

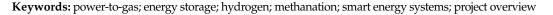


# **Review Review on the Status of the Research on Power-to-Gas Experimental Activities**

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**Abstract:** In recent years, power-to-gas technologies have been gaining ground and are increasingly proving their reliability. The possibility of implementing long-term energy storage and that of being able to capture and utilize carbon dioxide are currently too important to be ignored. However, systems of this type are not yet experiencing extensive realization in practice. In this study, an overview of the experimental research projects and the research and development activities that are currently part of the power-to-gas research line is presented. By means of a bibliographical and sitographical analysis, it was possible to identify the characteristics of these projects and their distinctive points. In addition, the main research targets distinguishing these projects are presented. This provides an insight into the research direction in this regard, where a certain technological maturity has been achieved and where there is still work to be done. The projects found and analyzed amount to 87, mostly at laboratory scale. From these, what is most noticeable is that research is currently focusing heavily on improving system efficiency and integration between components.



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## 1. Introduction

Power-to-gas (PtG) is an energy exploitation concept that began to expand during the last decade. It fits into the broader topic of sector integration, whereby the electricity and gas sectors are combined.

As nonprogrammable renewable energy sources (e.g., wind and photovoltaic) gradually gained a foothold in the energy market, in order to manage the energy surplus at certain times of the day and the deficit at others, storage methods began to be considered to a greater extent. In this context, PtG aims to be a viable technology for long-term storage in the form of chemical energy. In this case, in fact, the energy surplus of renewables is directed to an electrolyzer to produce green hydrogen, which can then be made to react with carbon dioxide to obtain the energy carrier methane.

Although the capture of carbon dioxide is, to date, a very expensive process [1], it allows the production of a renewable fuel that has a net zero impact on the environment, as is the case when using biomass. In addition, the energy carrier obtained has undoubted value since it can be used within the existing transport infrastructure for multiple purposes such as the production of electricity and heat, or for mobility.

The growing interest in PtG plants and in the involved processes is testified in various review papers. Specifically, in [2], a review of PtG and power-to-hydrogen plants focuses on technical characteristics, it analyzes the possible operation of the various components, and it evaluates how they are actually operated. Moreover, in [3], a selection of laboratory, pilot, and demonstration PtG projects is analyzed with particular focus on the methanation process. In [4], the authors consider plants producing hydrogen or other synthetic fuels and they assess the state of the art limited to technological and economic aspects. In [5],

reference is made to PtG demonstration projects in Europe. The focus is on the technologies adopted for the various phases. Then, the analysis is widened to power-to-x projects in [6] and other categorization criteria, e.g., carbon dioxide and electricity sources, are added. Finally, in [7], a similar analysis is extended worldwide.

It is noteworthy that in all the review articles cited, the focus is on plants by examining them from a technological point of view, often referring to the specifications of the various elements, or by evaluating their economic aspects.

None of them deal with the research and development activities of an experimental nature carried out on the plants and none discuss the state of experimental research on the subject.

#### Aim of the Paper

The intention of this paper is to fill this gap. In order to address this shortcoming, the review was trimmed by referring to research and development plants and to experimental research activities.

Therefore, the most important contribution of this work is to provide an overview with useful tools for researchers and companies working in the field. In this way, the reader would be able to determine which technologies are the most mature and adopted in practice, which research fronts are progressing most rapidly, and which gaps need to be filled with potential, further research. Broad areas of research are, therefore, identified, and each project is treated in relation to its research objective.

The aim of this work is also to provide a structured survey of the experimental research projects and research and development activities on this topic, highlighting the specifics of the research targets that characterize these projects. In addition, information has been acquired on companies and universities that have worked and/or are working on these projects, on the size of plants, and on the technologies adopted.

To achieve this purpose, a technical background is initially presented in this paper to provide the appropriate knowledge for a proper comprehension of the presented contents. Then, the methodologies adopted for the research are stated and, finally, the results obtained are discussed. The latter, for the sake of completeness, includes both quantitative technical data on the plants together with qualitative considerations on the research topics most widely discussed.

The main characteristics of the reviewed projects are summarized in the tables in Appendix A.

#### 2. Technical Background

Power-to-gas basically consists of three stages: electrolysis, carbon dioxide capture, and methanation [8]. Despite this, plants of this type do not have a well-defined architecture. In fact, for each of the mentioned stages, various technologies can be adopted, which can be very different from each other. One example is the methanation phase, which can be catalytic or biological [9], or the carbon dioxide capture phase, which can be mechanical, chemical, or thermal [10].

For this reason, it is considered appropriate here to provide an overview of the technologies that can currently be used in plants of this type.

#### 2.1. Electrolysis

The aim of the electrolysis step is the production of hydrogen. It is an electrochemical process in which the splitting of water molecules occurs. The electrolysis cell consists of two electrodes, the negatively charged one in which the reduction reaction takes place (namely, the cathode) and the positively charged one in which the oxidation reaction takes place (namely, the anode). The charge carriers vary depending on the technology used, but the overall reaction that occurs is generally as follows [11]:

$$H_2O \rightarrow H_2 + \frac{1}{2}O_2 \qquad \Delta H_r^0 = +285.8 \text{ kJ/mol}$$
 (1)

where the water phase can be liquid or in vapor form depending on whether a low- or high-temperature electrolyzer is used, while the molecular hydrogen and oxygen phases are gaseous.

Electrolyzers are mainly classified by their operational temperature, i.e., high- or low-temperature. Specifically, among low-temperature electrolyzers, the main types are alkaline and polymer electrolyte membranes (PEM). These electrolytic cells work at temperatures up to 90 °C. High-temperature technologies are, in contrast to the former, still under development. The main high-temperature electrolytic cell is the solid oxide electrolyte cell (SOEC), which is capable of working at temperatures between 700 °C and 900 °C [12].

From a PtG perspective, high-temperature electrolyzers establish the way for a further approach, that of co-electrolysis. In this case, in addition to steam, carbon dioxide is also split, and syngas (i.e., a mixture of carbon monoxide and hydrogen) is produced. Syngas can be further processed to produce synthetic fuels, including methane. As stated in [13], in a power-to-gas system, using an SOEC in co-electrolysis rather than steam-electrolysis can lead to an increase in the total system efficiency due to the greater heat available from the methanation of syngas than from the methanation of carbon dioxide alone.

#### 2.2. Carbon Dioxide Capture

One of the most critical points of the entire process is undoubtedly obtaining carbon dioxide. Carbon capture and utilization, in fact, currently represents one of the most expensive processes for a power-to-gas plant. Carbon dioxide can be taken from combustion processes or obtained as an industrial byproduct (it is worth highlighting here the presence of anaerobic digestion processes for biogas production). Another way could be to take atmospheric carbon dioxide directly from the atmosphere through a process known as direct air capture (DAC). This allows power-to-gas to be implemented even if there is no access to a specific plant producing carbon dioxide.

There are various methods that can be adopted for its capture. The main ones involve absorption (which can be chemical or physical), adsorption, or membrane systems. Cryogenic and algae-based methods are also being investigated [10].

The lower the concentration of carbon dioxide in the gas from which it is taken, the higher the cost of separation. This is why the use of atmospheric carbon dioxide, although it could be very useful, is currently extremely expensive [1] since its concentration is 420 ppm [14]. Despite this, as shown below, in practice there is no shortage of examples of plants making use of this technology. Another possibility, as yet unexplored in this field, could be to use air present in buildings which, as shown in [15], reaches values of more than 500 ppm.

#### 2.3. Methanation

The methanation stage is the final stage by which the desired gas is obtained. Reactors that can carry out methanation are divided into two large groups, catalytic and biological. In the catalytic ones, thermochemical conversion takes place, assisted by catalysis to facilitate the kinetics of the process. This procedure is known as the Sabatier process.

What takes place is the reaction between hydrogen and carbon dioxide that have already been obtained by the methods described above. Specifically, the reactions that occur are as follows:

$$CO_{2(g)} + 4 H_{2(g)} \rightleftharpoons CH_{4(g)} + 2 H_2O_{(g)} \qquad \Delta H_r^0 = -165.1 \text{ kJ/mol}$$
 (2)

$$CO_{(g)} + 3 H_{2(g)} \rightleftharpoons CH_{4(g)} + H_2O_{(g)} \qquad \Delta H_r^0 = -206.3 \text{ kJ/mol}$$
 (3)

$$CO_{2(g)} + H_{2(g)} \rightleftharpoons CO_{(g)} + H_2O_{(g)} \qquad \Delta H_r^0 = +41.2 \text{ kJ/mol}$$
(4)

$$2CO_{(g)} \rightleftharpoons C_{(s)} + CO_{2(g)} \qquad \Delta H_r^0 = -172.5 \text{ kJ/mol}$$
(5)

where the first two are the hydrogenation reactions of carbon dioxide and carbon monoxide, respectively, the third is the reverse water gas shift reaction, and the last is the Boudouard equilibrium reaction [11].

