

Highly exothermic process characteristics of catalytic reactors with integral heat exchange structures

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Abstract

In modern industrial practice, a variety of highly exothermic reactions are promoted by contacting of the reaction mixture in the gaseous or vapor phase with a heterogeneous catalyst. A need exists for improved catalytic structures employing integral heat exchange which will substantially widen the window or range of operating conditions under which such catalytic structures can be employed in highly exothermic processes like catalytic combustion or partial combustion. The highly exothermic process characteristics of catalytic reactors are investigated with integral heat exchange structures. Ethane mole fraction and gas-phase reaction rate profiles in catalytic reactors are presented, and ethane mole fraction, flow velocity, gas-phase reaction rate, and temperature contour plots are illustrated for catalytically supported thermal combustion systems. The present study aims to provide an improved reaction system and process for combustion of a fuel wherein catalytic combustion using a catalyst structure employing integral heat exchange affords a partially-combusted, gaseous product which is passed to a homogeneous combustion zone where complete combustion is promoted by means of a flame holder. Particular emphasis is placed upon the catalytic reactor configuration that allows the oxidation catalyst to be backside cooled by any fluid passing through the cooling conduits. The results indicate that the percentage of reaction completed in the exothermic catalytic reaction channel depends both upon the flow rate of the fuel-oxidant mixture through the exothermic catalytic reaction channel and upon the physical characteristics of the catalytic reactor. The tortuosity of the catalytic channels is increased by changing their cross-sectional area at a multiplicity of points along their longitudinal axes. The gas flow velocity entering the exothermic catalytic reaction channel should exceed the minimum required to prevent flashback into the fuel-oxidant stream upstream of the reactor if the fuel-oxidant mixture entering the exothermic catalytic reaction channel is within the limits of flammability. Catalytically-supported thermal combustion in the catalytic reactor is achieved by contacting at least a portion of the carbonaceous fuel intimately admixed with air with a solid oxidation catalyst having an operating temperature substantially above the instantaneous auto-ignition temperature of the fuel-air admixture. The film heat transfer coefficient provides useful means of characterizing the different flow geometries provided by the various flow channel configurations which distinguish the catalyst-coated channels from the catalyst-free channels of the catalyst structure. The total residence time in the combustion system should be sufficient to provide essentially complete combustion of the fuel, but not so long as to result in the formation of oxides of nitrogen.

Keywords: Catalytic reactors; Physical characteristics; Exothermic reactions; Heterogeneous catalysts; High temperatures; Thermal combustion

1. Introduction

In modern industrial practice, a variety of highly exothermic reactions are known to be promoted by contacting of the reaction mixture in the gaseous or vapor phase with a heterogeneous catalyst [1, 2].

In some cases, these exothermic reactions are carried out in catalyst-containing structures or vessels where external cooling must be supplied, in part, because of the inability to obtain sufficient heat transfer and the need to control the reaction within certain temperature constraints. In these cases, it is not considered practical to use a monolithic catalyst structure, where the unreacted portion of the reaction mixture supplies the cooling for the catalytic reaction, because existing catalyst structures do not provide an environment whereby the desired reaction can be optimized while removing the heat of reaction through heat exchange with unreacted reaction mixture under conditions where undesired reactions and catalyst overheating are avoided [3, 4]. Thus, the applicability of monolithic catalyst structures to many catalyzed exothermic reactions could clearly be enhanced if monolithic catalyst structures could be developed wherein the reaction zone environment and heat exchange between reacted and unreacted portions of the reaction mixture are improved.

Highly exothermic catalytic reactors with internal cooling have made rapid progress [5, 6]. While they have varying applications, the reactors are typified by exothermic reactions within the catalytic portion of the reactor and a cooling means to control the temperature within the catalytic portion to avoid a material failure, either of the substrate or the catalyst [7, 8]. Cooling in these reactors can be accomplished by a number of means, including placing the catalyst in a backside-cooled relationship with the cooling agent [9, 10]. A backside-cooling arrangement is particularly suitable for catalytic reactions that are both rapid and highly exothermic, for instance, catalytic combustion [11, 12]. In this arrangement, the catalyst substrate, typically a metal foil, is coated with an oxidation catalyst on only one side, the opposite side or backside remaining free of oxidation catalyst [13, 14]. The substrate is shaped and assembled, before or after catalyst coating, to create separate channels for exothermic reaction, in the channels coated with oxidation catalyst, or for cooling, in the channels free of oxidation catalyst [15, 16]. Fluid passing through the cooling channels removes a portion of the heat generated in the exothermic reaction channels.

In a typical catalytic combustor, active catalysts being supported on various substrates provide an effective means of initiating and stabilizing the combustion process when they are used with suitable mixtures of fuel and air [17, 18]. These combustion catalysts have several desirable characteristics: they are capable of minimizing nitrogen oxides emission and improving the pattern factor [19, 20]. However, one of their limitations is that their maximum operating temperature tends to be only marginally acceptable as a catalytic combustor inlet temperature [21, 22]. This limitation is inherent in the way the typical catalytic combustor operates. Catalysts initiate the combustion reaction at their surfaces and at temperatures lower than normal ignition temperature. However, once the reaction is initiated, it continues in the gas stream and persists beyond the catalyst in the form of afterburning [23, 24]. Simultaneously, the catalyst substrate temperature increases, resulting in an accelerated reaction which moves the reaction zone further upstream in the catalyst. The result may be damage of the catalyst and catalyst substrate if the fuel-air ratio is such as to give an excessive catalyst outlet temperature [25, 26]. There is an example of a backside-cooled catalytic reactor for use in a catalytic combustion system, with the basic method of splitting a given fuel-air mixture flow into catalytic and non-catalytic passages. This example is the use of a ceramic substrate with multiple parallel channels, generally of the same shape and size, in which the walls which border and define each catalytic channel are coated with an oxidation catalyst on the sides facing the catalytic channel, but are not coated with an oxidation catalyst on the sides facing adjacent non-catalytic channels. By this method, the percentage of total reactants catalyzed in the reactor is no greater than the percentage of catalytic channels [27, 28]. The average temperature rise through the reactor is thus limited [29, 30]. In addition, the wall temperatures of catalytic channels bordering adjacent non-catalytic channels are controlled through the use of backside cooling.

There are a number of ways of controlling the temperature, such as by dilution with excess air, by

controlled oxidation using one or more catalysts, or by staged combustion using variously lean or rich fuel mixtures [31, 32]. Combinations of these methods are also known. One widely attempted method is the use of multistage catalytic combustion [33, 34]. Most of these processes utilize multi-section catalysts with metal or metal oxide catalysts on ceramic catalyst carriers [35, 36]. It is, however, difficult to control the temperatures in these processes. Since the object of each of the processes is to produce a maximum amount of heat in a form which can be efficiently used in some later process, the combustion steps are essentially adiabatic. Consequently, a minor change in any of fuel rate, air rate, or operating processes in an early stage will cause significant changes in the latter stage temperatures. Very high temperatures place thermal strain on downstream catalytic elements [37, 38]. Platinum group metals are considered useful in catalytic combustion processes [39, 40]. A metal substrate is used for improved heat conduction to the backside cooling fluid, and for greater resistance to thermal shock. Aluminum-containing steels are cited as being preferred. This example is the use of non-similar shape and size channels, so that the flow split between catalytic and non-catalytic channels can be varied while retaining approximately half catalytic channels and half non-catalytic channels. Despite these changes, the fundamental structure, namely a multitude of catalytic channels and adjacent non-catalytic channels, is retained [41, 42]. The structure is refined in which periodic alterations in channel shape provide different wall heat transfer rates in the catalytic channels and non-catalytic channels [43, 44]. Again, however, the fundamental structure, namely a multitude of catalytic channels and adjacent non-catalytic channels, is retained. Furthermore, while the catalytic and non-catalytic channels have different shape and tortuosity, the average channel properties over some finite lengths are not varied in the longitudinal direction, so that the catalytic reactors taught are effectively one-dimensional or two-dimensional in terms of channel flow properties such as bulk heat transfer coefficient, velocity, or average cross-sectional shape or area.

There is also a clear need to improve the operability of monolithic catalyst structures in areas where they are currently used or proposed for use, such as the combustion or partial combustion of fuels or the catalytic treatment of exhaust emissions from internal combustion engines, to widen the range of operating conditions at which the desired catalytic conversions can be achieved. A catalyst structure made up of a series of adjacently disposed catalyst-coated and catalyst-free channels for passage of a flowing reaction mixture can be employed, wherein the catalytic and non-catalytic channels share a common wall such that integral heat exchange can be used to dissipate the reaction heat generated on the catalyst and thereby control or limit the temperature of the catalyst [45, 46]. That is, the heat produced on the catalyst in any given catalyst-coated channel flows through the common wall to the opposite non-catalytic surface to be dissipated into the flowing reaction mixture in the adjacent catalyst-free channel. The configuration of the catalytic channels differs from the non-catalytic channels in one or more critical respects, including the tortuosity of the flow channel, such that, when applied to catalytic combustion, catalytic and homogeneous combustion is promoted within the catalytic channels and not promoted or substantially limited in the non-catalytic channels while heat exchange is otherwise optimized [47, 48]. These uniquely configured catalyst structures substantially widen the window of operating parameters for catalytic combustion and partial combustion processes.

