

## ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

## SEMESTER PROJECT WITH GEM

Cleaning of biomass derived gasses for Solid Oxide Fuel Cell applications

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### Abstract

In the context of the Waste 2 Watts (W2W) European project, to design and install a biogas fed Solid Oxyde Fuel Cell (SOFC), this review focus on the requirements, costs and performance of a biogas cleaning unit for SOFC applications. The production cost and performance of Anaerobic Digestion (AD) and Biomass Gasification (BG) are compared. The concentration and variation of contaminants present in these gasses is reported. Their effect on a high temperature SOFC is considered and the methods for removing them presented. A literature review on the cost and performance of a biogas cleaning unit shows that it has a low impact on system performance (about 3% electricity losses) and high costs on the order of  $1'300 \text{ EUR}/kW_{th}$  for smaller scale systems. The main reasons for this high cost and ways to reduce it are discussed.

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## List of abbreviations

Gas Cleaning Section

 $\Delta P$  Pressure Drop

AC Activated Carbon Filter

General

bio-SNG bio Synthetic Natural Gas

CAPEX Capital Expenditures

CEPCI Chemical Engineering Plant Cost Index

LHV Lower Heating Value

**OPEX** Operating Expenditures

PSI Paul Scherrer Institut

RDF Refuse Derived Fuels

TOTEX Total Expenditures

WWTP Waste Water Treatment Plant

**Technologies** 

AD Anaerobic Digestion

BG Biomass Gasification

CHP Cogeneration Heat and Power

ICE Internal Combustion Engine

IGCC Integrated Gasification Combined Cycle

MFCF Molten Carbonate Fuel Cell

SOFC Solid Oxide Fuel Cell

#### 1 Introduction

Energy demand is steadily increasing, while Green House Gas emissions (GHG) have to be reduced. One way to tackle this problem is to produce energy from waste. Apart from burning waste in an incinerator, biogas may be produced from wet biomass via Anaerobic Digestion (AD) and woody biomass can be used to produce bio-SNG with Biomass Gasification (BG). Two carbon-neutral fuels that can be stored or upgraded and used for transport. Moreover, biogas can be used in Solid Oxide Fuel Cells (SOFC) for the Co-generation of Heat and Power (CHP), increasing the energy efficiency of the system, without the need to remove the CO<sub>2</sub> fraction. Since biomass is usually not available in high concentrations, except for Waste Water Treatment Plants (WWTP), it is interesting to focus on a small scale CHP installations, fed by biomass, since less transport improves the overall process efficiency.

This review is set in the context of the Waste2Watts Project (W2W) [26]. Figure 1 shows the overall process flow diagram of the W2W project. Cleaning takes place before the biogas is fed into the SOFC (Process 2) and refers to removing contaminants that my poison the anode or cathode. Upstream, an anaerobic digester operating at 60°C, fed by wet biomass produces the biogas. Alternatively a gasifier operating above 700°C producing bio Synthetic Natural Gas (bio-SNG) using woody biomass may replace the AD. Downstream, the clean biogas is consumed in a high-temperature SOFC operating between 700 and 850°C.

SOFC degradation is mainly driven by catalyst poisoning [15] and thermal cycling [11]. If both of these are controlled adequately, the life of the SOFC stack can be drastically increased. In the literature, the two main families of biogas contaminants that are most detrimental to the SOFC are Sulfurs and siloxanes [15] [4] [10], which have to be removed to below 1ppm and a few ppb respectively, to prevent fuel cell degradation over time. Research on this topic has focused on the impact on Ni/Al2O3 and Ni/YSZ catalysts, the most common type of SOFC in the industry, however, through doping, resistant to Sulfurs and other contaminants may be improved.

This review looks at biogas production and deep cleaning for SOFC applications specifically, with a focus on the latter. First, the cost of biogas and bio-SNG installations are presented, then the variation of contaminants and typical concentrations for a given biomass are identified. SOFC tolerance and degradation for different contaminants is evaluated and common cleaning techniques are presented. In the last part, the design, cost and performance of the biogas cleaning process is related to its scale, and integration opportunities are discussed.

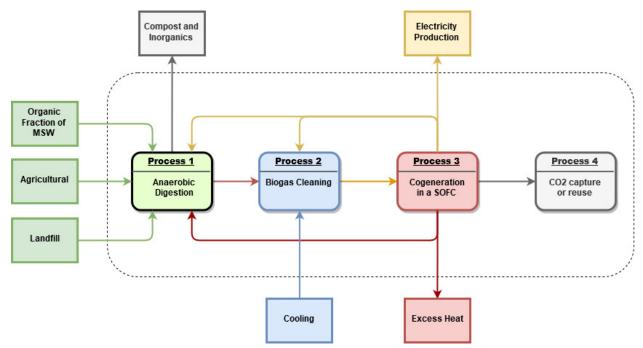


Figure 1: Overview of the Waste2Watts process

## 2 Biogas production

#### 2.1 Anaerobic Digestion

Anaerobic Digestion (AD) is a mature technology, with installation costs for the AD only at a 1  $MW_{el}$  plant, on the order of 1'000 EUR/ $kW_{th}$  or 5'000 EUR/ $m^3$ /h of biogas production capacity and the AD plus CHP system reach 30% to 36% electric efficiency [17], [12]. AD is expected to produce  $300m^3$  of biogas per tonne of biomass [14], this can vary depending on the biomass and the process. As of 2011, Germany was equipped with over 7'000 AD facilities totaling an installed capacity of  $2.4GW_{el}$  [12]. This system is economically viable at small scales, i.e.  $100 \ kW_{el}$  and below [8]. To export the biogas, it has to be upgraded, (filtering and removal of most CO2) the capital cost of an upgrading unit is prohibitively expensive at smaller scales [14]. Therefore most small scale AD systems use their biogas in a CHP unit, usually an internal combustion engine (ICE), to generate heat and electricity. The AD unit consumes about 15% of the energy produced [14].

