

DESIGN AND DEVELOPMENT OF A TOOL FOR SELECTING OPERATIONS TO OBTAIN BIOMETHANE FROM BIOGAS FROM DIFFERENT SOURCES

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ABSTRACT

Biomethane is a real alternative to natural gas and a clean way to valorize biogas obtained from organic waste in landfills or manure. It can be used as fuel in vehicles and boilers or injected in natural gas grid. Nevertheless, in Spain, biogas and biomethane are 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 has been designed. Modelized operations are absorption (with water, chemical and physical), pressure swing adsorption, membranes and dehydration. A step to determine if the biomethane obtained is suitable to be injected into the grid according to current Spanish regulation has been developed. To complete the design, a conventional simulation software can be used. The tool gives information about costs, consumptions and environmental impact of each selection. Pollutants modelled are those more common in biogas coming from manure although other contaminants hardly removable are considered: CO₂, CH₄, NH₃, SH₂, CO₂, O₂, N₂, H₂O and siloxanes. Unit operations have been modelled separately, and operating conditions can be easily modified by user. Some alarms have been settled to help user to make a correct selection. With this solution, a compromise between cost (compared to commercial solutions) and accuracy is met. This tool was used as a 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. This kind of solutions could help the deployment of biomethane in Spain and help installations to reduce its emissions by valorizing a residue.

Keywords: biomethane, circular economy, simulation tool.

1 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, the renewable gases such as biogas can play an important role to make the natural gas networks greener and more environmentally friendly. Spanish regulation to inject biomethane into the natural gas networks are quite restrictive, and although there was a change in our regulation to make easier the injection of gases coming from special sources (the minimum amount of CH₄ in gas to be injected was lowered from 95% to 90%), Spanish regulations are still the most hampering in Europe [2, 3]. Countries with less constricting normative have a higher penetration of non-conventional gases, such 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.

Spain is trying to correct this trend, and in the Plan Nacional Integrado de Energía y Clima 2021–2030 (Integrated National Plan for Energy and Climate) published last March, the role of biomethane as fuel and the importance of promote its use is enhanced [4].

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 and development (R&D) plus 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 low flows of biogas from different sources to obtain low-quality biomethane to be injected into the Spanish natural gas grid or to use it as fuel. In this study, we developed a simulation tool for a quick selection of treatments for biogas upgrading. From this tool, we obtain a first estimation of the performance of the selected treatments to determine whether the biomethane obtained meets regulations to be injected in natural gas grid or not. We also propose a novel approach to inject low-quality biomethane (not accomplishing legal limits) using the stage of grid injection at Metering & Regulating Stations (MRS) as a blending step where biomethane is mixed with natural gas, obtaining a gas which composition is accomplish Spanish. At latest, we compare results from our tool with a prototype to adjust theoretical efficiency obtained from tool.

2 OBJECTIVES

The main objective of this work is to develop a simulation tool where the unit operations based on adsorption, absorption and permeation are modelled using the physical equations that rules their performance and data available in bibliography. With this tool, a first quick selection of best unit operations for treating biogas of a given composition to reach a certain composition, high or low, can be made. From the results of the simulation tool, a pre-design of an upgrading pilot plant is obtained. In the framework of the project, a pilot plant of 1 m³N/h has been built combining several operations according to the results of the previous stage (simulation tool). Blending in MRS is studied as a way of obtaining high-quality biomethane starting from a low-quality biomethane. Unit operations considered and modelled are PSA with zeolites, water and soda scrubbing, chemical and physical absorption, active carbon filter, drying, membranes and grid injection. At this time, the tool is almost developed (all the unit operations are modelled), and we are working in small details or improvements of the simulation tool.

Restrictions to inject non-conventional gases into the Spanish grid are summarized in Table 1. Presence of dust or particles is not allowed. 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, the model automatically makes calculations to adequate temperature and or pressure and the energy consumption related to it.

3 EXPERIMENTAL

Simulation tool has been developed in Excel to make easier its usage for anyone. It has been structured in sheets. The first one 'PORTADA' (Front) is the core of the tool, as is where composition of biogas is entered and where the results of the sequence of unit operations are presented. An economical and environmental analysis is also developed and shown in this sheet. The tool was designed thinking that whether several unit operations are selected, the quality of the final gas will be higher. In sheet 'PORTADA,' the sequence of unit operations is selected. It is also possible save up to five simulations to compare results graphically. Substances modelled are those that are commonly present in biogas coming from manure and

Table 1: Gas quality specifications for gas injected into Spanish gas natural system.

