

Hydrogen from reformer gas a novel fuel and bridging technology: A combustion perspective

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Abstract

Constant-volume combustion experiments measuring laminar burning velocity are presented for combinations of methane, inert diluent and H₂/CO mixtures that would result from steam reforming of methane. The experiments illustrate the very attractive prospects of on-board steam reforming in natural gas powered vehicles that would employ exhaust gas recirculation (EGR) to improve combustion performance and reduce NO_x emissions. Laminar burning velocity in the partially reformed fuel stream can be maintained at levels similar to that of air/natural gas mixtures by increasing the prereforming of the fuel at increasing concentrations of EGR. Up to 40% dilution was tested, requiring that 53% of the methane fuel be reformed to maintain burning velocity. Calculations indicate NO_x levels are similar to scenarios with unreformed fuel.

The knowledge base from this and similar experiments is required to allow for the future adoption of on-board fuel reforming in IC engines. This is a critical intermediate step in introducing hydrogen as a fuel to the currently fossil-hydrocarbon oriented economy and fuel delivery infrastructure.

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1. Introduction

Practical difficulties of storage and transportation of hydrogen makes the use of on-board vehicular fuel reformers a very attractive alternative to systems which would employ hydrogen as a fuel source. Furthermore, fuel reforming for vehicles allows for a multifuel capability from fossil hydrocarbons or non-fossil biofuels, thus creating a gradual bridge for possibly phasing out fossil fuels without requiring the abrupt adoption of radically different technologies compared to systems on the road today. Partial reforming of fuels with poor evaporation characteristics (such as ethanol) makes new fuels more feasible in spark ignition engines, particularly in cold climates.

In this study, a combustion scenario is investigated for a methane fuelled spark ignition engine which uses exhaust gas recirculation (EGR) to reduce emissions in addition to

on-board steam reforming of the methane to hydrogen and carbon monoxide to counteract the drop in flame speed caused by using EGR dilution alone. The result is an overall reduction in emissions of oxides of nitrogen while illustrating a feasible scenario for phasing in more greenhouse friendly fuels to the existing market.

2. Background

The laminar burning velocity is a fundamental parameter to understand the combustion process and flame propagation. It is also a necessary input to model the turbulent combustion and pollutant formation. Precise values of laminar burning velocities are useful for practical application on internal combustion engines, which operate at pressure varying from 5 to 60 atm and unburnt gas temperature range from 500 to 1100 K [1]. Studies of burning velocities and flammability limits with some fuels with reformer gas addition are available [2–4], though data of burning velocities of methane/EGR/reformer gas mixtures are scarce, especially at elevated temperature and pressure to simulate the real engine operation. Furthermore, an interesting

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question remains whether the methane/EGR/reformer gas mixture loses the potential of low NO emission while maintaining the undiluted burning velocity.

Methane, a major component of natural gas, was used in this study for experimental practicality and potential application in natural gas engines. The high octane rating, low level of non-HC impurities and low carbon/hydrogen ratio of methane provide the natural gas engine fundamental emission advantages [5]. However, the natural gas engine has relatively high emission on oxides of nitrogen [6–8]. Thus, EGR is essential for a methane-fuelled engine. Unfortunately, the methane-fuelled engine has the poorest EGR tolerance compared with gasoline and methanol fuelled engines [9], with a maximum EGR tolerance 8% [6]. Clearly, reformer gas addition is necessary to broaden the EGR tolerance and improve the combustion stability.

Heimel [10] measured the burning velocity of hydrogen/air mixtures at elevated temperature, and Iijima and Takeno [11] measured that at elevated pressure, thus showing how hydrogen's high flame speed has potential for increasing flame speeds with H₂ in the mixture. Work by McLean et al. [12] confirmed this, thus indicating that reformer gas has potential to enhance the combustion of methane/diluent mixtures by its high burning velocity.

