Implementing of a Usable Tool for Selecting Operations to Upgrade Biogas to Biomethane

L. García Gómez, S. Luque, A. M Gutiérrez, and J. R, Arraibi

Abstract—In Spain, biomethane and biogas are still starting to be considered as an alternative to natural gas. A good way of promoting these renewable energies is supporting small and cheap treatment plants near to the place where the biogas is produced and where the biomethane can be used on site, fostering the circular economy. An easily usable simulation tool for selecting the best sequence of unit operations for treating biogas (based on adsorption, absorption, and membranes) has been designed. Pollutants modelled are CO$_2$, CH$_4$, NH$_3$, SH$_2$, CO$_2$, O$_2$, N$_2$, H$_2$O and siloxanes. This tool was used as first step to design a flexible and portable prototype for treating small flows of biogas as those produced in livestock which has been later built and is on operation.

Index Terms—Biogas upgrading, biomethane, circular economy, hybrid prototype.

I. INTRODUCTION

The European Union is fostering cleaner energy to make EU a global leader in renewable energy and ensure that the target of at least a 32% share of EU energy consumption coming from renewable energy sources is met by 2030 [1]. In this sense, renewable gases such as biogas can play an important role to make the natural gas networks greener and more environmentally friendly. To convert biogas into commercial grade natural gas fuel or biomethane, the most important step is the separation of CO$_2$ and CH$_4$ from biogas which is carried out by technologies based on unit operations like absorption or permeation. Biomethane is nowadays growing in importance as an alternative and `green´ fuel. It is obtained using several technologies to upgrade biogas, such as membranes, water scrubbing or pressure swing adsorption (PSA). In the Smart Green Gas project -a collaborative Research & Development +innovation study supported partially by the Centro para el Desarrollo Tecnológico Industrial (CDTI) that belongs to the Spanish Ministry of Economy and Competitiveness-, a hybrid flexible prototype is being designed to upgrade biogas from different sources with low flow to be able to inject low quality biomethane into the Spanish distribution natural gas network or to use it as fuel.

Spanish regulation to inject biomethane into the natural gas networks are quite restrictive and although there was a change in regulations making easier the injection of gases coming from special sources (the minimum amount of CH$_4$ in gas to be injected was lowered from 95 to 90%) are still the most hampering in Europe [2], [3]. Countries with less constraining normative, have a higher penetration of nonconventional gases, as biogas or biomethane, because treatments to meet composition of those gases with normative are not so exhaustive and therefore are economically and technically feasible.

II. OBJECTIVES

In this study we propose a novel approach to overcome this drawback by using the stage of grid injection at Metering & Regulating Stations (MRS) as a blending step to accommodate composition to Spanish regulations.

| Property                  | Units  | Minimum | Maximum |
|---------------------------|--------|---------|---------|
| Wobbe index               | kWh/m$^2$ | 13.403  | 16.058  |
| HHV                       | kWh/m$^3$ | 10.26   | 13.26   |
| Relative density          |        | 0.555   | 0.700   |
| S Total                   | mg/ m$^3$ | -       | 50      |
| H$_2$S+COS (as S)         | mg/ m$^3$ | -       | 15      |
| RSH (as S)                | mg/ m$^3$ | -       | 17      |
| O$_2$                     | mole %  | -       | 0.01*   |
| CO$_2$                    | mole %  | -       | 2.5     |
| H$_2$O (Dew point)        | °C a 70 bar | -  | +2      |
| HC (Dew point)            | °C a 1-70 bar | - | +5      |
| Dust Particles            |        | technically pure | |

During this project, a simulation tool was developed to make a quick preselection of the best unit operations to upgrade biogas. As blending in MRS can be used, it is not necessary to reach an extremely high quality in biomethane obtained that will lead probably to an unfeasible economically upgrading. Unit operations considered and modelled are:

- PSA with zeolites
- Water Scrubbing
- Water and Soda Scrubbing
- Amine Absorption
- Alcohol Absorption
- Active Carbon Filter

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- Drying with cooling unit
- Commercial membranes
- Grid Injection

Although initially materials developed during the project were tested, we considered that results obtained at lab scale will not be useful to model a real treatment (there were not enough materials and results were far to be commercial).

Most of the operations modelled are operated at high pressure or temperature, so auxiliary stages to adjust temperature or pressure are needed. To make simpler the use of the tool, model automatically makes the calculations of the adequation of temperature and or pressure and the energy consumption related to them.

