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Submerged aquatic plant (*Vallisneria spiralis* and *Egeria densa*) utilisation as a biogas cleaner and feedstock of co-digestion

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Abstract. Biogas was produced from sheep manure and two types of submerged aquatic plant (Vallisneria spiralis and Egeria densa). The gas cleaning was carried out by a water scrubber, where a significant part of CO_2 and H_2S can be separated from the gas. A part of water from the scrubber was circulated through an aquatic plant growth tank and the growth of used plants was examined.

Addition of E. densa to sheep manure increased gas yield by 8% and the mixing of sheep manure and V. spiralis resulted in 21% increase in gas yield. With the used scrubber, 70-80 vol% methane content can be reached in the cleaned biogas, and the water from the scrubber (which contained dissolved CO_2 and H_2S) resulted in 56-87% increase in size as opposed to 12-44% increase in the control group.

Keywords: biogas, biogas cleaning, Egeria densa, manure, sheep manure, Vallisneria spiralis

Introduction

Anaerobic digestion is a widely used treatment method for methane production from organic materials e.g. wastewater[1], manure [2], food waste [3], etc. During anaerobic digestion, the micro-organisms decompose the organic matter in the absence of oxygen, and its main product will be biogas. As the biogas has high methane content, its primary application is heat and power generation via combustion. Beside biogas, the digestate is a useful material too, it is typically used as a soil improvement agent.

Most of biomass (including aquatic plants) are suitable for biogas production. Many publications deal with the potential of anaerobic digestion of marine and freshwater plants. Algae may serve as potential sources of biogas, but these technologies have considerable difficulties [4]. In a review of Marquez et al. [5], the opportunities of anaerobic fermentation of different seaweeds are presented. According to them, high gas yield can be expected in case of most seaweed species, even with the use of bacteria for the degradation of terrestrial plants.

In addition to salt water plants, freshwater plants can also be used in anaerobic fermentation. In case of these plants, they are usually used or studied as raw material for biogas production. Most of these

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live above and on the surface of the water, such as water hyacinth [6], or the seven types of wetland aquatic plants which were used by Jiang et al. [7].

Aquatic plants can also be advantageously used for co-fermentation with other starting materials. According to the paper of Tubassum et al. [8], mixing natural and cultivated seaweeds with dairy slurry can increase biomethane efficiency during biogas production. Allen et al. [9] studied the co-digestion of fresh and dried green algae (*Ulva lactuca*) with dairy slurry at different ratios. Based on their observations, the disadvantage of sea weed fermentation can be reduced by the common fermentation.

Based on the present data, the aquatic plants may be appropriate for the co-digestion with another, lower quality feedstock. For example, such low-quality base material is sheep manure, according to literature [10] and our previous results, too [11]. In 2015, over 85 million sheep were recorded to be the part of the livestock population of the EU-28 [12]. Thus, despite being a low-quality base material, the exploitation of the manure of these livestock has a potential if mixed with another material.

In order to increase the efficiency of biogas utilization and to carry out the process in a more environmentally friendly way, the gas needs to be upgraded. One of the simplest way is the use of water scrubbing, which decreases the CO_2 and H_2S content of the biogas [13]. The reduction of CO_2 and H_2S content will significantly improve the quality of biogas by increasing the calorific value. In addition to water washing, the cleaning can be enhanced by using algae. Ramaraj and Dussadee summarized the biological biogas purifying methods and its results [14]. According to this review, gas cleaning with algae is self-sustaining with the addition of minimal nutrients and light, furthermore, it has several advantages over the conventional chemical CO_2 reducing methods [14]. Moreover, Salafudin et al. [15] combined the water scrubbing and the CO_2 binding ability of the plants. Using catfish pond water (which was overgrown with wild algae) as gas purifier, and the measured CO_2 capture efficiency of the cleaning system was 50%.

The aquatic plants used (*Valisneria spiralis* and *Egeria densa)* have high tolerance and have been widely distributed around the world [16]. The main properties of the aquatic plants are summarised in Table 1.

Properties	*Vallisneria spiralis*	*Egeria densa*
Regular height, cm	30-100	40-100
Optimal temperature for growing, °C	15-30	10-26
Water hardness	soft – very hard	soft – very hard
pH tolerance	6-9	5-10
Growth	Fast	Fast
Habitat	static or flowing freshwater habitats, (lakes, ponds, water courses, and wetlands)	inland lakes and rivers, often shallow, mild or warm, still or slow-moving waters

Table 1. Properties of V. spiralis and E. densa [16, 17].

