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(54) **METHOD FOR LOW NOX COMBUSTION OF SYNGAS / HIGH HYDROGEN FUELS**

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(57) **ABSTRACT**

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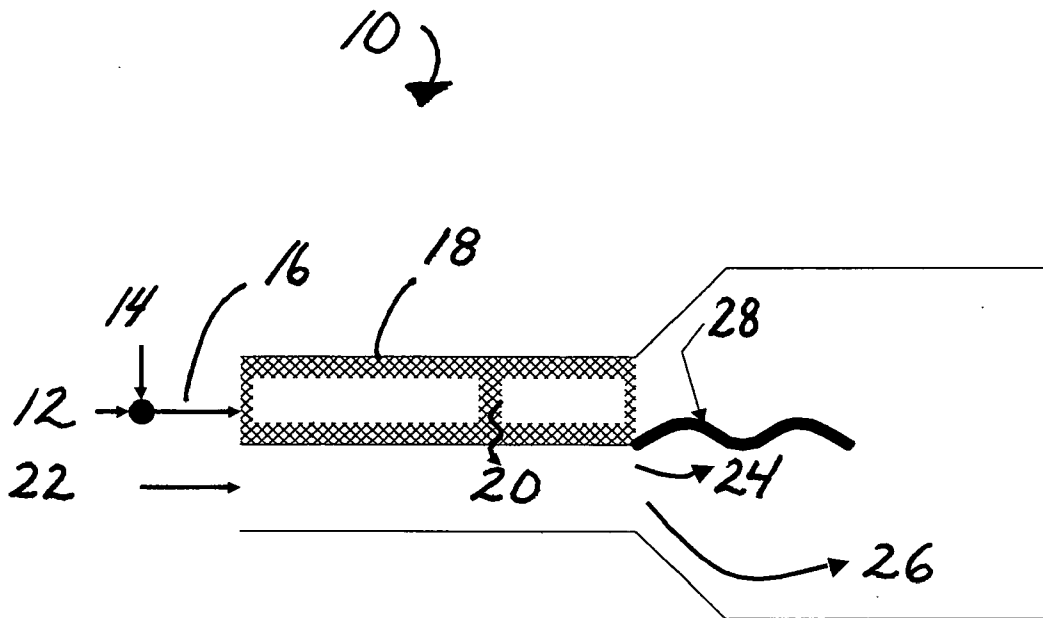
A method is provided for achieving low NOx in the operation of a non-premixed combustion system by reacting a fuel-rich mixture to produce partial reaction products plus heat, transferring a portion of the heat to a bypass air stream, and passing the cooled partial reaction products into non-premixed contact and combustion with a stoichiometric portion of the heated bypass air stream. A supply of fuel and a supply of air is provided; and a fuel-rich mixture is formed and reacted to produce partial reaction products plus a heat of reaction. A portion of the heat of reaction is transferred to a bypass air stream and the cooled partial reaction products are passed into non-premixed contact and combustion with a stoichiometric portion of the heated bypass air stream.

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Related U.S. Application Data

(63) Continuation-in-part of application No. 11/439,727, filed on May 23, 2006, now abandoned.



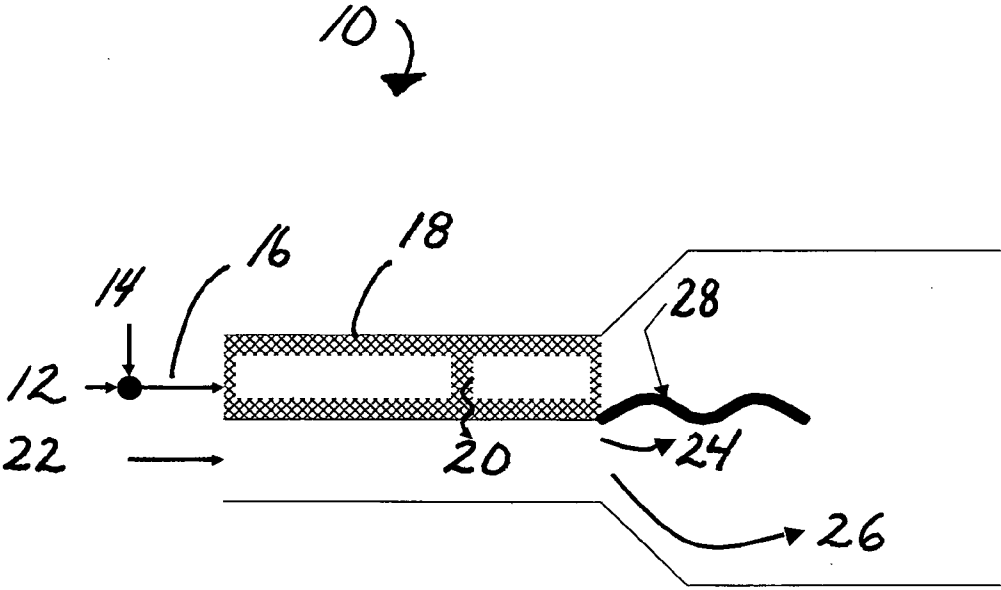


FIG. 1

Stoichiometric flame temperature prediction of methane for 100% oxygen conversion in reactor and 60% heat transfer to the cooling air.