In cases where the integral heat exchange structure is used to carry out catalytic partial combustion of a fuel followed by complete combustion after the catalyst, the catalyst must burn a portion of the fuel and produce an outlet gas sufficiently hot to induce homogeneous combustion after the catalyst [49, 50]. In addition, it is desirable that the catalyst not become too hot since this would shorten the life of the catalyst and limit the advantages to be gained from this approach. As the operating condition of the catalyst is changed, it is noted with the integral heat exchange structures that operating window of such catalysts are limited. That is, that the gas velocity or mass flow rate must be within a certain range to prevent catalyst overheating. In general, the backside-cooled catalytic reactors include a multitude of

catalytic channels, where each individual catalytic channel is in essence a separate catalytic reactor [51, 52]. As a result, variations in fuel-air ratio from channel to channel, due to imperfect premixing, for instance, can lead to different degrees of combustion and heat release in different channels. Likewise, variations in inlet temperature from channel to channel can also lead to variations in combustion behavior in different channels. Rate of reaction, catalyst light-off length, and maximum gas or surface temperature can all be affected by the temperature and fuel-air ratio at a channel inlet [53, 54]. In addition, manufacturing tolerances may result in unequal physical properties of different channels. Properties which may vary include channel size, wall thickness, catalyst or washcoat thickness, and catalyst loading; each of these may affect combustion behavior [55, 56]. In essence, multiple catalytic channels can produce widely varying degrees of catalytic combustion.

Because there is no mixing between separate catalytic channels in the backside-cooled reactors, the reactors suffer the above-mentioned disadvantages of sensitivity to premixing, for instance, the fuel-air ratio, and sensitivity to inlet temperature uniformity [57, 58]. Given that all real systems have some level of gas-stream non-uniformity, these sensitivities translate to a narrowed operating range. Structures and methods that provide an un-partitioned exothermic catalytic reaction channel and multiple cooling channels offer superior performance [59, 60]. The un-partitioned exothermic catalytic reaction channel allows for continual mixing of the fuel-oxidant stream within the channel leading to a more uniform combustion and a wider operating range. A catalytic reactor may employ an exothermic catalytic reaction channel cooled by numerous cooling channels, where the cooling fluid is a portion of the ultimate fuel-oxidant-product mixture [61, 62]. The structure of the reactor is more flexible, facilitating cross-stream area changes in the streamwise or longitudinal direction, since there is no constraint that walls contact each other to form multiple catalytic channels [63, 64]. Thus, the reactor can be used to vary the bulk fluid properties in the streamwise or longitudinal direction via cross-stream area changes. In particular, it may be desirable to reduce the velocity of the fuel-air mixture after it has entered the exothermic catalytic reaction channel, to provide greater residence time for reaction within the reactor, while maintaining sufficient velocity at the reactor inlet to prevent flashback to the fuel-oxidant mixture upstream of the reactor [65, 66]. Therefore, it is clear that a need exists for improved catalytic structures employing integral heat exchange which will substantially widen the window or range of operating conditions under which such catalytic structures can be employed in highly exothermic processes like catalytic combustion or partial combustion. It is necessary to capitalize on certain critical differences in the configuration of the catalytic and non-catalytic passageways or channels in an integral heat exchange structure to materially widen the operating window for such catalysts.

The present study relates to a catalyst structure employing integral heat exchange in an array of longitudinally disposed, adjacent reaction passageways or channels which are either catalyst-coated or catalyst-free, as well as a method for using the catalyst structure in highly exothermic processes, such as combustion or partial combustion processes. More particularly, the present study is directed to such a catalyst structure employing integral heat exchange wherein the catalytic and non-catalytic channels differ from each other in certain critical respects whereby the exothermic reaction in the catalytic channels and heat exchange between the catalytic and non-catalytic channels are optimized while undesired exothermic reaction in the non-catalytic channels is suppressed. The present study is focused primarily upon the integral heat exchange in an array of longitudinally disposed adjacent reaction passageways or channels, which are either catalyst-coated or catalyst-free, wherein the configuration of the catalyst-coated channels differs from the non-catalyst channels such that, when applied in exothermic reaction processes, such as catalytic combustion, the desired reaction is promoted in the catalytic channels and substantially limited in the non-catalyst channels. The present study aims to provide an improved reaction system and process for combustion of a fuel wherein catalytic

combustion using a catalyst structure employing integral heat exchange, preferably the improved catalyst structures, affords a partially-combusted, gaseous product which is passed to a homogeneous combustion zone where complete combustion is promoted by means of a flame holder. Particular emphasis is placed upon the catalytic reactor configuration that allows the oxidation catalyst to be backside cooled by any fluid passing through the cooling conduits.

2. Methods

In the present study, a catalytic reactor employs an un-partitioned exothermic catalytic reaction channel and multiple cooling conduits passing through the exothermic catalytic reaction channel. An oxidation catalyst is deposited on the exterior surfaces of the conduits within the exothermic catalytic reaction channel. This placement of the catalyst allows the oxidation catalyst to be backside cooled by any fluid passing through the cooling conduits. Backside cooling means that at each location where an oxidation catalyst is deposited on one surface of a wall no oxidation catalyst is deposited on an adjacent or opposite surface in contact with the cooling fluid, and a portion of the heat generated by reaction on the oxidation catalyst is conducted through the substrate to the adjacent or opposite surface that is in direct contact with the cooling fluid [67, 68]. The catalytic reactor can be made in numerous configurations with the following common elements [69, 70]. A casing forms the outer boundary, which can be of any shape. The reactor casing can be a single fabricated component, or can consist of two or more components joined together. Two or more conduits are placed within the casing such that one fluid stream can traverse an un-partitioned channel, the exothermic catalytic reaction channel, defined by the interior surface of the casing and the exterior surfaces of the conduits, and a number of separate fluid streams can traverse the passages defined by the interior surfaces of the conduits, without mixing occurring between fluid in the exothermic catalytic reaction channel and fluid in the conduits' interior passages. A heat transfer relationship exists between the fluid in the exothermic catalytic reaction channel and the fluids in the conduits' interior passages.

The structure of the integral heat exchange is represented physically in Figure 1 in which the catalytic reactor employs an arrangement of catalyzed and non-catalyzed substrate passages for providing passive cooling of the catalytic reactor. Such cooling permits the catalyst to function with higher reaction temperatures than otherwise possible. By applying a catalytic coating to a fraction of the walls of the parallel passages of a combustion catalyst substrate, the uncoated passages act to cool the common walls exposed to the reacting flow in the coated passages. Accordingly, the present design is directed to a catalytic reactor unit, which comprises the combination of a substrate composed of a plurality of generally parallel passages open at their opposite ends and exposed to a heated flow of fuel and air mixture therethrough and selected ones of the passages being coated with a catalyst and others of the passages being free of the catalyst so as to provide the substrate with an arrangement of catalyzed passages in which the mixture is catalytically reacted and non-catalyzed passages in which the mixture is substantially not reacted but instead provides passive cooling of the substrate. The substrate is composed of a plurality of intersecting walls defining the generally parallel passages being aligned in rows and columns. The walls have sections which border and define the respective passages. Each wall section is in common with two adjacent passages and has a pair of oppositely-facing surface regions, one of which is exposed to one of the two adjacent passages and the other exposed to the other of the two adjacent passages. The solid catalyst can have various forms and compositions and can be the types used to oxidize fuels in the presence of molecular oxygen. The catalyst can be in the form of relatively small, solid particles of various sizes and shapes, often in sizes below about one inch in the largest dimension, with a plurality of such particles being arranged together to form one or more catalyst masses or beds in the combustion zone. The catalyst is preferably of larger form and has a skeletal structure with gas flow paths therethrough.

