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Operational experience with the fuel processing system for fuel cell drives

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Abstract

Electric motor vehicle drive systems with polymer electrolyte fuel cells (PEFCs) for the conversion of chemical into electrical energy offer great advantages over internal combustion engines with respect to the emission of hydrocarbons, carbon monoxide and nitrogen oxides. Since the storage systems available for hydrogen, the "fuel" of the fuel cell, are insufficient, it is meaningful to produce the hydrogen on board the vehicle from a liquid energy carrier, such as methanol. At the Research Center Jülich such a drive system has been developed, which produces a hydrogen-rich gas from methanol and water, cleans this gas and converts it into electricity in a PEFC. This system and the operational experience on the basis of simulated and experimental results are presented here. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Fuel cell drive system; Fuel processing system; Compact methanol reformer; Catalytic burner; Dynamic drive simulation; Full fuel cycle

1. Introduction

Vehicle drive systems with fuel cells as electrochemical transducers in conjunction with new, on the whole low-carbon fuels can make a considerable contribution to reducing traffic-related emissions (above all CO_2) and to reducing the energy demand of the transport sector. The use of nonfossil fuel, e.g. renewably produced hydrogen, will inevitably lead to special advantages with respect to a sustainable coverage of energy needs and the greenhouse gas problems. However, the necessary reorientation in the energy economy involves a long-term perspective.

In the short and medium term, priority will probably be given to energy carriers other than hydrogen for the provision of fuel cell feed gases for the broad energy market, e.g. natural gas for stationary applications, and methanol or liquid hydrocarbons for mobile applications. In order to investigate drive systems with fuel cells with a view to finding a promising solution concerning their performance efficiency, Haldor Topsøe A/S (HTAS), Siemens AG and Research Center Jülich (FZJ) decided to design, construct and test a fuel cell drive system with methanol/steam reformer including catalytic burner (CMR), gas cleaning by a gas separation membrane, and polymer electrolyte fuel cell (PEFC) [1]. The choice of methanol as the fuel provides the possibility of storing a liquid fuel of high energy density at ambient pressure and ambient temperature [2]. The essential components of the complete fuel cell drive test rig built up within the framework of a European project cooperation between the three partners have the technical data given in Table 1. The setup and functions are described in detail in articles [3–5]. The subject matter of the present study is the description of the simulation and operation results of the drive system operated in the new European driving cycle (NEDC).

2. Dynamic drive simulation

In the present investigation, dynamic drive simulation had the task of providing a control profile for the mass flow controller of reactant supply (methanol and water) for the NEDC. For this purpose, test rig results from investigations into the stationary and dynamic operation of fuel gas production [4] were incorporated into the entire model of a vehicle drive with fuel cells in the form of characteristic curves and time constants of the control system elements. The starting point of the calculations is the simulation model of a vehicle drive according to Fig. 1 [6]. Essential parameters of the vehicle are summarized in Table 2.

In the simulation, the torque or the electrical power input of the electric motor is determined for each point in time of the driving cycle by means of the rated/actual vehicle speed

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Table 1
Technical data of the test rig components

Reformer	
Nominal power (kW$_{th}$)	50
Operational temperature (°C)	260–300
Nominal pressure (bar)	21
Catalytic burner	
Nominal power (kW$_{th}$)	16
Operational temperature (°C)	260–750
Nominal pressure (bar)	1.3
Gas separation membrane	
Nominal permeate flow (l$_N$/h)	800
Operational temperature (°C)	300–350
Nominal pressure (bar)	21
Fuel cell	
Nominal power (kW$_{el}$)	1
Operational temperature (°C)	30–70
Operational pressure (bar)	1.5

Table 2
Selected parameters for the simulation model of a vehicle drive with fuel cells according to Fig. 1

Total vehicle weight (kg)	1.475
Bulkhead area (m^2)	1.98
Drag coefficient	0.3
Rolling resistance coefficient	$f(v)$
Rated capacity of the fuel cell (kW)	65
Maximum H_2 capacity of the reformer (kW)	122

(basis: NEDC). Together with the power requirement of the power electronics and ancillary components, the gross electrical output of the fuel cell system is calculated, with which the hydrogen demand can be determined with known current–voltage characteristic of the fuel cell stack. This demand is covered from the actual fuel gas supply of the fuel gas production system or, because of the latter insufficient dynamics, from the gas storage tank.

