

Small-scale testing of a precious metal catalyst in the autothermal reforming of various hydrocarbon feeds

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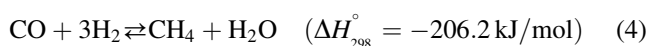
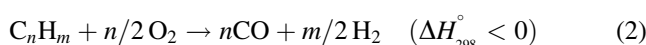
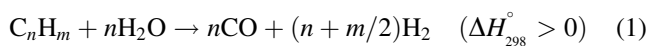
Abstract

The autothermal reforming of various hydrocarbon feeds with boiling ranges between 190 and 325 °C was examined on a precious metal catalyst in a small-scale tubular reactor to assess the feasibility of hydrogen production from diesel fuel. The experiments were performed at $n(\text{O}_2)/n(\text{C})$ and $n(\text{H}_2\text{O})/n(\text{C})$ ratios between 0.34–0.47 and 1.50–2.20, respectively. The space velocities (GHSV) amounted to between 13,000 and 18,000 h⁻¹. The hydrocarbon conversion, the composition of the product gas and the stability of the catalyst were determined as a function of the operating variables and the composition of the feed. The hydrocarbon conversion and catalyst bed temperatures increased with increasing $n(\text{O}_2)/n(\text{C})$ ratio. The $n(\text{H}_2\text{O})/n(\text{C})$ ratio showed a smaller effect on the hydrocarbon conversion than the $n(\text{O}_2)/n(\text{C})$ ratio. The composition of the product gas at conversions above 95% corresponded closely to the thermodynamic equilibrium. No decrease of conversion was observed for times on stream of approximately 20 h. The addition of 1-benzothiophene to a hydrocarbon feed yielding sulfur contents of 11 and 30 wt.ppm caused a decrease in the hydrocarbon conversion by deactivation of the catalyst. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Autothermal reforming; Hydrogen; Diesel fuel; Fuel cell

1. Introduction

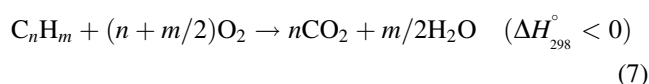
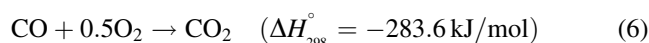
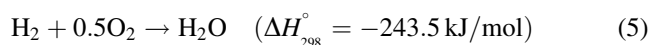
Hydrogen can be produced from alcohols and hydrocarbons by the process of autothermal reforming. Generally, the fuel is converted into a hydrogen-rich product gas using air and steam on a catalyst at high temperatures. The autothermal reforming of a hydrocarbon C_nH_m can be described by the following reactions:



The necessary heat for the endothermic steam reforming, Eq. (1), is supplied by the partial oxidation of hydrocarbons, Eq. (2). The shift reaction, Eq. (3), and the methanation reaction, Eq. (4), proceed simultaneously and yield a gas composition which corresponds to the thermodynamic equilibrium [1,2]. Important operation parameters for the

process are temperature, pressure, $n(\text{O}_2)/n(\text{C})$ and $n(\text{H}_2\text{O})/n(\text{C})$ ratio. These parameters should be chosen with the aim of avoiding the formation of carbon and minimizing the methane content of the product gas. Criteria for carbon-free operation can be found by means of thermodynamic calculations [3,4]. Nevertheless, the formation of carbonaceous deposits can occur in the autothermal reforming process of higher hydrocarbons [4].

It should be mentioned that the oxidation of the products hydrogen and carbon monoxide, Eqs. (5) and (6), may also occur. The result of a combination of Eq. (2) with Eqs. (5) and (6) is the total oxidation of hydrocarbons, Eq. (7). The consumption of oxygen for the total oxidation will enhance the amount of hydrocarbons to be converted by steam reforming.



For electric vehicles powered with a polymer electrolyte fuel cell on-board hydrogen production using liquid energy carriers has attracted much attention [3,5,6]. With regard to the existing infrastructure the use of gasoline or diesel fuel

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would be desirable. Both fuels are hydrocarbon mixtures containing alkanes, alkenes and aromatics. For example, diesel fuel with a boiling range between 200 and 380 °C can consist of several hundred different compounds.

