Field of Solar Hydrogen
Heat No. 13

Published in International Journal of Hydrogen Energy, Vol.19, No. 4, 1994

Test of a premixing radiant burner for the low NO_x combustion of natural gas / hydrogen mixtures

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Abstract

The subject of this work is the development and testing of premixing radiant burners operated either with natural gas or natural gas containing different hydrogen fractions. Emphasis is first placed on establishing a stable combustion process by varying the fuel gas composition, air ratio and burner load; furthermore, emissions should be as low as possible.

With respect to flashback-free operation, it is important to define the limits of hydrogen addition for the burner concept developed.

Nomenclature

m	Hydrogen ratio (mole fraction of hydrogen in fuel gas)
q	Power density (heat quantity relative to the outer surface of the fibrous structure)
CO	Carbon monoxide
CO_2	Carbon dioxide
H_2	Hydrogen
NO_x	Nitrogen oxide
O_2	Oxygen
UHC	Unburned hydrocarbons
λ	Air ratio

1. Introduction

The discussion of energy policy is increasingly focussing on environmental pollution caused by fossil energy carriers and on protecting the Earth's atmosphere from threatening changes. Concepts of economical energy use, efficient conversion of chemically bound energy into electricity or heat and the application of energy carriers free from carbon dioxide must be seen against this background. The influence of commercial fuels and hydrogen on specific carbon dioxide (CO_2) release is shown in *Fig. 1* where calculated CO_2 emissions are plotted for certain solid, liquid and gaseous fuels. The calculation only includes the CO_2 values of the input fuels due to combustion. Further CO_2 quantities arising e.g. from production, transport, preparation and distribution, have not been taken into consideration. The highest specific CO_2 emission of 350 g kWh^{-1} is found for lignite. Hard coal combustion involves only insignificantly lower CO_2 emissions amounting to approximately 330 g kWh^{-1}. Data on the composition of the coals are taken from Ref. [1]. Fuel oil EL (see specification in Ref. [2]) is already much more favourable, but inferior to methanol, if the processes preceeding combustion are neglected, as mentioned above [3]. The use of propane and especially natural gas [4] leads to significantly lower CO_2 values. If hydrogen is added to natural gas, then the CO_2 emissions initially only decrease gradually, and then more rapidly with increasing hydrogen ratio, reaching zero if m = 1. The ratio m represents the hydrogen mole fraction in the fuel gas. The hydrogen required for this purpose can be produced in electrolysis plants, for example, which may be operated by solar power or photovoltaics and also by electricity from wind converters and nuclear power plants [5]. For reasons of availability, it can only be gradually introduced into the heat market. The hydrogen fraction of existing natural gas networks could be gradually raised to a value which does not affect the safe operation of existing conventional burners. Exceeding this limit in mixed operation will affect the flame stability and operational safety. Consequently, these systems would have to be converted to cope with higher hydrogen fractions. Burners in which the flame stability plays a much less significant role are the so-called radiant burners. They are characterized by the fact, that a large portion of the heat produced is transferred by thermal radiation. The conversion of the fuel/air mixture takes place on the surface of porous bodies [6]. Extensive studies have been carried out, especially in the U.S.A. on the development of gas-fuelled radiant burners. The Institute of Energy Process Engineering has developed the catalytic burner on the basis of these system studies [7]. Work was initiated by the alcohol programme which, in addition to the production of energy alcohols, especially methanol, was also concerned with their utilization. Combustion experiments were carried out using modified conventional, as well as catalytic, burners. In the field of catalytic combustion, the programme was extended last year to also cover gaseous fuels, first of all natural gas H. At the request of Solar-Wasserstoff-Bayern GmbH (SWB) an offer was submitted for the delivery of a catalytic heating appliance for the solar plant at Neunburg vorm Wald. This device is to be operated at a maximum thermal load of 10 kW using natural gas H as well as a mixture of natural gas H and hydrogen and air as the oxidant. In view of the narrow time frame, a test rig was already converted for operation with the required fuel gas quantities and a larger load range prior to submitting the offer. Initial tests without a catalyst were carried out. They will be presented in the following.

