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ORIGINAL RESEARCH ARTICLE

Power-to-Gas: The role of chemical storage in an energy system with high shares of renewable energy

Markus Lehner[*], **Philipp Biegger, Ana Roza Medved**

Montanuniversität Leoben, Franz Josef-Straße 18, 8700 Leoben, Austria. E-mail: markus.lehner@unileoben.ac.at

ABSTRACT

The conversion of the energy supply to renewable sources (wind, photovoltaics) will increase the volatility in electricity generation in the future. In order to ensure a balanced power balance in the power grid, storage is required - not only for a short time, but also seasonally. The bidirectional coupling of existing energy infrastructure with the power grid can help here by using the electricity in electrolysis systems to produce hydrogen. The hydrogen can be mixed with natural gas in the existing infrastructure (gas storage, pipelines) to a limited extent or converted directly to methane in a gas-catalytic reaction, methanation, with carbon dioxide and/or carbon monoxide. By using the natural gas infrastructure, the electricity grids are relieved and renewable energies can also be stored over long periods of time. Another advantage of this technology, known as "Power-to-Gas", is that the methane produced in this way represents a sink for CO_2 emissions, as it replaces fossil sources and CO_2 is thus fed into a closed cycle.

Research in the field of Power-to-Gas technology is currently addressing technological advances both in the field of electrolysis and for the subsequent methanation, in particular to reduce investment costs. In the field of methanation, load-flexible processes are to be developed that are adapted to the fluctuating supply of hydrogen. The profitability of the Power-to-Gas process chain can be increased through synergistic integration into existing industrial processes. For example, an integrated smelting works offers a promising infrastructural environment, since, on the one hand, process gases containing carbon are produced in large quantities and, on the other hand, the oxygen as a by-product from the water electrolysis can be used directly. Such concepts suggest an economic application of Power-to-Gas technology in the near future.

Keywords: Power-to-Gas; Methanation; Renewable Energy; Storage

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1. Introduction and problem statement

One of the key challenges in the conversion of electricity generation to renewable energy is its spatially decentralized and temporally fluctuating occurrence, especially in the case of wind and photovoltaic systems. There are already times when the supply of renewable electricity can no longer be accommodated in the power grid, which leads to negative electricity prices or even to the temporary shutdown of systems. A further expansion of renewable energy generation is a declared socio-political goal[1], which envisages a share of renewable energy in gross final energy consumption of 75% and in electricity consumption of 97% for the EU in 2050. This results in a steadily increasing demand for renewable energy storage in the future[2]. Storage technologies can be categorized as electrical (e.g., capacitors), electrochemical (batteries), chemical (e.g., methane, methanol), mechanical (e.g., flywheel, compressed air storage), and thermal (e.g., district heating storage, latent heat storage) storage[3]. These storage technologies differ in terms of their storage duration, their volumetric storage density (storage capacity) and their level of technological development[4]. In this context, the need for storage depends both on the speed of expansion of renewable energy generation and on the development of other flexibility options, such as load management or grid expansion. However, it is obvious that different types of storage, e.g., short- and long-term storage, will be needed in the future.

Chemical storage units belong to the group of long-term storage units. The electrical energy is used either in a water electrolysis to produce hydrogen, or in a plasma or electric arc to synthesize C2 compounds (acetylene, ethylene) from methane, for example. While acetylene and ethylene can be further processed into products as chemical feedstocks, the hydrogen can be used directly as an energy carrier. It can be used in mobility (e.g., hydrogen-powered vehicles), for reconversion into electricity in fuel cells or corresponding gas turbines, or as a valuable reducing agent in the chemical, petrochemical and metallurgical industries. However, hydrogen as a chemical storage facility only makes sense if there is a corresponding infrastructure that may already be available locally, but first has to be built up nationwide. The admixture of hydrogen to natural gas to use the natural gas grid as storage infrastructure is limited[5]. However, hydrogen can also be further processed to methane, methanol or other higher hydrocarbons (Fischer-Tropsch synthesis) by incorporating a carbon source, such as CO_2 and/or CO, in chemical-catalytic or microbiological processes. The route to hydrogen or methane is referred to as Power-to-Gas, that to liquid energy carriers as Power-to-Liquid.

