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Design of a Natural Gas Steam Reforming Process for Hydrogen Production Powered by a Solar Furnace

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Abstract. The work involved in this paper has been developed as part of project NEST “Network for Energy Sustainable Technologies" framed in the PE program "Green Energies for the Future". It aims to develop technologies for the conversion and use of renewable sources that should be sustainable, both from an environmental and a social point of view. In particular, the paper shows the design of a process that employs the high temperature heat produced by a solar furnace, realized at Research Centre of ENEA in Portici, to supply a system for the hydrogen production based on the steam reforming natural gas process. In the direction of the decarbonization of the chemical industry, the solar concentration-based plant appears well compatible with the endothermic stage of a reforming based plant, in which the selected hydrocarbon reacts with steam to produce a hydrogen-rich syngas (to be refined in the further purification stages). [1]

Keywords: Solar Furnace, Green Hydrogen, Biogas Reforming, Solar Fuels

1. Introduction

With the Green New Deal, Europe has put itself forward to become the first climate-neutral continent in the world in 2050. The transition to a climate-neutral society will involve various sectors of society and the economy: energy, industry, civil, finance. It is now widely believed that one of the priorities is the progressive replacement of fossil fuels with fuels with a reduced carbon footprint. However, the replacement of fossil fuels derived from petrochemicals with biofuels, as in the case of natural gas with biomethane, may not be enough; and it will be necessary to introduce “renewable” hydrogen, for its direct use or to promote the production of “renewable synthetic fuels”. Given the leading role recognized to hydrogen in the decarbonization of the economy, it therefore becomes essential to explore all the technologies available for its “green” production, carrying out research and development activities on the most promising ones, pursuing objectives of reducing costs and increasing efficiency. To date, approximately 95 million tons (Mton) of hydrogen are produced worldwide [2], of which more than half a million tons in Italy alone; the trend in demand for hydrogen has been continuously growing for several decades by the chemical and refining industrial sector. In fact, most of the hydrogen (about 70 Mton) is used in the form of pure gas (for example for the synthesis of ammonia and for petrochemical processes) while a smaller part is used in a mixture with other gases (for example with CO, in synthesis gas or "syngas") for the synthesis of methanol or in steel plants for the reduction of iron-containing minerals. These 95 Mton of H_2 needed by industry today come partly as a co-product from other processes often carried out near the same plants where it is used, as a co-product of the cracking, reforming and gasification processes (of heavy hydrocarbon residues) in refineries, dehydrogenation plants (for example in olefin production units) and chlor-alkali plants. The hydrogen available as a co-product as a whole, however, is not enough to cover demand. Therefore, an additional quantity of hydrogen will have to be specifically produced. The most widespread process today is the steam reforming of natural gas, which covers about half of the hydrogen demand (>50 Mton/year of hydrogen are currently produced worldwide with this process) [3], while up to 92% of the worldwide hydrogen demand is satisfied through reforming processes of hydrocarbons [4]. Reforming based processes are based on the Steam Reforming (SR) reaction of fuels, in which hydrocarbons react with steam to produce hydrogen and carbon monoxide (see Equation 1).

$$CH_4 + H_2O \rightleftarrows 3H_2 + CO\Delta \qquad H_{SR}^0 = 206\ kJ \tag{1}$$

According to thermodynamics. the high reaction endothermicity impose to operate the process at high temperature, to assure adequate conversion values of the fuel.

Typically, the SR stage is followed by a further purification stage, the Water-Gas Shift (WGS) in which carbon dioxide produced in the former process is converted to carbon dioxide by producing further hydrogen (see Equation 2).

$$CO + H_2O \rightleftarrows H_2 + CO_2\Delta \qquad H_{WGS}^0 = -41\ kJ \tag{2}$$

Both processes (SR and WGS) should be carried out in catalytic reactors, in order to assure high reaction rates and process selectivity.