The specific physical and chemical fuel properties adapted for combustion in ignition and diesel engines must be taken into account for the design of an on-board reforming process. For example, the composition of the fuel might affect the choice of the $n(\text{O}_2)/n(\text{C})$ and the $n(\text{H}_2\text{O})/n(\text{C})$ ratio. Furthermore, aromatics and alkanes show different reactivities in the autothermal reforming process. For gasoline the hydrogen production from single compounds has been investigated between 600 and 800 °C for $n(\text{O}_2)/n(\text{C}) = 0.42$ and $n(\text{H}_2\text{O})/n(\text{C}) = 1.4$ [7]. At 600 °C, the production decreased in the following order: *iso*-octane > *n*-octane > methylcyclohexane > toluene > trimethylbenzene, showing that aromatics are less reactive than alkanes and cycloalkanes. Hydrogen production from the aromatic compounds increased at higher temperatures and the differences between the compounds were less pronounced.

Gasoline and diesel fuel contain sulfur compounds, which can cause deactivation of the catalyst by poisoning. Today in the European Union the sulfur content of diesel fuel is limited to 350 wt.ppm. Lower sulfur limits of 50 wt.ppm for gasoline and diesel fuel will be introduced in 2005. Even today diesel fuels with 10 wt.ppm are occasionally available. Various sulfur containing fuels could be successfully converted by autothermal reforming on precious metal catalysts. For example, No. 2 fuel oil containing 1200 wt.ppm sulfur was processed at temperatures between 940 and 780 °C in the catalyst bed with a conversion of 98.8% ($n(\text{O}_2)/n(\text{C}) = 0.378$ and $n(\text{H}_2\text{O})/n(\text{C}) = 2.57$) [8]. The long term behavior of the catalyst was not reported. Experiments with gasoline containing 50 wt.ppm sulfur were performed for approximately 1700 h [7]. During this operation time a decrease of the hydrogen content of the dry product gas from 39.0 to 37.5% was observed. A change in the hydrocarbon conversion was not described. Further successful tests with gasoline and diesel fuel with a sulfur content of 20 wt.ppm are reported without details in [9].

Autothermal reforming of gasoline has already led to the development of the first on-board reformers [9–13]. Information about the design of these reactors is rarely given. Obviously, in most cases metal or ceramic monoliths coated with a washcoat and the catalyst were employed. Precious metals have been recognized as suitable catalysts for the reaction [1,2,5–12,14,15]. In an earlier study nickel catalysts were also used for the autothermal reforming of *n*-tetradecane and benzene [4].

This paper presents results of the small-scale testing of a precious metal catalyst in the autothermal reforming of various hydrocarbon feeds with boiling ranges between 190 and 325 °C. The composition of the feeds is chosen with the aim of assessing the feasibility of the autothermal reforming of diesel fuel and of determining the appropriate operating variables for this process. The effect of changes of

operating variables, composition of feed and the sulfur content of the fuel on the hydrocarbon conversion, the composition of the product gas and the stability of the catalyst are determined.

2. Experimental

All experiments were performed in a tubular reactor (inner diameter 11 mm) surrounded by an electrically heated furnace. The use of a furnace was necessary to offset the reactor's heat losses. Hydrocarbons and water were vaporized and mixed in front of the tubular reactor in a vaporizer, which was heated by a second electric furnace. A nitrogen stream of 30 Nl/h was fed into the vaporizer to obtain a steady hydrocarbon/steam mixture. The air supply tube ended directly in front of the catalyst bed to prevent homogeneous oxidation reactions. The inner parts of the tubular reactor were made of titanium to avoid catalytic reactions on the stainless steel surface. The hot product gas was rapidly cooled to room temperature behind the catalyst bed to prevent post catalytic reactions. Water and liquid hydrocarbons were separated from the product gas in a condenser. All gas and liquid flows were controlled by flow-meters and the whole test system was operated by a process control system. The dry product gas was analyzed by means of a mass spectrometer. The time between two measurements amounted to approximately 30 s. The concentrations of the following gases were determined: H₂, CO₂, CO, CH₄, N₂, O₂, C₂H₄, C₂H₆, C₃H₆ (propene), Ar and H₂O.

