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_2O 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 CH4 in gas to be injected was lowered from 95 to 90%) are still the most hampering in Europe [2], [3]. Countries with less constricting 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.

Manuscript received August 23, 2021; revised September 7, 2021. This work was supported in part by the CDTI (in Spain) and FEDER funds.

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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.

TABLE I: Gas Quality Specifications for Gas Injected into the Spanish Natural Gas System [1]

Property	Units	Minimum	Maximum
Wobbe index	kWh/m^3	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_2S+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_2O (Dew point)	℃ a 70 bar	-	+2
HC (Dew point)	℃ 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

doi: 10.18178/jocet.2021.9.3.529

- 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
CH_4	mole%	90	-
CO	mole%	-	2
H_2	mole%	-	5
NH_3	mg/m^3	-	3
F/Cl	mg/m^3	-	10/1
Hg	$\mu g/m^3$	-	1
Siloxanes	mg/m^3	-	10
BTX	mg/m^3	-	500
Microorganisms	-	technically pure	
Dust Particles	-	technically pure	

(*): The maximum O_2 content could be 0.3 % mole if the [CO_2] is less than 2 % mole and the [H_2O] 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: CH_4, CO_2, H_2O, NH_3, SH_2, N_2, O_2 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 $kgCO_2$ equivalent is made by considering CH_4 average losses for each unit operation and calculating the equivalence of energy consumption to $kgCO_2$ eq. Calculus start by the condition of CO_2/SH_2 (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 specifical 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, '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 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 $kgCO_2$ eq., CH_4 losses, maintenance and operating costs and energy consumption. See Fig. 2.

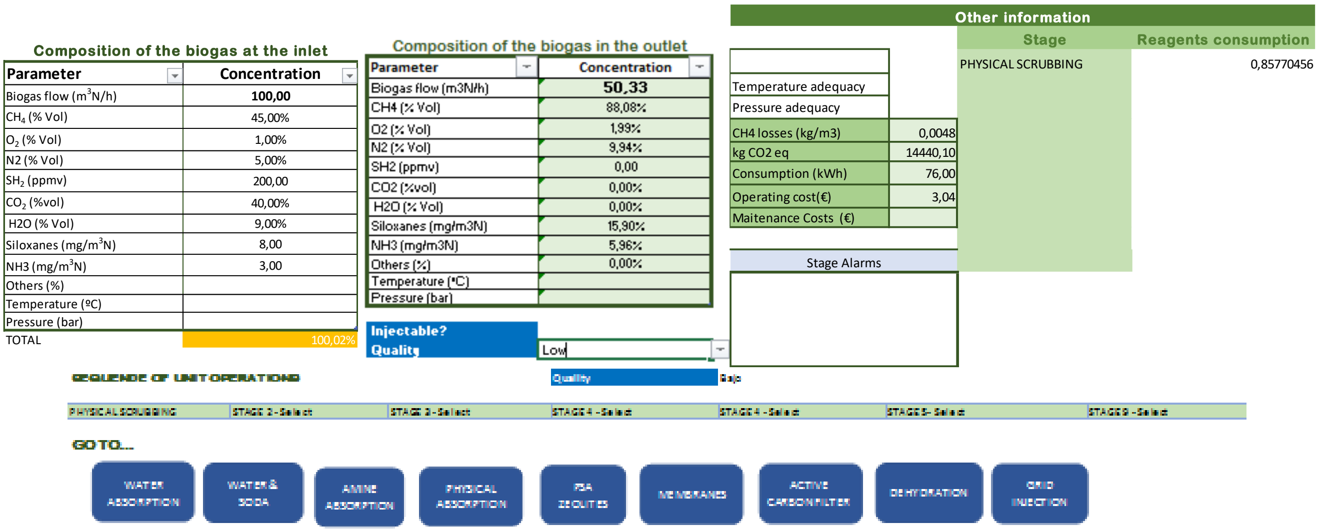

Fig. 1. Caption of tab 'PORTADA' (Front). It has been divided for a better comprehension.