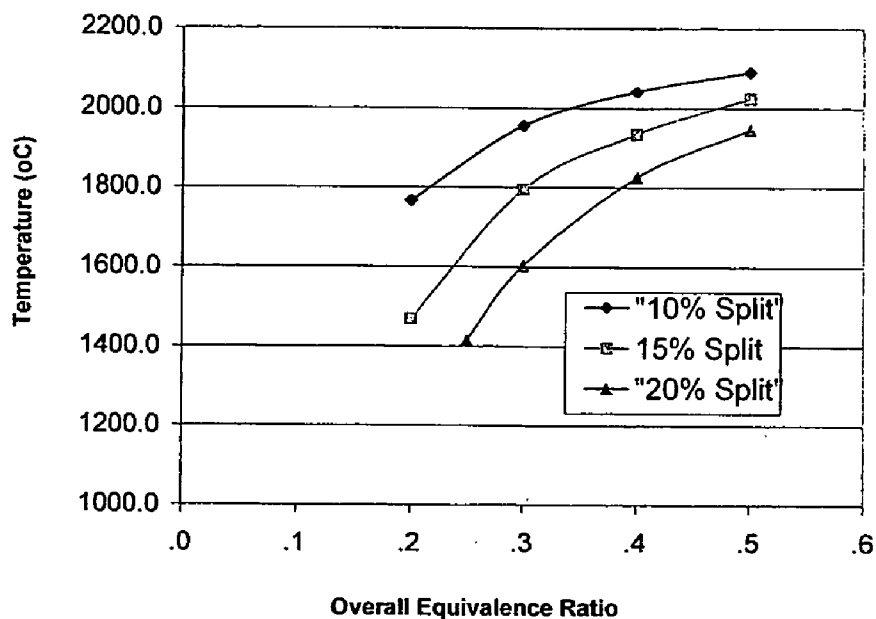


FIG. 2

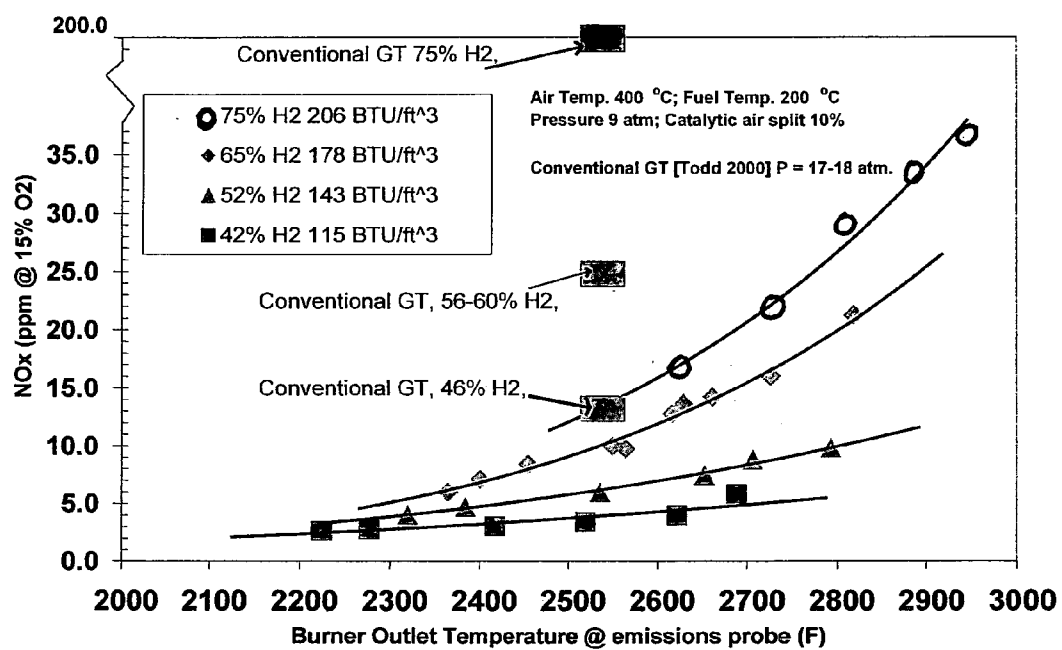


FIG. 3

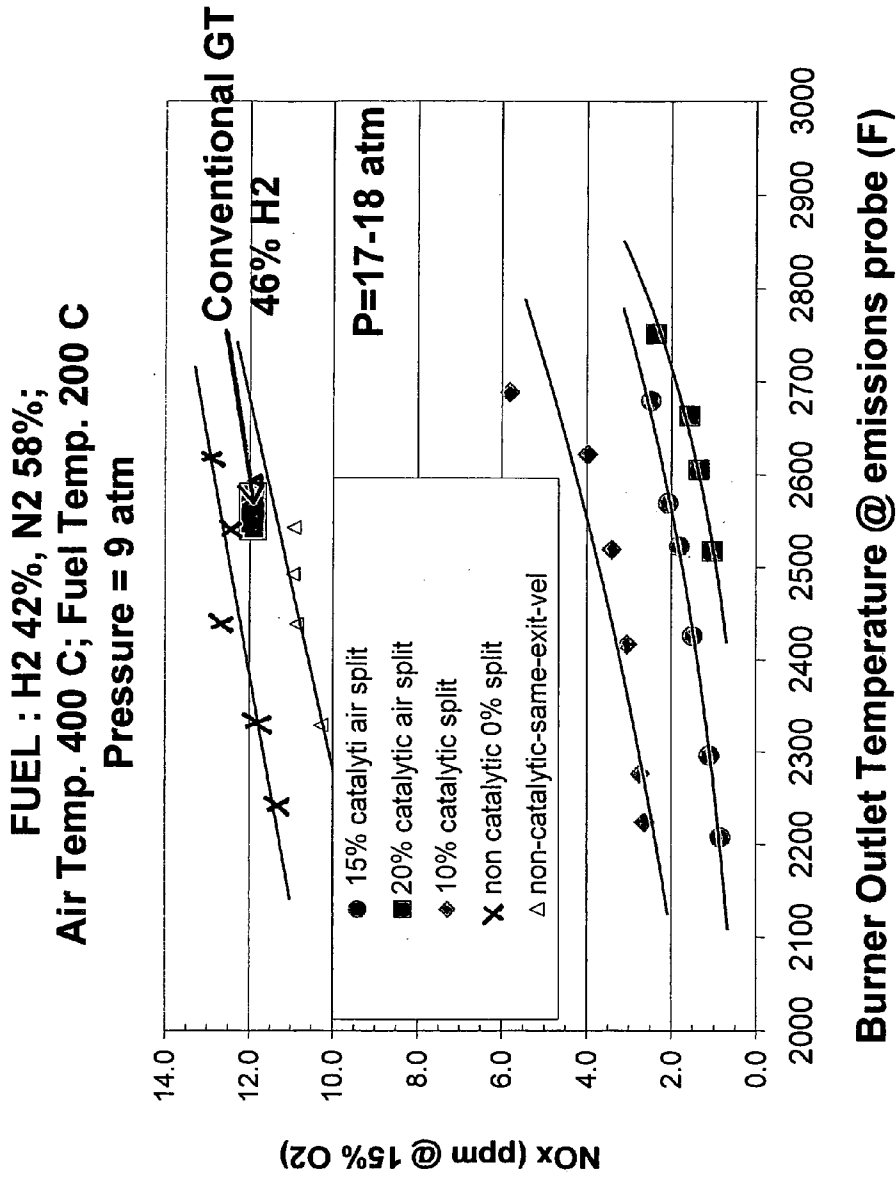


FIG 4

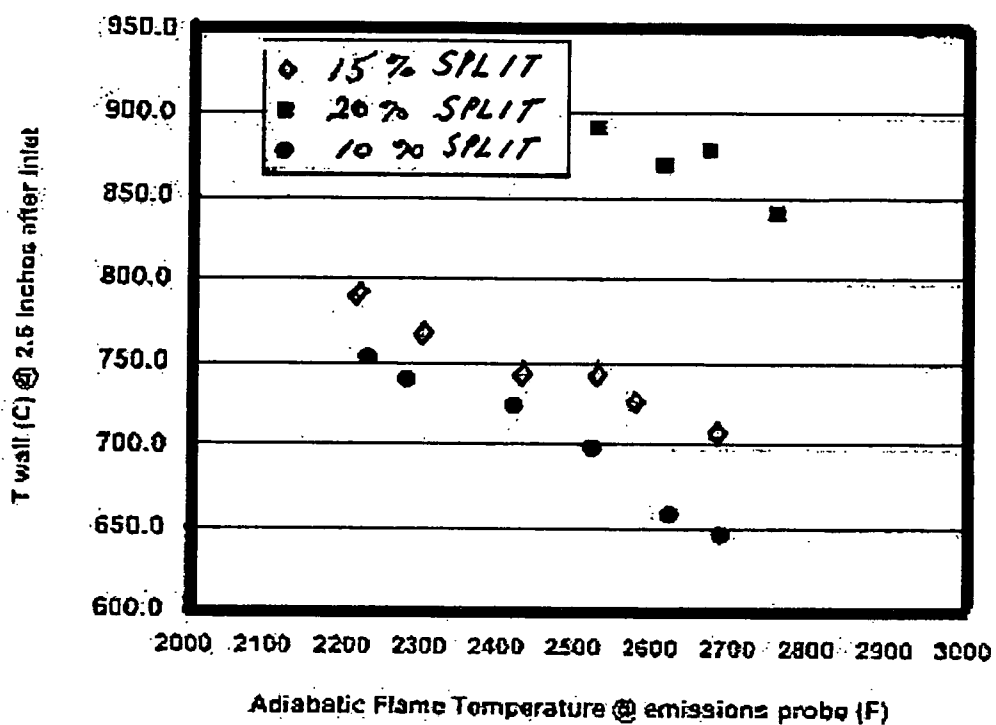


FIG. 5

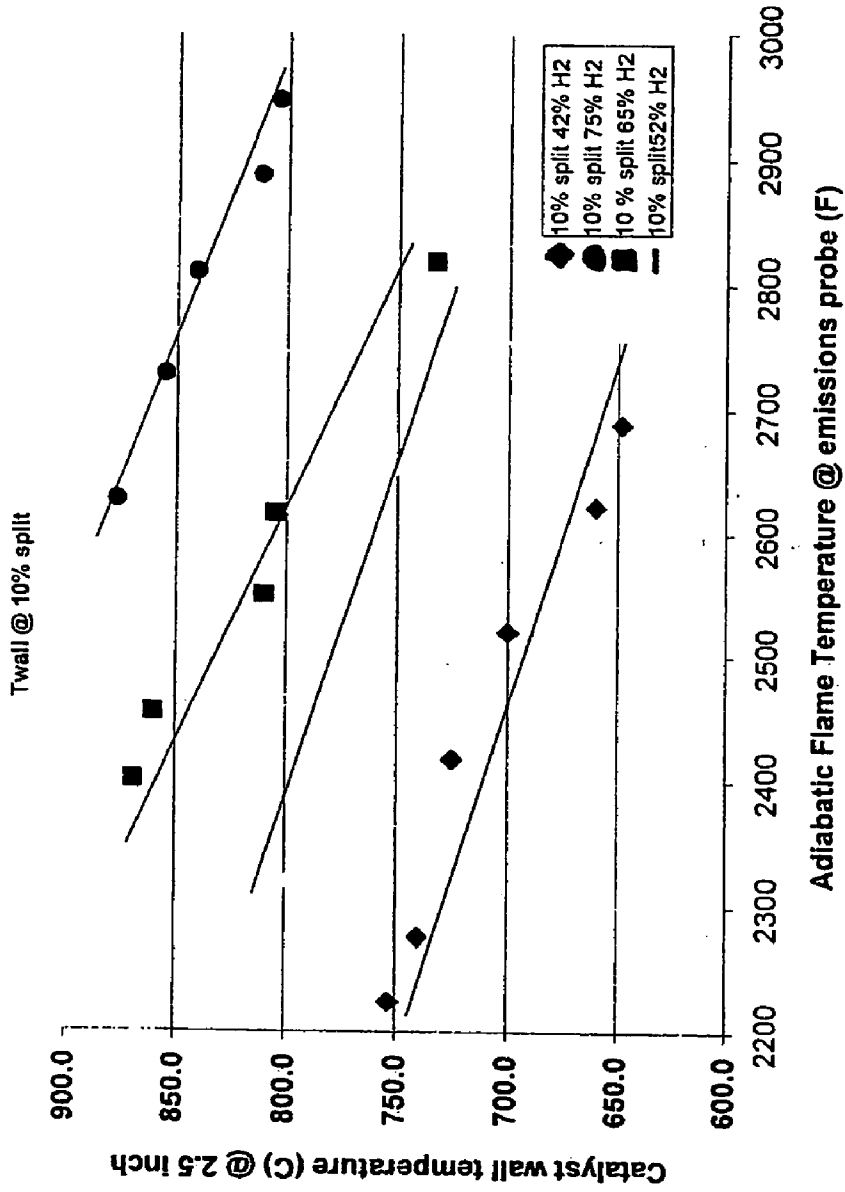


FIG. 6

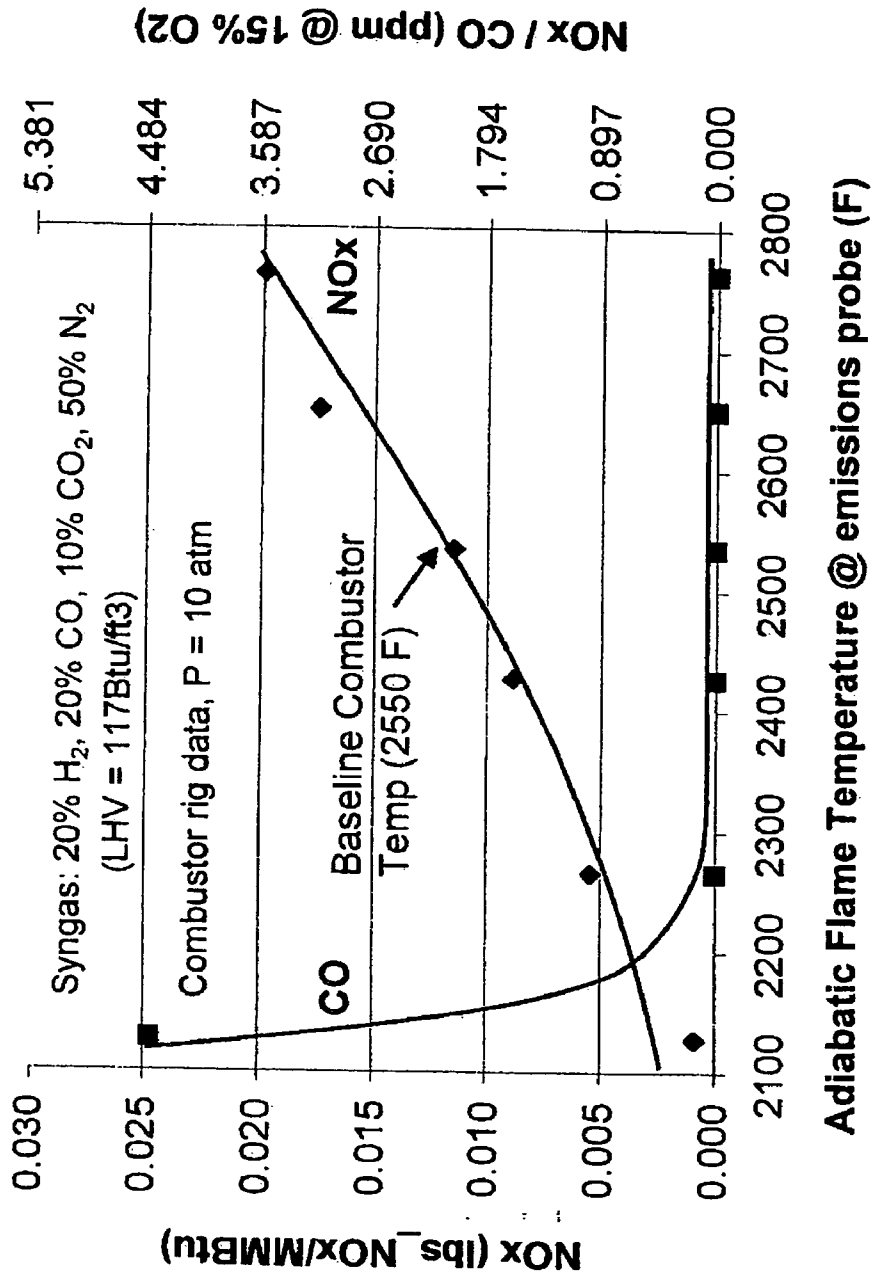


FIG. 7

METHOD FOR LOW NOX COMBUSTION OF SYNGAS / HIGH HYDROGEN FUELS

CROSS-REFERENCE

[0001] This application is a Continuation-In-Part of U.S. patent application Ser. No. 11/439,727 filed on May 23, 2006; which application in turn claims the benefit of U.S. Provisional Application No. 60/683,719 filed May 23, 2005.

GOVERNMENT RIGHTS

[0002] This invention was made with government support under U.S. Contract No. DE-FC26-05NT42647. The U.S. government holds certain rights in this invention.

