Experimental Development of the Set-Unit Concept on a Laboratory Scale for the H_2-Chain: Production-Storage-Compression-Utilization (Digi-HyPro)

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***Abstract* – The Digi-HyPro (Digitalized Hydrogen Process Chain for the Energy Transition) project's conceptual development of the SET-Unit investigates and facilitates the connection between the electric, gas, and mobility grid. This application report describes the experimental design of the Smart Energy Transition unit (SET-Unit), contemplating the bottom-up and top-down approaches. For the bottom-up approach, the design of core devices such as metal hydride-based hydrogen storage (MHS) and compressor (MHC) systems are shown. The gas separation system (GSS) concept is based on a hybrid process composed of membrane and pressure swing adsorption (PSA) for the gas grid coupling. Commercial anion exchange membrane electrolyzer (AEM-EL) and polymer exchange membrane fuel cell (PEM-FC) are assembled for the power grid connection. For the top-down approach, the first experimental SET-Unit composed of AEM-EL–MHS–PEM-FC in the nominal power range between 5 and 10 kWel and its control strategy for the optimal hydrogen and heat coupling is presented. All experimental development is carried out in the facilities of the Helmholtz-Zentrum Hereon in the frame of a cooperation agreement with the Helmut Schmidt University/University of the Federal Armed Forces.**

Keywords* – *Green hydrogen, hydrides, storage, compression, purification

NOMENCLATURE

AEM-EL	Anion Exchange Membrane Electrolyzer
CM	Cooling Module
EL	Electrolyzer
FC	Fuel Cell
FCM	Fuel Cell Module
FLC	Fuzzy Logic Controller
FSM	Finite State Machine
GDL	Gas Diffusion Layer
GSS	Gas Separation System
HCD	Humidity Control Device
HER	Hydrogen Evolution Reaction
HTF	Heat Transfer Fluid
HTTF1	Hydrogen Tank Testing Facility One
MHC	Metal Hydride Compressor
MHS	Metal Hydride Storage
MPC	Model Predictive Controller
OER	Oxygen Evolution Reaction
PED	Power Electronic Device
PEM-FC	Proton Exchange Membrane Fuel Cell
RHCs	Reactive Hydride Composites
WPD	Water Purification Device

I. INTRODUCTION

Hydrogen is a very light gas at standard pressure and temperature and is difficult to store. For compact storage, e.g. in vehicles, high pressures of up to 700 bar are necessary. Alternatively, hydrogen energy can be liquefied in a costly process whereby approx. 15-30% of the energy content of the hydrogen must be spent on liquefaction [1].

An energy-efficient and compact solution is hydrogen storage in metal hydrogen compounds, so-called metal hydrides. Compared to high-pressure hydrogen storage, twice the volumetric capacity (> 50 kg H_2/m^3) is achieved at much lower pressures, typically a few 10 bar [2]. Especially for stationary applications in urban areas, where land is limited and expensive and high restrictions regarding safety apply, metal hydride storage has a decisive advantage over the other storage methods. The storage is also stable over the long term, as there is almost no diffusion of gaseous hydrogen through the container wall due to the low storage pressure, and the metal hydrides do not cause any other side reaction. The hydrogen is stored 100% reversibly - completely independent of the time scale [1]. Interstitial metal hydrides benefit from absorbing and releasing hydrogen within a few seconds on a small scale [3]. However, in larger tanks, the transport of the heat of the reaction becomes rate-determining [4]. Therefore, metal hydride tanks must be optimally designed systematically and knowledge-based for the desired service.

In the Digi-HyPro project (Digitalized Hydrogen Process Chain for the Energy Transition), the digital and experimental coupling of all required components to connect the gas, power, heat, and mobility grid has been developed. The concept of the so-called SET-Unit can be seen in Figure 1. Our previous work [5] presented the bottom-up and top-down strategies for developing the SET-Unit concept. On the one hand, the bottom-up approach consists of units and subsystem-level model developments and their experimental setup designs and optimizations. On the other hand, the top-down approach consists of the conceptual design and development of the whole SET-Unit and the final conception considering an optimized automatic control strategy.

