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Investigating internal biological desulfurisation through dosage with ambient air

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Internal biological desulfurisation is a simple and cost-efficient procedure for extracting large amounts of hydrogen sulfide (H_2S) from biogas. Through introducing ambient air into the fermenter gas chamber, the H_2S in the chamber is metabolised by colonising aerobic sulfur bacteria into elementary sulfur and water.

In our study conducted in a fermenter at the "Unterer Lindenhof" research biogas plant, the reduction of H_2S concentration ("desulfurisation") through injecting different doses of air was investigated. It was demonstrated that a reduction in H_2S of around 90% was possible, although the amount of air required for this lay disproportionately higher than that required for a lesser degree of desulfurisation. With a 90% reduction, ratio of air flow to biogas flow was around 0.02 and thereby lay at the lower end of the recommended range of 0.02–0.08. With a reduction of 50%, this ratio lay at only 0.002.

An even higher degree of desulfurisation (> 90%) through introducing still more ambient air into the fermenter would have been theoretically possible, this is, however, prevented by a safety criterium that permits only a maximum measured O_2 concentration of 0.5% in the fermenter.

Keywords
Biogas, hydrogen sulfide, biological desulfurisation, air dosage

Sulfur is a component of all known living organisms, present mainly in some amino acids such as cysteine and methionine and their derivatives (Jacob 2003). Via substrate input, sulfur in bonded form enters the fermenter where it converts to sulfide, hydrosulfide and hydrogen sulfide via microbial metabolism and abiotic reactions. Measured as gaseous component in biogas plants, H_2S concentrations range from a few ppm to several thousand ppm (Reinelt 2017).

Even in smaller concentrations, H_2S represents a danger to health (a few ppm) or can be fatal (starting at just a few 100 rpm) (ATSDR 2006). Besides this, the presence of water and sulfur compounds can produce acidic condensate that corrodes gas transporting components in a biogas plant (Schnell 2003). This effect is increased by flexible operation of the thermal power station and associated cooling of the exhaust gas tract.

Through combustion of H_2S, in other words utilisation of biogas as fuel in the thermal power station, in particular sulfur dioxide and sulfur trioxide (SO_x), as well as derivatives, are produced from oxygen and hydrogen sulfide. This leads to acidification of oil and thus impairment of the engine oil lubrication effect. This effect has caused manufacturers to introduce oil additives aimed at resisting acidification.

Moreover, sulfur oxides are emitted through exhaust gases into the environment, causing damage to humans, animals, plants and construction materials (Mochidaa 2000).

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Extraction of H_2S from biogas, or avoidance of its production in the first place, is therefore of great importance for economic efficiency, operational safety and environmental protection. Various methods of desulfurisation in the sectors biogas, sewage and waste depot plants have become established:

- Through biological desulfurisation, hydrogen sulfide is converted by microorganisms and atmospheric oxygen. Both external and internal procedures are possible. The internal approach includes injection of atmospheric air into the fermenter gas container (Naegele 2013). Produced as a derivative of biological desulfurisation is elementary sulfur (Figure 1).
- With (internal) chemical desulfurisation, production of H2S is already prevented through precipitation of low-solubility sulfides. Applied most often in this respect are ferrous salts (ferrous chloride or ferrous hydroxide) which in most cases are added daily into the fermenter (Schneider 2002).
- With the (external) physical procedure, biogas is cleaned through gas washers or adsorption cascades (Osorio 2009).

In this study, the internal, biological desulfurisation is measured in a fermenter at the "Unterer Lindenhof" research biogas plant, whereby dosage of ambient air is varied and the resultant reduction in H_2S recorded. Conclusions were able to be made regarding total efficiency of desulfurisation and amount of air required.

Figure 1: Inside view of the fermenter. Through microbial degradation of the hydrogen sulphide, deposits of elementary sulfur are deposited on the ceiling. © State Institute of Agricultural Engineering and Bioenergy, University of Hohenheim.

