



Ultra-low NO_x emissions from catalytic hydrogen combustion

B. Fumey^{a,*}, T. Buetler^b, U.F. Vogt^{c,d}



^a Empa, Swiss Federal Laboratory for Materials Science and Technology, Urban Energy Systems, Ueberlandstrasse 129, 8600 Duebendorf, Switzerland

^b Empa, Swiss Federal Laboratory for Materials Science and Technology, Automotive Powertrain Technologies, Ueberlandstrasse 129, 8600 Duebendorf, Switzerland

^c Empa, Swiss Federal Laboratory for Materials Science and Technology, Materials for Energy Conversion, Ueberlandstrasse 129, 8600 Duebendorf, Switzerland

^d University of Freiburg, Crystallography, Hermann-Herder-Str. 5, D-79104 Freiburg i.Br, Germany

HIGHLIGHTS

- Flue gas analysis of catalytic oxidation of renewable hydrogen.
- Commercial Pt catalyst on highly porous SiC foam plates, design ready for up scaling.
- Stable operation with lean and very lean fuel mixture.
- Ultra low NO_x release, between 0.004 and 0.12 mg/kWh.
- Non premixing fuel and air supply.

ARTICLE INFO

Keywords:

Hydrogen
Catalytic combustion
Ultra-low NO_x
Non premixing
Domestic application

ABSTRACT

The objective of this work is to determine the nitrogen oxide emission in the flue gas of a catalytic hydrogen combustion process, operating without premixed hydrogen and air supply. The study was investigated on a novel designed gas under glass stove top burner, suitable for domestic kitchen applications. The basic catalytic burner assembly consists of two platinum coated silicon carbide (SiC) foam disks with a diameter of 150 mm, a thickness of 10 mm and a porosity of 60 and 80 pores per inch (ppi) respectively. The two catalytic SiC disks are stacked with 10 mm space between for a uniform air feeding and distribution. Hydrogen is supplied from below the assembly and air is blown in between the two Pt coated catalytic SiC disks, leading to a homogeneous air distribution and thus a uniform catalytic reaction of hydrogen and air. Tests are performed at hydrogen flow rates of 5, 10 and 15 NI/min, equivalent to 0.9, 1.8, 2.7 kW power, the hydrogen to oxygen ratios (φ) were fixed to 0.66, 0.5 and 0.33 respectively. Ultra-low nitrogen oxide emissions of 0.09 ppmv to 9.49 ppmv, equivalent to 0.007 to 0.37 mg/kWh are achieved with this novel developed catalytic combustion design. These values are significantly lower than the present EU regulation of 56 mg/kWh for combustion processes of gaseous fuels for heating applications. This result shows the very high potential of converting hydrogen to heat without harmful exhaust gases for a broad domestic application in decarbonised gas grids or stationary power to gas applications.

1. Introduction

Hydrogen (H₂) as a renewable energy carrier is globally promoted for renewable energy utilisation through storage, effecting considerable reduction of greenhouse gas emissions [1]. Due to this purpose, decarbonising gas supplies by delivering H₂, reform gas or H₂ enriched natural gas to homes is considered [2,3]. It is recognised, that replacing natural gas with hydrogen as fuel for building space heating, cooking and water heating, has the advantage of releasing only water vapour without any other harmful exhaust gases [4], like carbon dioxide (CO₂), carbon monoxide (CO), unburned hydrocarbons or soot. However, previous investigations have shown that open flame hydrogen burners

without catalytic combustion produce substantially more nitrogen oxides (NO_x) than equivalent natural gas burners, due to their high combustion temperature [5,6]. This is an important concern since nitrogen oxide (NO) and nitrogen dioxide (NO₂) are notable strong greenhouse gases. NO reacts in air to form NO₂ which in turn reacts with oxygen (O₂), catalysed by ultra-violet sun radiation, to form tropospheric ozone (O₃), a primary constitution of smog and again NO, thus forming a multiple cycle process. NO and NO₂ further forms nitric acid together with moisture from the air, resulting in acid rain and leading to damage of the ecosystem. Likewise, NO₂ is highly toxic for humans, effecting both lung and heart functions, wherefore NO_x emissions are generally subject to strict regulations [7].

