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clean fuel technology of the future today

H2 technology : White paper, Fuel processor technology

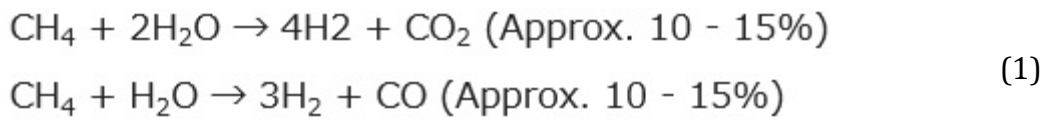**Date : 15/6/2015 version : 1.2**

Abstract

This work attempts a short and comprehensive description of Fuel Processors, mainly for transportation applications. The white paper is divided into two parts; the first covers a technical description of science and current products in the fuel processing field, the second part is a scientific literature review of published research works on the recent developments and achievements in the field.

Part A: Technical Review

Fuel processing and fuel processors working principle is quite simple; when fuels are burnt into a combustion chamber, a great part of the fuel remains unburnt or partially burnt and exists the chamber without utilization of its full energy content. This phenomenon leads to great energy losses, especially for the transportation sector. Fuel processors try to amend this faulty process by increasing the utilization of fuels energy content.

Fuel processors act on fuels to break the chains of hydrocarbons and alcohols [mainly] repeatedly until only hydrogen and carbon dioxide remain. All hydrocarbons have a general formula of C_CH_H and other organic compounds have similar formulas including mostly oxygen and nitrogen. Other elements are present as well as impurities such as sulphur. Fuel processors use a thermal or catalytic or thermo catalytic process to break down these formulas to H_2 and CO_2 (equation 1). Obviously, these are not the only compounds present in the final fuel mixture. The hydrogen- rich mixture contains, besides carbon dioxide, significant amounts of carbon monoxide, NO_x and SO_x which are harmful to the process, the environment and to human health.

$$CH_4 + 2H_2O \rightarrow 4H2 + CO_2 \text{ (Approx. 10 - 15\%)}$$
$$CH_4 + H_2O \rightarrow 3H_2 + CO \text{ (Approx. 10 - 15\%)}$$

(1)

Carbon monoxide produced in this reforming process is extremely toxic to human health; but more importantly, it is toxic for the reforming process as it is highly toxic [poison] for the reforming catalyst. Carbon monoxide is further treated with steam until is fully converted to carbon dioxide (equation 2). One note should be kept in mind; although carbon dioxide is of no concern for the transportation industry, it admittedly adds to the green house phenomenon and it is expected to be controlled soon [see also CO_2 emission rights]. NO_x and SO_x are highly controlled; however the reforming process seems to produce ultra low concentrations of these groups.

$$CO + 2H_2O \rightarrow H_2 + CO_2$$

(2)

$$CO + 1/2O \rightarrow CO_2$$
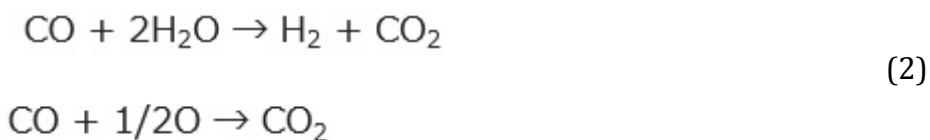
Theoretically any fuel can undergo reforming for utilization of its hydrogen content. Hydrogen has many advantages such as high energetic content, totally clean combustion [only water is the outcome] and unlimited sources [fuels, water, etc]. The only drawback of hydrogen use is its low volumetric energy content. This is why this process is so important; by utilizing common fuels with low combustion efficiency [20-60%], hydrogen is produced; hydrogen due to low size and gaseous phase reaches almost complete efficiency [no mass is wasted].

Up to date fuel processors have been manufactured for gasoline, ethanol, diesel, biofuels, etc. Obviously, based on the initial fuel, the efficiency gain will range widely. Fuel processors are based on different reactors, including catalytic reforming reactors (autothermal reforming reactors, partial oxidation reforming reactors, methane reforming and diesel steam reforming reactors), water gas shift reactors, selective CO preferential oxidation reactors (PROX), desulfurizers, and anode gas burners.