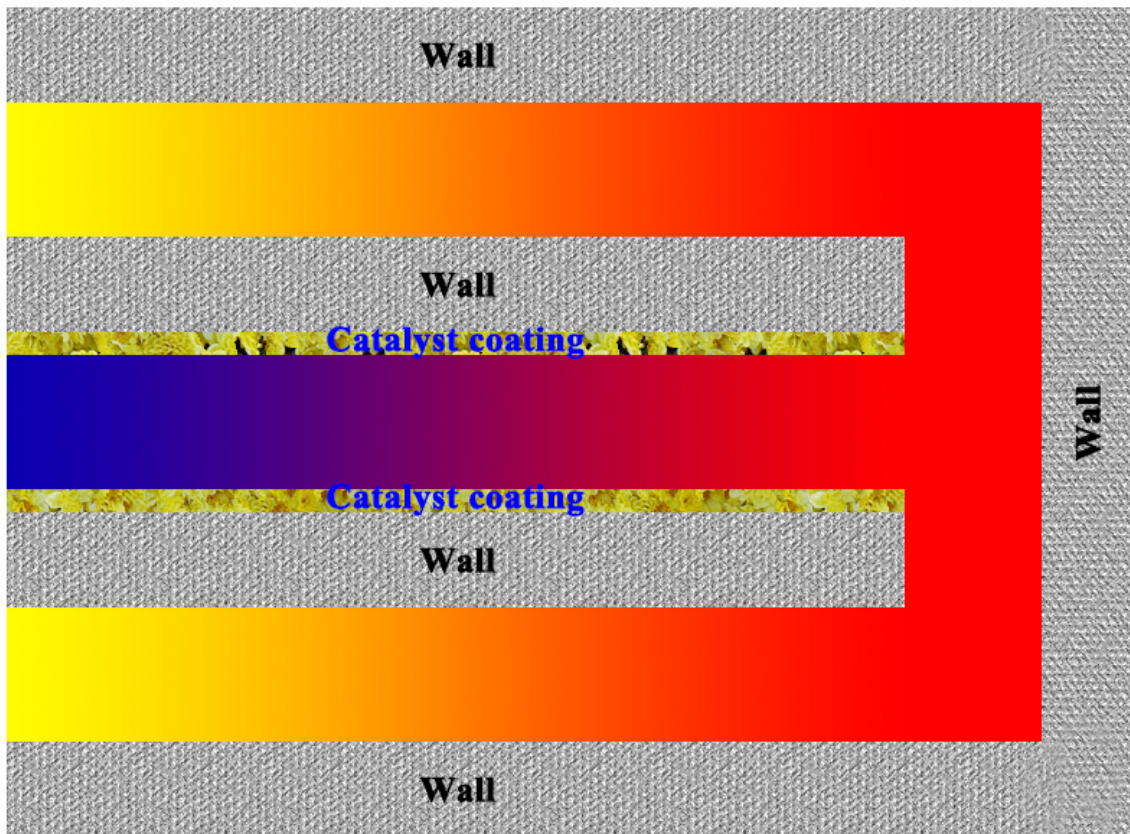


Figure 1. Physical representation of the integral heat exchange structure in which the catalytic reactor employs an arrangement of catalyzed and non-catalyzed substrate passages for providing passive cooling of the catalytic reactor.

The catalyst generally has one or more metal containing components which are catalytically active towards promoting the desired oxidation reactions. Furthermore, the catalyst coating is applied on selected ones of the wall surface regions exposed to certain ones of the passages, whereas selected others of the wall surfaces exposed to certain others of the passages are free of the catalyst coating. In such manner, the substrate is provided with the arrangement of catalyzed passages in which the mixture is catalytically reacted and non-catalyzed passages in which the mixture is substantially not reacted but instead provides passive cooling of the substrate. Also, the selected ones of the surface regions have catalyst coating thereon and the selected others of the surface regions being free of catalyst coating are on common wall sections such that a catalytic reaction can occur in those passages bordered by the catalyzed surface regions concurrently as cooling occurs in those passages being adjacent thereto and bordered by the non-catalyzed surface regions. Any arrangement of catalyzed and non-catalyzed passages is possible. In one arrangement, the catalyzed to non-catalyzed passages are in a ratio of one-to-one. In another arrangement, they are in a ratio of three-to-one.

The integral heat exchange within the catalytic reactor is illustrated schematically in Figure 2 in which at least a portion of the thermal combustion of the fuel takes place in the expansion zone of the catalytic reactor to counteract the cooling effect of the expansion of the gases. The catalyst structure desirably comprises a support and a combustion catalyst. The support is preferably metal. It may be corrugated and rolled or otherwise assembled in such a way that the combusting gas flows from end to end through the length of the corrugations. The catalyst is placed only on a portion of the corrugations in such a way that the catalyst is in heat exchange relationship to a surface having no catalyst. The heat produced on the catalyst flows thorough the structure wall to the flowing gas at the opposite non-catalytic wall. The heat also flows to the adjacent combusted gas. The catalyst and its structure

provide an exceptionally stable and temperature moderated structure having long life. The catalyst structure is particularly useful in fuel combustion processes and the fuel combustion processes. The structure is preferably a platinum-group, metal-based catalyst on a metal monolith. The metal monolith is assembled from or fabricated from metallic materials having a catalytic surface and an adjunct non-catalytic surface. One side of the catalyst structure component has catalyst upon it and the other side of the catalyst structure component is essentially catalyst-free. The preferred supports for this catalytic zone are metallic. Metallic supports in the form of honeycombs, spiral rolls of corrugated sheet, columnar, or other configurations having longitudinal channels or passageways permitting high space velocities with a minimal pressure drop are desirable in this service. They are malleable, may be mounted and attached to surrounding structures more readily, and offer lower flow resistance due to walls which are thinner than can be readily manufactured in ceramic supports. Another practical benefit attributable to metallic supports is the ability to survive thermal shock. Such thermal shocks occur in gas turbine operations when the turbine is started and stopped and, in particular, when the turbine must be rapidly shut down. In any event, the catalyst is deposited, or otherwise placed, on at least a portion of the walls within the metal supports' channels or passageways in the amounts specified. By the phrase "at least a portion" is meant that each channel need not be coated along its entire length. In some instances, catalyst placement along a portion of the length of the channel will be sufficient. Several types of support materials are satisfactory in this service: aluminum, aluminum-containing or aluminum-treated steels, ferrous alloys, certain stainless steels, or any high temperature metal alloy, including nickel or cobalt alloys where a catalyst layer can be deposited on the metal surface.

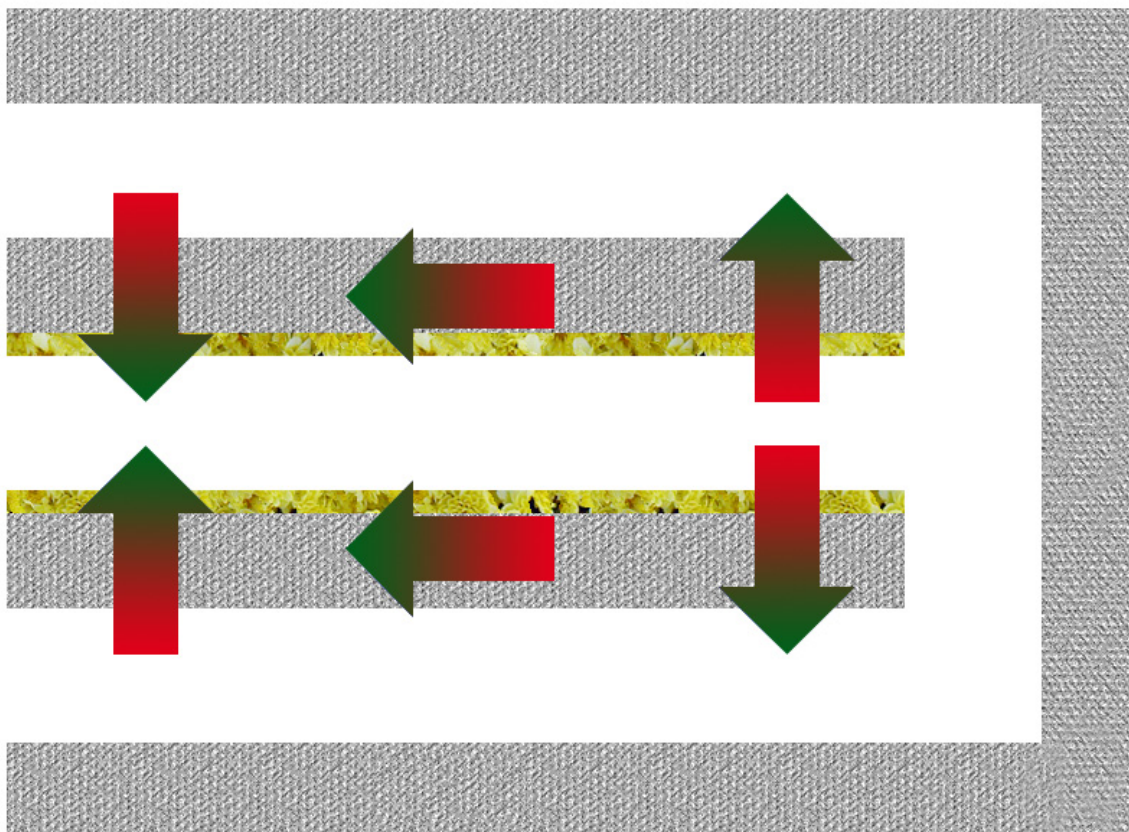


Figure 2. Schematic illustration of the integral heat exchange within the catalytic reactor in which at least a portion of the thermal combustion of the fuel takes place in the expansion zone of the catalytic reactor to counteract the cooling effect of the expansion of the gases.

The preferred materials are aluminum-containing steels. These steels contain sufficient dissolved aluminum so that, when oxidized, the aluminum forms alumina whiskers, crystals, or a layer on the

steel's surface to provide a rough and chemically reactive surface for better adherence of the washcoat. The washcoat may be applied using an approach, for instance, the application of gamma-alumina, zirconia, silica, or titania materials or mixed sols of at least two oxides containing aluminum, silicon, titanium, zirconium, and additives such as barium, cerium, lanthanum, chromium, or a variety of other components. For better adhesion of the washcoat, a primer layer can be applied containing hydrous oxides such as a dilute suspension of pseudo-boehmite alumina. The primed surface may be coated with a gamma-alumina suspension, dried, and calcined to form a high surface area adherent oxide layer on the metal surface. Most desirably, however is the use of a zirconia sol or suspension as the washcoat. Other refractory oxides, such as silica and titania, are also suitable. Most preferred for some platinum group metals, notably palladium, is a mixed zirconia-silica sol where the two have been mixed prior to application to the support. Silica appears to allow the zirconia to maintain the catalyst's stability, for instance, its activity, for a long period of time. The washcoat may be applied in the same fashion one would apply paint to a surface, for instance, by spraying, direct application, and dipping the support into the washcoat material. Aluminum structures are also suitable for use in this design and may be treated or coated in essentially the same manner. Aluminum alloys are somewhat more ductile and likely to deform or even to melt in the temperature operating envelope of the process. Consequently, they are fewer desirable supports but may be used if the temperature criteria can be met.