As can be seen, hydrogenation reactions are highly exothermic, which leads, according to Le Chatelier's principle, to the need to cool the reactor.

Methanators generally work at temperatures between 300 °C and 550 °C and at pressures ranging from 1 bar to 100 bar [16]. The materials used for catalysis are usually active metal particles (e.g., Ni, Fe, Ru) dispersed on a support consisting of a metal oxide such as alumina [17].

The most common types of catalytic methanator for these purposes are fixed-bed, fluidized-bed, and the so-called three-phase methanator. In the last, the solid catalyst (e.g., a powder) is suspended in a stable and inert liquid, and the third phase is represented by the processed gas. Fixed-bed reactors are adiabatic due to the poor capacity of the bed to conduct heat; this is the reason why there is often a need for a cooling section between the stages. To solve this problem, another solution is to use structured-type reactors such as monolith or honeycomb-type reactors, in which the internal metal structure is designed to facilitate heat transfer [18]. In addition, it is considered important to highlight the existence of studies on other types of methanators, such as sorption-enhanced reactors, in which water removal is carried out by adsorption directly inside the reactor, or the so-called photocatalytic methanation reactors, in which reactions are assisted by either solar or artificial light [9].

In biological-type reactors, the fundamental idea is very different from those mentioned above. In this case, catalysis is carried out by microorganisms, thus it is often referred to as a biocatalytic process. Operating conditions can range between 0 °C and 122 °C and between 1 bar and 10 bar [9]. The microorganisms used are the autotrophic hydrogenotrophic methanogens (archaeal bacteria), already known to be used in the methanogenic phase of the anaerobic digestion for the production of biogas. Based on this close relationship with anaerobic digesters for biogas production, there are two macrotypes of biological methanators, in situ and ex situ [16]. In the former, the hydrogen produced by electrolysis is injected directly into the digester to increase the total methane yield by converting the carbon dioxide present in the biogas. In the latter, a separate reactor is used in which hydrogen and carbon dioxide are fed under stoichiometric conditions. In the ex situ applications, the carbon dioxide input does not necessarily come from a biogas plant and the methanator can be a continuously stirred tank reactor, a trickle-bed reactor, or a reactor with hollow fiber membranes.

## 3. Methodology

The methodology adopted for the investigation involves an extensive analysis of the current literature on the subject of power-to-gas.

The project typologies taken into account in the analysis concern the following categories:

- *Research and development activities*: includes the completed plants, often commissioned by a company or a consortium, which have a demonstrative or prototypal nature.
- *Experimental research projects*: includes experimental campaigns carried out by universities or research centers that aim to increase the technical maturity of PtG plants, seen in their entirety or focusing on specific parts.

Moreover, the projects included in the review meet the following requirements:

- Aims toward the production of synthetic methane;
- Contributes to the increase in knowledge about power-to-gas plants—it is necessary that the project has an experimental or developmental character and documents the activities with papers or other deliverables;
- Refers either to the entire plant or to a single part. In the latter case, the experimental
  activity on a single component must be declaredly designed in order to obtain an
  overall improvement on the performance of the whole plant.

In order to effectively reorganize the information, a categorized table was first arranged to view as clearly as possible the plants and activities analyzed.

Secondly, review articles related to similar topics [2–6] were taken as a reference in order to start populating the table. The significant items of the other reviews were then reorganized in a suitable manner.

Subsequently, the table was filled in and expanded by means of experimental activities found in the literature and by a web analysis of currently existing plants.

Hence, an in-depth examination of the research objectives of the selected projects was carried out. A second table was then developed in which these objectives were grouped together, and the innovative characteristics of each project were noted. This represents the original contribution of the present paper to the existing literature.

For the sake of readability, these objectives were grouped into a number of broad areas, the research targets, categorized below:

- Heat management: includes all projects with the stated objective of optimizing heat handling, whether this is internal to a component or related to heat flow between several components.
- Operating condition optimization: concerns the conditions under which it would be good
  practice to operate a component and thus increase the efficiency of the components
  that compose a PtG system.
- *Materials*: consists of those projects in which materials are tested, e.g., for catalysis or water removal.
- *Carbon capture*: gathers together projects with the primary objective of finding an innovative way to sequester and utilize carbon dioxide.
- Design: concerns projects where the main aim is related to the design of a part of the plant or its complex.
- *Renewable fuel production*: includes those projects aiming to synthesize a green fuel.
- Integration within the energy system: collects those projects that tend to increase system efficiency by evaluating its integration into an energy network.
- Technical and economic feasibility: covers projects aiming to assess system viability, whether from a technical or economic point of view.

As can be seen from Table A2 in Appendix A, a single project can be placed into more than one category, if considered relevant.

#### 4. Results and Discussion

By means of the methodology discussed above, it was possible to find and evaluate 87 projects. As mentioned above, this number includes both research and development activities and experimental research projects. Each of them is dedicated to improving power-to-gas as a whole or with regard to individual components.

The results are reported and analyzed below.

#### 4.1. Projects Evaluated

The projects evaluated are listed in Table A1 in Appendix A. In this table, a set of both technical and nontechnical information is provided. The nontechnical information includes not only the name of the project, but also its location, start date, and the entities (whether universities or companies) involved in commissioning it.

#### 4.1.1. Temporal and Geographical Distribution

An initial important result can be found by analyzing the time trend concerning the years in which the PtG projects were launched.

As can be seen in Figure 1, power-to-gas plants began to emerge as early as the late 1990s. Although the PtG concept was not yet widespread and standardized, at Tohoku University [19] in Japan, a laboratory-scale plant was built to recycle carbon dioxide, which today can be considered as a power-to-gas system.

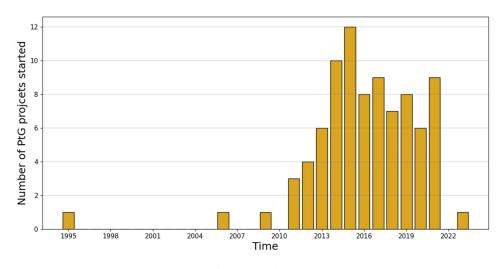


Figure 1. Power-to-gas projects grouped per start date.

However, the real surge of case studies on the subject only occurred after 2010, a date after which the power-to-gas concept began to be defined within the more general term power-to-x.

Great efforts for the realization of these projects were made in Europe and especially in Germany. In fact, Figure 2 shows the contribution that various countries worldwide have made and are making to the realization of power-to-gas plants.

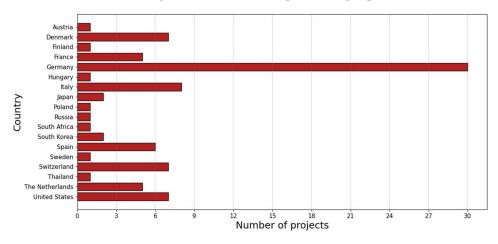


Figure 2. Power-to-gas projects grouped according to country.

Although Europe, and specifically Germany, constitutes the focus of the research, it is noticeable that interest in the subject extends worldwide and does not remain confined.

#### 4.1.2. Project Size

With reference to the nominal electrical power of electrolyzers, where present, it is possible to judge the size range of the projects under consideration. Knowing the size range over which a particular industrial process extends makes it possible to understand the degree of maturity of the process itself. As far as power-to-gas is concerned, it can be said that, at present, there is still much room for improvement. In fact, there is not yet the confidence regarding the technical–economic feasibility required to establish projects on a larger scale.

According to Figure 3, in fact, apart from a few cases, most projects have a size that is less than 1 MW.

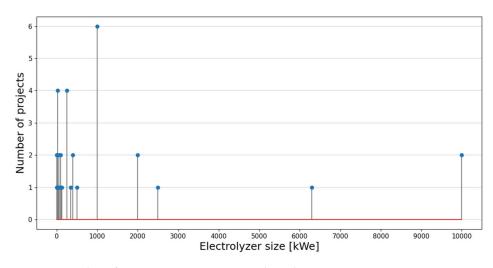


Figure 3. Number of power-to-gas projects per electrolyzer size.

This is due to the fact that electrolysis technology (and consequently power-to-gas) has received attention for large-scale applications only in recent years.

The graph shows that the majority of plants fall within the smallest range. Figure 4 focuses on this range, confirming the fact that at present, projects of this type are mostly still at a laboratory or demonstration level.

