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New electroscrubbing process for desulfurization

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A B S T R A C T
A new process for removing sulfur from gas using electricity is presented. The process combines electrochemical production of active oxidizing species with an oxidative scrubber for desulfurization. Reactions for the electrochemical cell and the scrubbing tower are presented and a scrubbing set-up with a continuous electrolyte recirculation is validated on synthetic biogas. The test shows that a gas flow of 200 L/h with 1330 ppm H2S can be cleaned down to non-detectable levels of sulfur in a single step, at a current efficiency of 29%. This is a very high removal percentage for a high concentration of H2S. Solid sulfur can be removed from the system after the experiments. The sulfur has traces of NaClO and no other products were detected. A simple model based on estimated gas side mass transfer coefficients were used to estimate a scrubber height of only a few centimeters. The experiments supports the model.

This technology allows for a robust implementation of sulfur free applications in a future green fuel and green chemistry like Power-to-X (PtX) and carbon capture and utilization (CCU). Sulfur compounds are poison for most heterogeneous catalysts, thus for carbon utilization, the carbon gas source has to be desulfurized.

1. Introduction

The renewable energy sector is booming and especially the biogas industry is growing [1]. Each year new biogas plants are constructed worldwide. In Europe alone there were 17,783 biogas plants in 2017 with ≥10,000 in Germany [2]. Compared to solar and wind energy, biogas does not suffer from fluctuating and unstable production rates. This makes biogas even more suitable as a main constituent in a fossil-free society.

Biogas consists roughly of 60% methane (CH4) and 40% carbon dioxide (CO2) with several impurities. The main impurity is hydrogen sulphide (H2S). Normal concentrations of H2S in biogas is above 1000 ppm. However, siloxanes and mercaptanes are also often found to a lesser extent in the biogas [3]. H2S is both flammable, toxic and corrosive [4]. Thus, independently of the downstream application, H2S has to be removed from the biogas.

Traditionally, biogas has been used in a combined heat and power engine for localized energy production. However, within the last decade, biogas upgrading has found its way to the market. Today, state-of-the-art upgrading of biogas refers to extraction of CH4 from biogas [5,6]. This type of CH4 is named biomethane [7] and is a renewable energy resource with many applications. On a commercial level, four different technologies are available for upgrading biogas: amine extraction, membrane separation, water scrubbing and pressure swing adsorption [8]. Future applications of biogas may differ significantly from the current state-of-the-art. Recently, the term “second generation biogas upgrading” has been proposed to distinguish between traditional biogas upgrading technologies and future upgrading technologies and processes [9].

Using biogas as carbon source for biofuels is currently being investigated [10–15]. Central to these applications is the reduction of CO2. Generally, there are three main methods for reducing CO2, i.e. biological, electrochemical and thermochemical. All technologies require removal of H2S from the biogas, with biological conversion tolerating more H2S as compared to thermochemical and electrochemical CO2 reduction reactions.

Thermochemical treatment of biogas remains under development. Several projects are investigating methanation of biogas [14,15]. Solid
oxide fuel cells using biogas are also being investigated as a possible application of biogas [16]. For reduction of CO$_2$ a nickel catalyst is most commonly used. It is well known that H$_2$S is poison to basically all catalysts [17,18], with concentrations as low as 10 ppm having devastating impact on the active catalytic sites. Thus, for future thermochemical upgrading of biogas, cost-efficient, deep and stable desulfurization is mandatory.

Several different desulfurization technologies are applied. Most technologies remove the H$_2$S from the biogas with the distinct exception of iron chloride desulfurization. The iron chloride salts are added to the biomass before entering the biogas reactor. The iron inactivates the H$_2$S and solidifies sulfur as iron sulfide. Biological desulfurization is widely used due to the low operating expenses (OPEX). The process has a rather large capital expenditure (CAPEX). Furthermore, the sulfur-oxidizing bacteria require nutrients and oxygen. Oxygen requires strict monitoring, as it is highly unwanted for upgrading purposes. Both iron chloride and biological desulfurization may lower the H$_2$S concentration from 2000 ppm to 100 ppm. An additional desulfurization polishing system is required in order to reach acceptable H$_2$S levels. Activated carbon can remove H$_2$S down to very low concentrations. A clear drawback of the carbon absorption technologies is a noticeable OPEX and toxic waste production due to limited regeneration possibilities. Activated carbon is too expensive to be used for bulk H$_2$S removal.

Generally, biogas plants are decentralized. This favors the need for desulfurization technologies with local regeneration and without waste generation. Regeneration could be performed in-situ either by air, e.g. biological desulfurization and the Lo-Cat process [19], by heat, e.g. chemical scrubbers or by electricity.

Among the desulfurization technologies used today, only the biological desulfurization technology uses local regeneration. However, the residual oxygen found in the cleaned gas is a large issue for the process [20]. For biogas plants with first generation upgrading facilities, this is a fundamental issue, as the maximum allowed oxygen content can be as low as 100 ppm for biomethane [21]. Another drawback of the biological cleaning process is the slow response time when it comes to fluctuating H$_2$S levels.

