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**September 25, 2018** 

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APPLICATION NUMBER: 62/549,816

FILING DATE: August 24, 2017

THE COUNTRY CODE AND NUMBER OF YOUR PRIORITY APPLICATION, TO BE USED FOR FILING ABROAD UNDER THE PARIS CONVENTION, IS *US62/549,816* 

By Authority of the

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Certifying Officer

# COMPOSITIONS, METHODS, AND APPARATUSES FOR CATALYTIC COMBUSTION

#### FIELD

[0001] The invention relates to the catalytic combustion of fuel mixtures. In particular, the invention relates to the catalytic combustion of clean fuel mixtures.

#### **BACKGROUND**

[0002] Various fuels have been used to harness energy. Traditionally, the majority of fuels are derived from fossil fuels. However, the use of fossil fuels may result in the generation of emissions of carbon dioxide and other greenhouse gases. There is growing evidence that these emissions may contribute to anthropogenic climate change, and to a decrease in overall air quality.

[0003] In response to these findings, efforts have been made to shift sources of energy from fossil fuels to fuels derived from cleaner sources. For example, coal fired equipment has been shut down in many nations in order to reduce emissions to improve air quality. This equipment may be found, for example, in power plants or district heating plants. These plants may contain other equipment, such as steam equipment and/or turbines, which may be converted for use with other fuels to generate electricity or heating.

One potential source of clean energy is hydrogen. Hydrogen may be produced in a number of ways, including steam reformation or electrolysis. When the electricity for electrolysis is derived from clean sources, such as nuclear, wind, tidal, or solar, the hydrogen produced does not result in the generation of carbon emissions. Hydrogen reacts with oxygen to form water and releases energy.

[0005] This reaction has been used by fuel cells to directly generate electricity. However, factors such as the degradation of membranes and complicated manufacturing processes for fuel cells have held back the adoption of hydrogen as a fuel. Hydrogen has also been used as a fuel in internal combustion engines. However, these systems may be complex and there are challenges with the safe handling of such systems.

[0006] Catalytic combustion systems have been used, but such systems often require complex or costly systems to initiate a self-sustaining reaction, such as preheating systems.

Even when self-sustaining reactions are started, these systems are often limited by temperatures that can be used due to ablation of catalytic material and concerns regarding the ignition of bulk fuel.

[0007] There is a need for improved systems and methods for utilizing clean sources of energy.

#### SUMMARY

[0008] In an aspect, there is provided a catalyst composition including a hydrogen oxidation catalyst (HOC); and an oxygen reduction catalyst (ORC). The catalyst is adapted for low temperature activation of a hydrogen combustion reaction.

[0009] In an aspect, there is provided a process for applying a catalyst composition including providing a substrate; applying a catalyst composition to the substrate to form a catalytic surface. The catalyst composition applied includes a hydrogen oxidation catalyst (HOC) and an oxygen reduction catalyst (ORC). The catalytic surface is adapted for low temperature activation of a hydrogen combustion reaction.

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[0010] In an aspect, there is provided a reactor apparatus. The apparatus includes a conduit having an inlet for receiving a first fluid material and an outlet; a heating element bonded to the conduit comprising a catalytic surface having a catalyst composition applied thereon for catalytically combusting a fuel mixture, the catalyst composition including a hydrogen oxidation catalyst (HOC) and an oxygen reduction catalyst (ORC); and a shell surrounding the conduit, the shell having an inlet for receiving a second fluid material and an outlet. One of the first fluid material and the second fluid material includes the fuel mixture and the other of the first fluid material and the second fluid material includes a heat exchange medium. Energy released by the catalytic combustion of the fuel mixture is transferred to the heat exchange medium. The catalyst composition is adapted to lower the activation energy of the combustion of the fuel mixture such that the reaction occurs under low temperatures.

[0011] In an aspect, there is provided a method for heating a heat exchange medium. A fuel mixture comprising a fuel and an oxidizer is supplied to a catalytic reactor having a catalytic heating element. The catalytic heating element includes a catalytic surface having a hydrogen oxidation catalyst (HOC), and an oxygen reduction catalyst (ORC), and is adapted to lower the activation energy of a combustion of the fuel mixture such that the combustion occurs at low

temperatures. The fuel mixture is catalytically combusted on the catalytic surface. The heat generated by the catalytic combustion is transferred to a heat exchange medium.

[0012] In an aspect, there is provided a catalytic combustor. The combustor includes a catalytic surface having a catalytic composition as described herein, or prepared by a process as described herein.

[0013] In an aspect, there is provided a method for catalytically combusting a fuel mixture. A fuel mixture comprising a fuel and an oxidizer to a catalytic combustor as described herein and is catalytically combusted on a catalytic surface of the catalytic combustor.

#### **BRIEF DESCRIPTION OF DRAWINGS**

[0014] Aspects of the invention will now be described with the following accompanying drawings.

[0015] Figure 1A is a schematic diagram illustrating a catalyst surface showing a continuous phase including a hydrogen oxidation catalyst and a discontinuous phase including an oxygen reduction catalyst.

[0016] Figure 1B is a schematic diagram illustrating a catalyst surface showing a continuous phase including a hydrogen oxidation catalyst and a discontinuous phase including an oxygen reduction catalyst where the total area of the discontinuous phase is the same as that shown in Figure 1A but having a greater interfacial perimeter.

[0017] Figure 2 is a block diagram illustrating a process for applying a catalyst composition according to an embodiment of the invention.

[0018] Figure 3 is an perspective view of a heat exchanger cross section according to an embodiment of the invention.

[0019] Figure 4 is a block diagram illustrating a process for heating a heat exchange medium according to an embodiment of the invention.

[0020] Figure 5A is a schematic diagram illustrating catalyst particles sprayed onto a substrate.

[0021] Figure 5B is a schematic diagram illustrating catalyst that is electroplated onto a substrate according to an embodiment of the invention.

[0022] Figure 6A is an SEM image showing the presence of palladium and tin on a substrate coated according to an embodiment of the invention.

[0023] Figure 6B is an SEM image showing the presence of palladium on a substrate coated according to an embodiment of the invention.

[0024] Figure 6C is an SEM image showing the presence of palladium and tin on a substrate coated according to an embodiment of the invention.

[0025] Figure 7 is a plot showing the temperature of a system operated according to a method according to an embodiment of the invention.

#### **DETAILED DESCRIPTION**

[0026] Throughout the following description specific details are set forth in order to provide a more thorough understanding to persons skilled in the art. However, well known elements may not have been shown or described in detail to avoid unnecessarily obscuring the disclosure. Accordingly, the description and drawings are to be regarded in an illustrative sense, rather than a restrictive sense.