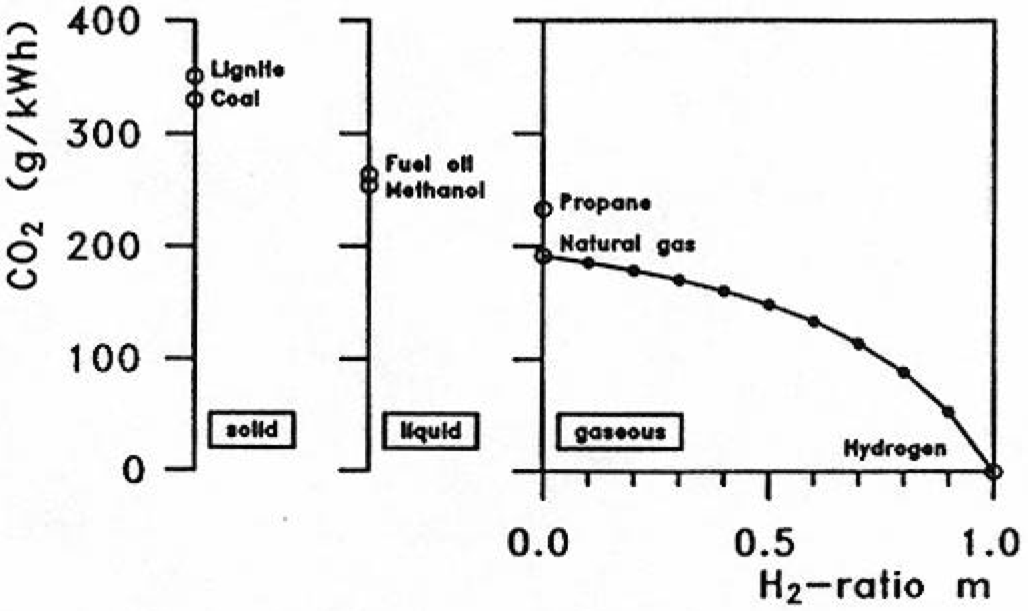

Fig. 1. Specific CO_2 emissions of different fuels.

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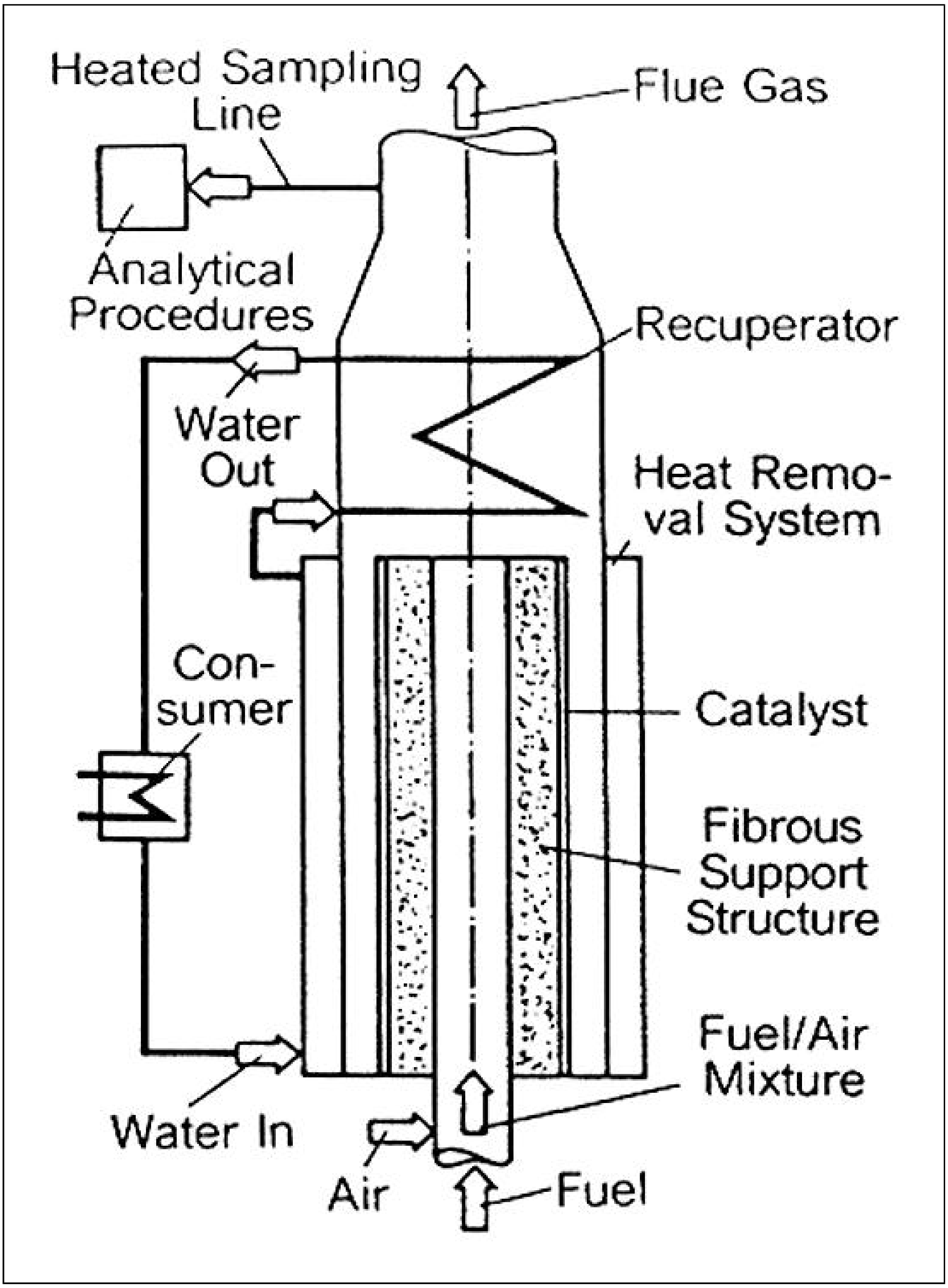