Electro-assisted scrubbers have local regeneration and do not produce any waste. Furthermore, they have the advantage that the electrochemical nature of the scrubber allows for fast adjustment adapting to fluctuating H$_2$S concentrations in the biogas. The term electroscrubbers is used to describe the gas cleaning processes where an electrochemical cell is used to support the scrubber. The term was introduced by Govindan et al. in 2012 [22].

Electrochemically assisted scrubbers are widely used [23]. The operation is based on electrochemical regeneration of the scrubbing medium. Usually, the scrubbing medium consists of a suspended metal that may be electrochemically oxidized to a high oxidation state. The metal with the high oxidation state is then used to remove e.g. H$_2$S and methyl mercaptans [24–27]. The application of electrochemistry has received increased interest as it is considered an environmentally friendly process for chemical production.

Currently, most of the electroscrubbing processes are being developed in the laboratory. However, Govindan et al. tested an electroscrubbing process for simultaneous removal of NO and SO$_2$ at semipilot scale [22]. The semipilot tests demonstrated the potential to use the process previously developed in the laboratory [28–32]. The successful upscaling of the process developed in the laboratory indicates that this could also be possible for other electroscrubbing processes.

Applying electro-assisted scrubbers to biogas could be beneficial when included in second generation biogas upgrading technologies. However, a water-based electrolyte would have to take the rather large amount of CO$_2$ in the biogas into account. CO$_2$ will be dissolved into the electrolyte and affect the pH-value. Generally, the pH-value is expected to reach a steady state level below neutral, due to the acidic properties of CO$_2$. Therefore, an electro-assisted scrubbing process operating at moderately low pH values would be preferred for biogas applications.

Electroscrubbers investigated in the literate generally have high removal rates ($\geq$90 %) for low concentration pollutants [33–35]. Removal rates as high as 100% have been found when the concentration of the pollutants is low (15 ppm). This could be an issue for H$_2$S removal from biogas. H$_2$S is generally found in high concentrations (greater than1000 ppm) in biogas. The electroscrubbing process is therefore required to have high removal rates, even at high concentrations if it should be used to remove H$_2$S from biogas.

Biogas has many impurities other than H$_2$S and an electro-assisted desulfurization process may additionally remove these impurities as well [24]. Generally, the desulfurization process is placed just downstream of the biogas reactor, and thus, will be the first downstream process encountering all types of impurities. Therefore, it may be expected that the electrolyte will require periodic replacement, depending on the amount of biogas impurities, the efficiency of the electrolyte regeneration, and the bulk electrolyte filtration process. For an expensive electrolyte, the filtration process may be optimized for complete impurity removal. Clearly, this will increase the operational cost of the system. Ideally, the electrolyte is cheap, sustainably disposable and may thus be replaced periodically.

In this paper we suggest a new and novel process which can be regenerated in-situ and does not generate waste. The aim is to develop a cheap, stable and robust process that selectively removes H$_2$S down to the very low ppm level, while leaving the CO$_2$ in the biogas. One of the main problems that the process should solve is removing H$_2$S at high concentrations with a high removal rate, i.e. remove H$_2$S from more than 1000 ppm down to very low concentrations. For similar processes, this has proved difficult.

The utilization of active chlorine as the oxidizing species is a novelty within the field of electroscrubbers. For the new process, suitable electrochemical and scrubbing reactions are proposed. The electrolyte system is tested on a lab-scale electroscrubber set-up interfaced with a gas mixture mimicking a real biogas.

The process is an electrochemically assisted scrubbing system. The solvent selectively oxidizes the H$_2$S content of the biogas into free sulfur. The process applies continuous solvent recirculation, which is regenerated in an undivided electrochemical cell. In Fig. 1, the mechanism is presented. The electrolyte comprises an aqueous solution and the electrochemical mechanism is (Fig. 1c):

**Anode [36]:**

\[
2\text{Cl}^- = \text{Cl}_2(aq) + 2e^- 
\]

**Cathode:**

\[
2\text{H}^+ + 2e^- = \text{H}_2(g) 
\]

Following the formation, chlorine reacts with water to form hypochlorite (Fig. 1d) [37]:

\[
\text{Cl}_2(aq) + \text{H}_2\text{O} = \text{H}^+ + \text{Cl}^- + \text{HClO}(aq) 
\]

Within the scrubber, the biogas interacts with the liquid. CH$_4$ has been assumed to be inert and is in the experiments considered interchangeable with other similar inert low-soluble gases such as N$_2$ or higher hydrocarbons. The CO$_2$ is expected to saturate the liquid over time due to CO$_2$ dissolution:

\[
\text{CO}_2(g) = \text{CO}_2(aq) 
\]

\[
\text{CO}_2(aq) + \text{H}_2\text{O} = \text{HCO}_3^- + \text{H}^+ 
\]

The hydrogen sulfide is expected to dissolve and dissociate (Fig. 1a) [38]:

\[
\text{H}_2\text{S}(g) = \text{H}_2\text{S}(aq) 
\]

\[
\text{H}_2\text{S}(aq) = \text{HS}^- + \text{H}^+ 
\]