The catalyst structure is comprised of a series of adjacently disposed catalyst-coated and catalyst-free channels for passage of a flowing reaction mixture wherein the channels at least partially coated with catalyst are in heat exchange relationship with adjacent catalyst-free channels and wherein the catalyst-coated channels have a configuration which forms a more tortuous flow passage for the reaction mixture than the flow passage formed by the catalyst-free channels. For convenience herein the terms "catalyst-coated channels" or "catalytic channels" in the catalyst structures may refer to single channels or groupings of adjacent channels which are all coated with catalyst on at least a portion of their surface, in effect a larger catalytic channel subdivided into a series of smaller channels by catalyst support walls or pervious or impervious barriers which may or may not be coated with catalyst. Similarly, the "catalyst-free channels" or "non-catalytic channels" may be a single channel or grouping of adjacent channels which are all not coated with catalyst, that is, a larger catalyst-free channel subdivided into a series of smaller channels by catalyst support walls or pervious or impervious barriers which are not coated with catalyst. In this regard, increased tortuosity of the flow passages formed by the catalyst-coated channels means that the catalyst-coated channels are designed such that at least a portion of the reaction mixture entering the catalyst-coated channels will undergo more changes in direction of flow as it traverses the length of the channel than will any similar portion of reaction mixture entering the catalyst-free channels. In practice, the increased tortuosity of the flow passage in the catalyst-coated channels can be accomplished by a variety of structural modifications to the channels including periodically altering their direction and changing their cross-sectional area along their longitudinal axis while the catalyst-free channels remain substantially straight and unaltered in cross-sectional area. Preferably the tortuosity of the catalyst-coated channels is increased by varying their cross-sectional area through repeated inward and outward bending of channels walls along the longitudinal axis of the channels or through the insertion of flaps, baffles or other obstructions at a plurality of points along the longitudinal axes of the channels to partially obstruct and divert the direction of reaction mixture flow in the channels.

3. Results and discussion

The ethane mole fraction profiles along the fluid centerline of the catalytic reactor are presented in Figure 3 with an integral heat exchange structure in an array of longitudinally disposed, adjacent reaction passageways or channels which are either catalyst-coated or catalyst-free. Regardless of the

specific catalytic reactor configuration, the reactor should be capable of providing good contact between the oxidation catalyst and the fuel-oxidant mixture in the exothermic catalytic reaction channel. For catalytic combustion applications, it is preferred that the reactor be sized such that the reaction of the fuel-oxidant mixture in the exothermic catalytic reaction channel should proceed more than 50 percent of the way to completion before exiting. For fuel-lean mixtures, this means that more than 50 percent of the fuel entering the channel should be consumed. Most preferably, more than 80 percent of the fuel entering the exothermic catalytic reaction channel should burn before exiting. The percentage of reaction completed in the exothermic catalytic reaction channel depends both upon the flow rate of the fuel-oxidant mixture through the exothermic catalytic reaction channel and upon the physical characteristics of the catalytic reactor. The chemical composition of the fuel-oxidant mixture may also affect the percentage of reaction completed, particularly if the rate of chemical reaction is significantly limiting when compared to the rate of mass transfer to the catalyst surface. With regard to percentage of reaction completed, important physical characteristics within the exothermic catalytic reaction channel include the rate of mass transfer to the oxidation catalyst surface, the ratio of oxidation catalyst surface area to reaction channel volume, and the activity of the oxidation catalyst. The catalyst-coated substrate may be fabricated from any of various high temperature materials. High temperature metal alloys are preferred, particularly alloys composed of iron, nickel, and cobalt, in combination with aluminum, chromium, and other alloying materials. High temperature nickel alloys are especially preferred. Other materials which may be used include ceramics, metal oxides, intermetallic materials, carbides, and nitrides. Metallic substrates are most preferred due to their excellent thermal conductivity, allowing effective backside cooling of the catalyst. The support material, preferably metallic or intermetallic, may be fabricated using conventional techniques to form a honeycomb structure, spiral rolls or stacked patterns of corrugated sheet, sometimes inter-layered with sheets which may be flat or of other configuration, or columnar or other configuration which allow for the presence of adjacent longitudinal channels which are designed to present flow channels in accordance with the design criteria set forth above. If intermetallic or metallic foil or corrugated sheet is employed, the catalyst will be applied to only one side of the sheet or foil or in some cases the foil or sheet will remain uncoated depending on the catalyst structure design chosen. Applying the catalyst to only one side of the foil or sheet, which is then fabricated into the catalyst structure, takes advantage of the integral heat exchange concept, allowing heat produced on the catalyst to flow through the structure wall into contact with the flowing gas at the opposite non-catalytic wall thereby facilitating heat removal from the catalyst and maintaining the catalyst temperature below the temperature for complete adiabatic reaction. In this regard, the adiabatic combustion temperature is the temperature of the gas mixture if the reaction mixture reacts completely and no heat is lost from the gas mixture. The critical difference in the design of the catalytic versus non-catalytic channels for the catalytic structure, in its most basic terms, is that the catalytic channels are designed so that the reaction mixture flow passages defined by the catalytic channels possess a higher or increased tortuosity over the corresponding flow passages formed by the non-catalytic channels. The concept of tortuosity, as used herein, is defined as the difference between the length of the path which a given portion of reaction mixture will travel through the passage formed by the channel as a result of changes in direction of the channel and changes in channel cross-sectional area versus the length of the path traveled by a similar portion of the reaction mixture in a channel of the same overall length without changes in direction or cross-sectional area, in other words, a straight channel of unaltered cross-sectional area. The deviations from a straight or linear path, of course, result in a longer or more tortuous path and the greater the deviations from a linear path the longer the traveled path will be. When applied to the catalyst structures, differences in tortuosity between catalytic and non-catalytic channels is determined by comparing the average tortuosity of all of the catalytic channels in the structure to the average tortuosity of all of the non-catalytic channels in the structures.

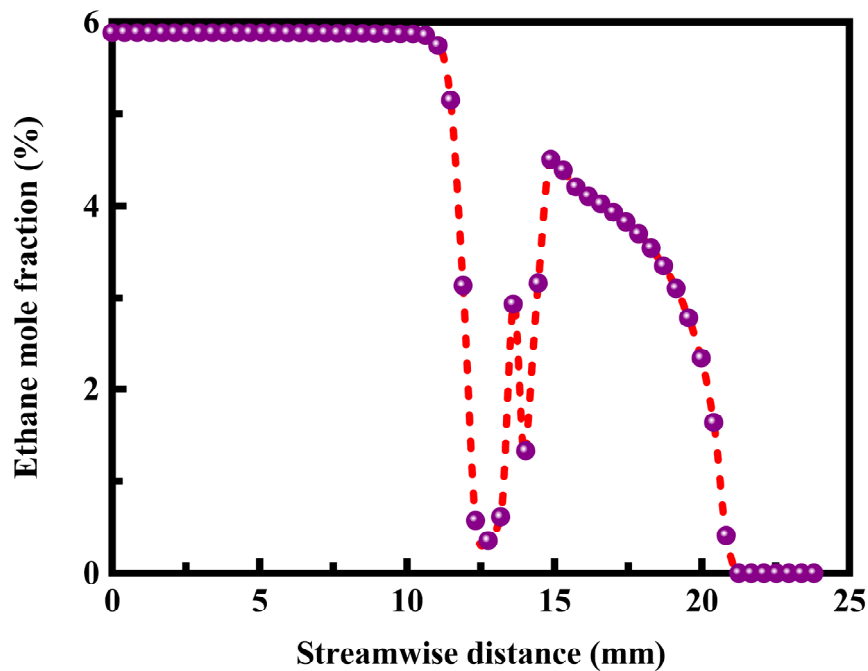


Figure 3. Ethane mole fraction profiles along the fluid centerline of the catalytic reactor with an integral heat exchange structure in an array of longitudinally disposed, adjacent reaction passageways or channels which are either catalyst-coated or catalyst-free.

