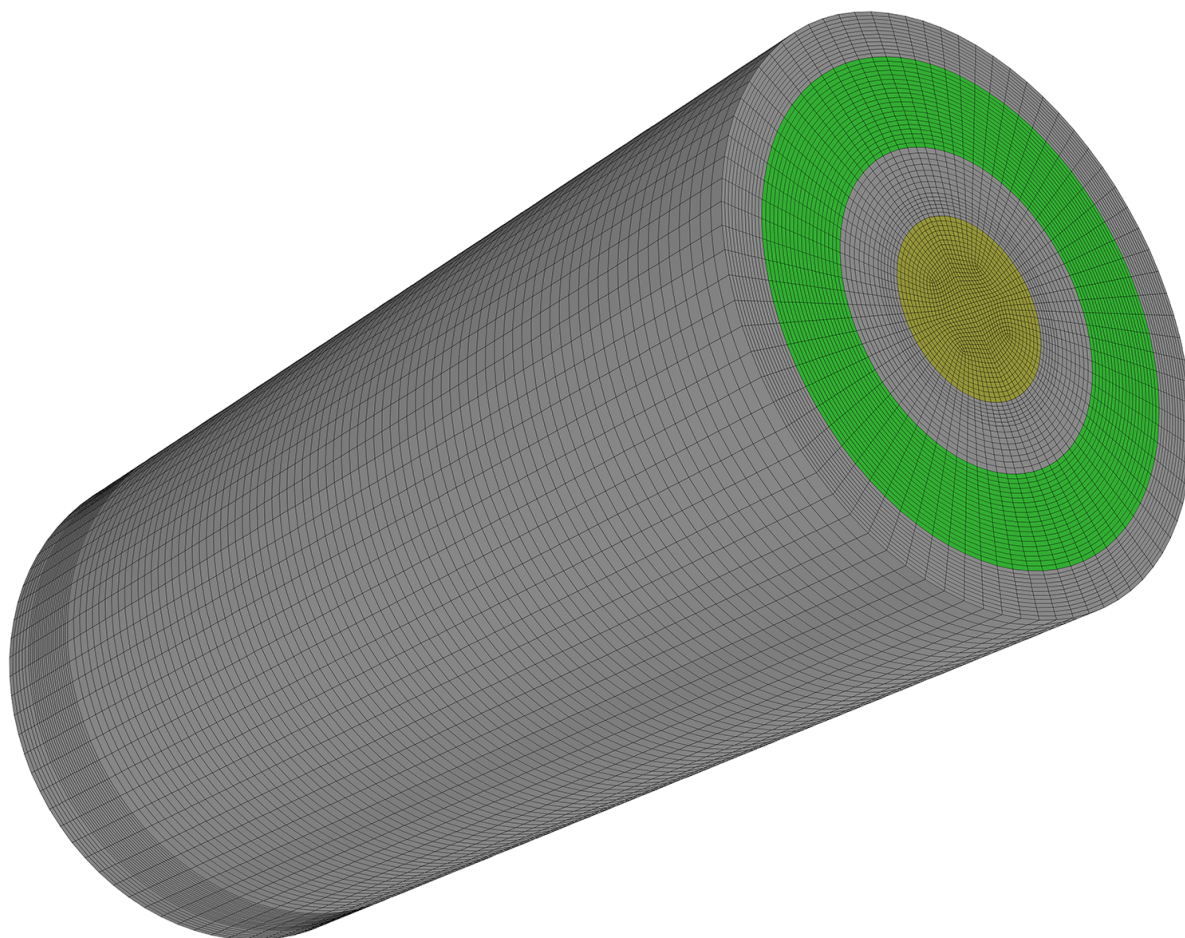


Figure 2. Schematic illustration of the typical computational mesh for the fluid and solid of the gas turbine combustor configured to combust propane for the reduction of nitrogen oxides emissions by heterogeneous catalysis.

Steady-steady simulations are performed, unless otherwise stated. The fluid density is calculated using the ideal gas law. The fluid viscosity, specific heat, and thermal conductivity are calculated from a mass fraction weighted average of species properties, and the specific heat of each species is calculated using a piecewise polynomial fit of temperature. The wall thermal conductivity and exterior convective heat loss coefficient are taken as independent parameters to understand how important thermal management is. The heat flux at the wall-fluid interface is computed using Fourier's law and continuity in temperature and heat flux links the fluid and solid phases. The left and right edges of the wall are assumed to be insulated. Newton's law of cooling is used at the outer edge of the wall. Computations are performed using meshes with varying nodal densities to determine the optimum node spacing and density that would give the desired accuracy and minimize computation time. As the mesh density increases, there is a convergence of the solution. The coarsest mesh fails to accurately capture the inflection point at the ignition point and the maximum temperature. Solutions obtained with meshes consisting of a few million nodes are reasonably accurate. The gas phase is assumed to be in quasi-steady-state. The walls are transiently modeled using energy diffusion and radiation. The gas phase is modeled using the forms of the momentum, energy, and species equations. To solve the conservation equations, a segregated solution solver with an under-relaxation method is used. The segregated solver first solves the momentum equations, then solves the continuity equation, and updates the pressure and mass flow rate. The energy and species equations are subsequently solved and convergence is checked. The latter is monitored through both the values of the residuals of the conservation equations and the difference between subsequent iterations of the solution. Numerical convergence is in general difficult because of the inherent stiffness of the chemistry as well as the disparity between the wall and the fluid heat conductivities. In order to assist convergence and compute extinction points, natural parameter continuation is implemented. The calculation time varies for these problems, depending on the initial guess as well as the parameter set.