The proprietary precious metal catalyst was supplied by dmc² Degussa Metals Catalysts Cerdec AG (spheres with an average diameter of 2 mm). The catalyst was placed in the reactor on a layer of Al₂O₃ grains. In each experiment 6.0 g of catalyst was employed (length of catalyst bed: 165 mm). Temperatures in the catalyst bed were measured by thermocouples at four fixed positions. A new sample of catalyst was used for each hydrocarbon feed. Table 1 shows the hydrocarbon feeds and their main chemical and physical properties. The alkane mixtures were produced by TOTAL FINA ELF (sold under the trade names of Hydroseal G 232 H and Hydroseal G 3 H) and consisted of *n*-, *iso*- and cycloalkanes with the C-number distributions indicated in Table 1. 1,2,3,4-Tetrahydronaphthalene (THN), decahydronaphthalene (DHN) and 1-benzothiophene were purchased from Aldrich.

The following were used as constant conditions for the experiments: hydrocarbon feed: 30 g/h, pressure: 1.6 bar (abs.) and reactor furnace temperature: 550 °C. The $n(\text{H}_2\text{O})/n(\text{C})$ and the $n(\text{O}_2)/n(\text{C})$ ratios were varied in the range of 1.50–2.20 and 0.34–0.47, respectively. The space velocities (GHSV) amounted to between 13,000 and 18,000 h⁻¹. Oxygen was always completely converted. The calculations of the hydrocarbon conversion were based on the CO₂, CO and CH₄ contents of the product gas. Condensable hydrocarbons leaving the reactor were considered

Table 1
Chemical and physical properties of the hydrocarbon feeds

Hydrocarbon feed	Composition of feed (wt.%)	Boiling range or boiling point (°C)	Sulfur content (wt.ppm)
C ₁₃ –C ₁₅	C ₁₃ –C ₁₅ -Alkane mixture/100	235–265	1
C ₁₃ –C ₁₉	C ₁₃ –C ₁₅ -Alkane mixture/50	235–265	1
	C ₁₄ –C ₁₉ -Alkane mixture/50	275–325	1
C ₁₃ –C ₁₅ /DHN	C ₁₃ –C ₁₅ -Alkane mixture/75	235–265	1
	Decahydronaphthalene/25	189–191	–
C ₁₃ –C ₁₅ /THN	C ₁₃ –C ₁₅ -Alkane mixture/85	235–265	1
	1,2,3,4-Tetrahydronaphthalene/15	207	–

as non-converted although the chemical structure of compounds might have been changed by isomerization, dehydrogenation or cracking reactions. Figures showing the product gas composition only contain the amount of nitrogen of the air stream and not the amount of nitrogen fed into the vaporizer.

Procedure: Before reaction the catalyst was reduced with hydrogen (1% H₂ in N₂) at approximately 300 °C. Then the water feed was started and, subsequently hydrocarbon and air were fed into the reactor. The oxidation of the hydrocarbon started at catalyst temperatures of 250 °C and higher. Finally, the furnace temperature was raised to 550 °C. For each feed autothermal reforming was performed at different $n(\text{O}_2)/n(\text{C})$ ratios with times on stream between 5 and 20 h for a selected ratio.

3. Results and discussion

The conditions of the experiments were chosen with the objective of working below complete hydrocarbon conversion, so that the influence of operating variables and the feed composition on the process could be determined by the conversion level and a possible decrease in conversion

caused by a deactivation of the catalyst could also be observed within a few hours. It should be pointed out that a hydrogen production process as part of a fuel cell system must be operated at complete hydrocarbon conversion to avoid poisoning of the catalytic processes for product gas purification and the fuel cell.

For the autothermal reforming of the C₁₃–C₁₅-feed the hydrocarbon conversion and the temperature at $x/L = 0.26$ in the catalyst bed versus time on stream are shown in Fig. 1 for a $n(\text{O}_2)/n(\text{C})$ ratio of 0.45. A steady conversion of approximately 96% was determined during 20 h on stream. The temperature in the catalyst bed decreased from the beginning to the end (Fig. 2), indicating that the exothermic oxidation reactions tended to proceed in the first part of the catalyst bed. Further reasons for the pronounced temperature profile are the heat losses of the reactor and the product gas cooling behind the catalyst bed. The composition of the dry product gas is given in Fig. 3. The concentrations amounted to 36.5% H₂, 15.2% CO₂, 7.0% CO, 1.3% CH₄ and 40.0% N₂. C₂H₄, C₂H₆ and C₃H₆ were not found. The CH₄ content was rather high, which can be attributed to the low end temperature of the catalyst bed. Due to the heat losses of the tubular reactor it was not possible to work at significantly higher end temperatures. A calculation revealed that the