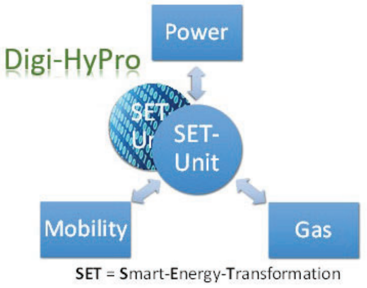

FIGURE 1: CONCEPTUAL VISUALIZATION OF THE SET-UNIT

Figure 2 shows a simplified diagram of the SET-Unit concept, including a water EL, water FC, MHS, MHC, and GSS. The system is designed to use and deliver electrical power during low and high-demand hours. Furthermore, mitigating and stabilizing the intermittency of power generation from renewable sources is possible. Hydrogen generated by the EL can be stored compactly and safely in the MHS or sent to the MHC to provide the mobility sector. The hydrogen can be separated from the mixed gas grid (natural gas + hydrogen) by the GSS. For power delivery, hydrogen can be converted in the FC.

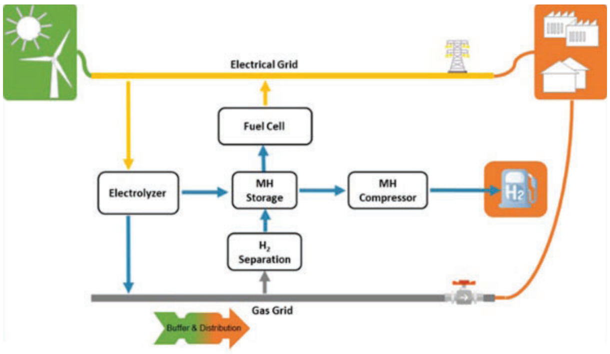

FIGURE 2: SIMPLIFIED DIAGRAM OF THE SET-UNIT WITH THE HYDROGEN-POWER COUPLING

This application report describes the conceptual and experimental development and assembly of the MHS system, MHC system, GSS, commercial AEM-EL and PEM-FC. Furthermore, the first experimental conception of the SET-Unit in the nominal power range between 5 and 10 kWel and its control strategy for the optimal hydrogen and heat coupling have been planned. All experimental development is carried out in the facilities of Helmholtz-Zentrum Hereon in the frame of a cooperation agreement with Helmut Schmidt University/University of the Federal Armed Forces.

II. DEVELOPMENT AND ASSEMBLY OF THE SET-UNIT COMPONENTS: BOTTOM-UP

This section is about the experimental development and assembly of the components included in the SET-Unit concept as a part of the bottom-up approach.

A. AEM-EL

Figure 3 (centre) shows an AEM-EL rack capable of producing green hydrogen and oxygen through electrochemical conversion of water while consuming electrical energy from renewable sources. This type of EL's water electrolysis occurs at low temperatures of ≤ 50 °C and in an alkaline environment created by a 1 wt.-% K(OH) electrolyte solution.