3. Results and discussion

The propane mole fraction contour plots in the gas turbine combustor are illustrated in Figure 3 for the reduction of nitrogen oxides emissions by heterogeneous catalysis. In order to meet the emission level requirements, for industrial low emission gas turbine engines, staged combustion is required in order to minimize the quantity of the oxides of nitrogen produced [59, 60]. 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 [61, 62]. The present design provides a gas turbine engine combustion chamber comprising a primary combustion zone and a secondary combustion zone downstream of the primary combustion zone. In this way, the temperatures in each stage can be reduced. Suitable temperature ranges in each stage are from 900 K to 1500 K, depending on the particular catalyst and support. An advantage of operating in this temperature range is that it is below the fixation temperature of nitrogen and consequently the combusted gases are free of nitrogen oxides. Additionally, catalytic combustion results in lower un-combusted fuel content. A further advantage of catalytic combustion is that it is possible to operate with minimum of air for combustion, namely excess oxygen in the combusted gases can be reduced almost to zero. The catalyst may be supported on a monolith. The preferred characteristics of the metallic monolith having a catalyst deposited thereon are that it presents low resistance to the passage of gases by virtue of its possession of a high ratio of open area to blocked area and that it has a high surface to volume ratio. Preferably, the metallic monolith is formed from one or more metals selected from the group comprising ruthenium, rhodium, palladium, iridium, and platinum. However, base metals may be used or base metal alloys which also contain a platinum group metal component may be used. Oxygen is the required element to support combustion. 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. In particular, if the operating temperature of the catalyst is increased substantially into the mass transfer limited region, the reaction rate again begins to increase rapidly with temperature. This is in apparent contradiction of the laws of mass transfer kinetics in catalytic reactions. The phenomenon may be explained by the fact that the temperature of the catalyst surface and the gas layer near the catalyst surface are above the instantaneous auto-ignition temperature of the mixture of fuel, air, and any inert gases and at a temperature at which thermal combustion occurs at a rate higher than the catalytic combustion rate. The fuel molecules entering this layer burn spontaneously without transport to the catalyst surface. As combustion progresses and the temperature increases, the layer in which thermal combustion occurs becomes deeper. Ultimately, substantially all of the gas in the catalytic region is raised to a temperature at which thermal combustion occurs in virtually the entire gas stream rather than just near the surface of the catalyst. Once this stage is reached within the catalyst, the thermal reaction appears to continue even without further contact of the gas with the catalyst.

