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N A. Abdivakhidova
Turin Polytechnic University in Tashkent

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N. A Abdivakhidova
Turin Polytechnic University in Tashkent
Little Ring Road St., 17, 100095, Tashkent, Uzbekistan

Abstract: *Electricity-to-gas conversion is a key area of interest for decarbonizing and increasing the flexibility of energy systems, as it can both absorb renewable electricity during periods of excess supply and provide backup energy during periods of excess demand. By combining the power-to-gas conversion with the natural gas network, the inherent flexibility of the line package can be exploited and some of the power variability can be transferred to the gas network. Also, as long as the gas injected into the gas grid is low-carbon, such as hydrogen from renewables to gas, then the overall greenhouse gas emissions from the gas grid can be reduced. This paper presents the first review of energy-to-gas conversion, reviewing real projects, and system modeling studies and comparing them based on scope, assumptions, and results. The review identified significant interest and potential for energy-to-gas conversion in conjunction with the gas grid, however there are still challenges to be overcome to find profitable business cases and address local and system-wide technical challenges. Although significant power-to-gas modeling has been done, more is needed to fully understand the impact of power-to-gas conversion and gas supply to the network on the operational behavior of the gas network, taking into account dynamic and spatial effects.*

Keywords: *gas networks, power-to-gas system, electrolysis, methanation.*

INTRODUCTION. The Power-to-Gas system uses renewable or surplus electricity for hydrogen production (Power-to-Hydrogen) using water electrolysis. This hydrogen can be injected directly to the gas or converted into, for example, synthetic methane, syngas, electricity, liquid fuels or chemicals. The reasons for using PtG are different as it mentioned in Tickler [1]. The main goal is to store energy for a long time, easily converting it into another stored energy, and at the same time reduce the load on electricity network controlled operation (flexible demand).

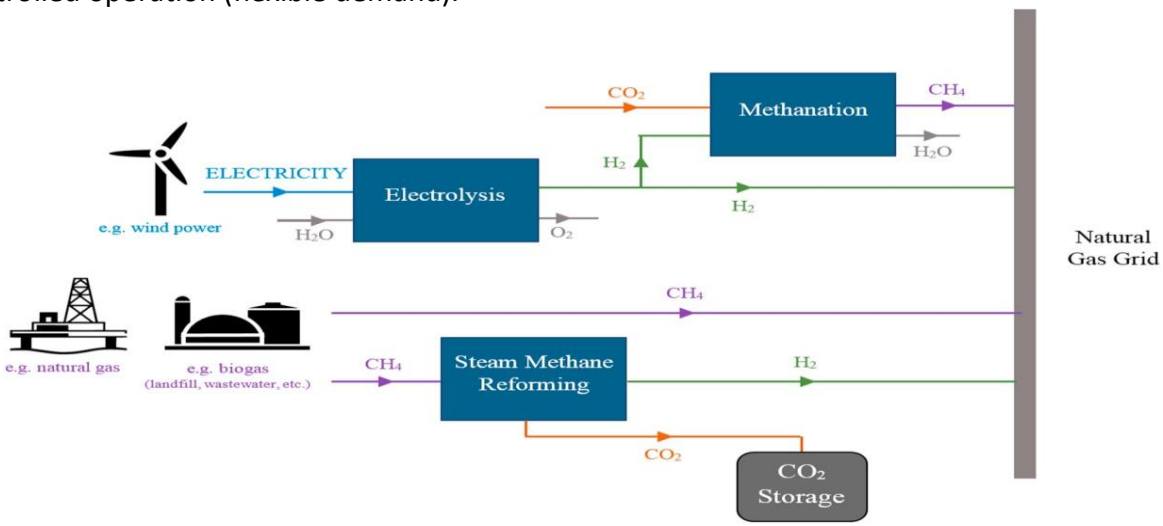

Fig. 1. Gas grid injection pathways, including: power-to-gas; hydrogen from steam methane reforming, direct injection of natural gas and bio methane; and synthetic methane from meth nation of hydrogen.

In addition, production renewable fuels for transport, household or industry, and chemical production can be a major factor for PtG. The Figure 1 gives a schematic overview of the necessary PtG pathways and components that have been discussed. In the following sections, the various components required for gas production, their specific PtG needs are discussed in more detail (e.g., various electrolysis technologies), their flowsheet (metalation) and the requirements for the transportation and distribution of gases in the natural gas network.

Production of hydrogen. Different concepts for the electrolytic production of hydrogen are already in commercial use available today. One version is the electrolysis of alkaline water. Like PEM (polymer electrolyte membrane) electrolysis behaves better at partial load, this type electrolysis has become the first choice for PtG [2]. However, for high powers it has not yet reached technology readiness, as did the alkaline electrolyze. In addition, a new type of electrolyze, SOEC (Solid Oxide Electrolyze) operating at high temperatures (700–800°C) is being studied for PtG applications.

