Production and conversion of liquid fuels and hydrogen from biomass and natural gas using microreactor technology

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Abstract

Microreactors have gained attention over the last two decades with respect to process intensification due to their enhanced mass and heat transfer characteristics and also due to their low inventory of reactants when reaction rates are increased. IMVT has identified this potential for energy related applications as early as 1997 and started projects on hydrogen and synthesis gas production. Several methods for catalyst application to microchannel walls have been developed for enabling the conduction of heterogeneously catalysed processes in microreactors.

Within this contribution, an outline of different processes and recent advances is given in the fields of catalyst preparation and lab-reactor construction, hydrogen and synthesis gas production as well as fuel synthesis from synthesis gas, either derived from biomass or stranded/off-shore gas. Additionally, the issues of conducting heterogeneously catalysed exothermic equilibrium reactions such as the water-gas shift for upgrading biomass-derived synthesis gas or methanol synthesis from the synthesis gas are discussed. Scaling up the reactors to pilot scale is addressed.

Introduction

Fuel cells allow the direct conversion of chemicals into electrical energy. Efficiencies above the Carnot efficiency of combustion engines can be reached. However, for low-temperature fuel cells hydrogen is required as feed. The problem so far is the storage of H\textsubscript{2}. Even with optimized metal hydride materials or new types of high pressure or cryogenic liquid tanks, the weight and volume of the entire hydrogen storage system is higher than for conventional fuels [1]. Thus, the use of hydrogen leads to a lower range of vehicles in comparison to an internal combustion engine.

H\textsubscript{2} production from resources in a chemical process on site is also problematic. Since H\textsubscript{2} demand can vary significantly over time, the system components like reformer and heat exchangers have to be able to cope with highly dynamic operation, and a high efficiency is required. This kind of operation is not possible with conventional chemical reactors, as they are optimized for steady state operation.

The Institute for Micro Process Engineering (IMVT) develops microstructured devices, which consist of several thousands of microchannels in metals since 1989. With the help of nanostructured catalyst layers on the channel walls these devices can be used as reactors.
Due to the small distance between the microchannels, the temperature of the hydrogen production system can be established within seconds. Extremely high amounts of heat and mass can be transferred in a short time (see Fig. 1). Thus, the equipment size, weight and energy losses can be reduced to a level that the overall efficiency of the system for generating of H₂ can almost reach the theoretical efficiency [2,3]. Current research addresses also the integration of Palladium membranes directly into the microreactor for hydrogen production. This approach may allow reduced mass transport limitations to the membrane, which should remove pure hydrogen. The reforming reaction, on the other hand, benefits from the removal of hydrogen since hydrogen is suppressing the reaction rate on the catalyst surface and reduces the maximum conversion due to the thermodynamic equilibrium between reactants and products.

Direct coupling of an exothermic reaction to the endothermic hydrogen production is favourable for the design of the H₂ production system. Heating of reformers via externally generated hot flue gas by homogeneous combustion is not feasible, since the hot gas temperature must be lowered by additional air dilution to a level below 1000 °C to avoid damaging the metallic microstructure. An important aspect is the formation of coke at specific reaction temperatures. However, for the production of clean hydrogen in microreactors a targeted "plugging" of microsystems with coke seems feasible.

![Fig. 1](image)

**Fig. 1:** Schematic of microchanneled plates in which reactions can be conducted nearly isothermal due to low heat transfer resistances (left) and schematic of catalyst coating on top of such microchannel walls possessing only a small temperature gradient ($\Delta T < 1 \text{ K}$) for a strongly exothermic reaction due to fast heat removal to the cooling fluid (right).

The second energy related application, which will be of great importance in the future, is the conversion of synthesis gas (“syngas”) into liquid fuels. With around 25% of world’s proven natural gas reserves located in remote areas offshore, conversion of so called “stranded gas” into transportable fuels and chemicals calls for new technologies. An analysis of satellite data, from which the amount of flared gas can be calculated, shows that roughly 0.04 m³ of natural gas is accompanied with 1 litre of crude oil [4]. The Fischer-Tropsch synthesis is one commercial pathway for the conversion of natural gas to
transportation fuels. However, for offshore production, the conversion technology should be compact, modular and light-weight. Microstructured reactors enable isothermal operation of the highly exothermic Fischer-Tropsch reaction (165 kJ/mol) which creates a variety of linear long-chained hydrocarbons. Hence, microstructured reactors could be especially suitable for highly active Fischer-Tropsch catalysts and product shaping. Taking into account some rough estimates from Life Cycle Assessment, calculations at KIT show that for an average off-shore platform the energy output can be increased by 2 % and that the CO₂ off-shore emission can be reduced to 40 % by such a compact process.