* Corresponding author.

E-mail address: benjamin.fumey@empa.ch (B. Fumey).

The three processes of NO_x formation from fuel combustion are, fuel based, thermal and prompt. Fuel based NO_x results from fuel bound nitrogen, generally found in coal or crude oil. Thermal NO_x is formed by the reaction of atmospheric nitrogen (N₂) and O₂ molecules in the flue gas at elevated temperature [8] and prompt NO_x results from the reaction of atmospheric N₂ with fuel fragments from carbon based fuels [9].

The combustion of hydrocarbon-based fuels can lead to the formation of all three types of NO_x, while the combustion of hydrogen, due to the absence of nitrogen, omits fuel based NO_x and the lack of carbon (C) prevents formation of prompt NO_x. Nevertheless, since thermal NO_x is fuel independent, it may be generated [6]. The rate of thermal NO_x formation depends on the combustion temperature, the fuel to air ratio as well as the residence time of the flue gas in the high temperature zone [10,11]. The principal reactions governing the formation of thermal NO_x, are:



Hereby, Eq. (1) is reaction rate limiting due to the very strong triple bond of the nitrogen molecule. Since the open H₂ flame can rise to temperatures of more than 2100 °C the required activation energy of 315 kJ/mol is reached and substantial formation of NO_x occurs.

The three general means to reduce thermal based NO_x are to reduce the combustion temperature, to decrease the flue gas residence time in the high temperature zone and to reduce the excess O₂ in the fuel to air mixture [12]. The most effective of these measures is the reduction of the combustion temperature.

Gaseous fuels are generally combusted through flame oxidation, flame less oxidation [13], catalytic oxidation [14] or hybrid oxidation, which consists partly of catalytic and partly flame oxidation [15,16], whereby catalytic oxidation generally enables the lowest combustion temperatures. In respect to fuel to oxidant ratio, distinction can be made between operation with rich fuel mixtures and lean fuel mixtures [17,18] as well as rich-lean fuel mixtures, where air is added in several steps during combustion [19]. Fuel rich combustion benefits from low O₂ content in the flue gas, thus potentially reducing NO_x formation. Fuel lean oxidation, on the other hand, reduces combustion temperature through cooling from excess air and thus also shows potential for NO_x reduction. Fuel and air can be supplied to the combustion zone either pre-mixed or non pre-mixed [17]. In this work catalytic oxidation of non pre-mixed lean fuel combustion is investigated.

In respect to catalytic oxidation of H₂, numerous studies have been performed. Ikea et al. [20] has investigated oxidation of H₂/air mixtures on platinum (Pt) plates to draw inferences concerning surface reaction mechanisms and rate parameters [21]. Appel et al. [22] performed similar work by the validation of various chemical reaction schemes in the catalytically stabilised combustion of H₂/air mixtures over Pt. Schefer et al. [23] investigated the combustion characteristics of lean H₂/air mixtures, flowing over a heated catalytic Pt plate. The objective of his work was to gain a better understanding of the interaction between fluid mechanic gas phase combustion and surface reaction.

Oxidation of pure H₂ and H₂ enriched fuels is studied on all power levels, from micro-reactors to large gas turbine machines. Micro-reactors for small portable power generation devices, fuelled with hydrogen [24] or hydrogen enriched hydrocarbons and syngas mixtures, [25] have been intensively studied experimentally and numerically during the last years. In general, gases are pre-mixed in micro-combustors and catalytic oxidation is involved. On a large scale, diffusion burners have been suggested in order to reach stable flame combustion of pure H₂ and H₂ enriched fuels [26]. Nevertheless, their generally high combustion temperature leads to excessive thermal NO_x formation. An alternative approach is found in lean premixed fuel combustion

in porous media, showing less NO_x emissions than purely flame combustion [27]. In this approach, it is found that a decrease in H₂ to air ratio reduces the NO_x generation [28]. Carroni and Griffin proposed a catalytically stabilised combustion, whereby part of the fuel is converted in a catalytic reactor and the remaining fuel is combusted in a subsequent gas phase burner [29]. In this approach, the catalyst is found to prevent flash back, an important feature in H₂ combustion due to its high flame speed [30,31].