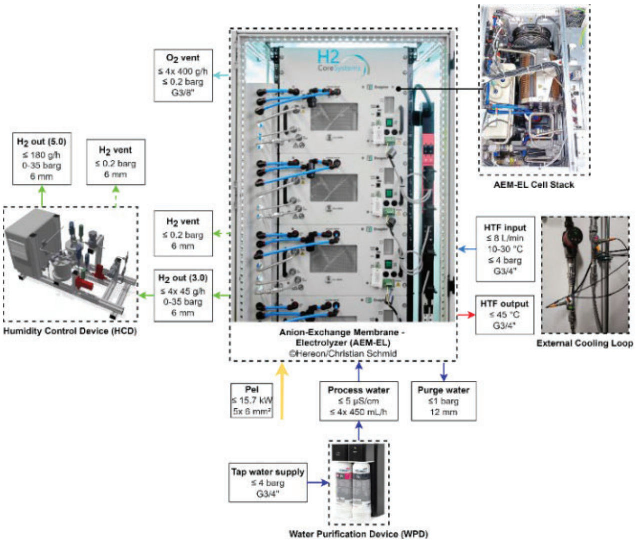

FIGURE 3: AEM-EL SYSTEM SETUP WITH EXTERNAL CONNECTIONS

The hydrogen and oxygen evolution reactions (HER and OER) are given in Equations (1) and (2). Noticeably, the hydroxide ions from the added electrolyte solution are not consumed for the overall electrochemical reactions [6].

$$\text{HER: } 2H_2O + 2e^- \rightarrow 2OH^- + H_2 \tag{1}$$

$$\text{OER: } 2OH^- \rightarrow H_2O + 0.5O_2 + 2e^- \tag{2}$$

The AEM-EL is based on four individual electrolyzer modules with approximately a net power of 2.4 kWel per module and 9.6 kWel in total. While in full load operation of the four electrolyzer modules, it will produce up to 0.18 kg/h hydrogen and 1.6 kg/h of oxygen with a water consumption of ca. 1.8 L/h.

As the hydrogen produced can contain up to 1,000 ppm water vapor, subsequent hydrogen drying is required to achieve a metal hydride-compatible hydrogen purity of 5.0 (⩽ 10 ppm water content). The used process-water and produced hydrogen must be treated for purity before and after the water-splitting process. The electrolysis water is provided by a water purification device (WPD), which can purify tap water through a combined water treatment process with several filtration, desalination, and ion exchange stages to achieve a final purity, measured by the electrical conductivity of ⩽ 5 µS/cm. For the hydrogen drying and purification process, a commercially available temperature swing adsorption dryer is used on the one hand, and on the other hand, a project-specific humidity control device (HCD) based on a water-affine polymer membrane to separate the residual water from the hydrogen content of the hydrogen gas mixture produced is used. The latter HCD measures the water content via dew point transmitters and the feed's mass flows, temperatures, and pressures, as well as retentate and permeate connections.

When the AEM-EL modules are operated, a certain amount of waste heat is generated due to the losses during water electrolysis. In particular, the anode and cathode activation overpotentials and the ionic conductivity of the membrane play a crucial role in this regard.

Due to the low-temperature electrolysis, the temperature of the recoverable waste heat is limited to 45 °C with a heat output of around 2.8 kW_{therm} in full-load operation. As the EL modules are liquid-cooled, a water-ethylene glycol mixture is used as heat transfer fluid (HTF). Depending on the given operating load profile and the HTF inlet temperature, the volume flow is adjusted between 4 to 8 L/min.

At a later stage of the project, a thermal-hydraulic coupling of the AEM-EL with the other heat-consuming or heat-supplying devices is planned to optimize the overall system efficiency of the SET-Unit.

B. *PEM-FC*

A water-cooled PEM-FC system from the Swedish company Powercell Group is used. It has a working range pressure between 3-6 bar to ensure proper hydrogen diffusion through the gas diffusion layer (GDL). It consumes 0.36 kg/h of hydrogen with a purity of $\geqslant$ 99.99% (3.5 grade) at a maximum net power and up to 260 m^3/h of ambient air for chemical reaction and ventilation.

The HER and OER are given in Equations (3) and (4). Hydrogen molecules dissociate and adsorb at the anode, oxidize to protons, and generate flowing electrons, which provide power. Then, protons diffuse through the membrane. Oxygen molecules adsorb at the cathode and react with the protons, producing water [7].

$$\text{HER: } H_2 \rightarrow 2H^+ + 2e^- \qquad (3)$$

$$\text{OER: } O_2 + 4H^+ + 4e^- \rightarrow 2H_2O \qquad (4)$$

Figure 4 shows the PEM-FC system, which comprises of three main modules: fuel cell module (FCM), power electronics device (PED), and cooling module (CM).