A highly efficient generation of fuels and platform chemicals from biomass with respect to energy balance and resources is necessary to avoid competition of fuel and food. Second generation biofuels and biochemicals, i.e. the conversion of gasified biomass to fuels and chemicals, would in principle enable a much higher biomass utilisation compared to processes like fermentation of corn. However, product shaping in the synthesis is again difficult due to mass transfer limitations and hot spots occurring in conventional reactors. Therefore new developments at IMVT comprise also systems for methanol synthesis, direct di-methyl ether synthesis and high temperature / high pressure shift reactions. The latter reaction can be applied for conditioning of the created syngas for further synthesis of fuels or chemicals.

A specific point which is addressed by IMVT for the latter type of reactions is that most studies in literature refer to just keeping the reactor temperature constant and thus enabling isothermal operation conditions. For exothermic equilibrium reactions, such as the water-gas shift or methanol synthesis, this is, however, not the optimum temperature profile. The first reaction may be necessary for the biomass conversion into fuels since dry biomass gasification yields a low H₂/CO ratio in the synthesis gas. The initial reaction rate is mainly influenced by the Arrhenius dependence and thus high reaction temperatures are favourable. With increasing conversion, approaching to equilibrium, however, will reduce the rate considerably and a lower reaction temperature should be employed. The maximum rate r as a function of the conversion X can be found by setting \[
\frac{dr(X)}{dT} = 0.
\]

By using counter-current cooling in microreactors, the optimum falling temperature profile may be approximated in an effective way. Laboratory microreactors are usually small in size, i.e. channel length is below 10 cm, so heat conduction in the microstructure may significantly contribute to the heat flux along the reaction zone. If this axial heat flux is within the range of the heat to be transferred, the desired temperature gradients may be significantly reduced. On the contrary, the conductive heat flux in the microstructure can be used on purpose to establish a certain temperature profile. Due to the fact that the heat production rate is usually decreasing with increasing conversion, heat removal by effective cooling may be higher than desired at the reactor outlet.

**Catalyst introduction and lab-reactor construction**

Over the last decades lot of effort has been invested into the development of methods to apply catalyst into microstructures or onto microchannel walls. Two reviews show the technology progress [5,6] and the difficulties and considerations which arise in individual chemical processes are described in a recent book chapter [7].
Two main principles are applied today: the introduction of fine powder catalyst in slit like arrangements, so called micro fixed bed or micro packed bed, and the application of catalyst coatings to microchannel walls. A SEM of the applied structures which can be filled is shown in Fig. 2 together with an image of a catalyst filling, taken through a glass window on top of the structure (normally a metal plate is on top of the pillars to form a slit). Examples from recent work at IMVT with regard to catalyst coating development are shown in Figure 3. Main objectives of the catalyst integration are high surface area, high activity of the catalytic active species, and improved adhesion on the wall amongst several other issues [7].

Fig. 2: SEM of a microstructured plate for catalyst powder integration (left) and image of a catalyst filling through a glass (right side).

Fig. 3: SEMs of microchanneled plates with different catalyst coatings.

Two types of lab-scale reactors are currently used at IMVT in the application field of hydrogen or fuel synthesis from biomass or remote natural gas. One type refers to integration of catalyst powder but is operated in cross flow with heat exchange fluid to operate at strictly isothermal conditions. The other type of lab-reactor can either be operated
with catalyst powder filling or with coated microchannel plates. This reactor allows exchange of the microstructure and is heated by cartridges and a temperature profile can be superposed along the channels to optimize local reaction rates.