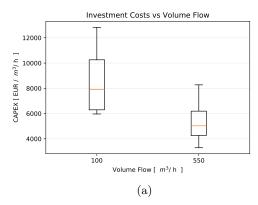
#### 2.1.1 Cost of Anaerobic Digestion

The specific investment cost (CAPEX) in EUR/ 'unit-of-capacity' and the specific annual operating costs (OPEX) in EUR/ 'unit-of-capacity' /year tend to decrease with scale for any chemichal process. The size of an AD process and its 'unit-of-capacity' may be expressed in  $m^3/h$  of biogas produced, alternatively the energy content of that biogas flow in  $kW_{th}$  can be used to take the quality of the produced gas into account. Publications mainly report the total investment cost of the AD and CHP system as an electric power plant without separating the cost of AD. The 'unit-of-capacity' of an entier power plant is always expressed in  $kW_{el}$ . The fraction of the CAPEX dedicated to the CHP is taken as 33%, as reported for the case of a 65  $kW_{el}$  AD project [8] (another 190  $kW_{el}$  installation reported the cost of the CHP unit as 36% of the total cost [8]). The same fraction is applied to the OPEX.

In order to compare the CAPEX of different installations with varying scale, the specific CAPEX is defined in EUR per  $kW_{th}$  as the ratio of the capital investment over the biogas production capacity of the AD. Since most publications report on the overall system, the nominal power is in  $kW_{el}$  of electricity generated by the CHP engine. The electric efficiency of a modern small scale ICE CHP is taken as  $\eta_{el,CHP} = 35\%$  as reported in [20]. The energy output of the AD is then described by equation 1. Where  $\dot{W}_{el,CHP}$  is the nominal power of the CHP engine and  $\dot{Q}_{biogas}$  is the nominal power of the AD.

$$\dot{Q}_{biogas} = \frac{\dot{W}_{el,CHP}}{\eta_{el,CHP}}$$
 [kW<sub>th</sub>] (1)

According to a study by the International Energy Agency (IEA) [14], AD facilities producing 100 to 500  $m^3$  of biogas per hour have a total investment cost of 710 to 470 EUR/ $kW_{th}$ , respectively. With a OPEX ranging from 12 to 41 EUR/ $kW_{th}/year$ . Currency transformations where made using a 10 year average exchange rate and the Chemical Engineering Plant Cost Index (CEPCI) of early 2019 with that of the reference year. Another study for the UK Department of Energy and Climate Change (DECC) [6] found a much higher range of CAPEX from 1'280 to 2'530 EUR/ $kW_{th}$  for installations with a capacity below 750  $kW_{th}$  (average 450  $kW_{th}$ ) and a corresponding OPEX ranging from 83 to 390 EUR/ $kW_{th}/year$ . Ranges found by other groups are presented in Table 1 and 5. These ranges both in terms of volume and energy are also presented in the form of box-plots in figures 2 and 3.



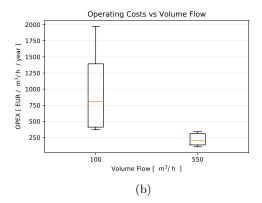
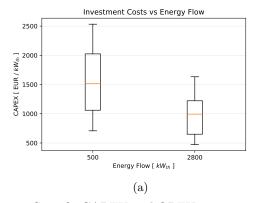


Figure 2: Specific CAPEX and OPEX ranges reported by different groups [14] [6] [17] [12] and groups in two sizes. (a) shows the CAPEX and (b) the OPEX, as a function of biogas production capacity in m<sup>3</sup>.



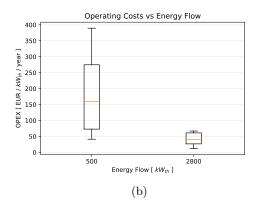
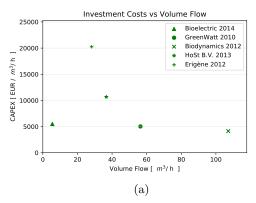


Figure 3: Specific CAPEX and OPEX ranges reported by different groups [14] [6] [17] [12] and groups in two sizes. (a) shows the CAPEX and (b) the OPEX, as a function of biogas production capacity in  $kW_{th}$ .

Table 1: Specific CAPEX and OPEX ranges reported by different groups [14] [6] [17] [12] are presented relative to the AD capacity in  $kW_{th}$ 

Source	CAPEX in EUR/ $kW_{th}$	OPEX in EUR/ $kW_{th}$ /year	SIZE in $kW_{th}$	Region
IEA [14]	470 to 710	12 to 40	500 to 2'500	World
DECC[6]	1'280 to 2'530	80 to 390	< 750	UK
ETRI [17]	650 to 1'630	25 to 70	2'800	$\mathrm{EU}$
IRENA [12]	510 to 1'220	25 to 60	1'000 to 10'000	World

In the context of the W2W project, it is important to look at smaller scale installations, when data is available, as scale affects the cost of most plants. Dobbelaere et al. [8] reported in 2015 on the economic viability of small scale AD projects in Easter Europe. Their findings are reported in figures 4. Since the W2W project requires a biogas production capacity on the order of 50  $kW_{th}$  or 10  $m^3/h$ , the CAPEX cost range would be from 1'000 to 4'000 EUR/ $kW_{th}$  and the OPEX from 50 to 200 EUR/ $kW_{th}$ /year.



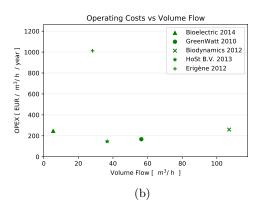
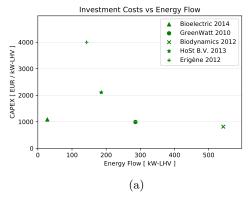


Figure 4: Cost of AD at smaller scales, as reported by [8]. (a) shows the CAPEX and (b) the OPEX, as a function of biogas production capacity in m<sup>3</sup>. Each shape corresponding to a specific case study, named after the AD plant manufacturer and the year of construction.



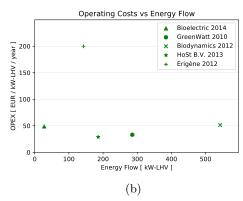


Figure 5: Cost of AD at smaller scales, as reported by [8]. (a) shows the CAPEX and (b) the OPEX, as a function of biogas production capacity in  $kW_{th}$ . Each shape corresponding to a specific case study, named after the AD plant manufacturer and the year of construction.