[0027] In an aspect, there is provided a catalyst composition comprising a hydrogen oxidation catalyst (HOC) and an oxygen reduction catalyst (ORC). The catalyst composition adapted for low temperature activation of a hydrogen combustion reaction.

[0028] In some embodiments, the HOC is a material on whose surface molecular hydrogen dissociates into hydrogen radicals. In some embodiments, the HOC is a noble metal. In some embodiments, the noble metal is palladium or platinum. In some embodiments, the noble metal is palladium.

[0029] In some embodiments, the ORC is a material on whose surface molecular oxygen dissociates into oxygen radicals. In some embodiments, the ORC is iron, zinc, silver, copper, tin, their oxides, or any combination thereof. In some embodiments, the ORC is SnO<sub>2</sub>.

[0030] In some embodiments, upon the introduction of molecular hydrogen and molecular oxygen to the catalyst composition, the HOC effects the dissociation of the molecular

hydrogen into hydrogen radicals and the ORC effects the dissociation of molecular oxygen into oxygen radicals. The transport of two of the hydrogen radicals to one of the oxygen radicals at the catalyst surface effects the formation of water. The overall reaction is exothermic and may be considered a catalytic combustion of hydrogen. Energy released by the catalytic hydrogen combustion may be harnessed for other purposes. In some embodiments, the energy released by the reaction is used to heat a heat exchange medium. In other embodiments, the energy released by the reaction is converted into work.

[0031] In some embodiments, the catalyst composition lowers activation energy of the hydrogen combustion reaction such that the reaction occurs at low temperatures. For example, the activation energy of the reaction may be lowered such that the reaction occurs at a temperature below 140 °C, 100 °C, 50 °C, 30 °C, 20 °C, 15 °C, or even below 10 °C. In some embodiments, the hydrogen combustion reaction occurring on the catalyst surface does not ignite bulk molecular hydrogen. In such embodiments, the catalytic combustion is a flameless catalytic combustion.

[0032] In some embodiments, the HOC and the ORC are configured to have high the interfacial perimeter. In some embodiments, the ratio of the surface area of the HOC to surface area of the ORC is between 9:1 and 4:1. In some embodiments, the ratio of the surface area of the HOC to the surface area of the ORC is about 20:3. In some embodiments, these surface area ratios of HOC and ORC provide sufficient interfacial perimeter and amounts of the catalytic material to lower the activation reaction such that the hydrogen combustion reaction may proceed at lower temperatures.

[0033] In some embodiments, with reference to Figs 1A and 1B, on a surface of the catalyst 100, the HOC forms a continuous surface area 110 and the ORC forms discontinuous surface areas 120. In some embodiments, HOC and the ORC are configured to increase the total interfacial perimeter between the continuous surface area and the discontinuous surface areas. For example, reducing the size of each discontinuous surface area while increasing the number of discontinuous surface areas results in increased the total interfacial perimeter for the same total discontinuous surface area.

[0034] When the activation energy for the catalytic hydrogen combustion reaction is lowered such that the reaction occurs at low temperatures, for example, at room temperature, the reaction may be initiated by merely introducing hydrogen and oxygen to the catalyst

composition without requiring a preheating or other initiation step. By removing preheating, the complexity and cost of a catalyst hydrogen combustion system may be reduced. Further, preheating introduces a potential risk of unintended premature hydrogen combustion, which may result in loss of efficiency or safety concerns.

[0035] In some embodiments, the catalyst composition is prepared according to a method as described below. In some embodiments, the catalyst composition is formed by electroplating.

[0036] In an aspect, there is provided a method **200** for preparing a catalyst composition. The process includes providing a substrate at **202**. At **208**, a catalyst composition is applied to the substrate to form a catalytic surface. The catalyst composition includes a hydrogen oxidation catalyst (HOC) and an oxygen reduction catalyst (ORC). The catalytic surface is adapted for low temperature activation of a hydrogen combustion reaction.

[0037] In some embodiments, catalyst composition includes an HOC area and an ORC area. In some embodiments, the catalytic surface includes an HOC area and an ORC area present in a ratio of between about 9:1 and about 4:1. In some embodiments, the ratio of the HOC area and the ORC area is about 20:3.

[0038] In some embodiments, the substrate is resistant to hydrogen embrittlement. In some embodiments, the substrate is a graphite, ceramic, austenitic stainless steel, aluminum, aluminum alloy, or copper alloy. In some embodiments, the substrate is graphite, ceramic, austenitic stainless steel, or copper alloy. In some embodiments, the substrate is graphite or austenitic stainless steel. In some embodiments, the substrate is graphite. In some embodiments, the substrate is 316 stainless steel. In some embodiments, the substrate is a heat conductor. The conduction of heat across the substrate may reduce accumulation of heat at a reaction site, thereby decreasing the formation of hot spots.

[0039] In some embodiments, the applying of the catalyst composition includes applying the HOC at **208A**, and applying the ORC at **208B**. In some embodiments, the HOC is applied prior to the application of the ORC. In some embodiments, the ORC is applied prior to the application of the HOC. In some embodiments, the HOC and the ORC are applied in a single step. In some embodiments, the applying of the catalyst composition includes coating the substrate with the HOC, the ORC, or both. In some embodiments, the coating includes

electroplating or applying a molten catalyst material to the substrate. In some embodiments, the coating includes electroplating.

[0040] In some embodiments, the applying of the catalyst composition results in the formation of a protective surface. For example, in some of embodiments the catalyst composition inhibits the oxidation of the substrate. As such, in some the applying of the catalyst may allow for the usage of a larger variety of substrate materials.

[0041] In some embodiments, the applying of the catalyst composition includes applying precursors of the HOC, the ORC, or both. In such embodiments, the process includes one or more finishing steps to convert the precursors to the HOC, the ORC, or both.

In some embodiments, the substrate has a textured surface. A textured surface may provide improved adhesion of the catalyst material onto the substrate. The surface texture balances the adhesion of the catalyst material to the substrate with thermal conduction and construction requirements. In some embodiments, the substrate surface has a roughness value  $R_a$  of approximately 12.5  $\mu$ m or a ISO Roughness Grade of N10.

[0043] In some embodiments, the substrate is roughened at **204** prior to the applying of any catalyst material to provide a desired texturing. In some embodiments, the substrate is roughened using a mechanical process, such as sanding, sand blasting, filing, scribing scratches, and knurling.

[0044] In some embodiments, the substrate is cleaned at **206** prior to or during the applying any catalyst material. The cleaning removes surface impurities from the substrate to improve adhesion of catalyst material. In some embodiments, the surface impurities include grease, residue from the mechanical roughening treatment, or both. In some embodiments, the cleaning includes electrolytic cleaning, washing with a solvent, or both.

