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# Production of Biomethane by Hydrogenation of $CO_2$ and CO

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## ABSTRACT

This study analyzes the production of biomethane obtained by hydrogenation with green hydrogen of the $CO_2$ contained in the biogas produced from organic wastes (by anaerobic digestion), or from wood wastes by pyrogasification. Green hydrogen, utilized in this process, is produced by water electrolysis, capitalizing on surplus renewable energy currently available. This paper presents recent advancements in industrial production, pilot plants, and projects, predominantly executed in Italy, focusing on the hydrogenation of bio-$CO_2$ and bio-CO into biomethane. Both microbial and heterogeneous catalysis methods are explored for this transformation. The implications of these advancements on the bioenergy sector and the potential for further innovation are also discussed, providing an insightful perspective on the current state and future direction of biomethane production.

## 1. INTRODUCTION

This study will deal with the production of biomethane by hydrogenation (with green hydrogen) of the $CO_2$ by-product of biomethane production by anaerobic digestion of organic wastes [1-5], and by hydrogenation of biogas (CO, $CO_2$, $CH_4$ and $H_2$) produced by pyrogasification of wood biomass [6-8]. The hydrogen defined as "green" is obtained by electrolysis of water, using electricity from renewable sources (mostly wind and photovoltaic), it can be produced and/or stored in periods of excess energy, to be used in hydrogenation reactions.

It must be acknowledged that these technologies, while promising, are still in the development phase at the industrial level. In a context, such as the current one, in which we want to reduce the use of hydrocarbons and promote hydrogen as the energy vector of the future, it may seem strange that we suggest to transform $CO_2$ into methane, using hydrogen. There are at least three reasons to pursue this course of action: 1) For at least three or four decades it will not be possible to completely replace the use of methane, which we are forced to import in large quantities; 2) Methane is a relatively "clean" hydrocarbon; 3) It will take many years for a massive and concentrated production of green hydrogen (such as that needed in the future for industry and transport), and for the development of suitable infrastructures for its use.

The current production of green $H_2$ is limited and fragmented, and paradoxically, gives rise to surpluses, as there are no structures to channel it to a user who has not yet formed. It is therefore logical to use this surplus of $H_2$ and focus (in a transition period that seems to be long) in the production of biomethane and methane by hydrogenation of $CO_2$ and CO. Moreover, the reason why it is needed to introduce additional hydrogen to hydrogenate $CO_2$ in biogas plants (obtained in an anaerobic digester), it is due to the fact that the produced biogas has only about 55% $CH_4$ and 45% $CO_2$, in fact the fed biomasses do not produce enough hydrogen for $CO_2$ hydrogenation. Actually, the $CO_2$, co-produced with biomethane, is generally released into the atmosphere after the separation of the biomethane and only in few cases is it recovered for industrial use [1-5].

In the pyrogasifcation plants of wood wastes, the produced biogas contains minor quantities of $CH_4$, $CO_2$ and $H_2$ and more CO, therefore, it is necessary to add high amounts of hydrogen to transform CO and $CO_2$ to biomethane [6-8].

Given the predictable strong development of biogas and biomethane plants, the amount of co-produced $CO_2$ will increase significantly. Its use will help to reduce emissions and increase the contribution of biogas towards sustainable solutions. Only a few process modifications are required for its separation and purification (upgrading).

The different technologies proposed for the hydrogenation of $CO_2$ to methane and biomethane are mentioned in the book by Prof. Guido Saracco (Turin Polytechnic), dedicated to Green Chemistry [9]. Furthermore, also in some recent articles [10, 11] and communications [12, 13] the different technologies for optimizing the hydrogenation of $CO_2$ to biomethane have been analyzed.

## 2. PRODUCTION OF GREEN $H_2$

Green hydrogen is produced by water electrolysis using electricity derived from the current surplus of renewable energy [14, 15]. The electrolyte is usually an aqueous solution of sodium hydroxide (NaOH) or potassium hydroxide (KOH), and the electrodes are made of nickel-plated steel; the cells work at temperatures between 50°C and 80°C and produce high purity hydrogen (up to 15 barg) [16]. Two advanced techniques of $H_2$ production are 3b the PEM and the AFC.