One of the important goals of PtG is the integration of unsustainable power generation from renewable energy sources (RES) to regional or national energy systems. Thus, the main problem of electrolysis in PtG is intermittent /dynamic work. All three considered cells, PEM, alkaline electrolysis and SOEC, are capable of withstanding load changes. Because alkaline and FEM electrolysis are more advanced than SOEC, they are discussed in more detail here.

In general, PEM electrolysis handles load changes better than alkaline electrolysis. Its minimum load is from 0 to 10%, while alkaline electrolyzes reach only 20–40% [3]. These values determined due to gas purity and subsequent safety considerations. When hydrogen appears at the anode where oxygen is produced, oxygen at the cathode becomes dangerous. A gaseous mixture of oxygen and hydrogen containing oxygen more than 4% in the volume spontaneously ignites. Therefore, in technical applications the maximum value of the oxygen content in hydrogen is set to 2 % in the volume. In alkaline electrolyzes, hydrogen reaches the cathode by diffusion through a diaphragm or because it is dissolved in potassium hydroxide, which is constantly stirred. In PEM electrolyzes, hydrogen diffuses through the membrane or is transported it dissolves in water. Because these effects occur at a constant rate, the independent from actual hydrogen production, the amount of impurities increases at lower loads. Both electrolyzes can theoretically operate under overload conditions.

Load changes affect the temperature and pressure levels in the cell and respectively, its effectiveness. Each electrolyze has an optimum operating temperature. At lower loads, less heat is generated and the optimum temperature may not be reached. In contrast, under overload conditions, additional cooling is not required. Changing the pressure has an inconsistent effect on efficiency. At high pressure levels the efficiency of the cell battery reduces, while the energy requirement for drying and compressing the gas after hydrogen production decreases. In particular, for alkaline electrolyzes under low pressures the system efficiency decreases. In the electrolyzes inherent in the system, hydrogen is compressed to 30 bar, which more efficient than an external compressor. However, these cells suffer more from system shutdowns. If there is not enough electricity for a minimum the load is provided by oscillating energy sources, the electrolyze must be completely switched off after a short time, including in case of depressurization. Then the cell must be rinsed to prevent unwanted reactions of the remaining hydrogen. In conclusion, shutdown results in loss of hydrogen as well as loss of pressure level, and subsequently the loss of efficiency [4] [5].

At low loads, in addition, the specific efficiency is significantly reduced. The plant balance, e.g. pumps and cooling, has a constant energy demand that is independent of hydrogen production. For loads below 30%, this results in a significantly higher energy consumption per kg of hydrogen [4]. In general, not only the electrolyze itself, but also

process control, power rectifier and plant balance was taken into account in design conditions. Along with efficiency losses, faster degradation of materials used for an electrolyze can be expected. There are currently no studies that quantify the degradation and explain the causes. However, with fluctuations consumed electricity electrical as well as mechanical parts of the electrolyze system are suspended.

MATERIAL AND METHODS. Specific investments (per kW of electricity consumed) for electrolyzes very much. Values for alkaline and PEM electrolyzes range from 900 and €4,200/kWh for 2010-2012 and is expected to drop to 250 and 900 euro/kWh in 2030 ([6]; [7]). This the large difference can be explained by the different technologies analyzed. AT 312 PART I Examining the challenges and scope of HSC design for now, PEM electrolyzes are still more expensive than alkaline electrolyzes.

However, for 2030 both values may be in the same range [6]. Other uncertainties are scaling and learning effects. Even although pots cost the same in small and large pots, plant balance (e.g. power supply, system control, gas dehumidification) is achieved reducing costs through scaling. Thus, future costs depend on the scenario chosen for pot sizes and absolute installed capacity. The unit costs of gas production depend not only on investments, but also strongly about the costs of purchasing the necessary electricity and hours of full load system. This also affects other parts of the PtG systems such as metalation.

Production of methane. Metalation is the conversion of carbon monoxide and carbon dioxide (CO_2) to methane (CH4) through hydrogenation. CO_2 metalation reactions were first discovered by Sabatier and Senders in 1902. The reaction for metalation is described in the Eq. 1.

$$CO_2 + 4H_2 = CH_4 + 2H_2O \quad (1)$$

If metalation is part of PtG system, carbon dioxide must be separated from industrial processes, energy plants or biogas plants, if they are to be carbon neutral. There are currently two different concepts available. The most common catalytic metalation at higher temperatures (starting from 250–300°C). In addition, biological metalation is possible.

Catalytic metalation**.** The above Sabatier reaction is most often carried out as a thermochemical catalytic reaction. The highest conversion rate of this reaction is achieved at low temperatures. However, to also provide a reasonable reaction rate, a catalyst is needed. Today, nickel catalysts are mainly used (other metals such as ruthenium) are possible ([8], [9]) with alumina as carrier material (also silicon dioxide, zeolites or test metals apply. They require a temperature of at least 300°C to operate. This reaction is highly exothermic (ΔH = 252.9 kJ/mol).

However, the catalyst begins to sinter at temperatures above 550°C ([8]). Thus, the main problem in reactor design is temperature control. The pressure level ranges from 1 to 200 bar [10]. In addition to temperature control, maximizing the conversion of switching to synthetic natural gas (LNG) is a primary goal of reactor design.