Since the endothermicity of the reforming reaction, the SR process should be thermally sustained by external heating duty: in the common steam reformer, further fuel is burned to produce the required heat to the process. It was estimated that for the conversion of 1 ton of methane in hydrogen (via SR reaction), around 0.35 ton of methane should be burned to sustain the required reaction endothermicity [5, 6], thus resulting in an overall efficiency reduction, as well as contributing to the environmental impact of the process.

For the decarbonisation of industrial sectors that require hydrogen, it will therefore be necessary to adopt other hydrogen production processes, which reduce the use of fossil fuels, and which do not cause greenhouse gas emissions. This need becomes even more relevant if we consider that, in perspective, the demand for hydrogen will continue to grow considerably driven by the “Hard-to-Abate” sectors. It will therefore be necessary to replace the hydrogen currently produced by traditional reforming processes with hydrogen that is possibly “renewable”, i.e. produced entirely from renewable energy sources. There are multiple processes to produce renewable hydrogen. The renewable energy source can be provided in the form of electricity (photovoltaic, wind, hydroelectric, geothermal, marine currents, solar thermodynamic, etc.) and/or heat (solar thermal, biomass combustion).

Among all the possible alternatives, this work aims to study the possibility of producing syngas through the steam reforming reaction using highly concentrated solar energy [7,8,9], and in particular the solar furnace already available at the ENEA Research Center in Portici (NA). In fact, the so-called "solar reforming" is considered a promising process to convert solar energy into high value-added products, such as syngas or hydrogen, to be used subsequently for the production of electricity, for example. For this purpose, the solar energy concentration system is essential, which, in this specific case, is made up of a heliostat, a primary concentrator and a receiver secondary concentrator. This system is able to produce a hot fluid (air, about 0.1 kg/s) at a temperature of about 600-850 °C that can be used to feed the hydrogen production process. In this paper a detailed description of the solar furnace and its working cycles are first detailed, then the preliminary design of the steam reforming system is described.

2. Plant description

In this paragraph a description of the main components of the plant is reported.

2.1 Solar furnace

The solar furnace (see Figure 1) consists of a 120sqm flat heliostat (see #1 in Figure 1) that reflects the light coming from the sun always in the direction parallel to the ground and with a North-South orientation, on a primary concentrator consisting of an array of hexagonal mirrors having a parabolic profile (see #2 in Figure 1).

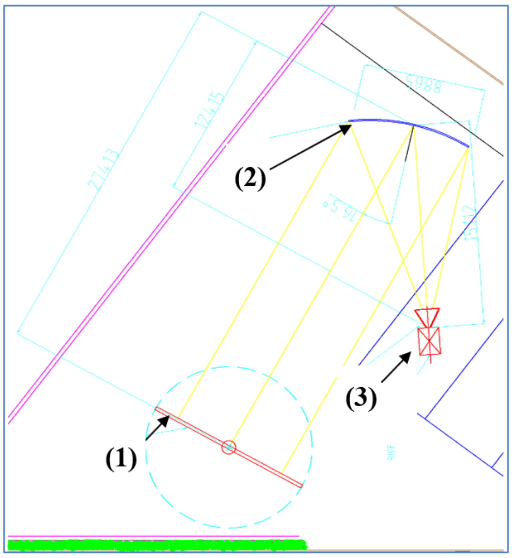

Figure 1. *Layout of the ENEA solar Furnace*

The light is concentrated in a receiver (see #3 in *Figure 1*), where thermal powers of the order of 30kW and concentrations of about 2000 suns are achieved [10,11].

This system is an off-axis optical concentrator since the segment joining the centres of the primary concentrator and the furnace is inclined by a solid angle of about 21° with respect to the direction of the light reflected by the heliostat. In the *Figure 2* the pictures of these main components are reported.