In the electrolyzer based on polymer electrolyte membrane (PEM) [17], the electrolyte is a solid plastic material; water reacts at the anode to form oxygen and positively charged hydrogen ions (protons). In the alkaline fuel cell (AFC) technology an aqueous KOH/NaOH solution is used as liquid electrolyte, with a mass fraction of 35-85 wt.%; the process generally operates at less than 100°C, to avoid water losses [18].

The hydrogenation of $CO_2$ can be realized with green hydrogen, by heterogeneous catalysis or by microorganisms; photoreduction can be used as well, but to a much lesser extent. A recent article [19] has highlighted (on the basis of quantitative and qualitative considerations) the potential deriving from the valorization of the $CO_2$ contained in biogas in Sweden, to increase the production of methane.

## 3. THE HYDROGENATION OF $CO_2$ TO METHANE

### 3.1 The catalytic hydrogenation

The catalytic hydrogenation of $CO_2$ to $CH_4$ ($CO_2 + 4H_2 = CH_4 + 2H_2O$) is historically called the "Sabatier reaction", from the name of its discoverer, the French chemist Paul Sabatier. Together with Jean-Baptiste Senderens, he carried out the reaction in 1887, by using nickel-based catalysts and working between 300 and 400°C, at about 3 Mpa [20, 21]. It is interesting to remember that several years later (in 1912) Sabatier received the Nobel prize for chemistry with the following motivation: "For his method of hydrogenating organic compounds in the presence of finely disintegrated metals, whereby the progress of organic chemistry has been greatly advanced in recent years". Catalytic methanation currently takes place at temperatures between 200 and 550°C and at pressures between 20 and 100 bar. The most used catalyst is nickel supported on $\gamma$-$Al_2O_3$, which has high activity, good selectivity for methane and high availability (therefore lower costs); however, it requires a high purity of the incoming gases, to avoid its poisoning by $NH_3$, chlorine, sulfur compounds and aromatics [22].

As a result of the strong exothermicity of the reaction (165 kJ/mole), the second problem of methanation is the choice of the reactor, which must guarantee a correct temperature management to avoid damaging of the catalyst. Other suitable catalysts can be based on Ru, Pd, Fe and Co; however, they too are sensitive to poisoning by $CO_2$ impurities.

Catalytic methanation can be used downstream in plants that produce biogas by pyro-gasification of wood or solid organic wastes, but also downstream the anaerobic digestion plants of organic wastes, where (after the purification), it is necessary to introduce a hydrogenation reactor, to hydrogenate the $CO_2$ separated from the methane [23]. Other types of catalysts have been investigated [24].

### 3.2 The hydrogenation of $CO_2$ with microorganisms

To obtain biomethane by hydrogenation of $CO_2$, it is also possible to use bio-methanation processes using methanogenic bacteria (archaebacteria type), which transform $CO_2$ into methane in the presence of hydrogen; this technology was discovered in Delft in 1906. Hydrogen, introduced into the biodigester, allows $CO_2$ to be almost completely hydrogenated [25, 26]. The main advantages of bio-methanation compared to the catalytic process are the lower reaction temperature and pressure, without the need to purify the $CO_2$ [2]. Methanogenic bacteria operate in very low oxygen conditions, between 35°C and 55°C, at atmospheric pressure, but with a $H_2/CO_2$ ratio of 7/1, almost double the theoretical need (Figure 1). The problem of the hydrogenation with bacteria is the low solubility of hydrogen in water, which is 40 times less than $CO_2$; therefore, it is necessary to increase the solubilization of hydrogen [1-5].