Numerous studies have already been carried out on the selected plants. For example, *V. spiralis* has high phytoremediation ability in case of polycyclic aromatic hydrocarbon (PAH) contaminated sediments [18]. Furthermore, this plant can accumulate high amount of cadmium, copper [19] and

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lead [20] from sediments. *E. densa* can accumulate the arsenic, zinc, aluminium [21] and selenium [22], as well as dissolved H_2S which has a positive effect for growth [23].

The goal of the experiments was to realize a sustainable lab-scale system, where the gas purification can be accomplished by aquatic plants that can be added to the fermenter as auxiliary feedstock. For this, *V. spiralis* and *E. densa* were used and the biogas production and composition were measured as well as the plants' length.

1. Materials and methods

The sheep manure originated from a livestock farm in Szendrő, and the plants (*V. spriralis* and *E. densa*) were obtained from local aquarium shop (Miskolc). The photographs of the used base materials and aquatic plants are shown in Figure 1., the results of ultimate and proximate analysis of the sheep manure, the two types of aquatic plants and the used mixtures can be seen in Table 2.

Figure 1. The used sheep manure (a), prepared E. densa (b) and the gas purifier V. spiralis (c) and Egeria densa (d).

	Parameter		Sheep manure	*Vallisneria spiralis*	*Egeria densa*	Sheep manure + 6 wt.% *Vallisneria spiralis (wet basis)*	Sheep manure + 6 wt.% *Egeria densa (wet basis)*
Dry matter	Nitrogen	wt.%	2.59	2.86	3.94	2.61	2.67
	Carbon		29.28	33.32	36.04	29.52	29.69
	Hydrogen		3.85	4.69	5.51	3.90	3.95
	Sulphur		0.70	0.65	0.44	0.70	0.68
	Oxygen		17.17	42.82	41.23	18.70	18.62
	Ash		46.42	15.66	12.48	44.57	44.38
	HHV	MJ/kg	9.87	10.38	12.75	-	-
Feed material	Moisture	wt.%	51.98	93.41	92.05	10	10
	pH	-	7.68	-	-	-	-

Table 2. Ultimate and proximate analysis of base materials.

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The schematic illustration of the lab-scale anaerobic digester system can be seen in Figure 2. The reactors were located in a 34 °C (mesophilic) water bath. The water scrubber operated without stopping during the experiment, but the water exchange between the water scrubber and the aquatic plant growing tank was batch. The water exchange worked 3 times a day for 60 minutes in total. This is 1 litre water change per day.