Fig. 2. Radiant burner

2. Description of the experimental facility

Figure 2 shows the main components of the test rig. The two fuel gases, natural gas H and hydrogen, are combined in a header and added to the combustion air in a mixer. The fuel gas/air mixture then flows in the radial direction through a cylindrical fibrous structure with a length of about 300 mm, an outer diameter of 100 mm and an inside diameter of 40 mm. The reaction zone is formed on the outer surface of the ceramic cylinder after spark ignition of the mixture. Apart from uniformly distributing the mixture, the fibrous body also has the task of separating the reaction zone from the incoming mixture and conducting as little heat as possible into the interior in the counter-flow direction due to its low thermal conductivity. A high proportion of the combustion heat is transferred by radiation to a heat exchanger arranged around the fibrous body. A further proportion is extracted from the flue-gas by a heat exchanger provided in the stack. The off-gas analysis system indicated permits the continuous determination of oxygen (O_2), carbon monoxide and carbon dioxide (CO, CO_2), nitrogen oxide (NO_x) and unburned hydrocarbons (UHC) as well as the discontinuous measurement of aldehydes and ketones using high-pressure liquid chromatography (HPLC).

3. Experimental procedure

The operating behaviour of the radiant burner was determined in short-term experiments as a function of various parameters. Several fibrous cylinders of different density were used. The emission values discussed further below apply to an uncoated body of 337 kg m^{-3}. Further parameters are the burner load relative to the outer burner surface, also referred to as power density *q*, the air ratio λ and the hydrogen ratio *m*. In general, the experiments have first been carried out with natural gas H, i.e. $m = 0$. Then hydrogen is added. The experimental aim is to determine the limits for the hydrogen fraction. One limiting factor is operational safety, which is no longer ensured if the reaction zone extends into the interior where the mixture is ignited (flashback). In this case, the fuel gas supply is switched off. The second limit is provided by the CO and UHC limit values, although the experiments are not switched off under these provisional test conditions if these limits are exceeded. The air ratio is varied between λ= I (stoichiometric) and 1.6.