The ethane mole fraction contour plots in the catalytic reactor are illustrated in Figure 4 with an integral heat exchange structure in an array of longitudinally disposed, adjacent reaction passageways or channels which are either catalyst-coated or catalyst-free. The design requires that the fluid exiting the exothermic catalytic reaction channel exit and the fluid exiting the conduit outlets must come into contact to permit some degree of mixing to form the ultimate combustion mixture. To accomplish this, the exothermic catalytic reaction channel exit and the conduit outlets must be proximately located. Proximately located means that the exits are spatially located to permit the fluids exiting the exit and the outlets to come in contact so some degree of mixing is possible prior to ultimate combustion. The design does not require symmetry in securing the upstream and downstream ends of the conduits and the different securing structures could be combined in a single structure. For instance, the conduits could be secured at the upstream end by a retaining structure and at the downstream end by an expanded conduit structure. In addition, the casing and the conduits may operate at different temperatures, resulting in different amounts of thermal expansion. Thus, clearance may be provided between the conduits and the upstream or downstream retainers, to allow for thermal growth of the conduits. The conduits may be secured to each other and to the casing only at one end, as for example at the inlet or upstream end, with the other end free to move longitudinally so that differential thermal growth of the conduits may be accommodated. If a clearance is allowed between the conduits and the retainers, or if differential thermal growth between adjacent conduits is expected, the conduits' expanded sections should be of sufficient axial length that lateral support and positioning of the conduits is provided even when adjacent tubes move in opposite axial directions to the maximum extent allowed by the clearance space or the expected difference in thermal growth. If the conduits penetrate or pass through the retainers, respectively, and if they are laterally positioned by the retainers, the expanded sections need not provide lateral support to the conduits. In this case, it may also be advantageous to allow the conduits to slide freely through at least one of the retainers, to allow for thermal expansion of the conduits. An oxidation catalyst is applied to the exterior of the conduits within the exothermic catalytic reaction channel. While the entire exterior of the conduits could be catalyst coated, as a practical matter catalyst coating should not be applied where the channels touch one

another or the casing. This allows close fabrication and assembly tolerances, without concern for variable coating thickness, and allows for welding or brazing of metal-to-metal contact points, if desired. In the catalyst structures, a variety of structure modifications can be made to the channels coated with catalyst to increase their tortuosity relative to the non-catalytic channels. In particular, the tortuosity of the catalytic channels can be increased by periodically changing their direction, for instance, by using channels having a zig-zag or wavy configuration or by repeatedly changing their cross-sectional area through periodic inward and outward bending of channel walls along their longitudinal axis or through the insertion of flaps, baffles or other obstructions to partially obstruct or divert the direction of reaction mixture flow at a plurality of points along the longitudinal axis of the channel. In some applications, it may be desirable to use a combination of changes in direction and changes in cross-sectional area to achieve an optimum difference in tortuosity but in all cases the tortuosity of the non-catalytic channel will be less on average than the tortuosity of the catalytic channels. Preferably, the tortuosity of the catalytic channels is increased by changing their cross-sectional area at a multiplicity of points along their longitudinal axes. One preferred way of accomplishing this change in tortuosity for the catalytic channels involves the use of a stacked arrangement of non-nesting corrugated sheets of catalyst support material which are corrugated in a herringbone pattern with at least a portion of one side of a given corrugated sheet facing and stacked against another corrugated sheet being coated with catalyst such that the stacked sheets in question form a plurality of catalytic channels. By stacking the corrugated sheets together in a non-nesting fashion, the channels formed by the stacked sheets alternately expand and contract in cross-sectional area along their longitudinal axis due to the inwardly and outwardly bending peaks and valleys formed by the herringbone pattern of the corrugated sheets. Other preferred ways of changing the cross-sectional area of the catalyst-coated channels include the periodic placement of flaps or baffles on alternate sides of the channels along their longitudinal axis or the use of screens or other partial obstructions in the flow path formed by the catalytic channels.

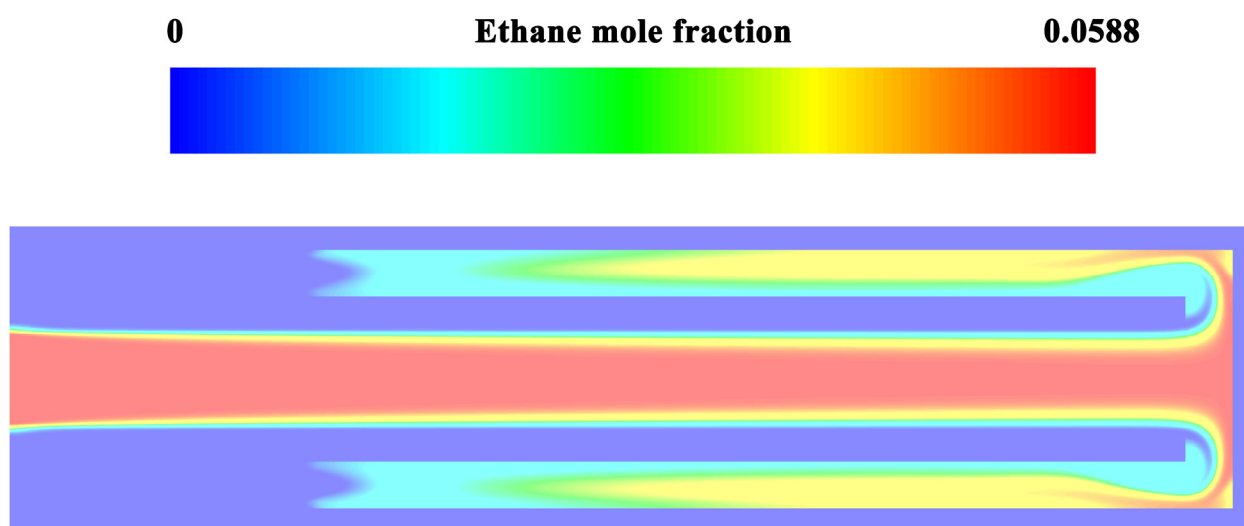


Figure 4. Ethane mole fraction contour plots in the catalytic reactor with an integral heat exchange structure in an array of longitudinally disposed, adjacent reaction passageways or channels which are either catalyst-coated or catalyst-free.

The velocity contour plots of flow in the catalytic reactor are illustrated in Figure 5 with an integral heat exchange structure in an array of longitudinally disposed, adjacent reaction passageways or channels which are either catalyst-coated or catalyst-free. A catalyst structure comprising a heat resistant support material composed of a plurality of common walls which form a multitude of adjacently disposed longitudinal channels for passage of a gaseous reaction mixture wherein at least a

part of the interior surface of at least a portion of the channels is coated with a catalyst and the interior surface of the remaining channels is not coated with any catalyst such that the interior surface of the catalyst-coated channels are in heat exchange relationship with the interior surface of adjacent catalyst-free channels. The preferred catalyst structure can be readily modified to increase the number and tortuosity of the catalytic channels by inserting additional corrugated sheets having a herringbone corrugation pattern between the two flat sheets with sharp peaked corrugations. If additional corrugated sheets are inserted in the repeat unit, they can be coated on one side of the other or remain uncoated depending on the catalyst structure desired. Catalytic channels and non-catalytic channels are formed by selectively coating one side of the two flat sheets and one side of one of the corrugated sheets with a catalyst. The reactor can incorporate streamwise variation of the fluid velocities. Specifically, the velocity of the fluid in the exothermic catalytic reaction channel can be decreased after the fluid enters the channel by streamwise geometric changes in the reactor wherein the channel's entrance flow area is less than the channel's cross-stream flow area at some streamwise location where the catalyst is deposited. The channel's entrance flow area is defined as the channel's cross-stream flow area immediately downstream of the most downstream conduit inlet. If the conduit wall thicknesses are approximately constant, streamwise changes in flow velocity can be produced by providing, for instance, either contracted sections of the conduits, expanded sections of the casing, or a combination of both. In a reactor employing expanded-end conduits, the cross-stream area of the conduits decreases just after the cooling flow enters the cooling passages, where the expanded conduit sections taper down to the nominal conduit size in the central portion of the reactor. As a result, the cooling flow velocity is increased to a value greater than its entrance velocity. Conversely, because the casing is of constant cross-sectional size, the cross-stream flow area of the exothermic catalytic reaction channel increases just after the fuel-oxidant mixture enters. As a result, the flow velocity in the exothermic catalytic reaction channel is decreased to a value less than its entrance velocity. The gas flow velocity entering the exothermic catalytic reaction channel should exceed the minimum required to prevent flashback into the fuel-oxidant stream upstream of the reactor if the fuel-oxidant mixture entering the exothermic catalytic reaction channel is within the limits of flammability. The laminar flame propagation velocity is typically less than 0.8 meters per second for hydrocarbon fuels in air, but the turbulent flame propagation velocity may exceed 8 meters per second and may approach 28 meters per second for highly turbulent flow. To prevent flashback, the gas flow velocity should exceed 8 to 28 meters per second at gas turbine engine conditions, or more if a safety margin is allowed. Because catalyst light-off becomes increasingly difficult with increasing velocity, it is desirable to reduce the velocity of the fuel-air stream once it has entered the exothermic catalytic reaction channel, by a streamwise variation of cross-stream area. The flow velocity of the fuel-air stream over the exothermic reaction surface in the exothermic catalytic reaction channel is nominally 28 meters per second or less. This reduction in velocity is achieved by a streamwise increase in the cross-stream area for flow over the exothermic reaction surface. Streamwise changes in cross-stream area are fixed by the geometry of the reactor, and do not change in time. The velocity of the cooling stream should exceed the maximum flame propagation velocity at the exit of the cooling conduits, if the cooling fluid exiting the cooling passages is within the limits of flammability, to prevent flashback from a downstream combustion chamber. If expanded conduit outlets are employed, it is also very important that the downstream increase in cross-stream area of the cooling conduits is sufficiently gradual that recirculation of the cooling flow does not occur, so that there is no possibility of flashback or flame holding in the downstream expanded conduit section. Typically, the cone angle for an axis-symmetric diffuser section should not exceed approximately 6 to 8 degrees for good pressure recovery and minimal recirculation. If continued backside cooling is a consideration, the angle should be especially shallow to ensure that there is no local separation of the cooling flow and concurrent loss of cooling effectiveness.