A typical reforming process [fuel processor] is given in Figure 1. Another commercial autothermal reactor is presented in Figure 2. The autothermal reactor contains a catalyst (green), which runs at 750°C bringing an LPG-air-steam mixture to chemical equilibrium. The feed stream is preheated using the hot reformate coming out of the catalyst using a recuperative heat exchanger (pink). The purple part upstream of the catalyst is the startup heater combined with flame arrestor. A whole enrichment process is given in Figure 3.
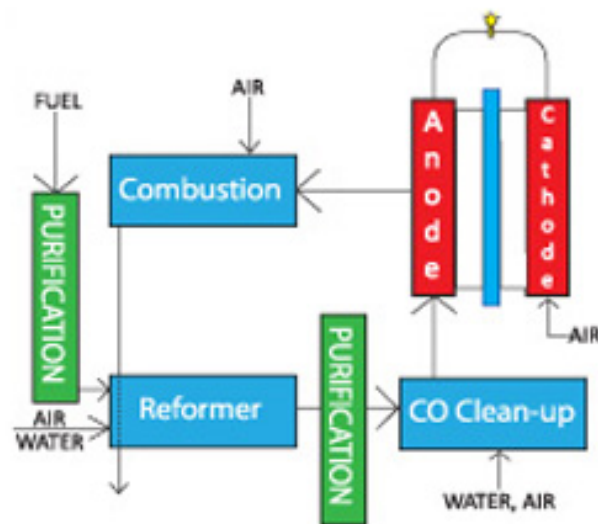
Figure 1: A typical reforming reactor
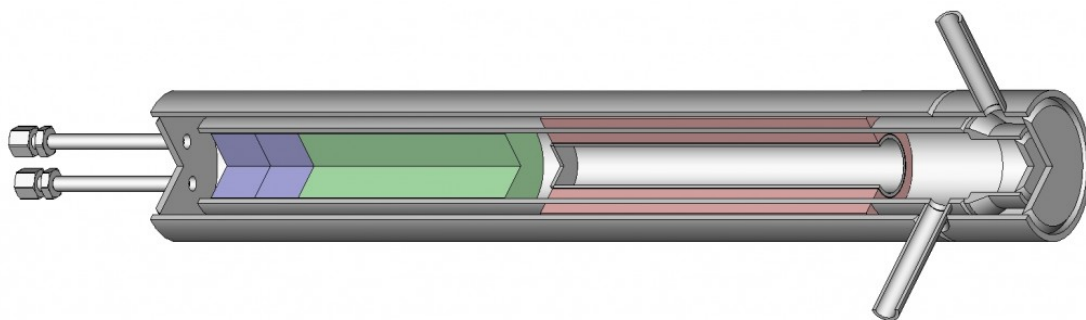
Figure 2: An autothermal reactor
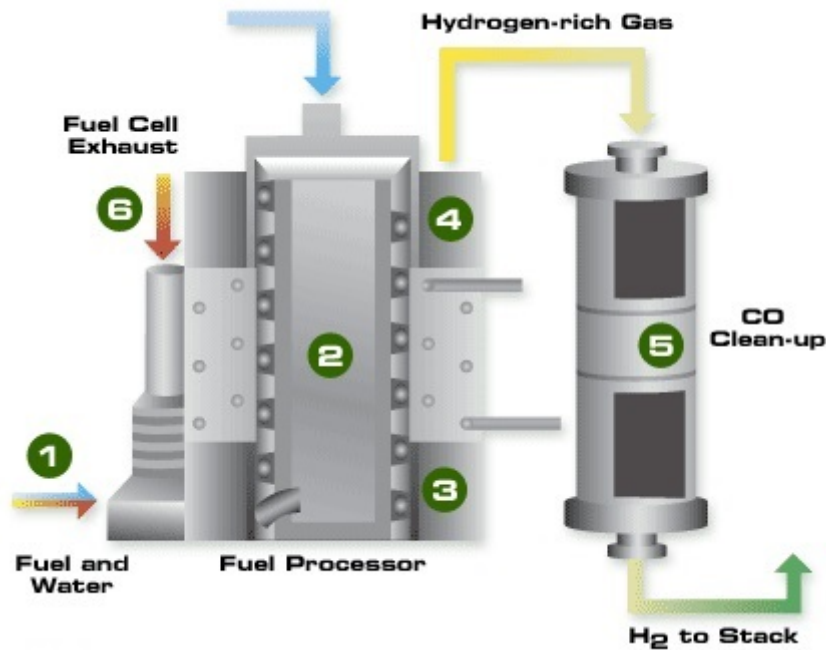
Figure 3: an overall process of the enriching process