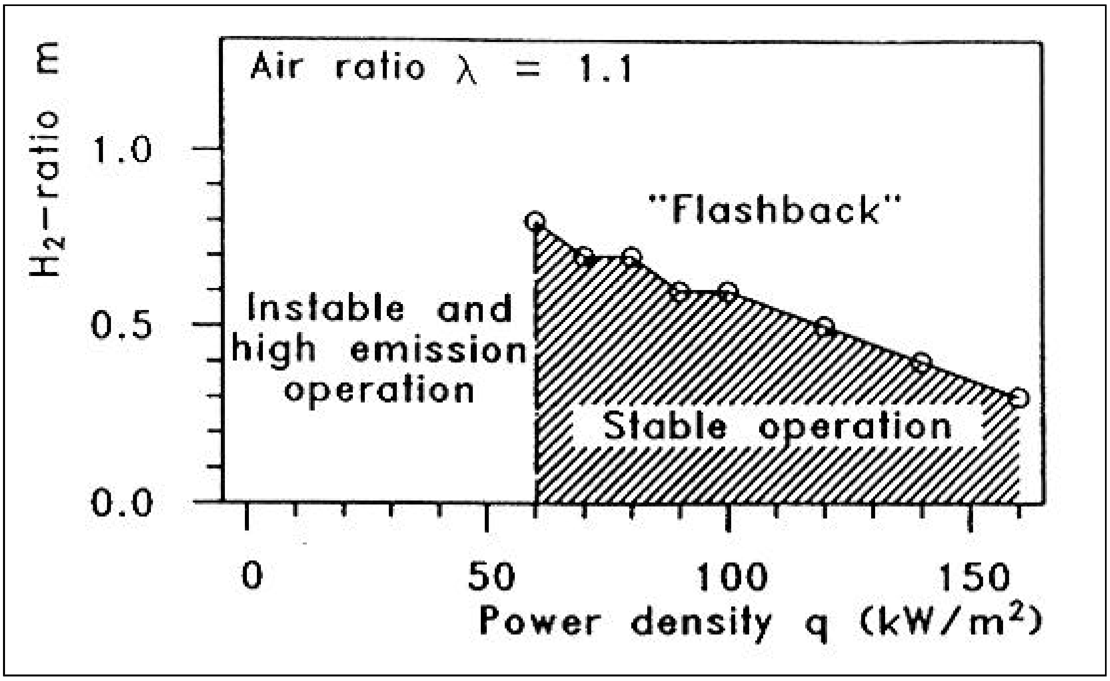

Fig. 3. H2, ratio vs Power density

4. Experimental results

Figure 3 shows the operating field of the burner body described in which the hydrogen ratio *m* is plotted as a function of the power density *q*. Safe operation without flashback is only possible in the hatched area. The possible hydrogen fraction decreases with increasing power density. The highest fraction achievable is $m = 0.8$ at a power density of 60 kW m^{-2}, whereas $q = 160$ kW m^{-2} only permits a ratio of $m = 0.3$. The flashback hazard in radiant burners is due to the low flow velocity of the mixture through the wall of the fibrous body. The maximum velocity at the highest flow rate is approximately 0.1 ms^{-1} and thus far below that in conventional burners. Since the laminar flame velocity of hydrogen is higher by about a factor of 7 than that of natural gas H (see Ref. [8]), the flashback hazard increases with elevated hydrogen fraction. The fact that radiant burners can be operated without flashback at all despite the low flow rates involved should be attributed, among other aspects, to the efficient heat removal from the reaction zone and the resultant low temperature level. As can be seen from *Fig. 4*, in which the CO emissions are shown as a function of the power density for the air ratio of 1.1, operation at high power densities was no longer possible for $m = 0.5$ (see also *Fig. 3*). At the lower power density of 70 kW m^{-2}, the CO emission was so high that it could no longer be represented on the selected scale. The significant decrease in CO concentration in proportion to the power density is to be attributed to better conversion of the fuel due to higher reaction temperatures. The diagram shows a clear graduation of the emissions up to a power density of 120 kWm^{-2}. Surprisingly, the CO concentrations increase with rising hydrogen fraction. The lowest values were measured for pure natural gas operation. With increasing hydrogen fraction, the reaction zone is shifted towards the incoming fuel due to the high flame velocity. This reduces the reaction temperatures and increases the fraction of unconverted fuel gas. The diagram suggests that the optimum is at a power density of about 120 kWm^{-2}.The emission values for this power density will therefore be discussed in more detail in the following.