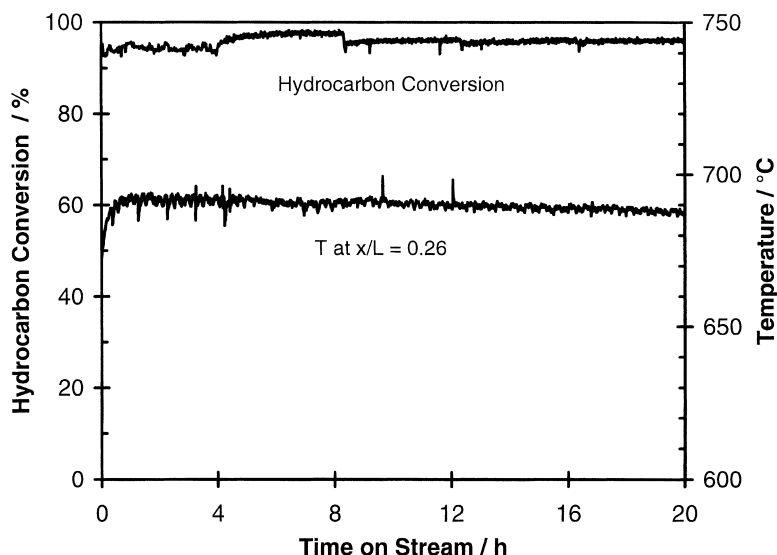


Fig. 1. Autothermal reforming of a C₁₃–C₁₅-alkane mixture ($n(\text{O}_2)/n(\text{C}) = 0.45$ and $n(\text{H}_2\text{O})/n(\text{C}) = 2.20$).

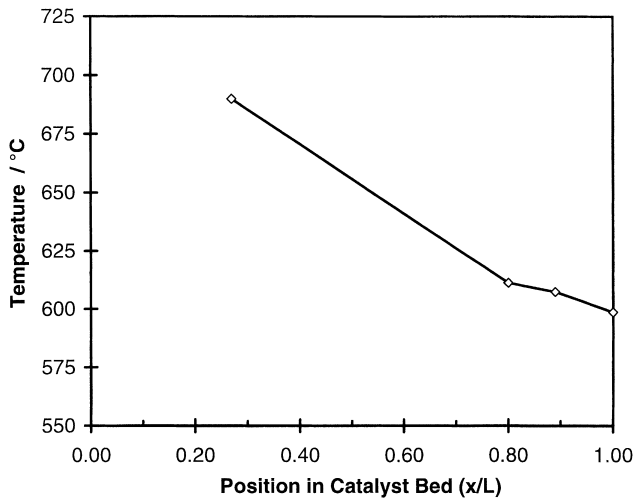


Fig. 2. Catalyst bed temperatures for the autothermal reforming of a C_{13} – C_{15} -alkane mixture ($n(O_2)/n(C) = 0.45$ and $n(H_2O)/n(C) = 2.20$).

composition of the product gas corresponds closely to the equilibrium if the end temperature of the catalyst bed is applied in the calculation. For the conditions given in Fig. 1 and *n*-tetradecane as hydrocarbon feed the equilibrium calculation yielded the following composition at a reaction temperature of 607 °C: 37.0% H_2 , 15.1% CO_2 , 6.5% CO , 1.8% CH_4 and 39.6% N_2 .

Diesel fuel is a complex hydrocarbon mixture with a boiling range between 200 and 380 °C. Compounds which can be found in diesel fuel include alkanes, aromatics and in smaller amounts alkenes with carbon numbers in the range of C_{10} – C_{22} . Due to their high cetane number alkanes are the most valuable compounds. Four different hydrocarbon feeds were chosen to examine the effect of the fuel composition on the autothermal reforming process. The C_{13} – C_{15} -feed and the C_{13} – C_{19} -feed are alkane mixtures with different boiling ranges. The third feed is composed of the C_{13} – C_{15} -alkane mixture and the cycloalkane DHN. THN was added to the