Part B: Scientific Literature Review

While hydrogen storage remains a critical issue for stand alone applications of the technology, researchers are developing onboard fuel processors, which can convert a variety of fuels into hydrogen to power these fuel cell vehicles. In a recent work [1], the feasibility study of a 100 kW on board fuel processor based on gasoline fuel using process simulation was presented. The steady state model was developed with the help of Aspen HYSYS to analyze the fuel processor and total system performance. The components of the fuel processor are the fuel reforming unit, CO clean-up unit and auxiliary units. Optimization studies were carried out by analyzing the influence of various operating parameters such as oxygen to carbon ratio, steam to carbon ratio, temperature and pressure on the process results and parameters. From the steady state model optimization using Aspen HYSYS, an optimized reaction composition in terms of hydrogen production and carbon monoxide concentration corresponds to oxygen to carbon ratio of 0.5 and steam to carbon ratio of 0.5. The fuel processor efficiency of 95.98% was obtained under these optimized conditions. The heat integration of the system using the composite curve, grand composite curve and utility composite curve were studied for the system. The most appropriate heat exchanger network from the generated ones was chosen and that was incorporated into the optimized flow

sheet of the 100 kW fuel processor. A completely heat integrated 100 kW fuel processor flow sheet using gasoline as fuel was thus successfully simulated and optimized.

Fuel cell systems based on liquid fuels are particularly suitable for auxiliary power generation due to the high energy density of the fuel and its easy storage, in contrast to gaseous hydrogen storage. A 3 kW PEM fuel cell system based on diesel steam reforming to be applied as an APU for caravans and yachts was developed in a recent work [2]. The start-up time of a fuel cell APU is of the greatest importance since a buffer battery has to supply electric power until the system is ready to take over.

The start-up time directly affects the battery capacity and consequently the system size, weight, and cost. In the presented work a novel start-up strategy for the steam reforming fuel processor is introduced. This approach utilizes the reactive heating of WGS reactors by using reformate from oxidative steam reforming (OSR) instead of the sequential heating of the fuel processor. The start-up procedure is demonstrated on a 10 kW steam reformer and a parameter study is carried out. The new procedure was tested on the complete fuel processor. Until SR operation, the total energy consumption sums up to up to 5.9 MJ fuel and 13 Ah (12 V) electric energy.

Several post-reforming processors have been used to minimize both sulfur poisoning and carbon deposition on the anode catalyst. In a recent work, the integrated diesel fuel processor comprised an autothermal reformer, a desulfurizer, and a post-reformer [3]. The autothermal reforming section in the integrated diesel fuel processor effectively decomposes aromatics, and converts fuel into H_2-rich syngas. Desulfurizer removes sulfur-containing compounds present in the diesel reformate. Finally, the post-reformer completely removes the light hydrocarbons, which are carbon precursors, in the diesel reformate. The diesel reformer, desulfurizer, and post-reformer were tested as microreactors for about 2500 h in an integrated mode. Efficiency of the overall process was kept intact for at least 2000 hours, and light hydrocarbons and sulfur-containing compounds were completely removed from the diesel reformate.

The reforming of biomass-derived oxygenates such as ethanol or glycerol was the subject of a research work [4]. The stability of catalyst formulations developed for glycerol reforming had not been adequately covered in the literature up to that date. In the case of propylene glycol, few studies had been presented. The practical processing of larger amounts of polyalcohols such as propylene glycol to synthesis gas for fuel cell applications in reactors larger than laboratory scale suited for distributed applications had also not been adequately reported. This investigation [4] dealt with the development of stable catalyst formulations for propylene glycol reforming and reformer and fuel processor development for the steam reforming of propylene glycol in the power range exceeding 17 kW thermal of the synthesis gas product. A self developed stable catalyst formulation was incorporated into a plate heat-exchanger reformer. The energy required to drive the endothermic steam reforming reaction was foreseen to originate from the combustion of fuel cell anode off-gas and additional propylene glycol in a future technical process. Therefore the demonstrator was composed of layers for energy supply carrying a micro-structured catalytic burner, which was fed with anode off-gas surrogate and reaction layers which carried catalyst for the oxidative steam reforming of propylene glycol.

