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AN INVESTIGATION OF A CATALYTICALLY-IGNITED AUTOTHERMAL REFORMER PRE-HEATING SYSTEM FOR HYDROGEN PRODUCTION IN A FUEL CELL POWERED VEHICLE

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ABSTRACT

Partial oxidation (POX), catalytic steam reforming (CSR) and autothermal reforming (ATR) are some of the main processes widely used to produce hydrogen. Autothermal Reformer also defines as a steam reforming reaction and a partial oxidation reaction that take place over microscopic distances at the same catalytic site thus avoiding complex heat exchanging. The steam reforming reaction absorbs part of the heat generated by the partial oxidation process reaction, limiting the maximum temperature in the reactor. For a catalytically ignited autothermal reformer system, catalysts need to be preheated to its light-off or catalytic ignition temperature before the reactions can be self-sustained. The objective of this research is to come out with the practical method for starting-up an autothermal reformer system in the shortest possible preheating time. A catalytic burner has been proposed to supply the required energy for pre-heating the autothermal reactor. This proposed catalytic burner will use available hydrogen and natural gas as fuel and platinum on the honeycomb monolith surface as catalyst. Platinum-type catalyst can readily reacts hydrogen with air under room temperature to produce heats that aid the catalyst to reach light off temperature. The energy from catalytic combustion will be used to preheat the autothermal reformer. Ceramic honeycomb monolith with platinum based washcoat will be used a burner system. The results will then be further verified using CFD-ACE code and as well as compared with that of conventional preheating methods such as electrical coil.

Keywords: Catalytic burner; hydrogen; natural gas; platinum; honeycomb monolith; washcoat.

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ABSTRAK

Tindakbalas Pengoksidaan separa (POX), pembentukan semula stim bermangkin (CSR), dan pembentukan semula autotermal (ATR) adalah sebilangan proses utama bagi penghasilan hidrogen. Pembentukan semula autotermal juga ditakrifkan sebagai tindakbalas pembentukan semula stim bermangkin dan pengoksidaan separa yang berlaku di dalam jarak mikroskopik pada kawasan pemangkinan yang sama bagi mencegah penukaran haba yang kompleks. Tindak balas pembentukan semula stim bermangkin menyerap haba yang dihasilkan oleh proses tindakbalas pengoksidaan separa. Oleh itu, suhu maksimum di dalam reactor dapat di hadkan. Bagi pencucuhan bermangkin sistem autothermal, mangkin itu perlu dipanaskan kepada suhu pencucuhannya sebelum tindakbalas penghasilan hidrogen boleh kekal dengan sendirinya. Objektif penyelidikan ini adalah untuk menghasilkan kaedah yang praktikal bagi permulaan pembentukan semula autotermal dalam masa yang singkat. Penunu bermangkin dicadang untuk membekalkan tenaga yang diperlukan untuk pra-pemanasan reactor autotermal. Penunu bermangkin yang dicadangkan ini menggunakan hidrogen yang sedia ada dan gas asli sebagai bahan api dan platinum pada permukaan *honeycomb monolit* sebagai pemangkin. Mangkin jenis platinum boleh bertindakbalas dengan hidrogen dan udara pada suhu bilik bagi menghasilkan haba untuk membantu mencapai suhu pencucuhan. Tenaga daripada penunu bermangkin akan digunakan untuk proses pra-pemanasan pembentukan semula autotermal. *Honeycomb monolit* seramik dengan platinum yang berasaskan saduran akan digunakan untuk system penunu. Seterusnya, kesahihan keputusan ini akan diperolehi dengan menggunakan kod CFD-ACE dan akan dibandingkan dengan kaedah pra-pemanasan konvinsional seperti gelung elektrik.

INTRODUCTION

The principle of the operation of catalytic combustion is simple. The catalyst is heated to the point where oxidation can begin and a mixture of fuel and air is passed through the system. Oxidation is initiated, the temperature rises and the system become self-sustaining (Trimm, 1991). Autothermal reforming also needs some total heat energy to activate the catalyst before it can be self-sustained for starting produce hydrogen. Normally, researchers use furnace or electrical heater to pre-heat catalyst or air in their experiment in the laboratory. In the automotive system, the electrical power is supplied by battery. The current vehicles are using battery 12 volts compared 220 volts for laboratory furnace. 12 volts battery is not enough to support electrical heater requirement for preheat catalyst up to 350 ^{O}C. Therefore, catalytic combustion is suggested to apply in the real problem for fuel cell vehicle. According to U.S. Department of Energy (2000), catalytic burner is use to supply heat to preheat autothermal reformer in fuel cell vehicle. The amount of heat will supply to autothermal reformer by catalytic burner.

Catalytic combustion required a minimum temperature approximately 350 °C to get proper combustion. Until recently, the necessary heat for start up was provided from several sources such as electrical heater, pilot burner, igniter, hot exhaust gas and compressor (Moallemi et.al 1999, Dalla Betta et. al 1995, Mandai and Gora, 1995, Furuya et. al, 1987). For example, automotive catalytic converter use hot exhaust gas to preheat the converter before it starts up. Due to the heat capacity of the exhaust system it takes about 2 minutes after the start of the engine before the temperature level is reached (Kirhner and Eigenberger, 1996). The main purpose of this research is to reduce the preheating time to less than 2 minute.