Research plant description

The investigation into internal biological desulfurisation was conducted in one of the two fermenters of the University of Hohenheim's "Unterer Lindenhof" research biogas plant in Eningen u. A. The basic technical construction of the plant is described by Mönch-Tegeder (2015). The cylindrical concrete container has a total volume of approx. 923 m^3 (inner diameter approx. 14 m). For the trial, this was filled to around 85 % capacity with fermentation substrate. Temperature within the reactor during the trial was 43.9 °C ± 1.1 °C. The trial was carried out over a period of 19 days (27.11.–15.12.2018). During the trial period, the following substrate was loaded daily into the fermenter:

- Cattle dung (FYM): 1200 kg ± 620 kg
- Maize silage: 2940 kg ± 520 kg
- Grass silage: 3000 kg ± 350 kg
- Horse manure: 690 kg ± 530 kg
- Grain: 580 kg ± 60 kg, mixed with 1580 kg ± 170 kg water
- Cattle and pig liquid manure: 5580 l ± 3060 l

The organic loading rate (OLR), based on the organic dry matter being fed into the fermenter container during the trial period, was approx. 3.61 kgoTS m^{-3} d^{-3}. The average length of time the substrate remained within the fermenter (hydraulic retention time, HRT) was approx. 50 days. Fluctuations in substrate loading rate had only a limited effect on biogas H_2S content (Table 1).

Gas measurement

The wet biogas volume flow, gas temperature and gas pressure were all continually measured in the gas pipeline at the fermenter outlet by a GDR 1404 gas monitor (Esters Elektronik, Deutschland). Volume flow was determined under standard conditions (1013.25 mbar, 0 °C, assuming an ideal gas and 100 % relative air moisture content) and based on the dryness of condition (Equation 1):

$$\dot{V}_{N,dry}^{biogas} = \dot{V}_{measured}^{biogas} \frac{T_0}{T_{measured}} \frac{p_{measured}}{p_0} \left(1 - \frac{p_{H2O}^{s}(T_{measured})}{p_{measured}}\right) \quad \text{(Eq. 1)}$$

with

$\dot{V}_{N,dry}^{biogas}$: dry biogas standard volume flow
$\dot{V}_{measured}^{biogas}$: measured, moist biogas volume flow
T_0: gas temperature in standard condition
p_0: gas pressure in standard condition
$T_{measured}$: gas temperature
$p_{measured}$: gas pressure
p_{H2O}^{s}: steam pressure from pure water

The composition of the dry biogas, i. e. following internal cooling of the gas and channelling of condensate into the measuring instrument, was determined approx. every two hours via a biogas analyser (InCa Bio 04, Union Instruments GmbH, Deutschland).

Biological desulfurisation

For biological desulfurisation, ambient air is injected via air pump (Takatsuki Hiblow HP 100, Japan) directly into the gas container of the fermenter. The air is injected at a single point through the fermenter wall near the fermenter ceiling. The air injection took place on the opposite side of the fermenter from the removal point of the produced biogas, in order to achieve an as long as possible retention time for injected air doses within the gas container. The (good) distribution of the air within the gas container is decisive for (effective) desulfurisation this aspect is planned to be the subject of future investigations. Up until now, no such studies are available in the literature. For standardisation of air flow, pressure and temperature data were collected from the nearby weather station "Eningen unter Achalm" and applied.

Desulfurisation occurs mainly on the fermenter ceiling and on the inner walls (Figure 1). No additional colonisation areas (e.g. using special colonisation grids or wooden constructions) were prepared for sulfur bacteria.

The minimum required amount of air $\dot{V}_N^{air(\min)}$ to achieve complete elimination of hydrogen sulfide can be estimated via mass flow balance and with the help of the reaction $H_2S + ½\ O_2 \rightarrow H_2O + S$ (Equation 2):

$$\frac{\dot{V}_N^{air(\min)}}{\dot{V}_{N,dry}^{biogas}} = \frac{1}{2}\frac{x_{H2S}^0}{x_{O2}^{air}} \qquad \text{(Eq. 2)}$$

with:

x_{H2S}^0: the concentration of H_2S in the dry biogas before desulfurisation
x_{O2}^{air}: the (constant) oxygen proportion of the injected ambient air.