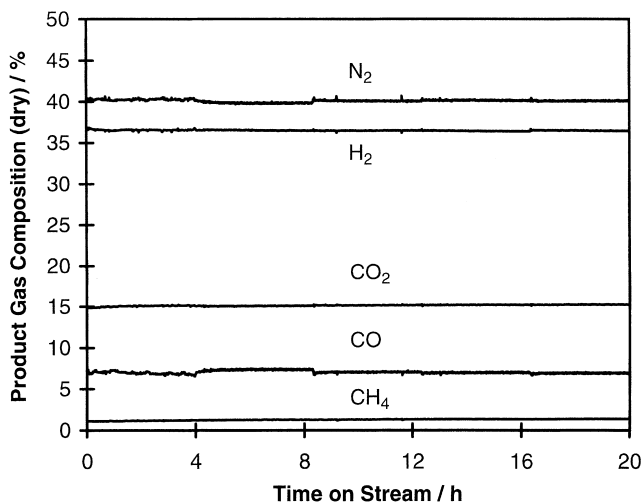


Fig. 3. Composition of the product gas ($n(O_2)/n(C) = 0.45$ and $n(H_2O)/n(C) = 2.20$).

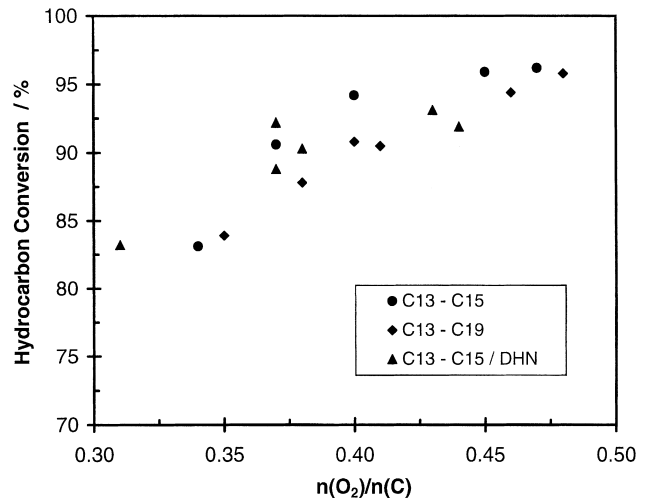


Fig. 4. Effect of $n(O_2)/n(C)$ ratio on hydrocarbon conversion for different alkane mixtures ($n(H_2O)/n(C) = 2.20$).

alkane mixture as an aromatic compound which is commonly found in diesel fuel. For each feed the hydrocarbon conversion was determined at different $n(O_2)/n(C)$ ratios (Figs. 4 and 5). No decrease of conversion was observed for times on stream of approximately 20 h. The conversion increased gradually when the air feed was enhanced. At high $n(O_2)/n(C)$ ratios the conversion approached 97%. Increasing the air feed also led to higher catalyst temperatures, which is shown for the C_{13} – C_{19} -alkane mixture in Fig. 6. The temperature increase was more pronounced in the first part of the reactor. The enhancement of the hydrocarbon conversion can be explained by a higher reaction rate of the oxidation compared to the steam reforming and the increase of the temperature. No pronounced differences of the hydrocarbon conversion as a function of the $n(O_2)/n(C)$ ratio could be detected between the feeds containing only alkanes (Fig. 4). The aromatic compound THN seems to have a detrimental effect on the conversion (Fig. 5).

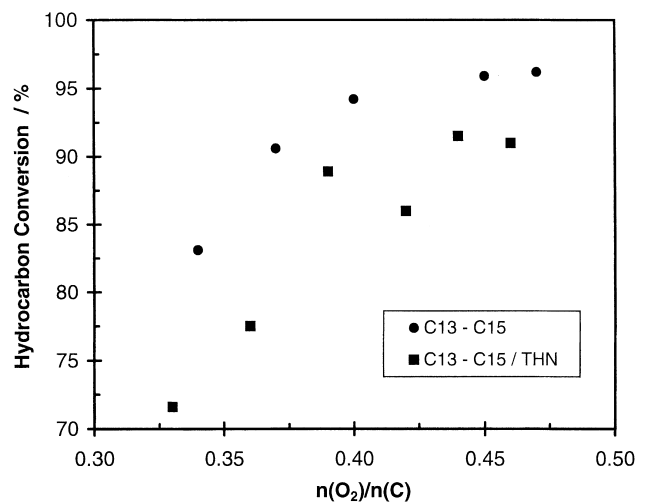


Fig. 5. Effect of the aromatic compound THN on the hydrocarbon conversion ($n(H_2O)/n(C) = 2.20$).