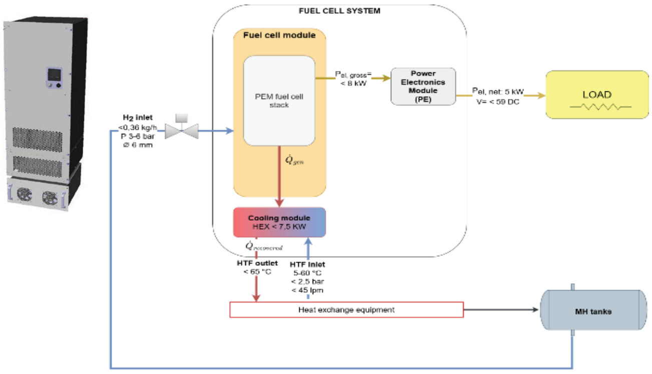

FIGURE 4: PEM-FC SYSTEM SETUP WITH EXTERNAL CONNECTIONS

A PEM-FC stack is the main component of the FCM producing up to 8 kW gross electrical power and an unregulated DC output voltage. Next, voltage regulation takes place in the PED delivering a maximum of 59 VDC and $\leqslant$ 5 kW Net Power to the end-user.

In the CM a brazed plate heat exchanger constitutes the interface between an external heating/cooling loop (for the option to recover the heat by the system) and an internal heating/cooling loop, both working together to regulate the temperature of the stack ($\leqslant$ 70 ºC) with a volume flow of up to 45 L/min.

The chosen HTF for the external heating/cooling loop will be a 50/50 water-glycol mixture. For the inlet of the CM, the operative temperature of the HTF is in the range between 5 to 60 °C and pressure up to 2.5 bar. The outlet of the HTF should be a maximum of 60 °C.

For the first stage of experiments, the PEM-FC system is tested in basic operation together with an air-cooled thermostat (LAUDA) with 8 kW of heating and 11 kW of cooling power for analysing steady and dynamic thermal behaviour of the PEM-FC system.

The PEM-FC system's second step is to be thermally coupled with the MHS by using the recovered heat in the external heating/cooling loop to release hydrogen from the MHS system.

C. *MHS*

1) *MHS material*

The material of choice for this project is the commercially available interstitial AB_2-metal alloy HydralloyC5 ($Ti_{0.95}Zr_{0.05}Mn_{1.46}V_{0.45}Fe_{0.09}$), which has several advantages for a hydrogen storage system. For instance, unlike other solid-state hydrogen storage materials, e.g. MgH_2 [8] or reactive hydride composites (RHCs) [9], it is stable against air and humidity and can be stored in ambient conditions without any protective atmosphere before activation for ease of handling. Furthermore, HydralloyC5 possesses fast kinetics (filling of a hydrogen tank possible within minutes) and a high volumetric capacity of > 60 kg/H_2 m^3 while still retaining the acceptable gravimetric capacity for a stationary system of about 1.6 wt%. Another advantage of HydralloyC5 is the thermodynamic properties of this material. As mentioned before, the AEM-EL for the SET-Unit can provide an operation temperature range between 20 and 30 ºC and an operation pressure between 30 and 35 bar. In contrast, the used PEM-FC has an operative temperature between 45 and 50 °C and needs a hydrogen back pressure between 3 and 6 bar for the operation. These operative conditions of the EL and FC are highlighted in Figure 5 as green rectangles and compared to the temperature-dependent equilibrium pressure of HydralloyC5.