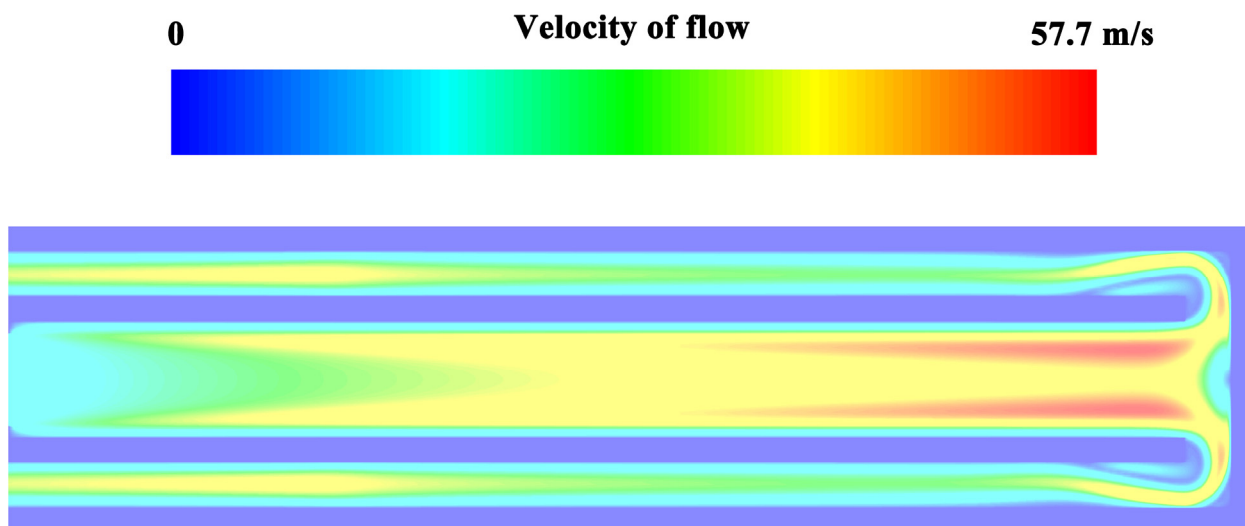


Figure 5. Velocity contour plots of flow in the catalytic reactor with an integral heat exchange structure in an array of longitudinally disposed, adjacent reaction passageways or channels which are either catalyst-coated or catalyst-free.

The gas-phase reaction rate profiles along the fluid centerline of the catalytic reactor are presented in Figure 6 with an integral heat exchange structure in an array of longitudinally disposed, adjacent reaction passageways or channels which are either catalyst-coated or catalyst-free. The method takes advantage of a sequence of fuel combustion zones, and improves the system by conducting at least a portion of the thermal combustion in a gas expansion zone of the catalytic reactor. Part of the thermal combustion may occur in both of these types of expansion areas. As a result, a continuous reheating effect occurs within the catalytic reactor and this system provides for the highly efficient use of the fuel, greater than if no combustion occurs in the expansion zone of the catalytic reactor, without the formation of undesirable amounts of nitrogen oxides during such combustion and the development of excessively high temperatures. Part of the catalytically supported thermal combustion of the fuel-air mixture is conducted upstream of the catalytic reactor by combustion of the mixture while passing through an insufficient amount of catalyst to effect complete combustion of the fuel prior to passing through the expansion zone. The partially combusted effluent from the catalyst, with or without substantial intermediate but incomplete further combustion, is introduced into a gas expansion zone of the catalytic reactor so that the partially combusted effluent is further thermally combusted while undergoing expansion in the catalytic reactor. Catalytically-supported thermal combustion in the catalytic reactor is achieved by contacting at least a portion of the carbonaceous fuel intimately admixed with air with a solid oxidation catalyst having an operating temperature substantially above the instantaneous auto-ignition temperature of the fuel-air admixture. At least a portion of the fuel is combusted in the catalytic reactor under essentially adiabatic conditions. Combustion in the catalytic reactor is characterized by the use of a fuel-air admixture having an adiabatic flame temperature substantially above the instantaneous auto-ignition temperature of the admixture but below a temperature that would result in any substantial formation of oxides of nitrogen. The adiabatic flame temperature is determined at catalyst inlet conditions. The resulting effluent is characterized by high thermal energy useful for generating power and by low amounts of atmospheric pollutants. Where desired, combustible fuel components, for instance, un-combusted fuel or intermediate combustion products contained in the effluent from the catalytic zone, or fuel-air admixture which has not passed through a catalytic zone, may be combusted in a thermal zone following the catalytic zone. Sustained catalytically-supported, thermal combustion occurs at a substantially lower temperature than in conventional adiabatic thermal combustion and therefore it is possible to operate without formation of

significant amounts of oxides of nitrogen. Combustion in the catalytic reactor is no longer limited by mass transfer as in the case of conventional catalytic combustion, and at the specified operating temperatures the reaction rate is substantially increased beyond the mass transfer limitation, for instance, at least about 8 times greater than the mass transfer limited rate. In the catalytic reactor, reaction rates of up to about 80 or more times the mass transfer limited rate may be attainable. Such high reaction rates permit high fuel space velocities which normally are not obtainable in catalytic reactions. One can employ, for instance, at least an amount of fuel equivalent in heating value to about 200 pounds of ethane per hour per cubic foot of catalyst, and this amount may be at least several times greater, for instance, an amount of fuel equivalent in heating value to at least about 800 pounds of ethane per hour per cubic foot of catalyst. There is, moreover, no necessity of maintaining fuel-to-air ratios in the flammable range, and consequently loss of combustion due to variations in the fuel-to-air ratio is not the problem it is in conventional combustors [71, 72]. When the fuel-air ratio in the cooling channels is within the limits of flammability, it is preferred that the cross-stream area for flow through the cooling channels is not increased after the cooling stream enters the conduits [73, 74]. The cross-stream area of the cooling passages may in fact be decreased to increase the cooling flow velocity for greater resistance to flashback or pre-ignition in the cooling portion of the reactor. The minimum residence time for pre-ignition to occur in the conduits is dependent upon fuel type, fuel-air ratio, temperature, and pressure, and can be measured experimentally or calculated on the basis of elementary chemical reaction rates, if known. Gas temperatures in the cooling portion of the reactor may rise to near the material limit of the catalyst and substrate, resulting in very short ignition delay times. To prevent pre-ignition in the cooling portion of the reactor, the gas residence time in the cooling portion of the reactor should be less than the ignition delay time. Increasing the cooling flow velocity within the cooling passages reduces the residence time of the cooling fluid within the reactor, and reduces the cooling fluid's propensity for pre-ignition. Similarly, decreasing the velocity of the fluid in the exothermic catalytic reaction channel provides increased residence time for reaction in a given length reactor, while the cooling fluid's residence time within the same length reactor remains at a smaller value, allowing a reduced propensity for pre-ignition of the cooling fluid. When applied to the catalysis of highly exothermic reactions, the catalyst structures are typically monolithic-type structures comprising a heat resistant support material composed of a plurality of common walls which form a multitude of adjacently disposed longitudinal channels for passage of a gaseous reaction mixture wherein at least a portion of the channels are coated on at least a part of their interior surface with a catalyst for the reaction mixture, catalyst-coated channels, and the remaining channels are not coated with catalyst on their interior surface, catalyst-free channels, such that the interior surface of the catalyst-coated channels are in heat exchange relationship with the interior surface of adjacent catalyst-free channels and wherein the catalyst-coated channels differ in configuration from the catalyst-free channels such that the desired reaction is promoted in the catalytic channels and suppressed in the non-catalytic channels of the catalytic reactor [75, 76]. In cases where the catalyst structure is employed in a catalytic combustion or partial combustion process, the critical difference in the design of the catalytic versus non-catalytic channels will insure more complete combustion of the fuel in the catalytic channels and minimum combustion in the non-catalytic channels over a wider range of linear velocity, inlet gas temperature and pressure [77, 78]. The highly exothermic process can be carried out in a single catalytic reaction zone employing the catalyst structure or in multiple catalytic reaction zones using catalyst structures designed specifically for each catalytic stage. In most cases the catalytic reaction zone will be followed by a homogeneous combustion zone in which the gas exiting from the earlier catalytic combustion zone is combusted under non-catalytic, non-flame conditions to afford the higher gas temperature. The homogeneous combustion zone is sized to achieve substantially complete combustion and to reduce the carbon monoxide level to the desired concentration.