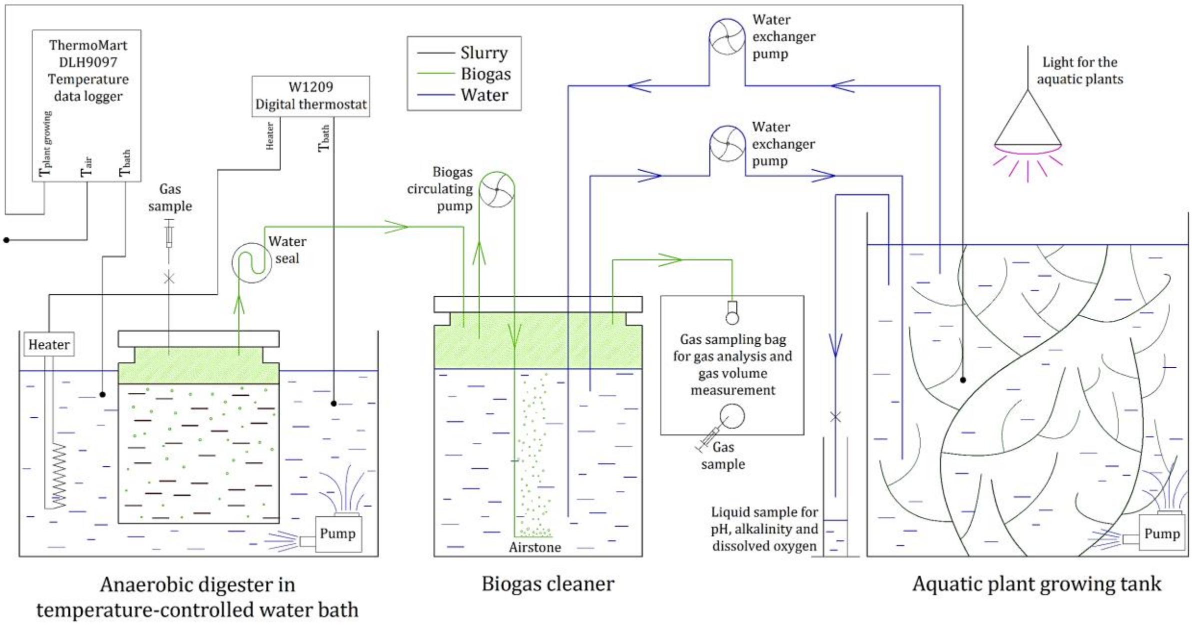

Figure 2. The lab-scale anaerobic digester system.

The produced gas was collected into a gas sampling bag and the amount of collected gas was measured every day. Gas samples were taken from both the sampling bags and reactors every 2-3 days and an Agilent 490 Micro-GC with a COX and a PPU modules were used for the analysis of CO_2, C_1-C_4, H_2, H_2S, CO, and O_2 content of the raw and cleaned biogas. The calculations of the Higher Heating Value (HHV) were based on the gas composition and the HHV of components. The plants were taken out of the water weekly and the total length of the plants, excluding sprouts, was measured. The measured parameters of the used plants at the beginning of the experiment can be seen in Table 3.

Properties	Cleaner		Control group	
	V. spiralis	*E. densa*	*V. spiralis*	*E. densa*
Number of sprouts, pcs	1	0	1	0
Length of all plants, cm	1105	155	1340	133
Number of leaves, pcs	47	-	58	-
Number of stems, pcs	3	5	3	5

Table 3. The initial sizes of used aquatic plants.

2. Results

Based on daily gas production, the cumulative gas production can be seen in Figure 3.

The gas formation started on the first day, due to the fact that the manure came from a part of dung storage that has been operating for a long time period and certain decomposition processes have already begun. The aerobic gas production due to these processes finished in a week and the anaerobic

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bacterium cultures growing in the next 10-15 days resulted in more intense gas production. The degradation of sheep manure started first, at 12-13 days, and with additive materials, the gas production was more significant after 15-16 days. The intensity of gas production started decreasing at day 34 in case of sheep manure and around day 36-38 for the mixtures. The lowest gas production was observed in case of the sheep manure, which was below 88 L gas/ kg dry matter. The addition of 6 wt.% *E. densa* resulted in 8% higher gas yield, while the addition of 6 wt.% *V. spiralis* induced 21% increase in the gas yield.