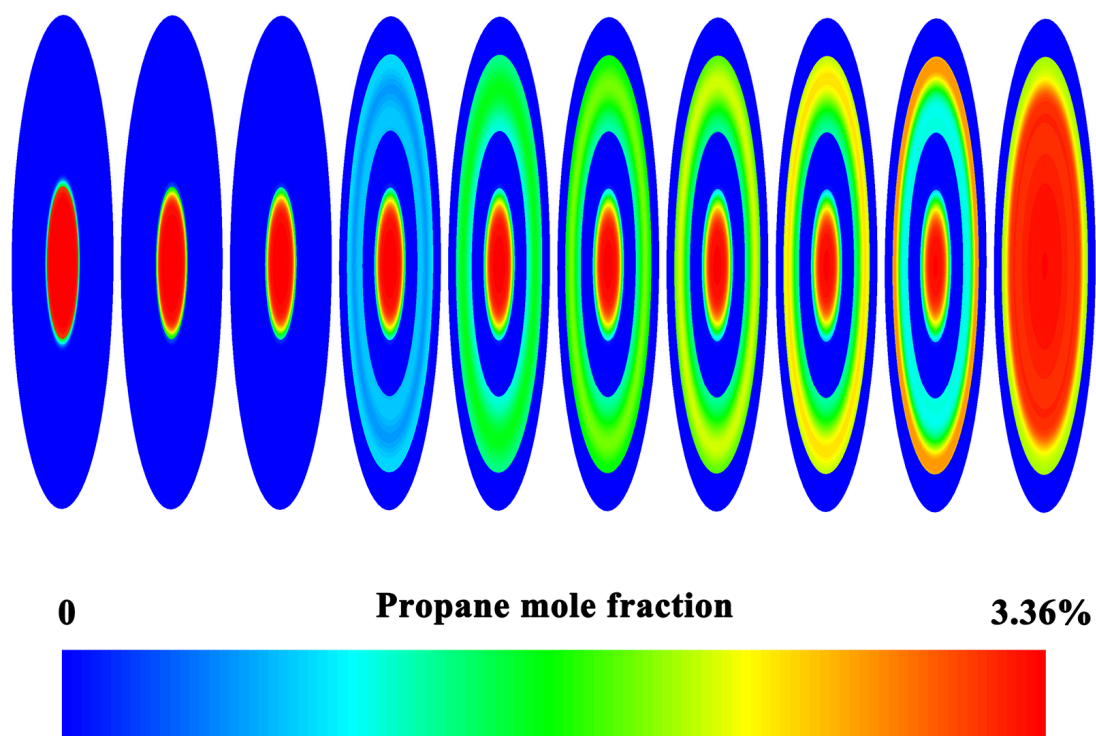


Figure 3. Propane mole fraction contour plots in the gas turbine combustor designed for the reduction of nitrogen oxides emissions by heterogeneous catalysis.

The temperature contour plots in the gas turbine combustor are illustrated in Figure 4 for the reduction of nitrogen oxides emissions by heterogeneous catalysis. While gas turbine engines employing purely thermal combustion have been used extensively as prime movers, especially in aircraft and stationary power plants, they have not been found to be commercially attractive for propelling land vehicles, such as trucks, buses and passenger cars [63, 64]. One reason for this is the inherent disadvantages of systems based purely on thermal combustion or conventional catalytic combustion [65, 66]. Catalysts are available for promoting catalytic combustion. These catalysts are useful in eliminating nitrogen oxides from gases which also contain some oxygen. In general, reaction of oxygen with combustible hydrocarbons or hydrogen provides sufficient heat to raise the temperature of the gas mixture so that nitrogen oxides present will decompose when the oxygen present is depleted by combustion. The reactions are all exothermic and provide considerable heat. Reactions between oxygen and hydrocarbon gases or between oxygen and hydrogen initiate at different temperatures. Plant efficiencies are markedly reduced and pollution limits for nitrogen oxides exceeded when

catalysts do not function. There is sufficient heat in gases leaving the catalytic combustion catalyst to reform a mixture of natural gas or other hydrocarbon and steam. Use of this heat via indirect heat exchange for reforming converts the incoming natural gas or hydrocarbon into a stream of gas containing a high percentage of hydrogen. This hydrogen is more than sufficient to initiate the catalytic combustion and to raise temperatures in the catalyst to a point where the remaining combustibles in the gas stream will also react with oxygen, reducing the oxygen content and further raising the temperature so that nitrogen oxide will decompose. According to the catalytically-supported, thermal combustion method, carbonaceous fuels can be combusted very efficiently at temperatures between about 1,200 K and about 2,000 K, for example, without the formation of substantial amounts of carbon monoxide or nitrogen oxides by a process designated catalytically-supported, thermal combustion. In conventional thermal combustion of carbonaceous fuels, a flammable mixture of fuel and air or fuel, air, and inert gases is contacted with an ignition source to ignite the mixture. Once ignited, the mixture continues to burn without further support from the ignition source. Flammable mixtures of carbonaceous fuels normally burn at relatively high temperatures. At these temperatures, substantial amounts of nitrogen oxides inevitably form if nitrogen is present, as is always the case when air is the source of oxygen for the combustion reaction. Mixtures of fuel and air or fuel, air, and inert gases which would theoretically burn at temperatures below about 2,080 K are too fuel-lean to support a stable flame and therefore cannot be satisfactorily burned in a conventional thermal combustion system.