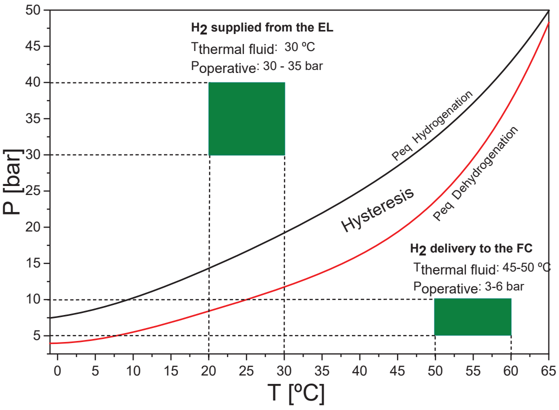

FIGURE 5: TEMPERATURE-DEPENDENT EQUILIBRIUM PRESSURE FOR THE USED MATERIAL HYDRALLOYC5 AND THE OPERATION PARAMETERS OF THE FUEL CELL AND ELECTROLYZER

The working conditions of HydralloyC5 are, therefore, at a "sweet spot" for an intermediate storage system: It can absorb hydrogen from the electrolyzer and feed the fuel cell without the need for an additional compressor system, which is known to be one of the most expensive parts during operation [10], or heat pump.

2) MH-tank and System

A digital twin was developed to design a metal hydride storage tank for a large-scale application using measured data from the lab scale (a few grams of material). With this digital twin, a large-scale storage vessel consisting of a standard pipe (168.3 mm diameter), an external heat exchanger, and an internal passive heat exchanger structure made from aluminium was designed to reduce the hydrogenation/dehydrogenation times and to provide the PEM-FC with the required flow mainly. The main focus was on the internal structure to compensate for the poor thermal conductivity of the storage material. Using a parametric analysis, this structure was optimized to reduce mass, dead volume, and costs [5].

This internal structure was then implemented inside a smaller prototype storage vessel that could store up to 250 g of hydrogen and compared to the same storage vessel with the same volumetric storage capacity without any internal heat exchanger structure. Those measurements showed that the internal structure can drastically improve storage kinetics.

The results of the prototype were used to improve the design of the internal structure, with a main focus on manufacturability and assembly on different scales. This overall design approach can be seen in Figure 6.

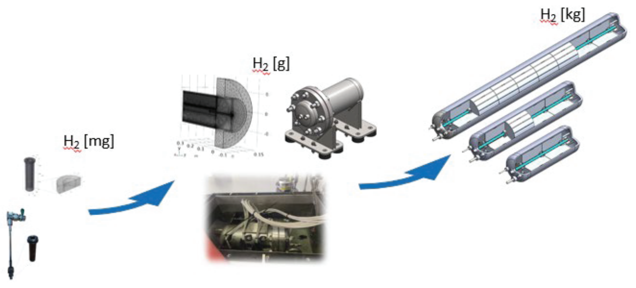

FIGURE 6: DESIGN APPROACH FOR THE MHS

Different sizes of the storage vessel (2 x 15 kg and 1 x 30 kg of storage material) were developed to build a MHS system, as shown in the flow diagram in Figure 7. This MHS system is designed to be coupled with the AEM-EL and PEM-FC, investigate the hydrogen and heat coupling, and evaluate different control strategies, as mentioned before.

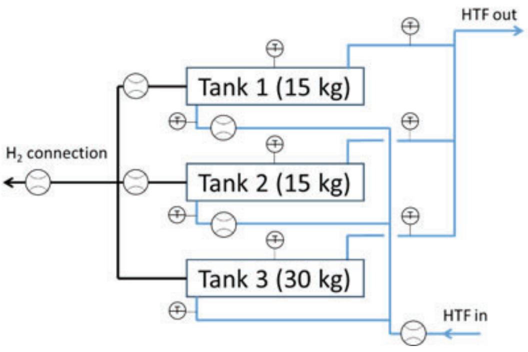

FIGURE 7: FLOW DIAGRAM OF MHS

D. MHC

1) MHC material

The goal of the compressor is to use hydrogen provided by the AEM-EL or MHS at around 30 bar and supply hydrogen at pressures of up to 350 bar for heavy-duty vehicles like trains and trucks. An MHC based on two stages was developed to achieve such a compression ratio.