Catalytic combustion has been vigorously explored as a route to the production of heat and energy in view of it's capability to achieve effective combustion at much lower temperature than in conventional flame combustion, thus allowing for the simultaneous ultra-low emission of NOx, CO and unburned hydrocarbons (UHC) (Forzatti and Groppi, 1999).

Catalytic combustion of hydrocarbons is becoming an increasingly viable technology in power generations, domestic heating appliances and chemical process heaters. (Dupont et.al , 2002, Choudhary et. al, 2002). In general, catalytic combustion applications involve heterogeneous catalyst and the catalyst retained within the combustion chamber. In heterogeneous catalysis the reaction occurs at the interface between the gas phase and the solid catalyst. This occurs not only at the external surface of the catalyst but also on the interior of the catalyst if it is porous.

Catalytic combustion also was introduced to reduce NOx and CO production from conventional combustion. It's also increase performance of combustion and the reactions almost reached complete combustion. Catalytic combustion includes several essential process or mechanism (Lee and Trimm, 1995, Hayes and Kolaczkowski, 1997):

I. Diffusion of the reactants from the gas phase to the catalytic surface
II. Adsorption of the reactants onto the catalytic surface
III. Movement of adsorbed species
IV. Reaction on the surface of the catalyst
V. Desorption of the product from the surface
VI. Diffusion of the products from the catalytic surface to the gas surface.

The objective of this research is to supply a total heat to autothermal reformer at cold-start process in short time to produce hydrogen. In this research, we propose catalytic burner as a heat supply system. The catalyst will be used is platinum coated on monolith honeycomb surface (Dupont et. al, 2002). Hydrogen is used to start combustion at room temperature (Deutschman et. al, 2000) and it act as a replacement for spark plug glow plug or electrical heater for started reaction. The experimental will run starts with a pure hydrogen/air flow through the monolith reactor that is ignited catalytically. Then, the methane feed is slowly increased and at the same time hydrogen concentration will be reduce. This research try to recognized the best optimum and minimum ratio for combination of hydrogen and methane in the started ignition process. We also want to know the level that catalytic burner needed external heat to started ignition in term of to get faster reaction time.

METHODLOGY

Both experimental and simulation were conducted simultaneously in this research to obtain the end result. The experimental approach is divided into two, which includes the washcoat preparation to prepare catalyst on honeycomb monolith surface and testing catalyst. The main focus of the experiment is the washcoat technique to produce non-porous catalyst by impregnation (Perego and Villa, 1997) and sol-gel method (Cho et. Al, 1998, Gonzalez et. al 1997).

This research will develop our own formulation of washcoat solution, technique and procedure to test various percentage weight of catalyst loading. For catalytic combustion, we used platinum as catalyst because platinum is the most commonly used as catalyst for the catalytic oxidation of methane (Goralski, 1998). For testing purpose, the self-made honeycomb monolith will compared with honeycomb monolith supply by Johnson Mattey (UK).

In this simulation approach, the study is including single channel monolith and the overall honeycomb monolith coated catalyst. The single channel model has been done developed by using CFD-GEOM. Now, the process is to define platinum and their properties in the material database before run the solver (CFD-ACE).