TABLE II: GAS QUALITY SPECIFICATIONS FOR GAS INJECTED INTO THE SPANISH NATURAL GAS SYSTEM (BOLETÍN OFICIAL DEL ESTADO, BOE OF 23 OCTOBER 2018). TABLE EXPRESSED IN REFERENCE CONDITIONS [0°C, V/(0°C, 1.01325 BAR)]

| Property   | Units   | Minimum | Maximum |
|------------|---------|---------|---------|
| CH₄        | mole%   | 90      | -       |
| CO         | mole%   | -       | 2       |
| H₂         | mole%   | -       | 5       |
| NH₃        | mg/m³   | -       | 3       |
| F/Cl       | mg/m³   | -       | 10/1    |
| Hg         | µg/m³   | -       | 1       |
| Siloxanes  | mg/m³   | -       | 10      |
| BTEX       | mg/m³   | -       | 500     |
| Microorgan isms | - | technically pure | |
| Dust       | -       | technically pure | |

(*): The maximum O₂ content could be 0.3 % mole if the [CO₂] is less than 2 % mole and the [H₂O] is not higher than 100 ppmv.

Restrictions to inject non-conventional gases into the Spanish grid are summarized in Table I and Table II.

III. DEVELOPMENT

A. Simulation Tool to Study the Upgrading of Biogas to Biomethane

Simulation tool has been developed in Excel for make easier its usage for any user. We studied a lot of references for developing the model of every unit operation, but we present only those which are mentioned here [4]-[6].

It has been structured in sheets. The first one is the core of the tool, as it is where composition of biogas is charged and where the results of the sequence of unit operations are presented. An economical and environmental analysis is also developed. The tool was designed thinking that when different unit operations are chosen the quality of the final gas will be higher. In this sheet the sequence of unit operations is selected. It is also possible save up to 5 simulations to compare results graphically.

Substances modelled are those with are commonly present in biogas coming from manure and some landfill gas: CH₄, CO₂, H₂O, NH₃, SH₂, N₂, O₂ and siloxanes. A box “other” has been enabled to complete composition to 100% if necessary. This decision was made because this work is being developed in the framework of a national investigation project where several Spanish companies participated. These companies give the following data:

Models of each unit operations are developed in an Excel Sheet independently. Each sheet contains information relevant about the performance of the unit operation: efficiency for every substance, working pressure and temperature. Economical costs are calculated considering maintenance costs, operating costs and investment costs [4]-[6]. Environmental asset expressed as kgCO₂ equivalent is made by considering CH₄ average losses for each unit operation and calculating the equivalence of energy consumption to kgCO₂ eq. Calculus start by the condition of CO₂/SH₂ (depending on the unit operation) in the outlet flow and depending on the grade of depuration the amount of solvent or adsorbent is calculated. The tool calculates automatically if it is necessary to accommodate pressure or temperature and an estimation of the effect of these accommodations are calculated through energy consumption. Lastly, a set of alarms specific for each operation are defined, so that the user of the tool is advised when one of the operation selected is not appropriate for the composition of biogas and a pre-treatment is needed or it is better change the selection.

As it has mentioned before, composition of biogas is introduced in tab ‘PORTADA’ (Front). The tool has been designed to have the main information in the same place. As it has been explained in this tab is also the information about the composition of the biomethane, consumptions, alarms, if it is necessary to adequate pressure or temperature, it is indicated also if there is a redundant stage (when similar unit operations have been selected and no extra depuration is achieved) …and the direct access (a link) to every unit operation. When a final step of grid injection is selected, an indication of whether the biomethane can be injected into the grid or not. There is also an option to indicate the quality of the biomethane to obtain: empty or ‘Sin determinar’ (not indicated) is the higher quality, ‘Limite legal’ for legal compliance and ‘Bajo’ (low) to upgrade the biogas to a CH₄ concentration lower than legal limit. The tool is being developed in Spanish. A caption of this tab is showed in Fig. 1. Once the biogas composition has been defined and the sequence of operations is selected, the tool starts calculations and give a final result, which is shown in ‘PORTADA’. The selection of unit operations can be done easily, only displaying the list of defined operations. To avoid repeat unit operations, the chosen operation disappears from the list for next steps. Up to ten operations can be picked. By clicking ‘Grabar Simulación’ (Save simulation) data (inputs, outputs) are copied in tab ‘Resumen de resultados’ (Summary of results) and represented graphically kgCO₂ eq., CH₄ losses, maintenance and operating costs and energy consumption. See Fig. 2.
Data from operating conditions of unit operations and alarms can be changed easily in the corresponding excel sheet. If an alarm is activated for one (or more) unit operation (it can be seen in ‘PORTADA’), by clicking the name of the operation in tab ‘PORTADA’ the user can move to the subsequent sheet to see why the alarm is activated. Results of each operation selected are gathered in tab ‘Cálculos etapas’ (calculations), where other relevant information is also compiled (consumptions, costs, ΔT, ΔP,...), as can be seen in Fig. 3. Global information about the upgrading process can be seen in ‘PORTADA’. As calculations are made based on the elimination of all the CO$_2$ present (there is a minimum concentration stablished in 1%), it is possible to obtain a biogas with ‘0’ SH$_2$ as the reagent is calculated for eliminate CO$_2$ and it is in excess in relation to SH$_2$. See Fig. 4. Available information in this chart is, as it has been mentioned, consumptions, economics of the process necessary reagent (solvent or adsorbent), costs, and alarms. In this case, the alarm is activated for ‘Absorción con alcoholes’ (physical absorption).