The hydride-forming alloy is the core of the MHC. Thus, four candidate materials have been identified to reach the target pressures at an assumed temperature operation range of the compressor between 10 and 90 °C. Two material candidates for each stage are listed in Table I, along with their respective thermodynamic properties.

TABLE I: PROPERTIES OF THE SELECTED HYDRIDE FORMING ALLOYS

Stage	***1st***	***1st***	***2nd***	***2nd***
Material	$MmNi_{4.15}Fe_{0.85}$ (AB_5)	**Hydralloy C5** (AB_2)	$TiCr_{1.9}$ (AB_2)	$TiCrMn_{0.7}Fe_{0.2}V_{0.1}$ (AB_2)
ΔH_{Des} [kJ/mol$_{H2}$]	25.3	26.5	26.19	20.6
ΔS_{Des} [J/mol$_{H2}$ K]	105	108.3	122	107
ΔH_{Abs} [kJ/mol$_{H2}$]	24.7	21.5	27	17.7
ΔS_{Abs} [J/mol$_{H2}$ K]	107	95.5	125.4	101
H_2 capacity *[wt.%]*	0.8 - 1.1	1.1 - 1.6	0.8 - 1.9	1.0-1.95
Literature sources	[11], Lab data	Lab data	[12],[13]	[14-15]

Besides the thermodynamic properties, other considerations in the material selection are to be considered. Material kinetics and the hydrogen capacity of the material play a role in the outlet flow of the compressor. The environmental impact of mining, purification, and refining the ores required to produce such materials is also crucial, and materials based on energy-intensive raw metals like pure lanthanum or vanadium should be avoided [16-17].

2) MHC system

In addition to examining individual components, the MHC, as part of the SET-Unit, is also being investigated as a system. System modeling and simulation are being driven forward to create a digital twin of the compressor and thus be able to carry out a wide range of optimization and scaling considerations. This includes the investigation of different operating modes, the dimensioning of the individual compressor components, such as the tanks of the two stages with each other, the scaling of the overall system size, and, finally, the investigation of the interaction of the compressor system with other modules of the SET-Unit. Parallel to these developments, two MHCs of different sizes are being experimentally developed, constructed, and evaluated. The smaller compressor contains 1-2 kg of MH material per stage and can be used to measure the performance of the material present for the first time in a larger scale than a few grams. Figure 8 shows a flow diagram of the 1-2 kg MHC batch system.

It is designed as a simple two-stage compressor with easily dismantled components to exchange the MH materials relatively quickly. It has comprehensive measurement technology for recording all hydrogen and heat transfer medium flows, pressures, and temperatures. The construction is complete, and the first investigations are ongoing work. Figure 9 shows the build setup.

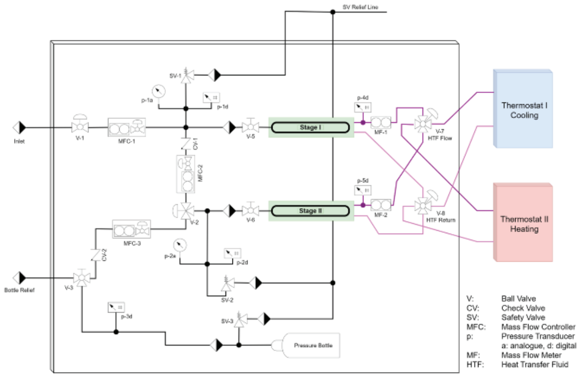

FIGURE 8: FLOW DIAGRAM OF 1-2 kg MHC BATCH SYSTEM

A further compressor on a significantly larger scale with approx. 30 kg of MH material per stage is currently being developed. It is being realized as a container structure and will be one of the few MH compressors in this application-related size range in the world. This compressor system is equipped with extensive control options and innovative concepts in its components, such as the heat exchanger in the metal hydride tank, which is crucial for its performance.