[0045] In some embodiments, the applying of the HOC **208A** includes coating the substrate. In some embodiments, the coating includes electroplating the substrate with the HOC. In some embodiments, the electroplating includes applying a current to a substrate immersed in a HOC electroplating solution comprising a salt of the HOC and a solvent.

[0046] In some embodiments, the salt of the HOC is an ammonium or chloride salt. In some embodiments, the salt of the HOC is ammonium palladium chloride or palladium chloride. In some embodiments, the salt is palladium chloride. In some embodiments, the salt of the HOC

is present at a concentration of between about 5 mM and about 15 mM. In some embodiments, the salt of the HOC is present at a concentration of between about 7 mM and about 10 mM. In some embodiments, the salt of the HOC is present at a concentration of about 8.8 mM.

[0047] In some embodiments, the solvent includes aqueous ammonia.

[0048] In some embodiments, the HOC electroplating solution includes a conductivity agent. In some embodiments, the conductivity agent includes NaCl.

[0049] In some embodiments, the electroplating includes applying a voltage of between about 5V and about 32V. In some embodiments, the electroplating includes applying a current of between about 0.3 A and about 3 A. In some embodiments, the electroplating is effected for about 15 minutes.

[0050] In some embodiments, the thickness of the HOC applied to the substrate is between about 100  $\mu$ m and about 1000  $\mu$ m. In some embodiments, the thickness of the HOC applied to the substrate is about 250  $\mu$ m.

In some embodiments, after the electroplating the HOC, the substrate is treated to stabilize the HOC. In some embodiments, the stability treatment includes heating the substrate with the HOC electrodeposited thereon. In some embodiments, the heating is effected at a temperature of about 100 °C.

[0052] In some embodiments, the applying of the ORC **208B** includes coating the substrate. In some embodiments, the coating includes electroplating the substrate with the ORC. In some embodiments, the electroplating includes applying a current to a substrate immersed in an ORC electroplating solution comprising a salt of the ORC and a solvent.

[0053] In some embodiments, the salt of the ORC is a chloride salt. In some embodiments, the salt of the ORC is stannous chloride. In some embodiments, the salt of the ORC is present at a concentration of between about 5 mM and about 10 mM. In some embodiments, the salt of the ORC is present at a concentration of about 6.6 mM.

[0054] In some embodiments, the salt is produced by dissolving an ORC salt precursor in the solvent. In some embodiments, the ORC salt precursor is an oxide. In some embodiments, the oxide is  $SnO_2$ .

[0055] In some embodiments, the solvent includes hydrochloric acid. In some embodiments, the ORC electroplating solution is prepared by dissolving SnO<sub>2</sub> in HCI. In some embodiments, the ORC electroplating solution is prepared by dissolving 1 g of SnO<sub>2</sub> in 800 mL of 32% HCI.

[0056] In some embodiments, the electroplating includes applying a voltage of between about 5V and about 32V. In some embodiments, the electroplating includes applying a current of between about 0.3 A and about 3 A. In some embodiments, the electroplating is effected for about 7 minutes.

[0057] In some embodiments, the thickness of the ORC applied to the substrate is between about 100  $\mu$ m and about 1000  $\mu$ m. In some embodiments, the thickness of the ORC applied to the substrate is about 250  $\mu$ m.

In some embodiments, the substrate is inspected after the applying of the HOC or the ORC to confirm the coverage of the HOC or the ORC. In some embodiments, the substrate is inspected using SEM/EDX. In some embodiments, if insufficient coverage is obtained, a secondary application of the HOC or the ORC is effected. In some embodiments, the heat treatment (described below) results in a reduction of coverage of the ORC relative to the HOC. For example, whereas up to 40% of surface of the substrate may be covered by the ORC after its application (i.e. a 6:4 ratio of HOC area to ORC area), only 30% of the surface may be covered by the ORC after heat treatment. Inspection prior to heat treatment may be used to determine whether the pre-heat treatment coverage of the ORC is sufficient to obtain a desired post-heat treatment coverage of the ORC. As such, in some embodiments, a target coverage ratio between the ORC and the HOC is higher after application of the catalyst material than after heat treatment.

[0059] A surface coated or electroplated with HOC, ORC, or combination thereof, may exhibit less hot spot formation when undergoing catalytic combustion as compared a surface having granules or particles of catalyst thereof applied thereon. Without wishing to be bound by theory, and with reference to Fig 5A and Fig 5B, it is believed that the applying of granular or particulate catalyst thereof results in relatively small contact areas with the underlying substrate. In contrast, the electroplating of the catalyst material results in the formation of thin catalyst layers where heat is easily transferred to the substrate. When a fuel mixture including a fuel and an oxidizer is introduced to the catalyst surface, the fuel undergoes a combustion process

whereby heat is released. If the heat is not transferred away from the catalyst, localized "hot spots" may be created, and may result in the ablation of catalyst from the surface, or in the ignition of the fuel mixture that can cause a flame or explosion. By increasing the contact area between the catalyst and the underlying substrate, heat may be more efficiently transferred away from the catalyst, reducing the formation of hot spots. The more even distribution of heat may allow the use of higher operating temperatures.

[0060] In some embodiments, the substrate having HOC and ORC applied thereon is subjected to one or more finishing steps at **210**. In some embodiments, the one or more finishing steps include heat treatment. In some embodiments, the heat treatment occurs under air or N<sub>2</sub>. In some embodiments, the heat treatment occurs under air. In some embodiments, the heat treatment is effected after the application of the HOC to the substrate, the application of the ORC to the substrate, or both. In some embodiments, the heat treatment includes a HOC heat treatment and an ORC heat treatment.

[0061] In some embodiments, heat treatment of the HOC is effected after the stability treatment. In some embodiments, heat treatment of the HOC is effected at a temperature of between about 800 °C and about 1200 °C. In some embodiments, the heat treatment of the HOC is effected at a temperature of about 900 °C. In some embodiments, the heat treatment is effected for between about 30 minutes and about 4 hours. In some embodiments, the heat treatment is effected for about one hour. In some embodiments, the heat treatment causes annealing of the HOC applied to the substrate.

[0062] In some embodiments, heat treatment of the ORC is effected after a stability treatment. In some embodiments, heat treatment of the ORC is effected at a temperature of between about 200 °C and about 600 °C. In some embodiments, the heat treatment of the ORC is effected at a temperature of about 400 °C. In some embodiments, the heat treatment is effected for between about 30 minutes and about 4 hours. In some embodiments, the heat treatment is effected for about one hour.

[0063] In some embodiments, the heat treatment activates or increases the catalytic activity of the HOC, the ORC, or both. In some embodiments, the heat treatment activates or increases the activity of the ORC. In some embodiments, the heat treatment effects calcination of the HOC, the ORC, or both. In some embodiments, an ORC precursor is applied and the heat treatment effects formation of the ORC, such as by converting the ORC precursor into the ORC.