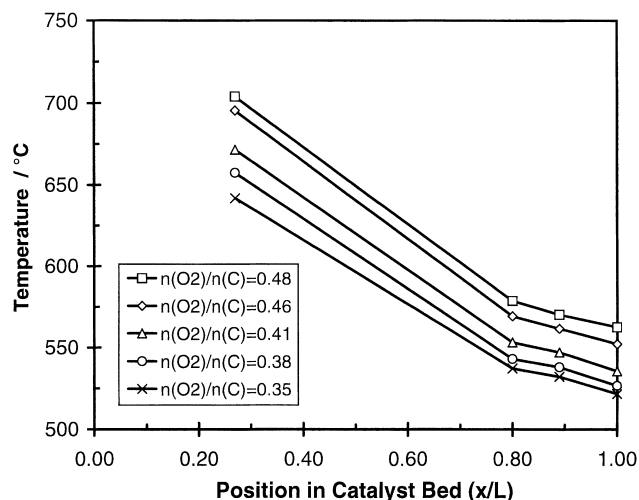


Fig. 6. Influence of the $n(\text{O}_2)/n(\text{C})$ ratio on the catalyst bed temperatures (feed: $\text{C}_{13}\text{--C}_{19}$ -alkane mixture and $n(\text{H}_2\text{O})/n(\text{C}) = 2.20$).

Particularly at low $n(\text{O}_2)/n(\text{C})$ ratios the conversion was lower compared with the pure $\text{C}_{13}\text{--C}_{15}$ -feed. This result might indicate that aromatic compounds have lower reaction rates in the oxidation or steam reforming reaction of the autothermal reforming process.

For the series of experiments with the $\text{C}_{13}\text{--C}_{15}$ -feed the composition of the product gas at different $n(\text{O}_2)/n(\text{C})$ ratios is given in Fig. 7. The composition will be affected by the $n(\text{O}_2)/n(\text{C})$ ratio and the temperature. Calculations showed that by increasing the $n(\text{O}_2)/n(\text{C})$ ratio from 0.34 to 0.47 and, simultaneously the temperature from 549 to 607 °C, corresponding to the end temperatures of the experiments, the dry product gas will contain more H_2 , CO and N_2 and less CO_2 and CH_4 for complete conversion of the hydrocarbon. However, the experimental results showed opposite tendencies. The H_2 and CO content decreased and the CO_2 and CH_4 content increased. A possible explanation for this result could be the low hydrocarbon conversion at $n(\text{O}_2)/n(\text{C})$

ratios below 0.45 (Fig. 4), which might lead to a lower H_2 and CO content and to a higher CO_2 and a CH_4 content in the product gas than predicted by the equilibrium calculations for complete conversion.

The choice of an appropriate $n(\text{H}_2\text{O})/n(\text{C})$ ratio for the autothermal process is a difficult task. A high $n(\text{H}_2\text{O})/n(\text{C})$ ratio suppresses the formation of methane and coke. But it is important to take into account the fact that water must be recovered from the anode and cathode exhaust gas streams of the fuel cell and, subsequently steam is generated in a vaporizer. With respect to the volume and weight of the condenser and vaporizer a low steam feed would be beneficial. Within the experiments the influence of the $n(\text{O}_2)/n(\text{C})$ ratios on the hydrocarbon conversion was examined for the $\text{C}_{13}\text{--C}_{19}$ -feed at different $n(\text{H}_2\text{O})/n(\text{C})$ ratios (Fig. 8). Again, for each $n(\text{H}_2\text{O})/n(\text{C})$ ratio the conversion increased with increasing air feed. At constant $n(\text{O}_2)/n(\text{C})$ ratio the conversion can be affected by a change in the partial pressures of the reactants, the catalyst temperature and the GHSV. With increasing $n(\text{H}_2\text{O})/n(\text{C})$ ratio the steam partial pressure and the GHSV increased while the catalyst temperature profiles did not change. The highest conversions were measured at $n(\text{H}_2\text{O})/n(\text{C}) = 2.20$ connected with the highest steam partial pressures and GHSV. At $n(\text{H}_2\text{O})/n(\text{C}) = 1.50$ the conversions were slightly higher than for $n(\text{H}_2\text{O})/n(\text{C}) = 1.80$. The effect of steam is more pronounced at lower $n(\text{O}_2)/n(\text{C})$ ratios. The results might indicate that a high steam partial pressure has a beneficial influence on the steam reforming reaction. At the lowest $n(\text{H}_2\text{O})/n(\text{C})$ ratio this effect could have been offset by the decrease in GHSV yielding slightly higher conversions for $n(\text{H}_2\text{O})/n(\text{C}) = 1.50$ than for $n(\text{H}_2\text{O})/n(\text{C}) = 1.80$.