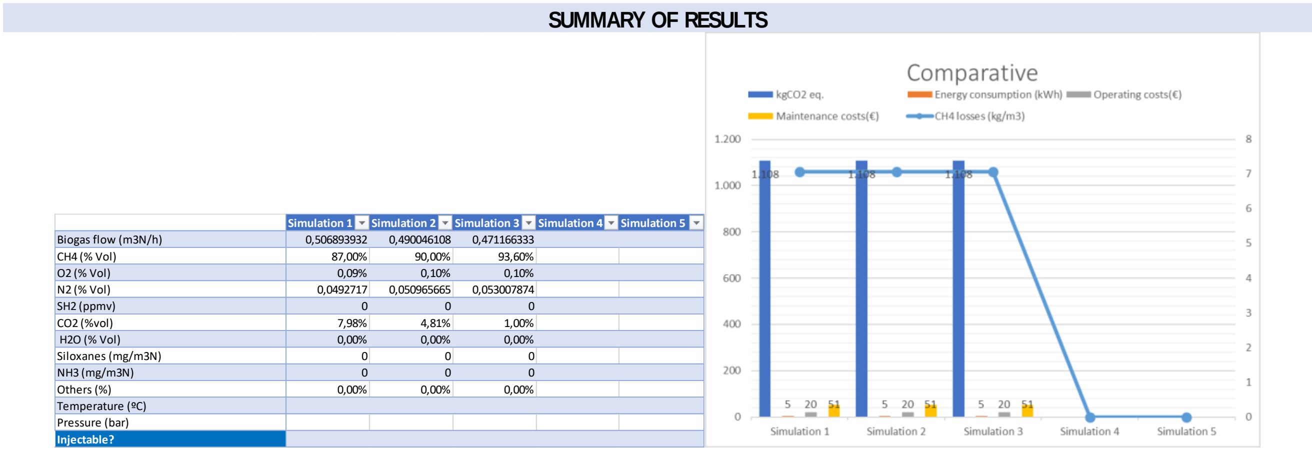

Fig. 2. Representation of results in 'Resumen de resultados' (summary results).

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 calculus 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 BIOGAS CONSIDERED TO DETERMINE COMPOUNDS TO BE MODELLED

Gas	Livestock waste (%)	Agricultural waste (%)	Sewage	Gas	Livestock waste (%)
CH_4	50-80	50-80	50-80	50-70	45-60
CO_2	30-50	30-50	20-50	30-50	40-60
H_2O	Saturated	Saturated	Saturated	Saturated	Saturated
H_2	0-2	0-2	0-5	0-2	0-0.2
SH_2	0-1%	100-700ppm	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(*)

