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Non-thermal gliding-arc plasma reforming of dodecane and hydroprocessed renewable diesel

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ABSTRACT

This paper focuses on reforming dodecane and hydroprocessed renewable diesel to hydrogen rich gas in a non-thermal gliding-arc plasma stabilized in a reverse vortex flow reformer. The liquid fuels were directly injected into the reaction chamber using an ultrasonic nozzle and entrained in the reverse vortex flow before passing through the plasma. Initial parametric tests were used to investigate the individual effects of varying power input, steam to carbon ratio, and equivalence ratio on reformer performance. Subsequent factorial tests varied these parameters to identify optimal specific energy requirements. Optimal reforming conditions for dodecane, a model diesel compound, resulted in specific energy requirements of 134.1 ± 1.1 kJ mol^{-1} H_2 produced, a H_2 yield of 65.0 ± 0.02%, and an efficiency of 37.0 ± 0.02%. Optimal conditions for hydroprocessed renewable diesel resulted in a specific energy requirement of 176.1 ± 3.8 kJ mol^{-1} H_2 produced, a H_2 yield of 64.2 ± 1.7%, and an efficiency of 35.0 ± 1.0% at 95% confidence intervals. Physical operating boundaries due to arc extinction were identified.
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Introduction

Recently, the alternative energy sector has experienced rapid growth because of increasing pressure from climate change awareness, rising fuel costs, and a need for domestic energy security [1,2]. New technologies have focused on producing energy that is accessible, environmental friendly, sustainable, secure, and can meet current and future projected energy needs [3]. Hydrogen is expected to play a large role in the energy economy of the future as it can be utilized in fuel cell applications, and in the synthesis of alternative fuels [1,3,4]. This paper explores the use of a non-thermal reverse vortex flow (RVF) gliding-arc reformer for liquid fuels. Tests were conducted using dodecane as a model diesel compound and hydroprocessed renewable diesel fuel. Parametric tests determined the effects of various system parameters, while factorial tests were utilized for system optimization.

Current fuel reforming technologies

Hydrogen is expected to be a prominent fuel in the future [1,3,4]. However current production methods are expensive, require complex and large machinery or costly catalysts, and require an expensive distribution infrastructure [1,3,5]. Steam, partial oxidation, and autothermal reforming constitute the major reforming technologies [2,3]. Hydrogen production via steam reforming of natural gas utilizes roughly one third of

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the fuel to support the parasitic energy requirement of the process. Ultimately this leads to an specific energy requirement (SER) of 325–354 kJ mol^{-1} of hydrogen produced [6,7].

Since the late 90's, interest in plasma reforming has grown [8]. Plasma reformers can operate in thermal equilibrium or non-thermal equilibrium. Thermal plasma reformers operate at high temperatures (5000–10,000 K), require power inputs of 1 kW–50 MW, require extensive measures to prevent electrode wear [9,10], and are most commonly used for metallurgical applications [11].

Non-thermal reformers can utilize various plasma forms such as glow discharge, corona discharge, silent discharge, dielectric barrier discharge, gliding arc, etc. Non-thermal plasma can operate near room temperature, at lower power inputs than thermal plasmas, and at or near atmospheric pressure [10]. They commonly have a much smaller physical foot print than traditional reforming technologies and can be used in onboard applications [12], all while maintaining high energy densities required for hydrocarbon reforming [13]. Within a non-thermal plasma, electrons exist at high temperatures, ranging from 10,000 to 100,000 K [2], while the protons and neutrons remain near room temperature [10,13]. Intermediate, 'warm' plasmas, however, with gas temperatures ranging from 2000 to 4000 K can also be utilized for controlled gas reforming [11]. Petitpas et al. produced an extensive comparison of plasma technologies highlighting various reformer efficiencies, fuel conversion rates, and energy requirements [2]. Gutsol et al.'s review of reforming technologies concluded that hydrogen must be produced for less than 109 kJ mol^{-1} [6] to be economically feasible. Many technologies are approaching this point [2].