Sulfur compounds in the hydrocarbon feed can cause deactivation of the precious metal catalyst in the autothermal reforming process. Therefore, the effect of a low sulfur content in the hydrocarbon feed on the hydrocarbon conversion was studied. The sulfur containing feeds were

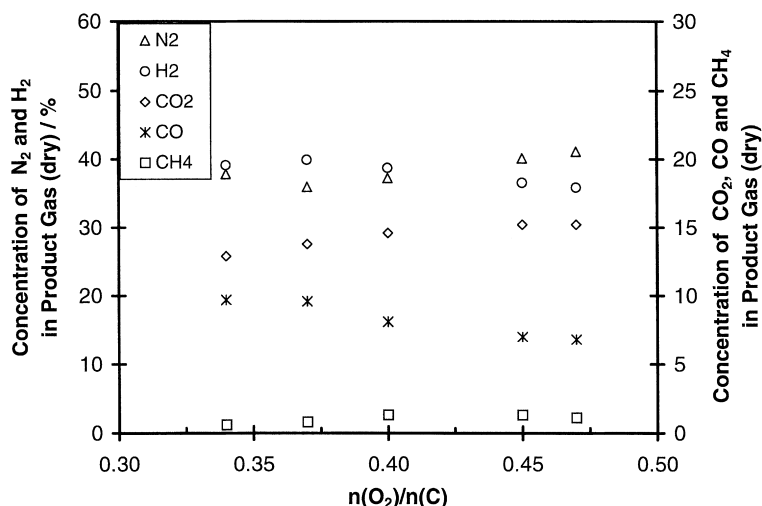


Fig. 7. Composition of the product gas at different $n(\text{O}_2)/n(\text{C})$ ratios ($n(\text{H}_2\text{O})/n(\text{C}) = 2.20$).

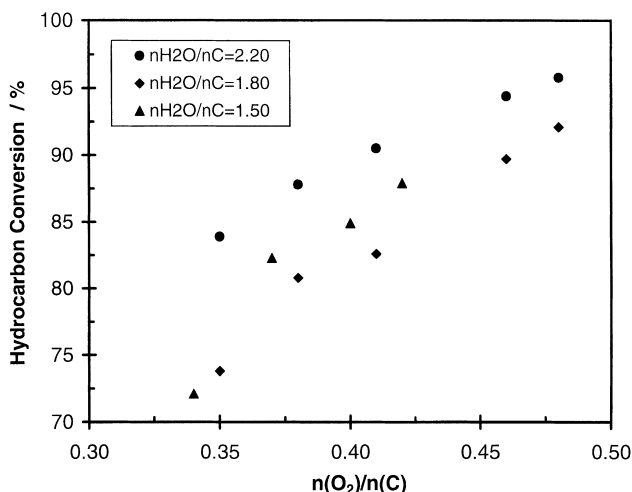


Fig. 8. Dependence of the hydrocarbon conversion on the $n(\text{O}_2)/n(\text{C})$ ratio at different $n(\text{H}_2\text{O})/n(\text{C})$ ratios (feed: C_{13} – C_{19} -alkane mixture).

prepared by the addition of small amounts of 1-benzothiophene to the C_{13} – C_{15} -alkane mixture to yield feeds with 11 and 30 wt.ppm sulfur. For both feeds a series of experiments was performed using a new sample of catalyst for each series. In a series of experiments the reaction was started with the pure C_{13} – C_{15} -alkane mixture and after approximately 20 h the sulfur containing feed was supplied for 20 h (Fig. 9). Finally, the feed was again switched back to the pure alkane mixture. Each change of feed was carried out without interrupting the reaction. Fig. 9 shows that when the sulfur containing feeds were supplied to the reactor the hydrocarbon conversion dropped sharply. The drop was faster and higher for the feed containing 30 wt.ppm sulfur. After some hours on stream the decrease of conversion slowed down and an almost constant conversion was attained. The decrease can likely be attributed to the poisoning of