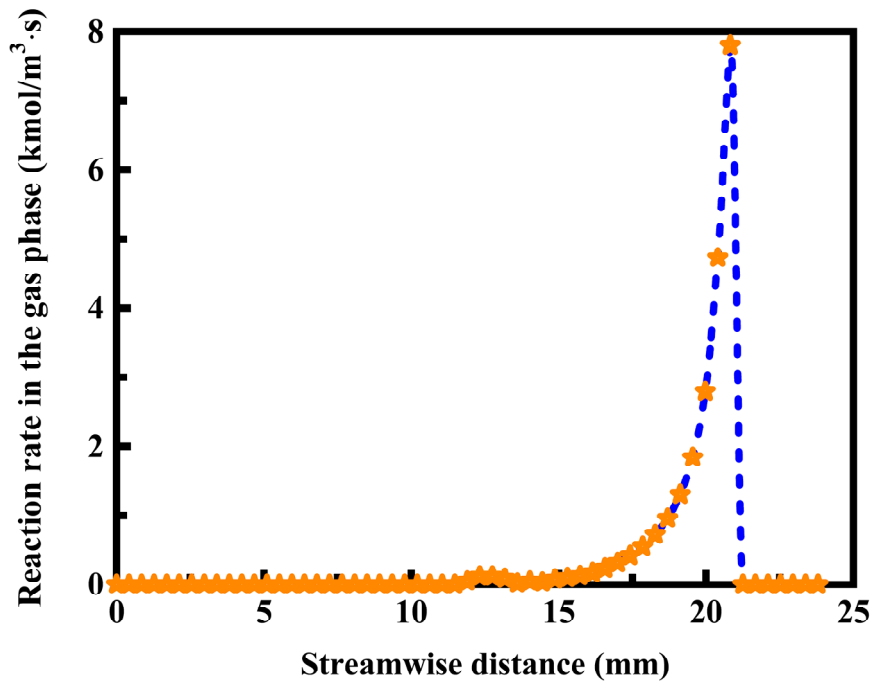


Figure 6. Gas-phase reaction rate profiles along the fluid centerline of the catalytic reactor with an integral heat exchange structure in an array of longitudinally disposed, adjacent reaction passageways or channels which are either catalyst-coated or catalyst-free.

The reaction rate contour plots in the gas phase of the catalytic reactor are illustrated in Figure 7 with an integral heat exchange structure in an array of longitudinally disposed, adjacent reaction passageways or channels which are either catalyst-coated or catalyst-free. In preferred catalyst structures, the channels coated with catalyst differ from the catalyst-free channels by having an average hydraulic diameter which is lower than the average hydraulic diameter of the catalyst-free channels and by having a higher film heat transfer coefficient than the catalyst-free channels. More preferably, the catalyst-coated channels have both a lower hydraulic diameter and a higher film heat transfer coefficient than the catalyst-free channels. Thus, for the catalyst structures, the average hydraulic diameter can be determined by first finding the hydraulic diameter for all of the catalyst-coated channels in the structure by calculating the average hydraulic diameter for any given channel over its entire length and then determining the average hydraulic diameter for the catalyst-coated channels by totaling up all of the calculated hydraulic diameters for the individual channels, multiplied by a weighing factor representing the fractional open frontal area for that channel. Following the same procedure, the average hydraulic diameter for the catalyst-free channels in the structure can also be determined. The finding that the catalyst-coated channels most advantageously have a lower average hydraulic diameter than the catalyst-free channels can be explained, in part, by the fact that the catalyst-coated channels desirably have a surface to volume ratio which is higher than that of the catalyst-free channels, since hydraulic diameter bears an inverse relationship to surface to volume ratio. Further, in the catalyst structures, the difference in average hydraulic diameter of the catalyst-coated channels and catalyst-free channels gives an indication that the catalyst-free channels, on average, must be more open channeled and therefore, the gas flow through these channels is less affected by changes in the channel diameter than the catalyst-coated channels, again, in part, because of the higher surface to volume ratios in the catalyst-coated channels. Preferably, the numeric ratio of the average hydraulic diameter of the catalyst-coated channels to the average hydraulic diameter of the catalyst-free channels, that is, average hydraulic diameter of catalyst-coated channels divided by average hydraulic diameter of catalyst-free channels is between about 0.08 and about 0.88 and, most preferably, the ratio of average hydraulic diameter of catalyst-coated channels to catalyst-free channels is between about 0.2

and 0.8. The film heat transfer coefficient is a dimension-less value, which is measured experimentally by flowing gas, for instance, air or air-fuel mixtures, at a given inlet temperature through an appropriate test structure having the specified channel geometry and temperature and measuring the outlet gas temperature. Since the gas composition, flow rates, pressures and temperatures in the catalytic and non-catalytic channels of the catalyst structure are very similar, the film heat transfer coefficient provides useful means of characterizing the different flow geometries provided by the various flow channel configurations which distinguish the catalyst-coated channels from the catalyst-free channels of the catalyst structure. Since these different flow geometries, in turn, are related to the tortuosity of the flow path formed by the channels, the film heat transfer coefficient provides some measure of tortuosity as it is employed in the catalyst structures. While one could conceive of a variety of methods to measure or otherwise determine the film heat transfer coefficient in the catalyst structures, one convenient method would involve constructing an experimental test structure, for instance, a solid thick metal structure, with internal space machined to simulate the desired channel shape; and then to test it in environments where the wall temperature is essentially constant from inlet to outlet or varies from inlet to outlet and is measured at several points along the channel length in the structure. The catalyst structure is particularly useful when equipped with appropriate catalytic materials for use in a combustion or partial combustion process wherein a fuel, in gaseous or vaporous form, is typically partially combusted in the catalyst structure followed by complete homogeneous combustion downstream of the catalyst. With the catalyst structure, it is possible to obtain more complete combustion of fuel in the catalytic channels with minimum combustion in the non-catalytic channels over a wider range of linear velocities, gas inlet temperatures and pressures than has here-to-for been possible with catalyst structures of the previous designs, including those employing integral heat exchange. Accordingly, the design also encompasses an improved catalyst structure for use in the combustion or partial combustion of a combustible fuel, as well as a process for combusting a mixture of a combustible fuel and air or oxygen-containing gas, using the catalyst structure.

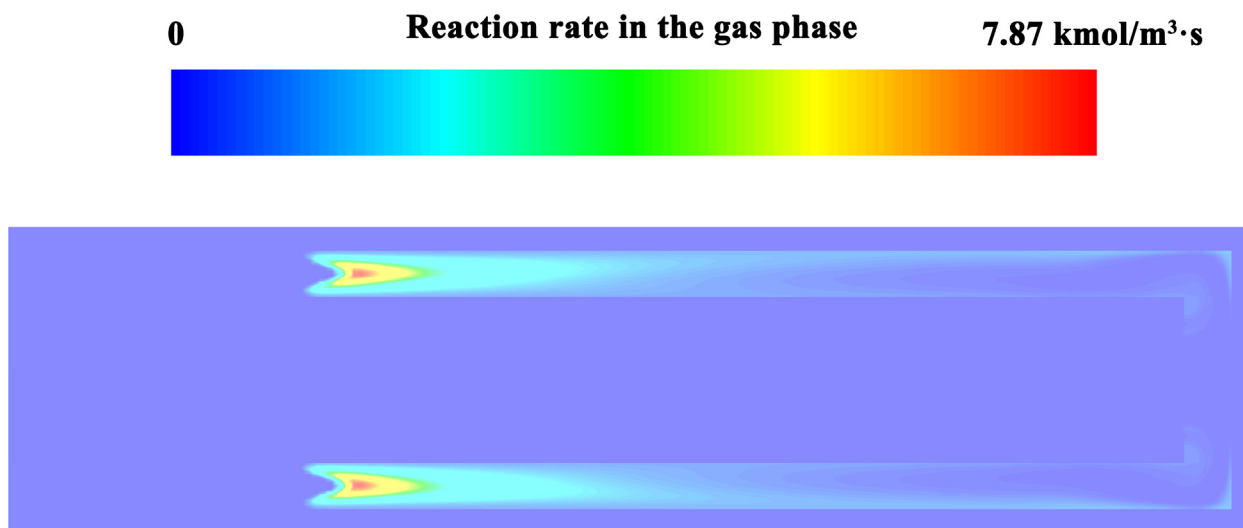


Figure 7. Reaction rate contour plots in the gas phase of the catalytic reactor with an integral heat exchange structure in an array of longitudinally disposed, adjacent reaction passageways or channels which are either catalyst-coated or catalyst-free.