A novel concept of diesel fuel processing was proposed for the stable operation of solid oxide fuel cells (SOFCs) in a breakthrough work [5]. High temperature operation of SOFCs can lead to the capability of internal reforming with fuel flexibility. SOFCs can directly use CH_4 and CO as fuels given sufficient steam feeds due to catalytic reaction on the SOFC anode. However, heavier hydrocarbons than CH_4, such as ethylene, ethane, propane, etc., induce carbon deposition on the Ni-based anode of SOFCs. In the case of an ethylene steam reforming reaction on the Ni-based catalyst, the rate of carbon deposition is faster than it is when hydrocarbons, including aromatics, are used. Hence, the removal of light hydrocarbons (over C1-hydrocarbons), especially ethylene, with the reformate gases of heavy hydrocarbons (diesel, gasoline, kerosene, and JP-8) is important for stable operation of SOFCs. A novel approach, called the "post-reformer", was introduced for removing the light hydrocarbons (over C1-hydrocarbons) with the reformate gas stream. The CGO-Ru (3.0wt.%) catalyst was selected as the post-reforming catalyst because it shows high selectivity for removing light hydrocarbons (over C1-hydrocarbons) and achieving the high reforming efficiency. The diesel reformer and post-reformer are continuously operated for

about 200h in an integrated mode. The reforming performance did not degrade, and light hydrocarbons (over C1-hydrocarbons) in the diesel reformate were completely removed.

Microstructured reactors for water–gas shift and the preferential oxidation of carbon monoxide were developed for a fuel processing/fuel cell system running on iso-octane and designed for an electrical power output of 5 kW (el) [6]. The target application was an automotive auxiliary power unit (APU). The work covered both catalyst and reactor development. A platinum/ceria catalyst was applied for water–gas shift, while platinum on zeolite/alumina carrier served as catalyst for preferential oxidation. These catalysts were introduced into the final full size prototype reactors, which were constructed from microstructured stainless steel foils. Testing in a pilot scale test rig revealed conversion close to the thermodynamic equilibrium for the water–gas shift reactors at a WHSV range of $17–41$ N dm^3 h^{-1} gcat^{-1}.

The preferential oxidation, which was performed at higher WHSV in the range of $48–98$ N dm^3 h^{-1} gcat^{-1} revealed up to 90% conversion in a first stage, while in the second stage reactor the carbon monoxide content of the reformate was decreased to less than 50 ppm. The reactors were then incorporated into a complete bread-board fuel processor of 5 kWel power equivalent, which comprised also of an autothermal reformer reactor. The fuel processor was operated at a steam to carbon ratio of 3.3 and an oxygen to carbon ratio of 0.67. Under these conditions, it converted the iso-octane feed completely to purified reformate with an overall efficiency of 74%.

The development of a compact ethanol fuel processor for small scale high temperature polymer electrolyte membrane fuel cell (HT-PEMFC) systems with 200-500W electrical power output was reported [7]. Promising markets for reformer fuel cell systems based on ethanol are mobile or portable leisure and security power supply applications as well as small scale stationary off grid power supply and backup power. Main components of the fuel processor to be developed were the reformer reactor, the shift converter, a catalytic burner and heat exchangers. Development focused in particular on the homogeneous evaporation

of the liquid reactants ethanol and water for the reformer and burner and on the development of an efficient and autarkic start-up method, respectively. Theoretical as well as experimental work has been carried out for all main components separately including for example catalyst screening and evaporator performance tests in a first project period. Afterwards all components have been assembled to a complete fuel processor which has been qualified with various operation parameter set-ups. A theoretically defined basic operation point could practically be confirmed. The overall start-up time to receive reformate gas with appropriate quality to feed an HT-PEMFC ($x(CO) < 2\%$) takes around 30 min. At steady state operation the hydrogen power output is around 900 W with $H-2$ and CO fractions of 41.2% and 1.5%, respectively.