**Coupling Exothermic and Endothermic Reactions for H₂-Generation**

*Figure 4: Pressure drop calculation for a steam reformer at different mass flows of hot flue gas and component height as a function of number structured metal plates.*

The process of steam reforming of natural gas or other hydrocarbons either derived from fossil or biomass resources is endothermic. Heat must be efficiently introduced into the reactor system to facilitate the reaction and in order to generate a high net energy output with regard to the overall system including the fuel cell for electricity generation. Microreactors are proven to possess extraordinary heat exchange capabilities, but this heat transfer is accompanied with higher pressure drop. Figure 4 shows the dependence of the pressure loss and the stack height - equivalent to the size of the reactor - depending on the number of microstructured metal sheets for a gasoline reformer with a flue gas heating (homogeneous combustion of gasoline) to produce a hydrogen-rich gas. The calculation takes into account full conversion for reforming. The two different flue gas fluxes (38.5 and 12.5 kg / h) for the pressure drop calculation are equivalent to different reformer inlet temperatures (850 °C and 1100 °C) and temperature differences between inlet and outlet of the flue gas, respectively. With a low mass flow of flue gas, equivalent to a very high inlet temperature of the flue gas and a possible damage of the microstructure, a pressure drop boundary condition of 30 mbar ("maximum") can be achieved with a number of 70 metal sheets. On the other hand, with a higher flux the boundary conditions for the maximum size of the reformer (less than 20 cm stacking height) cannot be obtained.

*Fig. 4: Pressure drop calculation for a steam reformer at different mass flows of hot flue gas and component height as a function of number structured metal plates.*
Due to the problem of optimization of the pressure loss in combination with the flue gas temperature, a direct coupling of an exothermic reaction with the endothermic steps in hydrogen production in microreactors is a preferred heat integration method. A total oxidation could be carried out at nearly-stoichiometric condition without a temperature maximum as occurring with homogeneous combustion.

A schematic of the overall process for the production of H\textsubscript{2} for fuel cells from liquid hydrocarbon mixtures such as Diesel under the provision of a combined endo- and exothermic process for steam reforming is shown in Figure 5. The fuel is mixed with evaporated water and converted in the reformer into a hydrogen rich gas. The reformate is cooled, and is cleaned, for example, by shift reaction and selective oxidation to remove carbon monoxide (CO). The cleaned gas is used for the production of electrical energy in a PEM fuel cell (PEFC). The residual H\textsubscript{2} in the fuel cell off-gas can be burnt and used for heating and evaporation of the reactant flows or to meet the energy demand of the steam reforming.

Taking into account the efficiency of the individual steps yields an overall efficiency, including that of the fuel cell, of 22%. This is significantly higher than for a system of diesel engine and generator [8] with 4.5%.

However, in order to perform such a coupling of endothermic and exothermic reactions, the system must be optimized for operation with the evaporator and the reformer. Because of the different reaction rates of combustion of hydrogen (the anode exhaust of the fuel cell included) and reforming, this is not trivial. To start a system from ambient temperature requires a very active noble metal catalyst. With such a catalyst the hydrogen conversion can be very fast at final operating temperature of the reformer, i.e. in a few millimeters of microchannel length full conversion can be reached at a residence of only 3 ms. Steam reforming, which requires according to our experience 50 to 300 ms residence time [9,10], possesses a fundamentally different heat demand. To balance heat supply and demand, hydrogen can be mixed with air in sections. To be able to balance heat generation and consumption by this procedure, kinetic models are needed. Power law models or

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**Fig. 5**: Flowchart of the overall system for generating electrical energy with direct thermal coupling of combustion and evaporation and condensation on the example of diesel steam reforming.
elementary step models are produced in collaboration with other research groups. Isothermally designed experiments in test reactors where the catalyst layers are produced on exchangeable microstructured sheets serve for the verification of the models [11,12]. An example of such a microstructured diesel reformer with integrated catalytic combustion and its characteristics was previously published [13].

In addition to maximizing the volume fraction of catalyst, an important aspect is the optimization of the catalyst productivity. The mass ratio of catalyst and reactor volume or weight of microstructure reactors is rather low [14]. IMVT already achieved values for the catalyst productivity of up to 40 g\textsubscript{hydrocarbon} / (g\textsubscript{cat} h). The productivity and the volume fraction ultimately decide on a minimization of system weight, the ratio m\textsubscript{steel} / F\textsubscript{in}, i.e. the mass of steel from the reactor in relation to a given hydrocarbon flux. This ratio should be in the range of about 5 g\textsubscript{steel} h / g\textsubscript{hydrocarbon}.