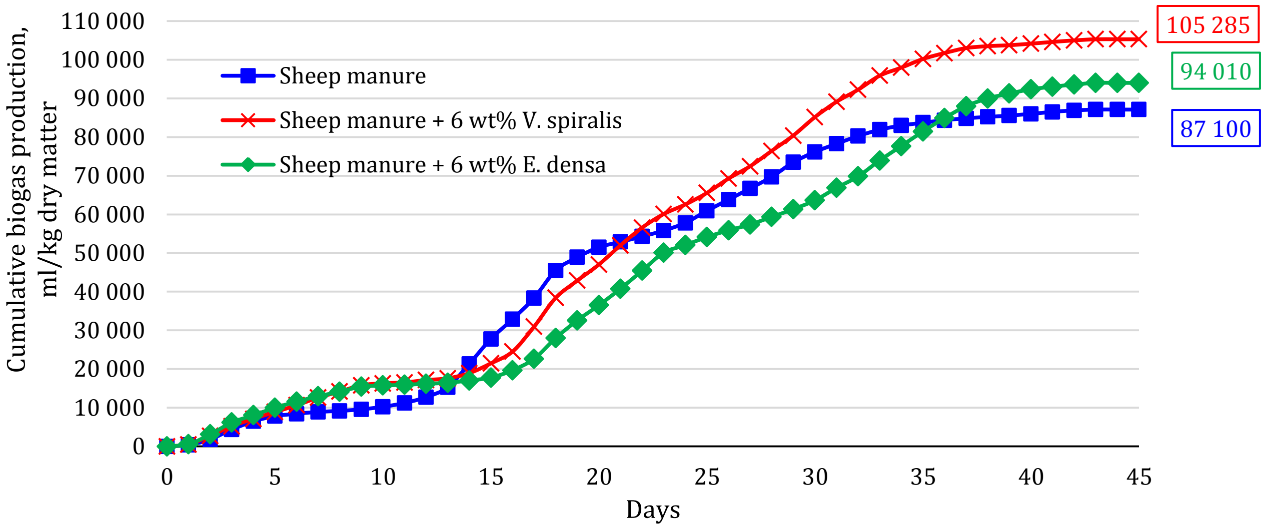

Figure 3. Cumulative biogas production.

The comparison of the composition of the formed biogas with the purified biogas composition is illustrated in Figure 4.

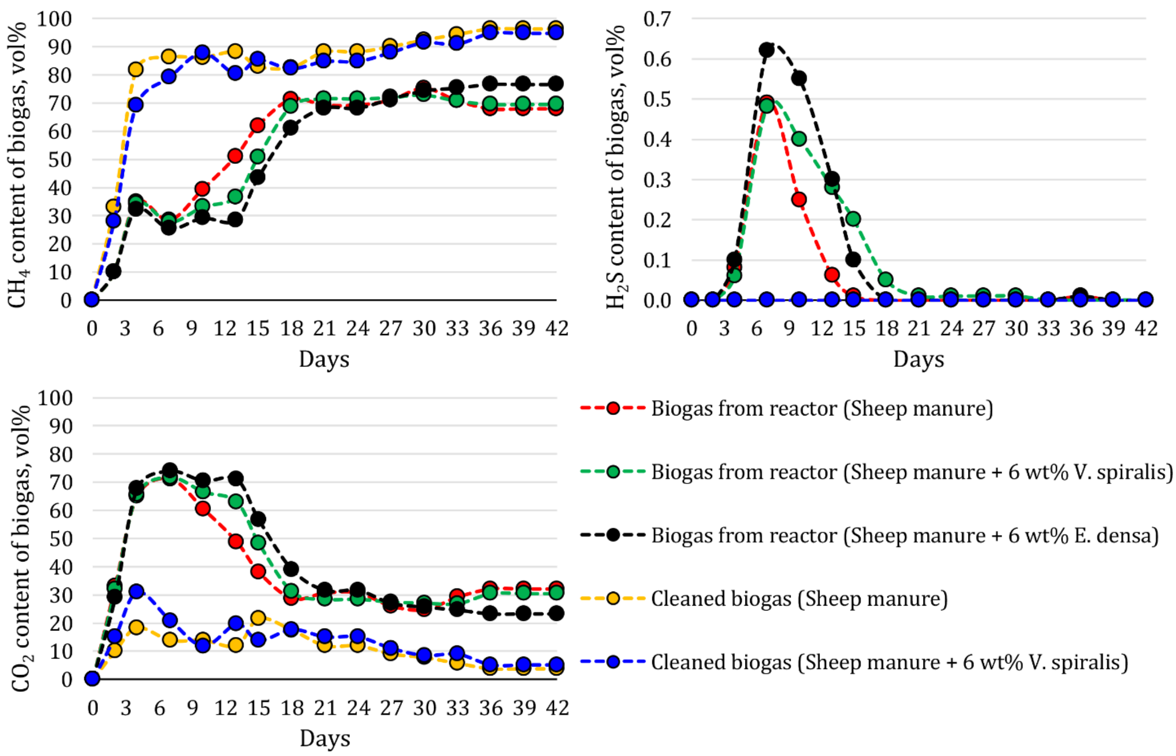

Figure 4. The changes of gas composition during the experiment.

Based on Figure 4., the produced gas mainly contained CH_4 and CO_2, in 99 vol%. The CH_4 content in every reactor reached its maximum after 18-20 days, after it was around 70 vol%. Due to the slower degradability of fresh additives, the increase in methane content was the fastest in case of sheep manure, however, the highest final CH_4 content arose from the sheep manure-*E. densa* mixture. Figure 4 clearly shows the effect of scrubbing, as a stable 80-90 vol% CH_4 content can be reached with its use. In the last days of the experiment, with a small amount of gas formation, the methane content of the purified gas reached 95 vol%.