A simulative work analyzed the impact of fuel type on the energy efficiency of systems composed by a fuel processor for hydrogen production and a PEM (proton exchange membrane) fuel cell [8]. Two fuel processors were simulated; one employs steam reforming to produce hydrogen, the other one autothermal reforming. In both cases, fuel processing is completed by two water gas shift units and one preferential CO oxidation unit. Five classes of fuels were considered, i.e. alkanes, alkenes and alkynes, alcohols and aromatics and steam to carbon and oxygen to carbon inlet ratios, reforming temperature, fuel cell split fraction and exhaust gas temperature are explored as operative parameters. For each fuel considered, Aspen Plus (R) was used to calculate the operative conditions that maximize the energy efficiency of the systems. For each system, the data were employed to identify an analytic expression to calculate the best possible energy efficiency given the elementary composition of the fuel and its lower heating value. The expressions proved to hold true for a broad range of fuel types.

Microstructured reactors provide significant potential for the reduction of system size, system complexity and catalyst costs [9]. Scaled-up micro-channel reactors containing thousands of channels create less scale-up issues compared to conventional (fixed bed) systems. They allow a careful heat management during start-up and normal operation, which increases the service life of the catalyst. Complete fuel processors are under development applying steam reforming, partial oxidation and partial de-hydrogenation reactions on a large variety of fuels such as methanol, ethanol, natural gas, LPG, gasoline, kerosene and diesel. The systems size varies between 100 W and about 20 kW thermal power of the hydrogen produced. All development steps required for building a fuel processor had been addressed; reactor and system simulation and design, catalyst

development, catalyst stability against impurities in the fuel and set-up/testing of complete fuel processors for systems up to 13 kW thermal power output.

The choice of fuel converted to hydrogen mainly depends on the application and existing infrastructure. While trucks have diesel fuel readily available, LPG is the right choice for caravans. Having demonstrated the feasibility of compact fuel processing, the aspects of future mass production for such devices for consumer applications have been addressed. Complete fuel processor systems (e.g. a 5 kW diesel fuel processor, a 250W fuel processor/ fuel cell system running on LPG as fuel which will be commercialized end of 2010 and a 100 W methanol fuel processor operated with a high temperature PEM fuel cell) and novel components such as a reactor for the partial de-hydrogenation of kerosene were presented in detail.

The performances of four different auxiliary power unit (APU) schemes, based on a 5 kWe net proton exchange membrane fuel cell (PEM-FC) stack, were evaluated and compared in a comprehensive work [10]. The fuel processor section of each APU was characterized by a reformer (autothermal ATR or steam SR), a non-isothermal water gas shift (NI-WGS) reactor and a final syngas catalytic clean-up step: the CO preferential oxidation (PROX) reactor or the CO selective methanation (SMET) one. Furthermore, three hydrocarbon fuels, the most commonly found in service stations (gasoline, light diesel oil and natural gas) were considered as primary fuels. The comparison was carried out examining the results obtained by a series of steady-state system simulations in Aspen Plus® of the four different APU schemes by varying the fed fuel. From the calculated data, the performance of CO-PROX was not very different compared to that of the CO-SMET, but the performance of the SR based APUs was higher than the scheme of the ATR based APUs.

The most promising APU scheme with respect to an overall performance target was the scheme fed with natural gas and characterized by a fuel processor chain consisting of SR, NI-WGS and CO-SMET reactors. This processing reactors scheme together with the fuel cell section, notwithstanding having practically the same energy efficiency of the scheme with SR, NI-WGS and CO-PROX reactors, ensures a less complex scheme, higher hydrogen concentration in the syngas, lower air mass rate consumption, absence of nitrogen in the

syngas and higher potential power of the stack anode exhaust. The stack anode exhaust was recycled to the fuel processor section, thanks to the presence of methane produced in the final clean-up methanation reactor.

A novel partial oxidation (POX) catalyst and butanol-to-hydrogen fuel processor sub-system concept have been developed to fill the need for person-portable power systems with high energy density for extended duration operation [11]. The system is designed to reform isobutanol through partial oxidation with air. A multi-stage carbon monoxide removal train was developed to convert the CO present in the reformate stream. The resulting hydrogen stream is suitable for integration with a PEM fuel cell for person-portable power system applications. An integrated system design approach was undertaken to optimize the fuel processing system configuration and operating conditions.