![Image](image.jpg)

**Fig. 6:** Hydrogen distribution holes with increasing spacing to approximate the heat demand of methanol steam reforming

Figure 6 shows, based on modeling, the distribution of interconnecting holes for hydrogen transfer into the channel for the conduction of the exothermic reaction. In this design the consideration of the pressure loss in the hydrogen distribution channels below the transfer holes and the pressure loss in the combustion channel in relation to the pressure loss in each transfer hole is important. The intended flow per transfer hole should be equal. An alternative to the design shown by etching is the use of even smaller holes made by laser structuring. Examples of such reactor designs can be found in literature [15]. Since the lab–scale systems either combine reforming or evaporation and heat supply, the heat losses may, depending on the temperature level of the process, be between negligible [15] and dominant [13,16].
Example: pure hydrogen from cracking propane

The opinion prevails that coking or hydrcarbon cracking reactions are not compatible - in the sense of avoiding solids and associated plugging - with microsystems. However, coke formation was only observed in substantial amounts in steam reforming of diesel and ethanol, although this has not led to blockage of the channels. This is even valid for situations when the vapor dosage failed during the experiments. It appeared that in this context, the stability of the catalyst and the choice of the reactor material is more important. For example, systems comprising alumina were irreversible damaged in absence of steam, whereas cerium oxide layers showed no reduction in catalyst activity. This was attributed to the reported cerium oxide oxygen storage effect [11,12]. A complete regeneration of the cerium-based catalyst systems was possible by short-term oxygen treatment at reforming temperature. Nicrofer 3220H or the American Alloy 800 H show little catalytic function to form coke and low adherence of coke, while stainless steel like 1.4404 well promotes coke formation.

Based on this experience, a system was built to be used specifically for the cracking reactions of propane for hydrogen production. An advantage of this technology was that the reaction product almost exclusively consisted of hydrogen gas. The catalyst system was a vanadium oxide coated stainless steel net integrated in a Nicrofer reactor. The stainless steel catalyzes an initial coke formation and coke itself becomes the active catalyst. For a permanent production of hydrogen, regeneration of the catalyst is required, i.e. at least two separate reaction zones for cracking and regeneration are needed. These zones have to be alternatively switched. For additional warming-up and maintaining the operating temperature, combustion of the fuel cell anode exhaust gas or of the regeneration exhaust gas is necessary. The exhaust gas from regeneration is mainly containing carbon monoxide. In principle, the following two reaction equations are valid for cracking (1) and regeneration (2).

\[
\begin{align*}
\text{C}_3\text{H}_8 & = 4 \text{H}_2 + 3 \text{C} & \Delta H_R & = + 104 \text{ kJ/mol C}_3\text{H}_8 \\
3 \text{C} + 1.5 \text{O}_2 & = 3 \text{CO} & \Delta H_R & = 3 \times (- 110.5 \text{ kJ/mol C})
\end{align*}
\]

Figure 7 shows the basic structure of the developed apparatus and a photograph of the system which was designed for a flow rate of 3 lN / min. The reactor contains alternating stacks of four pieces of steel net and two microstructured plates for the dosing of combustion air into the combustion channels. The orientation of the steel net changes after each combustion zone by 90° so that two passages, one for regeneration and one for cracking are formed.

Currently, the time requirements for regeneration and for cracking are different. The regeneration phase is about five times longer than the cracking. In addition, the required length of the steel net is significantly shorter, i.e. carbon deposits are found only at the entrance. Thus the total number of required passages and switching actions are higher. It is necessary in the current design to burn considerable amounts of anode off-gas to maintain the temperature of the system due to the different time scales of regeneration and cracking (for details see [17]). The design for such reactors has to be modified in the future.
Integration of membranes for in-situ hydrogen separation while reforming

The reforming reaction produces a hydrogen-rich gas containing also CO, which has to be removed in several steps (see Figure 5) in order not to poison the PEM fuel cell while feeding the hydrogen. An alternative to several clean-up processes like water gas shift, selective oxidation or methanation, is the application of a dense palladium membrane. To provide a high flux of hydrogen over the membrane a high pressure gradient and a thin membrane are necessary. Applying high pressure, however, shifts the equilibrium of the reforming reaction to the reactants and full conversion is not possible, and the membranes get easily damaged. By integrating the membrane into the microreactor these disadvantages can be avoided. The membrane can be easily supported in the small channels with regard to pressure cracks and the reaction can be facilitated through in-situ removal of hydrogen. Three different approaches are currently addressed at IMVT:

a) Integration of palladium thin foils directly between metal microchannels (for T < 400°C)

b) Integration of palladium thin foils on a porous metal support with ceramic porous diffusion barrier layer (prevents diffusion of steel components into Pd or vice versa) on top of microchannels (for T > 400°C)

c) Preparation of palladium thin films on a porous metal support with ceramic porous diffusion barrier layer on top of microchannels (for T > 400°C)

