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Lignite as additive in anaerobic digestion of liquid dairy manure without an external inoculum: Impact on BMP tests and fertilizer properties

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ABSTRACT

This study simultaneously investigated the effects of lignite as an additive in biogas applications and the suitability of the obtained digestate to produce organic nitrogen fertilizer. Lignite was added to the anaerobic digestion of liquid dairy manure without an external inoculum at a rate of 3.75 w.-% and a particle size of (1250-2500) µm. Maximum methane production rate, methane potential, and lag phase time were determined using the modified Gompertz model, respectively. Subsequently, the digestate was separated into liquid and solid phases, and nitrogen distribution and loadings were examined. Lignite addition increased the max. methane production rate by +34.7% and decreased the lag phase time by -15.17%. The absolute amount of nitrogen attached to the solid phase, which can be obtained from 1 t digestate after centrifugation, increased by 95.36% from 0.767 kg_N/t_D (DM) to 1.499 kg_N/t_D (DM + L) for lignite addition.

Keywords: Anaerobic digestion, Renewable Energy, Lignite, Biogas, Nutrient recovery

NOMENCLATURE

Abbreviations	
AD	Anaerobic digestion
TS	Total solids
VS	Volatile solids
FM	Fresh matter
TN	Total nitrogen
TC	Total carbon
LSR	Liquid-to-solid-phase-ratio
D	Digestate

1. INTRODUCTION

Biogas produced from anaerobic digestion (AD) of agricultural residues is a reliable and environmentally friendly source of renewable energy. In addition, AD minimizes the emission of odor, NH_3, N_2O, and CH_4 of manure during storage [1,2]. Untreated dairy manure and digestate can be used as organic fertilizer in livestock regions. Due to high water content, long transport distances often are uneconomical. Hence, excess manure in livestock regions as well as a demand for mineral fertilizers in arable farming regions occur. Phase-separation of the digestate and subsequent drying of the solid phase can decrease transportation costs and consequently reduce the demand for mineral fertilizers. An increase in total mass and concentration of nitrogen in the dry solid phase can improve the rentability of separation units to produce organic fertilizers due to lower transportation costs and increased solid fertilizer quality and quantity. In addition, reduced nitrogen loadings in liquid digestate lower the risk of nitrogen leaching in livestock regions.

Carbon-rich materials, like biochar or lignite, have recently been discussed in research for various applications. Used as soil conditioner, the storage capacity of soils for water and plant nutrients can be increased and greenhouse gas emissions can be reduced [3]. Furthermore, carbon-rich materials can be used as a carrier for nitrogen to create a slow-release fertilizer, decreasing nitrogen leaching and ammonia vaporization from the soil [4].

The use of lignite as an additive during anaerobic digestion can increase biogas production and stabilize the biochemical process [5,6]. By adsorption of nitrogen

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present in the digestate, carbon-rich materials can improve the digestion conditions and simultaneously increases the amount of nitrogen attached to the solid phase after separation. In this study, a combined approach is presented, simultaneously investigating the impact of lignite addition during anaerobic digestion on methane production and fertilizer properties.

2. MATERIAL AND METHODS

2.1 Liquid dairy manure and lignite

Liquid dairy manure (DM) was used as substrate and inoculum and collected from a dairy farm and biogas plant site in Hamminkeln, Germany. Lignite was collected at an RWE Power AG powerplant in Niederaußem, Germany. DM was sieved (< 2 mm) to remove larger solid organic matter and increase homogeneity. Lignite was sieved to a particle size of (1.25 – 2.50) mm. Total solid (TS) and volatile solid (VS) content of DM and lignite at the beginning and the end of AD were analyzed in triplicate according to DIN EN 12880 [7] and DIN EN 12879 [8]. Total nitrogen (TN) and total carbon (TC) content of DM and lignite were measured in dry matter using a CN analyzer (Vario MAX cube, Elementar). The chemical and physical properties of DM and lignite are given in Table 1.

