Experiment and model results of a first bidirectional active temperature control module with metal hydrides for a fuel cell

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Abstract. Many temperature control techniques are based on active heating and passive cooling and thus have a lack of active bidirectional controllability. Whereas, several applications need a precise bidirectional temperature control. The present study demonstrates a novel bidirectional and active temperature control unit (TCU). This unit heats and cools a fluid with the reversible reaction between metals and hydrogen. The reaction of metal hydrides is characterized by a pressure temperature correlation. Hence, the thermal power and the temperature can be regulated by the gas pressure in the TCU. This enables temperature controllers with the pressure as actuating variable on the metal hydride. In this study, an existing $LaNi_5$ reactor is modelled as 0D model. A PID controller is implemented in the simulation and in the experimental setup to stabilize the temperature with the regulation of the pressure in the reactor. Results from experiment and simulation show the stabilization of the temperature.

1. Introduction

Plenty applications and mechanisms are known to stabilize or control the temperature with thermoelectric, magnetic, phase change or thermochemical effects. Until now many of the applications lack an active and bidirectional control [1-3]. Active implies it is changeable and directly implementable, whereas bidirectional implies that it can switch between heating and cooling. One example of the lack of active and bidirectional control is the common liquid cooling of e.g. a fuel cell, which uses the ambient as heat sink [2].

In the present contribution we present a novel bidirectional and active temperature control unit (TCU) based on metal hydrides reacting with hydrogen, thus it is especially suitable for hydrogen applications such as fuel cells (FC). Metal hydrides are a compound of specific metals and hydrogen. The thermochemical reaction of metal hydrides is a reversible gas solid reaction as given in equation (1), where MH stands for metal hydrides, ΔH_R is the reaction enthalpy, x and y stand for arbitrary integers. The absorption of hydrogen is exothermal and the desorption endothermal, thus the reaction can be used to heat or cool its environment. Furthermore, ab- and desorption depend on pressure and temperature, which enables to control the temperature by adjusting the pressure.

$$MH_x + \frac{1}{2}yH_2 \leftrightarrows MH_{x+y} + \Delta H_R \tag{1}$$

In the present TCU, the metal alloy $LaNi_5$ is used. Figure 1 illustrates the principal of the TCU with the example of a FC. On the left side the heating and on the right side the cooling mechanism is shown. A heat transfer fluid (HTF) passes through the TCU, which is also connected to the hydrogen supply and the FC. The hydrogen flows in or out of the TCU to ab- or desorb depending on the fluid inlet temperature and thus the required cooling or heating process.
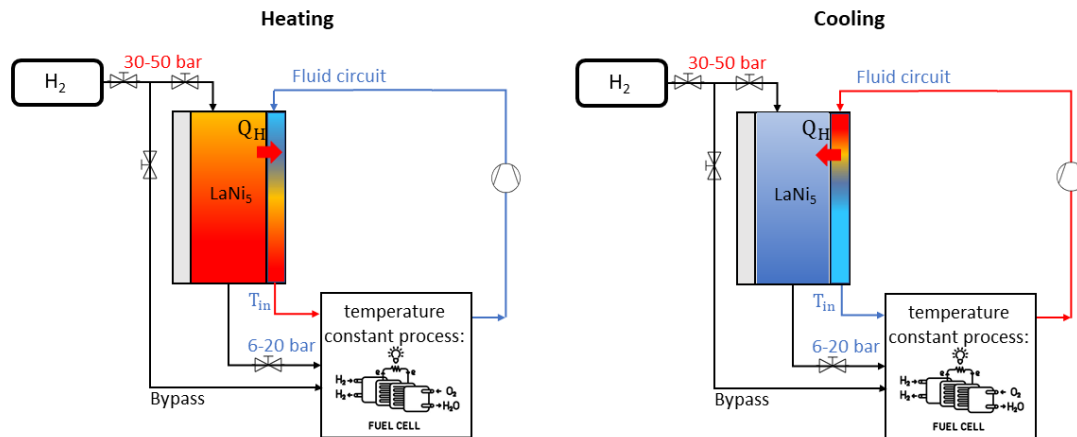
**Figure 1.** Schematic TCU approach in a fuel cell cooling circuit.