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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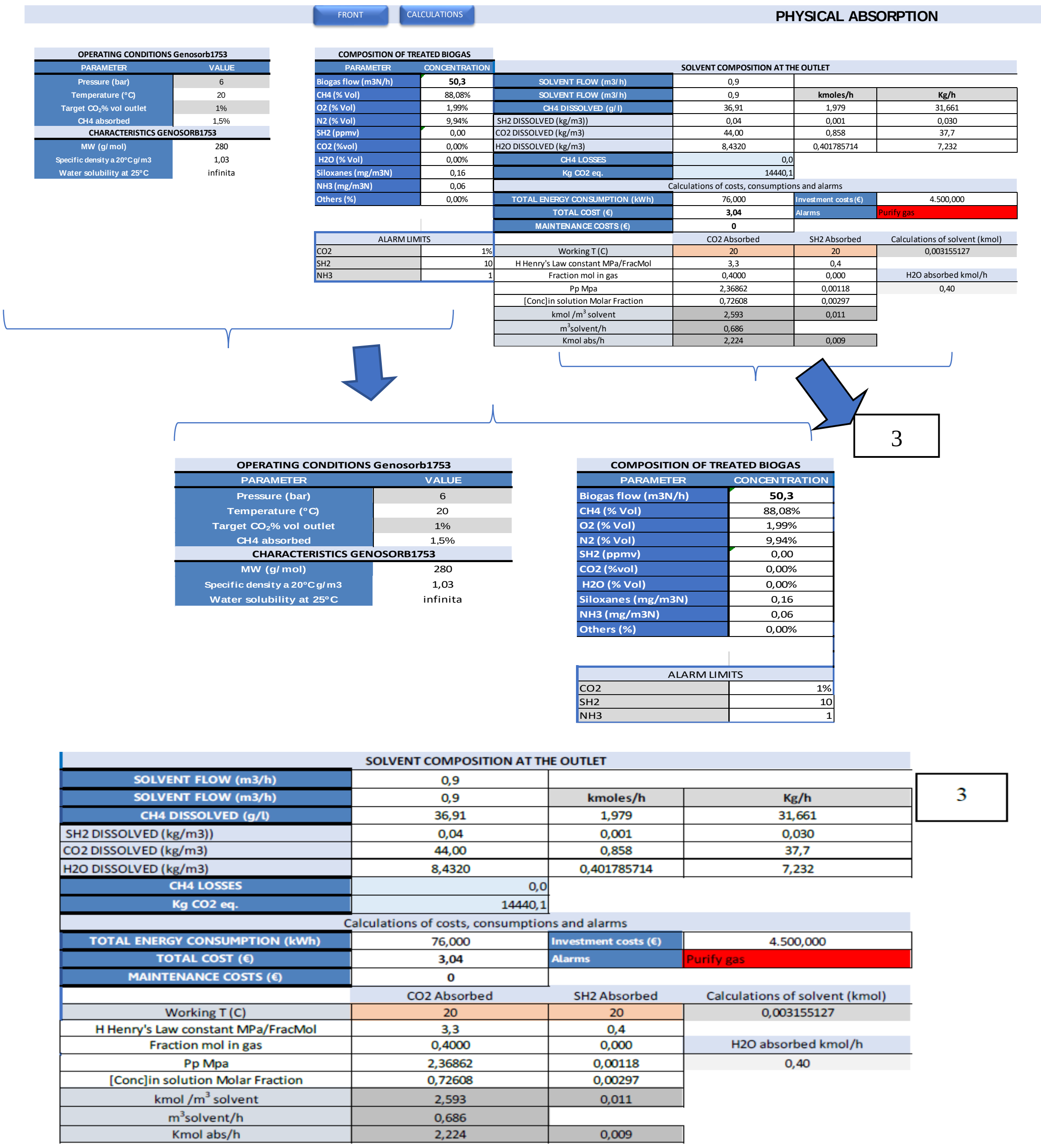
	Initial	SECUENCE OF STAGES								Final
SEQUENCE	1	2	3	4	5	6	7	8	9	10	
Biogas flow (m3N/h)	1,00	0,920068452	0,909910819	0,506893919							0,51
CH4 (% Vol)	45,00%	48,91%	49,46%	0,87							87,00%
O2 (% Vol)	1,00%	1,09%	1,10%	0,000949774							0,09%
N2 (% Vol)	5,00%	5,43%	5,49%	0,049271701							4,93%
SH2 (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 (ºC)	0,00	40	50	5							
Pressure (bar)	0,00	1	8	6							
ΔT (ºC)		40,00	10,00	-45,00							Temperature adequacy
ΔP (ºC)		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	5	6	7	8	9	10	FINAL

Fig. 3. View of tab 'Cálculos etapas' (calculations).



OPERATING CONDITIONS Genosorb1753	
PARAMETER	VALUE
Pressure (bar)	6
Temperature (°C)	20
Target CO_2% vol outlet	1%
CH4 absorbed	1,5%
CHARACTERISTICS GENOSORB1753	
MW (g/mol)	280
Specific density a 20°C g/m3	1,03
Water solubility at 25°C	infinita

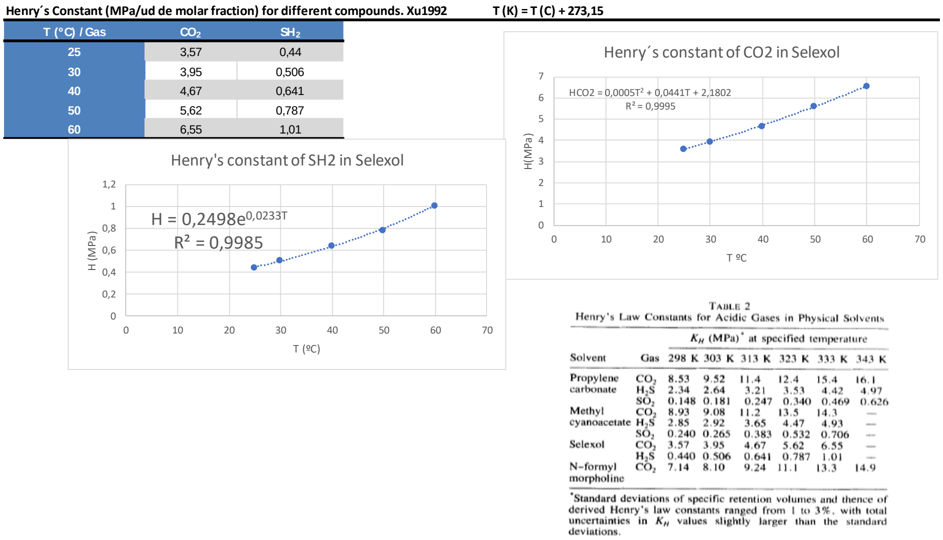
COMPOSITION OF TREATED BIOGAS	
PARAMETER	CONCENTRATION
Biogas flow (m3N/h)	50,3
CH4 (% Vol)	88,08%
O2 (% Vol)	1,99%
N2 (% Vol)	9,94%
SH2 (ppmv)	0,00
CO2 (%vol)	0,00%
H2O (% Vol)	0,00%
Siloxanes (mg/m3N)	0,16
NH3 (mg/m3N)	0,06
Others (%)	0,00%