And subsequently being oxidized by hypochlorite (Fig. 1b):
reacts with water to form hypochlorite (Eq. (3)).

\[
\text{HS}_2\text{S} + \text{H}_2\text{O} \rightarrow \text{H}_2\text{SO}_3 + \text{H}_2\text{S}
\]

The dissociated sulfide is oxidized by hypochlorite (Eq. (8)). The elemental sulfur formed is absorbed into the scrubber and is dissolved and dissociated (Eq. (6) and (7)).

\[
\text{H}_2\text{S}_2\text{O}_3 \rightarrow \frac{1}{2}\text{H}_2\text{SO}_4 + \frac{1}{2}\text{H}_2\text{O} + \text{H}_2\text{S}
\]

\[
\text{H}_2\text{S} + \text{H}_2\text{O} \rightarrow \text{H}_2\text{SO}_4
\]

\[
\text{HS}_2\text{O}_3 + \text{H}_2\text{O} \rightarrow \text{H}_2\text{SO}_4 + \text{H}_2\text{S}
\]

2. Scrubber model

A simple model for determination of the scrubber height will be constructed. The aim of the model is to estimate a scaling up of the electroscrubber. The model will be based on the calculation of a scrubber height from the Height of a Transfer Unit (HTU) and the Number of Transfer Units (NTU). The model will be based on the calculation of an electroscrubber. The model will be based on the calculation of a scrubber height from the Height of a Transfer Unit (HTU) and the Number of Transfer Units (NTU).

2.1. Height of scrubber

The scrubber height (h) can be calculated based on the Height of a Transfer Unit (HTU) times the Number of Transfer Units (NTU) [39]:

\[
h = \text{NTU} \times \text{HTU}
\]

When the mass transfer from the gas to the scrubbing liquid is low, the molar gas flow (G_m), the molar liquid flow (L_m) and the overall gas phase mass transfer coefficient (K_g) can be assumed to be constant. For these conditions, both the equilibrium line and operating line, see Fig. 2(a), are straight [39]. The NTU and HTU can be calculated under these assumptions as [39]:

\[
\text{NTU} = \frac{y_i - y_u}{\Delta y_m} \quad (11)
\]

\[
\text{HTU} = \frac{G_m}{AaK_g} \quad (12)
\]

where \(y_i\) is the mole fraction of the pollutant in the gas at the inlet of the bottom of the scrubber (see Fig. 2(a)), \(y_u\) is the mole fraction of the pollutant in the gas at the outlet of the top of the scrubber, \(A\) is the cross sectional area of the scrubber, \(a\) the specific surface area of the packing material and the log mean concentration difference (\(\Delta y_m\)) is given by [39]:

\[
\Delta y_m = \frac{(y_i - y_u^*) - (y_u - y_u^*)}{\ln(y_i - y_u^*) - \ln(y_u - y_u^*)} \quad (13)
\]

where \(y_u^*\) is the mole fraction of the pollutant in the gas that is in equilibrium with the liquid at the bottom (i) or the top of the scrubber (u). Thus, the difference, \(y_i - y_u^*\), corresponds to the driving force of the mass transfer at the bottom of the scrubber and vice versa for \(y_u - y_u^*\).

The values \(A, a, G_m, y_i\) and \(y_u\) are either given (see Fig. 2(b)), e.g. \(G_m, y_i\) and \(y_u\), or part of the design of the scrubber, e.g. \(A\) and \(a\). \(K_g\) is calculated as [40]:

\[
K_g = \frac{1}{\frac{y_i^*}{y_u^*} + \frac{y_u^*}{y_i^*}} \quad (14)
\]

where \(k_{i/g}\) is the mole fraction mass transfer coefficient in the liquid (l) or the gas phase (g), \(E\) is the enhancement factor and \(m\) is the equilibrium constant, given by \(y = mx\) where \(x\) is the mole fraction of the of the pollutant in the solution, see Fig. 2(a). The enhancement factor, \(E\), indicates the improved performance of a reactive solvent when compared with a non-reactive solvent. A reactive solvent has \(E \gg 1\), and this is expected to be the case for electroscrubbers. The enhancement factor is a well developed concept that has been a subject in several text books, including Danckwerts [40] and Astaria [41].