Figure 2. *From left to the right: view of flat heliostat, primary concentrator and receiver input hole*

The receiver consists of a double-walled cavity where the light enters from a front hole and impacts the internal walls, heating them. These latter are cooled by a flow of air that circulates in a circuit created in a cavity between the internal and external walls. Under nominal working conditions, with direct solar radiation of 1000W/sqm, the heat removed by the receiver is capable of heating a flow of 0.1 kg/sec of air from 620°C to 850°C.

2.2 Thermal System

The heat produced by the furnace is used by a thermal system built in correspondence of the primary concentrator at ground level. It includes the following main components: air compressor, heat recovery unit, thermal storage system, auxiliary heating and user.

Figure 3. *7,5kW air compressor and its control box mounted in the plant*

The compressor (see *Figure 3*) is of the rotary type and equipped with inverter, has an electrical maximum absorption of 7.5kW and pushes the external air into the thermal circuit. It has only to provide the prevalence needed to overcome the circuit pressure losses: its design working point foresees a 0.1kg/sec flow rate with an operating pressure of 300mbar.

The heat recovery unit (*Figure 4*) is a compact air-to-air plate exchanger, necessary to preheat the external air processed by the compressor by exchanging heat with the flow of hot air exiting the user and which is then released into the atmosphere.

Its nominal operating conditions are:

- Hot side: Tin: 700 °C - Tout: 120 °C – P: 1.1 bar– flow rate: 0.1 kg/s.
- Cold side: Tin: 70 °C - Tout: 620 °C – P: 1.5 bar (g) flow rate: 0.1 kg/s.
- Pressure drop @0.1kg/s: 40mbar on each branch.

In order to minimize thermal losses, an external coating was created consisting of 4 layers of specific insulating material for high temperatures (Microtherm Slatted 20 mm thick) covered by a steel sheet shell.

The heat storage tank (*Figure 5*) is a component realized in AISI 316 steel having the shape of a vertical tube made up of three elements: two plenums, one lower and one upper, connected to the circuit pipes and a central body. The accumulation is made up of alumina spheres in the central part with a diameter between 12mm and 14mm with a minimum content of alumina oxide equal to 99.0%. The total mass of alumina is approximately of 1900kg, which allows for the accumulation about of 120kWh of thermal energy. The thermal insulation is of the same type as that used for the heat recovery unit and has been created both internally and externally to the metal shell. In the central body, 12 thermocouples with a long rod to measure the temperature at the centre of the tank and 12 with a shorter rod to measure the temperature at half the radius of the cylinder have been installed along the entire height of the cylinder. Several thermodynamic simulations carried out by means Comsol Multiphysics software (version 5.6) showed that the efficiency of the storage, measured as the ratio between extracted and injected energy system was about of 85% [12].

Figure 4. *Heat recovery unit installed in thermal plant*

Figure 5. *View of the inside of the tank (left) during assembly operations with the two rows of thermocouples visible and from the outside (right)*

The thermal circuit is also equipped with a backup heating system (*Figure 6*) for the heat transfer fluid. It serves to stabilize the inlet temperature of the heat transfer fluid to the solar receiver to avoid thermal shocks and to allow the target process temperature of 850°C to be reached in the various operating conditions at the receiver outlet. This system consists of three 15 kW electric heaters assembled using an equipment specially designed to separate the inlet flow to power the individual heaters and remixing it at the outlet.