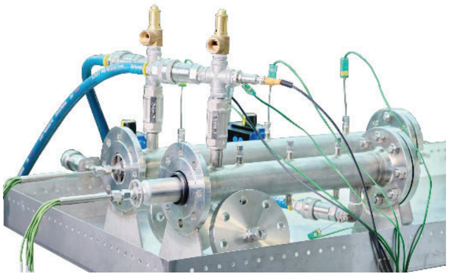

FIGURE 9: 1-2 kg MHC BATCH DEVELOPED SETUP

E. GSS

A so-called hybrid process that uses membrane and pressure swing adsorption (PSA) techniques is considered the most promising configuration. Figure 10 presents the concept of the GSS and the hybrid process. The bulk separation of hydrogen is accomplished in the membrane stage, while the following PSA process is employed to achieve the required H_2 purity. The hybrid process was developed using process simulation tools [5]. The model consists of a H_2 selective membrane stage combined with a 4-bed PSA system. Moreover, it is planned to build the GSS system in a container structure.

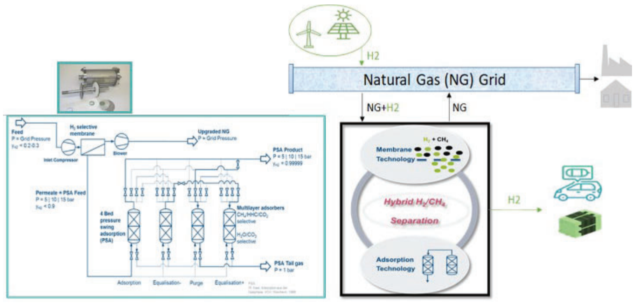

FIGURE 10: GAS SEPARATION SYSTEM CONCEPT (ADAPTED FROM [5])

III. DEVELOPMENT OF THE FIRST SET-UNIT COUPLING: TOP-DOWN

This section describes the first approach for the experimental development of the SET-Unit, including the coupling of the AEM-EL, PEM-FC, and MHS as a top-down strategy. The coupling includes the hydrogen management and the heat exchange, as well as the conception of the control strategy to optimize the system's operation.

A. *Infrastructure: Assembly of the Set-Unit*

Figure 11 – left shows the building and layout at the Helmholtz-Zentrum Hereon, where the first SET-Unit approach has been developed. The spatial distribution of the current and planned test facilities, including the existing and future pipelines, is also presented (Figure 11 – right). As seen, the MHS, the AEM-EL, and an open container for energy storage are placed in one room (R106). The PEM-FC with all the required peripherical devices, a thermostat (LAUDA), the needed control cabins, and the hydrogen management (HTTF1: hydrogen tank testing facility one) are placed outside of this room in room 101. The AEM-EL–MHS–PEM-FC SET-Unit is hydrogen and thermally coupled. The hydrogen management is done through the HTTF1 and the developed control cabins. The thermal coupling is designed to improve the overall system's efficiency by using the waste heat from the AEM-EL and PEM FC through the energy storage container (open container) and the thermostat.