2.2. Estimation of the mole fraction gas side mass transfer coefficient

An approximation of \(k_g\) is obtained by using the method derived in Absorption Towers by Morris and Jackson [42], as presented in Kemiske enhedsoperationer by Kjarboe et al. [39]. Several different methods for calculations of \(k_g\) exist. This includes models far more complicated that what have been used here. Initial calculations and assumptions indicated that the more advanced models were not required to give a fair estimation of the height. The implementation of more complicated models was therefore not applied. According to Morris and Jackson, \(k_g\) can be calculated [39,42]:

\[
k_g = k_{i/g} \frac{\rho_g}{M_g} \quad (15)
\]

where \(M_g\) is the molar mass of the gas, \(\rho_g\) is the density of the gas and \(k_{i/g}\) is the mole fraction mass transfer coefficient in the gas phase. \(k_{i/g}\) can be calculated as:

\[
k_{i/g} = 0.04C_p Re^{-0.25}Sc^{-0.1} \frac{V}{4\text{Re}^{0.5}} \quad (16)
\]

where \(C_p\) is the gas film factor, \(x_{lim}\) is the logarithm of the average fraction of the solvent (B) in the liquid film, \(V\) is the superficial gas flow.
velocity, $Re$ is Reynold’s number and $Sc$ is Schmidt’s number. The superficial gas velocity is given by:

$$v = \frac{G_v}{A} \tag{17}$$

where $G_v$ is the gas volume flow. $Sc$ is given by:

$$Sc = \frac{\eta_v}{D_{cg} \rho_v} \tag{18}$$

where $\eta_v$ is the viscosity of the gas and $D_{cg}$ is the diffusion coefficient of the gas phase. Reynold’s number is given by:

$$Re = \frac{d_b \nu \rho_i}{\eta_i} \tag{19}$$

where, $d_b$ is the hydraulic diameters of the packing material given by:

$$d_b = \frac{4 \epsilon}{a} \tag{20}$$

where $\epsilon$ is the porosity of the packing material.

### 2.3. Calculation of the electroscrubber height

$K_p$ for the laboratory electroscrubber is calculated under several assumptions. It is assumed that $L_{in}, G_m$ and $K_p$ are constant. The oxidation of HS is reported as fast and has a high $K$-value. Therefore, the concentration of the pollutant, i.e. HS, in the liquid may be assumed to be $\sim 0$ [43]. The high reactivity may be assumed to result in a high enhancement factor $E$. The equilibrium between H$_2$S in the gas phase and in the liquid phase for the given oxidative system is expected to result in very low $m$-values. The partial pressure of the H$_2$S in the gas phase is very small for any concentration in the liquid phase. This is equivalent to a very small $m$-value. For a large $E$ and a small $m$, $K_p = k_p$ according to Eq. (14). This is in accordance with Ramm’s rule [39]. $K_p$ is then calculated as $k_p$ using Eq. (15). The physical parameters used for the calculation of $k_p$ are presented in Table 1.

### 3. Materials and methods for the developed electrochemically scrubber system

In Fig. 3, the set-up during the experiments is presented. Details regarding the scrubbing system, electrochemical cell and the gas flow & analysis is found in the sections below. Overall, a gas-mixture of nitrogen (N$_2$) and H$_2$S is mixed with CO$_2$. N$_2$ is used instead of CH$_4$ to avoid flammable and explosive mixtures exiting the scrubbing system in the laboratory. Within the electroscrubber, the CH$_4$ is not an explosive risk as no O$_2$ is present. Both N$_2$ and CH$_4$ are expected to be inert gasses in the process. The mixed gas then enters a home-made scrubbing set-up where it interacts with the electrolyte. Here, oxidative species from the liquid is used to oxidize H$_2$S. The oxidative species is regenerated in the electrochemical cell that is positioned in the bottom of the scrubber. When the liquid is regenerated, it is pumped back into the scrubber. The liquid goes through this cycle in less than 50 s, ensuring a fast response to changes in operating parameters, such as the applied potential.

#### 3.1. Scrubbing system

A scrubbing system was constructed using standardized tubing devices and equipment based on an internal diameter of 110 mm. The tower and liquid container were made from PMMA (Plexiglas) allowing for visual inspection of the liquid flow and electrochemical cell under operation. A water circulation pump with a brushless motor capable of 5.5 m lift was used together with a showerhead which resulted in a liquid flow of 1.26 L/min. evenly distributed within the scrubbing tower. Simple uncoated Lightweight Expanded Clay Aggregate (LECA) balls with a density between 210 and 280 kg/m$^3$ were used as packing material with suitable gas-to-liquid wetting abilities.

#### 3.2. Electrochemical cell

The undivided electrochemical cell was positioned in the bottom of the scrubber unit and consisted of two graphite electrodes from Frederiksen Scientific A/S, with a diameter of 6 mm and a total length of 250 mm. 150 mm of the electrodes were inside the reactor and 100 mm outside for connection purposes. No degradation of the carbon electrodes were observed during the experiments. The voltage and current were applied by a Voltcraft LSP-1403 80 W switching DC power supply. An electrolyte consisting of 1 L of demineralized water mixed with 200 g NaCl was used within the system. The concentration of solvent was chosen as a high amount of NaCl to ensure a high conductivity of the solvent. However, over-saturation of the solvent is also unwanted and the NaCl concentration was therefore kept below the maximum value (360 g NaCl per liter of water).
Table 1

Parameters used for calculation of scrubber height.