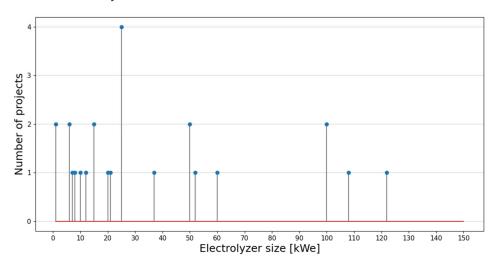


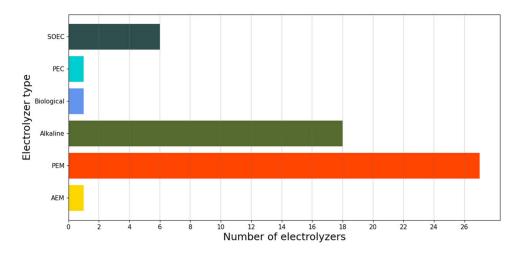
Figure 4. Number of power-to-gas projects per electrolyzer size (focused on smaller sizes).

4.1.3. Component Typology

With reference to Section 2 regarding the technical background, it can be seen that the set of processes that make up PtG can be conducted in various ways. Although the components of the system perform the same task, they can differ considerably in operating principles (e.g., biological or catalytic methanation).

The work presented here may be useful in clarifying which types of technology are actually used and to what extent.

The electrolysis phase makes use of various technologies, detailed in Figure 5.



**Figure 5.** Number of power-to-gas projects per electrolyzer type (PEM = proton exchange membrane, SOEC = solid oxide electrolysis cell, PEC = photoelectrochemical water splitting, AEM = anionic exchange membrane).

The analysis shows that most plants use PEM-type electrolyzers, followed by alkalinetype and SOEC. However, it is noticeable that there are attempts to incorporate electrolytic cells with a lower level of technological maturity into a power-to-gas system. Specifically, in [20,21] photoelectrochemical water splitting is used, in [22] an anionic exchange membrane electrolytic cell is exploited, and in [23] a bioelectrochemical system for electromethanogenesis is employed. It is also worth mentioning that PEM and alkaline electrolyzers are used in combination in the *Jupiter 1000* project [24].

The large presence of PEM-type electrolyzers can be explained due to the fact that they have better dynamic performance [25] and can therefore be more easily powered by nonprogrammable renewable energy sources such as wind and photovoltaic.

As far as methanation is concerned, on the other hand, the results are reported in Figure 6.

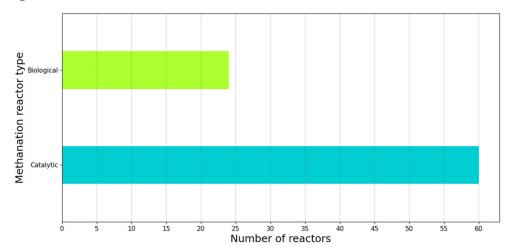
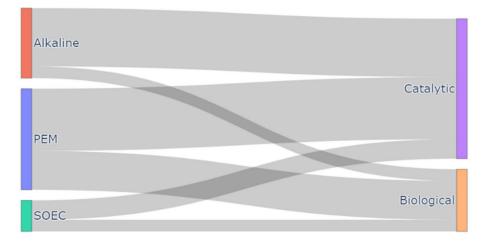


Figure 6. Grouping power-to-gas projects per methanation technology.

The vast majority of methanators are catalytic, but the presence of biological methanators is not negligible. Within the category of catalytic methanators, however, there is a great variety of architectures. In fact, there are many types of catalytic reactor, as reported in Section 2.3. In this case, the choice of the reactor is often based on considerations including the management of the system as a whole.

The choice of a particular type of electrolyzer in combination with a particular type of methanator depends on many factors, and it is noticeable that there is still no standardized



choice. Therefore, Figure 7 shows the choices that have been made in various projects for the coupling of the two components.

Figure 7. Sankey diagram showing coupling between electrolysis and methanation technologies.

As can be seen, there is much flexibility for coupling. However, while it is true that PEM and SOEC electrolyzers are almost equally divided between catalytic and biological methanation, on the other hand, there is a high percentage of alkaline electrolyzers that are coupled with a catalytic methanation system. This behavior is presumably due to an easier techno-economic handling of the components.

#### 4.2. Research Targets

The implementation of a demonstration plant or an experimental or a research and development activity is characterized by well-defined research objectives that contribute to the overall knowledge of the topic. These research objectives were grouped into categories of research targets, as illustrated in Section 3 regarding the methodology adopted.

With reference to the categories listed in Section 3, Figure 8 shows the distribution of the considered projects within the research targets.

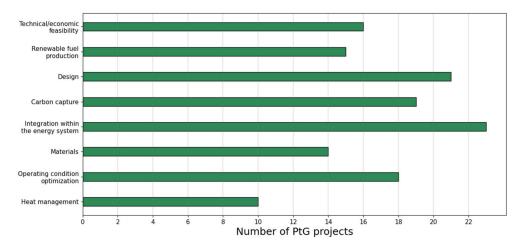
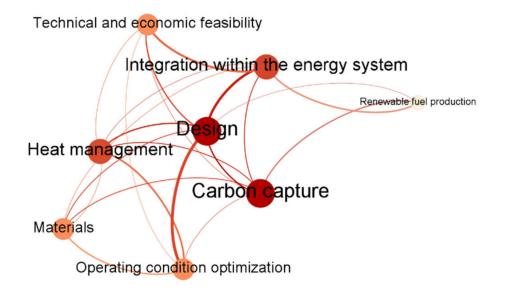
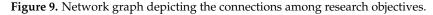


Figure 8. Number power-to-gas projects per research target.

The distribution is fairly homogeneous, proving that, at present, there is a need for improvement in all directions.

To gain a more comprehensive view, it might be useful to analyze these categories by means of a network graph. Figure 9 refers to the projects with research objectives that fall into multiple categories only. The size of the nodes represents the number of projects falling into that category while the thickness of the connecting lines represents the number of projects that share those two categories. In this way, it is possible to qualitatively assess which research lines are most closely related so they can be dealt with in the same project.





It can be seen that the objectives of carbon capture and design are among the most frequent in situations where multiple targets are combined. The opposite is the case where the objective is to produce a renewable fuel. This graph reveals, for instance, that in cases where design is one of the targets, often there is also the presence of investigations into the optimization of the operating conditions or the attempt to integrate PtG into a broader energy system. However, the tendency is that there is no preferential relationship between two categories in particular. Each node, in fact, has connections with almost all the others, and there are few cases of combinations of targets that are never treated together.

In the following, each of these research targets is considered in more detail.

#### 4.2.1. Heat Management

In a system such as power-to-gas, accurate heat management is of crucial importance for proper and efficient operation. In fact, as previously mentioned, the system components are generally characterized by a certain amount of waste heat power, whether due to dissipation or exothermic reactions.

Specifically concerning this issue, in [26,27] the plant design focuses on waste heat recovery. Similar activity takes place in [28] where, in particular, the thermal energy coming from the electrolyzer and from the methane compression phase is used to optimize the organic process in the fermenter.

In [29,30], the focus is on the inclusion of heat exchangers in the system. In [29], a plate-and-shell condenser is inserted for the removal of reaction steam. In [30], instead, within the *CoSin* project, it is the methanation reactor itself (microstructured-type) that fulfills the function of a heat exchanger and, within it, the direct utilization of biogas manages to reduce the presence of hot spots. In addition, the system includes preheating of the gases entering the reactor and a mechanism for condensation.

*El upgraded biogas* [31,32] and *HELMETH* [12] deal with PtG systems that make use of high-temperature electrolyzers (SOEC type). Therefore, in both cases, it was decided to produce the electrolyzer's feed steam by means of methanation heat. In the case of *HELMETH*, this achieves an overall system efficiency of 76%.

There is no shortage of studies on heat management within components. In [33], the heat transfer of a bubbling fluidized bed reactor is studied both by CFD simulation and experimentally. In addition, the effect of hydrodynamics and reaction kinetics on the heat

transfer coefficient is identified. The authors in [34] address the thermal regeneration of absorber material for the direct capture of carbon dioxide from air. Lastly, in [35], regarding a microchannel reactor, the use of hydrogen as a cooling agent for the reactor, in addition to its function as a reactant, is evaluated.

#### 4.2.2. Operating Condition Optimization

Managing several interconnected components, the study of power-to-gas plants cannot ignore the pursuit of optimization of the conditions under which they are to be operated. Often, the optimization concerns the methanation phase in order to have the highest possible quality and yield possible, but there is no lack of research on the optimization of the electrolysis process within a system with the characteristics of PtG.

The effect of temperature, stoichiometric ratio of reactants, pressure, and gas hourly space velocity is investigated in [29,36,37], within the  $CO_2$ -SNG pilot plant [38], in [39], in [40] within the project *PID Eng&Tech*, and in [41,42] in *Power-to-Gas 250* with the aim of achieving maximum reaction yield and gas quality. In these cases, these parameters (whether some or all of them) are calibrated in such a way that the methanation reactor is optimally operable in the power-to-gas context.