ALARM LIMITS	
CO2	1%
SH2	10
NH3	1

SOLVENT COMPOSITION AT THE OUTLET			
SOLVENT FLOW (m3/h)	0,9		
SOLVENT FLOW (m3/h)	0,9	kmoles/h	Kg/h
CH4 DISSOLVED (g/l)	36,91	1,979	31,661
SH2 DISSOLVED (kg/m3))	0,04	0,001	0,030
CO2 DISSOLVED (kg/m3)	44,00	0,858	37,7
H2O DISSOLVED (kg/m3)	8,4320	0,401785714	7,232
CH4 LOSSES	0,0		
Kg CO2 eq.	14440,1		
Calculations of costs, consumptions and alarms			
TOTAL ENERGY CONSUMPTION (kWh)	76,000	Investment costs (€)	4.500,000
TOTAL COST (€)	3,04	Alarms	Purify gas
MAINTENANCE COSTS (€)	0		
	CO2 Absorbed	SH2 Absorbed	Calculations of solvent (kmol)
Working T (C)	20	20	0,003155127
H Henry's Law constant MPa/FracMol	3,3	0,4	
Fraction mol in gas	0,4000	0,000	H2O absorbed kmol/h
Pp Mpa	2,36862	0,00118	0,40
[Conc]in solution Molar Fraction	0,72608	0,00297	
kmol /m^3 solvent	2,593	0,011	
m^{3}solvent/h	0,686		
Kmol abs/h	2,224	0,009	

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 'Alarmas' (alarms) will turn in red 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.


Henry´s Constant (MPa/ud de molar fraction) for different compounds. Xu1992 T (K) = T (C) + 273,15

T (°C) / Gas	CO_2	SH_2
25	3,57	0,44
30	3,95	0,506
40	4,67	0,641
50	5,62	0,787
60	6,55	1,01

TABLE 2
Henry's Law Constants for Acidic Gases in Physical Solvents

Solvent	Gas	298 K	303 K	313 K	323 K	333 K	343 K
		K_H (MPa)* at specified temperature					
Propylene carbonate	CO_2	8.53	9.52	11.4	12.4	15.4	16.1
	H_2S	2.34	2.64	3.21	3.53	4.42	4.97
	SO_2	0.148	0.181	0.247	0.340	0.469	0.626
Methyl cyanoacetate	CO_2	8.93	9.08	11.2	13.5	14.3	—
	H_2S	2.85	2.92	3.65	4.47	4.93	—
	SO_2	0.240	0.265	0.383	0.532	0.706	—
Selexol	CO_2	3.57	3.95	4.67	5.62	6.55	—
	H_2S	0.440	0.506	0.641	0.787	1.01	—
N-formyl morpholine	CO_2	7.14	8.10	9.24	11.1	13.3	14.9

*Standard deviations of specific retention volumes and thence of derived Henry's law constants ranged from 1 to 3%, with total uncertainties in K_H values slightly larger than the standard deviations.