| Parameter                        | Symbol | Unit | Value | Note |
|----------------------------------|--------|------|-------|------|
| Molar gas flow                   | \( G_g \) | mol/s | Varies, see Table 2 | Measured |
| Molar liquid flow                | \( L_w \) | mol/s | 0.8 ± 0.1 | Measured and calculated from 20 wt% NaCl and 80 wt% H₂O as liquid, Supplier information |
| Radius of scrubber               | \( r \) | m | \( 55 \times 10^{-3} \) | Calculated as \( \frac{1}{2} \) diameter of material, \( \frac{1}{2} \) radius of material |
| Gas density                      | \( \rho_g \) | kg/m³ | 1.71 | Calculated as 90% CO₂ and 10% N₂ based on numbers from [44] |
| Gas viscosity                    | \( \eta_g \) | Pa s | \( 15.3 \times 10^{-6} \) | Calculated as 90% CO₂ and 10% N₂ based on numbers from [44] |
| Porosity                         | \( \epsilon \) | Fraction | 0.38 | Based on data from [45] |
| Diameter of packing material     | \( D_p \) | m | 0.025 ± 0.001 | Measured |
| Specific surface area            | \( a \) | m²/m³ | 149 | Calculated as \( a = \frac{1}{2} \) from Table 3 |
| Initial concentration of pollutant | \( y_i \) | Mole fraction | \( (1330 ± 70) \times 10^{-6} \) | Model biogas concentration |
| Final concentration of pollutant  | \( y_f \) | Mole fraction | 50 × 10⁻⁶ | Detection limit of the gas analysis equipment |
| Equilibrium end concentration    | \( y_e^* \) | Mole fraction | \( 5 \times 10^{-9} \) | \( y_e^* = y_i \), factor used: 10.000 |
| Equilibrium start concentration  | \( y_o^* \) | Mole fraction | \( 1.33 \times 10^{-7} \) | \( y_o^* = y_f \), factor used: 10.000 |
| Logarithm of the average fraction | \( x_{bio} \) | Unit less | 1 | Assumed value due to low concentrations of the pollutant |
| Gas film factor                  | \( C_g \) | Unit less | 2.5 | Assumed, based on values from [39] |
| Diffusion coefficient of the gas phase | \( D_{dg} \) | m²/s | \( 5.4 \times 10^{-4} \) | Based on models from [46,47] |
| Molar mass of gas                | \( M_g \) | g/mol | 42.2 | Calculated from density, assuming ideal gas behavior |

### 3.4. Scanning electron microscopy (SEM) analysis

The SEM used for analyzing the sulfur content was a Hitachi tabletop SEM TM3000 equipped with energy dispersive x-ray spectroscopy (EDS). The imaging was performed using backscattered electrons at 15 kV. The image was acquired at a working distance of 9500 μm with a magnification of 1000. The EDS analysis was performed at 15 kV with a mapping time of 500 s.

### 4. Results and discussion

Overall, the process proved capable of removing H₂S from a synthetic biogas. The effect of current, gas flow and the current efficiency was investigated in the sections below to give a better understanding of the process. The sulfur produced in the process is also analyzed in the sections below.

In Table 2, an overview of the results are presented. The scrubber is compared with the theoretical height calculated in section 2. Under the assumption presented, the height of the laboratory electrosrubber is modelled to be between 2.4 and 5 cm. This is a very low scrubber height. In the laboratory, scrubbers of similar height were tested by removing the packing elements to create an active scrubber height of only a few centimeters. The results are observed to support the model as shown in the figures below.

### 4.1. Effect of current

In theory, the current required for the oxidation of sulfide to sulfur \( (i_{th}) \) can be calculated applying Faraday’s law [48]:

\[
i_{th} = \frac{n_{sulfur}}{t} F = \frac{\gamma_{sulfur} \rho V}{RT} \frac{P}{F} \cdot F
\]

where \( n_{sulfur} \) is the amount of sulfur, \( t \) is time, \( \gamma_{sulfur} \) is the concentration of sulfur in the gas, \( p \) is the pressure, \( V \) the volume, \( R \) the gas constant, \( T \) the temperature, \( F \) is the Faraday’s constant and \( z \) is the number of electrons required for the reaction and is equal to 2, according to Eq. (1). It is assumed that H₂S may be considered an ideal gas at room temperature and atmospheric pressure. A 100% Faraday efficiency is assumed in combination with 100% scrubber efficiency in order to calculate the theoretical current. In reality, lower efficiencies may be expected. Typically, chloralkali plants are known to have efficiencies in the range of 50-75% [36] (3400–2100 kWh/ton Cl₂).

Lower current efficiency may be caused by undesired side reactions. This could include anodic O₂ generation [49]:

\[2H₂O = O₂(g) + 4H^+ + 4e^-\]

Or the oxidation of sulfur:

\[S + 4 H₂O = H₂SO₄ + 6H^+ + 6e^-\]

Or cathodic chlorine reduction:

\[Cl₂ + 2e^- = 2Cl^-\]

\[ClO^- + 2e^- + 2H^+ = Cl^- + H₂O\]

All of which will decrease the current efficiency.

A decrease in chloride concentration could also decrease the current efficiency. If there are not enough chloride ions present at the anode surface, side reactions will take place instead. Throughout the experiments, the same solvent was used without adding chloride. No changes to the current efficiency was observed over the course of the experiments.
content of the biogas. From Fig. 4 it is evident that this seems to be the case.