#### 2.2 Gasification

Biomass Gasification (BG) has a few advantages over AD. In the context of W2W, the gasifier operates at similar temperatures as the SOFC, reducing the exergy losses when heating the 60°C AD with waste heat at 800°C. Gasification may process woody biomass, which has a higher energy concentration than wet biomass, and is otherwise composted or turner into wood pellets and combusted. Large scale coal gasification is a mature technology, however, woody biomass is never found in concentrations as high as coal and BG plants have to be scaled down. BG is a less widespread technology than AD. As of 2011, only 50 wood gasification plants where built and 25 planned in Germany [7]. The net electric efficiency of a BG power plant is expected to range from 31% [12] to 35% for Integrated Gasification Combined Cycle (IGCC) [17]. The capital cost of a Biomass Gasification plant vary depending on the technology used. The investment cost vary from 1'200 EUR/ $kW_{th}$ , or 6'000 EUR/ $m^3$ , up to 4'000 EUR/ $kW_{th}$ , or 20'000 EUR/ $m^3$ , for a  $MW_{el}$  scale system.

#### 2.2.1 Cost of Gasification

M. Kraussler et al, investigated the cost of BioSNG production, based on dual fluidized bed steam gasification of wood chips. They assumed the bio-SNG gas to be composed of about 40% H2, 25% CO, 20% CO2 and 10% CH4 [13]. Their results for the OPEX and CAPEX of bio-SNG production, including cleaning and upgrading are show in figure 6.

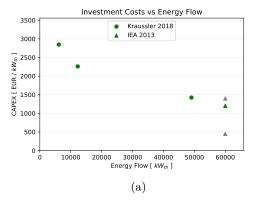
The IEA reported in 2013 that a 420  $MW_{th}$  wood gasification plant will cost between 3'400 and 4'000 EUR/ $kW_{th}$  and produce 1'900  $m^3$  of Bio-SNG per dry tonne of wood chip [14]. This investment cost includes gas

cleaning and upgrading.

In the Energy Technology Reference Indicator projections (ETRI 2014) the investment cost necessary for a 60  $MW_{th}$  BG plant is estimated to be between 450 and 1'400 EUR/ $kW_{th}$ , with a median at 1'200, without gas cleaning equipment [17].

Another group studying the option of BG in the context of developing countries, found that BG is viable at lower scales (10 to 100  $kW_{el}$ ) [7], and that the investment cost vary depending on the region of the project, from 150 EUR/ $kW_{el}$  to EUR  $3,000/kW_{el}$ .

Small scale BG installations are available online at 2'500 EUR/ $kW_{el}$  [5] for a 50  $kW_{el}$  plant. With a specified life of only 4 years, 5 replacements would be necessary to sustain the production for 20 years, pushing the investment cost to 12'500 EUR/ $kW_{el}$ .



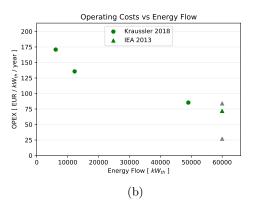


Figure 6: Cost of wood chip gasification as a function of plant capacity in  $kW_{th}$  of bio-SNG (LHV based), based on the results of [13] and [14]. (a) shows the CAPEX and (b) the OPEX which was assumed to be 0.06% of the CAPEX in [13]. The grey triangles show the extreme values from IEA and green is the median value.

## 3 Biogas contaminants and cleaning

#### 3.1 Origin and variation of biogas contaminants

The production of biogas, although widespread with more than 17'400 plants in Europe [21], most of which are based on Anaerobic Digestion (AD), is still an active research field. Variations in biogas volume flow and quality is part of most AD plants. The same is true for contaminants present in the biogas. As highlighted in Figure 7, the concentration of H<sub>2</sub>S varies with time on an hourly basis from 100 to 1'850 ppm. The concentration and presence of each contaminant depends on the biomass used and the production cite [1]. Since it also varies in time, quantifying contaminants requires multiple samplings over days or months which is costly, especially when trying to detect trace elements.

The Paul Scherrer Institute (PSI) is part of the W2W project and contributed by measuring the concentration of contaminants in biogas derived from different feed stock at different plants. Their results for the main chemical species are summarized in Table 2. Results from other research groups for WWTP derived biogas are also presented. H<sub>2</sub>S is the main contaminant, ranging from concentrations of 4ppm to 6500ppm. Other sulfure compounds such as Thiols, COS, CS<sub>2</sub> and DMS are often present at ppb levels. Then Siloxanes, mainly D4, D5 and L3 are present at ppb levels or more depending on the biomass used. When measuring Siloxane concentration of WWTP from different countries, researches found values of total Si ranging from 0.8 to 136.4 mg Si/m³ [15]. Other Hydrocarbons (HC) such as Aromatic and cyclic HC and light hydrocarbons other than methane range from 0.1 to 1600 mg/m³. Halogens, mainly HCl, appear mostly in landfill biogas. In general, agriculture derived biogas have the lowest concentration of contaminants and landfill biogas the highest. The biogas is also saturated with water [4] when leaving the digester.

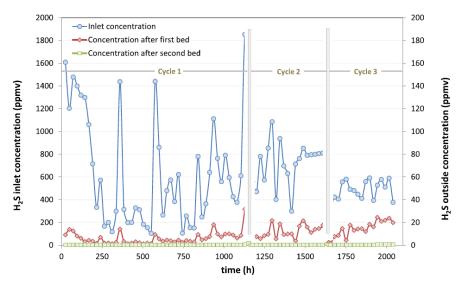


Figure 7: Online measurement of the  $H_2S$  concentration at the outlet of a WWTP in blue and at the outlet of the  $H_2S$  scrubber in red, as measured by [4].

**Table 2:** Summary of the main contaminants in biogas from different sources as reported by W2W Work Group 1 from PSI [1] and other sources [16] [9] [22] [4]. Siloxanes are manly of type D4 and D5. Other HC include cyclic Hydrocarbons (HC) and aromatic HC.

Spieces	$\mathbf{Unit}$	Agric	ulture	OFI	MSW	WV	VTP	Lan	dfill
		average	range	average	range	average	range	average	range
Methane	%  v/v	57	50 - 70	58	45 - 70	64	55 - 65	53	40 - 70
$H_2S$	$\mathrm{ppm}_v$	750	4 - 6'500	700	10 - 3'000	640	10 - 1'852	150	20 - 5'000
Thiols	$ppm_v$	3	0.5 - 10	2	0.3 - 6	0.6	0.2 - 1	3.5	0 - 8
Siloxanes	$mg/m^3$	< 0.02	-	1	0 - 4	14.4	9 - 42	3	0.1 - 11
Other HC	$mg/m^3$	1.1	0.1 - 2	260	50 - 600	36.5	29 - 44	500	20 - 1'600
Halogens	$\mathrm{mg/m^3}$	-	-	-	-	-	-	85	2 - 300

#### 3.2 SOFC degradation from contamination

In their 2017 review, A. Lanzini et al. [15] reported on SOFC deactivation due to contaminants. They identified the Ni-anode, as the most sensitive par of the fuel cell stack and reported on the following contaminants.