H_2S formation was significant in the first 20 days of the experiment, during this period the maximum content of H_2S was 0.5-0.6. vol%. Despite the two plants had a lower sulphur content than the manure (Table 2.), in case of the manure the H_2S content of the biogas was the lowest, which increased with the additives. After 20 days, the amount of H_2S was below the limit of detection (0.01 vol%), however, the H_2S content of purified biogas was under 0.01 vol% during the whole duration of the experiment.

In addition to the components shown, some gas samples also contained hydrogen (max. 0.06 vol%), carbon monoxide (0.07 vol%) and other hydrocarbons (max. 0.03 vol%).

Based on the gas composition, the HHVs were calculated and Figure 5. shows the changes of HHV in relation to the duration of the experiment. During the first 20 days, the HHV increased and then its value was 28±2 MJ/m^3 as long as the experiment lasted. After the gas cleaning, the HHV increase was significant. By day 7, the HHV was over 30 MJ/m^3 and it rose to 35-40 MJ/m^3 in the last 12 days.

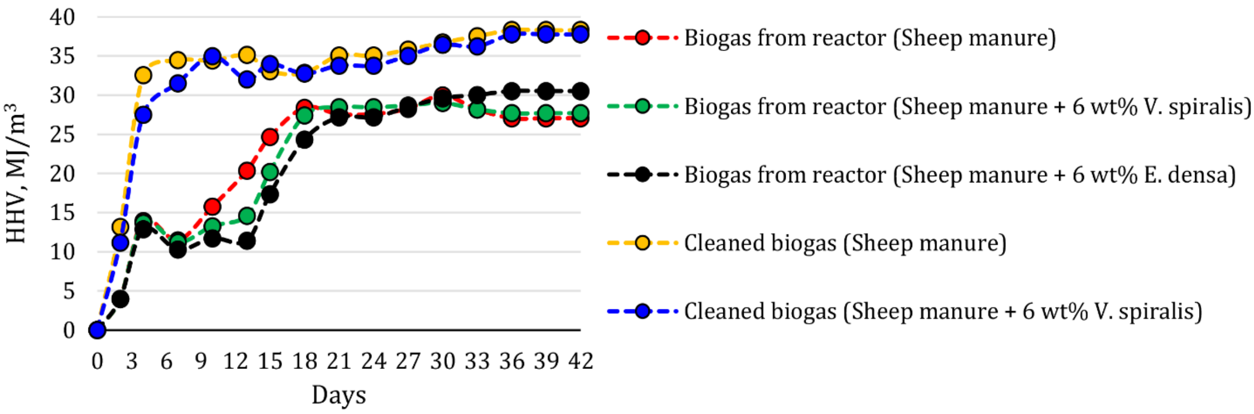

Figure 5. The changes of HHV of biogas during the experiment.

The water of the scrubber and of the plant growing tank was partially exchanged. The sizes of the plants growing in the exchanged water and the control group plants are illustrated in Figure 6. 100% refers to the total length of leaves (for *V. spiralis*) and stems (for *E. densa*). Plants that also received water from scrubber grew significantly faster than the plants of the control group. In case of *V. spiralis,* for 6 weeks, the growing was 75% faster but the number of sprouts was less (with 3) in case of cleaner plants than in the control group. In case of *E. densa,* the difference was 12% and the number of sprouts was higher (with 5) as a result of the scrubber water.

In case of *V. spiralis*, at the beginning of the experiment, cut leaves of uniform size did not grow any further, and some of them started to die slowly. In addition, the plants produced more sprouts in both tanks, but the size of the new ones had not been added to the size of the original plant. This caused the reduction in plant size towards the end of the experiment.