The stationary operating performance of the methanol steam reformer is derived from experimentally determined characteristics for methanol conversion and CO content as a function of specific hydrogen production (Fig. 2). The dynamic properties of reactant supply and the tubular reactor were investigated at the test rig by feeding step functions of the reactant supply. The time response thus determined is included in the model in the form of transfer functions. The operational performance of the Pd/Ag membrane is incorporated into the simulation model in accordance with earlier laboratory experiments [4]. An experimentally determined curve is available for modeling the current–voltage characteristic of the fuel cell [3]. The power density is fixed at 2.2 kW/m^2 at nominal load (65 kW$_{el}$).

Performance scaling of the components of the drive is oriented to the maximum hydrogen capacity of the CMR of 61 kW (related to the lower heating value H_l). In order to achieve the capacity range of present-day passenger cars for the entire drive, it is assumed that the CMR represents one of two stages of a future reformer. The maximum hydrogen capacity of the simulated reformer is thus 122 kW (H_l). In addition, the scaling of the drive is selected so that the maximum methanol quantity supplied in the driving cycle (NEDC) corresponds to that of the test rig (12 kg$_{MeOH}$/h). Based on the results of the dynamic analysis of fuel gas production, an operating strategy was selected which, due to the large time constants, exclusively takes the actual filling level of the gas storage tank into account. Within the permissible pressure range from 20 to 40 bar, the control of the reformer, i.e. the supply of methanol and water, is regulated linearly. The maximum amount of methanol–water mixture is correspondingly fed to the reformer when the gas storage pressure reaches the lower limit. The rating of the gas storage system ensures that actual values do not drop below this lower limit even in full load operation.

Fig. 3 shows the control profile for the CMR resulting from the calculations. Since the driving cycle is run in endless operation (restart after 1200 s) each new driving cycle starts with dropping reactant supply in the range from

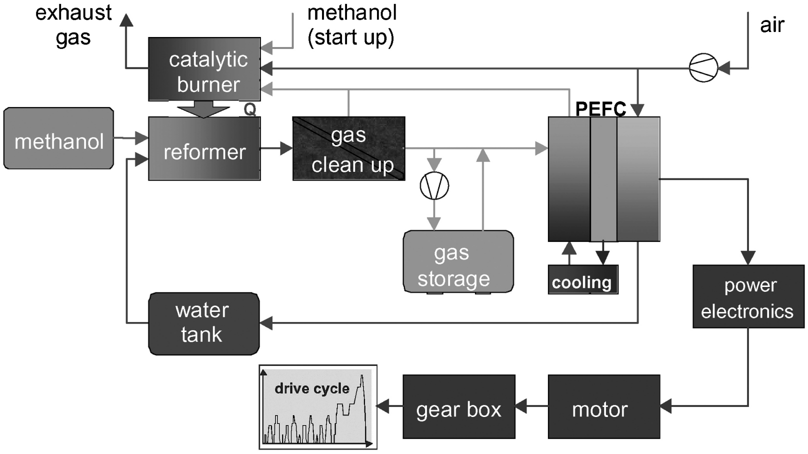

Fig. 1. Fuel cell drive system with H_2 generation and gas storage for the improvement of the dynamic properties (configuration according to simulation model).