In [43] specifically, the operating parameters are adjusted to achieve optimal carbon dioxide conversion in terms of methane purity and selectivity. All this is performed in a bench-scale fluidized bed reactor. In [30,44] as part of the *CoSin* project, the focus is on the purity of the methane output and specifically on obtaining a gas capable of meeting the requirements for being fed into the grid. In [45], instead, part of the *COSYMA* project, the operating conditions are investigated for good management of the methanation reactor during the long term and to avoid deactivation of the catalyst.

Significant research efforts are also dedicated to biological reactors. In this case, in addition to the parameters discussed for catalytic reactors [46], it is also necessary to monitor other values such as the pH of the reaction environment and the nutrients for the bacteria, as can be seen in [47] from the *SYMBIO* project and in [48].

In [49], however, optimal conversion is studied under more particular conditions because the archaeal bacteria are immobilized on a biofilm. Furthermore, in [50] by means of a biotrickling filter, faster methane production rates are claimed to be achieved than with other bioreactors.

The operating conditions of electrolyzers applied to power-to-gas are mainly investigated in high-temperature electrolysis cells as the level of technological maturity is lower in comparison with low-temperature cells. Specifically, in *HELMETH* [12] the conditions of an SOEC are examined in terms of steam conversion and energy demands under varying pressure. In [51], instead, co-electrolysis with subsequent methanation of the syngas produced is implemented. In this case, the operating conditions of the two components were modified individually in order to improve the selectivity for synthetic natural gas production.

#### 4.2.3. Materials

From the literature analysis that was carried out, a considerable amount of activity was noticed concerning the study of materials. These mainly concern catalysis of the methanation phase, degradation, sorbents for carbon dioxide, and water.

Catalysts for applications in power-to-gas are studied in [52–54]. In [52], a honeycomb catalyst is developed as part of the *EE-Methan aus CO*<sub>2</sub> project to process carbon dioxide from flue gases; the aim is to increase the life of the catalyst and avoid poisoning problems. In [53], a powder catalyst is developed by varying the nickel content in order to achieve increased interaction with the substrate. In [54], a comparison is made between a homemade catalyst and various commercial ones. Within the *COSYMA* project [45], moreover, the long-term deactivation of the catalyst is investigated.

The study on catalyst supports is also of equal importance, as shown by the activities described in [40,55,56]. Since the methanation reaction is exothermic, the stability of the catalyst could soon become compromised without adequate support.

The capture of carbon dioxide is analyzed in [34] where direct air capture is implemented using a  $K_2CO_3/Al_2O_3$  composite sorbent, and in [57], where the performance of a dual function material is evaluated, which handles both the capture of carbon dioxide and the methanation step. Another bifunctional material is employed in [58] in which it acts as both catalyst and water removal. The removal of water also takes place in [59] as part of the *EDGaR Synthetic methane* project and within the *PID Eng&Tech* project in [39] in which sorption-enhanced methanation is implemented via adsorbers based on zeolites or other materials. In contrast, in [60], water is removed with thin-film composite membranes within a catalytic membrane reactor.

Other studies focus on the development of metal honeycomb catalysts for better heat dissipation as in *STORE&GO Germany* [61], on the degradation affecting the materials of a solid oxide electrolysis cell during the long term [62], and on the development of special filters for biological methanation [50].

#### 4.2.4. Carbon Capture

The possibility of enabling carbon dioxide capture, alongside an option for long-term storage, is one of the major strengths of power-to-gas technology. For this reason, many of the projects evaluated have carbon capture as a major issue.

A number of studies concerns carbon dioxide recycling from flue gases. This is carried out in [36,57], in [63] within the CCU P2C Salzbergen project, and in [26] as part of the  $CO_2$  Conversion to Methane project. In [20,21] from the project Reduction & reuse of  $CO_2$ : renewable fuels for efficient electricity production, carbon dioxide is collected from a nearby cement plant, while in Jupiter 1000 [24] it comes from a nearby facility, and in [64] carbon dioxide is recycled from the flue gases of a conventional power plant. The utilization of flue gases from lignite combustion in a power plant is studied in CO2RRECT [65] and in [66] as part of the  $CO_2$ -Methanation of flue gas project. In [67], within the BIT3G project, in addition to implementing carbon dioxide recycling, the carbon footprint of the entire process is evaluated.

An interesting step forward has been taken in [19,68–70] in which carbon dioxide is obtained directly from the combustion of the gas produced. This makes the process increasingly circular and self-sufficient.

In contrast, in [71,72] the intention is to take carbon dioxide from biogas by means of gas cleaning and reuse it to further lower emissions.

If it is not possible to have a source such as those mentioned above, carbon dioxide can be extracted from the atmosphere via DAC, although it is present in low concentrations. This route has been pursued in [34,73,74].

#### 4.2.5. Design

In the power-to-gas sector, as the plants have not yet reached full technical maturity, their design lies within the research objectives. The design may concern the whole plant or part of it.

One of the components that has received the most emphasis is the methanation reactor. As far as catalytic reactors are concerned, they are studied in [75] within the *EDGaR Synthetic methane* project, and in [51] where the reactor is coupled with a co-electrolysis system and prototypes are also built in [44,70]. In [26], the design of the system includes both carbon capture and methanation.

Biological reactors are not far behind, in fact, it is found that in [49] a new type of trickle bed reactor is used for biological methanation, in [23] a prototype of a bioelectrochemical system for electromethanogenesis is constructed, a biocatalytic reactor is also developed in [76] as part of the *P2G-BioCat* project, and in [48] there is the implementation of a thermophilic anaerobic trickle bed reactor. In addition, there is also a fixed-bed bioreactor constructed from low-cost materials [46].

Undoubtedly, however, it is noticeable that, in general, the main focus is on the power-to-gas system as a whole. The first example was in 1995 at Tohoku University [19],

where a photovoltaic-powered seawater electrolyzer, a combustion flue gas carbon dioxide removal system, and a catalytic methanation reactor were used. Subsequently, several similar plants were proposed. In terms of those that can be defined as demonstration or pilot projects, it is worth mentioning *EE-Methan aus CO*<sub>2</sub> [52], *El upgraded biogas* [31], *RENOVAGAS* [22], *Towards the Methane Society* [77], *CO*<sub>2</sub>-*SNG pilot plant* [38], and *UC Irvine power-to-gas* (*P2G*) [78]. Moreover, *DemoSNG* can be highlighted since the plant was placed in a shipping container [79].

On laboratory-scale level, in [80] a process for the conversion of solar energy to hydrocarbons is proposed, in [81] a power-to-gas process coupled with an oxycombustion is implemented, and in [29], the system has been integrated into a mobile apparatus.

#### 4.2.6. Renewable Fuel Production

Many of the power-to-gas projects presented here allow the production of sustainable synthetic fuels that can be used in various applications.

A primary application is the development of fuels for mobility. The goal of clean mobility is stated in the *BioPower2Gas* [72] and in *W2P2G* [82]. In *Power to Gas biogas booster* [83], the aim is to convert 69 municipal vehicles to be fueled by green natural gas, while in [84], within the *Audi e-gas* project, the aim is to produce synthetic compressed natural gas to power a specific line of cars. Again, in *SYNFUEL* [85], the purpose is to produce sustainable fuels, and in [86], regarding the *Minerve* project, the aim is to produce synthetic compressed natural gas for transportation or boilers.

In two of the cases evaluated, the objective is to produce liquefied natural gas: *Ingrid— STORE&GO* [73] and *Energieversorgung Lübesse* [87].

In [88,89], as part of *RENERG*<sup>2</sup>, the aim is to synthesize a hydrogen/compressed natural gas blending to be used in combined heat and power systems, and in *Smart Grid Labor* [90], the fuel is used in combined heat and power systems or for long-term storage. In [22,91,92], the objective is instead to feed gas into the distribution network.

Lastly, in [93], the production of propellant on Mars is identified as a possible application, and in [67], within the *BIT3G* project, the production of fuels in the form of both methane and ammonia is identified as a possible application.

#### 4.2.7. Integration within the Energy System

The collection of papers obtained during this research shows that there is great focus on the integration of the power-to-gas plants into broader energy system networks, so that their full potential can be exploited.

Among the most studied topics, the coupling of the electrolyzer with nonprogrammable renewable sources seems to play a significant role. In [94], regarding the *UC Irvine power-to-gas (P2G)* demonstration project, the dynamic behavior of a PEM electrolyzer coupled with renewable sources is examined. The integration with photovoltaic is investigated in [95], while the coexistence of a PtG with renewables and other storage systems is evaluated in *Small-Scale Demonstrator in Sion* [80] and in the *Morbach* project [96]. In [97], within the *EDGaR Synthetic methane* project, the trend of both energy surplus and deficit is analyzed. In [98–100], the inclusion of PtG in the system is evaluated with reference to the grid-balancing issue.