Gas quality specifications for gas injected into the Spanish natural gas system (0°C, 1.01325 bar) [2]				Gas quality specifications for gas from non-conventional sources injected into the Spanish natural gas system) (0°C, 1.01325 bar) [3]			
Property	Units	Min.	Max.	Property	Units	Min.	Max.
Wobbe index	kWh/m ³	13.403	16.058	CH ₄	mole %	90	–
HHV	kWh/m ³	10.26	13.26	CO	mole %		2
Rel. density	mg/m ³	0.555	0.700	H ₂	mole %	–	5
S Total	mg/m ³	–	50	NH ₃	mg/m ³	–	3
H ₂ S + COS (as S)	mg/m ³	–	15	F/Cl	mg/m ³	–	10/1
RSH (as S)	mg/m ³	–	17	Hg	µg/m ³	–	1
O ₂	mole %	–	0.01*	Siloxanes	mg/m ³	–	10
CO ₂	mole %	–	2.5	BTX	mg/m ³	–	500
H ₂ O (dew point)	°C 70 bar	–	+2	Microog.	–	Technically pure	
HC (dew point)	°C 1–70 bar	–	+5				

Table 2: Average compositions considered in this investigation.

Gas	Livestock waste (%)	Agricultural waste (%)	Sewage sludge (%)	Urban waste (%)	Landfill gas (%)
CH ₄	50–80	50–80	50–80	50–70	45–60
CO ₂	30–50	30–50	20–50	30–50	40–60
N ₂	0–1	0–1	0–3	0–1	2–5
H ₂ O	Saturated	Saturated	Saturated	Saturated	Saturated
H ₂	0–2	0–2	0–5	0–2	0–0.2
SH ₂	0–1%	100–700 ppm	0–1	0–8	0–1
NH ₃	Traces	Traces	Traces	Traces	0.1–1%
CO	0–1	0–1	0–1	0–1	0–0.2
N ₂	0–1	0–1	0–3	0–1	2–5
O ₂	0–1	0–1	0–1	0–1	0.1–1
Organic compounds	Traces	Traces	Traces	Traces	0.01–06*

some landfill gas: CH_4 , CO_2 , H_2O , NH_3 , SH_2 , N_2 , O_2 and siloxanes. A box 'others' has been enabled to complete composition to 100% if necessary. Companies participating in the R&D project described in the introduction give the following composition for biogas.

Models of each unit operation are developed independently in different Excel sheet. 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. Environmental asset expressed as kgCO_2 equivalent is made by considering CH_4 average losses for each unit operation and calculating the equivalence of energy consumption with kgCO_2 eq. Calculus start by the desired concentration of CO_2/SH_2 (depending on the unit operation) in the outlet of the unit operation, 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 adaptations is calculated through energy consumption. Lastly, a set of alarms specific for each operation is defined so that the user of the tool is advised if one of the operations selected is not appropriate for the composition of biogas and a pre-treatment is needed.

The tool has been designed to have the main information in tab 'PORTADA'. As it has been explained in this tab is also available the information about the composition of the biomethane, consumptions and alarms, whether it is necessary to adequate pressure or temperature or not. It is also indicated if there is a redundant stage (when similar unit operations have been selected and no extra depuration is achieved), and in this sheet, there is direct access (a link to go to the sheet where that unit operation is modelled) for each unit operation. If a final step of grid injection is selected, an indication of whether the biomethane can be injected into the grid or not (for high flows of biomethane) and final composition. There is also an option to indicate the quality of the biomethane to obtain: empty or 'Sin determinar' (not indicated) is the higher quality, 'Límite legal' for legal compliance and 'Bajo' (low) to upgrade the biogas to a CH_4 concentration lower than legal limit. The tool is being developed in Spanish. A caption of this tab is shown in Fig. 1.