Among the available fuel processing options, systems based on both partial oxidation and auto-thermal reforming (ATR) of isobutanol fuel were investigated. The former, i.e. partial oxidation reforming, results in a simpler but less efficient design to implement. The latter, i.e. autothermal reforming, is a more efficient approach for conversion of isobutanol to hydrogen for the fuel cell anode. A systematic design study has determined the optimal operating conditions for the partial oxidation and autothermal-based systems. Energy density, in terms of W-h/kg and W-h/L, is a critical parameter for person-portable power systems. These systems must be lightweight and compact, especially for military applications to avoid hindering a soldier's movements in the field. The butanol-fueled system under development has an inherent advantage due to the increased energy density of the fuel, as compared to methanol-fueled systems.

A complete, integrated sub-system was designed using 3-D CAD modeling software to minimize the size and weight of the reformer. 3-D CAD renderings were created for both the partial oxidation and autothermal-based butanol fuel processor systems. The POX-based butanol reforming system is estimated to achieve an energy density of 810 W-h/L and 850 W-h/kg, while the ATR-based system is estimated to achieve an energy density of 1,000 W-h/L and 2,000 W-h/kg. The concept feasibility of reforming isobutanol to a hydrogen-rich stream via partial oxidation was experimentally demonstrated. Novel catalyst formulations

and catalyst support structures have been developed and tested. A Rh/Ce-based reforming catalyst effectively converted isobutanol to an H_2-rich stream with no coke formation under the desired system operating conditions. The catalyst was also demonstrated to be regenerable, should carbon deposition occur.

An autothermally-reformed, gasoline-fueled automotive polymer electrolyte fuel cell (PEFC) system has been modeled and analyzed for the fuel processor and total system performance [12]. The purpose of the study was to identify the influence of various operating parameters on the system performance and to investigate related tradeoff scenarios. The effects of the following parameters were included in the analysis: operating pressure (3 and 1 atm), reforming temperature (1000-1300 K), water-to-fuel and air-to-fuel reactant feed ratios, electrochemical fuel utilization, and thermal integration of the fuel processor and the fuel cell stack subsystems. The analyses were also used to evaluate the impact of those parameters on the concentrations of methane and carbon monoxide in the processed reformate. Both of these gases can be reduced to low levels with adequate water-to-carbon used in the fuel processor. Since these two species represent corresponding amounts of hydrogen that would not be available for electrochemical oxidation in the fuel cell stack, it is important to maintain them at low levels.

Subject to the assumptions used in the analyses, particularly that of thermodynamic equilibrium, it was determined that reforming temperatures of 1100 K, a water-to-carbon mole ratio of 1.5-2.5, and the use of fuel cell exhaust energy in the fuel processor subsystem can yield fuel processor efficiencies of 82-84%, and total system efficiencies of 40-42% could be achieved. For the atmospheric pressure system, if the exhaust energy is not used in the fuel processor subsystem, the fuel processor efficiency would drop to 75-82% and the total system efficiency would drop below 40%. At higher reforming temperatures, say 1300 K, the fuel processor efficiency would decrease to 78%, and the total system efficiency would drop below 39%, even with the use of the fuel cell stack exhaust energy.

The performance of a metal-based solid oxide fuel cell (SOFC) coupled with an integrated diesel fuel processor, with a focus on operating stability was presented [13]. The reformate was produced by an autothermal reformer (ATR), desulfurizer, and post-reformer using commercial diesel. Diesel reformate possesses the characteristics of low fuel concentration

and high steam quantity due to its fuel processing condition for stable operation. These characteristics lead to high oxygen partial pressure at the anode which causes the oxidation of the cell's metallic component. Various approaches, such as thermodynamic calculation, temperature-programmed reduction (TPR) analysis, and the electrochemical performance of a single cell, were used to prove the oxidation phenomenon in this experiment. As a result, the key factors for stable operation were discovered, and a metal-based SOFC single stack with an area of 50-mm × 50-mm was successfully operated for 1000 h at a 4%/1000 h degradation rate under a modified condition of diesel reformate.