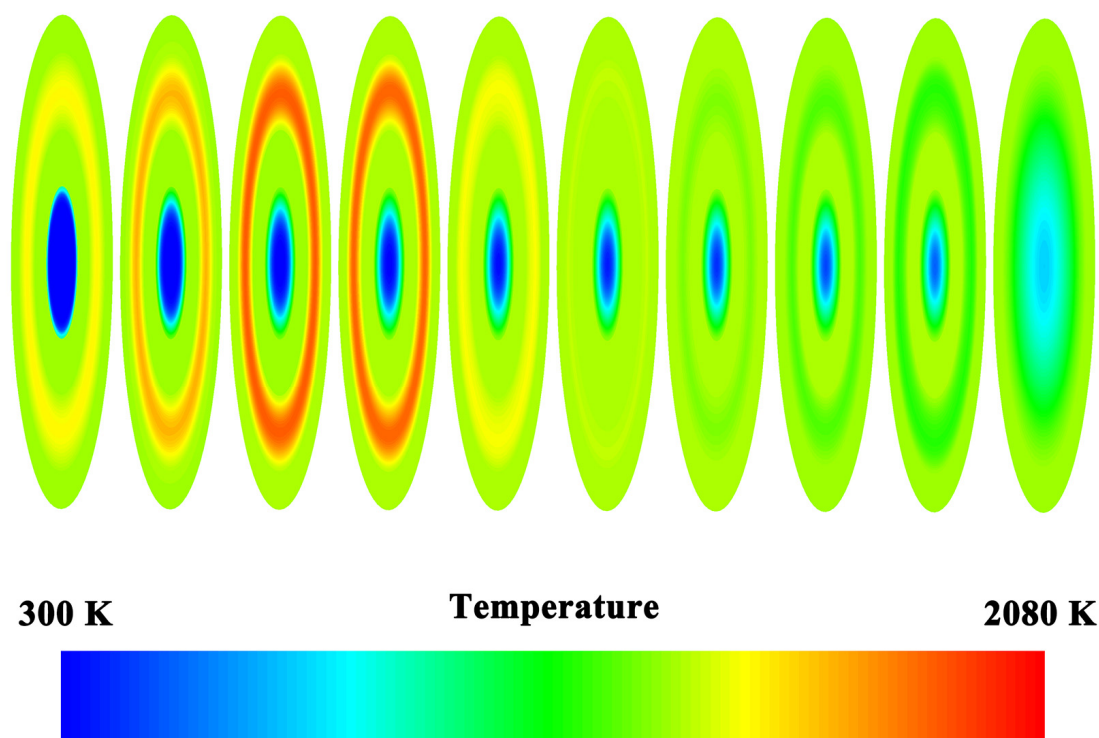


Figure 4. Temperature contour plots in the gas turbine combustor designed for the reduction of nitrogen oxides emissions by heterogeneous catalysis.

The propane mole fraction profiles along the fluid centerline of the gas turbine combustor are presented in Figure 5 under different fluid velocity conditions. In conventional catalytic combustion, the fuel is burned at relatively low temperatures [67, 68]. However, catalytic combustion is regarded as having limited value as a source of thermal energy [69, 70]. In the first place, conventional catalytic combustion proceeds relatively slowly so that impractically large amounts of catalyst would be required to produce enough combustion effluent gases to drive a turbine or to consume the large amounts of fuel required in most large furnace applications. In the second place, the reaction temperatures normally associated with conventional catalytic combustion are too low for efficient

transfer of heat for many purposes. Typically, catalytic combustion is also relatively inefficient, so that significant amounts of fuel are incompletely combusted or left un-combusted unless low space velocities in the catalyst are employed. Catalytic combustion is a chemical process whereby a combustible species, in the gas phase, is reacted over a solid catalyst to completely oxidize the target molecules. For molecules containing only carbon and hydrogen or molecules containing carbon, hydrogen, and oxygen, the products of catalytic combustion are solely carbon dioxide and water. Catalytic combustion is frequently, but not exclusively, employed in applications where the concentration of the target species is below its lower ignition limit. When combustible gases are below their lower ignition limit then the mixture will not ignite when exposed to an ignition source. At such dilute concentrations, it is more efficient to use catalysts to react, convert, or combust, the target compounds because a catalyst can facilitate complete oxidation of the target species at a temperature significantly lower than the auto-ignition temperature of the molecule. It is generally desirable to develop catalysts that are capable of facilitating catalytic combustion at the lowest possible temperature in order to reduce the energy costs associated with operating a catalytic combustion system. Complete catalytic combustion of a target species can only occur when oxygen gas is found in molar stoichiometric excess; a condition which is easily met when the target species is present in trace quantity in air. Even in a reduced-oxygen environment, provided more moles of oxygen gas are present compared to moles of carbon atoms to be combusted then complete combustion of the target species can be realized. The required residence time of the gases in the space between the catalyst and the inlet of the turbine expansion zone is a function of the temperature of the gases exiting the catalyst. In any event, the gas residence time between the exit of the upstream oxidation zone and the inlet of the turbine gas expansion zone may be minimal and is such that at least a significant amount of the combustion takes place in the turbine gas expansion zone. If desirable, this residence time may be so small that at least a major portion of the total combustion occurring subsequent to the upstream catalyst zone is in the turbine expansion zone. The fuel is combusted in contact with free or molecular oxygen and free or molecular nitrogen. The fuel may occur or be obtained in admixture with components which are essentially inert in the oxidation system. The fuel has a relatively high energy content and is of a nature which permits the preparation of the oxidation feed streams.