- Sulfur (mainly H<sub>2</sub>S, but also other sulfur compounds) reduce the performance of the SOFC, even at ppb levels. Low level sulfur was reported to be reversible. Irreversible degradation was observed for a constant 2ppm sulfur poisoning for 500h.
- Siloxanes causes silicium deposition and catalyst deactivation, by decomposing at high temperatures. At 3ppm large performance drops over short periods of time were observed. Even at 69ppb, a group measured a degradation of 5% per 1'000h. The authors conclude that siloxanes have to be completely removed to protect the SOFC.
- Light hydrocarbons other than methane can cause carbon deposition on the Ni reforming catalyst. Rapid catalyst deactivate (Ni/AL<sub>2</sub>O<sub>3</sub>) was observed when feeding C2 and C3 hydrocarbons at 800ppm for 25 hours and stable activity was observed at below 200pmm.
- Halogens, (HCl, CH<sub>3</sub>Cl and Cl<sub>2</sub>) where reported to cause degradation by some research teams. One group measured no degradation for a one hour exposure to 100ppm HCl, but 1.6% increase of Area Specific Resistance, over a 200h test run at the same concentration. Degradation is reported to be more severe for CH<sub>3</sub>Cl and Cl<sub>2</sub> compared to HCl. But no consistent trend appears in the literature for SOFC poisoning by Halogens.

To prevent SOFC degradation over long periods of operation, the priority for many research teams [4] [10] [25] is the removal of sulfur compounds down to ppb levels and the complete removal of siloxanes. Small amounts of sulfur and carbon deposition over a year of continuous operation may be acceptable, since the performance may be recovered by regeneration during a plant shut down. But siloxane poisoning will accumulate over time, limiting the lifetime of the SOFC. Large concentrations of any harmful contaminant is not reversible and must be avoided.

### 3.3 Available gas cleaning methods

Many WWTP use in-situ sulfur removal methods through micro aeration of the digester to promote bacterial consumption of sulfur compounds. The addition of iron salts in the sludge of the digester is also used to precipitate sulfur compounds. In-situ methods are usually effective and explain why many WWTP produce biogas with low levels of sulfur (10ppm H<sub>2</sub>S). Okoro et al. [18] identified precipitation with iron salts and micro-aeration as the most cost effective methods of removing sulfur from digester biogas. A 2017 review by Mehr et al. [16] also identified micro-aeration as the best option for sulfur removal. In-situ methods should be the priority for sulfur removal, since adsorption methods will result in hazardous waste and a high disposal costs.

For a biogas containing high concentrations of  $H_2S$  (on the order of 100pmm), a scrubbing step is necessary. Iron-based adsorption filters present a viable option for  $H_2S$  scrubbing [4]. These can be regenerated a limited number of times (3 in the case of [4]), which doubled the filter's life and reduce toxic waste handling. These filters operate with a humid gas.

For siloxane removal, an polishing step is necessary. Activated carbon (AC) adsorption filter was the option of choice for some research groups [4] [10]. At this step, it is important that the H<sub>2</sub>S is low (on the order of 10ppm) because H<sub>2</sub>S compete for adsorption with siloxane on AC filters. Water also competes with siloxane adsorption, therefor the gas has to be completely dried before the polishing step, and since reducing temperature can improve adsorption, this should be done with a chiller. Siloxane competes with other compounds for adsorption (and among each siloxane compound) this can cause 'roll-up' where weakly absorbed compounds such as siloxane L2 are released in high concentration when replaced by other compounds as the filter fills up. The adsorption capacity of siloxanes falls when inlet concentrations are low, requiring an oversized adsorption filter.

If other sulfur compounds such as DMS are present in notable concentrations (order of 1 mg/m $^3$ ) a Hydro Desulfurization (HDS) step may be necessary to turn these compounds into H $_2$ S, before they can be captured by the polisher. HDS requires heat and hydrogen, that may be generated by the SOFC, although pre-heating and an initial amount of hydrogen is required for start-up.

#### 3.4 System integration

The main energy consumers of the cleaning unit are the chiller and the blower. If the SOFC is equipped with a heat driven micro-turbine for re-circulation, this micro-turbine may also drive the compressor of the chiller and the fan of the blower. Increasing the overall efficiency.

Many researchers such as [10] include a gas holder in their system design. Since both the biogas flow rate vary and the contaminant concentration vary as seen in Figure 7, a gas holder can help reduce this variation. Holding the biogas reduce the need of flaring during production peaks and facilitate the control of the SOFC, keeping it at nominal load longer. Figure 7 also shows that bursts of concentration in sulfur causes some sulfur to escape the iron-based adsorption beds. Holding the gas on a daily basis can reduce those peaks and therefor reduce the risk of break-through.

#### 3.5 Biomass Gasification

The composition of bio-SNG is different from biogas, as shown in figure 8. Contaminants such as Sulfur compounds and HCl are also present, along with solid particles tars and alkain metals. The gas exit the gasifier at high temperature, and to preserve a high efficiency, it should be cleaned at high temperature, before entering the SOFC.

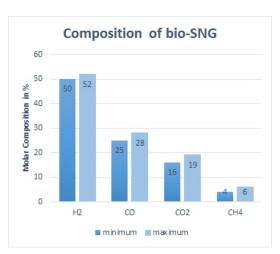


Figure 8: Typical composition for bio-SNG produced by gasification as reported in [3].

In their extensive review, Aravind et al [2], identified the main contaminants present in biomass gasification produced gas and their effect on SOFC performance. They also discussed ways to deal with these contaminants at high temperature, that are summarized below.