Because of secondary and subsequent reactions (e.g. production of sulfates and sulfites, Naegele 2013), the minimum amount of air so calculated does not necessarily represent the actual minimum amount for complete desulfurisation. Rather, the answer arrived at in this way serves as a reference value for later evaluation of trial results.

During the trial period, biogas flow represented an average of approx. 94 m^3N/h. The H_2S concentration in produced biogas before desulfurisation was approx. 240 ppm. With an oxygen concentration in the ambient air of approx. 21 % this, according to Equation 2, produced a value of approx. 0.0006 for the described stoichiometric ratio for minimum air flow to biogas flow. Recommendations from the literature for this ratio lie by approx. 0.02 to 0.08 (Polster 2006) and are therefore higher by the factors 33 to 133 than the minimum required amount of air.

Conducting the experiment and evaluating results

Various air flows were applied in the investigation, and the subsequent reduction of H_2S concentration Δx_{H2S} measured. Each air flow was maintained at a constant level over a period of 36 h until a stationary value for the new H_2S concentration had been measured. This value (x_{H2S}) was then applied for assessing the extent of desulfurisation.

Between one air flow setting and the next, the pumps were switched off so that the value of the H_2S concentration could be determined without biological desulfurisation (x^0_{H2S}). After some two days, a stationary value for the initial concentration was determined and then applied in each case to assess desulfurisation efficacy.

Applied for estimating desulfurisation efficiency, the desulfurisation efficiency η, defined as the relative H_2S reduction, i.e. the reduction of H_2S concentration based on the initial concentration (Equation 3), was:

$$\eta = \frac{\Delta x_{H2S}}{x^0_{H2S}} = \frac{x^0_{H2S} - x_{H2S}}{x^0_{H2S}} = 1 - \frac{x_{H2S}}{x^0_{H2S}} \qquad \text{(Eq. 3)}$$

Hereby, it is implicitly assumed that the biogas flow in the respective experiment block is constant. Also not explicitly taken into account is the reduction in concentration through dilution due to the air injection: The theoretical reduction in concentration through dilution was, in our trials, a maximum of approx. 2 % and could therefore be ignored in comparison to the effects of biological desulfurisation. The maximum dilution effect on a biogas component i can be theoretically estimated with Equation 4:

$$\Delta x_i^{dilution} = x_i^0 - x_i = x_i^0 \left(1 - \frac{1}{1 + \frac{\dot{V}_N^{air}}{\dot{V}_{N,dry}^{biogas}}} \right) \qquad \text{(Gl. 4)}$$

with

$\Delta x_i^{dilution}$: the reduction of concentration of the component i based on the air injection,

x_i^0 : the concentration of i before the air injection

x_i : the concentration of i after the air injection without any reactions that had taken place

For estimation of maximum diluting effect it was assumed that the volume and concentration changing reactions are negligible compared with the changes to volume or concentration through air injection

With Equations 2 and 4 is produced Equation 5:

$$\Delta x_i^{dilution} = x_i^0 \left(1 - \frac{1}{1 + \frac{1}{2} \frac{\dot{V}_N^{air}}{\dot{V}_N^{air(\min)}} \frac{x_{H2S}^0}{x_{O2}^{air}}} \right) = x_i^0 \left(1 - \frac{1}{1 + \frac{1}{2} \Gamma \frac{x_{H2S}^0}{x_{O2}^{air}}} \right) \quad \text{(Eq. 5)}$$

This applies specifically for the maximum H_2S reduction through dilution (Equation 6):

$$\Delta \eta^{dilution} = \frac{\Delta x_{H2S}^{dilution}}{x_{H2S}^0} = 1 - \frac{1}{1 + \frac{1}{2} \Gamma \frac{x_{H2S}^0}{x_{O2}^{air}}} \quad \text{(Eq. 6)}$$

Analogous to air figure λ during combustion processes is thereby $\frac{\dot{V}_N^{Luft}}{\dot{V}_N^{Luft(\min)}}$ the (dimensionless) air injection dosage Γ.