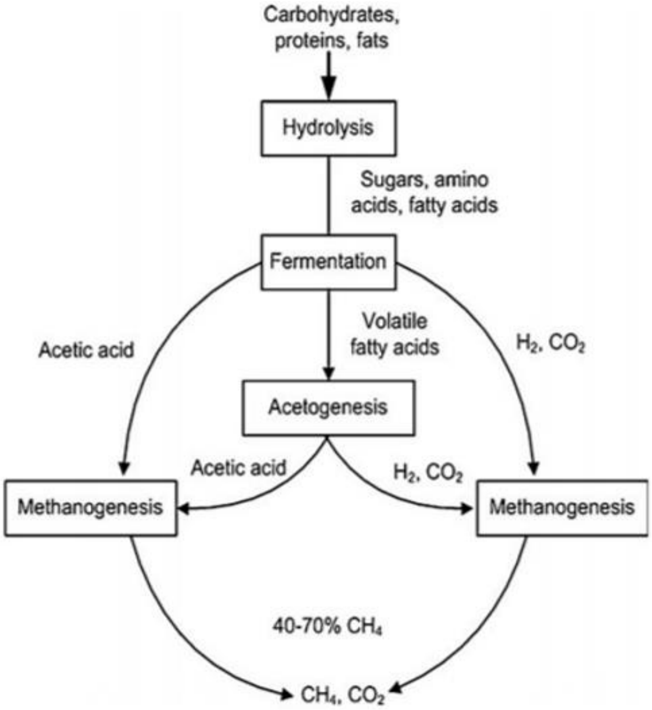

**Figure 1.** Process flow of the degradation of organic material through anaerobic digestion

The methanation of the biogas can be carried out inside the digester (in-situ), by injecting hydrogen and carefully controlling the operating parameters, or by feeding the biogas with green hydrogen in a second external reactor (ex-situ) in which methanogenic microorganisms are yet present. In this second hydrogenation reactor (after the anaerobic digester) the $CO_2$, obtained together with biomethane, can be hydrogenated to methane (see Figure 2).

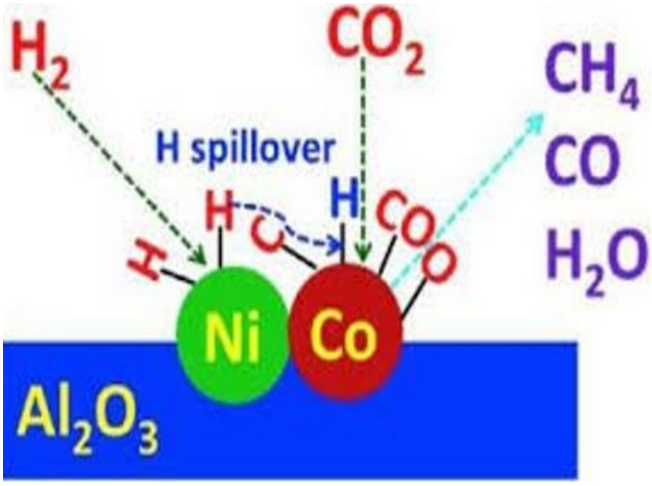

**Figure 2.** Hydrogenation of $CO_2$ coproduced with biomethane

## 4. PROJECTS OF $CO_2$ HYDROGENATION TO BIOMETHANE IN UNITS FOR ANAEROBIC DIGESTION OF ORGANIC WASTES

In Indre et Loire (France) a plant for the production of biomethane was started applying the Methycentre technology; the $CO_2$ from the anaerobic digester is hydrogenated catalytically with green hydrogen, and it is separated from the methane in the purification plant [27].

In Duchy, near Saint-Florentin (France), a plant with Hycaunais technology has been built, where the $CO_2$ co-produced with methane [28] in the anaerobic digestion unit of organic wastes, is hydrogenated (after separation from methane) to biomethane with green hydrogen, in a reactor containing selected microorganisms [29].

A recent article [30] reports that Fraunhofer researchers of the Institute for Microengineering and Microsystems (Mainz, Germany) have found an efficient way to produce biomethane by hydrogenating $CO_2$ contained in the biogas. Inside the hydrogenation reactor, a structure of coated microchannels was created with the catalyst, in which to flow $H_2$ and $CO_2$; with this reactor the surface area is increased and, promoting the contact between the gases and the catalyst; therefore, also the yield in biomethane is increased.

The "Acea Pinerolese Industriale" has two biomethane plants in Pinerolo (Turin, Italy), the biomethane is produced from the organic fraction of municipal solid wastes (OFMSW). In this industrial site, a project called "ProGeo" started in 2016, to build an experimental hydrogenation plant of $CO_2$ to biomethane, with green hydrogen; the $CO_2$, produced by the anaerobic digestion of organic wastes, has been separated from the biomethane in the upgrading stage, by absorption in water, after cooling and compression of the biogas [31, 32]. The realization of the pilot plant of ProGeo project was preceded by the Prometeo project, which had the objective of producing green $H_2$ by electrolysis of water, by using the excess energy produced by renewable energies (wind and photovoltaic). In the ProGeo project the catalytic hydrogenation of $CO_2$ takes place in a single stage (intercooled with recirculation) with a subsequent purification system, to increase the purity of the methane and to recirculate the unconverted $CO_2$ in the hydrogenation reactor. This hydrogenation technology allows a reduction in plant costs and an increase in $H_2$ conversion.