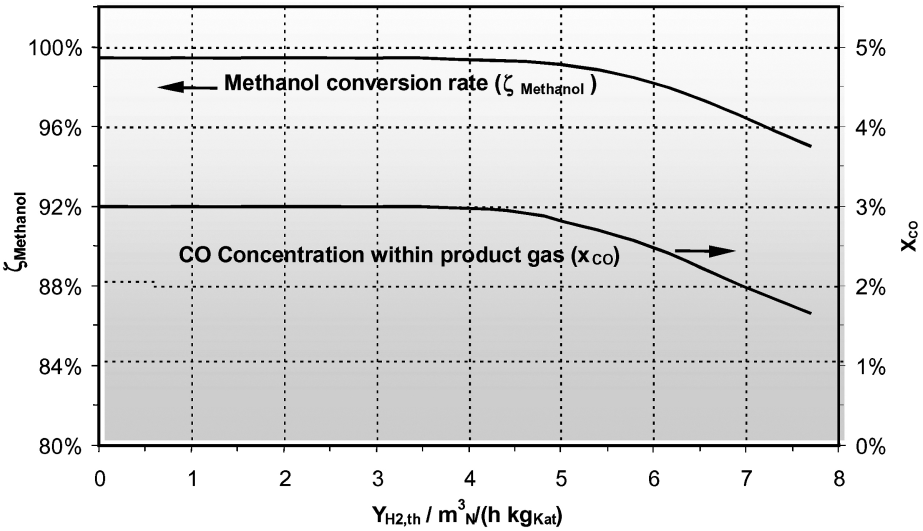

Fig. 2. Characteristic curves for methanol conversion and CO behavior as a function of specific hydrogen production $Y_{H_2,th}$.

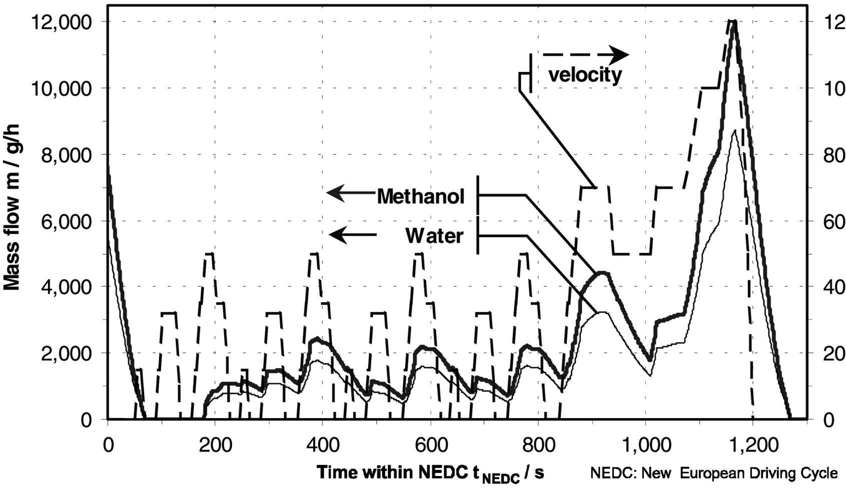

Fig. 3. Control of the reformer preset values of the mass flows of methanol and water as a function of time in the NEDC (length of the driving cycle 1200 s according to old process).

0 to 70 s to recharge the gas storage system. After reaching the upper limit of the gas storage pressure of 40 bar, the reactant supply is interrupted according to the operating strategy. Since, however, due to the large time constants of fuel gas production, hydrogen continues to be produced and must be interim-stored, the gas storage system will be overcharged. As a result, no reactant is supplied between 70 and 180 s. Only after dropping below the upper limit of the gas storage pressure will methanol–water mixture be charged again.

On the whole, the dynamics of the system (here especially of fuel gas production) governs the degree of hybridization of the drive system: large time constants lead to a large storage tank in the design, which means additional weight and costs for the vehicle. On the other hand, the operation of the gas storage system requires an increased demand of electric power for the compression of the hydrogen to be stored. The independence of the control of the fuel gas production system of the actual performance of the fuel cell, however, opens up the possibility of avoiding power ranges with low efficiencies and thus improving the energy balance by the application of an optimized operating strategy. Nevertheless, an improvement of the dynamics of the fuel cell system appears desirable.