A mathematical model in Monod equation form was adapted to the measured values from our trial, $\eta(\Gamma)$ (Equation 7):

$$\eta(\Gamma) = \frac{\Gamma - \beta}{\Gamma - \beta + k} \quad \text{(Eq. 7)}$$

In Equation 7, β thus describes the minimum air flow required so that a measurable desulfurisation can take place. This value can, e. g., be produced through short blasts of the injected air, or through secondary reactions of the oxygen. *k* is a model parameter by which $\eta(\beta + k) = 0{,}5$ gilt. *k* determines decisively the form of the model curve and is dependent on very many parameters such as temperature, kinetic limitations, colonisation areas, microorganism nutrient supply, fermenter geometry, location of air injection or the period of time for, and distribution of, air within the gas container. The smaller *k* is, the more effective the biological desulfurisation. Additionally assumed for this model is 100 % desulfurisation efficiency by very large amounts of air ($\eta(\Gamma \to \infty) = 1$).

Results and discussion

Table 1 summarises the results of the desulfurisation investigation. Summarised for each trial block is: The air standard volume flow applied ($\dot{V}_N^{air}$), the dry biogas standard volume flow ($\dot{V}_{N,dry}^{biogas}$), the H_2S concentration before desulfurisation (x_{H2S}^0), the H_2S concentration after desulfurisation (x_{H2S}), the desulfurisation efficiency (η), the ratio of configurated air flow to theoretical minimum air flow (Γ) and the ratio of configurated air flow to biogas flow $\frac{\dot{V}_N^{air}}{\dot{V}_{N,dry}^{biogas}}$.

Table 1: Trial results of biologic desulfurisation

Versuchsblock	$\dot{V}_N^{air}$ ($l_N\ h^{-1}$)	$\dot{V}_{N,dry}^{biogas}$ ($m^3_N\ h^{-1}$)	x_{H2S}^0 (ppm)	x_{H2S} (ppm)	η	Γ	$\frac{\dot{V}_N^{air}}{\dot{V}_{N,dry}^{biogas}}$
I	462	78.8	226	46	0.796	10.90	0.0059
II	130	95.8	209	130	0.378	2.73	0.0014
III	226	87.5	219	75	0.658	4.95	0.0026
IV	736	108	232	37	0.841	12.34	0.0068
V	2105	103.6	249	35	0.859	34.27	0.0203
VI	116	111.3	250	193	0.228	1.75	0.0010
VII	69	113.3	241	206	0.145	1.06	0.0006

In Figure 2 the desulfurisation efficiency η is presented as a function of the air injection dosage Γ. The dimensionless presentation comprises all important parameters of biological desulfurisation and permits comparison with other biogas plants. Through this type of presentation, evaluation of desulfurisation is possible independently of the initial H_2S concentration and from the amount of biogas produced. Also included is the dilution effect which was negligible in our trials (Figure 2, thin red line).

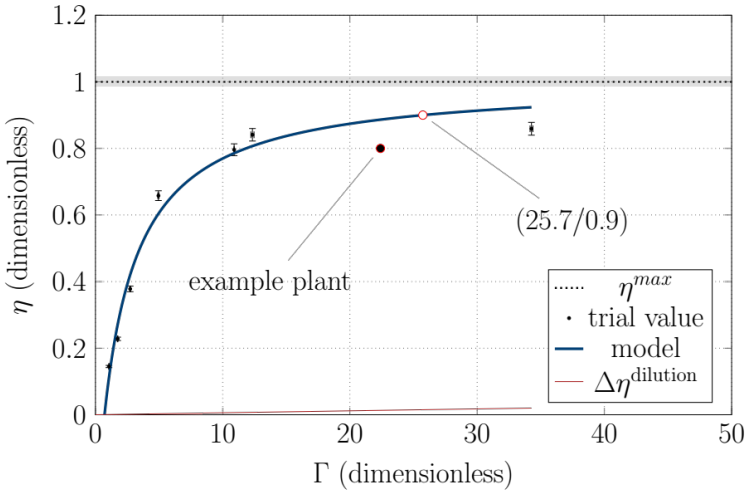

Figure 2: Process of the desulfurisation efficiency over the dosage of injected air. The upward H_2S reduction trend progressively reduces as air flow increases. The possible maximum (i. e. absolute) desulfurisation takes place where η^{max} = 1 statt. $\Delta\eta^{dilution}$ is here determined for x_{H2S}^0 = 250 ppm ppm according to Equation 6. The positioning of the example plant is described in the text.