FIELD OF THE INVENTION

[0003] The present invention relates to a method for ultra-low NOx combustion of high-hydrogen content fuels. In particular, the present invention comprises removing heat from the partial reaction products produced by reacting a fuel-rich mixture and transferring a portion of that heat into a cooling air stream. By removing heat from the partial reaction products, the stoichiometric flame temperature is reduced. More particularly, the present invention provides a method for lowering the stoichiometric flame temperature of cooled partial reaction products and passing such cooled partial reaction products into non-premixed contact and combustion with a stoichiometric portion of heated the air stream that was heated by the transfer of heat from the partial reaction of the fuel-rich mixture.

BACKGROUND OF THE INVENTION

[0004] With energy usage directly related to economic growth, there has been a steady increase in the need for increased energy supplies. In the U.S., coal is abundant and comparatively low in cost. Unfortunately, conventional coal-fired steam plants, which are a major source of electrical power, are inefficient and pollute the air. Thus, there is a pressing need for cleaner, more efficient coal-fired power plants. Accordingly, Integrated Gasification Combined Cycle ("IGCC") coal technology systems have been developed which can achieve significantly improved efficiencies in comparison to conventional steam plants. In such a system, syngas (a mixture of hydrogen and carbon monoxide) is produced by partial oxidation of coal or other carbonaceous fuel. This allows cleanup of sulfur and other impurities, including mercury, before combustion.

[0005] Concern over global warming resulting from carbon dioxide emissions from human activity, primarily the combustion of fossil fuels, has led to the need to sequester carbon. If carbon sequestration is desired, the carbon monoxide can be reacted with steam using the water gas shift reaction to form carbon dioxide and hydrogen. Carbon dioxide may then be recovered using conventional technologies known in the art. This allows pre-combustion recovery of carbon dioxide for sequestration.

