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Novel compact steam reformer for fuel cells with heat generation by catalytic combustion augmented by induction heating[1]

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Abstract

The overall objective of the project is to investigate, design and test an improved steam reformer for fuel cell power plants. Catalytic combustion is used to minimise the disadvantages of conventional external reforming. By integrating various technologies, smaller-scale reformer, lower emission levels and reduced start-up time along with improved temperature control can be achieved.

Catalytic combustion is used to generate the heat required for the reforming reaction, thus avoiding NO_x formation. The heat can be generated locally and made available for the reforming reaction using a thermally conductive substrate.

The first results of the catalytic test reactor show good conversion rates for both the combustion and the reforming reaction. Based on the hydrogen production rate, the initial objectives for the volume of the reformer have proven to be feasible.

The results are used to build a 20 kW integrated test reactor.

Keywords: Methane-steam reformer; Catalytic combustion; Induction heating; Fuel cell feed

1. Introduction

Fuel cells are very clean high-efficient electricity producing systems. The fuel processor normally consists of a steam reformer, in which methane is converted into hydrogen.

In natural gas reforming the following reaction occurs:

$$CH_4 + H_2O \rightleftarrows CO + 3H_2, \quad \Delta H_0 = -206\,\mathrm{kJ/mol}.$$

In order to sustain this highly endothermic reaction, external heat supply is necessary.

The application of conventional natural gas steam reforming has some disadvantages. The reformer, for instance, is large; about 30–40% of the volume of the fuel cell system is taken up by the fuel processor and the heat exchanging equipment.

This large size requires heavy investments and long start-up times.

In fuel cell units, NO_x emissions only stem from the fuel combustion process, during which heat is produced for the endothermic process.

Reductions in size, costs and start-up time for reformers will increase the attractiveness of fuel cell systems.

The target values for the novel compact reformer are based on a significant improvement as compared to

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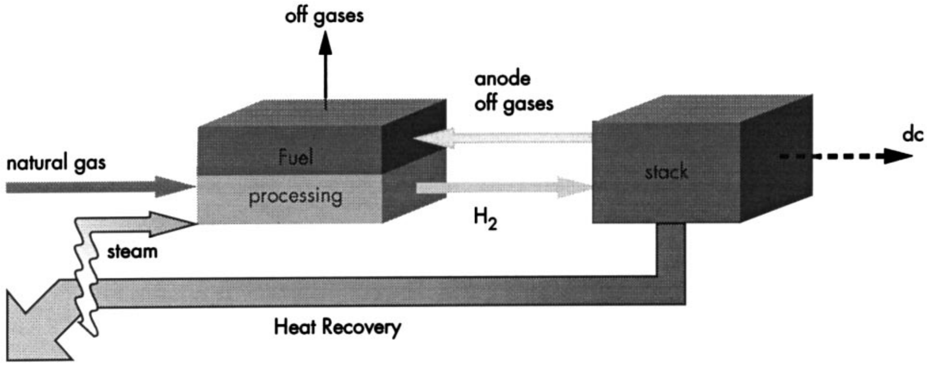

Fig. 1. Simplified scheme for integration of the reformer in the fuel cell plant.

state-of-the-art reformers in the year 1994. In this project, targets were set for volume, start-up time and costs.

The volume target was set at 5 l/kW$_e$.

Of the different fuel cell types, the solid polymer fuel cell (SPFC) was selected as the best option for combination with the novel compact reformer. For the SPFC a significant cost reduction is foreseen and it may therefore have good market perspectives, in particular for small-scale CHP units. These units need a short start-up time and a compact design.

A simplified flow chart for a fuel cell plant is given in Fig. 1.

The anode off gases are fed into the combustor. The anode off gases contain carbon dioxide, hydrogen, water, nitrogen and methane. If necessary, a small part of the combustor feed can be supplied by a natural gas flow.

In order to achieve zero emissions, catalytic combustion is applied as a heat source to sustain the endothermic reaction. By using metal monoliths and sintered metals as a heat conductive support, heat transfer and heat production are being integrated, resulting in a compact design.

A scheme of this concept is given in Fig. 2.

The combustion and reforming reactions take place in a metal plate reactor with intimate contact between the combustion and the reformer side.

By applying a coil around the reactor, induction is used as a heating source for start-up, to achieve uniform heating and as additional heat supply under load changes.