- Solid particles from 10 to 0.2  $\mu$ m are present in varying amounts (1 to 10 % weight) depending on mineral and inorganic elements present in the biomass and the operation of the gasifier. They should be removed down to a few ppmw since they might accumulate in the pores of the SOFC anode due to their size and reduce its performance over time. Larger particles are easily removed using cyclones but for sub-micron particles ceramic filters are the best option.
- Tars concentration vary according to the gasification process. Updraft gasifiers have concentrations of 20 to  $100 \text{ g/m}^3$  whereas downdraft versions have only 0.01 to  $1.5 \text{ g/m}^3$  of tars in their produced gas. Later research [19] concluded that tars are detrimental to the anode and cause carbon deposition. A combination of nickel-based and dolomite-based catalysts is recommended by [2] to remove tars. Tars can also be dealt with by adding oxygen and operating at higher temperatures. Tar elimination should be done at high temperature to avoid tar condensation.
- Sulfur in the form of H2S with some COS are also present in bio-SNG. Levels of 20 to 200ppm where reported. To remove the sulfur, zinc-based sorbents where considered as the best option for their high temperature stability, removing sulfur to 4ppmv when operated at 600°C. Recent studies indicate that this concentration is too high, and the temperature may have to be further decreased to reduce the sulfur concentration to acceptable levels.
- HCl is present in quantities varying from 90 to 200ppm for wood derived gas. As for sulfur, it is difficult to remove HCl at elevated temperature, Na<sup>2</sup>CO<sup>3</sup> was selected as the best candidate, due to its good performance at 600°C, removing HCl to 1ppm and low salt emission (1.4ppm of NaCl).
- Alkalin metal compounds are present on the level of a few ppm. They cause corrosion and are typically limited to 0.1ppm for gas turbines. They should be removed to 1ppm or below to prevent fuel cell degradation. To this effect the authors recommend activated alumina, an alkali getter that can operate at 850°C.

Based on their findings, Aravind et al [2], proposed the gas cleaning system for bio-SNG shown in figure 9. A 600°C bottleneck is observed, due to the two sorption filters for which the contaminant capture rate is higher at lower temperatures. More steps are required to clean bio-SNG than biogas, since water condensation is not a problem at high temperatures, there is no need for drying. For bio-SNG, 4 to 5 steps are required, even if tar concentration is low due to a downdraft design, against 2 to 3 steps for biogas.

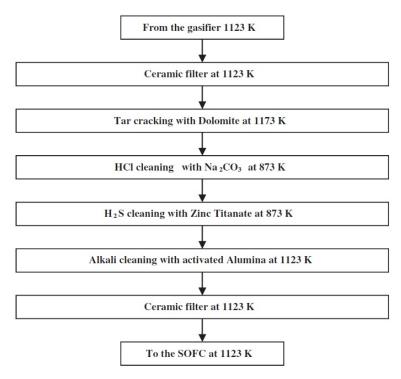


Figure 9: Biomass Gasification derived gas cleaning system proposed by [2].

## 4 Cost of biogas cleaning

#### 4.1 Literature review on biogas cleaning costs and performance

Four papers containing key engineering and economic parameters on biogas deep cleaning for SOFC applications are reviewed. Their methods and scenarios are evaluated and graded from closest to furthest relative to the W2W project. Parameters for specific cost and performance are presented as functions of scale. Results are discusses and summarized. A notebook with explicit data handling of each parameter is presented in the annex.

In 2013, Trendewicz et al. [25] conducted a techno-economic analysis of the utilization of biogas from a US based Waste Water Treatment Plant (WWTP) in a SOFC at large scale. They modeled the process in Aspen Plus for installations with an electric capacity ranging from 330kW to 6.14MW net output. The biogas cleaning process was modeled based on Molten Carbonate Fuel Cell (MCFC) applications for biogas. It included water removal, H2S and siloxanes removal down to a level of  $2.82 \text{ mg/m}^3$  each (about 100ppb for H2S and 300ppb for siloxanes). The authors identified a pressure drop of 10kPa through the gas cleaning section. The reported investment cost and maintenance cost were 920 EUR/ $kW_{el}$  and 100 EUR/ $kW_{el}$ /year respectively for the  $330kW_{el}$  situation. They reported a total of 13 facilities and 7.9MW installed capacity in the US, of WWTP with cogeneration, using fuel cells. Fluctuations on biogas production was collected from a WWTP and a hourly fluctuation of +20% to -20% was observed over a year, as well as fluctuations of +10% to -10% for monthly biogas production. The authors compared the economics of fuel cell applications to for WWTP biogas usage to other technologies and found that although fuel cells would produce much less emissions and have a higher electric efficiency, they where not yet competitive from an economic point of view, even at large scale.

N. de Arespacochaga et al. [4] published in 2014 a detailed study on the process of biogas cleaning for SOFC applications. They designed and installed a small scale cleaning unit on the line of an existing WWTP anaerobic digester, corresponding to  $30kW_{el}$  net electric output. They measured the performance and resilience of their biogas cleaning design. The set-up was (i) two regenerable iron-based adsorbent unit to remove H2S, in lead lag positions with on line sulfur measures (ii) a biogas drying unit to remove moisture and (iii) an activated carbon unit to remove the remaining trace components (siloxanes, linear and aromatic hydrocarbons). H2S was removed from a inlet concentration of 400ppm (average) to 1'800ppm (maximum) to and outlet concentration below 0.5ppm (average 0.1ppm). Siloxanes, Linear HC and aromatic HC where removed to trace levels of 1.4 to 0 ppb. The test was conducted for 3'600 hours. The iron-based adsorbent was regenerated 3 times with hot air drying and its performance decreased. Economic comparisons between different scenarios highlighted the benefit of biogas pre-treatment using a bio-trickling filter. Investing on the pre-treatment had a higher investment cost of 2'000 EUR/ $kW_{el}$ , a lower operating cost of 50 EUR/ $kW_{el}$ /year, and lower total cost compared the the scenarios with no pre-treatment.

In 2014, Tjaden et al. [24] modeled a  $25kW_{el}$  electric SOFC-driven cogeneration plant, supplied by biogas from organic and agricultural waste. They reported on a pressure drop of 15 kPa in the cleaning section and did not report on economic parameters.