Once the composition of the biogas has been defined and the sequence of operations selected, the tool starts calculations and gives a result, which is shown in 'PORTADA'. The selection of unit operations can be done easily, only displaying the list of defined operations. To avoid repeating unit operations, the selected operation disappears from the list of the next stage. Up to ten operations can be chosen.

By clicking 'Grabar Simulación' (save simulation), data (inputs, outputs) are copied in tab 'Resumen de resultados' (summary of results) and represented graphically kgCO_2 eq., CH_4 losses, maintenance and operating costs and energy consumption. See Fig. 2.

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, by clicking in the name of the operation in tab 'PORTADA', the user can move to the corresponding 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 calculus are made based on the elimination of almost all the CO_2 present (there is a minimum concentration established in 1%), it is possible to obtain a biogas with '0' SH_2 as the solvent is calculated to eliminate CO_2 and it is in excess in relation to SH_2 .

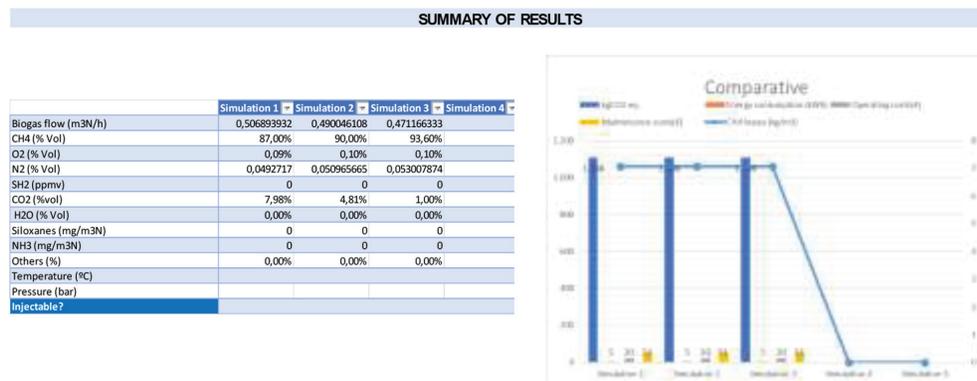


Figure 2: Results shown in ‘Resumen de resultados’ (Summary of Results), also depicted graphically to compare different treatments.

SEQUENCE	Initial	SEQUENCE OF STAGES										Final
	1	2	3	4	5	6	7	8	9	10		
Biogas flow (m3N/h)	1,00	0,920068452	0,909910819	0,506893919								0,51
CH4 (% Vol)	45,00%	48,91%	49,46%	0,87								87,00%
O2 (% Vol)	1,00%	1,09%	1,10%	0,000949774								0,09%
N2 (% Vol)	5,00%	5,43%	5,49%	0,049271701								4,93%
SH2 (ppmv)	200,00	217,3751307	3,5	0								0,00%
CO2 (%vol)	40,00%	43,48%	43,96%	0,079778525								7,98%
H2O (% Vol)	9,00%	1,07%	0,00%	0								0,00%
Siloxanes (mg/m3N)	8,00	0	0	0								0,00%
NH3 (mg/m3N)	3,00	0	0	0								0,00%
Others (%)	0,00%	0	0	0								0,00%
Temperature (°C)	0,00	40	50	5								
Pressure (bar)	0,00	1	8	6								
ΔT (°C)		40,00	10,00	-45,00								Temperature adequacy
ΔP (°C)		1,00	7,00	-2,00								Pressure adequacy
m3/h solvent-kg/h reagent		3,644860733	0,001052771	3,777084554								
CH4 losses(kg/m3)		0	1,81441E-06	7,065019899								7,065021714
kg CO2 eq		855	52,44393986	200,2303346								1,107,67
Operating consumption (kWh)		4,5	0,276020536	0,272973246								5,048993781
Operating costs		20,25	0,011040821	0,01091893								20,27
Investment costs		3000	0,003959333	2700								5,700,00
Maintenance costs		0	0,005960376	50,95500585								50,96
	1	2	3	4	5	6	7	8	9	10		FINAL

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

For every unit operation in upper side, there is an information about the process (operating conditions, characteristics of solvent/adsorbent ...). Likewise, composition of final gas and solvent, as well as the amount of reactive needed for a given concentration of CO₂ or SH₂ at the outlet gas, is shown. Compounds for which alarms have been settled as well as the limit for the alarm are also indicated there. Two buttons at the top of the Excel allow the navigation to the pages ‘PORTADA’ (Front) and ‘Cálculos etapas’ (calculations).