3. Experimental results

The profile of the reformer reactant flows shown in Fig. 3 and determined by dynamic simulation was used as a preset value input for activating the mass flow controller in the drive test rig. For the segment of the NEDC representing the load range of long-haul driving, the time response shown in Fig. 4 is obtained for the methanol entering the reformer as

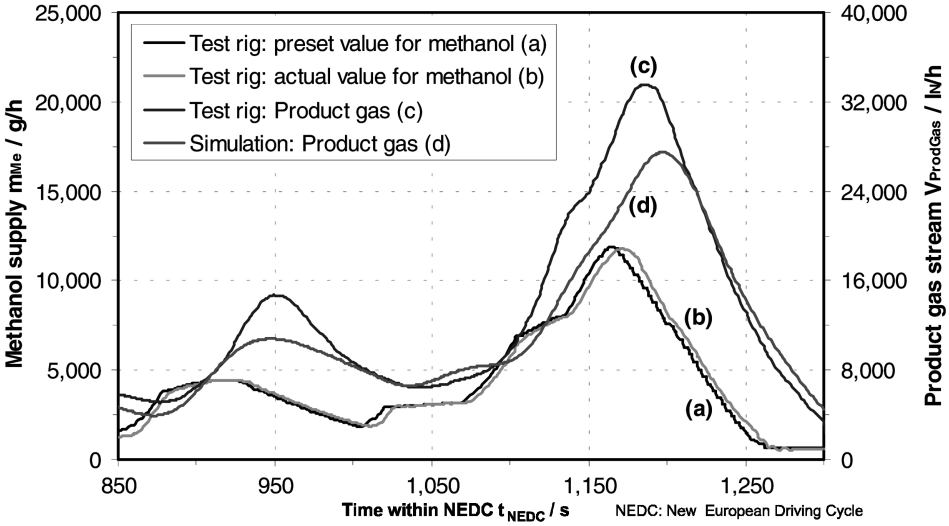

Fig. 4. Time curves of methanol mass and product mass flow for the long-haul part of the NEDC.

well as for the product gas actually produced in the reformer and for the simulated product gas stream. Relative to methanol supply it can be seen that the time lag between preset value feed and adjustment of the desired mass flow can range between 2 and 10 s, depending on the load. This is a consequence of the time constants of the mass flow controller resulting for the respective load steps, so that in a real drive system the flow controller will have to be replaced by a faster porportioning unit. Considering the curve for the product gas stream it is remarkable to note that the maximum flows determined experimentally clearly exceed those of the simulation. This is a consequence of the varying reformer temperature over time in the experiment caused by great load changes. The idealized approach of the model-based simulation, on the other hand, assumes constant temperature over the entire cycle. In the time ranges of low load variations (about 1000–1100 s) and decreasing load (about 1200–1300 s) good agreement can be observed between simulation and experiment.

Important quantities for evaluating the quality of methanol and steam reforming are the methanol conversion rate and the CO fraction contained in the product gas. Fig. 5 shows the variation of these two quantities over the entire NEDC. The conversion rate reaches values between 99.6 and 100% over nearly the entire core range of the driving cycle. In this range the CO concentrations are between 2.7 and 4.6%. The small variation range of the two quantities is a consequence of the moderate load change demands made on the CMR due to control. Both quantities only decrease to the minimum value of 93.5% for methanol conversion and about 0.8% for CO content in the range of full load between 1150 and 1200 s.

Another important parameter for evaluating the performance efficiency of fuel gas production is CMR efficiency. CMR efficiency is defined as the quotient of the energy content of the hydrogen produced (lower heating value) and that of the burnable components supplied to reformer and catalytic burner. The evaluation of these parameters for the entire driving cycle provides a CMR efficiency of 77.3%.