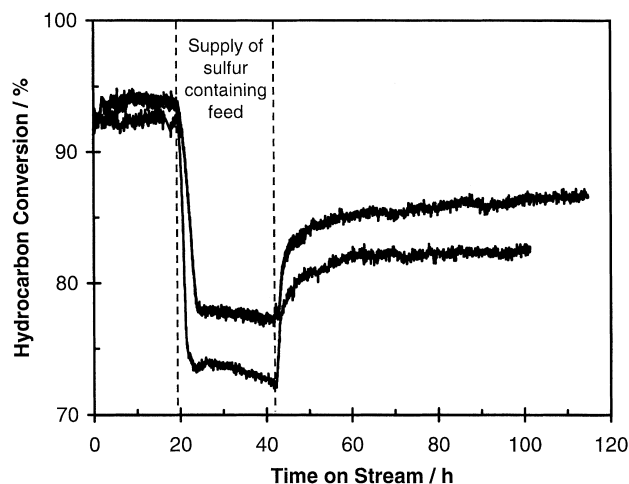


Fig. 9. Influence of the sulfur content in the C_{13} – C_{15} -alkane mixture on the hydrocarbon conversion ($n(\text{O}_2)/n(\text{C}) = 0.43$ and $n(\text{H}_2\text{O})/n(\text{C}) = 2.20$; upper curve: 11 wt.ppm sulfur and lower curve: 30 wt.ppm sulfur; operation with sulfur containing feed between approximately 20 and 40 h on stream).

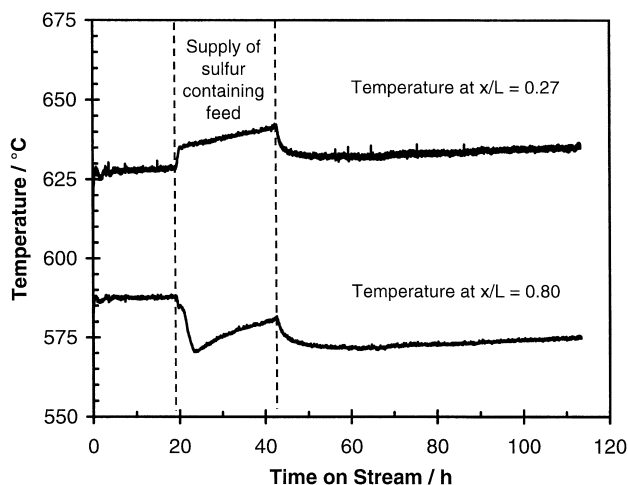


Fig. 10. Influence of the sulfur content in the feed on the catalyst bed temperatures (sulfur content: 11 wt.ppm, $n(\text{O}_2)/n(\text{C}) = 0.43$ and $n(\text{H}_2\text{O})/n(\text{C}) = 2.20$).

the catalyst by a gradual adsorption of sulfur. The poisoning seems to be partly reversible, since the conversion increased sharply when the feed was switched back to the pure alkane mixture. The original level of conversion could not be attained again, which might be attributable to an irreversible sulfur poisoning or to a very slow desorption of sulfur due to the low end temperature in the catalyst bed of approximately 580 °C. The change of feeds also showed an influence on the catalyst temperature (Fig. 10). The temperature at $x/L = 0.27$ increased by approximately 20 °C while the end temperature decreased by approximately 30 °C. A possible explanation of these observations is a suppression of the endothermic steam reforming reaction by sulfur poisoning and a shift of the oxidation reactions to the first part of the catalyst bed. During operation with the sulfur containing feeds the temperature in the catalyst bed increased gradually and the conversion decreased slightly (Fig. 9), which could have been caused by a slow shift to more total oxidation of hydrocarbons. The subsequent change to the pure alkane feed led to an enhancement of the steam reforming reaction causing the temperature decrease after approximately 42 h on stream.

4. Conclusions

The hydrocarbon mixtures employed can be converted by autothermal reforming into a hydrogen-rich product gas with a wide range of operating variables. Without the heat losses of the small-scale tubular reactor higher reaction temperatures and a less pronounced temperature profile will enable operation of a larger reactor at higher GHSV and also decrease the tendency for sulfur to adsorb on the catalyst. Therefore, the autothermal reforming of ultra-low sulfur diesel fuel (approximately 10 wt.ppm sulfur) with the precious metal catalyst seems to be a feasible process for the production of hydrogen in a polymer electrolyte fuel cell

system. Preliminary results of the operation of an autothermal reforming reactor with a power of approximately 5 kW confirmed this opinion.

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