In order to determine the desulfurisation efficiency of an arbitrary plant in Figure 2 the following parameters are required: (dry) biogas standard volume flow, the air flow dosage to be injected and the H_2S concentrations before and after desulfurisation. As example, a plant with biogas production of 150 m^3N/h, an air injection dosage of 4 m^3N/h, a H_2S concentration of 500 ppm before desulfurisation and 100 ppm after desulfurisation delivers a value for the ratio of adjusted air flow to theoretical minimum air flow Γ of 22.4 as well as a desulfurisation efficiency η of 0.8.

The model given in Equation 7 is matched to the measured values from our trial. The minimum flow β is determined at 0.71and the model parameter k at 2.78. Based on the model it can be deduced from this experiment that through injection of the stoichiometric air dosage, an H_2S reduction of approx. 9.3 % can be calculated. Additionally, it can be estimated that 90 % of the maximum desulfurisation efficiency can be achieved by an air flow ratio Γ of approx. 25.7, i. e. the fermenter would have 25.7 times the air volume flow injected which, stoichiometrically, would have been necessary for a complete desulfurisation. Through further increase of air dosage, desulfurisation efficiency increases only to a limited extent. Oxygen utilisation in this respect is very incomplete, so that the O_2 proportion in the biogas can, first of all, exceed the safety-relevant threshold. Secondly, through the oxygen oversupply, reactions with sulfur and water may occur, leading to the aforementioned acidification and corrosion.

The ratio of $\frac{\dot{V}_N^{air}}{\dot{V}_{N,dry}^{biogas}}$ with an approx. 90 % desulfurisation gives, in our trials, approx. 0.02 and, with that, lies at the bottom of the recommendation range of 0.02–0.08.

In the trials of Naegele (2013), the external biological desulfurisation in a percolating filter reactor was more efficient: In this case with a reactor temperature of approx. 40 °C a very high desulfurisation (> 95 %) was achieved with a ratio through $\frac{\dot{V}_N^{air}}{\dot{V}_{N,dry}^{biogas}}$ of approx. 0.024. At this small air injection dosage the dilution effect on biogas composition is negligible (Polster 2006). Thus, the theoretical reduction of CH_4 concentration by $\frac{\dot{V}_N^{air}}{\dot{V}_{N,dry}^{biogas}}$ = 0,02 , and within a typical methane proportion for regenerative source energy production plants of 52 %, result in a maximum of approx. 1 percent. In our trials, this small dilution effect was not verifiable in that it lay within the concentration fluctuations of the biogas composition (e. g. because of the fluctuations in the substrate composition, or the reactor temperature). Higher doses of injected air would have, however, led to a measurable reduction of CH_4 concentration, as also determined by Polster (2006) und Naegele (2013). For example, by $\frac{\dot{V}_N^{air}}{\dot{V}_{N,dry}^{biogas}}$ = 0.08 and 52 % methane proportion, a theoretical maximum reduction of the CH_4 concentration of approx. 4 percent would be possible. The reduction of gas quality, the technical safety aspect and the higher risk of producing acidic condensate must all be considered where there is to be a higher air injection dosage.

For consideration, Figure 3 shows the theoretical maximum reduction of the methane concentration through dilution with an initial value of x_{CH4}^0 = 52 %, e. g. for two different initial concentrations x_{H2S}^0 = 250 ppm and 1000 ppm. Moreover, the reduction of H_2S concentration through biological desulfurisation with the model parameter values from our trials (β = 0.71, k = 2.78, without dilution effect) is demonstrated. Here, the reductions are based on the dimensionless air injection dosage $\frac{\dot{V}_N^{air}}{\dot{V}_N^{air\,(min)}}$.