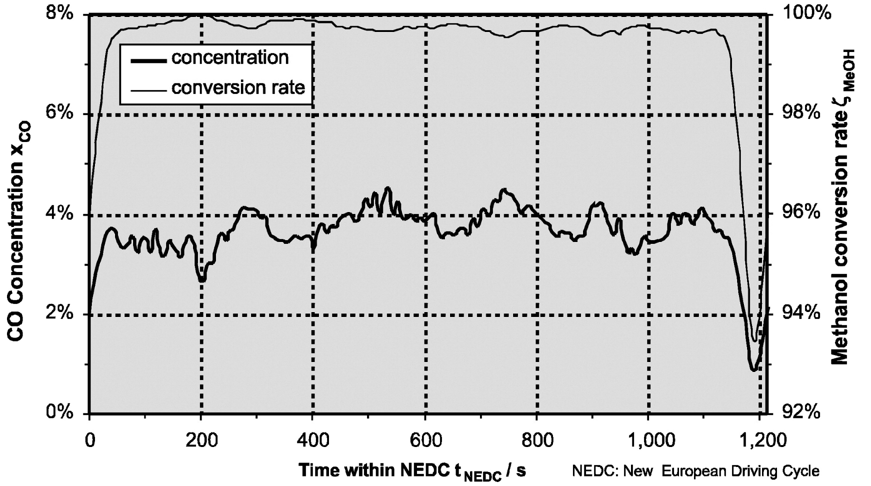

Fig. 5. Time curves of methanol conversion by the CMR and CO concentration in the product gas for the entire NEDC.

Table 3
Pollutant emissions of the catalytic burner and future limit values

Emission	Actual experimental results (mg/km)	Pre-experimental results [7] (mg/km)	SULEV standard (mg/km)
CO	1.8	0.3	625
NO_X	0.3	<0.01	12
UHC	3.2	0.9	6

In this case too, a deviation from the simulation result can be explained by the variation of reformer temperature during the cycle.

In addition to being the process heat source, the catalytic burner of the CMR is also the after burning unit for burnable residual gases arising in the system. In the drive system under consideration, these are the retained fraction of the gas separation membrane and the residual anode gas of the fuel cell. Since the membrane and fuel cell used in the test rig only have a fraction of the capacity of the CMR at their disposal, the residual gas to be fed to the catalytic burner when driving through the NEDC is made available from compressed gas cylinders. The respective time-dependent composition of the mixture was derived from the simulation results. For the catalytic burner fuelled in this way, the relevant off-gas pollutants (CO, NO_X and unburned hydrocarbons, UHC) were continuously measured to assess the off-gas quality. The results of actual measurements are shown in Table 3 together with earlier measurements of a separate burner unit fed with a constant fuel gas composition over the cycle [7] and with the future limit values of the super ultra low emission vehicle (SULEV) standard. The higher pollutant emissions of the actual measurements compared to the pre-experiments are a consequence of the variation of the fuel gas composition. In addition, not yet optimal mixture preparation and distribution could also have had a negative effect on fuel gas conversion. Nevertheless, the specific emission values are clearly below the future limit values, for CO and NO_X even by more than one order of magnitude.

4. Conclusion

The application of dynamic simulation for the description of the time-dependent process flows in a drive system with fuel cell made it possible to operate a fuel cell system with H_2 generation as a test rig according to standardized NEDC requirements. Preset value inputs for controlling the reformer reactants, methanol and water, as well as the residual gases to be fed to the catalytic burner were provided by the simulation result. Cycle operation of the CMR showed good agreement with the simulation results in partial ranges with minor load variations on the basis of the product gas flow. Significant differences are only observed in the case of greatly changing performance requirements. This behavior can be attributed to the variations occurring in reformer temperature. The CO concentrations measured in the product gas and the methanol conversion of the reformer show minor variations over wide ranges of the cycle. The only exception is the full load range at the end of the cycle, which displays the lowest values. A CMR efficiency of 77.3% was determined for the NEDC from the mass flows measured in front of and behind the CMR. The cycle-related pollutant emission values amount to 1.8 mg/km for CO, 0.3 mg/km for NO_X and 3.2 mg/km for UHC and are thus clearly below those of the future SULEV standard.

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