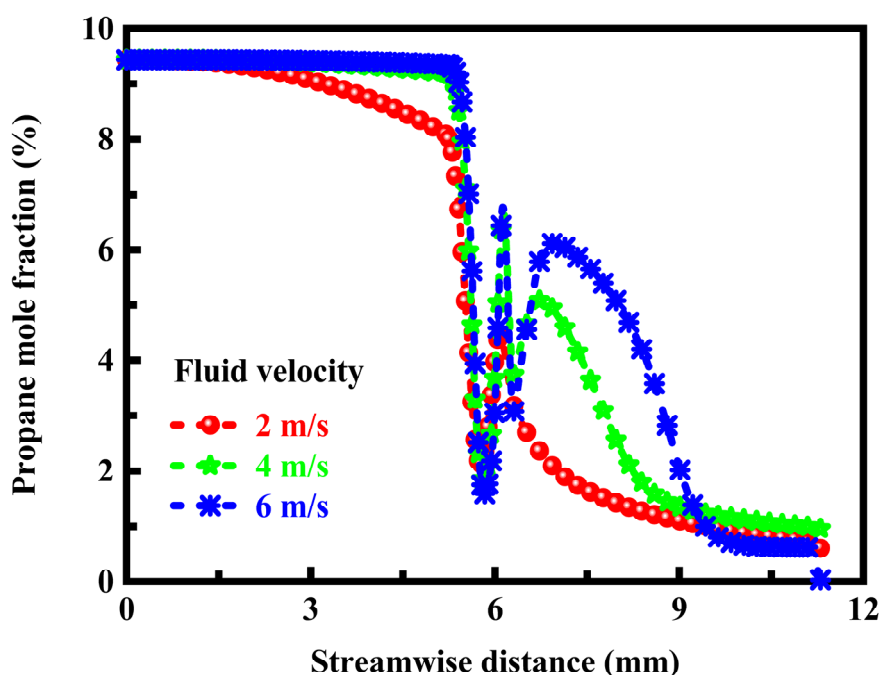


Figure 5. Propane mole fraction profiles along the fluid centerline of the gas turbine combustor under different fluid velocity conditions.

The homogeneous reaction rate profiles along the fluid centerline of the gas turbine combustor are presented in Figure 6 under different fluid velocity conditions. 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 [71, 72]. 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 [73, 74]. The channels through the unitary body or skeletal structure can be of any shape and size consistent with the desired superficial surface and should be large enough to permit relatively free passage of the gas mixture. The channels may be parallel, or generally parallel, and extend through the support from one side to an opposite side, such channels being separated from one another by preferably thin walls. The channels may also be multi-directional and may even communicate with one or more adjacent channels. The channel inlet openings can be distributed across essentially the entire face or cross-section of the support subject to initial contact with the gas to be oxidized. The unitary, skeletal structure support type of oxidation catalyst can be characterized by having a plurality of flow channels or paths extending therethrough in the general direction of gas flow. The flow channels need not pass straight through the catalyst structure and may contain flow diverters or spoilers. The skeletal structure support is preferably constructed of a substantially chemically inert, rigid, solid material capable of maintaining its shape and strength at high temperatures. The support may have a low thermal coefficient of expansion and low thermal conductivity. Often, the skeletal support is porous but its surface may be relatively non-porous, and it may be desirable to roughen its surface so that it holds the catalyst coating better, especially if the support is relatively non-porous. The support may be metallic or ceramic in nature or a combination thereof. The combustion gases are exhausted to atmosphere after giving up some of their remaining heat to the incoming air provided from the compressor. Operating the combustion process in a very lean condition 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. In a catalytic combustor, fuel is burned at relatively low temperatures. While emissions can be reduced by combustion at these temperatures, the utilization of catalytic combustion is 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. In conventional thermal combustion systems, a fuel and air in flammable proportions are contacted with an ignition source to ignite the mixture which will then continue to burn. Flammable mixtures of most fuels are normally burned at relatively high temperatures, 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 decreased 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 combustion systems utilizing a catalyst, there is little or no nitrogen oxides formed in a system which burns the fuel at relatively low temperatures. Such combustion heretofore has been generally regarded as having limited practicality in providing a source of power as a consequence of the need to employ amounts of catalyst so large as to make a system unduly large and cumbersome. Consequently, combustion utilizing a catalyst has been limited generally to such operations as treating tail gas streams of nitric acid plants. The catalytically supported thermal combustion involves essentially adiabatic combustion of a mixture of fuel and air or fuel, air, and inert gases in the presence of a solid oxidation catalyst operating at a temperature substantially above the instantaneous auto-ignition temperature of the mixture, but below a temperature which would result in any substantial formation of oxides of nitrogen under the conditions existing in the catalyst.