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

		m3/h solvent	0,7	0,9		Final Flow Nm3/h	
Composition of biogas at the inlet		Design factor	1,25			50,3	
Biogas flow (m3N/h)	**100,00**	**m3**	**kMoles**	**kmol absorbed**	**kmol outlet**	**m3 outlet**	**% Outlet**
CH4 (% Vol)	45,00%	45,0	2,0	0,0301	1,9788	44,3	88,08%
O2 (% Vol)	1,00%	1,0	0,0446	0	0,0446	1,0	1,99%
N2 (% Vol)	5,00%	5,0	0,2232	0	0,2232	5,0	9,94%
SH2 (ppmv)	200,00	0,020	0,0009	0,00	0,0000	0,0	0,00%
CO2 (%vol)	40,00%	40,00	1,786	1,79	0,0000	0,0	0,00%
H2O (% Vol)	9,00%	9,0	0,402	0,40	0,0000	0,0	0,00%
Siloxanes (mg/m3N)	8,00	0,00	0,000	0,00	0,0000	0,0	0,00%
NH3 (mg/m3N)	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	1,000

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 biomethane storage system with pressure regulator.
- An injection flow meter.
- A regulating electro-valve.
- A Metering & Regulating Station (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.
- A gas mixture analyzer 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 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.

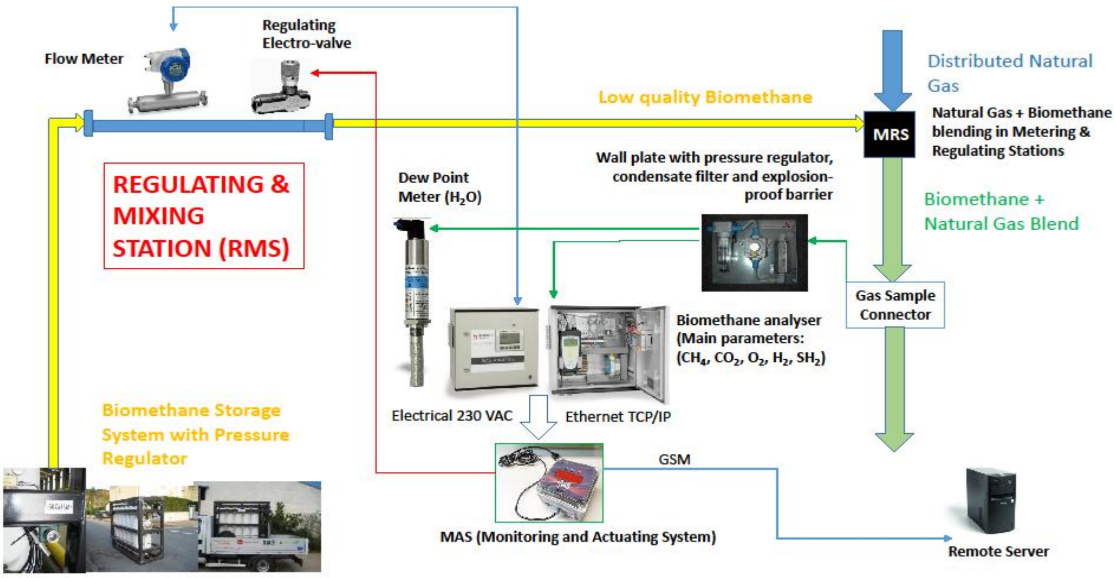


Fig. 7. Scheme for injection of biomethane in gas grid.

IV. Results

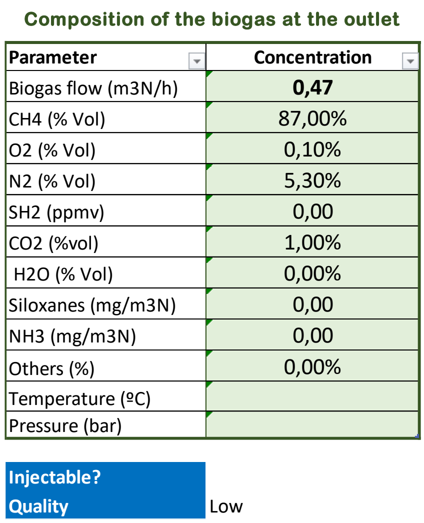
Final results of the simulation for the pre-design of the hybrid prototype are shown in tab 'PORTADA' as well as consumptions, economical and environmental impact and alarms. Information given by the tool is shown in Fig. 8. For a given composition (for instance, a landfill gas) several trials were done 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 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 distribution network.

TABLE IV: Inlet and Outlet Compositions 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_2O	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
CH_4 (%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_2O (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


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? Quality: 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.

Acknowledgment

We would like to express our gratitude to Nortegas Energía Distribución SAU and to the 'Vertedero de Lapatx' managed by Urola Erdiko Lapatx Zabortegia S.A.

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