In some embodiments, tin is applied to the substrate, and the heat treatment effects the formation of stannous oxide.

[0064] Having reference to Figure 3, in an aspect, there is provided a reactor 300. The reactor 300 includes a conduit 310 having an inlet 312 for receiving a first fluid material and an outlet 314; and a shell 330 surrounding the conduit 310 having an inlet 332 for receiving a second fluid material and an outlet 334. One of the first fluid material and the second fluid material is a fuel mixture. The other of the first fluid material and the second fluid material is a heat exchange medium. The conduit 310 includes heating element 316 having disposed thereon a catalyst composition including a hydrogen oxidation catalyst (HOC) and an oxygen reduction catalyst (ORC). The heating element 316 is configured to effect the catalytic combustion of the fuel mixture and transfer the energy released by the catalytic combustion to the heat exchange medium. In some embodiments, the catalyst composition lowers the activation energy of the catalytic combustion such that it occurs at low temperatures.

[0065] In some embodiments, the catalyst composition is a catalyst composition as described above. In some embodiments, the heating element **316** includes a substrate having the HOC and the ORC applied thereon. In some embodiments, the HOC and the ORC are applied to the substrate using the method as described above. In some embodiments, the ratio of the HOC to the ORC is between about 9:1 and about 4:1 by surface area. In some embodiments, the ratio of the HOC to the ORC is about 20:3 by surface area.

[0066] In some embodiments, the fuel mixture includes a fuel and an oxidizer. In some embodiments, the fuel includes hydrogen, a hydrocarbon material, or a mixture thereof. In some embodiments, the hydrocarbon material is a renewable hydrocarbon material, such as biodiesel, biogas, or algae fuel. In some embodiments, the fuel is hydrogen. In some embodiments, the oxidizer includes oxygen, oxygen-enriched air or air. In some embodiments, the oxidizer is oxygen.

In some embodiments, the inlet receiving the fuel mixture comprises fuel inlet and an oxidizer inlet. In this manner, the fuel mixture does not need to be premixed. By introducing the components of the fuel mixture at the reactor, the composition of the fuel mixture may be better controlled. Any changes to the composition of the fuel mixture introduced to the reactor are effected more rapidly since transport delays may be minimized. Further, without an

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oxidizer present with the fuel, the transport of the fuel and oxidizer separately may be relatively safer than transporting the fuel mixture.

[0068] In some embodiments, the heat exchange medium includes water or helium. In some embodiments, the heat exchange medium includes or is water. In such embodiments, catalytic combustion of the fuel mixture heats the water. In some embodiments, the water is heated to generate steam. Steam generated by the heat exchanger 300 may be used for various purposes. For example, the steam may be used to drive turbines to generate electricity, to provide energy in district heating applications, or in other applications requiring steam.

[0069] In some embodiments, at least a portion of the heating element **116** is integral to a surface of the conduit **310**. In some embodiments, at least a portion of the heating element **116** is bonded to a surface of the conduit **310**.

In some embodiments, the heating element **316** includes a plurality of fins **318**. The addition of fins may increase the surface area on which the catalytic hydrogen combustion can take place. In some embodiments, one or more of the fins are rippled or have a concertina shape to further increase the surface area. In some embodiments, the plurality of fins **318** is bonded to conduit **310**. The bonding of the fins facilitates the transfer of heat, for example via conduction, from the fins **318** to the conduit **310** and any material in contact therewith. For example, where catalytic combustion of the fuel mixture occurs on the surface of the fins **318**, the heat released by the reaction may be transferred to the heat exchange medium via the conduit **310**.

In some embodiments, the heat exchanger 300 includes a distributor 320 for distributing the fuel mixture proximate to the heating element 316. The distribution of the fuel mixture to the heating element 316 may better introduce uncombusted hydrogen to the surface of the heating element 316. In some embodiments, the distributor 320 is fluidically coupled to the inlet (312 or 332) that receives the fuel mixture. In some embodiments, the distributor 320 includes a plurality of hooked pipes, each defining orifices for delivering the fuel mixture to the heating element 316. In some embodiments, the distributor 320 varies the composition of the fuel mixture delivered to different zones of the heating element 316 such that each zone is provided with an individualized fuel mixture. For example, each of the plurality of pipes may deliver an independent fuel mixture.

[0072] In some embodiments, the conduit **310** includes a plurality of tubes **322**. In this manner, the rate of heat transfer to the heat exchange medium is increased by maximizing the ratio of the surface area to the volume. In some embodiments, the tubes **322** are arranged in a single pass arrangement whereby fluid received at one end of the conduit **310** is conducted to another end. In some embodiments, the tubes are arranged in a multi-pass arrangement whereby fluid received at one end of the conduit is conducted to the other end and back to the first end, and optionally for more passes.

[0073] In some embodiments, the conduit **310** receives the heat exchange medium and the shell **330** receives the fuel mixture. In such embodiments, the heating element **316** is disposed on the outside of the conduit **310** and exposed to the interior volume of the shell **330**. Heat generated by the catalytic combustion of the fuel mixture is transferred to the heat exchange medium disposed within the conduit **310**.

[0074] In some embodiments, the catalytic combustion of the fuel mixture effects heating of the shell 330. In some embodiments, a preheater 340 in contact with an exterior surface of the shell 330 transfers heat released by the catalytic combustion of the fuel mixture to the fuel mixture or the heat exchange medium prior to its introduction into the conduit 310 or the shell 330. In some embodiments, the preheater 340 is disposed upstream of the conduit 310 and preheats the heat exchange medium prior to its introduction into the upstream conduit 310. In some embodiments, the heat exchange medium is heated to a temperature of about 90 °C prior to its introduction into the inlet.

[0075] In some embodiments, unreacted fuel in the fuel mixture is recycled back into the heat exchanger. The recycling of fuel reduces the amount of uncombusted fuel that must be treated or stored. In some embodiments, a downstream condenser (not shown) removes water from the combustion products. By removing the water from the combustion products, the recycled fuel may contain less thermal diluent. Thermal diluent may cause a decrease in the operating temperature or an increase in the formation of condensation on the catalytic surface, which may reduce or inhibit combustion.

[0076] In some of those embodiments where the fuel is hydrogen, components configured for contact therewith are made from materials resistant to hydrogen embrittlement. For example, hydrogen embrittlement-resistant materials include austenitic stainless steel, copper, copper alloys, aluminum, aluminum alloys, and ceramics.