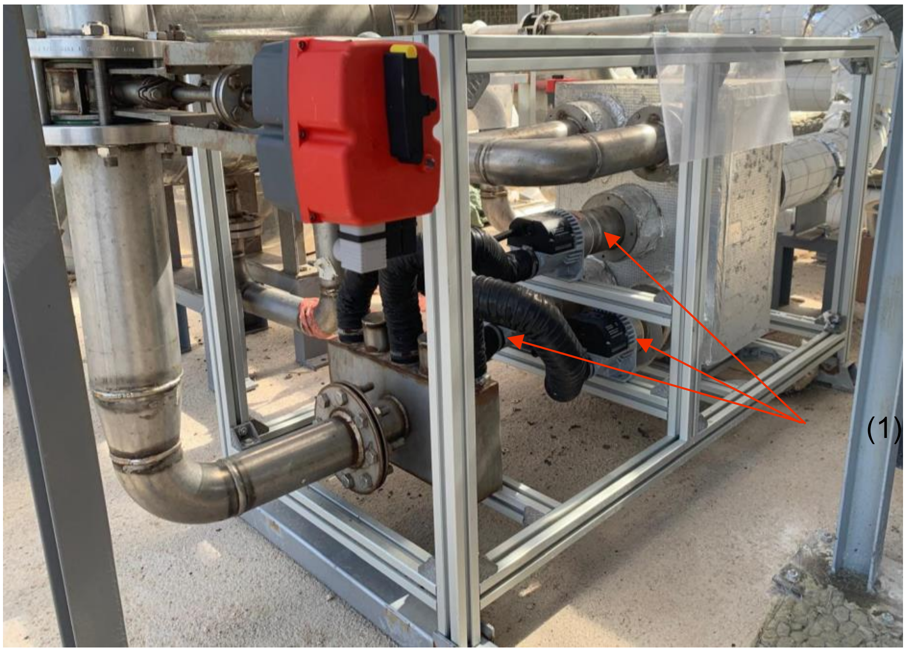

Figure 6*. View of the equipment with the auxiliary heating systems assembled on it (1)*

The user system is an air/water exchanger that serves to simulate the use of heat produced by the furnace or by the auxiliary heating system in a possible user. On one side it is connected to the high temperature air circuit by means of DN80 flanges, while on the other one is connected to a hydraulic circuit that will allow the heat to be disposed of and the air temperature to be lowered from 850°C to 700°C. This exchanger has a series of hydraulic circuits that can be activated in order to increase or decrease the air outlet temperature and therefore simulate loads of different sizes.

The nominal operating point is as follows:

- Hot side (air): Tin: 850 °C - Tout: 700 °C – P: 0.2 bar(g) – flow rate: 0.1 kg/s.
- Cold side (water): Tin: 20 °C - Tout: 30 °C – P: 2.5 bar(g) flow rate: 0.5 kg/s.

The thermal system has been designed to allow different operating configurations described below, thanks to several automatic valves placed along the circuit which allow to choose the air flow path.

2.2.1 Direct Supply Configuration

In the direct feed configuration (*Figure 7*), the heat produced in the receiver at 850°C (1) is used in a dissipation system that simulates its use and cools the flow up to 620°C (2). Subsequently, the flow exiting the user releases its heat, preheating the incoming flow via an exchanger-recuperator (3). This incoming flow has previously been processed through the compressor (4).

2.2.2 Heat storage loading

When the furnace (*Figure 8*) is working, but there is no need to use the produced heat, the thermal accumulation can be loaded. In this configuration, the passage in the user (3) is replaced by that in the thermal accumulation tank (5). All the rest of the cycle is exactly the same as in the previous case.