When calculations are made based on a physical or chemical principle, related information is also gathered. For example, in Absorption steps, Henry’s Law is represented graphically to obtain a regression equation to make calculations. For adsorption, data from equilibrium are also depicted to obtain an equation to model the process. An example of this is shown in Fig. 6 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 ‘Alarmas’ (alarms) will turn on red, and in the lower side of the page, it is possible to check what compound(s) is over the limit (Fig. 6). In

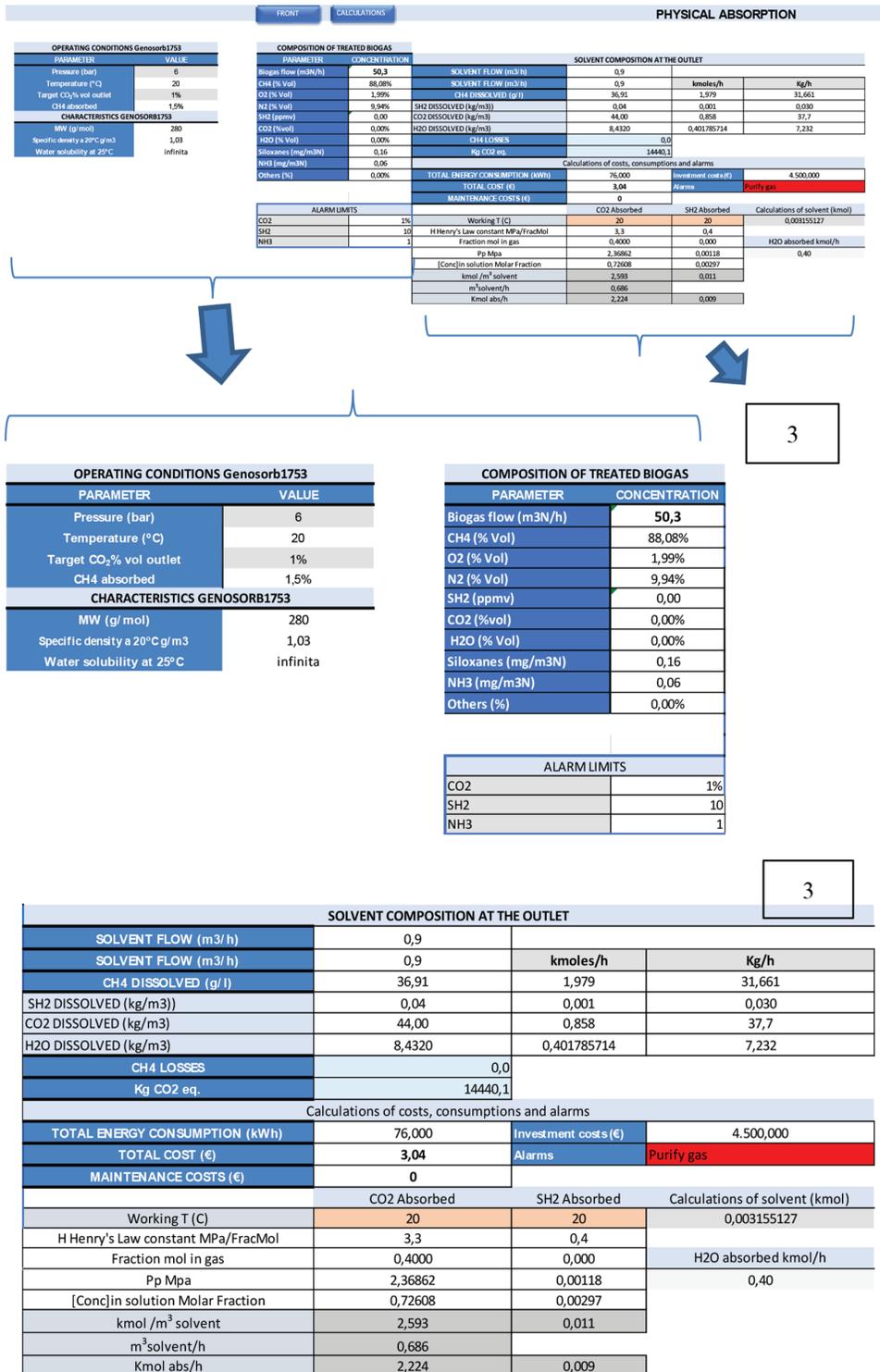


Figure 4: Upper side of one unit operation (physical absorption), as an example.