[0077] In some of those embodiments where the shell **330** receives the fuel mixture, the interior of the shell is coated with a reflective material to reduce or minimize radiative heat loss. In some embodiments, the heat exchanger includes insulation surrounding the shell. In some embodiments, the insulation is glass wool.

[0078] In an aspect, there is provided a method **400** for heating a heat exchange medium. At **402**, a catalytic surface comprising a hydrogen oxidation catalyst (HOC) and an oxygen reduction catalyst (ORC). The catalytic surface is adapted to lower the activation energy of a combustion of a fuel mixture comprising a fuel and an oxidizer such that the combustion occurs at low temperatures. At **404**, the fuel mixture is supplied to the catalytic surface. At **406**, the fuel mixture is combusted on the catalytic surface. At **408**, the heat generated by the catalytic combustion is transferred to a heat exchange medium.

[0079] In some embodiments, the HOC and the ORC are present in a ratio of between about 9:1 and about 4:1 by surface area. In some embodiments, the HOC and the ORC are present in a ratio of about 20:3.

[0080] In some embodiments, the fuel mixture includes a fuel and an oxidizer. In some embodiments, the fuel includes hydrogen, a hydrocarbon material, or a mixture thereof. In some embodiments, the hydrocarbon material is a renewable hydrocarbon material, such as biodiesel, biogas, or algae fuel. In some embodiments, the fuel is hydrogen. In some embodiments, the oxidizer includes oxygen, oxygen-enriched air or air. In some embodiments, the oxidizer is oxygen.

[0081] In some embodiments, combustion products of the catalytic combustion include water. Accumulation of water on the catalytic surface reduces the surface available to catalyze the combustion of the fuel mixture. In some embodiments, the catalytic combustion occurs at a low pressure. Lower pressures may decrease the boiling point of the water. The lowered boiling point may decrease the accumulation liquid water on the catalytic surface by increasing the rate of vaporization of the water.

[0082] In some embodiments, the supplying of the fuel mixture includes modulating the composition of the fuel mixture. In some embodiments, the supplying of the fuel mixture includes a start-up phase **404A**, an operating phase **404B** or both.

During the start-up phase **404A**, the catalytic surface is disposed at an initial temperature. In some embodiments, the initial temperature of the catalyst surface is below 100 °C, 50 °C, 30 °C, 20 °C, 15 °C, or even below 10 °C. In some embodiments, the composition of the fuel mixture provided during the start-up phase **404A** is relatively oxygen-rich. A relatively oxygen-rich fuel mixture may undergo catalytic combustion at a lower temperature than a fuel-rich mixture. In some embodiments, the ratio of oxygen to fuel of the fuel mixture supplied during the start-up phase is between about 1:2 to about 1:1. In some embodiments, the ratio of oxygen to fuel of the fuel mixture supplied during the start-up phase is about 11:14. The catalytic combustion effects heating of the catalytic surface. In some embodiments, the start-up phase lasts for up to one hour, 45 minutes, 30 minutes, or even 20 minutes.

Having reference to Figure 7, in a particular embodiment of the invention, a fuel mixture was supplied to a reactor 400. The initial temperature of was ambient temperature (approximately 25 °C). The temperature rose slowly for about 10 seconds until it reached approximately 35 °C, at which point the temperature started increasingly more rapidly. Without wishing to be bound by theory, it is believed that at the initial temperature, the energy was sufficient to activate the reaction, but at relatively low reaction rates. As the reaction proceeded, energy 'was released by the exothermic catalytic combustion reaction. This resulted in the observed increase in temperature and provided additional energy, which increased reaction rates and led to faster increases in the temperature. In contrast, in a system fed the same fuel mixture, but where palladium was the sole catalytic material in a similar heating element, there was no observed increase in temperature over ambient temperatures, even after a period of more than one hour.

In some embodiments, once the temperature of the catalytic surface exceeds about 140 °C the process enters an operating phase **404B**. In some embodiments, the composition of the fuel mixture is relatively fuel-rich. In some embodiments, catalytic combustion with a relatively fuel-rich fuel mixture may occur at higher operating temperatures as no excess oxidizer is present to act as a thermal diluent. In some embodiments, the ratio of oxygen to fuel of the fuel mixture supplied during the operating phase is between about 2:5 and about 2:3. In some embodiments, the ratio of oxygen to fuel of the fuel mixture supplied during the operating phase is about 3:4. In some embodiments, the catalytic combustion during the operating phase occurs on the catalytic surface having an operating temperature of higher than 300 °C, 400 °C, 500 °C, or even 600 °C. In some embodiments, the operating temperature is higher than 600 °C. Higher catalytic surface temperatures result in a greater thermal gradient

with the heat exchange medium, thereby increasing the amount of heat transferred to the heat exchange medium.

[0086] In some embodiments, the fuel mixture has limited or no thermal diluent. For example, air is a mixture of gases including nitrogen, oxygen, and carbon dioxide. The non-oxygen gases in air have limited reactivity and may act as thermal diluents absorbing some of the heat released by catalytic combustion such that the temperatures that can be achieved by the catalytic combustion of the fuel mixture is lower than when the fuel mixture includes limited or no thermal diluent. Further, at high operating temperatures, the presence of nitrogen can result in the formation of undesirable NO<sub>x</sub>. As such, in some embodiments, the oxidizer is oxygen-enriched air, or oxygen. In some embodiments, the oxidizer is oxygen.

[0087] In some embodiments, the catalytic combustion is effected at a pressure lower than atmospheric. In some embodiments, the catalytic combustion is effected at a pressure of less than 100 kPa, 90 kPa, 80 kPa, 70 kPa, 60 kPa, or even less than 50 kPa. At pressures lower than atmospheric pressures, the boiling point of water is lowered, thereby reducing the likelihood of the accumulation of liquid water on the catalytic surface. Further, at lower pressures, the combustibility of the bulk fuel mixture may be reduced, which may result in greater safety.

[0088] In some embodiments, the heat exchange medium is water. In some embodiments, the transfer of the heat to the water results in the formation of steam. Steam may be used, for example, to drive turbines, to provide energy for district heating systems, or in desalination operations.

[0089] In some embodiments, the catalytic surface is at least a portion of the surface of a heating element of the heat exchanger described above. In some embodiments, the catalytic surface is prepared according a method as described above. In some embodiments, the catalytic surface includes a catalyst composition as described above.

[0090] In an aspect, there is provided a catalytic combustor. The catalytic combustor includes a catalytic surface. The catalytic surface includes a catalytic composition as described above or is prepared by a process as described above.