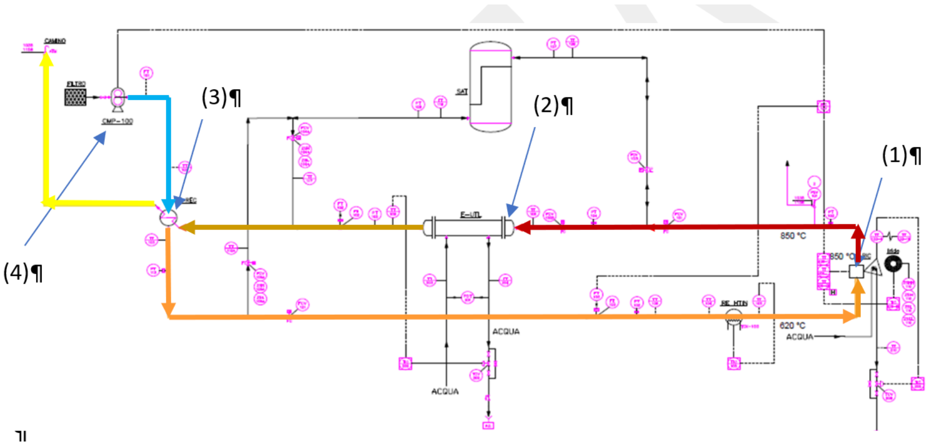

Figure 7. *Operating diagram of the system in the direct supply configuration*

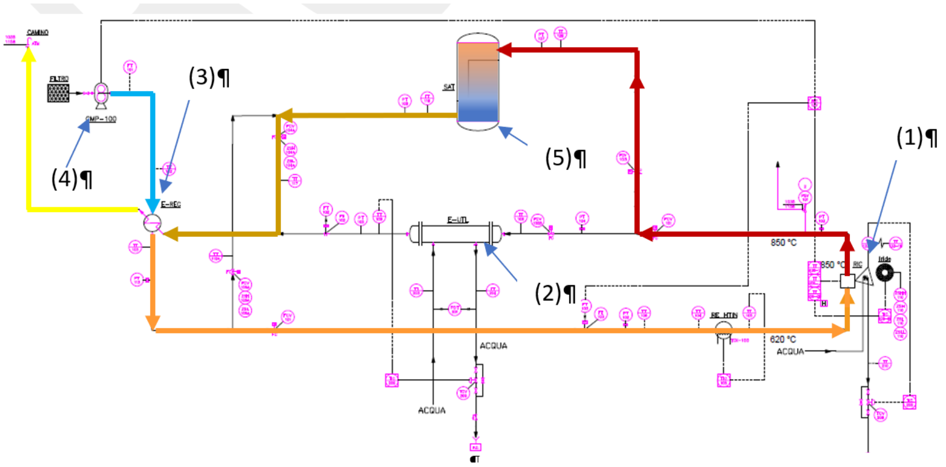

Figure 8. Operating diagram of the system in heat storage charging configuration

2.2.3 Heat storage unloading

When there is no solar radiation and you want to use the thermal energy accumulated in the storage system, the valves on the pipe allow to change the path as shown in the following figure, heating the flow in the storage (5) and then cooling it in the user. In this layout, the receiver is obviously excluded from operation.

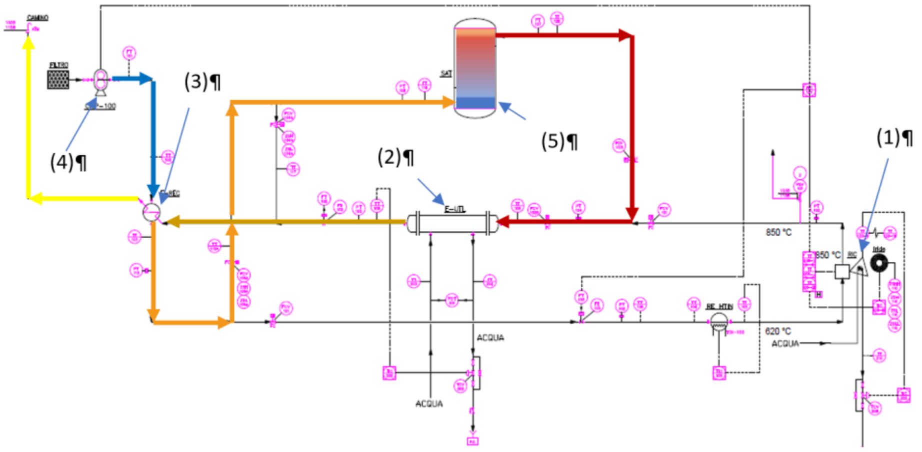

Figure 9. Operating diagram of the system in heat storage discharging configuration