2. Experimental Set Up

The experiment for the TCU is designed to yield data to validate the model of the reactor and calculate the controller parameters. Furthermore, experiments with the TCU extend the knowledge about the reaction kinetic and heat exchange in the designed unit. The temperature of the HTF, here water glycol, can be controlled by a thermostat bath. Thereby, two different experiment settings are possible: one with an isothermal HTF inlet flow temperature to validate the model, another with a temperature deflection of the HTF inlet flow as disturbance to test the control loop.

During the experiment thermocouples measure the temperature of the water glycol before and after the reactor. An additional thermocouple records the temperature inside the $LaNi_5$ reactor. A pressure sensor is located in the tubing to the reactor measuring the hydrogen pressure for the reaction. Figure 2 shows the reactor on the right hand-side and the experimental setup on the left. In between the three tubes of the reactor and the shell flows the HTF. The metal hydride is placed inside the three tubes between their fins. Hydrogen passes through the filters to the metal hydride or vice versa.

The first experiments are full load absorptions and desorptions. The temperature of the HTF inlet flow is 45°C and the ab-and desorption pressure are 8 bar and 1 bar for the isothermal HTF inlet flow experiments, respectively. Additionally, part load experiments with two consecutive ab-and desorption

of ca. 40% loading were repeated with an implemented PID controller and a temperature disturbance of the HTF inlet flow.
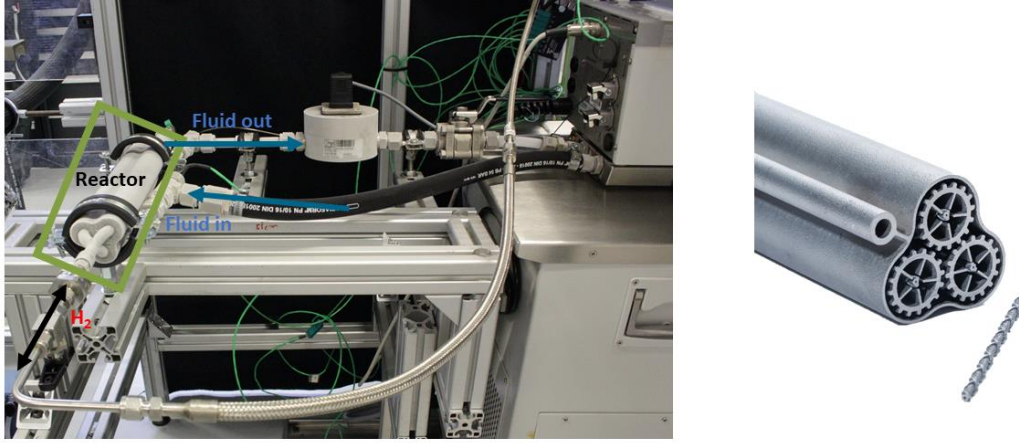
Figure 2. Experimental set up with reactor and thermostat bath on the left hand-side and on the right hand-side the cross-sectional view of the reactor.

3. Simulation model
A mathematical model has been developed to describe the reaction kinetic and the heat transfer of the TCU based on energy and mass balances. Subsequently, a controller system is implemented in the model to simulate the adaptation of the hydrogen pressure depending on the inlet temperature to stabilize the outlet temperature.