[0006] As a result of the high flame speed of hydrogen, flashback is an issue with premixed dry low NOx combustion systems. Flashback remains an issue with the use of syngas as well. Regardless of whether carbon dioxide is recovered or whether air or oxygen are used for syngas production, hydrogen content of the gas typically is too high to allow use of conventional dry low NOx premixed combustion for NOx control. Therefore, diffusion flame combustion is used typi-

cally with steam or nitrogen added as a diluent to the syngas from oxygen blown gasifiers to minimize NOx. Even so, exhaust gas cleanup still may be required. Thus, such systems, though cleaner and more efficient, typically cannot achieve present standards for NOx emissions without removal of NOx.

[0007] A further problem is that the presence of diluent in the fuel increases mass flow through a turbine often requiring the bleeding off of compressor discharge air. Since bleed off of compressor air must be limited to allow sufficient air for combustion and turbine cooling, the amount of diluent which can be added to the fuel is limited. Typically, NOx cannot be reduced below about ten parts per million ("ppm") without operational problems, including limited flame stability.

[0008] There are further efficiency loss issues. If nitrogen is added to dilute the fuel gas, there is an energy penalty related to the need to compress the nitrogen to the pressure required for mixing with the fuel gas. In addition, use of syngas in a gas turbine designed for natural gas increases turbine mass flow even without syngas dilution. Typically, to avoid excessive loads on the turbine rotor, operation is at a reduced turbine inlet temperature and/or with bleed of compressed air from the turbine compressor.

[0009] In one example of the prior art wherein a combustion process is disclosed for the reduction of NOx, U.S. Pat. No. 3,826,078 to Quigg (hereinafter referred to as "Quigg"), the prior art discloses a combustion process comprising raising the temperature of a secondary air stream without removing heat from the rich burn primary zone. Quigg further discloses that "it is preferred that the transition from the fuel-rich condition in the primary combustion zone to the fuel-lean condition in the secondary zone be sharp or rapid, e.g., be effected as quickly as possible." (See Quigg, Column 14, lines 63-67). This system is well known in the art and is commonly referred to as "Rich Burn/Quick Quench."

[0010] However, raising the temperature of a secondary air stream without removing heat from the rich burn primary zone, as taught by Quigg, raises the stoichiometric flame temperature, and in turn, elevates the NOx emissions of a combustion system. In particular, Quigg discloses a method comprising partial oxidation in an upfront stage of a combustor; followed by a quick quench, or dilution, with secondary air followed by adding heated air before passing the combustion products and all of the heat of reaction to a turbine. Quigg attempts to control the temperature of combustion products to achieve low NOx, and compatibility with materials used in gas turbines, with this Rich Burn/Quick Quench method. However, the process is not easily controlled and treads a fine balance between extinguishing the flame completely and not reducing the temperature sufficiently, the latter problem leading to higher NOx and damage to a turbine.

[0011] Accordingly, it is an object of this invention to provide an improved combustion system for achieving low NOx combustion. It is another object of the present invention to provide a method of achieving low NOx in the operation of a non-premixed combustion system. It is yet another object of the present invention to provide a method for achieving low stoichiometric flame front temperature and, in turn, reduced combustion temperature and reduced NOx emissions.

SUMMARY OF THE INVENTION

[0012] In sum, a method of achieving low NOx in the operation of a non-premixed combustion system according to the present invention comprises reacting a fuel-rich mixture

to produce partial reaction products plus heat, transferring a portion of the heat to a bypass air stream, and passing the cooled partial reaction products into non-premixed contact and combustion with a stoichiometric portion of the heated bypass air stream.

[0013] In a preferred embodiment of the present invention, a supply of fuel is provided, preferably hydrogen gas wherein such hydrogen gas comprises elemental hydrogen. A supply of air is provided and split into a first percentage of air and a second percentage of air. A fuel-rich mixture of the fuel and the first percentage of air is formed. The fuel-rich mixture is reacted to produce partial reaction products comprising a portion of reacted fuel and a portion of unreacted fuel plus a heat of reaction. A portion of the heat of reaction is concurrently transferred to the second percentage of air thereby producing a heated second percentage of air and cooled partial reaction products. The cooled partial reaction products are passed into non-premixed contact and combustion with a stoichiometric portion of the heated second percentage of air.

[0014] In one preferred embodiment of the present invention, the non-premixed contact and combustion of the cooled partial reaction products with a stoichiometric portion of the heated second percentage of air defines a reduced flame front temperature. Preferably, the reduced flame front temperature is at least 200 degrees Celsius lower than that of the portion of unreacted fuel.