Subsequently, the same company has been involved in a second project, called Spotlight [33] to realize the production of biomethane using $CO_2$, green hydrogen and a sunlight photonic device, to carry out the hydrogenation. The aim of the project is the development of a photonic device that uses sunlight to convert $CO_2$ (produced by ACEA in the anaerobic digester) to biomethane, with green hydrogen; a syngas containing CO (that can be used to produce methanol) is also produced by reverse water gas shift.

Another company, the Italian ENEA, has studied the possibility of increasing the amount of biomethane produced in an anaerobic digestion plant of organic wastes, by introducing green $H_2$ into the biodigester, to increase the hydrogenation of $CO_2$; the process is based on the use of methanogenic hydrogen-genophilic bacteria, which are already used for the production of biomethane [34] in a project called + GA. With this technology not only does the amount of $CH_4$ increase, but the emission of $CO_2$ into the atmosphere is avoided, and $CO_2$ separation costs from methane are reduced in the process of upgrading the biogas. To carry out these further hydrogenations of $CO_2$ in the anaerobic digester, ENEA developed new technological solutions able to facilitate the solubilization of the added hydrogen and to increase its degree of assimilation by selected microorganisms; practically, a hydrodynamic cavitator is used which improves the dissolution of the gases in the liquid phase. In theory, the $CO_2$ that is not hydrogenated by the outgoing microorganisms from upgrading, could be used in a downstream plant for chemical methanation, producing more biomethane. The +GAS project started on 1 September 2016 and it has been completed on 31 August 2018 coordinated by ENEA which, with this project, won the prize of the Emilia-Romagna Region for innovative and strategic industrial research solutions in the field of energy. In the future, pilot and demonstration plants will have to be built to verify the industrial feasibility of the process.

On 6 September 2022 news arrived [35] that in Bologna, in 2023, the Hera company with the “Pietro Fiorentini” group have to build a biomethane production plant by hydrogenation with microorganisms (technology of the German subsidiary MicroPyros) of the $CO_2$ produced by the anaerobic digestion plant for sewage sludge: that is a large plant present since years in Corticella (Bologna). In Corticella, $H_2$ will be produced by electrolysis of purified waste-water, by using electricity produced in excess from renewable sources; the biomethane produced will be used by 1200 families in the area; the plant will be called SynBioS (Syngas Biological Storage) with an investment of 10 million euros.

## 5. PYROGASIFICATION TECHNOLOGIES TO PRODUCE BIOGAS AND ITS TRANSFORMATION INTO BIOMETHANE

The pyrogasification process can be divided into four phases: drying of the charge, pyrolysis, oxidation/reduction (gasification) and partial purification [6-8] (Figure 3). Pyrolysis is the biomass treatment, between 650-750°C in the absence of oxygen, with short contact times; the gas produced contains $CO_2$, CO and $H_2$ (approx. 85%), a liquid (approx. 5%) and a solid (approx. 5-10%). The partial purification of the syngas consists in the elimination of some by-products, such as dust (vegetable charcoal or biochar), ashes and tars. The biogas obtained is sent to an internal combustion engine (more rarely a turbine) to produce energy.