Figure 1 shows an overview of the Power-to-Gas technology[6]. The main advantage of converting renewable electricity into methane lies in the possibility of unlimited use of the existing natural gas grid and natural gas storage facilities as well as in the diverse, technologically fully developed use of methane or natural gas as an energy source for reconversion, for heat generation, in mobility or even as a chemical feedstock.

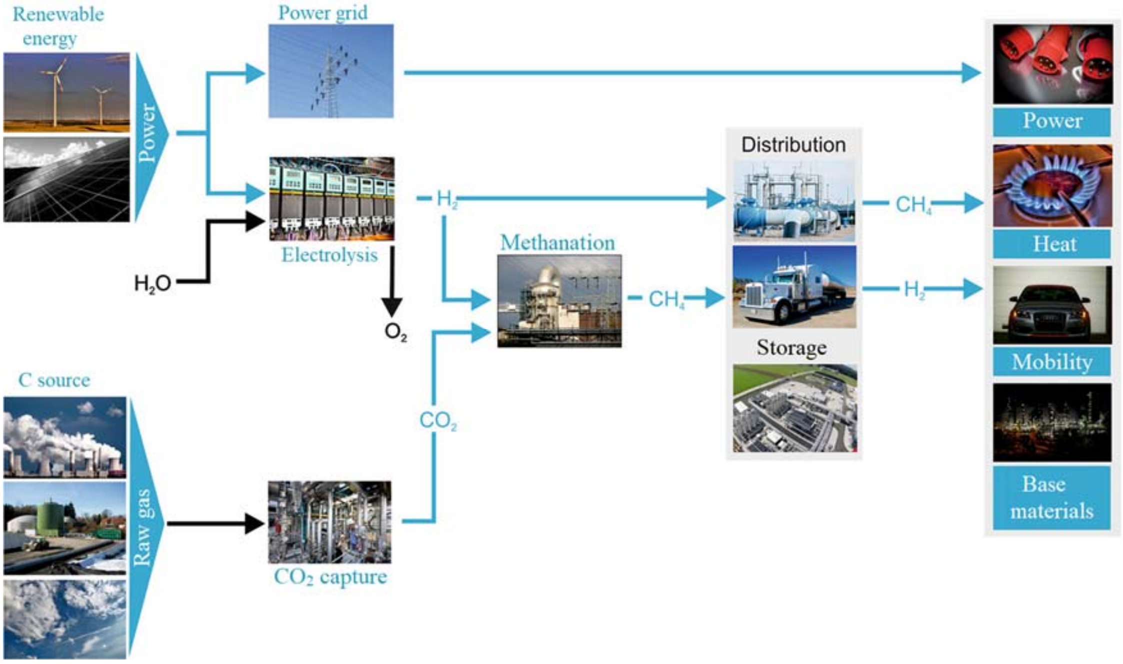

Figure 1. Concept of Power-to-Gas technology.

The research tasks for the entire Power-to-Gas process chain are, on the one hand, the dynamic operating behavior, especially for methanation, the economic efficiency, the (overall) efficiency, and the provision of a suitable carbon source for methanation[7]. While the first process step, the water electrolysis, can be operated very dynamically for the case of PEM (polymer electrolyte membrane) electrolysis and thus reacting flexibly to a fluctuating electricity supply, this is not state of the art especially for catalytic methanation in fixed bed or fluidized bed reactors[7]. The provision of a carbon source can significantly affect the economics, so special attention must be paid to the site with the specific infrastructural environment for a Power-to-Gas plant. Ongoing research activities at the University of Leoben are currently dealing with these requirements, some of which are briefly presented below.