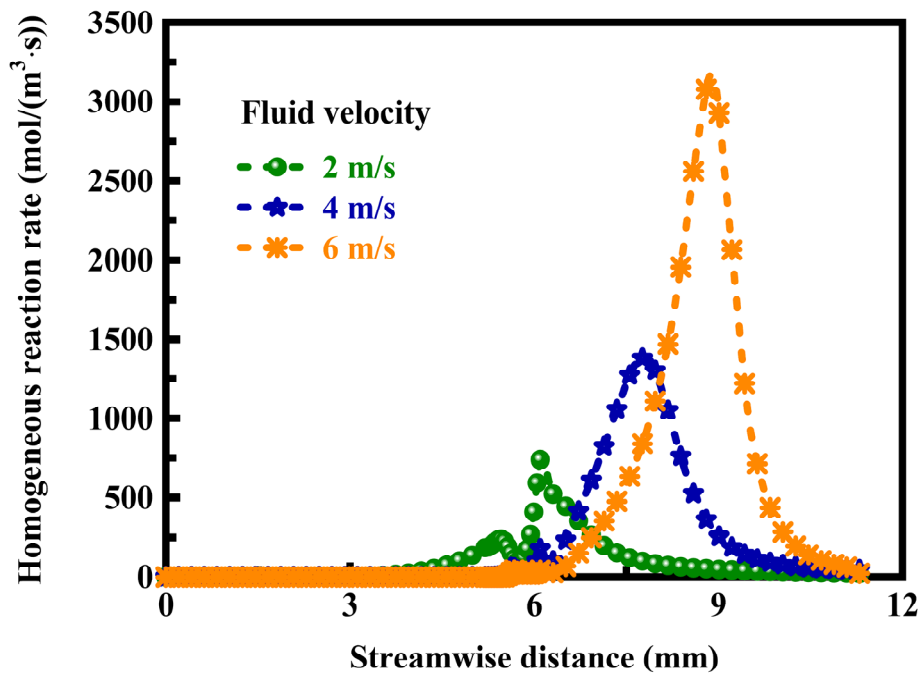


Figure 6. Homogeneous reaction rate profiles along the fluid centerline of the gas turbine combustor under different fluid velocity conditions.

The pressure contour plots in the gas turbine combustor are illustrated in Figure 7 for the reduction of nitrogen oxides emissions by heterogeneous catalysis. At relatively low temperatures, the catalytic reaction rate increases exponentially with temperature. As the temperature is raised further, the reaction rate enters a transition zone in which the rate at which the fuel and oxygen are being transferred to the catalytic surface begins to limit further increases in the reaction rate. As the temperature is raised still further, the reaction rate enters a so-called mass transfer limited zone in which the reactants cannot be transferred to the catalytic surface fast enough to keep up with the catalytic surface reaction and the reaction rate levels off regardless of further temperature increases. In the mass transfer limited zone, the reaction rate cannot be increased by increasing the activity of the catalyst because catalytic activity is not determinative of the reaction rate. The only apparent way to increase the reaction rate in a mass transfer limited reaction is to increase mass transfer. However, this typically requires an increase in the pressure drop across the catalyst and consequently a substantial loss of energy. Sufficient pressure drop may not even be available to provide the desired reaction rate. Of course, more mass transfer can be affected, and hence more energy can always be produced by increasing the amount of catalyst surface. However, this results in catalyst configurations of such size and complexity that the cost is prohibitive and the body of the catalyst is unwieldy. For example, in the case of gas turbine engines, the catalytic reactor might very well be larger than the engine itself. In general, conventional adiabatic, thermal combustion systems operate at such high temperatures in the combustion zone that undesirable nitrogen oxides, 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. 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. Catalytic combustion systems, though, are capable of achieving ultra-low emissions [75, 76]. However, catalytic combustion systems are not able to offer the accuracy and controllability of the air staging system over a wide range of power levels, fuel properties and ambient operating conditions [77, 78]. The system may be operated in different manners to allow for low and high-power operation, as well as according to a controlled schedule that may be

programmed. Under low power operation oxidation does not occur in the catalyst section. However, the mixing of the fuel and air in the fuel preparation and mixing section is enhanced by the presence of the catalyst. As the engine power level increases the compressor outlet air temperature will become high enough to activate the catalyst, and partial oxidation reactions will occur.