**TABLE III: COMPOSITIONS OF BIOMAS CONSIDERED TO DETERMINE COMPOUNDS TO BE MODELLED**

| Gas      | Livestock waste (%) | Agricultural waste (%) | Sewage     | Gas    | Livestock waste (%) |
|----------|---------------------|------------------------|------------|--------|---------------------|
| CH$_4$   | 50-80               | 50-80                  | 50-70      | 45-60  |                     |

It is possible go quickly to that step by clicking on the blue button ‘Absorción con alcoholes’ (physical absorption). Then, the appearance of the excel tab for this unit operation, it is very similar for each, see Fig. 4. For every unit operation in upper side of the corresponding tab, information about de process (operating conditions, characteristics of reagent) can be found. Likewise, composition of final gas and solvent, as well as the given concentration of CO$_2$ or SH$_2$ at the outlet gas is showed. Compounds for which alarms have been settled as well as the limit for the alarm are also indicated there.

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Fig. 3. View of tab ‘Cálculos etapas’ (calculations).

Fig. 4. Upper side of one unit operation (physical absorption), as an example
Two buttons at the top of the tab of every unit operation allowing the navigation to the pages ‘PORTADA’ and ‘Cálculos etapas’ (calculations). When calculations are made based on a physical or chemical principle, related information is also found. For example, for Absorption Henry’s Law is represented graphically to obtain an equation trough a regression to make calculations. For adsorption, data from equilibrium is also depicted to obtain an equation to model the process. An example of this is shown in Fig. 5 for physical absorption. Some of the references used to develop the tool are [5]-[13], besides the specific bibliography used to model each unit operation that we don’t summarize in this document because of its extension. If there is any alarm, then the box next to ‘Alarms’ (alarms) will turn in red in Fig. 6 and in the lower side of the page it is possible check what compound/s is over the limit in the inlet Fig. 6. In table of Fig. 6, are made all the calculations as well as the amount of solvent/adsorbent needed and final flow of biogas.

**Fig. 5.** Henry’s Law for absorption of different substances in Selexol.

![Henry's constant of SH2 in Selexol](image)

**Fig. 6.** Lower part of a sheet of one unit operation.

**B. Regulating & Mixing Station (RMS)**

In this project the RMS was studied as another stage for biogas upgrading. One of the limiting aspects in Spain to inject biogas to the grid is that in most of cases achieve the composition allowed by regulations is not affordable.

However, if the last step of the sequence of unit operations is a blending process in RMS where the flow of biomethane to inject is small in comparison to the flow of natural gas in the grid it is simpler to accomplish with current law in the outlet pipe of the RMS.

Design to incorporate RMS as a blending system to ‘purify’ the biogas (Fig. 7):
- **A biogas storage system with pressure regulator.**
- **An injection flow meter.**
- **A regulating electro-valve.**
- **A Metering & Regulating Station (MRS) in which the biogas is injected.**
- **A gas sample connector.**
- **A wall plate with a pressure regulator, condensate filter and explosion proof barrier.**
- **A gas mixture dew point meter.**
- **A gas mixture analyzer of CH4, CO2, O2 and SH2.**
- **An intelligent continuous Monitoring and Actuating System (MAS) to inject biomethane in a smart way by controlling the gas parameters in mixture and enacting on the total volume of the biomethane injected.**
- **A remote server.**

In order to guarantee that the mixture meets the gas quality specifications at the Metering & Regulating Station (MRS) output, we have to continuously measure the injected biomethane flow to be able to act on different parameters: if the limit value of a parameter is breached, the intelligent system will act on the flow rate by decreasing the amount of biomethane injected so that the mixture meets the minimum quality standards of the gas distributed in the Spanish network.