2. Load-flexible methanation

During methanation, H_2 and CO_2 or CO are converted to CH_4 and water vapour. These reactions are strongly exothermic, higher pressures and low temperatures shift the equilibrium position towards the products. Lowering the temperature below 200 °C slows down the kinetics and leads to undesirable formation of nickel tetracarbonyl. Common catalytically active species are nickel, ruthenium or iron compounds[6].

$$CO + 3H_2 \leftrightarrow CH_4 + H_2O \quad \Delta H_R^0 = -206 \text{ kJ/mol}$$

$$CO_2 + 4H_2 \leftrightarrow CH_4 + 2H_2O \quad \Delta H_R^0 = -164 \text{ kJ/mol}$$

The reactions usually take place in fixed-bed or fluidized-bed reactors. Fluidized beds enable an almost isothermal mode of operation, since the heat of reaction can be dissipated from the fluidized bed, but are practically not load-flexible. In addition, the swirled catalyst is subjected to a high abrasive load. In fixed-bed reactors, special measures must be taken to prevent excessive local temperature increases, e.g., by recirculating the gases or by intercooling. Classic fixed-bed reactors also have little load flexibility.

For the application of methanation within the Power-to-Gas process chain, it is advantageous to have a process that is highly load-flexible, for which the reactor can be kept fully or partially on stand-by, and for which easy scale-up is possible by modularizing the reactor. These requirements result from the fluctuating power supply, to which PEM electrolysis in particular can respond well, exhibiting cold start times of a few seconds to minutes, and can be operated very dynamically[7]. In addition, the electrolysis unit can be assembled modularly to the desired nominal power. When coupled with conventional methanation processes, very large intermediate storage facilities must be provided for the hydrogen produced in the electrolysis in order to be able to operate the catalytic methanation evenly and continuously.

In the course of a Research Studio Austria, a methanation process based on ceramic honeycomb catalysts was developed at the University of Leoben together with the industrial partner Christof Projects GmbH, which combines the above requirements[8]. In this process, the ceramic honeycombs simultaneously serve as catalysts and heat accumulators for the exothermic heat of reaction (**Figure 2**). Cordierite, which has a high thermal shock resistance and heat storage capacity, is particularly suitable as a material for the honeycombs.

The ceramic honeycomb catalysts are arranged in a reactor divided into two or more chambers, which can be flowed together or individually (**Figure 3**). If only part of the chambers is traversed, the load can be reduced very far, e.g., lowered to 20% of the nominal load depending on the parallel existing chambers. By cyclically switching between the chambers, all reactor segments remain at the reaction temperature, since the exothermic reaction heat is stored in the honeycomb. The honeycombs can be arranged in several layers one on top of the other, between which cooling and/or gases can be supplied or removed[9]. The ceramic honeycombs have been extensively tested and after a short development time, they almost reach the performance of conventional catalysts[8].

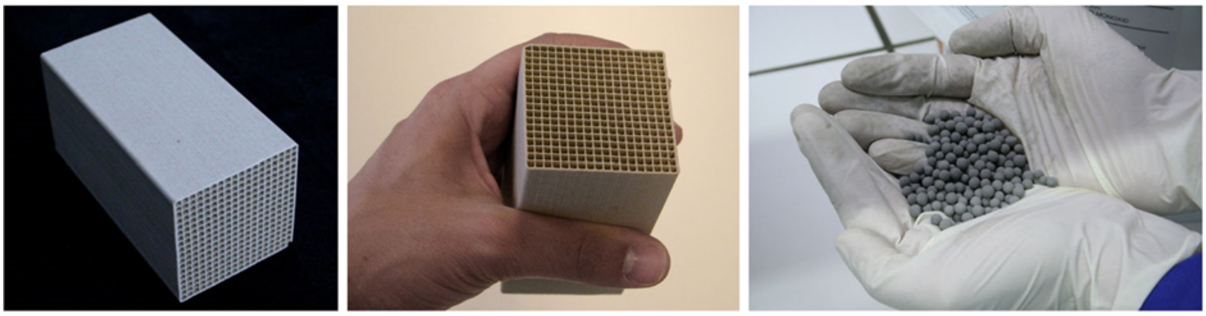

Figure 2. Honeycomb catalyst (left and center), commercial bulk catalyst (right).