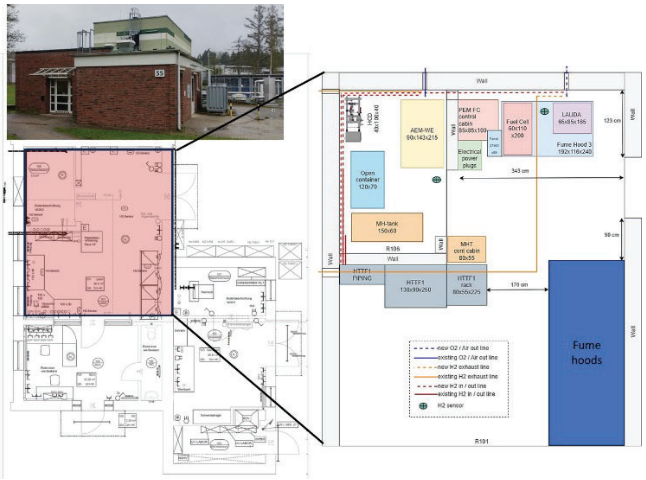

FIGURE 11: ESPACIAL DISTRIBUTION OF THE SYSTEM IN THE LABORATORY

B. *Control Strategy Concept*

Optimizing the control strategy of the SET-Unit is challenging because of the several system variables and market conditions that affect its performance. The approach here focuses on the thermal balance of one of the core elements: the MHS. The temperature control system is significantly responsible for the hydrogen absorption rate from the AEM-EL, the storage capacity of the MHS, the hydrogen flow and pressure to the PEM-FC, and, thus, the whole system's efficiency. As seen in Figure 13, a finite-state machine (FSM) and two types of controllers are used: a model predictive controller (MPC) and a fuzzy logic controller (FLC). The FSM ensures that each device operates within its defined states, guaranteeing safety by requiring certain conditions to be met before transitioning to the next state. This methodical approach is crucial in maintaining operational integrity and preventing accidents by strictly adhering to predefined safety protocols. The MPC is based on the system behaviour, and it determines in advance the control indication difference to the ideal operating parameters, such as pressure, temperature, and hydrogen flow, depending on external demand. The MPC also considers the objectives of the operating strategy as prefixes and the dynamic demand, such as maximizing hydrogen storage/supply, electricity consumption/supply (peak shaving), or thermal energy storage/supply. Implementing dual fuzzy logic controllers (FLC) in the system presents an approach to managing thermal and flow dynamics complexities within the SET-Unit. The first FLC's role is balancing the internal and external heat contributions, ensuring optimal utilization of the buffer storage, and maintaining the desired flow rates of the heat transfer fluid. This is achieved by monitoring pressure, temperature, and hydrogen flow deviations of critical system performance parameters. The second FLC complements the first by assessing the operational status of the system's components, guiding the procedural operations such as the activation or deactivation of additional tanks. This evaluation considers the ideal operational parameters and incorporates empirical data, environmental factors, and market trends, which are essential for adaptive and responsive control. The control strategy's effectiveness, developed through simulation, will undergo rigorous validation, emphasizing its potential for stand-alone applications and its comparative advantage over other control methods. This comprehensive control of an intelligent energy transformation unit based on hydrogen is unique to date and promises to maximize potential synergy approaches. [17-19].

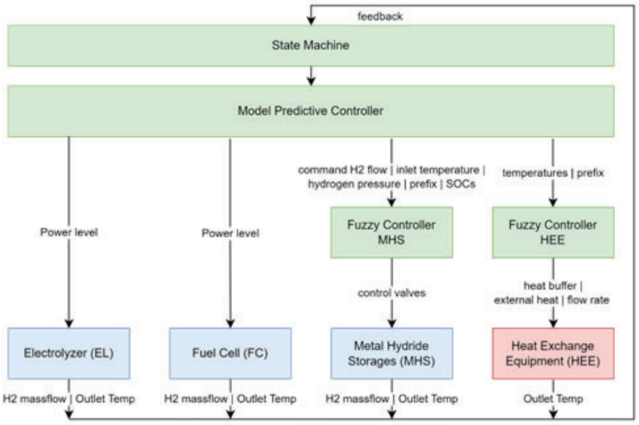

FIGURE 13: CONTROL STRATEGY FOR THE INTEGRATED SYSTEM

IV. CONCLUSION

This application report provides an overview about the approaches used to develop the first SET-Unit approach. The scope covers from the development and assembly in the laboratory of the different single components, i.e., MHS, MHC, AEM-EL, PEM-FC and GSS, to the hydrogen and thermal coupling of the AEM-EL–MHS–PEM-FC in their nominal scale of 5-10 kWel and employing advanced control strategies. As shown, the experimental SET-Unit is in a high degree of development and the concretion for the first proof of concept is going to be achieved soon. Following to this initial proof of concept, it is planned to scale-up the SET-Unit to a scale of 20-30 kWel and the coupling of devices such as MHC and GSS are envisioned.

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