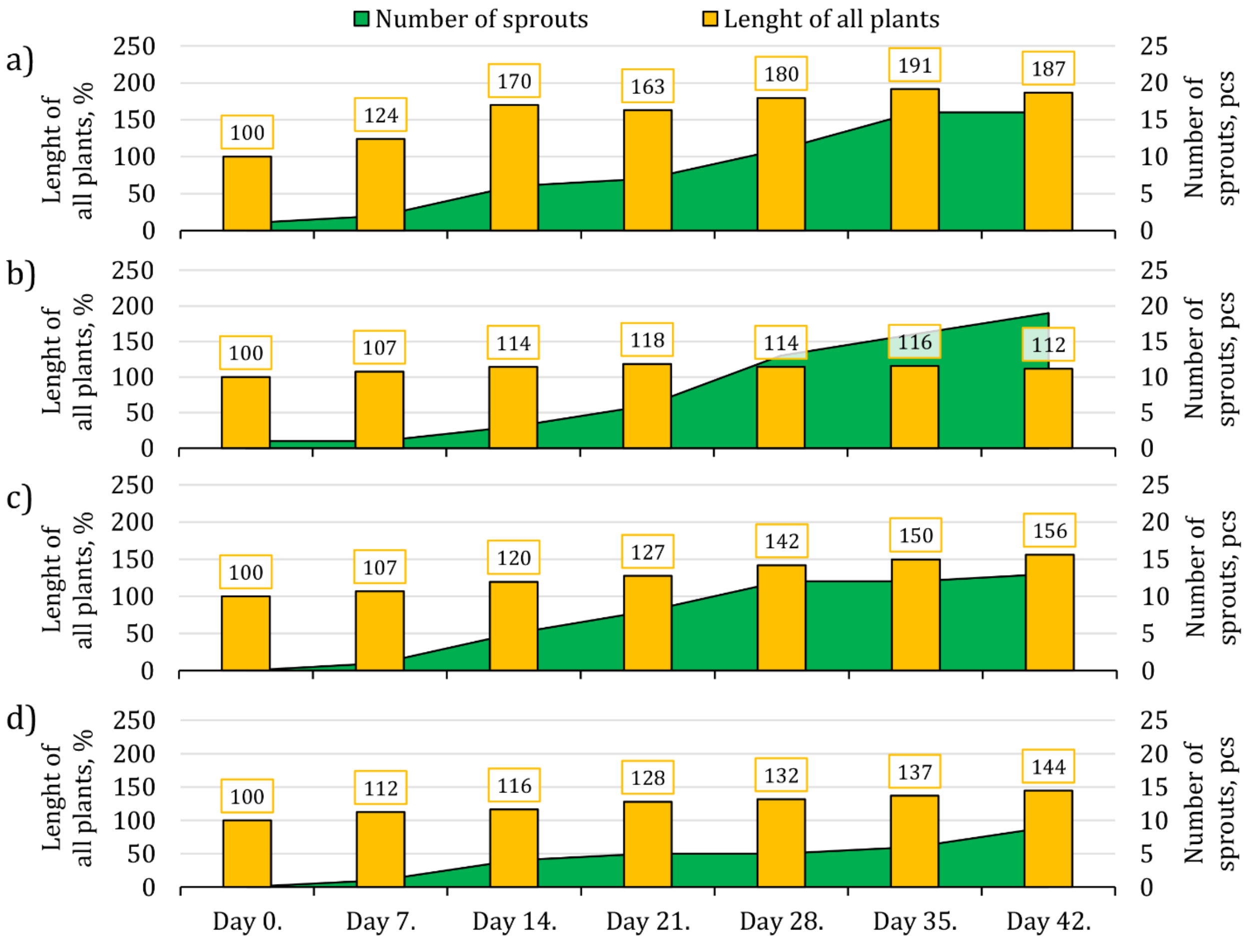

Figure 6. Plant growth and number of new sprouts during the experiment (a – V. spiralis, cleaner; b – V. spiralis, control group; c – E. densa, cleaner; d – E. densa, control group).

3. Conclusion

Sheep manure was fermented at 34 °C with and without aquatic plants (*Vallisneria spiralis* and *Egeria densa*) and these plants were used as gas cleaner too. In the case of mixtures, the gas formation was more difficult to start, but in the case of both additives a larger amount of gas was formed. 6 wt.% *V. spiralis* increased the gas quantity with 21%, and 6 wt.% *E. densa* with 8%. During all experiments, the methane content reached a maximum of 67-73 vol% and this period lasted 12 days. In contrast, the additives increased the H_2S content of the gas.

The water of scrubber absorbed significant amount of CO_2, thereby, the CO_2 content of gas decreased to 5-10 vol% by the end of the experiment. In addition, the H_2S content of the gas dropped below 0.01 vol%.

From the water of the scrubber, the size of *V. spiralis* increased by 87% in 6 weeks, while plants in the control group grew by only 12%. In case of *E. densa* the water of the scrubber caused 56% plant

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growth and significantly more sprouts were produced by the cleaning plants than the plants of control group.

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