For approach (a) the steps from the microstructured plates and the membrane, to a laser welded membrane assembly, and the module in which the membrane assembly gets tested are shown in Figure 8.
Examples from BtL Technology

For the production of liquid synfuels from biomass feedstocks (BtL) and low-hydrogen fossil raw materials, a water gas shift reaction step carried out at high pressure and temperature may be advantageous for minimizing energy losses. If gasification yields an insufficient H₂/CO ratio for fuel synthesis and high pressure can be applied, subsequent gas cleaning and water gas shift should be carried out at elevated pressure and above the temperature level of the synthesis.

For the laboratory tests a new microstructured reactor design was built, which allows testing of catalysts powder in a micro fixed bed or as sheets with channel wall catalyst coating at elevated pressure and temperature (up to 50 bars, 600°C). Wall coated catalysts of 5 wt.% Pt/CeO₂, Pt/CeO₂/Al₂O₃ (Fig. 9) and 5 wt.% Ru/ZrO₂ have been produced by incipient wetness impregnation and sol-gel technology.

Fig. 9: Scanning Electron Microscopy image of the applied Pt/CeO₂/Al₂O₃ coating on 200 x 200 μm microchannels
The platinum catalyst systems were shown to be highly active under operating conditions above 400°C, and equilibrium yields of CO₂ can be reached at 500 to 600°C. At elevated pressure, the reaction rate increases due to increased partial pressure of the reactants (Fig. 10). Negligible hydrocarbon product concentrations such as methane were found which would be thermodynamically attractive under the process conditions [18]. Also coke formation happened in the microreactor inlet which has been found to be formed in the gas phase during heating up the gas mixture. The Ruthenium catalyst system enables high reaction rates below 400°C (see also [19]).

Fig. 10: Measured yields of CO₂ compared to equilibrium values for the Pt/CeO₂/Al₂O₃ coating at different pressures, modified residence time of 4e-4 g*min/cm³, and a feed composition of 32% CO, 10% CO₂, 18% H₂, 10% N₂, 30% H₂O

In the conversion of syngas to methanol, the applied pressure is usually around 50-100 bar, since the thermodynamic equilibrium is unfavourable at lower pressure. In terms of the chemical equilibrium shift, an option to methanol separation arises, by simultaneous conversion of methanol to di-methyl ether (DME). This one-step process for DME synthesis is thermodynamically favourable, and a higher CO conversion rate can be achieved over hybrid catalysts, even operating at lower pressures and higher temperatures than a two step process. However, there is a clear increase in the potential for thermal runaway of the catalyst bed due to the lower methanol partial pressure and the associated CO conversion rate increase. The application of microstructured reactors would reduce this risk considerably, and thus process intensification with optimized process conditions seems possible. The advantages may be useful for compact GTL (gas-to-liquid) or remote BTL (biomass-to-liquid) applications.

Various methods have been investigated for the deposition of a mixture of the methanol synthesis catalyst and the dehydration catalyst. For the preparation of the methanol synthesis catalyst Cu/ZnO/Al₂O₃ a method of stepwise co-precipitation of the metal nitrates was followed at elevated temperature of 65°C [20]. For the dehydration
catalyst direct Al₂O₃ deposition was performed by a sol-gel synthesis developed at IMVT [21], and alternatively H-ZSM5 catalysts were produced by hydrothermal synthesis.

From XRD it was found that the alumina prepared by sol-gel and calcined at different temperatures showed no transition from the amorphous state into gamma alumina up to 700°C, but was converted into the delta and alpha phase at 800 and 1000°C respectively. This is in agreement with ammonia desorption experiments where also negligible acidic function was found. The HZSM-5, created by growth on silicalite seeds, exhibited good crystallinity and reasonable acidity but its growth on stainless steel plates was much more difficult. Almost homogenous growth of the zeolites has been reached by initial sol-gel coating, followed by silicalite seeding and hydrothermal synthesis of the H-ZSM5 (Fig. 3). For the copper catalyst system compositions of 65/25/10 wt.% of CuO/ZnO/Al₂O₃ were obtained with BET surface areas of approximately 125 m²/g.