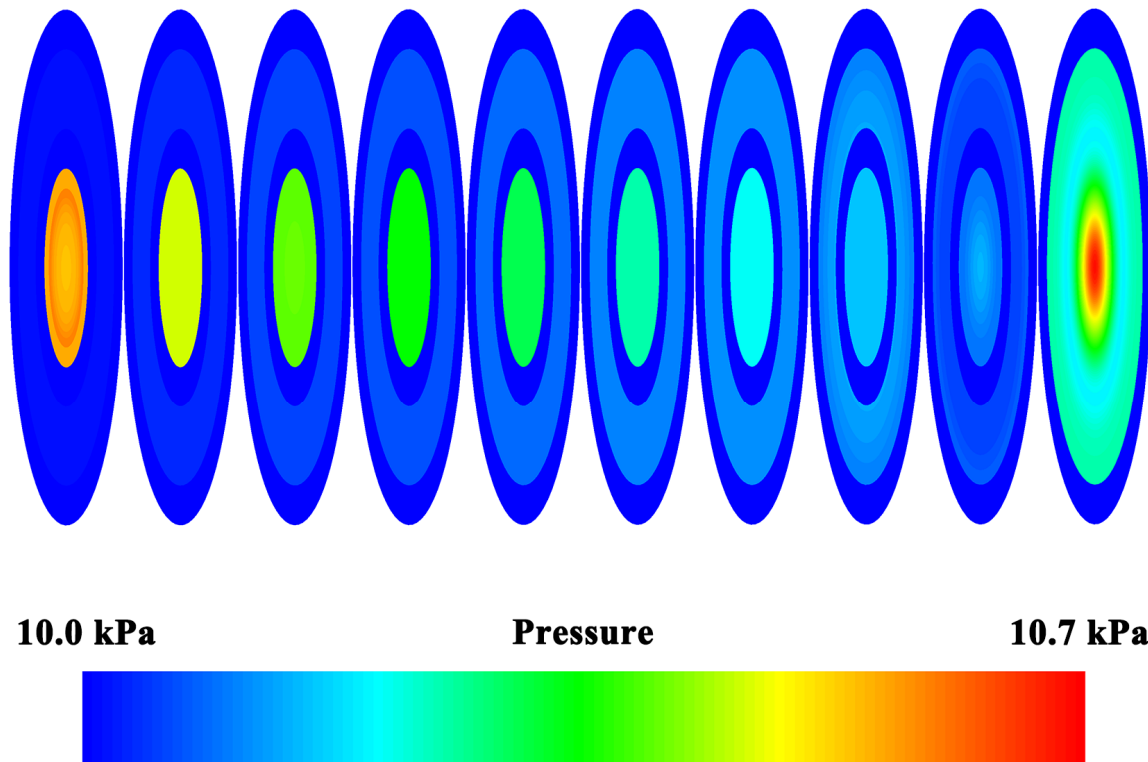


Figure 7. Pressure contour plots in the gas turbine combustor designed for the reduction of nitrogen oxides emissions by heterogeneous catalysis.

The chemical enthalpy contour plots in the gas turbine combustor are illustrated in Figure 8 for the reduction of nitrogen oxides emissions by heterogeneous catalysis. Nitrogen oxides formation is thermodynamically favored at high temperatures. 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. The limits of the operating temperature are governed largely by residence time and pressure. The instantaneous auto-ignition temperature of the mixture is defined herein to mean the temperature at which the ignition lag of the mixture entering the catalyst is negligible relative to the residence time in the combustion zone of the mixture undergoing combustion. Essentially adiabatic combustion means in this case that the operating temperature of the catalyst does not differ by more than about 80 K, from the adiabatic flame temperature of the mixture due to heat losses from the catalyst. Among the unique advantages of the catalytically supported thermal combustion in the presence of a catalyst is the fact that mixtures of fuel and air which are too fuel-lean for ordinary thermal combustion can be burned efficiently. Since the temperature of combustion for a given fuel at any set of conditions is dependent largely on the proportions of fuel, of oxygen available for combustion, and of inert gases in the mixture to be burned, it becomes practical to burn mixtures which are characterized by much lower flame temperatures. In particular, carbonaceous fuels can be burned very efficiently and at thermal reaction rates at temperatures in the range from about 1,200 K to about 2,000 K. At these temperatures, very little if any nitrogen oxides are formed. In addition, because the catalytically supported thermal combustion is stable over a wide range of mixtures, it is possible to select or control reaction temperature over a correspondingly wide range by

selecting or controlling the relative proportions of the gases in the mixture. The method may be carried out in various ways, including heating the catalyst body by electrical means, or by first thermally combusting a fuel and air mixture and applying the heat produced to the catalyst body. Once a catalyst temperature is reached at which the catalyst will function to sustain mass transfer limited operation, the combustion of fuel in the presence of the catalyst will bring it rapidly to the required operating temperature. Once operating temperature is reached, the catalyst will provide for sustained combustion of the fuel vapor. After the catalyst body reaches a temperature at which it will sustain mass transfer limited operation, the aforementioned application of heat to the catalyst body is no longer necessary and an admixture of unburned fuel and air is introduced into the system to establish the supported thermal combustion to provide a motive fluid for a turbine or heat to a furnace. No serious start-up problem normally is presented. The operation is substantially continuous and it is necessary to start the system only at infrequent intervals. Consequently, the substantial emissions of atmospheric pollutants which tend to occur in start-ups are not serious because of the small number of infrequent start-ups. While this pollution may be tolerated in stationary operations which are normally used continuously and for long periods of time, it cannot be tolerated in the vehicular type of installation where start-ups are frequent, due to intermittent operation [79, 80]. Also, the start-up must be rapid in order to be as efficient as in the conventional present-day automobile [81, 82]. The mixture of unburned fuel and air is not introduced to the catalyst body until it has reached a temperature at which it will sustain the desired rapid combustion. Such preferred procedure minimizes pollutant emissions during start-up. Once combustion in the zone containing the catalyst is achieved, the fuel-air admixture is passed to the catalyst at a gas velocity, prior to or at the inlet to the catalyst, in excess of the maximum flame propagating velocity. This avoids flash-back that causes the formation of nitrogen oxides [83, 84]. Various means have been proposed in the past to prevent such flash backs from traveling back to the gas supply source [85, 86]. Preferably, this velocity is maintained adjacent to the catalyst inlet [87, 88]. Suitable linear gas velocities are usually above about 8 meters per second, but considerably higher velocities may be required depending upon such factors as temperature, pressure, and composition.