In 2018, S. Giarola et al. [10] performed a techno-economic evaluation for retrofitting a sub- $MW_{el}$  WWTP, to accommodate an AD, biogas holder, biogas cleaning system in two parts to co-generate heat and power using a SOFC, in Europe. The design of the cleaning system contains a blower, a dryer, sulfur and silicium removal with inline sampling, followed by a second dryer and a polisher. Both the scrubbing and polishing filters are in a lead-lag setup. The cleaning layout is based on the EU-project 'DEMOSOFC' as presented in Figure 10. The nominal SOFC capacity was  $175kW_{el}$ . Three scenarios are presented for the costing of the cleanup system: (A) current manufacturing volume of 100 units. (B) short-term manufacturing volume of 1'000 units. (C) Target manufacturing volume of 10'000. (unit refers to a biogas cleaning unit for SOFC applications) The reported costs for the current scenario were 917 EUR/ $kW_{el}$  for the investment cost and 76 EUR/ $kW_{el}$ /year for the operating cost.

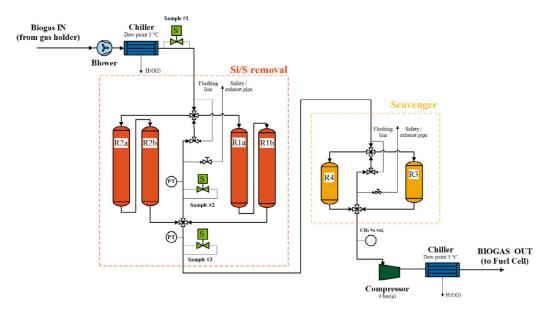


Figure 10: The DEMOSOFC cleaning unit as presented in [10], for a  $25kW_{el}$  electric SOFC, where the biogas is produced by AD and stored in a gas holder, before entering this section.

#### 4.2 Methodology

The gas cleaning section cost depends on the nominal gas flow rate (i.e. capacity) and contaminant concentration it is designed for. To compare the economic results of the different papers reviewed, the unit for the capacity of the cleaning section is expressed in therms of biogas flow rate in m<sup>3</sup>/h. The specific investment cost (CAPEX) in EUR/m<sup>3</sup>/h and the specific annual operating costs (OPEX) in EUR/m<sup>3</sup>/h/year are initially assumed to be a function of capacity. The concentration and nature of contaminants is not taken into account in the cost calculation. Currency transformations follow equation 2, the Chemical Engineering Plant Cost Index (CEPCI) for early 2019 is used as the current value, and unless specified otherwise, the year of publication of the study is used as a the reference year. Currency exchange rates are calculated using a 10-year average exchange rate (2009 - 2019). The CAPEX and OPEX can be combined using formula 3 in order to compare the total annual costs or TOTEX of each scenario in each paper. Where  $C_{inv}$  is the specific investment cost in EUR/m<sup>3</sup> and  $\tau$  an annualization factor. With a 2.5% interest rate i and a 20 years life span n. i and n are chosen as per Giarola et al. [10]. In an effort of transparency and documentation, all calculations are attached as supplement to this work, in the form of jupyter notebooks.

$$c_p = c_p^{ref} \cdot \frac{I_{CEPCI}^{current}}{I_{CEPCI}^{ref}}$$

$$TOTEX = OPEX + \tau \cdot C_{inv}$$

$$[EUR/m^3/year]$$
(2)

$$TOTEX = OPEX + \tau \cdot C_{inv}$$
 [ $EUR/m^3/year$ ] (3)

$$\tau = \frac{i(1+i)^n}{(1+i)^n - 1}$$
 [-/year]

Are spacechaga et al. [4] presented the following scaling law (5) for the investment cost as a function the volumetric flow rate of biogas, with a scaling factor  $\gamma = 0.6$ . Where  $\dot{V}$  and  $\dot{V}_{ref}$  are the current and reference biogas flow rates in  $m^3/h$ .

$$C_{inv}(\dot{V}) = C_{inv}(\dot{V}_{ref}) \cdot \left(\frac{\dot{V}}{\dot{V}_{ref}}\right)^{\gamma}$$
 [EUR/m<sup>3</sup>]

Concerning operating costs, if the replacement of adsorbent filters is assumed to be included in the investment cost, then the operating cost is driven by the energy requirements of the dryer and the blower. Which is directly proportional to the biogas flow rate, i.e.  $\gamma = 1$ . Therefore equation 6 can be used, where  $C_{op}$  is the annual operating cost or OPEX.

$$C_{inv}(\dot{V}) = C_{inv}(\dot{V}_{ref}) \cdot \left(\frac{\dot{V}}{\dot{V}_{ref}}\right)$$
 [EUR/m³/year] (6)

#### 4.3 Results

In Figure 11, the investment cost scaling law is compared to the reported investment cost of two other studies. The scaling law with a gamma of 0.6 as in [4] is used to find the theoretical value for the cleaning section of the W2W project, shown as a yellow star and compared with the results of Work Group 2 [1] as a blue square. The data points from the two other papers are within the 30% range, shown in blue dotted lines, a common range for rough economic estimations. However the resulting CAPEX is more than twice that of Work Group 2.

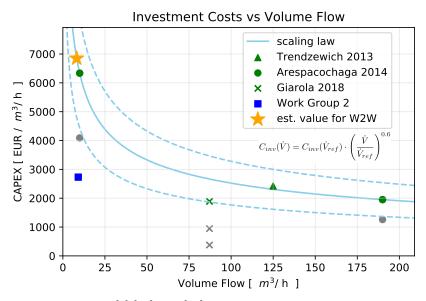


Figure 11: The CAPEX reported in [4] [10] and [25], for a biogas deep cleaning unit feeding an SOFC, are plotted as a function of the biogas flow rate in normal cubic meters per hour. The most likely scenario of each paper is in green, the others in grey. The shapes represent each paper, with circle as the reference. The scaling law proposed by [4], is plotted as a solid blue line, with a 30% margin of error in dashed blue lines. The yellow star is the theoretical specific investment cost of the W2W project and it is calculated using the scaling law for a SOFC capacity of  $25kW_{el}$ . The result is compared with the values of Work Group 2 [1] as a blue square.

The specific operating cost is assumed to remain constant with scale, as shown in Figure 12. Since the OPEX of both likely scenarios (in green) from the reference paper [4] where more than 30% below the other studies, it was likely underestimated. To correct this, the average OPEX of all three papers is used. The new OPEX includes all likely scenarios within the 30% margin, as shown on Figure 12. However, this OPEX is less than half of what Work Group 2 found, as shown by the blue square.