3.1. Reactor Model description
The reactor is simplified as a lumped 0D-model. Figure 3 shows schematically the reactor's heat and mass flows. Based on the boundaries of Figure 3, the balance of the cooling fluid is derived from the energy balance of Equation (2). Hereby, T_{HTF} represents the outlet temperature, $T_{\text{HTF,in}}(t)$ the inlet temperature, ρ_{HTF} is the density, $c_{p,\text{HTF}}$ is the specific heat capacity, V_{HTF} is the Volume and $\dot{m}_{\text{HTF}}$ the mass flow of the HTF. kA refers to the heat transfer coefficient between metal hydride and HTF.

$$\frac{dT_{\text{HTF}}}{dt} = \frac{1}{\rho_{\text{HTF}} c_{p,\text{HTF}} V_{\text{HTF}}} \left[kA\Delta T_{\text{MH,HTF}} + \dot{m}_{\text{HTF}} c_{p,\text{HTF}} \left(T_{\text{HTF,in}}(t) - T_{\text{HTF}} \right) \right] \qquad (2)$$

The energy balance for the metal hydride in Equation (3) describes the temperature change in the metal hydride (MH). m_{MH} is the mass and $c_{v,\text{MH}}$ is the isochore specific heat capacity of the metal hydride. $\Delta T_{\text{MH,HTF}}$ is the average logarithmic temperature difference between metal hydride and HTF. Furthermore, x is the conversion rate of the metal hydride and M_{H_2} the molar mass of hydrogen.

$$\frac{dT_{\text{MH}}}{dt} = \frac{1}{m_{\text{MH}} c_{v,\text{MH}}} \left(-kA\Delta T_{\text{MH,HTF}} + \frac{\partial x}{\partial t} \frac{m_{\text{MH}}}{100 M_{\text{H}_2}} \Delta H_{\text{R}} \right) \qquad (3)$$

Furthermore, the dynamic of the metal hydride reaction by pressure or temperature change is defined by a kinetic equation as in Equation (4) with K_{Ar} as Arrhenius factor and E_{A} as the activation energy of the metal hydride [4]. p_{G} is the pressure of the reaction and p_{eq} is the equilibrium pressure for the reaction at a specific metal hydride temperature.

$$\frac{dx}{dt} = \begin{cases} K_{Ar}\exp\left(\dfrac{-E_A}{RT_{MH}}\right)\ln\left(\dfrac{p_{GR}}{p_{eq}(T_{MH})}\right)(x_{max} - x), & \text{Absorption} \\[3mm] K_{Ar}\exp\left(\dfrac{-E_A}{RT_{MH}}\right)\dfrac{p_{GR} - p_{eq}(T_{MH})}{p_{eq}(T_{MH})}x, & \text{Desorption} \end{cases} \tag{4}$$

The required equilibrium pressure is calculated with the Van't Hoff equation extended by an additional term for the dependency of the metal hydride's loading state [5],[6].

The parameter kA was roughly calculated with the reactor's geometry and material properties. The parameter K_{Ar} was fitted on experimental data of isothermal full ab-and desorption.
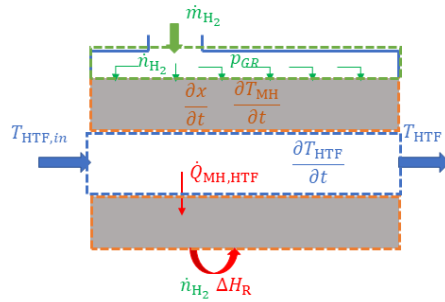

Figure 3. Schematic heat and mass transfers in the metal hydride reactor.

3.2. Controller description

The controller consists of two control loops: an outer control loop adjusting the hydrogen pressure and an inner control loop adjusting the hydrogen flow according to the required pressure as shown in Figure 4. The outer control loop consists of one PID controller, whose input is the temperature difference between outlet temperature of the HTF and setpoint. The manipulated variable is the demanded reactor pressure to reach the desired value of the metal hydride temperature for heating or cooling the HTF to the setpoint temperature. The inner control loop consists of two separate PID controller, one for increasing and one for decreasing the gas pressure in the reactor by control of the inlet and outlet hydrogen mass flow rate. Switching between the inlet or outlet PID controller depends on the manipulated variable of the outer control loop, the hydrogen pressure. If the manipulated variable of the outer control loop is higher than the reactor pressure, the inlet controller will adjust the inflow of the hydrogen and thus the reactor pressure and vice-versa.