In domestic use of hydrogen, low-temperature flameless catalytic combustion appears to be an effective approach due to the fact, that H₂ is much more readily catalytically oxidised than any other fuel [14]. Application specific required temperatures are well below 100 °C for space heating and domestic hot water heating and below 700 °C for cooking applications. Haruta et al. studied the utilisation of H₂ for this purpose, focusing on H₂/air pre-mixing [14,32]. It was found that catalytic combustion of H₂ has the big advantage of simple temperature adjustment, especially at respectively low oxidation temperatures. However, non-uniform temperature distribution over the catalyst surface can be an issue, requiring special attention in respect to design features [33].

Pangborn et al. have designed catalytic burners for domestic application, whereby H₂ and air was premixed and care was taken not to exceed the self-ignition temperature in order to prevent flame flashback [34,35]. Carlos and Amell studied the performance of a ceramic porous media burner, operating in a surface combustion mode for home appliance [36]. Dahm [37] proposed a non pre-mixed diffusive burner with passive air supply, and Gromann et al. [38] developed a non-stationary H₂ cooker with finger like porous H₂ distributors. It is found that burners with passive air supply have a low area specific power ratio due to the limited oxygen disposability.

In literature, a call for practical experiences in catalytic oxidation of hydrogen is postulated [39]. Zhen et al. pointed to the lack of work done on small-scale hydrogen burners and low temperature air combustion for practical application [40]. Ekins recognised uncertainties concerning the risks associated with H₂ as a consumer fuel in buildings [41]. Altfelt and Pinchback emphasised the need of operation data for adjusted and new H₂ fuel based domestic appliances [42].

In this work the flue gas analysis in respect to NO_x formation of a novel catalytic diffusion burner, designed for domestic application is presented. A new approach of catalytic diffusion oxidation on Pt coated porous SiC foams with non-premixing H₂ and air supply is followed. Air is forced by an air pump system, allowing a high area specific power ratio at lean fuel conditions.

2. Experiment

2.1. Burner design

The stove top burner consists of 4 highly porous silicon carbide (SiC) ceramic foam disks. These are installed in a step type round stainless steel casing as shown in Fig. 1. The steps function as support for the porous SiC ceramic disks and the increase in diameter guarantees a uniform H₂ contact to the catalytic Pt coated surface of the SiC foams. Fig. 1a shows the H₂ supply from the bottom centre of the burner casing with an open space for initial hydrogen distribution. A diffuser SiC disk, labelled in the figure as disk 1, with a porosity of 100 pores per inch (ppi) covers this space and leads to a homogeneous H₂ distribution (Fig. 1b). The follow up disk 2 is a Pt coated catalytic combustion SiC disk with a porosity of 80 ppi, (Fig. 1c), forming the primary combustion area. Fig. 1d shows the installed perforated air supply tubes, for air distribution perpendicular to the hydrogen inlet flow. To improve distribution of air and hydrogen, a secondary distributor disk 3, with a porosity of 40 ppi is placed over the air supply tubes (Fig. 1e) followed by the second and final Pt coated catalytic active disk with 60 ppi, as shown in Fig. 1f and labelled as disk 4. Disks 1 and 3 serve to reach a homogeneous distribution of both hydrogen and air, this is an

Download English Version:

<https://daneshyari.com/en/article/6680799>

Download Persian Version:

<https://daneshyari.com/article/6680799>

[Daneshyari.com](https://daneshyari.com)