Although biomethane may not have the required quality, the mixture must comply with the minimum national gas quality standards.
IV. RESULTS

Final results of the simulation for the pre-design of the hybrid prototype are shown in tab ‘PORTADA’ as well as consumptions, economical and environmental impact and alarms. Information given by the tool is shown in Fig. 8. For a given composition (for instance, a landfill gas) several trials were done being possible to record up to 5 results (trials). To execute a simulation with an average composition from a landfill gas, the following cheap treatments should be used: dehydrating, active carbon filter, PSA with zeolites operated at 6 bar. The composition of the biogas and of the biomethane obtained are shown in Table IV. However, if the gas blending solution is considered as the last upgrading stage in compliance with the Spanish natural gas regulations, then, by varying the volume ratio of the mixture the permitted results are obtained in the MRS gas output. See Table V. As the biomethane composition is not in accord with the current Spanish regulations, a last stage of blending with distributed natural gas will be used to adjust the biomethane composition to the Spanish standards on biomethane injection into the natural gas distribution network.

### TABLE IV: Inlet and Outlet Compositions of the Landfill Gas Modelled

| Property | Units  | Inlet | Outlet |
|----------|--------|-------|--------|
| CH₄      | vol%   | 45    | 87.00  |
| O₂       | vol%   | 1     | 0.10   |
| N₂       | vol%   | 5     | 5.30   |
| NH₃      | mg/m³N | 3     | 0      |
| SH₂      | ppmv   | 200   | 0      |
| CO₂      | vol%   | 40    | 7.8    |
| H₂O      | vol%   | 9     | 0      |

### TABLE V: Composition of the Natural Gas + Biomethane Blending in the MRS Gas Output

| MRS Gas Input | Biomethane injected in the MRS | MRS Gas Output |
|---------------|--------------------------------|----------------|
| CH₄ (%mole)   | 97                             | 87             |

Fig. 7. Scheme for injection of biomethane in gas grid.

![Diagram](image)

**Ethane (%mole)** 1.1 0 0.99  
**Propane (%mole)** 0.1 0 0.09  
**N₂ (%mole)** 0.7 4.93 1.123  
**CO₂ (%mole)** 1 7.98 1.698  
**O₂ (%mole)** 0 0.09 0.009  
**SH₂ (ppmv)** 5 0 4.5  
**H₂O (ppmv)** 0 0 0  
**NH₃ (mg/m³N)** 0 0 0  
**Siloxanes (mg/m³N)** 0 0 0  
**Total (%mole)** 100 100 100  
**Volume ratio (m³N)** 9 1 10  
**HHV (KWh/m³N)** 10.44 9.14 10.31  
**Molecular weight** 16.8 18.2 16.94  
**Relative density** 0.581 0.616 0.5845  
**Wobbe index (kWh/m³N)** 13.69 11.65 13.49

**Composition of the biogas at the outlet**

| Parameter | Concentration |
|-----------|---------------|
| Biogas flow (m³N/h) | 0.47 |
| CH₄ (% Vol) | 87.00% |
| O₂ (% Vol) | 0.10% |
| N₂ (% Vol) | 5.30% |
| SH₂ (ppmv) | 0.00 |
| CO₂ (%vol) | 1.00% |
| H₂O (% Vol) | 0.00% |
| Siloxanes (mg/m³N) | 0.00 |
| NH₃ (mg/m³N) | 0.00 |
| Others (%) | 0.00% |
| Temperature (ºC) | |
| Pressure (bar) | |

**Injectable?** Low

Fig. 8. Simulated composition of the biomethane at the outlet of the prototype.
V. CONCLUSION

Spanish regulations to inject biomethane into the natural gas network are so restrictive that it makes the biomethane injection in most cases economically and technically unfeasible due to the high technological investment cost to upgrade biogas to the required quality levels. In this project, we propose a novel approach to overcome this drawback using the last stage of grid injection (blending) into natural gas Metering & Regulating Stations (MRS) as an additional upgrading operation. We will use this stage to adapt the composition of the resulting blended gas to current strict Spanish regulations. This solution will help to the development of biomethane in Spain as a feasible renewable energy and the growth of small new projects near to the biogas source, for example, livestock. The variability of biogas flows and quality in these cases is saved using systems for gas storage and a final step of blending with natural gas.

To this aim and as a previous stage a simulation tool was developed to make a rapid preselection of the best operations to upgrade biogas to low or high quality biomethane given a known composition of biogas. A portable hybrid prototype was designed and constructed to test its behaviour in the upgrading of a typical landfill gas. Results of real test were like data obtained with the simulation tool, that supports the theoretical models developed.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

AUTHOR CONTRIBUTIONS

LGG worked on the development of the simulation tool and on the design and tests of the prototype and wrote the paper; S.L worked on the simulation tool and revised the paper. A.M.G worked on the simulation tool and on the design and tests of the prototype and had revised the paper; J.R.A worked on the design of the prototype and the tests; all authors had approved the final version.

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