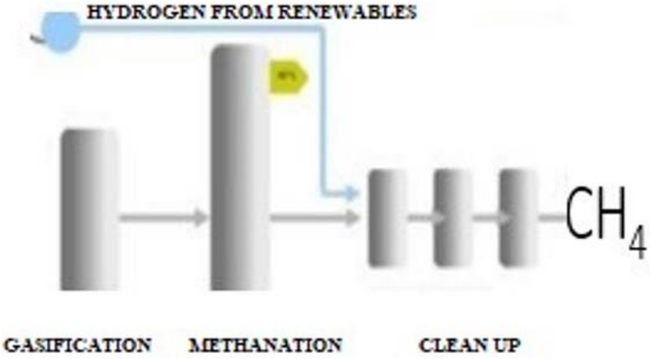

**Figure 3.** Production of biomethane by pyrogasification of wood wastes

Among the co-products of syngas there are traces of $CH_4$ and of other hydrocarbons (both paraffinic and aromatic) depending on the technology and raw material used; other compounds, such as $NH_3$, $H_2SO_4$, HCl, organic substances and various impurities must be eliminated before hydrogenation to biomethane. The production of syngas does not release any emission into the atmosphere and does not produce wastes as dangerous liquids. The syngas obtained is used today in various plants in Italy and in Europe, for the production of electricity and heat. The problem is that the production of biomethane is limited by the high costs of hpurification of the biogas. The sustained elevation in methane prices may potentially enhance its appeal in the market.

### 5.1 Examples of pyrogasifcation plants

In Gothenburg (Sweden), it has been realized the first demonstration plant for the production of syngas by pyrogasification of lignocellulosic biomass and subsequent hydrogenation of the obtained biogas [36]. The plant, called "Gobigas", went into operation on December 2014, with the production of 20 mW/h of biomethane (2200 $Nm^3/h$) [37].

A pilot plant for the production of biomethane by pyrogasification of lignocellulosic wastes and solid agri-food wastes was built in January 2018 in Saint-Fons (near Lyon, France) by the Engie company, with the “Gaya” technology [38]. Suitable wastes were collected within a radius of 50-70 km from Saint-Fons. The plant produces methane (55-75%) and $CO_2$ (20-45%), the separation of $CO_2$ is done with membranes, after the synthesis of biomethane.

In 2018, ENEA has built an experimental biomethane production plant at Casaccia (Rome) for the pyrogasifcation of wood wastes and subsequent hydrogenation of the biogas [39, 40]. In this pilot plant the pyrogasification is accompanied by a hydrogen production plant, obtained by electrolysis of water; $H_2$ is introduced into the hydrogenation reactors, already existing downstream the pyrogasifier, increasing the quantity of methane, produced by hydrogenation of CO and $CO_2$ not converted because of insufficient hydrogen in the biogas plant.

## 6. CONCLUSIONS

The interest in increasing the production of biomethane by hydrogenating the $CO_2$ (co-produced in biomethane plants) is not only due to find alternatives to the methane that arrives with gas pipelines. In the future it is important to carry out these processes for the following reasons: 1) to use organic and wood waste, producing biogas, in order not to waste natural raw materials; 2) to decrease $CO_2$ emissions, whose production will increase together with that of biogas; 3) to use any surplus of renewable energy to produce hydrogen that can be used to hydrogenate $CO_2$.

Furthermore, the current increase in the price of methane of fossil origin makes the production of biomethane with the technologies described in this note competitive. In the case of production of methane, the interest is to decrease the consumption of fossil fuels. The use of biogas creates emissions of $CO_2$: those produced by the combustion of biomethane and those contained in the biogas itself; however, it must be taken into account that they do not come from fossil fuels and therefore do not go against the European restrictions. The production of biogas brings limited energy contribution to the area production, but still has the great advantage of reducing the amount of materials sent to dump. The purification of the biomethane contained in the biogas opens up a much wider energy horizon, which allows $CH_4$ to be introduced into the national network, reducing dependence on imports from abroad. However, a decisive contribution to our methane needs can only come from the development of larger plants equipped for separation and disposal of $CO_2$. In the world, 90% of biomethane is obtained by upgrading the biogas obtained from the anaerobic digestion of organic wastes (agriculture, animal, urban organics, and by-products of agricultural industries) while 10% is obtained by pyrogasification of woody biomass, and subsequent hydrogenation, regarding the biomethane obtained by hydrogenation of the biogas produced by pyrogasification of wood wastes or solid organic wastes, currently the biogas is essentially used to produce electricity and heat. The big problem in the production of biomethane is the high costs for the purification of the obtained biogas.

On 18 July 2022 it was announced that the Italian company NextChem (active in the green chemistry and energy transition sector and controlled by the Italian group Marie Tecnimont) will build a plant at the port of Le Havre (France) by 2025, this will be the world biggest production of biomethane by pyrogasification of woody biomass, with an output of 15.4 million $m^3$ of biomethane (40).

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