[0091] In some embodiments, the heat generated by the operation of the catalytic combustor is used as a heat source to effect work. In some embodiments, the heat generated

by the operation of the catalytic combustor is used to drive a Stirling Engine, a turbine, a phase change operation for air conditioning, a district heating system, or a desalination system. For example, in some embodiments, the catalytic combustor is a heating element incorporated into reactor 300. In another embodiment, the catalytic combustor is a graphite block having a hole adapted to receive a expansion cylinder of a Stirling engine. In some embodiments, the heat generated by the operation of the catalytic combustor is used to generate steam. For example, the catalytic combustor may be integrated into a reactor as described above.

[0092] In an aspect, there is provided a method for catalytically combusting a fuel mixture. A fuel mixture including a fuel and an oxidizer is supplied to a catalytic combustor as described above and catalytically combusted on the catalytic surface of the catalytic combustor. In some embodiments, the catalytic combustion is effected similarly to the heating of a heat exchange medium as described above except that the heat is not necessarily transferred to the heat exchange medium.

#### **EXAMPLES**

### Example 1 - Preparation of a catalyst composition

[0093] A disc was cut from 316 stainless steel sheet having a thickness of 0.8 mm. Mounting holes were drilled into the disc. The surface of the disc was mechanically roughened to an ISO roughness Grade N10 using carborundum paper having progressively finer grits from 30 through 800.

Work

[0094] An HOC electroplating solution was prepared by dissolving 1 g of PdCl in 800 mL of 30% ammonia. The disc and a palladium electrode of similar area were mechanically suspended in the solution and an electrical current of 1 A at 30 V was applied for 15 minutes. The objective was to obtain a uniform coverage of palladium approximately 250 µm in thickness.

[0095] Once the plating was complete the disc was removed, rinsed with distilled water and gently patted dry with a delicate task wiper to remove excess remaining liquid. The disc was then baked in an industrial furnace at 100 °C for approximately 15 minutes to remove residual electroplating solution.

[0096] A visual inspection of the surface was performed to confirm that a uniform coating was applied with no visible substrate. If the coverage was marginal, a multi-point SEM/EDX inspection was performed. The electroplating process was repeated if required.

[0097] Once coverage was confirmed, the disc was annealed at 900 °C for 1 hour and then allowed to cool to room temperature.

[0098] An ORC electroplating solution was prepared by dissolving 1 g of SnO<sub>2</sub> in 800 mL of 32% HCl.

[0099] The cooled disk was mechanically suspended in the ORC electroplating solution and an electrical current of 10V at 3A was applied across the disc and a palladium electrode of similar dimensions for 7 minutes.

[00100] Once the ORC electroplating was complete, the disc was removed and rinsed with distilled water. The inspection process was repeated to confirm whether the electroplating deposited tin to at least 30% of the surface.

[00101] When the coverage was confirmed, the disc was annealed at 400 °C for 1 hour.

[00102] An SEM/EDX image showing the catalyst composition as applied to a substrate is provided at Figures 6A-C. The settings used to obtain the image were as follows:

[00103] Acc. Voltage: 15.0 kV

[00104] Resolution: 512 x 512 pixels

[00105] Viewed Resolution: 50%

[00106] Process Time: 5

[00107] Image Width: 1.567 mm

[00108] Mixed Map: Palladium La1 (red), Tin La1 (green).

#### Example 2 – Catalytic combustion with the catalytic composition

[00109] A reactor having a catalytic composition applied to a heater disposed therein, as shown in Figure 3 was used. The system was run such that the conduit received water and the shell received the fuel mixture.

[00110] The system was primed with water from a tank using a pump until it flowed out of the steam/water outlet through an open valve. The water subsystem outlet valve was then

closed. The pressure in the internal environment of the shell was lowered to obtain a vacuum of about 2 kPa(a). Hydrogen and oxygen were then introduced to the internal cavity though a regulator and controlled with a mass flow controller. The flow was initially set to deliver the hydrogen and oxygen in a ratio of about 14:11. A series of thermocouples were used to monitor internal and external temperatures.

[00111] Having reference to Fig 7, the internal temperature was initially about 25 °C. For approximately 10 seconds, there was a relatively slow initial temperature increase. At a temperature of between approximately 40 °C and 60 °C, the rate of temperature increase accelerated significantly. The temperature leveled off at between approximately 350 °C and 400 °C.

[00112] The water was circulated into the subsystem and the steam pressure was controlled by opening the output valve to maintain a steam pressure of below 800 kPa.

[00113] The flow the fuel mixture was then adjusted to increase the amount of  $H_2$  delivered and then the amount of  $O_2$  delivered until the vacuum system is was longer able to maintain an internal pressure of below approximately 100 kPa(a).

[00114] The temperature increased to approximately 600 °C and high quality steam was produced.

[00115] While a number of exemplary aspects and embodiments have been discussed above, those of skill in the art will recognize certain modifications, permutations, additions and sub-combinations thereof are possible. In the above description, for purposes of explanation, numerous details are set forth in order to provide a thorough understanding of the present disclosure. However, it will be apparent to one skilled in the art that these specific details are not required in order to practice the present disclosure. Although certain dimensions and materials are described for implementing the disclosed example embodiments, other suitable dimensions and/or materials may be used within the scope of this disclosure. All such modifications and variations, including all suitable current and future changes in technology, are believed to be within the sphere and scope of the present disclosure. All references mentioned are hereby incorporated by reference in their entirety.

#### WHAT IS CLAIMED IS:

1. A catalyst composition comprising:

a hydrogen oxidation catalyst (HOC); and an oxygen reduction catalyst (ORC);

wherein the catalyst is adapted for low temperature activation of a hydrogen combustion reaction.

- 2. The catalyst composition of claim 1, wherein the ratio of the surface area of the HOC to surface area of the ORC is between about 9:1 and about 4:1.
- 3. The catalyst composition of claim 2, wherein the ratio of the surface area of the HOC to the surface area of the ORC is about 20:3.
- 4. The catalyst composition of claim 1, wherein the HOC and the ORC are formed by electrodeposition.
- 5. The catalyst composition of claim 1, wherein the catalyst is adapted to activate hydrogen combustion at a temperature of below about 140 °C.
- 6. The catalyst composition of claim 5, wherein the catalyst is adapted to activate hydrogen combustion at a temperature of below 20 °C.
- 7. The catalyst composition of claim 1, wherein the HOC is a noble metal.
- 8. The catalyst composition of claim 7, wherein the HOC is platinum or palladium.
- 9. The catalyst composition of claim 8, wherein the HOC is palladium.
- 10. The catalyst composition of claim 1, wherein the ORC is stannous oxide.
- A process for applying a catalyst composition comprising:
   providing a substrate;

applying a catalyst composition to the substrate to form a catalytic surface, wherein the catalyst composition comprises a hydrogen oxidation catalyst (HOC) and an oxygen reduction catalyst (ORC);

wherein the catalytic surface is adapted for low temperature activation of a hydrogen combustion reaction.