In *Audi e-gas* [84], in addition to evaluating the utilization of surplus energy from an offshore wind farm, heat is transferred to a nearby biogas plant, which in return supplies carbon dioxide. The idea of implementing system integration in order to gain access to carbon dioxide is also put into practice in *bioCONNECT* [101] and *DemoSNG* [79], in which there is a relationship with a bioethanol plant and a biomass gasification plant, respectively. The same strategy is implemented with biogas plants in [102,103] and in *Methanation at Eichhof* [104]. In the latter, moreover, the methane produced is converted back into electricity that is sold to the grid. In addition, it is reported that in [21], carbon dioxide is taken from a cement factory and in *Swisspower Hybridkraftwerk* [92] from a wastewater treatment plant.

A further option is to exploit the oxygen produced by electrolysis for other uses. Specifically, in [81] it is employed to implement oxyfuel combustion, which also occurs in the *Exytron* demonstration project [68] where oxyfuel combustion is implemented in a gas turbine to produce electricity. In the latter, there is also waste heat recovery, which is used for domestic hot water production and space heating. Furthermore, in *SYNFUEL* [85] it is used for gasification.

The integration with a district heating plant is studied in *El upgraded biogas* [31], in *Power-to-Gas project in Rozenburg* [105] where the gas is burned in a boiler that provides heat to nearby buildings, and in *Hybrid power plant Aarmatt—STORE&GO Switzerland* [106] where a strong interconnection between the electricity, water, gas, and district heating networks is investigated.

Furthermore, power-to-gas plants are integrated into renewable energy communities in *Energieversorgung Lübesse* [87] and in *INFINITY1* [107].

#### 4.2.8. Technical and Economic Feasibility

Common to all demonstration plants, the technical–economic feasibility in some projects takes center stage among the research objectives.

The intention to make power-to-gas economically viable is found in [108,109] and in *Methfuel* [110]. The *bioCONNECT* project [101] exploits the balancing market and savings given by the European Union Allowances share regarding carbon dioxide. Grid balancing is also explored in *Power to gas Hungary* [111], while in *El upgraded biogas* [31] the challenge is to make a plant using SOEC viable.

The feasibility is investigated from a technical point of view in [105] and in *CO2RRECT* [65]. Specifically, in the latter, the off-design behavior of electrolyzers is studied and the performance of the catalyst when treating flue gas is evaluated. Furthermore, in the *HyCAUNAIS* project [112], the dynamic operation is carried out to ensure that the variability of the power supply is followed. The profitability of a PtG plant with biocatalytic methanation is investigated in *ORBIT* [113] and in *P2G-Foulum* project [114].

Still in relation to this type of plant, in [99,100] both a technical and economic evaluation is presented as the performance is analyzed to determine whether biological methanation is commercially viable. Techno-economic evaluations are also carried out in [80,115] and in *Power to Gas* 250 [41,42], where the process is optimized both to obtain a good gas quality output and to minimize costs. In addition, in [116] of *MethyCentre* environmental aspects are also studied, and the regulatory aspects are investigated in *P2G-BioCat* [76].

#### 4.3. Discussion

From the previous paragraphs, it can be seen that studies in the PtG field are greatly diversified, starting from component tests to the investigation of the integration of the plant in the energy system. Undoubtedly, this has led to a considerable development of this technology. Nevertheless, it is also important to assess the limitations that distinguish these projects.

A major limitation found in the projects analyzed is that improvements are often not tested during long-run operation. Often, operational problems that were not previously foreseen appear only in the long term.

Furthermore, the wide variety of components used does not allow a standard to be defined and thus a benchmark to be set. Through the latter, improvements could be quantified and compared.

Considering a more specific case, it could be possible to discuss heat management. In this context, the coupling of a PtG plant with thermal users of various types on both the residential and industrial levels could be further explored. Selling excess thermal energy could lead to a reduction in operational costs whose real impact would be interesting to investigate.

It is precisely the assessment of the real impacts, especially those that cannot be assumed a priori, that is the most important driver of experimental-type research and makes this necessary to give adequate maturity to a technology. It appears from the analysis above that, at present, there is still ground to cover.

#### 5. Conclusions

Studies on power-to-gas technology have been increasing significantly over the last few years. The intense research on the subject is justified by the multiple advantages that PtG can offer. Beyond the synthesis of a green fuel, there is the possibility of long-term storage, carbon sequestration, and a contribution to the energy-balancing market. All these goals can be reached, producing a well-known energy carrier that is renewable and that can be used for many applications.

This paper analyzes plants and experimental activities that are currently in place and highlights the main research directions on this topic.

It should be pointed out that most of the 87 projects that have been reviewed started in Europe after 2010, testifying the current innovative nature of this technology. Although plant sizes are still relatively small (most projects are characterized by a power output lower than 100 kW), there is a great variety in the technologies used for each process. It is found, indeed, that in addition to the best-known technologies, there are also many research activities on alternative and original technologies. This finding can be observed in the technologies of each stage of the process under study.

What can be deduced from this investigation is that power-to-gas research objectives span many areas. Great efforts can be seen regarding the study of materials, especially for the catalysis of methanation, and regarding the optimization of the conditions under which the various components operate. There is no shortage of projects that aim to assess the techno-economic feasibility, to tackle design issues, or to produce a green synthetic fuel. However, it is found that most projects aim to increase the efficiency of the system, both through the proper use of waste heat and, above all, through the correct integration of the power-to-gas process into energy networks.

Several projects, in fact, exploit interconnection with renewable energy plants; some use industrial facilities for the supply of carbon dioxide with an advantage on both sides, while others have been able to exploit waste heat in district heating networks.

Nonetheless, it can be seen that in many projects, research has developed involving several different areas, which proves that power-to-gas technologies require a strong multidisciplinary approach and a wide range of expertise.

What emerges most of all, however, is that this technology is currently still innovative and therefore further theoretical and experimental studies and more technical experience are needed.

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## Abbreviations

ARAID	Fundacion Agencia Aragonesa para La Investigacion y El Desarrollo
AEM	Anionic exchange membrane
BMWi	Bundesministerium für Wirtschaft und Energie
BTS	Bayer Technology Services
BTU	Brandenburgische Technische Universität Cottbus
CEA	Commissariat à l'énergie atomique et aux énergies alternatives
CFD	Computational fluid dynamics
CNR-ITAE	Consiglio Nazionale delle Ricerche-Istituto di Tecnologie Avanzate per l'Energia
CoSPE	Center of Sustainable Process Engineering
DAC	Direct air capture
DNV	Det Norske Veritas
DTU-Environment	Danmarks Tekniske Universitet
DVGW	German Technical and Scientific Association for Gas and Water
ECN	Energy Research Centre of the Netherlands
EMPA	Eidgenössische Materialprüfungs- und Forschungsanstalt
EnBW	Energie Baden-Württemberg
	Agenzia nazionale per le nuove tecnologie, l'energia e lo
ENEA	sviluppo economico sostenibile
EPFL	Ecole Polytechnique Federale de Lausanne
Fraunhofer ISE	Fraunhofer-Institut für Solare Energiesysteme
Fraunhofer IWES	Fraunhofer-Institut für Windenergiesysteme
HSR	Hochschule für Technik Rapperswil
	Institute of Catalysis and Petrochemistry—Consejo Superior de
ICP-CSIC	Investigaciones Científicas
INSTM	National Interuniversity Consortium of Materials Science and Technology
IREC	Catalonia Institute for Energy Research
JKU Linz	Johannes Kepler University Linz
KIT	Karlsruhe Institute of Technology
NFCRC	National Fuel Cell Research Center
NREL	National Renewable Energy Laboratory
PEC	Photoelectrochemical water splitting
PEM	Proton exchange membrane
PFI technical centre	Prüf- und Forschungsinstitut Pirmasens e.V.
PSI	Paul Scherrer Institut
PSL—Research University	Université Paris Sciences et Lettres
PtG	Power-to-gas
PTTEP	PTT Exploration and Production Public Company Limited
PV	Photovoltaic
SDU	Syddansk Universitet
SIAD	Società Italiana Acetilene e Derivati
SoCalGas	Southern California Gas Company
SOEC	Solid oxide electrolytic cell
TKI Gas	Turkish Coal Operations Authority
TU Clausthal	Technische Universität Clausthal
TU Delft	Technische Universiteit Delft
TU Munich	Technische Universität München
TU Wien	Technischen Universität Wien
UCI	University of California at Irvine
ZHAW	Zürcher Hochschule für Angewandte Wissenschaften
	Zactor Electoriale ful filige malate model of fullen

## Appendix A

This appendix provides a structured tabulated view of the data found. Specifically, Table A1 presents all the general information of the projects (e.g., start date, company or university involved, location, etc.) together with some technical remarks (e.g., typology of the technology for electrolysis and methanation, sources for carbon dioxide and electricity, etc.). In Table A2, the research targets of the various projects are listed.