Therefore, different types of reactors are used or researched. Fixed bed reactors are state of the art meth nation equipment [10]. To control the temperature, several reactors (from two to five) are connected in series with intermediate flow cooling (adiabatic calculation) (Alternatively, direct cooled fixed bed reactors, i.e. shell-and-tube and plate reactors, can be used as prototypes for isothermal design [9].

Rapid load changes and subsequent temperature changes during metalation are not a problem for the catalyst. However, modern catalysts have a maximum lifetime of only 24,000 hours. In addition, after a period of inactivity, the reactor needs several days to restart, and the reactor must be flushed with hydrogen or some other inert gas before

shutdown [10]. The specific investment (per kW of methane capacity) for catalytic metalation is also difficult to determine. For 2030, the expected range is between 200 and 600 EUR/kW.

RESULTS AND DISCUSSION. Biological metalation. Instead of the thermochemical catalytic process described above, the Sabatier reaction can also be carried out by the metabolic processes of methanogenic archaea, chemoautotrophic microorganisms. This is a common process that is part of the production of biogas in anaerobic digesters. Therefore, different ways of implementing this reaction in biological reactors are possible.

First, hydrogen is added to the biogas boiler or after the digester to increase the methane yield. Thus, the concentration of methane in biogas can be increased from 50% to 75% ([10] [11]). However, the higher partial pressure of hydrogen in the digester prevents the decomposition of the biomass. Thus, the addition of hydrogen is limited ([12]).

As a second option, the meth nation can also take place in a separate microorganism reactor fed with pure carbon dioxide or raw biogas. As in a biogas reactor, microorganisms need thermophilic conditions (about 65°C). Archaea working under mesophilic (33–45°C) or hyperthermophiles (65–85°C) conditions are used. At higher temperatures, the solubility of hydrogen in water decreases, although [11] gases (hydrogen and carbon dioxide) must be dissolved in water for processing by microorganisms. The pH value should remain between 6.8 and 7.2, which must be carefully monitored as carbon dioxide lowers it. An effective method for pH control has yet to be developed ([10]). In addition, strictly anaerobic conditions are necessary, since oxygen kills microorganisms. Since microorganisms are more resistant to impurities than nickel catalyst in thermochemical meth nation, biological meth nation can be fed with raw biogas and hydrogen without any pre-treatment.

However, before LNG is supplied to the gas network, it must be purified, for example, from sulfur components. In test reactors, higher methane yields have been achieved using pure carbon dioxide compared to biogas. Also, the metalation reactor can be smaller if pure carbon dioxide is used. After metalation, only 85%–95% of the produced gas is methane (The rest consists of hydrogen and carbon dioxide, which, after purification, are returned to the metalation process. Due to incomplete reactions of the starting materials, the efficiency of converting hydrogen to methane is reduced. In addition, 18% of the added hydrogen is converted into heat rather than methane. Various types of reactors are being developed for this process. The most common for this application is the continuous stirred reactor. This type, however, needs extra energy to get everything mixed up.

The most important task at the moment for biological metalation is entry of gaseous hydrogen to microorganisms due to low solubility hydrogen in water. On the other hand, the first test showed that archaea are resistant to sudden changes in temperature, pressure and load [10]. Even starting after a long period of inactivity is possible without problems.

Further research is needed to study dynamic behavior. However, it can be assumed that the limiting factor will be the process control system, and not the process itself Compared to thermochemical metalation, biological metalation occurs at a slower rate due to lower temperature, resistance to gas-liquid mass transfer, and back mixing. This results in large reactor volumes when a similar volume flow must be achieved. At present, biological metalation is still on a pilot and demonstration scale. The investment required for biological metalation for small plants (5MW SNG) is in the same range as for catalytic metalation. However, the scaling effects are clearly more pronounced for catalytic metalation [11]. According to Götz [10], biological metalation is technically the most beneficial for small reactor sizes. As shown in the paper If the main purpose of PtG is the production of fuels and chemicals, alkaline electrolyzes are preferred. If, on the contrary, the focus of the project is the use of surplus energy and ancillary services for the electricity grid, then PEM pots,

despite their higher cost, are more common. In addition, new electrolyzes (SOEC, SPE) are being developed to improve efficiency.

To avoid restrictions on the hydrogen that can be given by gas restriction, pay special attention to the production of other gases, such as the metalation and co-electrolysis pathways. The technology for this is completely different. Instead of storing electrical energy, chemical energy in the form of gas (especially methane) is much more preferable and more convenient. But the price of production of synthetic methane is not so low. If the electrical energy is taken from renewables (wind, photovoltaic panels), production of hydrogen could be profitable.

CONCLUSION. Hydrogen could be directly added to the gas grid, could be used for metalation reaction, or sometimes hydrogen becomes a fuel for transport system. In terms of speed of reaction, catalytic method for producing methane is better because of high temperature range and also much easier to control the reaction. Biological method is still in the development. Depending on economic issue and existing infrastructure we can follow different scenarios for PtG applications. However, until now, all technologies take place under certain conditions of offenses.

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