- 12. The process of claim 11, wherein the catalytic surface includes an HOC area and an ORC area having a ratio of between about 9:1 and about 4:1.
- 13. The process of claim 12, wherein the ratio of the HOC area to the ORC area is about 20:3.
- 14. The process of claim 11, wherein the applying of the catalyst composition comprises applying of the HOC and applying of the ORC.
- 15. The process of claim 11, wherein the applying of the catalyst composition includes electroplating the HOC, the ORC, or both to the substrate.
- 16. The process of claim 11, further comprising heating treatment of the catalyst-applied substrate.
- 17. A reactor apparatus comprising:

a conduit including:

an inlet for receiving a first fluid material, and

an outlet, and

a heating element bonded to the conduit comprising a catalytic surface having applied thereon, a catalyst composition for catalytically combusting a fuel mixture, the catalyst composition including a hydrogen oxidation catalyst (HOC) and an oxygen reduction catalyst (ORC); and

a shell surrounding the conduit, the shell having an inlet for receiving a second fluid material and an outlet;

wherein one of the first fluid material and the second fluid material includes the fuel mixture and the other of the first fluid material and the second fluid material includes a heat exchange medium.

wherein energy released by the catalytic combustion of the fuel mixture is transferred to the heat exchange medium, and

wherein the catalyst composition is adapted to lower the activation energy of the combustion of the fuel mixture such that the reaction occurs under low temperatures.

- 18. The reactor of claim 17, wherein the catalytic surface includes an HOC area and an ORC area having a ratio of between about 9:1 and about 4:1.
- 19. The reactor of claim 18, wherein the ratio of the HOC area to the ORC area is about 20:3.
- 20. The reactor of claim 17 wherein the fuel mixture comprises hydrogen and oxygen.
- 21. The reactor of claim 20, wherein the fuel mixture consists of hydrogen and oxygen.
- 22. The reactor of claim 17 wherein the heat exchange medium comprises water.
- 23. The reactor of claim 22, wherein the transfer of the energy to the water effects vaporization to produce steam.
- 24. The reactor of claim 17, wherein the heating element further comprises at least one fin, and at least a portion of the catalyst surface is on the at least one fin.
- 25. The reactor of claim 17, further comprising a distributor for distributing the fuel mixture proximate to the heating element.
- 26. The reactor of claim 17, further comprising a preheater upstream of and in thermal contact with the reactor, for preheating the heat exchange medium or the fuel mixture.
- 27. A method for heating a heat exchange medium comprising:

supplying a fuel mixture comprising a fuel and an oxidizer to a catalytic reactor having a catalytic heating element including a catalytic surface having a hydrogen oxidation catalyst (HOC), and a oxygen reduction catalyst (ORC), the catalytic surface adapted to lower the

activation energy of a combustion of the fuel mixture such that the combustion occurs at low temperatures;

catalytically combusting the fuel mixture on the catalytic surface; and transferring the heat generated by the catalytic combustion to a heat exchange medium.

- 28. The process of claim 27, wherein the HOC and the ORC are present on the catalytic surface in a ratio of between about 9:1 and about 4:1 by surface area.
- 29. The process of claim 28, wherein the HOC and the ORC are present on the catalytic surface in a ratio of about 20:3 by surface area.
- 30. The process of claim 27, wherein the fuel is hydrogen and the oxidizer is oxygen.
- 31. The process of claim 27, wherein the catalytic combustion occurs at a pressure below atmospheric pressure.
- 32. A catalytic combustor including a catalytic surface including a catalytic composition as defined any one of claims 1 to 10 or prepared by a process as defined in any one of claims 11, to 16.
- 33. A method for catalytically combusting a fuel mixture comprising:

5

11.

supplying a fuel mixture comprising a fuel and an oxidizer to a catalytic combustor as defined in claim 32; and

catalytically combusting the fuel mixture on the catalytic surface.

### **ABSTRACT**

There is provided a catalyst composition including a hydrogen oxidation catalyst and an oxygen reduction catalyst. The catalyst is adapted for low temperature activation of a hydrogen combustion reaction.

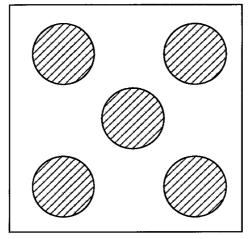


FIG 1A

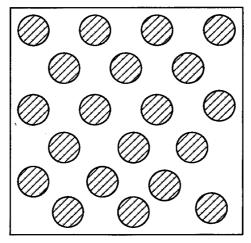
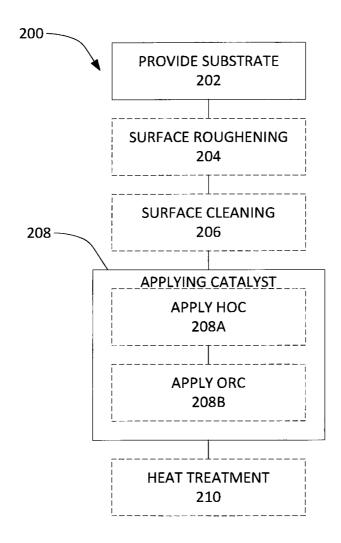
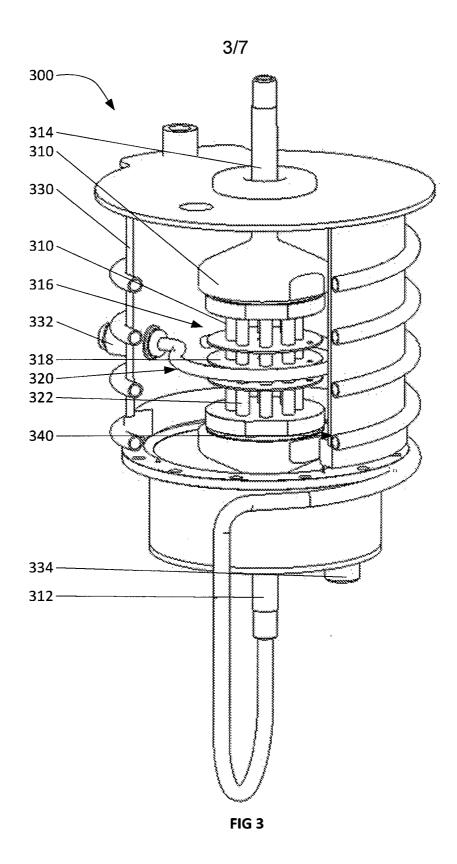