Project Name	Location	Country Code	Company/University	Start Date	Electrolyzer Power (kWel)	Electrolysis Type	Electricity Source	Methanation Type	CO <sub>2</sub> Source	Ref.
Activity and stability of powder and monolith-coated NiGDC catalysts for CO <sub>2</sub> methanation	Messina	ITA	CNR-ITAE, University "Mediterranea" of Reggio Calabria, INSTM	'Mediterranea" of Reggio 2017 Catalytic		Bottled CO <sub>2</sub>	[53]			
Audi e-gas	Wertle	DEU	Audi, ETOGAS GmbH, McPhy	2013	6300	Alkaline	Wind	Catalytic (fixed bed)	Biogas	[84]
Biocatalytic methanation of hydrogen and carbon dioxide in a fixed bed bioreactor	Helsinki	FIN	Natural Resources Institute Finland	2015	n/a	PEM	n/a	Biological	Bottled CO <sub>2</sub>	[46]
Biocatalytic methanation of hydrogen and carbon dioxide in an anaerobic three-phase system	Cottbus	DEU	Brandenburg University of Technology Cottbus-Senftenberg	2014	n/a	n/a	n/a	Biological	Biogas	[49]
bioCONNECT	Lemgo	DEU	Technische Hochschule Ostwestfalen-Lippe	2016	n/a	PEM	Renewable	Biological	Flue gas	[101]
Bioelectrochemical systems for energy storage: A scaled-up power-to- gas approach	Terrassa	ESP	LEITAT Technological Center, Universitat Politècnica de Catalunya, Universitat Autònoma de Barcelona	2019	n/a	Biological	Grid	Bioelectrochemical	Biogas	[23]
BioPower2Gas	Allendorf	DEU	Microbenergy, Viessmann, Schmack	2014	400	PEM	Waste treatment	Biological	Biogas	[72]
BIT3G project	Perugia	ITA	University of Perugia, TU Delft	2018	n/a	PEM	Renewable	Catalytic	Flue gas	[67]
Catalytic methanation of carbon dioxide captured from ambient air	Novosibirsk	RUS	Boreskov Institute of Catalysis, Novosibirsk State University	2018	-	Bottled hydrogen	-	Catalytic	Air	[34]
CCU P2C Salzbergen	Salzbergen	DEU	H&R Chemisch Pharmazeutische Spezialitäten GmbH, BMWi	2024	n/a	n/a	Renewable	n/a	Flue gas	[63]
CO <sub>2</sub> Conversion to Methane Project	Rayong province	THA	PTTEP, Hitachi Zosen	2015	n/a	Alkaline	Renewable	Catalytic (fixed bed)	n/a	[26]
CO <sub>2</sub> methanation in a bench-scale bubbling fluidized bed reactor using Ni-based catalyst and its exothermic heat transfer analysis	Daejeon	KOR	Korea Institute of Energy Research, Korea Electric Power Corporation Research Institute	2020	-	Bottled hydrogen	-	Catalytic (bubbling fluidized bed)	Bottled CO <sub>2</sub>	[43]

**Table A1.** General information on the selected projects (n/a stands for information not available).

Project Name	Location	Country Code	Company/University	Start Date	Electrolyzer Power (kWel)	Electrolysis Type	Electricity Source	Methanation Type	CO <sub>2</sub> Source	Ref.
CO <sub>2</sub> recycling by reaction with renewably generated hydrogen	Reno	USA	Desert Research Institute	2009	1	PEM	PV	Catalytic	Bottled CO <sub>2</sub>	[36]
CO <sub>2</sub> -Methanation of flue gas	Brandenburg	DEU	BTU, Panta Rhei	2013	-	Bottled hydrogen	n/a	Catalytic (fixed bed)	Flue gas	[66]
CO2RRECT	Niederaußem	DEU	BTS, RWE Power, Siemens	2013	100	PEM	Renewable	Catalytic	Flue gas	[65]
CO <sub>2</sub> -SNG pilot plant	Łaziska	POL	Institute for Chemical Processing of Coal, TAURON Wytwarzanie S.A.	2019	122	Alkaline	Grid	Catalytic (microchannel)	Flue gas	[38]
CoSin project	Sant Adria de Besos	ESP	IREC, Ineratec GmbH, Accio	2018	37	Alkaline	Renewable	Catalytic	Biogas	[30,117]
COSYMA	Werdhölzli (Zürich)	CHE	PSI	2017	n/a	n/a	Renewable	Catalytic (bubbling fluidized bed)	Biogas	[45]
DemoSNG	Köping	SWE	DVGW, KIT	2014	50	PEM	Renewable	Catalytic (honeycomb catalyst)	Biomass gasification	[79]
Dual function materials for CO <sub>2</sub> capture and conversion using renewable H2	New York	USA	Columbia University in the City of New York	2014	-	Bottled hydrogen	Renewable	Catalytic (fixed bed)	Flue gas	[57]
Economic assessment of a power-to-substitute-natural- gas process including high-temperature steam electrolysis	Grenoble	FRA	CEA, PSL—Research University	2015	n/a	SOEC	n/a	n/a	n/a	[62]
EDGaR synthetic methane project	n/a	NLD	ECN, TU Delft, Hanze Technical University	2015	n/a	SOEC	n/a	Catalytic (fixed bed)	Biomass gasification	[59,75,97,118]
EE-Methan aus CO <sub>2</sub>	Leoben	AUT	TU Wien, JKU Linz, Montanuniversität Leoben	2015	-	Bottled hydrogen	Renewable	Catalytic (honeycomb)	Biogas	[10]
Einsatz der Biologischen Methanisierung für Power-to-Gas-Konzepte: Hochdruckmethanisierung von H2	Hohenheim	DEU	University of Hohenheim, KIT	2016	n/a	n/a	n/a	Biological	Biogas	[108,109]
El upgraded biogas	Foulum	DNK	Haldor Topsøe	2016	50	SOEC	Grid	Catalytic	Biogas	[31,32]
Energieversorgung Lübesse	Lübesse	DEU	Exytron, Lübesse Energie GmbH	2023	10000	Alkaline	Wind	Catalytic	Flue gas, biogas	[87]
Experiment and numerical analysis of catalytic CO <sub>2</sub> methanation in bubbling fluidized bed reactor	Anseong	KOR	CoSPE, Korea Institute of Energy Research, Korea National University of Transportation	2021	-	-	-	Catalytic (bubbling fluidized bed)	Bottled CO <sub>2</sub>	[33]

Project Name	Location	Country Code	Company/University	Start Date	Electrolyzer Power (kWel)	Electrolysis Type	Electricity Source	Methanation Type	CO <sub>2</sub> Source	Ref.
Experimental analysis of photovoltaic integration with a proton exchange membrane electrolysis system for power-to-gas	Irvine	USA	University of California	2017	7	PEM	PV	-	-	[95]
Exytron demonstration project	Rostock	DEU	Exytron	2015	21	Alkaline	PV	Catalytic	Flue gas	[68]
Forschungsanlage am Technikum des PFI	Pirmasens	DEU	TU Clausthal, PFI Technical Centre	2016	2500	Alkaline	Renewable	Biological	Biogas	[102,119]
HELMETH	Karlsruhe	DEU	KIT, Sunfire GmbH	2018	15	SOEC	n/a	Catalytic (fixed bed)	n/a	[12]
High performance biological methanation in a thermophilic anaerobic trickle bed reactor	Garching	DEU	TU Munich, Bavarian State Research Center for Agriculture	2017	-	Bottled hydrogen	-	Biological	Bottled CO <sub>2</sub>	[48]
High-Performance Biogas Upgrading Using a Biotrickling Filter and Hydrogenotrophic Methanogens	Durham	USA	Duke University	2017	-	Bottled hydrogen	-	Biological	Bottled CO <sub>2</sub>	[50]
Hybrid power plant Aarmatt—STORE&GO Switzerland	Aarmatt	CHE	Regio Energie	2015	350	PEM	PV	Biological	Biogas	[106]
Hybrid power plant Falkenhagen—STORE&GO Germany	Falkenhagen	DEU	Uniper Energy Storage GmbH, KIT	2013	2000	Alkaline	Wind	Catalytic (honeycomb)	Flue gas	[61]
HyCAUNAIS Project	Saint- Florentin	FRA	Storengy, AREVA H2Gen, University of Franche Comté, Engie	2019	1000	PEM	Wind	Biological	Biogas	[112]
INFINITY 1	Pfaffenhofen a. d. Ilm	DEU	n/a	2020	1000	PEM	Renewable	Biological	Biogas	[107]
Ingrid—STORE&GO Italy	Troia	ITA	Engineering, Hydrogenics, Climeworks	2017	1000	PEM	Wind, PV	Catalytic (micromethanation)	Air	[73]
Integrated Co-Electrolysis and Syngas Methanation for the Direct Production of Synthetic Natural Gas from CO <sub>2</sub> and H <sub>2</sub> O	Aachen	DEU	Aachen University, Eichel Institut für Energie	2021	n/a	SOEC	Renewable	Catalytic (fixed bed)	n/a	[51]
Intensification of catalytic CO <sub>2</sub> methanation mediated by in situ water removal through a high-temperature polymeric thin-film	Valencia	ESP	Universitat Politecnica de Valencia, Institute of Membrane Research, University of Twente	2021	-	Bottled hydrogen	-	Catalytic	Bottled CO <sub>2</sub>	[60]