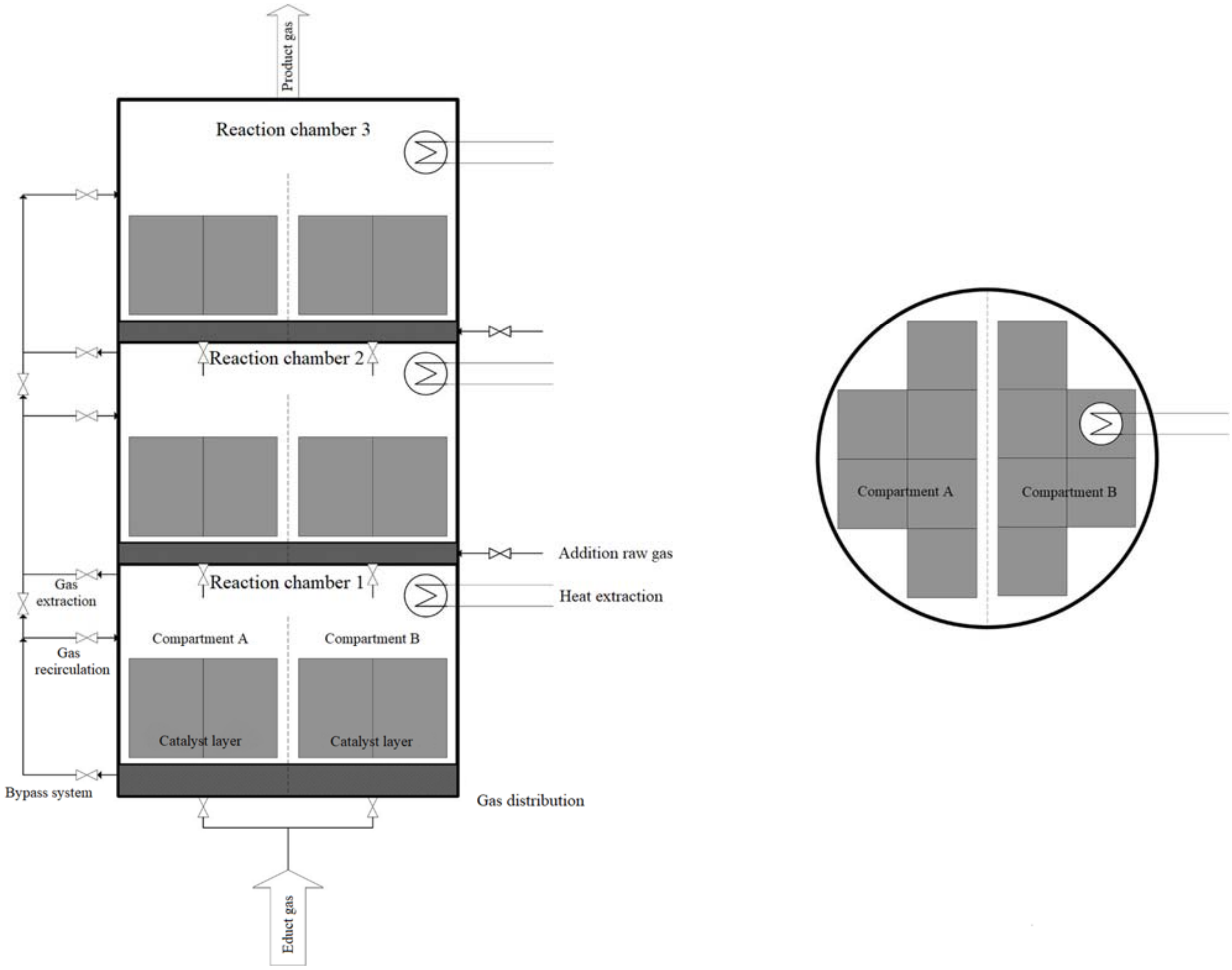

Figure 3. Schematic of the load-flexible methanation process with ceramic honeycomb catalysts.