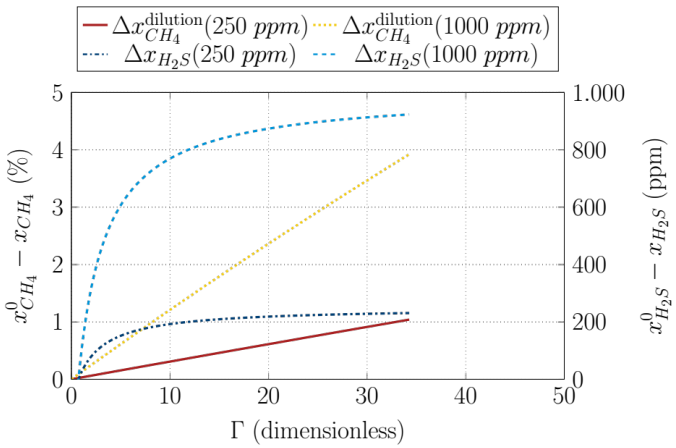

Figure 3: The process of H_2S concentration reduction through biological desulfurisation with the model values from our trials. Additionally applied here is the theoretical maximum reduction of the methane concentration as a result of air injection. This example was calculated for x_{CH4}^0 = 52 % and x_{H2S}^0 = 250 ppm/1.000 ppm with Equation 5 as well as Equations 3 and 7.

Figure 4, on the other hand, shows theoretical maximum reduction of the methane concentration through dilution with an initial value of x_{CH4}^0 = 52 %, based on the ratio $\frac{\dot{V}_N^{air}}{\dot{V}_{N,dry}^{biogas}}$.

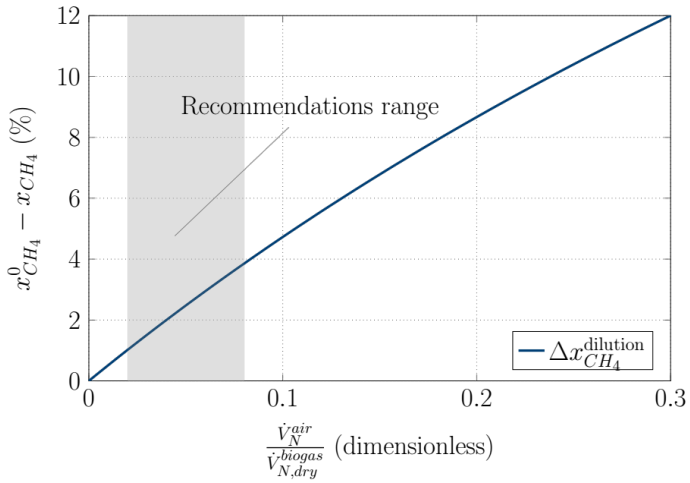

Figure 4: Theoretical process of methane concentration reduction through air injection. This example is calculated for x_{CH4}^0 = 52 % according to Equation 4. On the left side of the recommendation range, the reduction represents approx. 1 percent and, on the right border, approx. 4 percent.

Conclusions

Increased H_2S content in biogas can lead to serious damage to the gas transporting components within a plant, or to increased emissions of harmfull gases after combustion. For this reason, H_2S proportion in biogas must be substantially reduced. With internal biological desulfurisation exists a simple, cost efficient and effective technique for the removal of large amounts of H_2S from biogas.

In the literature, a reference value for the ratio of air flow to biogas flow of from 0.02 to 0.08 is given. Demonstrated in our trials and other trials (Naegele 2013) is that even a relatively low air injection dosage is sufficient for achievement of very high desulfurisation efficiency.

With increasing desulfurisation efficiency, the amount of air required increases over-proportionately. However, too much injected air can lead to an increase in O_2 concentration in the reactor chamber, an increased danger of acidification and corrosion and to a reduction in methane content. The effective distribution of the injected air onto the sulfur bacteria is therefore of decisive importance to achieve high desulfurisation and thus limited air requirement. The influence of air distribution and various bacteria colonisation surfaces should therefore be the subject of future investigations.

The above-mentioned reasons and limitations demonstrate that the technique presented here makes complete biological desulfurisation practically impossible. However, because modern power/heating plants are encouraged to achieve improved exhaust emission standards and a flexible operational procedure towards an almost complete elimination thereof, air injection dosage can only be a module in desulfurisation and must be combined in the future with external processes, for instance with active carbon filters.

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