In the case of Fischer-Tropsch synthesis (FT) it was demonstrated by IMVT and collaboration partners that very active catalysts can be operated under conditions, which can only be applied in conventional reaction engineering equipment under 1:20 dilution ratio of the catalyst to effectively remove the reaction heat [22]. Since in some cases a by-product formation on the microchannel walls can be observed due to inherent high surface area to volume ratio, an investigation on wall influences on the reaction performance is often necessary. For the FT application absolutely no influence of the wall material was observed. Neither the product distribution nor the deactivation of the catalyst was modified. While showing that the pressure drop over the applied packed bed catalyst is reasonably low, even for the small particles sizes of 50-150 μm and several meters of bed length, the huge potential of the technology becomes obvious. Taking into account the same catalyst loading for the applied microsystem as for conventional reaction engineering equipment it becomes clear that over 90% size reduction is feasible and compact systems can be built.

### Shaping reaction rates for exothermic equilibrium reactions

As stated in the introduction, microreactors can offer more than just enabling isothermal conditions. For exothermic equilibrium reactions a decreasing temperature profile would be advantageous for following the optimum trajectory of the reaction rate in the conversion-temperature plot. However, too effective cooling in counter-current mode could lead to quenching of the reaction - starting from the reactor outlet.

To avoid such quenching, a more detailed investigation of the importance of the axial heat flux along the reactor is needed. Investigating a stack of multiple microstructured foils for the reaction and only one cooling foil (asymmetric stacking), the effect of quenching can be reduced. A semi-porous body model indicates that an increase of the foil ratio (reaction vs. cooling) increases the axial temperature gradients, while the temperature of the stack of reaction foils in such an arrangement remains almost constant at any axial position in the direction of the heat transfer.

In the water gas shift reaction, a kinetic rate expression for a copper catalyst system [23] with the pre-exponential factor adjusted to obtain conversions close to experimental results was applied at IMVT so far. The model suggests that the expected conversion for a linear decrease of temperature is above the conversion for isothermal operation both for low and high operating temperature (Fig. 11).
Fig. 11: Calculated conversion for isothermal operation at low and high temperature and for a linear falling temperature profile for the water gas shift reaction.

Scaling up microreactor technology to pilot operation

The microreactor interior channels and structures have to be reasonably small to obtain high heat and mass transfer capabilities. However, the outer size of the reactors doesn’t necessarily have to be small – microchannels or microstructures can be multiplied to a larger stack with more plates and more channels per plate. In previous projects on liquid phase reactions IMVT was able to show that a reactor for 1.5 t/h of throughput can be built and operated [24]. For heterogeneously catalysed gas phase processes like the fuel production from syngas, the different density of the fluid creates the need for up-scaling the outer size of the microreactors even further. Lab-scale reactors have been built in the range of 2 g of catalyst and 200 g of catalyst per reactor have been applied for technical scale. 2011 an even larger diffusion bonding oven for fabrication of pilot scale reactors has been installed at IMVT. A scale with 7 to 30 kg catalyst seems feasible in one microchannel plate stack. This turns out to be large enough to produce at least 10 kg/h of FT product within one module. A reactor system for water gas shift has been built for 5 m³/h throughput of syngas, which will be operated with biomass-derived syngas from an entrained flow gasifier in the near future.

Conclusions

The current contribution shows that microreactor technology has great potential for energy applications. Two major applications are in the focus at IMVT: flexible, small scale hydrogen production for fuel cell applications and compact systems fuel synthesis, either from biomass or off-shore natural gas. The demand for compact systems in both applications may only be fulfilled by advanced catalyst design enabling high reaction rates and by optimized temperature profiles in the reactor. The latter is of high importance for
exothermic equilibrium reactions, such as methanol synthesis and water gas shift reaction. Finally, by expanded facilities for fabrication, a great potential for demonstration of the technology on pilot scale is available.

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(von dort gelangt man zum Archiv des AKE) eingesehen werden. Allen, die zu diesem Sammelband beigetragen haben, sei an dieser Stelle sehr herzlich gedankt.

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Hardo Bruhns