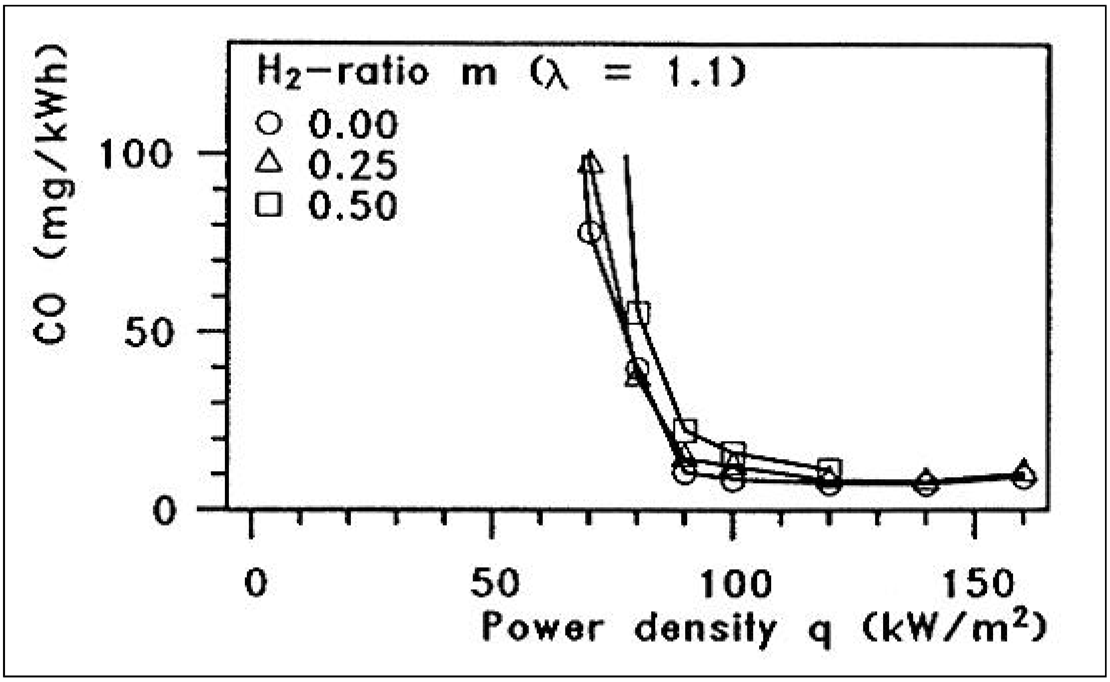

Fig. 4. CO emissions vs air ratio

The following *Figs 5, 6* and *7* show the CO, NO_x and UHC emissions as a function of the air ratio. The burner can be operated stoichiometrically without any difficulties. Under these conditions, the CO concentrations range between approximately 10 and 27 mg kWh^{-1}(*Fig. 5*). They then decrease to a minimum at air ratios between 1.05 and 1.1. This is in agreement with earlier experiments using methanol [7].