FIG 1B



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FIG 2



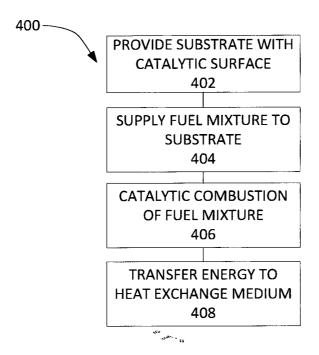
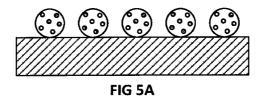
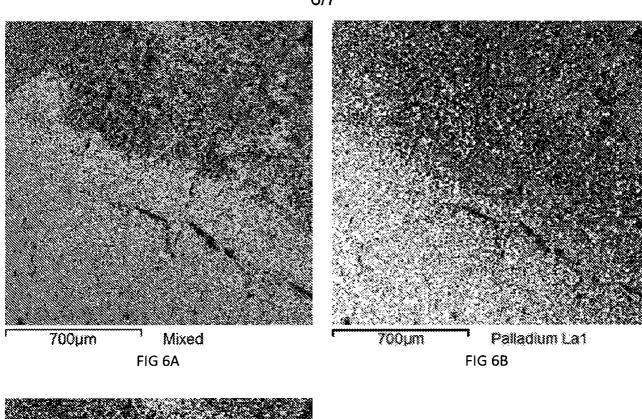
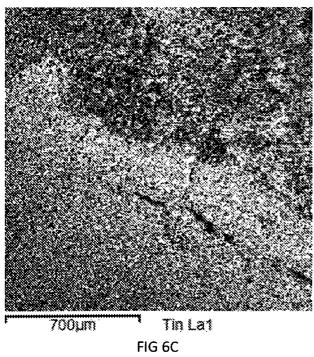


FIG 4







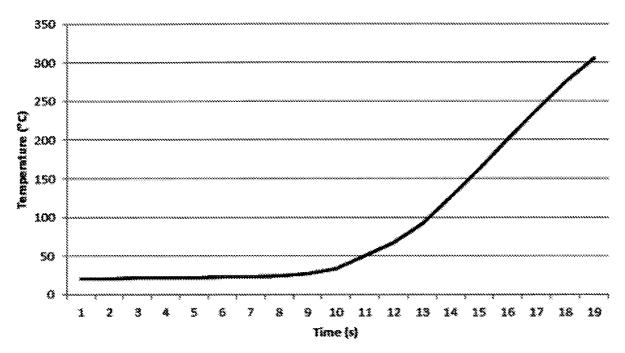


FIG 7

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Application Data Sheet 37 CFR 1.76		Attorne	y Docket Number	10242-0012		
		Applica	tion Number			
Title of Invention	Compositions, Methods, a	nd Apparatus	es for Catalytic Comb	oustion		
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		/	Application Number							7	
Title of Invention	Title of Invention Compositions, Methods, and Apparatuses for Catalytic Combustion										
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Subject Matter		Utility	TL								<b>-</b>
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Application Data Sheet 37 CFR 1.76		Attorney Docket Number	10242-0012
		Application Number	
Title of Invention	Compositions, Methods, and Apparatuses for Catalytic Combustion		

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This section allows for the applicant to claim priority to a foreign application. Providing this information in the application data sheet constitutes the claim for priority as required by 35 U.S.C. 119(b) and 37 CFR 1.55. When priority is claimed to a foreign application that is eligible for retrieval under the priority document exchange program (PDX)<sup>1</sup> the information will be used by the Office to automatically attempt retrieval pursuant to 37 CFR 1.55(i)(1) and (2). Under the PDX program, applicant bears the ultimate responsibility for ensuring that a copy of the foreign application is received by the Office from the participating foreign intellectual property office, or a certified copy of the foreign priority application is filed, within the time period specified in 37 CFR 1.55(g)(1).

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# Statement under 37 CFR 1.55 or 1.78 for AIA (First Inventor to File) Transition Applications

This application (1) claims priority to or the benefit of an application filed before March 16, 2013 and (2) also
contains, or contained at any time, a claim to a claimed invention that has an effective filing date on or after March
16, 2013.
 NOTE: By providing this statement under 37 CFR 1.55 or 1.78, this application, with a filing date on or after March
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Application Data Sheet 37 CFR 1.76		Attorney Docket Number	10242-0012
		Application Number	
Title of Invention	Compositions, Methods, and Apparatuses for Catalytic Combustion		

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When this Application Data Sheet is properly signed and filed with the application, applicant has provided written authority to permit a participating foreign intellectual property (IP) office access to the instant application-as-filed (see paragraph A in subsection 1 below) and the European Patent Office (EPO) access to any search results from the instant application (see paragraph B in subsection 1 below).

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U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number. Attorney Docket Number 10242-0012 **Application Data Sheet 37 CFR 1.76 Application Number** Compositions, Methods, and Apparatuses for Catalytic Combustion Title of Invention Assignee Complete this section if assignee information, including non-applicant assignee information, is desired to be included on the patent application publication. An assignee-applicant identified in the "Applicant Information" section will appear on the patent application publication as an applicant. For an assignee-applicant, complete this section only if identification as an assignee is also desired on the patent application publication. Remove If the Assignee or Non-Applicant Assignee is an Organization check here. **Family Name Suffix** Middle Name **Prefix** Given Name Mailing Address Information For Assignee including Non-Applicant Assignee: Address 1 Address 2 City State/Province Postal Code Country i Phone Number Fax Number **Email Address** Additional Assignee or Non-Applicant Assignee Data may be generated within this form by Add selecting the Add button. Remove Signature: NOTE: This Application Data Sheet must be signed in accordance with 37 CFR 1.33(b). However, if this Application Data Sheet is submitted with the <u>INITIAL</u> filing of the application <u>and</u> either box A or B is <u>not</u> checked in subsection 2 of the "Authorization or Opt-Out of Authorization to Permit Access" section, then this form must also be signed in accordance with 37 CFR 1.14(c). This Application Data Sheet must be signed by a patent practitioner if one or more of the applicants is a juristic entity (e.g., corporation or association). If the applicant is two or more joint inventors, this form must be signed by a patent practitioner, all joint inventors who are the applicant, or one or more joint inventor-applicants who have been given power of attorney (e.g., see USPTO Form PTO/AIA/81) on behalf of all joint inventor-applicants. See 37 CFR 1.4(d) for the manner of making signatures and certifications. Signature /James David Johnson/ Date (YYYY-MM-DD) 2017-08-24 First Name James **Last Name** Johnson Registration Number 47685

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Application Data Sheet 37 CFR 1.76		Attorney Docket Number	10242-0012
		Application Number	
Title of Invention	Compositions, Methods, and Apparatuses for Catalytic Combustion		

This collection of information is required by 37 CFR 1.76. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 23 minutes to complete, including gathering, preparing, and submitting the completed application data sheet form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.** 

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