While the blue triangles in Fig. 4 show the CO$_2$ content in the synthetic biogas entering the system, the green triangles show the CO$_2$ content exiting the system. It is observed that throughout the experiment of almost 4 h, the CO$_2$ content in and out of the system remains the same within the uncertainties of the measurements. This indicates that CO$_2$ is not removed from the synthetic biogas. In other words, there is no correlation between the applied current and the removal of CO$_2$. Once the system is saturated with CO$_2$, it will no longer interact with the CO$_2$ content of the biogas. From Fig. 4 it is evident that this seems to be the case.

The black squares in Fig. 4 show the H$_2$S concentration that enters the scrubber, while the red circles show the H$_2$S concentration exiting the system. It is observed that after an initialization of approximately 30 min, the initially applied current of 3.46 times the theoretical current is sufficient to removal all the H$_2$S from the synthetic biogas. This result was confirmed by a non-detectable H$_2$S concentration in 5 subsequent analyses. Following this, the current was decreased to a value 2.26 times the theoretical current. The system responded quickly, as the next two data points show a H$_2$S concentration of 350 ppm while. The applied current is then changed again, to a value between the two previous set-points, i.e. 2.82 times the theoretical current. Again, the system responded quickly to the change as the H$_2$S concentration in the exiting gas decreased to 160 ppm.

Fig. 4 shows that the system is capable of removing H$_2$S down to non-detectable concentrations. In order to do this, it is required to adjust the current of the electrochemical cell. At lower applied currents, the system is not capable to remove all H$_2$S. However, significant removal of H$_2$S is observed for all applied currents. The system is also observed to adjust quickly to changes in the operating parameters. When the applied current is changed, after 2 and 3 h, the system is adjusted within 20 min. This is observed, as the H$_2$S concentration is changed and appears stabilized when the next data-point is obtained 20 min. after the change in the applied current. This is expected, as the liquid goes through a full cycle almost 25 times within 20 min.

From Fig. 4, it is also observed that in order to remove H$_2$S completely, currents more than 3 times higher the theoretical currents are required. It is expected, that upon optimization of the electrochemical cell, the current efficiency will increase. An optimization could include the utilization of better suited electrodes and a membrane to separate the cathode and anode.

### Table 2

Overview over parameters for the experiments regarding scrubber height. The standard deviation has been calculated using the n-1 method. For all concentration data points, the uncertainty has either been the standard deviation or 50 ppm, depending on which is the larger number. *the measured value was zero, indicating that the concentration was lower than the detection limit (0.5 ppm).

| Gas flow [10$^{-4}$ mol/s] | Inlet H$_2$S value modelled [ppm] | Inlet H$_2$S value measured [ppm] | Standard deviation | Outlet H$_2$S value measured [ppm] | Standard deviation | Scrubber height theory [cm] | Scrubber height test [cm] | Removal rate [%] | Current [A] |
|--------------------------|---------------------------------|---------------------------------|-------------------|-------------------------------|-------------------|---------------------------|-------------------|----------------|------------|
| 0.67                     | 500                             | 468                             | 14                | 33                            | 11                | 2.37                      | 2.1 ± 0.2         | 93 ± 15         | 0.183      |
| 0.78                     | 500                             | 495                             | 2.5               | 135                           | 9.6               | 2.46                      | 2.1 ± 0.2         | 73 ± 14         | 0.215      |
| 1.12                     | 500                             | 461                             | 3.7               | 190                           | 0.82              | 2.69                      | 2.1 ± 0.2         | 59 ± 15         | 0.308      |
| 2.24                     | 1330                            | 1262                            | 21                | 346                           | 19                | 4.56                      | 3.2 ± 0.2         | 73 ± 6          | 1.24       |
| 2.24                     | 1330                            | 1327                            | 84                | 156                           | 11                | 4.56                      | 3.2 ± 0.2         | 88 ± 7          | 1.62       |
| 2.24                     | 1330                            | 1331                            | 31                | Not detected*                 | 0*                | 4.56                      | 3.2 ± 0.2         | 100 ± 5         | 2          |
| 2.80                     | 1330                            | 1908                            | 10                | 626                           | 31                | 4.82                      | 3.2 ± 0.2         | 67 ± 4          | 2.4        |
| 2.80                     | 1330                            | 1912                            | 18                | 74                            | 22                | 4.82                      | 3.2 ± 0.2         | 96 ± 4          | 3.66       |
| 3.26                     | 1330                            | 2035                            | 14                | 840.5                         | 25                | 5.05                      | 3.2 ± 0.2         | 59 ± 3          | 2.8        |

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In Fig. 4, the gas composition is presented for different applied currents relative to the theoretical minimum current ($i/i_{theory}$) for a constant gas flow of 200 L/h. The data was collected through a 4 h experiment where three different applied currents were tested. The background color indicates the applied current, which is either 2.26 (red), 2.82 (blue) or 3.46 (orange) times the theoretical current.