Houseman and Cerini demonstrated the technical feasibility of on-board steam reforming of gasoline or methanol to generate hydrogen for automobiles [13]. Steam reforming of methane has been developed and a catalyst, such as Ni-YSZ, was strongly recommended to reduce the reaction temperature [14,15]. An improved novel membrane reformer [16] was able to generate a large volume of hydrogen by steam reforming of methane at relative low temperature. Jamal et al. [17] concluded that steam reforming generated the maximum quantity of hydrogen and no nitrogen was involved. On the other hand, partial oxidation produced less hydrogen than steam reforming despite an exothermic reaction. For example, the products of partial oxidation of gasoline were only composed of a quarter of hydrogen, a quarter of carbon monoxide and half of nitrogen [2,4]. Meanwhile, additional CO was slightly effective in improving combustion efficiency, emissions and stability [2]. Steam reforming is able to produce the maximum 75% hydrogen ideally and it also has the economic benefits, a comparatively low costs and relatively high efficiency of hydrogen production [18].

3. Method and apparatus

A preheated cylindrical combustion chamber was used to measure the laminar burning velocity of methane/diluent/H₂ – CO mixtures for a range of pressures and temperatures. The equivalence ratios of all experimental mixtures were unity, to match the operational needs of catalytic converters. The EGR diluent was simulated by CO₂ and N₂ and reformer gas was simulated by H₂ and CO based on the assumed steam reformation of methane:

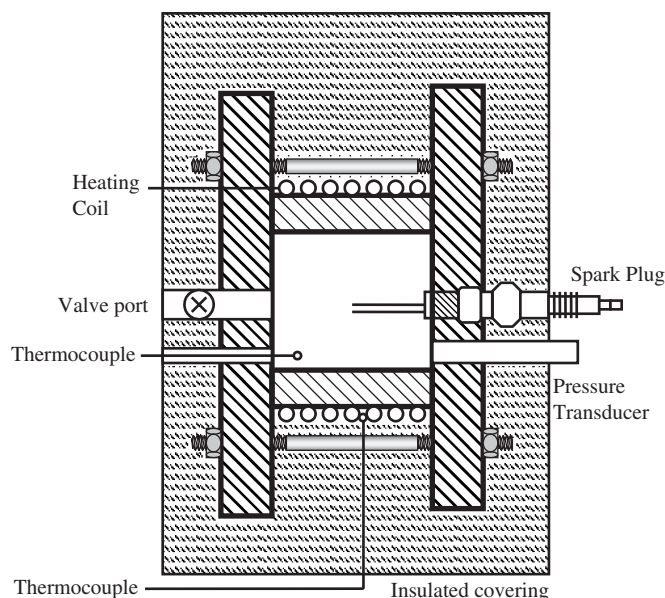


Fig. 1. Experimental apparatus (right) showing combustion cell and instrumentation.

A preheated high pressure cylinder combustion chamber with central spark ignition [19] was used (Fig. 1). The chamber volume was 1.1 L with 114 mm equal diameter and length. The heavily insulated chamber was wrapped in electric heating elements to facilitate a range of initial temperature from 298 to 473 K and initial pressure from 1 to 5 atm. An extended-electrode spark plug was located along the axis of the chamber and insulated by heat shrink tubing to ensure the central spark.

EGR diluent was simulated by 18.5% CO₂ and 81.5% N₂ and the stoichiometric reformer gas mixture was simulated by 22.1% H₂, 7.4% CO, and 70.5% air. The stoichiometric methane/air mixture consisted of 9.5% methane and 90.5% air. The diluent composition was slightly different with that used by others ($\frac{14}{86}$ [20] or $\frac{15}{85}$ [21]), which only simulated the “dry” exhaust gas and ignored the significant water content of real exhaust gas.

A previously described [22–24] multizone thermodynamic equilibrium model was used to calculate the laminar burning velocity from the pressure trace. The cylinder gas was discretized into 1500 equal elements. For each element, energy balance and equilibrium calculations were performed. In order to minimize the stretch and ignition effects, the measured burning velocities from flame radius 45–57 mm were fitted to the model and then extrapolated to initial experimental pressure to obtain the burning velocity [25].

Since the measurement difference between NO and NO_x was negligible in spark ignition engines which burn near-stoichiometric mixture [26], only nitric oxide was simulated here. Ignoring trace fuel bound nitrogen, the NO production was predicted with the extended Zeldovich mechanism, the primary mechanism that governs NO formation in combustion of near stoichiometric fuel–air mixture [26].

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