Adaptation to liquid fuels

Previous work explored parameters affecting the performance and optimization of non-thermal, reverse vortex flow, stabilized gliding-arc plasma reforming of methane [14]. This paper explores the performance and optimization of the same system adapted for the reforming of selected liquid fuels. Liquid fuels can be distributed via existing fuel infrastructure and then reformed to produce hydrogen on site.

The two fuels of particular interest for ship board applications are marine diesel (F-76) and hydroprocessed renewable diesel (HRD-76). Dodecane was chosen as a model liquid diesel compound in the development of a liquid fuel injection system. HRD-76 was the primary biofuel used in the recent Great Green Fleet exercises [15]. Reforming F-76, HRD-76, or their blends, could allow the Navy to transport a liquid fuel that could be reformed into hydrogen rich gas for shipboard applications.

Materials and methods

Fig. 1 shows the test bed for RVF non-thermal, gliding-arc plasma reforming of liquid fuels. The system used for methane reforming previously described [14] was modified to accommodate liquid fuels. Changes to the three major component groups, reactant delivery, reformer and power supply, and product conditioning, are highlighted below.

Reactant delivery

A piston pump (SmoothFlow Pump 100 (LF), Microfluidica, Glendale, WI) provided precise fuel flow control to a nozzle injection system. The fuel is atomized upon injection at the top of the reaction chamber by an ultrasonic nozzle system powered by a digital signal generator (model HT40K50T, Sonaer Ultrasonics, Farmingdale, NY). The system atomized liquid fuel into 25 μm diameter droplets using 1.15–2.25 W power input over a range of fuel flow rates from 0.44 to 1.65 ml min^{-1}. Droplet sizes were measured using a phase-Doppler particle sizing system (Aerometrics, Sunnyvale, CA). Increasing power to the nozzle tip decreased the mean droplet diameter while increasing axial and radial droplet velocity, and resulted in liquid accumulation on the reactor walls and/or excessive unreacted fuel directly exiting the reactor. Particles with 25 μm diameter were entrained within the interior vortex of the reverse vortex flow, and were transported along the flow lines through the plasma arc.

Reformer and power supply

The power supply for the reactor underwent minor changes when the reformer was adapted for use with liquid fuels. The only change to the power supply section was the substitution of an updated variable transformer (Model 3PN1510B-DVAM, ISE, Inc., Cleveland, OH).

Fig. 2 shows the updated reformer, modified for the introduction of liquid fuels into the reaction chamber. A non-conducting, top electrode mount made from zirconia ceramic (AmZirOx 86, Astro Met Zirconium Oxide, Cincinnati, OH) electrically insulated the ultrasonic nozzle from the upper electrode. AmZirOx 86 zirconia ceramic was chosen due to its electrical resistivity, low thermal expansion, and chemical inertness. Removing the upper electrode mount from the Macor top plate allowed radial insertion of the upper electrode into the reaction chamber. The 42 mm ID quartz tube reactor body is sealed at all mounting points with silicone O-rings.

The stainless steel tangential inlet plate used in the earlier study was replaced by one made of zirconia (AmZirOx 86, Astro Met Zirconium Oxide, Cincinnati, OH) to avoid electrical discharge between it and the top electrode. The bottom electrode lead was inserted through a pilot hole on the side of the plate (not shown in Fig. 2) and made radial contact with the Inconel bottom electrode. All gas ports were 316 stainless steel Swagelok fittings inserted into the ceramic with pipe thread connections.

The ultrasonic nozzle was attached to an aluminum mounting plate fixed to the Macor™ top plate with PTFE threaded rods and sealed with a Kalrez® o-ring.

A funnel shape was added to the bottom of the quartz post reaction chamber to drain any unreformed liquids out of the reactor. A high temperature three-way valve (model MPT100-SS-T-SSS-N-15, Control Resources, Salisbury NC) at the reactor exit directed the product stream into the reformate conditioning and analysis section or into a bypass to exhaust during non-steady state periods. This allowed the system to come to steady state without contaminating the impinger train used for liquid analysis.

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