3. Design of the reformer unit

The availability of a hot fluid stream, characterized by a temperature between 700°C and 850°C, able to supply a net power of about 20kW, well suits with a reaction stage in which syngas could be produced by steam reforming of hydrocarbons. From a theoretical point of view, natural gas steam reforming could be carried out at temperature within 650-800°C, with an appreciable fuel conversion [4]. Preliminary energetic evaluations assessed that the power generated by the solar furnace could assure a potential hydrogen production (by the reforming stage) up to 24 Nm^3/h by processing 7.15 Nm^3/h of natural gas, or 18.5 Nm^3/h of hydrogen by processing 10.36 Nm^3/h of biogas, by feeding both hydrocarbon and water at room conditions. In order to strike the goal of a sustainable process, the reaction system concept focus on a totally self-powered process, since both reactants pre-heating and steam reforming reaction will totally thermally be sustained by the energy produced by the furnace. Obviously, the reforming system will be able to work in the direct supply and in the heat storage unloading configurations (see paragraph 2.2.1 and 2.2.3) by replacing the user with the reformer heat exchanger. A key issue of the process is the maximization of the heat transfer rate from the heating medium to the reaction volume: in fact, the high endothermicity of reforming reaction results in the highest heat duty in the reaction section. As a principle, the increasing of heat flux toward the reforming area could be achieved by an increased heat exchange area as well as by reducing the heat transfer resistances: both concepts will be considered in the reaction system design. In fact, efforts will be spent in the reaction volume geometry optimization, in order to strike the requirement of a high surface-to-volume ratio, responsible for the heat transfer promotion [13], without penalizing pressure drops and fluid dynamics in the reaction volume. On the other hand, innovative structured catalytic systems will be adopted, characterized by high thermal conductivity and able to maximize the solid-gas mass transfer: such approach both promotes the heat transfer to the reaction gas, and emphasizes the reaction mechanisms on the catalytic surface, thus resulting in an increasing in the catalytic reaction rate, that in turn allows a reduction in the rection volume [14]. In addition, to assure a good approach to thermodynamic equilibrium, particular attention should be paid to the catalytic formulation: the appropriate combination of a chemical support, chemical active phase and promoters/stabilizers are able to reduce the reaction temperature threshold, and contribute to achieve a catalytic system able to effectively operate with different fuel typologies and feed ratios, and to exhibit acceptable performances in terms of stability and coke deposition resistance [15].

A not negligible aspect of the reaction section is the thermal management of the flux [16]: basically, one of the project requisites is to fed reactants at room temperature, while produced syngas is available at temperature close to 750-800°C. Therefore, sensible heat could be recovered and exploited to pre-heat reactants (aside the heating through the furnace hot stream). Therefore, the thermal integration of involved process streams also plays a crucial role in the reaction section design. The basic scheme of the plant is shown in the Figure 10, from which it can be seen how the system is divided into three sections:

1. **Feeding section:** is constituted by a battery of Mass Flow Controllers (MFC) aimed to control the reactants supply to the reaction system. Aside the reforming reactants (methane, carbon dioxide for the biogas simulation, liquid water), hydrogen, oxygen and nitrogen could be also delivered to the catalytic volume for several catalyst treatment processes (programmed temperature reduction, TPR, and programmed temperature oxidation, TPO) for the activation and/or regeneration procedures. The gaseous reactants are available in high pressure (up to 250 bar) cylinders and delivered to the MFCs after a pressure reducer and a shut-off valve, while demi-water was stocked in a tank and pressurized through a rotative pump.

2. **Reaction section**, comprising the catalytic reactor and the heat exchange system for the reactants pre-heating.

The reactor is stainless steel made, consisting of tubes with an internal diameter of 3 cm placed in a cylindrical shell. The reaction takes place tube side, while in the shell some septa force the heating gas in transversal trajectories, in order to enhance the heat exchange. The reactor is completed by highly conductive structured catalysts, designed and realized by ProCEED labs at University of Salerno, and inserted in the inner tubes, assuring a deep contact between catalysts and tube walls in order to maximise the heat flux toward the reaction volume.