Project Name	Location	Country Code	Company/University	Start Date	Electrolyzer Power (kWel)	Electrolysis Type	Electricity Source	Methanation Type	CO <sub>2</sub> Source	Ref.
Jupiter 1000	Fos-sur-Mer	FRA	GRTgaz	2018	1000	Alkaline/PEM	Renewable	Catalytic	Flue gas	[24]
Klimafreundliches Wohnen	Augsburg	DEU	Exytron	2019	52	Alkaline	PV	Catalytic	Flue gas	[69]
Laboratory-scale experimental tests of power to gas-oxycombustion hybridization system design and preliminary results	Zaragoza	ESP	Universidad de Zaragoza, ARAID	2021	n/a	PEM	PV	Catalytic (fixed bed)	Flue gas	[81]
Laboratory-scale reactor in Fraunhofer IWES	Kassel	DEU	Fraunhofer IWES	2016	25	Alkaline	Wind	Catalytic (fixed bed)	Air	[98]
MeGa-stoRE	Lemvig	DNK	Aarhus University, GreenHydrogen	2013	6	Alkaline	Renewable	Catalytic	Biogas	[71]
Methanation at Eichhof	Bad Hersfeld	DEU	Eichhof Agricultural Training and Research Center, Fraunhofer IWES	2012	25	PEM	Renewable	Catalytic	Biogas	[104]
Methanation of carbon dioxide by hydrogen reduction using the Sabatier process in microchannel reactors	Richland	USA	Pacific Northwest National Laboratory, Colorado School of Mines	2006	-	Bottled hydrogen	-	Catalytic (microchannel)	Bottled CO <sub>2</sub>	[93]
Methanation of recovered oxyfuel-CO <sub>2</sub> from Ketzin and of flue gas emitted by conventional power plants	Cottbus	DEU	BTU Cottbus	2015	n/a	n/a	n/a	Catalytic (fixed bed)	Flue gas	[64,120]
Methanation potential suitable catalyst and optimized process conditions for upgrading biogas to reach gas grid requirements	Flensburg	DEU	Hochschule Flensburg	2019	-	Bottled hydrogen	-	Catalytic	Biogas	[44]
MethFuel	Frankfurt	DEU	Elogen, Infraserv Höchst, iGas energy	2020	1000	PEM	Renewable	Catalytic (bubble column reactor)	Flue gas, Air	[110]
MethyCentre	Angé	FRA	Storengy	2021	250	PEM	Grid	Catalytic (millistructured)	Biogas	[116]
Minerve	Nantes	FRA	AFUL Chantrerie, GRTgaz, Polytech Nantes	2018	12	PEM	PV	Catalytic (fixed bed)	Bottled CO <sub>2</sub>	[86]
Modeling of Laboratory Steam Methane Reforming and CO <sub>2</sub> Methanation Reactors	Genova	ITA	University of Genova, INSTM	2020	-	n/a	-	Catalytic	n/a	[54]

Project Name	Location	Country Code	Company/University	Start Date	Electrolyzer Power (kWel)	Electrolysis Type	Electricity Source	Methanation Type	CO <sub>2</sub> Source	Ref.
Morbach	Morbach	DEU	EtoGas, ZSW, juwi	2011	25	Alkaline	Wind, PV	Catalytic	Biogas	[96]
ORBIT	Ibbenbüren	DEU	Universität Regensburg, Friedrich Alexander University	2020	1	n/a	PV	Biological	Biogas	[113]
P2G movable modular plant operation on synthetic methane production from CO <sub>2</sub> and hydrogen from renewable sources	Casaccia	ITA	ENEA, Politecnico di Milano	2019	-	Bottled hydrogen	-	Catalytic (fixed bed)	Bottled CO <sub>2</sub>	[29]
P2G Solar Energy Storage RD&D	Golden	USA	NREL, SoCalGas	2014	250	PEM	PV	Biological	Bottled CO <sub>2</sub>	[99,100]
P2G-BioCat	Avedøre	DNK	Electrochaea, Hydrogenics	2016	1000	Alkaline	Renewable	Biological	Biogas	[76]
P2G-Foulum Project	Foulum	DNK	Electrochaea, E.ON	2013	250	n/a	Renewable	Biological	Biogas	[114]
PEGASUS Project	n/a	ITA	ENEA, Società Gasdotti Italia SpA, SIAD	2019	n/a	n/a	Renewable	Catalytic	Biogas	[91]
Performance analysis of Sabatier reaction on direct hydrogen inlet rates based on solar-to-gas conversion system	Miyazaki	JPN	University of Miyazaki	2021	n/a	PEM	PV	Catalytic	Bottled CO <sub>2</sub>	[37]
PID Eng&Tech	Bilbao	ESP	University of the Basque Country	2021	-	Bottled hydrogen	-	Catalytic	Bottled CO <sub>2</sub>	[39,40,55]
Pilot plant—Tohoku Institute of Technology	Sendai	JPN	Tohoku University	1998	n/a	Alkaline	PV	Catalytic	Flue gas	[19]
Power to Flex	Groningen	NLD	Provincie Groningen, Nordwest Gruppe, Oosterhof Holman	2016	n/a	Alkaline	Renewable	Catalytic	Biogas	[28]
Power to Gas 250	Stuttgart	DEU	ETOGAS, ZSW, Fraunhofer IWES	2012	250	Alkaline	Renewable	Catalytic	n/a	[41,42]
Power to Gas at Eucolino	Schwandorf	DEU	Viessmann, Microbenergy, Schmack	2012	108	n/a	n/a	Biological	Biogas	[27]
Power to Gas biogas booster	Straubing	DEU	MicroPyros	2015	10	n/a	Renewable	Biological	Biogas	[83]
Power to gas BTU Cottbus	Nordhackstedt	DEU	BTU, Flensburg University of Applied Sciences	2012	500	Alkaline	Wind	Biological	Biogas	[103]
Power to gas Hungary	n/a	HUN	Electrochaea, MVM, University of Chicago	2017	10000	n/a	n/a	Biological	Biogas	[111]

Project Name	Location	Country Code	Company/University	Start Date	Electrolyzer Power (kWel)	Electrolysis Type	Electricity Source	Methanation Type	CO <sub>2</sub> Source	Ref.
Power-to-Gas project in Rozenburg	Rozenburg	NLD	DNV, TKI Gas	2014	8,3	PEM	PV	Catalytic (fixed bed)	Bottled CO <sub>2</sub>	[105]
Power-to-Methane HSR	Rapperswil	CHE	HSR, Climeworks	2015	25	Alkaline	PV	Catalytic	Air	[74]
ProGeo	Perugia	ITA	University of Perugia, ENEA	2019	20	n/a	-	Catalytic	Flue gas	[70,121]
Pure methane from CO <sub>2</sub> hydrogenation using a sorption-enhanced process with catalyst-zeolite bifunctional materials	Delft	NLD	Delft University of Technology, Åbo Akademi University	2021	-	Bottled hydrogen	-	Catalytic	Bottled CO <sub>2</sub>	[58]
Reduction and reuse of CO <sub>2</sub> : renewable fuels for efficient electricity production	Zurich	CHE	ZHAW, EMPA	2014	n/a	PEC	n/a	Catalytic (fixed bed)	Flue gas	[21]
RENERG <sup>2</sup>	Villigen	CHE	PSI, ZHAW, EMPA	2016	100	PEM	PV	Catalytic	Biomass gasification	[88,89]
RENOVAGAS Project	Jerez de la Frontera	ESP	Enagas, ICP-CSIC	2014	15	Alkaline (anionic exchange membrane)	Renewable	Catalytic (microchannel)	Biogas	[22]
Small-Scale Demonstrator in Sion	Sion	CHE	EPFL, EMPA	2017	3,6	PEM	PV	Catalytic	Bottled CO <sub>2</sub>	[80]
Smart Grid Labor	Hamburg	DEU	Hamburg University of Applied Sciences	2015	n/a	PEM	n/a	Biological	n/a	[90]
Storage of electric energy from renewable sources in the natural gas grid—water electrolysis and synthesis of gas components	Baden- Wurtemberg	DEU	DVGW, EnBW, Fraunhofer ISE	2011	6	PEM	Wind, PV	Catalytic (fixed bed)	n/a	[115]
Study of the role of chemical support and structured carrier on the CO <sub>2</sub> methanation reaction	Fisciano	ITA	University of Salerno	2018	-	Bottled hydrogen	-	Catalytic	Bottled CO <sub>2</sub>	[56]
Swisspower Hybridkraftwerk	Diekiton	CHE	Swisspower, Limeco	2021	2000	PEM	Waste treatment	Biological	Flue gas	[78,92]
SYMBIO	Lyngby	DNK	DTU-Environment, SDU	2014	n/a	n/a	Wind	Biological	Biogas	[47]
SYNFUEL	Lyngby	DNK	Haldor Topsoe, Technical University of Denmark, Aalborg University	2015	n/a	SOEC	Wind	Catalytic	Biomass gasification	[85]