The temperature contour plots in the catalytic reactor are illustrated in Figure 8 with an integral heat exchange structure in an array of longitudinally disposed, adjacent reaction passageways or channels which are either catalyst-coated or catalyst-free. The catalyst in the catalytically supported thermal combustion generally operates at a temperature approximating the theoretical adiabatic flame temperature of the fuel-air admixture charged to the combustion zone. The entire catalyst may not be at

these temperatures, but preferably a major portion, or essentially all, of the catalyst surface is at such operating temperatures. These temperatures are usually in the range of about 1200 K to 2000 K. The temperature of the catalyst zone is controlled by controlling the composition and initial temperature of the fuel-air admixture, namely adiabatic flame temperature, as well as the uniformity of the mixture. Relatively higher energy fuels can be admixed with larger amounts of air in order to maintain the desired temperature in a combustion zone. At the higher end of the temperature range, shorter residence times of the gas in the combustion zone appear to be desirable in order to lessen the chance of forming oxides of nitrogen. The residence time is governed largely by space throughput, pressure, and temperature, and generally is measured in milliseconds. The residence time of the gases in the catalytic combustion zone and any subsequent thermal combustion zone may be below about 0.08 second, preferably below about 0.02 second. The gas space velocity under standard temperature and pressure conditions may often be, for instance, in the range of about 0.8 to 8 or more million cubic feet of total gas per cubic foot of total combustion zone per hour. The total residence time in the combustion system should be sufficient to provide essentially complete combustion of the fuel, but not so long as to result in the formation of oxides of nitrogen. In a preferred aspect, the catalyst structure can be further characterized by catalyst-coated channels that differ from the catalyst-free channels in one or more critical structural defining elements which, in turn, take advantage of, and expand upon, the concept of the increased tortuosity of the catalyst-coated channels. In particular, the preferred catalyst structure typically employs a plurality of longitudinally disposed channels coated on at least a portion of their interior surface with catalyst, that is, catalyst-coated channels, in heat exchange relationship with adjacent channels not coated with catalyst or catalyst-free channels wherein the catalyst-coated channels have an average hydraulic diameter which is lower than the average hydraulic diameter of the catalyst-free channels and the catalyst-coated channels have a higher film heat transfer coefficient than the catalyst-free channels [79, 80]. The average hydraulic diameter, which is defined as four times the average cross-sectional area of all of the channels of a particular type, for instance, catalyst-coated channels, in the catalyst structure divided by the average wetted perimeter of all of the channels of that type in the catalyst structure, is reflective of the finding that the catalyst-free channels are most advantageously designed to have a larger hydraulic diameter and to be less affected by changes in configuration than the catalyst-coated channels [81, 82]. The film heat transfer coefficient is an experimentally determined value which correlates with, and expands upon the tortuosity of the average catalyst-coated channel versus that of the average catalyst-free channel in the catalyst structure [83, 84]. Further optimization of the catalyst structure is obtained if, in addition to controlling the average hydraulic diameter and the film heat transfer coefficient as set forth above, the heat transfer surface area between the catalyst-coated channels and the catalyst-free channels is controlled such that the heat transfer surface area between the catalyst-coated channels and catalyst-free channels divided by the total channel volume in the catalyst structure is greater than about 0.8 per millimeter [85, 86]. A catalytic reactor thus provided with such passive substrate cooling will be able to operate with a richer mixture of fuel and air and at lower velocities without overheating and damaging the catalyst or catalyst substrate [87, 88]. This, in effect, serves to raise the maximum temperature of the catalyst. Another advantage of the arrangement is that the reacting passages provide stable, high temperature, continuous, and uniform ignition sources for the balance of the unreacted mixture which then burns at the desired high temperature just downstream of the catalytic reactor. In effect, the unit is a hybrid of a catalytic reactor and a flame holder. Any hot surface acts as a catalyst to some degree, hence even the non-catalyzed passages may tend to provide some surface combustion. This effect will be minimized by selecting a ceramic base material with minimal catalytic properties. It may also be possible to control the boundary layer, decrease the surface area, decrease the residence time, and perhaps even provide a chain breaking or ignition delaying surface. The catalytic elements can be engineered to

provide the reactivity across the unit best tailored to the fuel preparation zone characteristics, or to the requirements of the catalytic reactor inlet pattern factor.

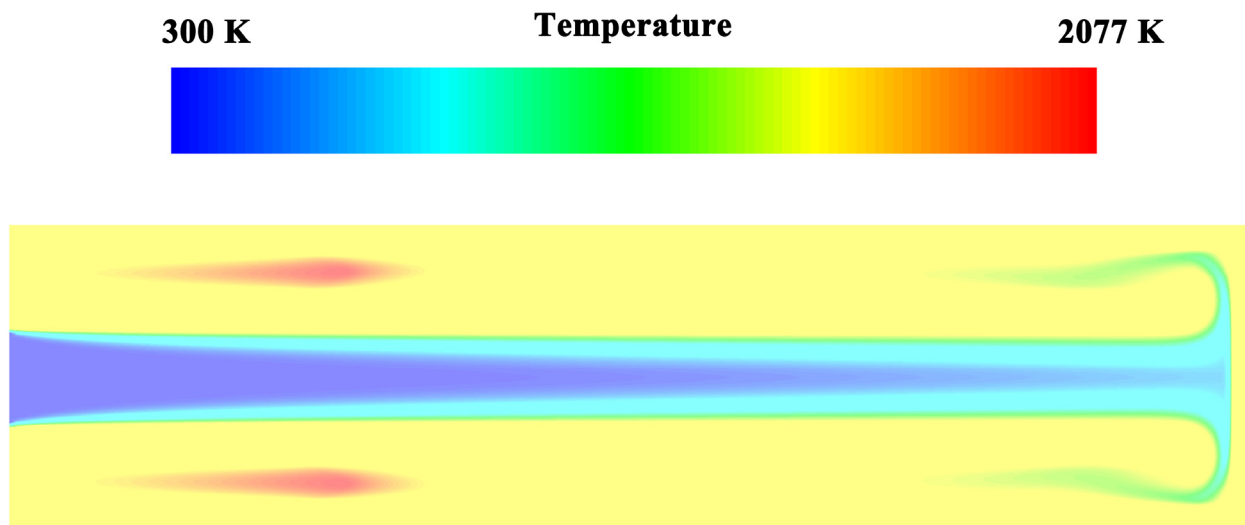


Figure 8. Temperature contour plots in the catalytic reactor with an integral heat exchange structure in an array of longitudinally disposed, adjacent reaction passageways or channels which are either catalyst-coated or catalyst-free.

4. Conclusions

The highly exothermic process characteristics of catalytic reactors are investigated with integral heat exchange structures. Ethane mole fraction and gas-phase reaction rate profiles in catalytic reactors are presented for a catalytic reactor with an integral heat exchange structure, and ethane mole fraction, flow velocity, gas-phase reaction rate, and temperature contour plots are illustrated for catalytically supported thermal combustion systems. The present study aims to provide an improved reaction system and process for combustion of a fuel wherein catalytic combustion using a catalyst structure employing integral heat exchange affords a partially-combusted, gaseous product which is passed to a homogeneous combustion zone where complete combustion is promoted by means of a flame holder. Particular emphasis is placed upon the catalytic reactor configuration that allows the oxidation catalyst to be backside cooled by any fluid passing through the cooling conduits. The major conclusions are summarized as follows:

- The percentage of reaction completed in the exothermic catalytic reaction channel depends both upon the flow rate of the fuel-oxidant mixture through the exothermic catalytic reaction channel and upon the physical characteristics of the catalytic reactor. The critical difference in the design of the catalytic versus non-catalytic channels for the catalytic structure, in its most basic terms, is that the catalytic channels are designed so that the reaction mixture flow passages defined by the catalytic channels possess a higher or increased tortuosity over the corresponding flow passages formed by the non-catalytic channels.
- The tortuosity of the catalytic channels is increased by changing their cross-sectional area at a multiplicity of points along their longitudinal axes. One preferred way of accomplishing this change in tortuosity for the catalytic channels involves the use of a stacked arrangement of non-nesting corrugated sheets of catalyst support material which are corrugated in a herringbone pattern with at least a portion of one side of a given corrugated sheet facing and stacked against another corrugated sheet being coated with catalyst such that the stacked sheets in question form a plurality of catalytic channels.
- The gas flow velocity entering the exothermic catalytic reaction channel should exceed the

minimum required to prevent flashback into the fuel-oxidant stream upstream of the reactor if the fuel-oxidant mixture entering the exothermic catalytic reaction channel is within the limits of flammability. The velocity of the cooling stream should exceed the maximum flame propagation velocity at the exit of the cooling conduits, if the cooling fluid exiting the cooling passages is within the limits of flammability, to prevent flashback from a downstream combustion chamber.

- Catalytically-supported thermal combustion in the catalytic reactor is achieved by contacting at least a portion of the carbonaceous fuel intimately admixed with air with a solid oxidation catalyst having an operating temperature substantially above the instantaneous auto-ignition temperature of the fuel-air admixture. Combustion in the catalytic reactor is characterized by the use of a fuel-air admixture having an adiabatic flame temperature substantially above the instantaneous auto-ignition temperature of the admixture but below a temperature that would result in any substantial formation of oxides of nitrogen.
- The film heat transfer coefficient provides useful means of characterizing the different flow geometries provided by the various flow channel configurations which distinguish the catalyst-coated channels from the catalyst-free channels of the catalyst structure. Since these different flow geometries, in turn, are related to the tortuosity of the flow path formed by the channels, the film heat transfer coefficient provides some measure of tortuosity as it is employed in the catalyst structures
- The temperature of the catalyst zone is controlled by controlling the composition and initial temperature of the fuel-air admixture as well as the uniformity of the mixture. The total residence time in the combustion system should be sufficient to provide essentially complete combustion of the fuel, but not so long as to result in the formation of oxides of nitrogen. Any hot surface acts as a catalyst to some degree, hence even the non-catalyzed passages may tend to provide some surface combustion. This effect will be minimized by selecting a ceramic base material with minimal catalytic properties. It may also be possible to control the boundary layer, decrease the surface area and the residence time, and perhaps even provide a chain breaking or ignition delaying surface.

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