Close to the reactor, the heat exchanger system is placed, in which sensible heat is transferred from hot fluid from solar furnace and from the exhaust gas to the reactants. The heating system is arranged in order to maximize the heat recovery from the exhaust products, and to maximize the temperature in the catalytic volume, in order to promote the hydrocarbons conversion according to thermodynamic constrains.

Other devices are placed in the reaction section, to assure an accurate monitoring of the process, in particular:

- probes for measuring temperature;
- probes for measuring pressure.

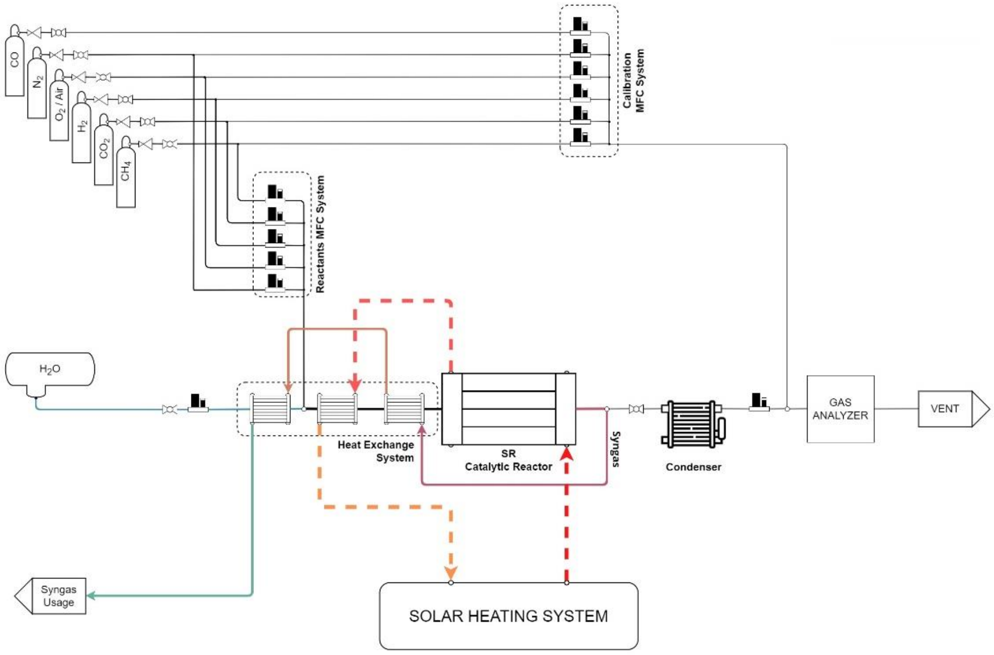

Figure 10. *Schematic diagram of the methane steam reforming plant*

3. **Analysis section,** in which the composition of the achieved syngas was evaluated. It is constituted by a dedicated multigas analyser, able to detect the volumetric fraction of hydrogen, methane, carbon monoxide, carbon dioxide and oxygen. A sampling line from the exhaust gas from the reaction section is delivered to the analysis section, after a drying stage carried out in a cryogenic bath device. The sampling flow rate was controlled by dedicated MFC, placed downstream the drying unit. A dedicated MFCs block was connected to the pure gas cylinders, in order to create gas mixture to deliver to the gas analyser for the calibration procedures.

The syngas coming out of the reaction stage well fits requirements for a Molten Carbonate Fuel Cell (MCFC) or Solid Oxide Fuel Cell (SOFC) for the electrical power generation.