Tab	le A1	. Cont.
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Project Name	Location	Country Code	Company/University	Start Date	Electrolyzer Power (kWel)	Electrolysis Type	Electricity Source	Methanation Type	CO <sub>2</sub> Source	Ref.
Thermal management and methanation performance of a microchannel-based Sabatier reactor-heat exchanger utilising renewable hydrogen	Potchefstroom	ZAF	North-West University	2020	8	PEM	PV	Catalytic (microchannel)	Bottled CO <sub>2</sub>	[35]
Towards the Methane Society	Midtjylland- Region	DNK	PlanEnergi, Haldor Topsoe, Aarhus University	2011	n/a	n/a	Wind	Catalytic	Biogas	[77]
UC Irvine power-to-gas (P2G) demonstration project	Irvine, Golden	USA	SoCalGas, UCI, NREL, NFCRC	2017	60	PEM	Wind, PV	Biological	Biogas	[94]
W2P2G	Wijster	NLD	Attero, Gasunie, Audi	2014	400	n/a	Waste treatment	Catalytic	Biogas	[82]

## Table A2. Research targets of the selected projects.

Project Name	Heat Management	Operating Condition Optimization	Materials	Carbon Capture	Design	Renewable Fuel Production	Integration within the Energy System	Technical and Economic Feasibility
Activity and stability of powder and monolith-coated NiGDC catalysts for CO <sub>2</sub> methanation			~					
Audi e-gas						~	~	
Biocatalytic methanation of hydrogen and carbon dioxide in a fixed bed bioreactor		~			~			
Biocatalytic methanation of hydrogen and carbon dioxide in an anaerobic three-phase system		~			~			
bioCONNECT							~	<b>v</b>
Bioelectrochemical systems for energy storage: A scaled-up power-to-gas approach					~			
BioPower2Gas				~		~		
BIT3G project				~		~		
Catalytic methanation of carbon dioxide captured from ambient air	~		~	~				
CCU P2C Salzbergen				~				
CO <sub>2</sub> Conversion to Methane Project	~			<b>v</b>	~			

Project Name	Heat Management	Operating Condition Optimization	Materials	Carbon Capture	Design	Renewable Fuel Production	Integration within the Energy System	Technical and Economic Feasibility
CO <sub>2</sub> methanation in a bench-scale bubbling fluidized bed reactor using Ni-based catalyst and its exothermic heat transfer analysis		V						
CO <sub>2</sub> recycling by reaction with renewably generated hydrogen		V		v				
CO <sub>2</sub> -Methanation of flue gas				<b>v</b>				
CO2RRECT				<b>v</b>				<b>v</b>
CO <sub>2</sub> -SNG pilot plant		v			~			
CoSin project	~	v						
COSYMA		v	~					
DemoSNG					~		<b>v</b>	
Dual function materials for CO <sub>2</sub> capture and conversion using renewable H2			~	v				
Economic assessment of a power-to-substitute-natural-gas process including high-temperature steam electrolysis			~					
EDGaR synthetic methane project			~		~		<b>v</b>	
EE-Methan aus CO <sub>2</sub>			~		~			
Einsatz der Biologischen Methanisierung für Power-to-Gas-Konzepte: Hochdruckmethanisierung von H2								v
El upgraded biogas	<ul> <li>✓</li> </ul>				~		~	<ul> <li>✓</li> </ul>
Energieversorgung Lübesse						<b>v</b>	~	
Experiment and numerical analysis of catalytic CO <sub>2</sub> methanation in bubbling fluidized bed reactor	~							
Experimental analysis of photovoltaic integration with a proton exchange membrane electrolysis system for power-to-gas							V	
Exytron demonstration project				~			~	
Forschungsanlage am Technikum des PFI							~	
HELMETH	~	<b>v</b>						
High performance biological methanation in a thermophilic anaerobic trickle bed reactor		V			~			
High-Performance Biogas Upgrading Using a Biotrickling Filter and Hydrogenotrophic Methanogens		~	~					

Project Name	Heat Management	Operating Condition Optimization	Materials	Carbon Capture	Design	Renewable Fuel Production	Integration within the Energy System	Technical and Economic Feasibility
Hybrid power plant Aarmatt—STORE&GO Switzerland							<b>v</b>	
Hybrid power plant Falkenhagen—STORE&GO Germany			~					
HyCAUNAIS Project								~
INFINITY 1							~	
Ingrid—STORE&GO Italy				~		~		
Integrated Co-Electrolysis and Syngas Methanation for the Direct Production of Synthetic Natural Gas from $CO_2$ and $H_2O$		V			~			
Intensification of catalytic CO <sub>2</sub> methanation mediated by in situ water removal through a high-temperature polymeric thin-film			v					
Jupiter 1000				~				
Klimafreundliches Wohnen				<b>v</b>				
Laboratory-scale experimental tests of power to gas–oxycombustion hybridization system design and preliminary results					V		V	
Laboratory-scale reactor in Fraunhofer IWES							~	
MeGa-stoRE				~				
Methanation at Eichhof							~	
Methanation of carbon dioxide by hydrogen reduction using the Sabatier process in microchannel reactors						~		
Methanation of recovered oxyfuel–CO <sub>2</sub> from Ketzin and of flue gas emitted by conventional power plants				V				
Methanation potential suitable catalyst and optimized process conditions for upgrading biogas to reach gas grid requirements		V			v			
MethFuel								<b>v</b>
MethyCentre								<b>v</b>
Minerve						~		
Modeling of Laboratory Steam Methane Reforming and CO <sub>2</sub> Methanation Reactors			v					
Morbach							~	
ORBIT								v
P2G movable modular plant operation on synthetic methane production from CO <sub>2</sub> and hydrogen from renewables sources	V	V			V			

Project Name	Heat Management	Operating Condition Optimization	Materials	Carbon Capture	Design	Renewable Fuel Production	Integration within the Energy System	Technical and Economic Feasibility
P2G Solar Energy Storage RD & D							~	~
P2G-BioCat					~			~
P2G-Foulum Project								<b>v</b>
PEGASUS Project						<b>v</b>		
Performance analysis of Sabatier reaction on direct hydrogen inlet rates based on solar-to-gas conversion system		V						
PID Eng&Tech		<b>v</b>	~					
Pilot plant—Tohoku Institute of Technology				~	~			
Powe to Flex	<b>v</b>							
Power to Gas 250		<b>v</b>						~
Power to Gas at Eucolino	<b>v</b>							
Power to Gas biogas booster						<b>v</b>		
Power to gas BTU Cottbus							~	
Power to gas Hungary								<b>v</b>
Power-to-Gas project in Rozenburg							<b>v</b>	<b>v</b>
Power-to-Methane HSR				~				
ProGeo				~	~			
Pure methane from CO <sub>2</sub> hydrogenation using a sorption-enhanced process with catalyst–zeolite bifunctional materials			v					
Reduction and reuse of CO <sub>2</sub> : renewable fuels for efficient electricity production				V			~	
RENERG2						<b>v</b>		
RENOVAGAS Project					~	<b>v</b>		
Small-Scale Demonstrator in Sion					~		~	<b>v</b>
Smart Grid Labor						~		
Storage of electric energy from renewable sources in the natural gas grid—water electrolysis and synthesis of gas components								v
Study of the role of chemical support and structured carrier on the $\mathrm{CO}_2$ methanation reaction			~					
Swisspower Hybridkraftwerk						V	<b>v</b>	

Project Name	Heat Management	Operating Condition Optimization	Materials	Carbon Capture	Design	Renewable Fuel Production	Integration within the Energy System	Technical and Economic Feasibility
SYMBIO		<b>v</b>						
SYNFUEL						<b>v</b>	~	
Thermal management and methanation performance of a microchannel-based Sabatier reactor-heat exchanger utilising renewable hydrogen	V	V						
Towards the Methane Society					~			
UC Irvine power-to-gas (P2G) demonstration project					~		V	
W2P2G						<b>v</b>		

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