The PID controller parameters of the outer control loop are optimized with the combined model of reactor and controller. The optimized parameters were implemented in the experimental setup. In contrast, the parameters of the PID controllers of the inner control loop are set to the default parameters, as the exact controller behaviour are unknown in the experimental setup due to the unknown regulation from pressure to hydrogen mass flow as manipulated variable.
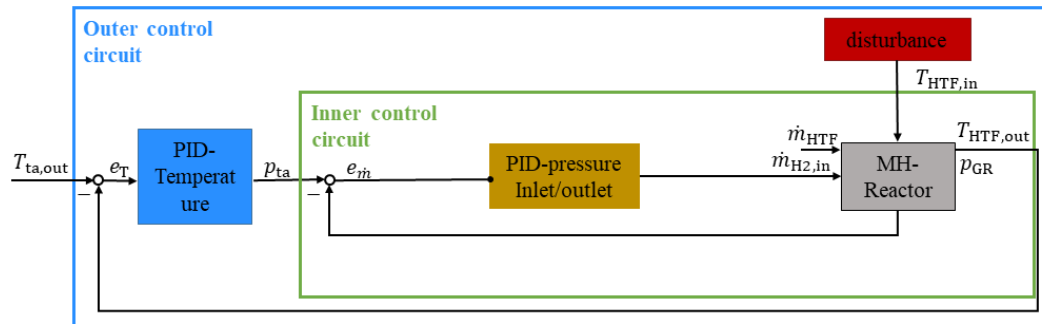
Figure 4. Schematic control loop for the TCU.

4. Results

In the following subsection the reactor model is validated with experimental data. Moreover, the simulation and experimental results with the control loop during part loadings with temperature disturbance are compared and analysed.

4.1. Validation of the model

Figure 5 shows on the left-hand side the loading of the metal hydride. On the right-hand side the heat flow from metal hydride to HTF over time for complete ab-and desorption experiments with constant temperature at 45° C is shown. Both graphs illustrate two reproduced experiments as line and the simulation's results of the experiments as dots. The loading of the metal hydride is calculated using a mass balance of the incoming hydrogen, whereas the heat flow is calculated using the energy balance. Accordingly, the heat flow and the loading of the metal hydrides are independent results. The dotted and the line graphs are on both sides almost identical, thus the simulation is considered to be validated.
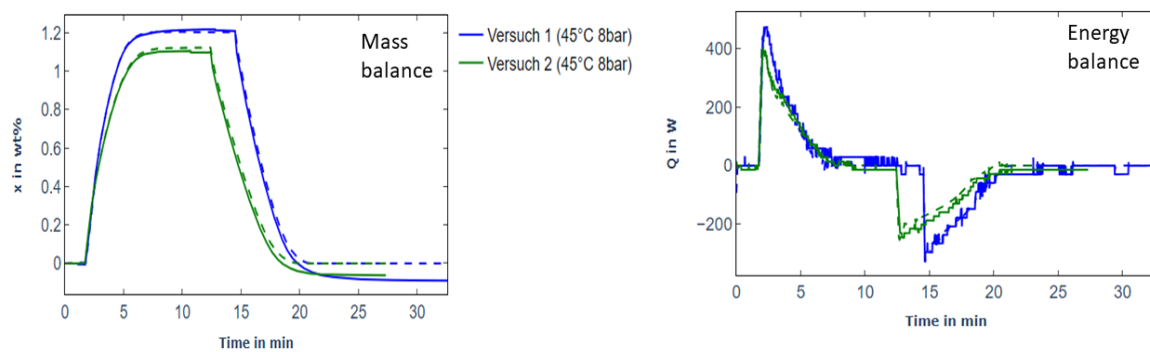

Figure 5. Compared results of the simulation and experiment at 45° C to validate the model.