3. Power-to-Gas in the integrated steel mill

In Europe, primary steel production is mainly carried out via the route of a so-called integrated steel mill. The designation of this route is based on the interconnection of several aggregates or production stages at one location. An integrated metallurgical plant is characterized by complex material and energy flow characteristics. Energy-rich gases containing CO, CO_2 and H_2, so-called co-product gases, are produced by various processes. These include, for example, crucible gas from the steel mill, coke oven gas from the coking plant and blast furnace gas from the blast furnace. According to the state of the art, these are energetically utilized after purification and are characterized by different compositions, e.g., with regard to the content of CO, CO_2, H_2 and nitrogen (N_2). The C or H_2 content shows significant differences, as shown in **Table 1**.

Table 1. Composition and quantity of various joint gases at voestalpine Stahl, Linz[10]

Species	Crucible gas	Coke oven gas	Blast furnace gas
Max. Volume flow [Nm^3/h]	75,000	65,000	800,000
CO_2 [Vol-%]	19.6	1.2	21.9
CO [Vol-%]	52.0	5.8	25.8
H_2 [Vol-%]	0.3	66.2	5.9
CH_4 [Vol-%]	–	21.9	0.5
N_2 [Vol-%]	28.1	~2.9[a]	~45.9
C_xH_y [Vol-%]	–	~2.0[a]	–
H_2S	–	4–12 g/Nm^3[a]	14 g/Nm^3[a]
NH_3	–	6–8 g/Nm^3[a]	–

[a] Values from [11].

A significant increase in the energy efficiency of the steel mill process can be achieved by coupling conventional steel production with Power-to-Gas technology[12]. The potential for increasing efficiency is the subject of a research project currently underway. Renewable electricity is integrated into the production process via water electrolysis. H_2 is thus available from electrolysis to reduce CO or CO_2 from the co-produced gases. Methanation produces methane as a product, which can be used both within the integrated smelter and for feeding into the natural gas grid. Oxygen from the electrolysis can be used internally in the smelter. In the course of an FFG project starting in 2017, optimal process chains and interconnection variants for the integration of Power-to-Gas in a steel mill will be developed at the University of Leoben together with voestalpine Linz and Donawitz, K1-MET GmbH as well as the Vienna University of Technology and the Energy Institute at the JKU Linz. The target of the optimization is to reduce the total CO_2 emissions of the integrated steel mill while increasing energy efficiency. In addition, experimental proof will have to be provided that joint gases are suitable as reactants for methanation in terms of their composition and their quantitative occurrence.

Figure 4 shows an exemplary principle sketch of how the interconnection between an integrated smelter and a Power-to-Gas plant for the production of a methane-rich product gas can look like. The dashed lines represent various possible product recycling routes. The solid lines mean variants of the basic material flows in the planned process chain.