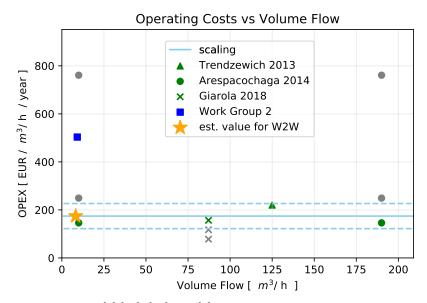


Figure 12: The OPEX reported in [4] [10], [25] and [1] for a biogas deep cleaning unit feeding an SOFC, are plotted as a function of the biogas flow rate in normal cubic meters per hour. For details on shapes and colors of data points, see Figure 11

The TOTEX can be calculated in two ways. Either by calculating and plotting directly the TOTEX of each study, then correcting the scaling law proposed by Arespacochaga et al. to account for the contribution of the constant OPEX. Then this scaling law is used to calculate the value at the size of the W2W project. The results for this method is show in Figure 13 and shows poor correlation with the study by Trendzewich et al. Alternatively the scaling law for the TOTEX can be calculated as a sum of the OPEX and CAPEX functions found previously using equation 3. This results in a similar scaling law with a higher reference TOTEX as seen in Figure 14, and does give a slightly better correlation with results from other papers.

To be able to compare these results, with previous results for AD and gasification costs, costs are converted to per  $kW_{th}$  of the biogas Lower Heat Value (LHV). The TOTEX values in terms of energy is presented in Figure 15. All results are summarized in table 3 and 4 in terms of volume flow and energy flow respectively. Comparing the combined results from [4] [10], [25] and the values of Work Group 2, the TOTEX match, but CAPEX and OPEX diverge. It may be that Work Group 2 considered the cost of purchasing all filters as a replacement cost, attributing this to OPEX, while other groups considered the cost of all filters for a lifetime operation as part of the CAPEX. In their detailed calculations, the replacement of adsorbent cost is almost 80% of the OPEX. This would explain why the CAPEX of Work Group 2 is much lower at small scale, and the OPEX much higher, that other sources.

**Table 3:** Summary of the best values of specific costs for a biogas deep cleaning unit feeding an SOFC from each source, in terms of volume flow [4] [10], [25] and [1].

Volume Flow	Capacity in m <sup>3</sup> /h	CAPEX in EUR/ m <sup>3</sup> /h	OPEX in EUR/ $m^3/h$ /year
Fit result	8	6'850	170
Work Group 2	9	2'730	500
Arespacochaga 2014	10	6'340	150
Trendzewich 2013	125	2'420	220
Giarola 2018	87	1'890	160

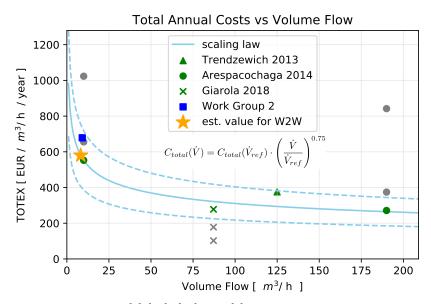


Figure 13: The TOTEX reported in [4] [10], [25] and [1] for a biogas deep cleaning unit feeding an SOFC, are plotted as a function of the biogas flow. The scaling law proposed by Arespacochaga et al. is corrected to fit the TOTEX values of the reference paper, a scaling factor of 0.75 is obtained. It is plotted as a solid blue line, with a 30% margin of error in dashed blue lines. The yellow star is the theoretical total cost of the W2W project and it is calculated using the scaling law for a SOFC capacity of 25 kW. For details on shapes and colors of data points, see Figure 11.

**Table 4:** Summary of the best values of specific costs for a biogas deep cleaning unit feeding an SOFC from each source, in terms of energy flow [4] [10], [25] and [1].

Energy Flow	Capacity in $kW_{th}$	CAPEX in EUR/ $kW_{th}$	OPEX in EUR/ $kW_{th}$ /year
Fit result	42	1'350	40
Work Group 2	46	540	100
Arespacochaga 2014	51	1'240	30
Trendzewich 2013	640	470	40
Giarola 2018	292	570	50

#### 4.4 Performance

The main performance parameter for a filtration unit is the pressure drop  $\Delta P$  across the entire unit. This arise mainly from the adsorption filters but other parts such as pipe connections, valves and the chiller contribute. Trendewicz et al. [25] reported a pressure drop of 10kPa across the cleaning unit and 5kPa across piping of the entire plant, for a large scale installation. Tjaden et al. [24] reported a pressure loss of 15kPa across the cleaning section for a 25kW system. Finally this work's reference paper [4] did not report a pressure drop but instead a yearly electricity consumption for parts of the cleaning section. Assuming a 90% plant capacity factor and that a single blower for the whole cleaning section consumes all of this electricity, it would require 0.83kW of power. It is possible to lookup available data sheets from blower manufacturers to deduce the pressure drop, such as in Figure 16. Following the purple line (50hz current) the over-pressure for this power consumption is approximately 15kPa. Power to pressure graphs vary from a blower to another, this model was the smallest available that could consume this power (up to 1kW) and deliver the necessary gas flow rate (10m<sup>3</sup>/h).

Unfortunately these tables only exist for air, while the blower has to compress biogas. To get a better approximation, the pressure drop can be calculated as a function of the work done and the, gas properties and inlet conditions. The temperature rise across the compressor, is dependant on equation 7, where the mass flow rate is known from 8. The three biogas properties  $\rho$ ,  $C_p$  and  $C_v$  are all calculated by summing the property of

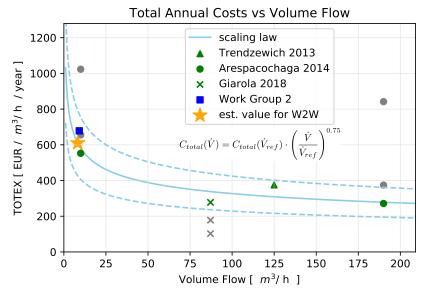


Figure 14: The TOTEX reported in [4] [10], [25] and [1] for a biogas deep cleaning unit feeding an SOFC, are plotted as a function of the biogas flow. The scaling law proposed by Arespacochaga et al. for CAPEX and the average OPEX are combined to get a TOTEX scaling law. It is plotted as a solid blue line, with a 30% margin of error in dashed blue lines. For details on shapes and colors of data points, see Figure 11.