Fig. 3. The experimental set-up for testing of the electrochemical removal of H$_2$S.
4.2. Removal percentage

The removal percentage is observed to increase with current. In order to analyze the correlation between current and removal percentage, data is compared across different gas flow rates, gas concentration and applied currents. For this comparison, the mass of the removed sulfur (S) is calculated as:

$$ S = \frac{y_{\text{sulfur}}(\text{in}) - y_{\text{sulfur}}(\text{out})}{y_{\text{sulfur}}(\text{in})} \cdot \frac{pV}{RT} \cdot M_{\text{sulfur}} $$  \hspace{1cm} (26)

where $M_{\text{sulfur}}$ is the molar mass of sulfur (32.1 g/mole) and H$_2$S is assumed to be an ideal gas. For comparison across parameters, S is used to determine the removal percentage of the system.

In Fig. 5, the black squares show the sulfur removed (S) as a function of the current density. The same electrochemical cell was used for all experiments, and an increase in current is therefore directly connected to an increase in current density. For each data-point, the light red bars indicate the total amount of sulfur in the gas during the experiment. It is therefore possible to see both the absolute amount of sulfur removed in g/h and the removal percentage. For instance, during the experiment performed with a current density of 2.0 A, 0.37 g/h of sulfur was removed. This corresponded to a complete removal of H$_2$S from the gas. Similarly, the experiment performed with a current density of 2.8 A resulted in the removal of 0.50 g/h. However, this only corresponded to sulfur removal rate of 59%.

A linear dependency is observed between the absolute sulfur removal (S) and the applied current (I). This linear dependency indicates that other parameters such as concentration of H$_2$S and flow rate of H$_2$S do not affect the overall efficiency of the system. The corresponding linear fit has a $R^2$-value of 0.993 a slope of $0.169 \pm 2 \times 10^{-3}$ g/(hA), and an interception at $6 \times 10^{-4}$. Thus, no matter the other parameters of the experiments performed, 0.17 g/h sulfur were removed per ampere applied. This value is 3.54 times lower than the theoretical value, which results in an overall current efficiency of 28.3% of the cleaning process. This efficiency is the combined efficiency of the electrochemical cell and the scrubber and may be improved upon optimizing the system. For the scrubber, better suited random packing material may increase the surface area for gas/liquid interaction, and it may also be possible to choose a more optimal packing material. The electrochemical cell may be further optimized by changing the electrode materials and surface area of the electrodes.

The linear dependency between the applied current and the measured efficiency indicates that the unwanted reactions consuming energy also increase with applied current. The reaction efficiency inside the scrubbing tower is not expected to depend on the current in the electrochemical cell, and it may therefore be assumed that the unwanted side reactions at the electrodes are the main reason for the loss of efficiency. Carbon electrodes were used for these experiments, and optimized surface treated electrodes could hinder the side reactions. This could help increase the current efficiency of the system.

4.3. Effect of gas flow and removal percentage

The removal percentage ($\Delta S$) is calculated using simple mass balance principles, based on gas-side H$_2$S concentrations, entering and exiting the system:

$$ \Delta S = \frac{y_{\text{sulfur}}(\text{in}) - y_{\text{sulfur}}(\text{out})}{y_{\text{sulfur}}(\text{in})} $$ \hspace{1cm} (27)

where, $y_{\text{sulfur}}$ is the H$_2$S mole fraction in the biogas.
Fig. 6 show ΔS as a function of gas flow and applied current. The red circles show ΔS from experiments performed with \( \frac{i}{i_{\text{theory}}} = 3.5 \pm 0.2 \) times the theoretical current, while the black squares show ΔS from experiments with \( \frac{i}{i_{\text{theory}}} = 2.2 \pm 0.2 \). It is observed at the 200 L/h and 250 L/h gas flows that an increase in \( \frac{i}{i_{\text{theory}}} \) results in an increase in the ΔS. It is also observed that increasing the gas flow decreases the removal percentage. This is showed for different flows and for different \( \frac{i}{i_{\text{theory}}} \)-values. It is observed, that for a gas flow of 200 L/h, an \( \frac{i}{i_{\text{theory}}} \)-value of 3.5 \pm 0.2 is sufficient to achieve ΔS = 100%. From the two arrows in Fig. 6, it is shown how ΔS can be decreased:

- Increased gas flow (arrow 1 in Fig. 6)

If the gas flow is increased from 200 L/h to 250 L/h, the ΔS decreases. This indicates that the reaction becomes limited by kinetics or mass transfer inside the scrubbing tower. This will be discussed below.

- Decreased the applied current (arrow 2 in Fig. 6)

If the \( \frac{i}{i_{\text{theory}}} \)-value is decreased to 2.2 \pm 0.2, ΔS decreases. This indicates that there is not enough oxidizing agent in the electrolyte.