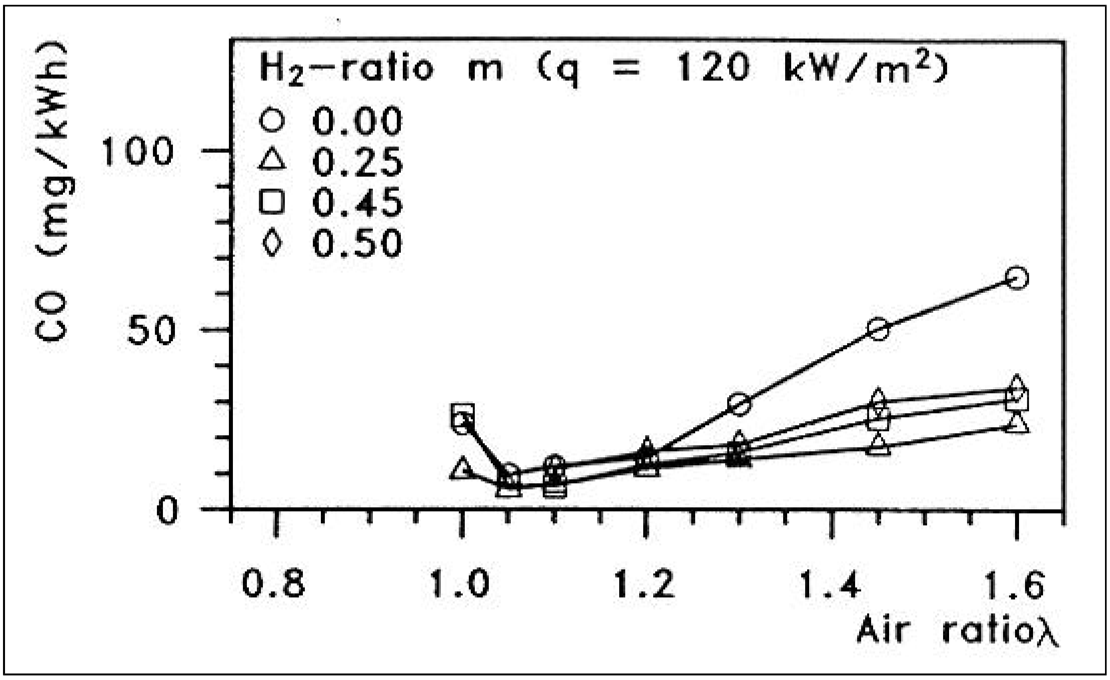

Fig. 5. CO emissions vs air ratio

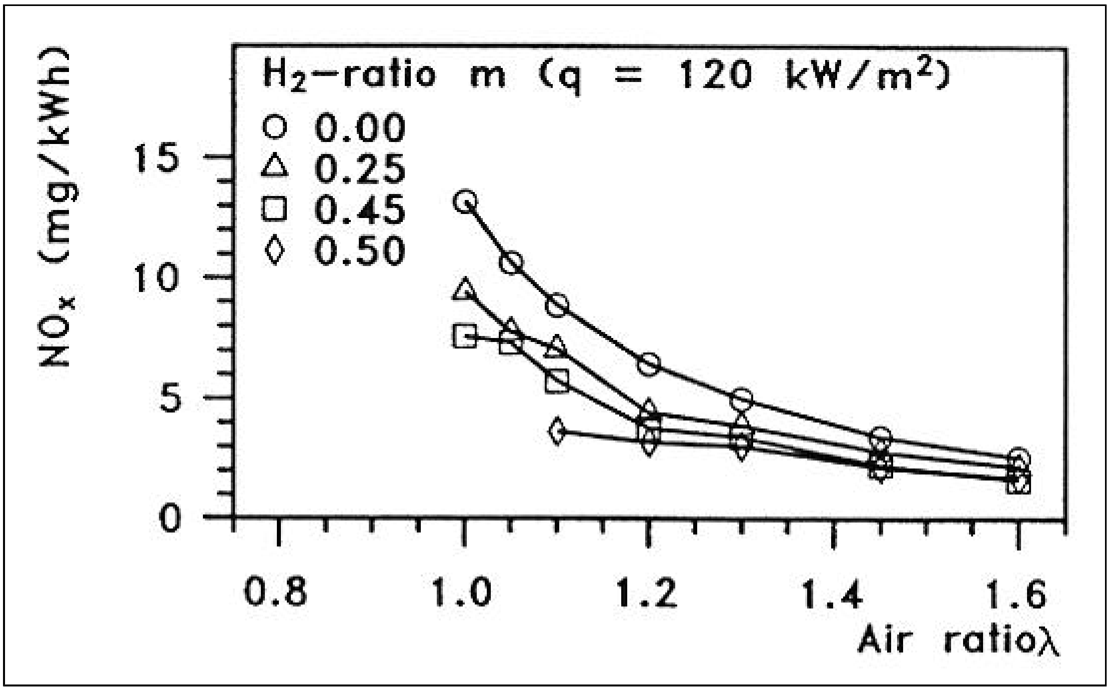

Fig. 6. NO_x emissions vs air ratio

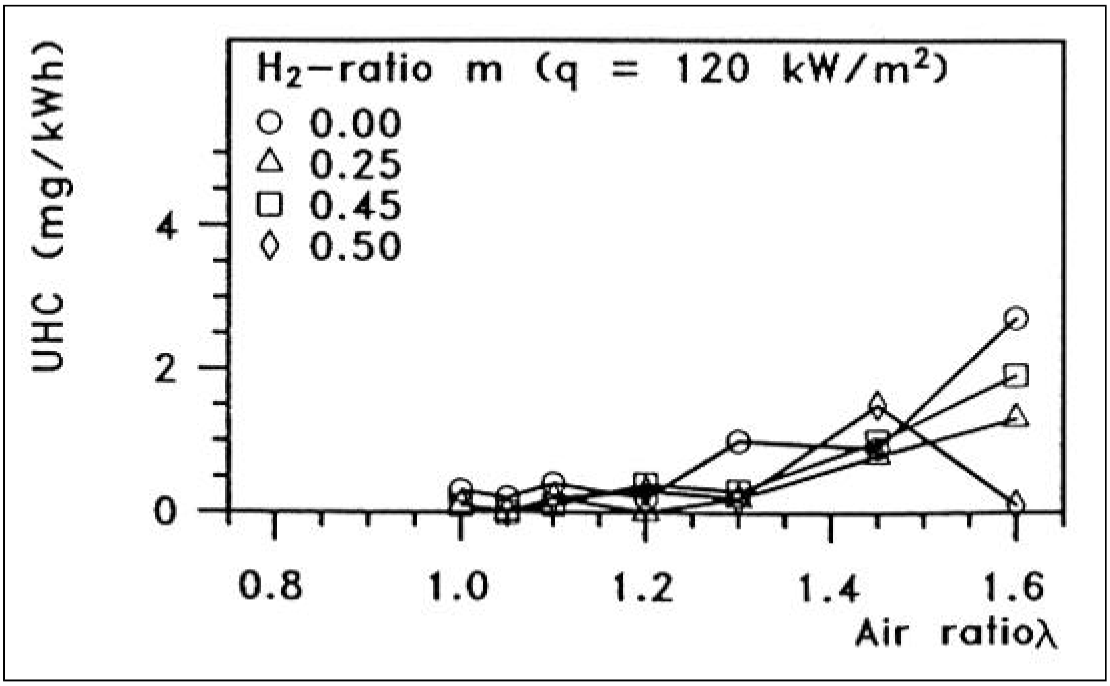

Fig. 7. UHC emissions vs air ratio

The CO concentrations then steadily increase due to shorter residence times, reversing the trend just outlined. Pure natural gas leads to the highest emissions. A small hydrogen fraction of 25 % for example causes an increase of about 5 mg kWh^{-1} at $\lambda = 1.1$ to about 25 mg kWh^{-1}. Higher hydrogen fractions lead to higher values. The NO_x emissions are clearly graduated according to fuel gas and hydrogen fractions (see *Fig. 6*). Higher hydrogen quantities lead to lower NOx values at any air ratio due to reduced reaction temperatures. The excess air at air ratios greater than 1 causes the values to decrease due to the cooling effect. A suitable catalyst will further reduce these concentrations, which are already low. The unburned hydrocarbons measured as propane equivalent and shown in *Fig. 7* are below 0.5 mg kWh^{-1} in the lower range of air ratios up to 1.2. A clear classification of the individual measuring results does not appear meaningful and would be rather incidental. Even the highest concentration of 2.5 mg kWh^{-1} measured at $\lambda = 1.6$ for natural gas operation is tolerable. Although a separation of the hydrocarbons is possible and was indeed done, the results will not be presented and discussed here.

5. Summary

The investigations carried out so far with a radiant burner as a precursor of the catalytic burner to be developed for operation with natural gas H and mixtures of natural gas H and hydrogen have shown that the system used can be operated with pure natural gas and hydrogen fractions between 0.25 and 0.8. Operation with higher fractions up to 100 % hydrogen is not yet possible at present. However, this goal could be achievable by using a sufficiently active catalyst in conjunction with burner operation at low power densities, e.g. 60 kWm^{-2} or other measures e.g. putting in the inner part of the porous structure a thin wall with holes. The combustion efficiency is higher than 99.99 %.

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