4. Conclusions

In this work, the design of a system for the production of hydrogen by reforming process of biogas or natural gas, powered entirely by the heat supplied by the solar furnace realized and operating at ENEA research Centre in Portici, has been described. The solar furnace, consisting of a dual-stage solar concentration system and a cavity receiver, allows to heat a 0.1 kg/s air flow from 620°C to 850°C. This energy is currently used by a thermal plant for testing and characterizing a high-temperature thermal storage system made of alumina spheres. The nominal capacity of the storage is 120 kWh: this energy currently allows the furnace to operate for about 4 hours in the absence of solar radiation. The thermal circuit is very flexible thanks to the presence of servo-controlled valves and allows different operating configurations: 1) the use of the heat produced by the receiver on an external load; 2) the loading of the thermal storage; 3) the use of the heat accumulated in the storage to power the load. The system is completed by an auxiliary heating system consisting of 3 15kW electric heaters that can be used to stabilize the temperature of the heat flow on days with variable solar irradiance, or to work in the complete absence of radiation for the execution of urgent tests. All the piping is made of AISI310 stainless steel insulated with a special microporous material that allows obtaining a very low thermal conductance (0.025W/sqmK) even at the high working temperatures of the system.

The reforming system for hydrogen production has been designed based on the characteristics of the solar furnace. Using the available thermal power, a thermochemical cycle has been sized for the production of 24 Nm^3/h of hydrogen by processing 7.15 Nm^3/h of natural gas, or 18.5 Nm^3/h of hydrogen by processing 10.36 Nm^3/h of biogas. The core of the system will be the reactor that must be built in order to facilitate the heat exchange between reagents and the hot flow coming from the furnace. It is planned to build a crossflow tube and shell exchanger, in which the reagents are circulating inside the tubes that are heated from the outside. Special catalysts, inserted in the tubes, will allow to lower the threshold temperature of the reaction. The crucial point is to choose a geometry of this component with a high surface-to-volume ratio and that allows to optimize the heat exchange process. The Syngas produced will be passed inside further heat exchangers in order to preheat the reagents before entering the reactor. Similarly, the air hot flow, before re-entering the furnace circuit, will be used to preheat such reagents. A specific section of the plant is devoted to the chemical characterization of the produced syngas by using a multigas analyser, able to detect the volumetric fraction of hydrogen, methane, carbon monoxide, carbon dioxide and oxygen. All the reagents will be supplied via gas-cylinders connected to mass flow controllers whose purpose will be to supply the correct quantities to simulate different types of incoming gas (e.g. natural gas, bio-methane or bio-gas). The syngas coming out of the reaction stage well fits requirements for a Molten Carbonate Fuel Cell (MCFC) or Solid Oxide Fuel Cell (SOFC) for the electrical power generation.

The pilot system here presented will allow testing an innovative approach for the production of hydrogen by reforming hydrocarbons using entirely energy coming from renewable sources. As already mentioned, most of the hydrogen in use today comes from this process and therefore the approach followed in this study could play an important role in the decarbonization of this industrial sector.

Furthermore, this application could give new impetus to high concentration CSP systems that are essential due to the very high temperatures involved that cannot be reached by other renewable plants.

In the coming months, a detailed design of the components and in particular of the reactor will be carried out, in order to be able to have the prototype built and integrated into the solar furnace in a short time.

Data availability statement

The data on which this publication is based are reported in the references below.

Author contributions

Conceptualization: C. Cancro, M. Atrigna, A. Ricca, L. Mongibello; Data curation: C. Cancro, A. Ricca, L. Mongibello; Formal analysis: A. Borriello, V. Palladino; Investigation: C. Cancro, M. Atrigna, G. Ciniglio, A. Borriello; Methology: C. Cancro, A. Ricca; Software: L. Mongibello, M. Atrigna; Resource: G. Ciniglio, A. Borriello; Supervision: C. Cancro; Validation: C. Cancro, A. Ricca; Writing – original draft: C. Cancro, A. Ricca, M. Atrigna; Writing – review & editing: V. Palladino, G. Ciniglio

Competing interests

The authors declare that they have no competing interests.

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