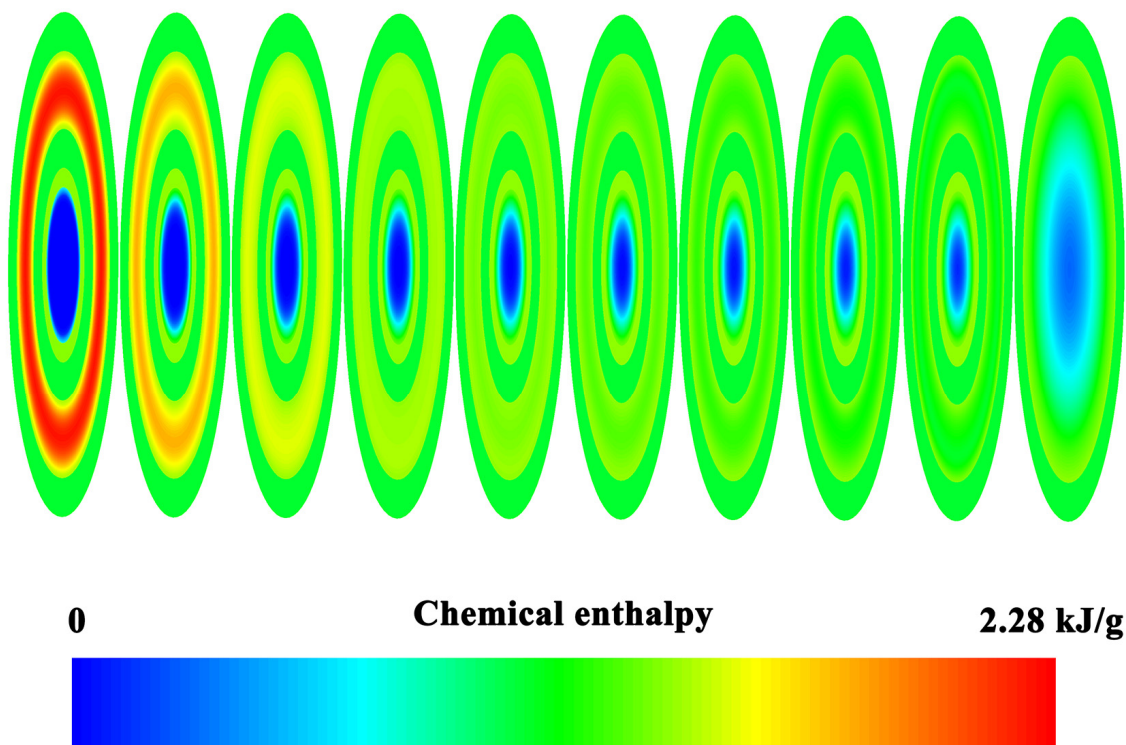


Figure 8. Chemical enthalpy contour plots in the gas turbine combustor designed for the reduction of nitrogen oxides emissions by heterogeneous catalysis.

4. Conclusions

Steady-steady simulations are performed using computational fluid dynamics. The fluid viscosity, specific heat, and thermal conductivity are calculated from a mass fraction weighted average of species properties, and the specific heat of each species is calculated using a piecewise polynomial fit of temperature. Natural parameter continuation is performed by moving from one stationary solution to another. Knowledge of critical parameters gains a fundamental understanding of the essential factors affecting the stability of the combustion process. Particular emphasis is placed upon the sustained combustion of at least a portion of fuel under essentially adiabatic conditions at a rate which surmounts the mass transfer limitation. The major conclusions are summarized as follows:

- 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.
- Flammable mixtures of carbonaceous fuels normally burn at relatively high temperatures, and substantial amounts of nitrogen oxides inevitably form if nitrogen is present.
- Complete catalytic combustion of a target species can only occur when oxygen gas is found in molar stoichiometric excess; a condition which is easily met when the target species is present in trace quantity in air.
- In combustion systems utilizing a catalyst, there is little or no nitrogen oxides formed in a system which burns the fuel at relatively low temperatures.
- In the mass transfer limited zone, the reaction rate cannot be increased by increasing the activity of the catalyst because catalytic activity is not determinative of the reaction rate.
- Among the unique advantages of the catalytically supported thermal combustion in the presence of a catalyst is the fact that mixtures of fuel and air which are too fuel-lean for ordinary thermal combustion can be burned efficiently.

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