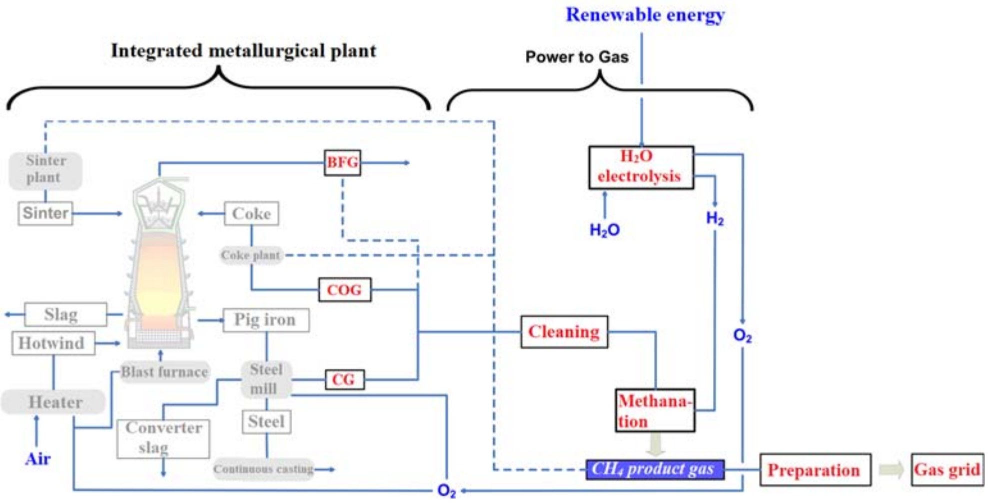

Figure 4. Coupling between an integrated smelter and the power-to-gas concept for methane production. Abbreviations: BFG: Blast furnace gas; COG: Coke oven gas; CG: Crucible gas.

Due to their composition, crucible gas and coke oven gas are the main potential co-gas streams for Power-to-Gas conversion (**Table 1**). In the concept of an interconnection between the smelter and the Power-to-Gas plant, the educt gases are fed to a gas purification system before being used in the catalytic methanation process, where impurities are removed, such as dust or sulfur compounds, which can have a poisoning effect on the catalyst of the methanation plant. Above all, sulfur compounds such as H_2S (for contents see **Table 1**) are known to be catalyst poisons. In any case, the amounts of H_2S must be determined precisely and site-specifically in order to prevent catalyst deactivation. Dust removal is also necessary, since too high a dust content causes the active catalyst surface to become coated, thus inhibiting the catalytic effect. Additional H_2 is provided by water electrolysis, feeding renewable energy into the integrated smelter. The by-product of electrolysis is oxygen, which is fed into the smelter and can be used there. For example, the oxygen could be injected directly into the blast furnace or it could be added to the air which is heated in the hot blast stove and then fed to the blast furnace as a so-called hot blast. Other possible uses for the oxygen would be in the steel mill to freshen the pig iron. This would result in a further increase in production efficiency, as the energy-intensive air separation process could be dispensed with or its size reduced. The hydrogen from the electrolysis enters the methanation plant with the joint gas mixture. The methane-rich product gas formed there can, after optional processing, be fed into the natural gas grid or alternatively used as a substitute for fossil natural gas within the steel mill. However, it is also possible to use the product gas internally in the steel mill without further purification.

This example makes it clear that embedding a Power-to-Gas system in a corresponding infrastructural environment offers significant advantages that can significantly increase the overall efficiency and, in particular, the overall profitability of chemical storage of renewable energy. The development of optimal interconnection variants as well as an exact quantification of, for example, the CO_2 savings potential is the subject of current research.

4. Conclusions

The expansion of renewable energy generation increasingly requires storage options. At present, it is still unclear to what extent, on which time axis and which storage technologies will be needed. However, there is no doubt that an energy supply with a high share of renewables will not be able to manage without short- and long-term storage. Among long-term storage facilities, Power-to-Gas offers the opportunity to draw on the well-developed infrastructure of the natural gas grid. This allows investments in transport and storage infrastructure to be avoided, as well as building on fully developed utilization technologies.

Within the Power-to-Gas process chain, it is important to be able to operate both electrolysis and catalytic methanation dynamically and with load flexibility. A newly developed methanation process uses ceramic honeycombs as a heat accumulator and catalyst. This enables a previously unrealizable load flexibility of methanation. The economic viability of the Power-to-Gas process chain can be increased by exploiting synergies with an optimal infrastructural environment. Integrated smelters represent a promising possibility for this, which on the one hand have suitable carbon sources for methanation and on the other hand possibilities for using the oxygen as a by-product of electrolysis and the energy-rich methanation products. In addition to the integration of renewable energy into the production process as well as its chemical storage, a great potential for reducing CO_2 emissions from energy-intensive industrial production arises. Similar potentials can be assumed, for example, in the cement or refractory industries.

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Conflict of interest

The authors declared that they have no conflict of interest.

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