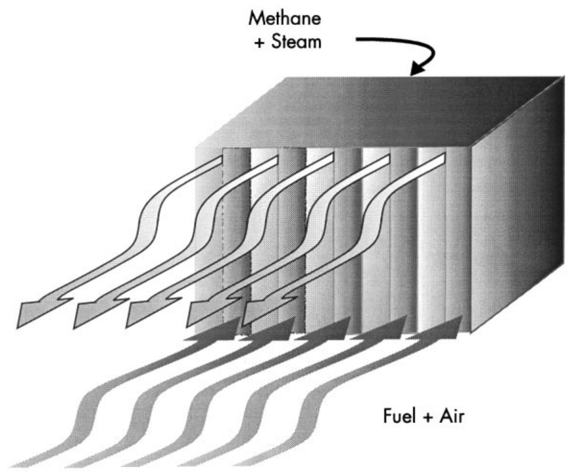

Fig. 2. Concept of reforming with heat supply by catalytic combustion through heat conductive substrates.

2. Project outline

The main activities in the project can be summarised as follows:

- Development of reformer and combustion catalysts.
- Coating development.
- Substrate material research.
- Induction heating of metallic substrates in a test rig.
- Modelling of heat transfer.
- Techno-economic assessment.
- Design, building and testing of a catalytic test rig.
- Design, building and testing of an integrated reactor.

The results of the research into combustion catalysts and of the catalytic test rig will be discussed in this paper. These results will be used for the final integrated reactor design.

3. Experimental

The combustion catalyst was selected from various precious metal compositions. Several powder metal catalysts were tested for their methane conversion efficiency, at a space velocity of 3000 h^{-1} at 1 bar in a mixture of 0.5% methane, 10% oxygen and balance nitrogen. The catalyst volume used was 1 cm^3. In order to account for the effect of degradation by water and by high temperatures a test procedure was developed to select suitable catalysts.

In order to simulate the interaction between the reforming and the combustion reaction a catalyst test rig was built. In the predesign stage, the plate reactor configuration was identified as the most suitable.

Calculations of the reactor's dimensions were based on reaction kinetics and heat transfer. The test reactor is shown in Fig. 3.

The inner part of the metal reactor consists of alternate foils of corrugated and flat high temperature steel, thus creating a metal monolith structure. The foils were bound together by means of an oxidation treatment. The CPSI (channels per inch^2) number of the monoliths is 250. Combustion takes place in the outer two parts of the sandwich structure.

A washcoat of a precious metal mixture was applied on the metal support of the combustion sections. The composition of this precious metal mixture was slightly different from the powder catalyst described in the activity tests but the metal mixture shows similar behaviour.

The reformer section was washcoated with a reformer catalyst. Both reformer and combustion coating were prepared by Degussa AG.

The reactor was tested with a co-flow arrangement of the combustion and reforming sections.

The composition of the inlet gas for the reformer and combustion sections is outlined in Table 1.

4. Results and discussion

4.1. Activity tests

Tests showed good behaviour for a bimetallic precious metal catalyst.

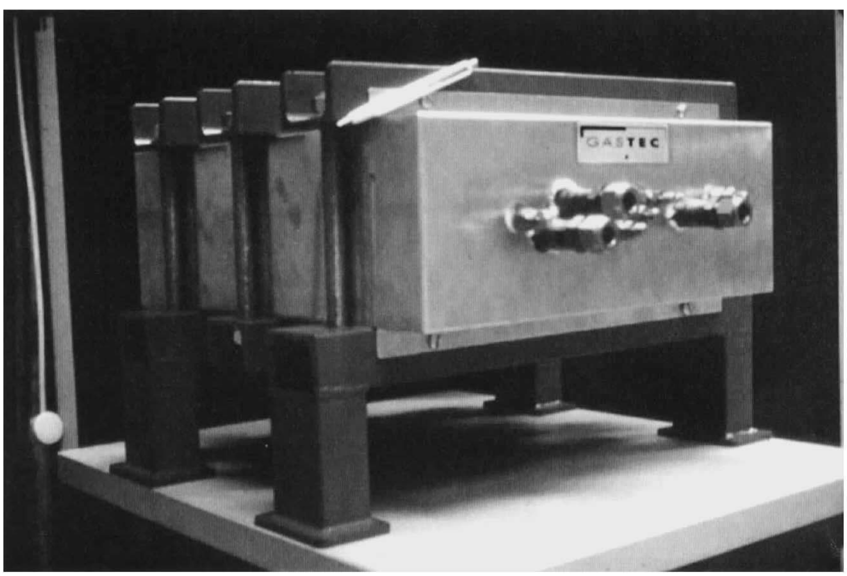

Fig. 3. The catalytic test reactor.