[0015] The supply of fuel comprises hydrogen and further may comprise syngas. The fuel may comprise from about 20 percent hydrogen to about 75 percent hydrogen, or even up to 100 percent hydrogen. The amount of air in the fuel-rich mixture represents at least about twenty percent of the total supply of air. The fuel-rich mixture may be reacted in a backside-cooled catalytic reactor; and the second percentage of air may provide the backside cooling.

BRIEF DESCRIPTION OF THE DRAWINGS

[0016] FIG. 1 provides a diagrammatic representation of the combustion of a fuel in accordance with the present invention.

[0017] FIG. 2 provides a graphical representation of the overall equivalence ratio versus temperature during the combustion of methane.

[0018] FIGS. 3-4 provide a graphical representation of the burner outlet temperature versus NOx in ppm.

[0019] FIGS. 5-6 provide a graphical representation of the adiabatic flame temperature versus the temperature at the wall of the reactor at various locations.

[0020] FIG. 7 provides a graphical representation of tests results obtained from the operation of a device according to the present invention.

DETAILED DESCRIPTION OF THE INVENTION

[0021] The present invention comprises a method of achieving low NOx in the operation of a non-premixed combustion system comprising obtaining a supply of hydrogen gas and a supply of air. The supply of air is split into a first percentage of air and a second percentage of air, and a fuel-rich mixture of the fuel and the first percentage of air is formed. The fuel-rich mixture is reacted to produce partial reaction products consisting essentially of reacted fuel and remaining unreacted fuel plus a heat of reaction, and a portion of the heat of reaction is transferred to the second percentage of air thereby producing a heated second percentage of air and

cooled partial reaction products. The cooled partial reaction products are combusted on contact with a stoichiometric portion of the heated second percentage of air in a non-premixed flame such that a second portion of the heated second percentage of air bypasses the non-premixed flame thereby reducing heat liberated in the non-premixed flame.

[0022] One embodiment of the present invention 10 is shown in FIG. 1. As shown for the embodiment in FIG. 1, a first percentage of air 12 comprising a twenty percent split of the combustion air is mixed with the fuel 14 to form a fuel rich mixture 16 having an equivalence ratio of two, where a ratio of one is stoichiometric. Thus, a portion of a second percentage of air 22 comprising the balance of the combustion air, where such portion is approximately equal to a corresponding twenty percent of the combustion air will be required to complete combustion of the fuel-rich mixture. Complete conversion of the oxygen is assumed in a catalytic reactor 18, preferably a backside cooled catalytic reactor thereby producing partial reaction products comprising reacted fuel and remaining unreacted fuel. Sixty percent of the heat of reaction 20 is transferred to a second percentage of air wherein such air provides the backside cooling for catalytic reactor 18. On contact of the reacted fuel with a stoichiometric portion of the remaining combustion air, only a second twenty percent of air 24 is required. The balance of the combustion air 22 bypasses the flame front. Thus, forty five percent 26 of the reaction heat bypasses the non-premixed flame 28 thereby reducing the heat liberated in the flame by about twenty percent.

[0023] For conventional hydrocarbon fuels, including methane, the reduction in the heat liberated in the flame is not near enough for low NOx production in modern gas turbines. As shown in FIG. 2, even with a twenty percent air split, the adiabatic flame temperature of methane is not reduced below 1600 celsius at equivalence ratios greater than 0.3. Modern industrial and utility gas turbines require primary combustion zone equivalence ratios of greater than 0.4. For such fuels, the ultra-low NOx levels possible with lean premixed combustion, such as are possible with the method of '040, are preferred.

[0024] An important aspect of the present invention is that, in contrast to methane, the adiabatic stoichiometric flame temperature of high hydrogen content fuels can be reduced sufficiently to allow ultra low NOx diffusion flame combustion, even for the highest inlet temperature gas turbines thus allowing wide turndown. At the operating temperatures of many turbines, low NOx is achievable with air splits as low as ten or fifteen percent. With the need for carbon sequestration becoming increasingly important, the art has turned to carbon-free hydrogen such as can be produced from syngas. Nitrogen dilution of the fuel may be used for NOx control. Unfortunately, a high dilution is required to reach even ten to 15 ppm NOx.

[0025] As shown in FIG. 3, reducing the hydrogen concentration from 100 percent to 75 percent yields an unacceptably high 200 ppm NOx with conventional combustion, as demonstrated by Todd, D. M., and Battista, R. A., (2000) "Demonstrated Applicability of Hydrogen Fuel for Gas Turbines", Proceedings of Gasification 4 the Future, Noordwijk, Netherland. The data from this reference are shown in FIGS. 3 and 4 and denoted as "Conventional GT". Dilution to 46 percent hydrogen is required even to approach the ten ppm level; the same level that results with 75 percent hydrogen using the method of this invention employing only a ten percent air split. As shown in FIG. 4, increasing the split from ten to

twenty percent increases the amount of fuel reacted and reduces NO_x by greater than a factor of more than two. It should be recognized that increasing the air split for a given fuel flow increases catalytic heat release and, in turn, increases the catalyst temperature. Accordingly, more heat is transferred to the cooling combustion air stream thereby decreasing the stoichiometric flame temperature of the fuel stream and the NO_x production as shown in FIG. 4.