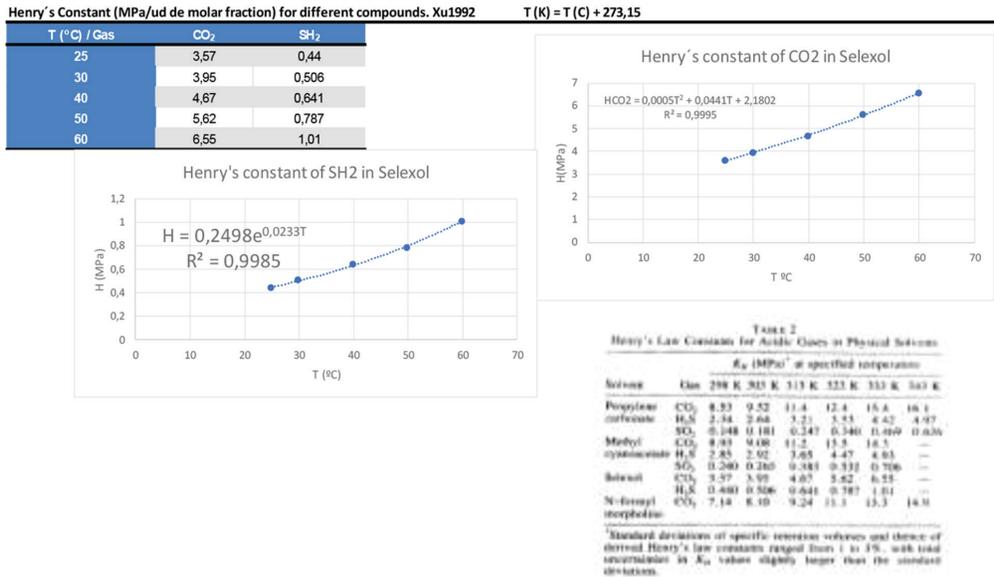


Figure 5: Henry's Law for absorption of different substances in Selexol.

Composition of biogas at the inlet		m ³ /h solvent		Final Flow Nm ³ /h			
100,00		0,7	0,9	50,3			
Design factor		1,25					
Biogas flow (m ³ N/h)	100,00	m ³	kMoles	kmol absorbed	kmol outlet	m ³ outlet	% Outlet
CH ₄ (% Vol)	45,00%	45,0	2,0	0,0301	1,9788	44,3	88,08%
O ₂ (% Vol)	1,00%	1,0	0,0446	0	0,0446	1,0	1,99%
N ₂ (% Vol)	5,00%	5,0	0,2232	0	0,2232	5,0	9,94%
SH ₂ (ppmv)	200,00	0,020	0,0009	0,00	0,0000	0,0	0,00%
CO ₂ (%vol)	40,00%	40,00	1,786	1,79	0,0000	0,0	0,00%
H ₂ O (% Vol)	9,00%	9,0	0,402	0,40	0,0000	0,0	0,00%
Siloxanes (mg/m ³ N)	8,00	0,00	0,000	0,00	0,0000	0,0	0,00%
NH ₃ (mg/m ³ N)	3,00	0,0	0,000	0,00	0,0000	0,0	0,00%
Others (%)	0,00%	0,000	0,000	0,00	0,000	0,0	0,00%
Total		4,465	2,219	2,247		50,326	1,00%

Figure 6: Lower part of a sheet of one unit operation.

the table, all the calculations are made including the amount of solvent/adsorbent needed and final flow of biogas.

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 to achieve the composition allowed by regulations is not affordable. To overcome this, 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 is designed to accomplish with current law in the outlet pipe of the RMS. Design to incorporate RMS as a blending system to 'purify' the biogas schematically, as shown in Fig. 7, consists of:

- A biomethane storage system with pressure regulator.
- An injection flow meter.
- A regulating electro-valve.
- A MRS in which the biomethane 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.