each component multiplied by its volumetric fraction  $f_c$ , as in equation 9. For the isentropic compression of ideal gasses, we can write equation 10. Rewriting this equation to explicitly get the pressure drop  $\Delta P$  yields equation 12. The work  $\dot{W}$  provided to the compressor, the gas flow rate  $\dot{V}$  and the gas composition are given in the reference paper [4]. The inlet temperature is assumed to be 60° (thermophillic AD) and at atmospheric pressure. The gas properties  $\rho$ ,  $C_p$  and  $C_v$  all depend strongly on temperature, but at first approximation,  $\Delta T$  is low enough (10°) to be neglected. The resulting pressure drop is close to 15kPa, but the result will vary depending on the chosen inlet temperature.

$$\dot{W} = \dot{m} \cdot C_p \cdot \Delta T \tag{7}$$

$$\dot{m} = \dot{V} \cdot \rho \tag{8}$$

$$\rho_{biogas} = \sum_{c=1}^{\#component} f_c \cdot \rho_c \tag{9}$$

$$\frac{P_1}{P_2} = \left(\frac{T_1}{T_2}\right)^{\frac{\gamma}{\gamma - 1}} \tag{10}$$

$$\gamma = \frac{C_p}{C_v} \tag{11}$$

$$\Delta P = P_1 \cdot \left[ \left( \frac{T_1 + \Delta T}{T_1} \right)^{\frac{\gamma}{\gamma - 1}} - 1 \right]$$
(12)

The energy consumed in the cleaning section to compensate for the pressure drop through the adsorption filters is low relative to the nominal power of the entire process. In the reference case [4] the electric consumption to counteract the pressure drop is 1.5% of the theoretical electric output and the consumption for the dryer is another 1.5% of that same output. In total, about 3% of the energy is lost to the biogas cleaning and drying section.

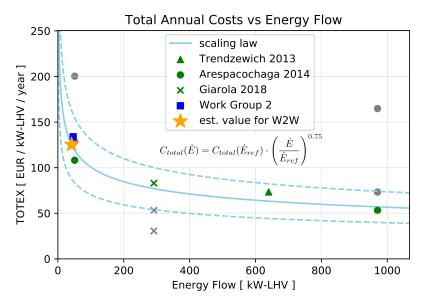


Figure 15: The TOTEX reported in [4] [10], [25] and [1] for a biogas deep cleaning unit feeding an SOFC, are plotted as a function of the energy value of the biogas flow. The scaling law proposed by Arespacochaga et al. for CAPEX and the average OPEX are combined to get a TOTEX scaling law. For details on shapes and colors of data points, see Figure 14.

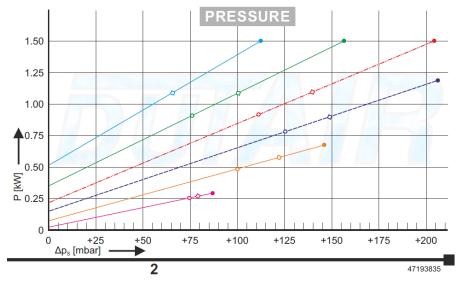


Figure 16: Pressure vs Power graph for the dual channel blower DB313 model from DUTAIR [23]. The purple dashed line is valid for  $50 \mathrm{Hz}$  (EU based) current. For a power of  $0.83 \mathrm{kW}$ , this corresponds to a over-pressure of a little over  $15~\mathrm{kPa}$ 

## 5 Conclusion

The CAPEX of AD is 500 to 2'500 EUR/ $kW_{th}$  with a median at 1'000 EUR/ $kW_{th}$ , the OPEX is 20 to 400 EUR/ $kW_{th}$ , with a median at 50 EUR/ $kW_{th}$ /year for smaller scale systems. Concerning gasification of biomass, one source reported a CAPEX below 1'000 EUR/ $kW_{th}$ . Other sources reported a CAPEX ranging from 1'200 to 4'000 EUR/ $kW_{th}$  with values on the order of 3'000 EUR/ $kW_{th}$  for smaller scale systems. The OPEX ranged from 75 to 175 EUR/ $kW_{th}$ /year. Although studies and reports on the cost of BG are sparse, these results indicate that AD is a cheaper process, something that is expected since the installed capacity of AD plants is much higher than the installed capacity of BG plant.

Concerning the cost of the cleaning unit, for the biogas or bio-SNG produced, it was not possible to compare the costs for AD and BG produced gas, due to the lack of studies on the cost of bio-SNG cleaning for SOFC utilization. The CAPEX of cleaning biogas, was estimated to vary from 500 to 1'300 EUR/ $kW_{th}$  and the OPEX from 30 to 100 EUR/ $kW_{th}$ /year. These values may seem high when compared to the cost of AD, but they are in the same range as the cost of biogas upgrading for medium scale installations (upgrading CAPEX is 500 EUR/ $kW_{th}$  for facilities processing up to 1'000  $m^3/h$  of biogas but for small scale facilities the CAPEX can reach 2'600 EUR/ $kW_{th}$  according to the IEA [14]).

Using today's SOFC in an AD CHP plant, it is possible to almost double the electric efficiency of the prime move (35% for an ICE against 60% for an SOFC), while the cleaning section required has a small impact on performance (approximately 3% electric losses). Half of the losses are due to a pressure drop on the order of 15kPa through the adsorption filters, compensated by a blower and the other half come from the chiller consumption. The main barrier to the adoption of this technology is the high cost of designing a cleaning section, that ensures the longevity of the SOFC. This include costs of preliminary sampling at the biogas production site at hourly frequency over months. The only other option, is to oversize the cleaning section, and change filters early, to prevent risks of breakthrough. A gas holder, may help reduce the variation in the biogas quality and contaminant concentration and the sampling frequency required.

The continued development of standardized biogas cleaning units for SOFC applications and measurement of pollutant concentrations at different biogas sites, as is done in the W2W project, will drive the cost down. Moreover, a key to decreasing the design and installation cost is the development of an affordable and reliable in-line gas analysis system for trace elements. The lead lag configuration with a sulfur and siloxanes detectors in the middle, would ensure that filters are changed when they break through and not before or after, maximizing their life while ensuring no break-through to the SOFC. With such a device it is possible to have two gas holders at the outlet of the AD, with a sulfur detector and a switch, to store biogas with high sulfur levels in a secondary gas holder, meant for flaring and heating of the AD, further reducing the size of the cleaning unit.

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