To ensure the SOFC's stable operation using the reformate, it is recommended that the operating condition should be considered from a viewpoint combining fuel composition, oxygen partial pressure, and temperature. The modified condition for the long-term operation of a metal-based SOFC using diesel reformate was also described.

The use of a solid-oxide fuel cell (SOFC) to provide auxiliary power for heavy duty trucks can increase fuel efficiency and reduce emissions by reducing engine idling time was discussed in a research work [14]. SOFC's that directly oxidize hydrocarbon fuels have lower power densities than do SOFC's that operate from hydrocarbon reformate, and since the SOFC is a costly component, maximizing the fuel cell power density provides benefits in reducing the overall APU system cost. Thus current SOFC APU systems require the reformation of higher hydrocarbons for the most efficient and cost effect fuel cell system. The objective of this research is to develop the technology to enable diesel reforming for SOFC truck APU applications. Diesel fuel can be reformed into a H_2 and CO-rich fuel feed stream for a SOFC

by autothermal reforming (ATR), a combination of catalytic partial oxidation (CPOx), and steam reforming (SR).

The typical autothermal reformer is an adiabatic, heterogeneous catalytic reactor and the challenges in its design, operation and durability on diesel fuel are manifold. These challenges begin with the vaporization and mixing of diesel fuel with air and steam where fuel pyrolysis can occur and improper mixing leads to hot and cold spots, which contribute to carbon formation and incomplete fuel conversion. The exotherm of the partial oxidation reaction can generate temperatures in excess of 800 ^{0}C, a temperature at which catalysts rapidly sinter, thus reducing their lifetime. The temperature rise can be reduced by the

steam reforming endotherm, but this requires the addition of water along with proper design to balance the kinetic rates. Carbon formation during operation and startup can lead to catalyst deactivation and fouling of downstream components, thus reducing durability of the fuel processor. Water addition helps to reduce carbon formation, but a key issue is the source of the water onboard a vehicle. Additionally, changes in diesel fuel composition, such as seasonal changes affect the reactor operation and design considerations.

Argonne National Laboratory is developing a fuel processor for converting liquid hydrocarbon fuels to a hydrogen-rich product suitable for a polymer electrolyte fuel cell stack [15]. The processor uses an autothermal reformer to convert the feed to a mixture of hydrogen, carbon dioxide, carbon monoxide and water with trace quantities of other components. The carbon monoxide in the product gas is then converted to carbon dioxide in water-gas shift and preferential oxidation reactors. Fuels that have been tested include standard and low-sulfur gasoline and diesel fuel, and Fischer-Tropsch fuels. Iso-octane and n-hexadecane were also examined as surrogates for gasoline and diesel, respectively.

Complete conversion of gasoline was achieved at 750 C in a micro reactor over a novel catalyst developed at Argonne. Diesel fuel was completely converted at 850 ^{0}C, over this same catalyst. Product streams contained greater than 60% hydrogen on a dry, nitrogen-

free basis with iso-octane, gasoline, and n-hexadecane. For a diesel fuel, product streams contained >50% hydrogen on a dry, nitrogen-free basis. The catalyst activity did not significantly decrease over >16 hours operation with the diesel fuel feed. Coke formation was not observed. The carbon monoxide fraction of the product gas could be reduced to as low as 1% on a dry, nitrogen-free basis when the water-gas shift reactors were used in tandem with the reformer.

Fuel cell requires hydrogen as its fuel source for generating power. Hydrogen for use in auxiliary power units is produced in a fuel processor by the catalytic reforming of hydrocarbons. Diesel, jet fuel, gasoline, as well as natural gas, are potential fuels that all have existing infrastructure of manufacture and distribution, for hydrogen production for fuel cell applications. It is well known that essentially all hydrocarbon feeds contain sulfur at different concentrations. In addition to coking, sulfur poisoning is the main force for

deativation of pre-reforming and reforming catalysts. A research work aimed to develop, test and characterize efficient catalysts for hydrogen generation from diesel autothermal reforming [16]. Bimetallic catalysts exhibited superior performance compared to the commercial catalyst and the monometallic counterparts. BET, TPD, TPR, and XPS were utilized for surface analysis of these formulations, which showed that the enhanced stability is due to a strong metal–metal and metal–support interaction in the catalyst.

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