4.2. Results and comparison control loop

The implemented control loop in the simulation and in the experimental setup are supposed to stabilize the outlet temperature of the HTF at 45° C even when a fluctuating inlet temperature is imposed. Figure 6 shows a graph of the temperature over the time, where the green line is the setpoint, the black and orange line are the HTF inlet flow temperature during simulation (dashed) and experiment (line), respectively. The dashed blue and red line refer to the outlet temperature of the HTF from simulation and experiment, respectively. As the imposed disturbance, the inlet temperature, deflects between ± 2K from the setpoint. The simulation result decreases maximal 0.5 K and oscillates lower than the setpoint during the two absorptions, while it oscillates higher than the setpoint during the two desorptions. Compared to this, the experimental results oscillate around the setpoint but with higher amplitudes than the simultaneous results during the absorption. During the desorption, the experimental results are as well as the simulation results oscillating above the setpoint, but the amplitudes of the oscillations from

the experiment are higher than from the simulation. After two ca. 40% absorptions followed by two desorption, the HTF outlet flow temperature cannot be stabilized at the end of the second desorption, because not enough hydrogen was saved during absorption to desorb fast enough during the entire desorption. This shows the problem of the batch process and the current limitations of thermal energy for cooling or heating with the TCU. If the loading reached the limit, no further ab-or desorption is possible. Thus, not reaching these limits is preferable. Overall, simulation and experiment proof that a stabilization of the temperature outlet is possible using the introduced TCU based on metal hydrides.
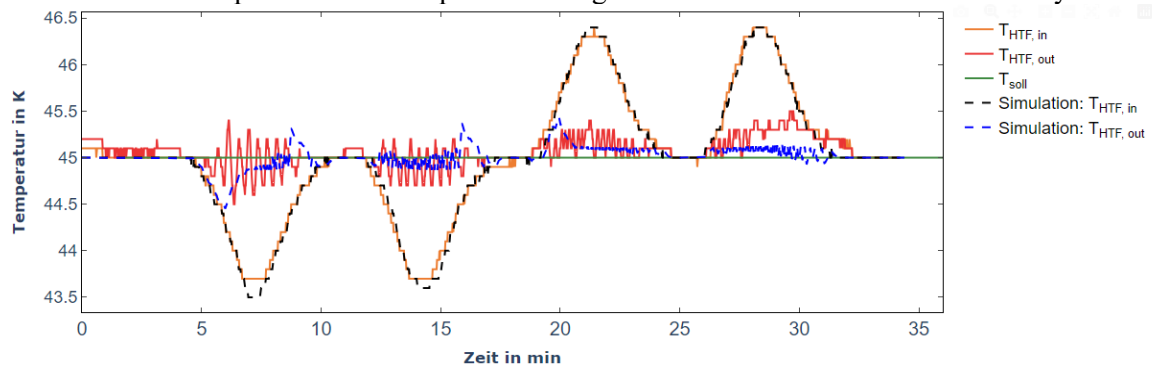
Figure 6. Simulation and experimental HTF temperatures with a PID controller.

5. Conclusion

In this study we present the stabilization of a temperature with a metal hydride reactor and the required control loop as a TCU. A model of the reactor was validated with experimental results of full ab- and desorptions at 45° C. Furthermore, partial ab- and desorptions were performed in simulation and experiment. A comparison of the results proves that the implemented PID controller is able to stabilize the temperature outlet of the HTF. One remained problem is the unknown loading status leading to limiting ab- and desorption, which can be solved with an online status calculator. Another remained problem is the oscillation of the outlet temperature. A promising solution is an internal model control for the oscillation of the outlet temperature.

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