[0026] The maximum allowable air split is determined by the allowable material temperatures. Thus, as shown in FIG. 5, the catalytic wall temperature increases as air split is increased from ten to twenty percent. FIG. 6 shows that wall temperature increases with increased hydrogen content of nitrogen-diluted hydrogen for a given air split. This allows for higher air splits for lower hydrogen content fuel.

[0027] FIG. 7 provides the results of tests performed at conditions simulating operation of the IGCC unit at Tampa Electric Polk power station using the method of this invention. Emissions results at 2550° F. adiabatic flame temperature correspond to baseload operating temperature. At this condition, NO_x emissions were 0.011 lbs/MMBtu or 2.0 ppm corrected to 15% O₂. CO emissions were near zero. As reported on the web, GE report page 12 GER-4219 (May 2003) by R Jones and N. Shilling, this unit operates at less than 25 ppm NO_x; however, post combustion clean-up is required for as low as 10 ppm NO_x. As shown in FIG. 7, a wide turndown at low emissions is provided. In addition, very low NO_x at temperatures hundreds of degrees higher than the Tampa unit combustion temperature are possible.

1. A method of achieving low NO_x in operation of a non-premixed combustion system comprising:

- a) providing a supply of fuel comprising hydrogen gas;
- b) providing a supply of air and splitting the supply of air into a first percentage of air and a second percentage of air;
- c) forming a fuel-rich mixture of the fuel and the first percentage of air;
- d) reacting the fuel-rich mixture to produce partial reaction products comprising a portion of reacted fuel and a portion of unreacted fuel plus a heat of reaction and transferring a portion of the heat of reaction to the second percentage of air thereby producing a heated second percentage of air and cooled partial reaction products; and
- e) passing the cooled partial reaction products into non-premixed contact and combustion with a stoichiometric portion of the heated second percentage of air.

2. The method of claim 1 wherein the non-premixed contact and combustion of the cooled partial reaction products with a stoichiometric portion of the heated second percentage of air defines a reduced flame front temperature.

3. The method of claim 2 wherein the reduced flame front temperature is at least 200 degrees Celsius lower than that of the portion of unreacted fuel.

4. The method of claim 1 wherein the fuel comprises syngas.

5. The method of claim 1 wherein the fuel comprises from about 20 percent hydrogen to about 75 percent hydrogen.

6. The method of claim 1 wherein the fuel comprises from about 75 percent hydrogen to about 100 percent hydrogen.

7. The method of claim 1 wherein the amount of air in the fuel-rich mixture represents at least about twenty percent of the total supply of air.

8. The method of claim 1 wherein step (d) further comprises reacting the fuel-rich mixture in a backside-cooled catalytic reactor.

9. The method of claim 6 wherein step (d) further comprises reacting the fuel-rich mixture in a backside-cooled catalytic reactor and the second percentage of air provides backside cooling.

10. A method of achieving low NO_x in operation of a non-premixed combustion system comprising:

- a. obtaining a supply of fuel comprising hydrogen gas;
- b. obtaining a supply of air and splitting the supply of air into a first percentage of air and a second percentage of air;
- c. forming a fuel-rich mixture of the fuel and the first percentage of air;
- d. reacting the fuel-rich mixture to produce partial reaction products consisting essentially of reacted fuel and remaining unreacted fuel plus a heat of reaction and transferring a portion of the heat of reaction to the second percentage of air thereby producing a heated second percentage of air and cooled partial reaction products; and
- e. combusting the cooled partial reaction products on contact with a stoichiometric portion of the heated second percentage of air in a non-premixed flame such that a second portion of the heated second percentage of air bypasses the non-premixed flame thereby reducing heat liberated in the non-premixed flame.

11. The method of claim 10 wherein the non-premixed contact and combustion of the cooled partial reaction products with a stoichiometric portion of the heated second percentage of air defines a reduced flame front temperature.

12. The method of claim 11 wherein the reduced flame front temperature is at least 200 degrees Celsius lower than that of the portion of unreacted fuel.

13. The method of claim 11 wherein the fuel is syngas.

14. The method of claim 11 wherein the fuel comprises from 20 percent to 75 percent hydrogen.

15. The method of claim 11 wherein the fuel comprises from about 75 percent hydrogen to about 100 percent hydrogen.

16. The method of claim 11 wherein the amount of air in the fuel-rich mixture represents at least about twenty percent of the total supply of air.

17. The method of claim 11 wherein step (d) further comprises reacting the fuel-rich mixture in a backside-cooled catalytic reactor.

18. The method of claim 17 wherein step (d) further comprises reacting the fuel-rich mixture in a backside-cooled catalytic reactor and the second percentage of air provides backside cooling.

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