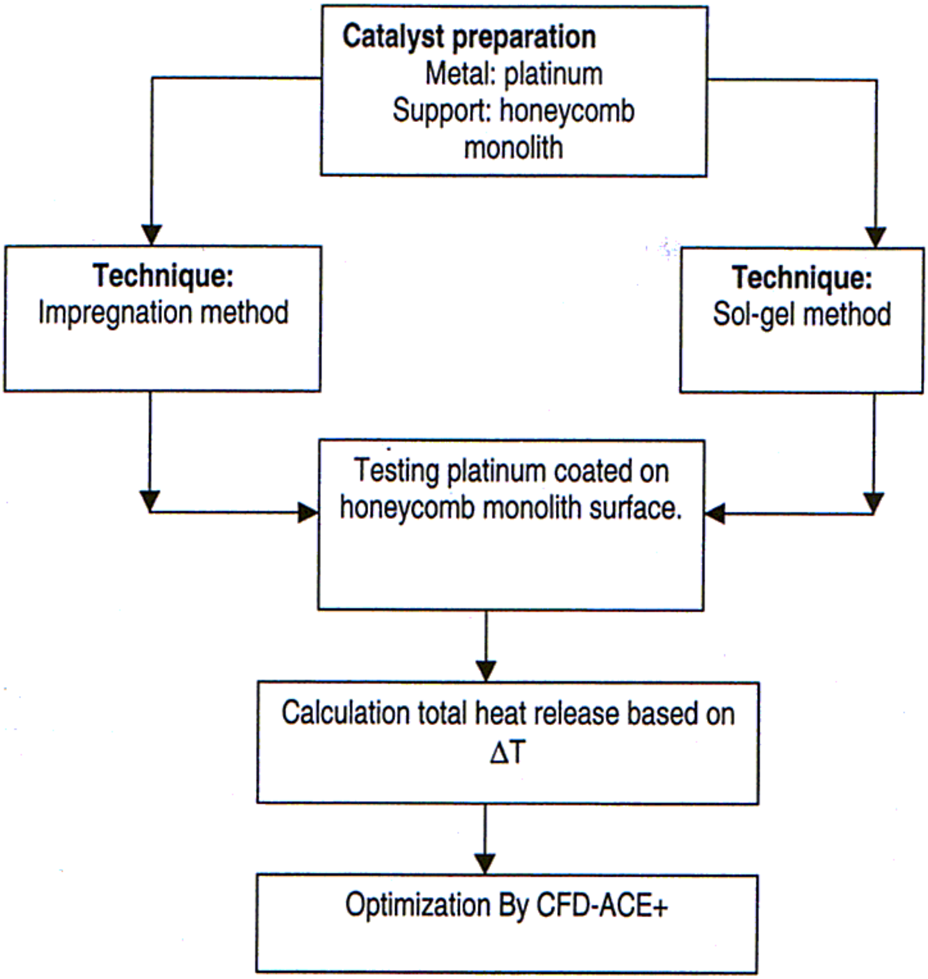

FIGURE 1 Flow chart of research methodology

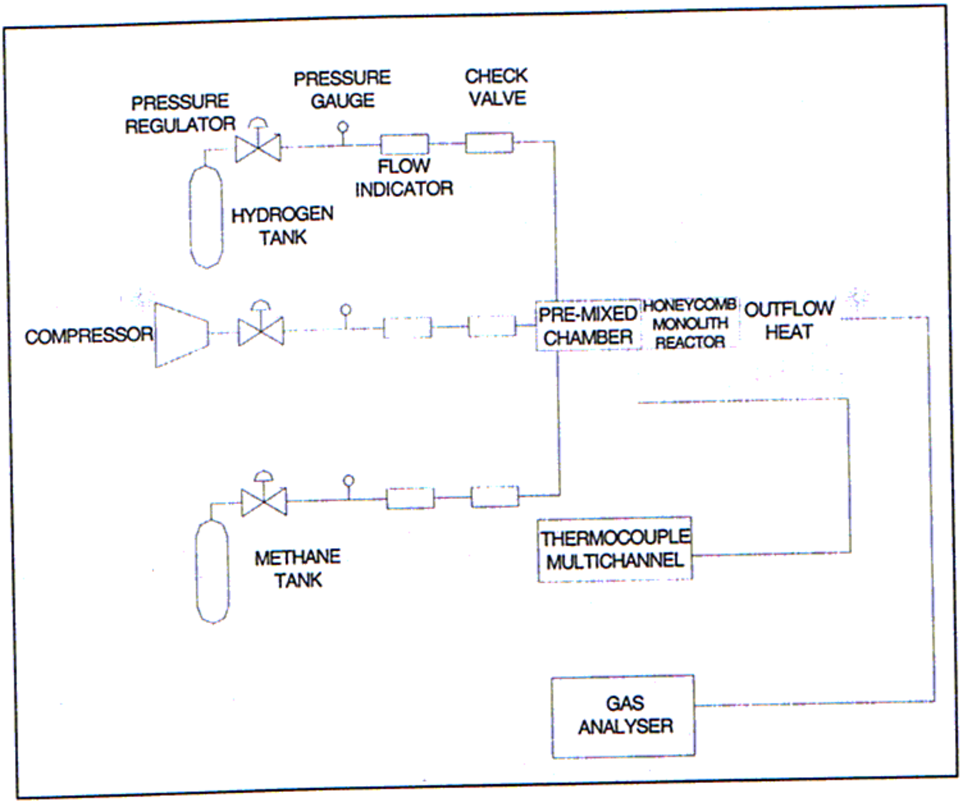

FIGURE 2 Experimental rig for catalytic combustion system

EXPECTED RESULT

Total oxidation will be occurs on the surface when hydrogen reacts with platinum surface at room temperature immediately. This reaction produce amount of heat to aid catalytic burner reached the ignition temperature. When reaching the ignition temperature of the heterogeneous reaction, the temperature of the catalyst rises rapidly because of heat release by the exothermic surface reaction. The heat release will be transferred to an autothermal reformer to preheat the system.

Refer to Deutschman paper; the catalytic burner can be reached uniform temperature at 1040 K in 50 s. We expected the total time including preheating time is less than 2 minute. This temperature and time can be manipulated by controlling some parameters such as oxygen flow rate, methane flow rate, hydrogen flow rate and catalyst loading.

CONCLUSION

This research will be reached the objective in development of new preheating system for autothermal reforming reactor. Combination of hydrogen and methane can reduce the ignition temperature of fuels (Yurum, 1995). This catalytic burner will produce amount of heat depends on autothermal reforming requirement for start-up process without using external heat device.

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