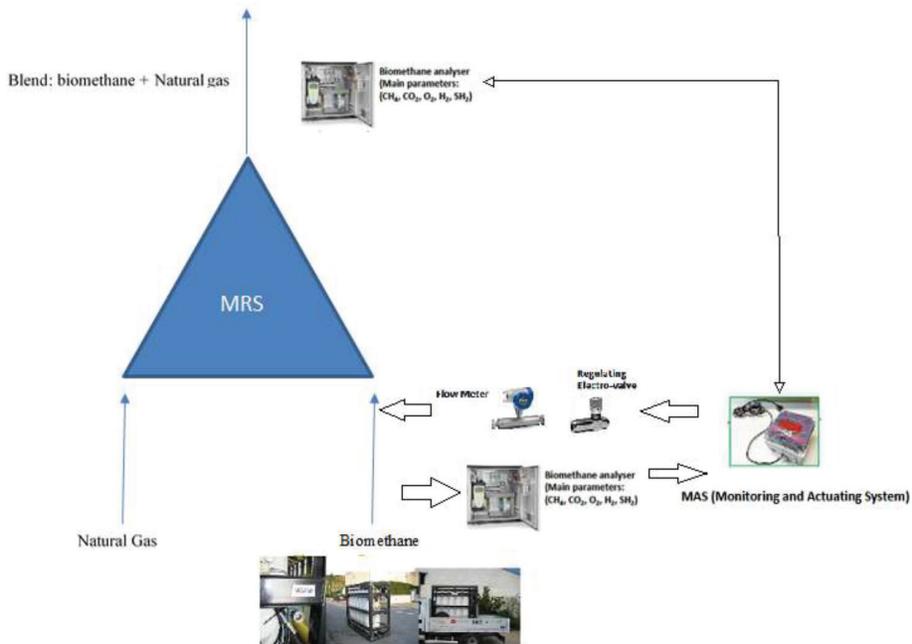


Figure 7: Scheme of the injection system.

- A gas mixture analyser of CH_4 , CO_2 , O_2 , H_2 and SH_2 .
- 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 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 have to 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.

4 RESULTS AND DISCUSSION

A simulation with an average composition of a landfill gas was done. Several treatments were modelled, being the most suitable (for a cheap solution) the one combining dehydration, active carbon filter and PSA with zeolites operated at 6 bar (Fig. 8).

Composition of the biogas and the biomethane obtained from the models is shown in Table 3. Results obtained in the tool developed are shown in Fig. 9.

These results were got using the option 'Bajo' (Low) in tab 'PORTADA', to obtain a cheap biomethane that meets legal limits thanks to a final step of blending in RMS.

Results of intermediate stages are collected in Fig. 9. Thus, and according to the current Spanish regulations, the injection of biomethane into the natural gas distribution network is not feasible. See again the last column of Fig. 9. But 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 4.

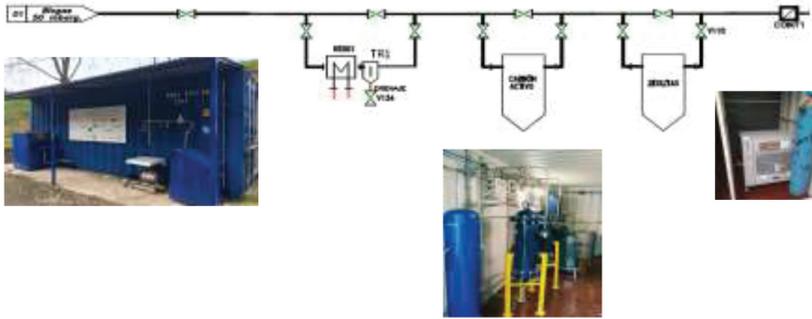


Figure 8: Prototype.

Table 3: Inlet and outlet composition of the landfill gas modelled.