Table 1
Inlet gas composition for the reformer and combustion sections of the catalytic test rig

	Combustion section	Reformer section
Inlet temperature (°C)	530	380
CH_4 flow (Nl/min)	1–4	2–8
Air flow (Nl/min)	300	–
N_2 flow (Nl/min)	–	2–8
Steam flow (Nl/min)	–	20

After conditioning at 450°C the temperatures for 50% and 90% conversion are 290°C and 310°C, respectively.

The activity in a 30% water feed after an ageing treatment at 900°C in a 30% water feed was lower. The temperatures at 50% and 90% conversion are 375°C and 400°C, respectively.

The activity in a 30% water feed is sufficiently high for operation in the novel compact reformer. However, the initial activity can be recovered by exposure to a dry feed.

4.2. *Catalytic test rig results*

The temperature was measured at some points in the centre of the reactor (see Figs. 4 and 5).

Good heat transfer was observed between the combustion and the reforming sections.

At a short distance behind the entrance of the reactor, the temperature levels of the reformer and combustion sections were equal. The conversion of the combustion and reforming reactions are more than 99.98% and 97%, respectively. The NO_x levels are within the ppb range.

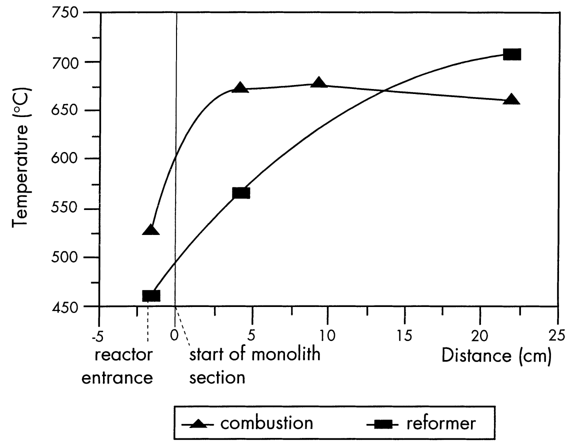

Fig. 5. Temperature measurements in the combustion and reformer sections.

Based on the hydrogen output at the reformer section a volume of less than 1.5 l/kW_e for the reactor internals is feasible. Assuming a three-fold volume for the total reformer, the volume target of 5 l/kW_e seems within reach.

Further improvements are possible by improved contacts between the metallic substrate and the reactor walls as well as by increasing the input temperature. Improved catalysts formulations will be implemented in the final reactor.

Furthermore, the relative heat loss to the environment is very high for this test reactor. An increased

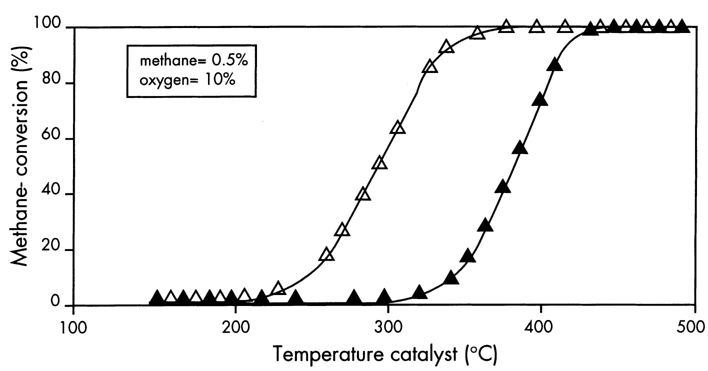

Fig. 4. Activity test of a precious metal mixture in 0.5% methane in nitrogen: (1) after conditioning, (2) after ageing and in 30% water.

number of sections will be used for the final reactor, thus leading to a more cubic shape and a relatively lower heat loss to the environment.

5. Conclusions

Utilisation of a precious metal mixture may lead to a good methane conversion even in a 30% water-saturated methane mixture at low temperatures.

The concept of a zero-emission reactor with coated metallic internals and metallic contacts between the combustion and reforming sections is operating satisfactorily.

First results show that the volume targets for the compact reformer with heat generation by catalytic combustion are feasible. A 20 kW integrated reactor will be tested in a demonstration project.

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