The electroscrubber is observed to be limited by the two different mechanisms above; the amount of oxidizing species (applied current) and the contact time (gas flow) inside the scrubber. The amount of the electrolyte is represented as the \( \frac{i}{i_{\text{theory}}} \)-value. In theory, an \( \frac{i}{i_{\text{theory}}} \)-value of 1 should be sufficient for complete removal of H₂S. At an \( \frac{i}{i_{\text{theory}}} \)-value of 1, the oxidizing species is produced in a 1:1 ratio with H₂S. If the current efficiency is 100%, then \( \frac{i}{i_{\text{theory}}} = 1 \) is enough for a complete removal. Due to unwanted side reactions and reverse discharge reactions, the expected efficiency is lower than 100%. In Fig. 6, a low \( \frac{i}{i_{\text{theory}}} \)-value results in a low capacity of the electrolyte and therefore lower ΔS. The gas flow shows the limitations in mass transfer and reaction kinetics. As expected, if the gas flow is too high, the electroscrubber does not have the time to remove all the H₂S. From these experiments, it was determined that 0.30 L packing material was required for a gas flow of 200 L/h. This corresponds to a space velocity of:

\[
\frac{200 \text{L/h}}{0.30 \text{L}} = 667 \text{h}^{-1}
\]  

(28)

It may be assumed, that this value was dependent on the removal reaction and independent on the scrubber design. In other words, the space velocity can be used for scaling up the electroscrubber.

4.4. Sulfur

Throughout the experiments, elemental sulfur was formed as described in Eq. (8). Apart from sulfur, no other product was found. Due to the low flow rates and concentrations, however, less than 5 g of sulfur were formed throughout the experiments. As expected, the sulfur was observed to float on top of the electrolyte [50]. While operating the system, the liquid was not filtered, allowing for the sulfur to be collected on the electrolyte surface. Upon finishing the experiments, sulfur was collected from the electrolyte and analyzed EDS and SEM, as seen in Fig. 7. In Table 3, the elemental configuration is presented, as NaCl is part of the dried sulfur. The oxygen content could indicate that some of the sulfur has been oxidized to sulfate. This reaction could both take place inside the reactor as an unwanted side reaction or when the sulfur was exposed to air upon dissembling the scrubbing unit. However, no rapid reaction between elemental sulfur and atmospheric oxygen was observed. Another explanation for the oxygen content could be that hypochlorite (NaClO) is present in the sulfur instead of NaCl.

For continuous operation with batch recirculation, a filtration system for removal of elemental sulfur is required. A system operating with a mixture of NaCl and water can have periodical change of liquid without large expenses and thus, the filtration system should not increase the operational cost (OPEX) significantly. The elemental sulfur may be used as fertilizer either by mixing it with the degassed manure returning to the farms or by a separate sulfur handling system. If the quality of the sulfur is high enough, other applications within the chemical industry may even create a revenue from the scrubbing system.

Table 3

| Element | S    | O    | Na   | Cl   |
|---------|------|------|------|------|
| Atomic% | 72 ± 3 | 11 ± 1 | 8.9 ± 0.3 | 8.1 ± 0.4 |

5. Conclusions

Overall, the electroscrubbing desulfurization process based on a low pH electrolyte proved capable of removing H₂S from a synthetic biogas containing CO₂ and N₂ as replacement for CH₄. For a 200 L/h gas flow with 1330 ppm H₂S, a complete removal to below detection limit was achieved using a current of 4.5 A, corresponding to 3.5 times the theoretical current value.
By adjusting the applied potential, the resulting current could be regulated to ensure a complete removal of H₂S. This result indicates that the process is flexible and can be adjusted to fluctuations in the H₂S concentrations. The electroscrubber was found to have a quick response time, allowing for real-time adjustments.

A linear correlation was found between applied current and removal percentage. A constant value of 0.17 g sulfur was removed per ampere per hour independent on other parameters, i.e. the gas flow rate, H₂S concentration, and applied current. This value corresponds to an overall current efficiency of 28.3%. It is expected that the efficiency can be further improved by optimizing the electrode materials and surface area of the electrodes.

The electroscrubber was determined to have a space velocity of 667 h⁻¹, which can be used for scaling up the process. This is a high space velocity when compared with biological processes that usually has a regulated to ensure a complete removal of H₂S. The work reported in this paper.

The column height for the process was calculated using basic mass transfer theory. The height was found to be extraordinarily small due to carbonate formation due to the solvent saturated with no unwanted oxygen generation at the anode. During 14 days of operation, unwanted side reactions were also described, and these may decrease the current efficiency. One of these unwanted side reactions is oxygen development. During 14 days of operation, the sulfur removal was found to suffic for sulfur removal.

The new electroscrubbing process can be used for cleaning of carbon sources for e.g. CCO or catalytic processes such as the Sabatier reaction.

CRediT authorship contribution statement

Sebastian Nis Bay Villadsen: Investigation, Conceptualization, Writing – original draft, Visualization. Malene Ahrensberg Kaab: Investigation, Formal analysis. Lars Pleth Nielsen: Writing – review & editing, Supervision. Per Møller: Conceptualization, Supervision, Funding acquisition. Philip Loldrup Fosbol: Supervision, Methodology, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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