Property	Units	Inlet	Outlet
CH ₄	vol%	45	87.00
O ₂	vol%	1	0.10
N ₂	vol%	5	5.30
NH ₃	mg/m ³ N	3	0
SH ₂	ppmv	200	0
CO ₂	vol%	40	7.8
H ₂ O	vol%	9	0

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 (°C)	
Pressure (bar)	

Injectable? Quality Low

New simulation

Record simulation

Other information	
Stage	Reagents consumption
DEHYDRATION	3,644860733
ACTIVE CARBON FILTER	0,001052771
PSA ZEOLITES	3,777084554

Temperature adequacy	
Pressure adequacy	
CH4 losses (kg/m3)	7,0650
kg CO2 eq	1107,67
Consumption (kWh)	5,05
Operating cost(€)	20,27
Maintenance Costs (€)	50,96

Stage Alarms	
PSA ZEOLITES	Purify gas

Figure 9: Final results and information given by the tool.

Table 4: Composition of the natural gas + biomethane blending in the MRS gas output.

	MRS gas input	Biomethane injected in the MRS	MRS gas output
Methane (%mole)	97	87	96
Ethane (%mole)	1.1	0	0.99
Propane (%mole)	0.1	0	0.09
N ₂ (%mole)	0.7	4.93	1.123
CO ₂ (%mole)	1	7.98	1.698
O ₂ (%mole)	0	0.09	0.009
SH ₂ (ppmv)	5	0	4.5
H ₂ O (ppmv)	0	0	0
NH ₃ (mg/m ³ N)	0	0	0
Siloxanes (mg/m ³ N)	0	0	0
Total (%mole)	100	100	100
Volume ratio (m ³ N)	9	1	10
HHV (KWh/m ³ N)	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/m ³ N)	13.69	11.65	13.49
Methane number	92.7	76.6	91.09

As the biomethane composition is not in accordance 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 line. The quality of biogas obtained affects to treatment cost as it reduces the amount of chemicals as well as power consumption.

The prototype constructed was tested under real conditions with a landfill gas. Composition of the biomethane in the outlet of the prototype and its change with time was measured employing an Agilent 490 Micro Gas Chromatograph. We are not allowed to show data from the composition of the biogas in the inlet and the upgraded biomethane, but we can affirm that the performance of the pilot plant achieved a 95%, being the minimum molar fraction of CO₂ in the outlet next to 1% (this value is set up in the tool as final concentration to start calculations) using a zeolite 13X.

The pilot plant was designed to be portable to make it more usable and affordable, as it could be shared with nearby biogas generating facilities.

SEQUENCE	1	2	3	4	Final
Biogas flow (m3N/h)	1,00	0,920068452	0,909910819	0,471166333	0,47
CH4 (% Vol)	45,00%	48,91%	49,46%	0,87	0,00%
O2 (% Vol)	1,00%	1,09%	1,10%	0,001021793	0,10%
N2 (% Vol)	5,00%	5,43%	5,49%	0,053007874	5,30%
SH2 (ppmv)	200,00	217,3751307	3,5	0	0,00%
CO2 (%vol)	40,00%	43,48%	43,96%	0,01	94,59%
H2O (% Vol)	9,00%	1,07%	0,00%	0	0,00%
Siloxanes (mg/m3N)	8,00	0	0	0	0,00%
NH3 (mg/m3N)	3,00	0	0	0	0,00%
Others (%)	0,00%	0	0	0	0,00%
Temperature (°C)	0,00	40	50	5	
Pressure (bar)	0,00	1	8	6	
ΔT (°C)		40,00	10,00	-45,00	Temperature adequacy
ΔP (°C)		1,00	7,00	-2,00	Pressure adequacy
m3/h solvent-kg/h reagent		3,644860733	0,001052771	3,777084554	
CH4 losses(kg/m3)		0	1,81441E-06	7,065019899	7,065021714
Kg CO2 eq		855	52,44393986	200,2303346	1,107,67
Operating consumption (kWh)		4,5	0,276020536	0,272973246	5,048993781
Operating costs		20,25	0,011040821	0,01091893	20,27
Investment costs		3000	0,003959333	2700	5.700,00
Maintenance costs		0	0,005060376	50,95500585	50,96
	1	2	3	4	FINAL

Figure 10: Results of intermediate stages.

5 CONCLUSIONS

Spanish regulations to inject biomethane into the natural gas grid 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 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, e.g. live-stock. The variability of biogas flows and quality in these cases are 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 pre-selection 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.

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