Table 1: Chemical and physical properties of liquid dairy manure and lignite

Parameter	DM	Lignite
TS (% FM)	5.91 ± 0.003	45.20 ± 0.341
VS (% TS)	74.22 ± 0.107	82.53 ± 0.783
TN (% TS)	3.84 ± 0.005	0.58 ± 0.014
TC (% TS)	39.66 ± 0.040	54.79 ± 0.221

2.2 Anaerobic digestion

Biochemical methane-potential (BMP) tests were performed to determine the methane production from DM and DM with a lignite addition rate of 3.75 wt.-% of the digestate, respectively. The BMP tests were carried out according to VDI 4630 [9]. 500 ml glass bottles were used as digesters. The produced biogas volume was measured daily with a 400 ml eudiometer according to VDI 4630 [9] and converted to dry standard conditions. Biogas composition was analyzed with an uncertainty for CH_4 of 1.52 mol-% (k = 2.32) using a gas chromatograph (Focus GC, Thermo Fisher Scientifics Inc., USA). The GC is equipped with a micro-packed column (ShinCarbon ST 100/120, Restek Ltd.) and a thermal conductivity detector and operates with helium as carrier gas.

All tests were performed in triplicate in a climate chamber at a temperature of 39 ± 1°C. 300ml liquid dairy manure was filled into the digester. For lignite addition, DM was mixed with 15 g lignite. Afterwards, all samples were filled up with deionized water to the working volume of the digesters. The prepared samples were labeled DM (liquid dairy manure) and DM + L (liquid dairy manure with lignite).

2.3 Nitrogen distribution

After digestion, the digestate was separated into a liquid and solid phase using a lab-scale centrifuge (Centrifuge 5804, Eppendorf AG) and dried at 105°C for 12h. Rotation frequency and centrifugation time were kept constant for all samples. After centrifugation, the mass balance of the solid and liquid phases was determined for all samples. TN and TC content of the solid phase was measured in dry matter similar to DM and lignite. TN and TC of liquid phase digestate were analyzed photometrically (TOC-L, Shimadzu).

2.4 Data analysis

The modified Gompertz model was fitted to the experimental data in order to compare the kinetics of methane production with and without lignite addition:

$$B = B_0 \cdot exp\left\{-exp\left[\frac{R_m \cdot e}{B_0} \cdot (\lambda - t) + 1\right]\right\}, \qquad (1)$$

where B is the cumulated CH_4 production ($L \cdot kg_{VS}^{-1}$) over digestion time (d), B_o is the maximum CH_4 production potential ($L \cdot kg_{VS}^{-1}$), λ is the lag phase (d), R_m is the maximum CH_4 production rate ($L \cdot kg_{VS}^{-1} \cdot d^{-1}$), t is time (d) and e is the Euler´s number. The model describes the cell density during microbial growth and is commonly used for simulations of CH_4 production in AD batch tests.

Single-factor analysis of variance (ANOVA) was conducted in Microsoft Excel and used for data analyses. Comparison of sample arithmetical means was done by binary t-tests between the reference sample DM and DM + L at $\alpha = 0.05$. The coefficient of determination R^2 was calculated to determine the accuracy of the modified Gompertz model.

3. RESULTS

3.1 Biogas and CH_4 production

The cumulated methane production over time is shown in Figure 1. Experimental maximum CH_4

Table 2: Experimental maximum CH_4 production during BMP tests and predicted parameters using the modified Gompertz model

Sample	Measured CH_4 production ($L \cdot kg^{-1}$-VS)	Predicted max. CH_4 production (B_0) ($L \cdot kg^{-1}$-VS)	Lag-Phase (λ)	max. CH_4 production rate (R_m) ($L \cdot kg^{-1}$-$VS \cdot d^{-1}$)	Model R^2
DM	260.6 ± 0.77	291.5 ± 3.38	5.757 ± 0.27	9.12 ± 0.13	0.9961 ± 0.00
DM + L	268.3 ± 2.57	275.0 ± 3.19	4.884 ± 0.06	12.28 ± 0.03	0.9971 ± 0.00

production during BMP tests and predicted parameters using the modified Gompertz model are shown in Table 2. High values of the coefficient of determination R^2 (> 0.996) indicate a good fit of the model to the experimental data.

The addition of lignite to the digestion of liquid dairy manure without an external inoculum accelerated the digestion process. However, final experimental methane volumes converge for samples with and without lignite (Table 2). The addition of 3.75 wt.-% lignite (DM + L) caused an increase in the measured specific methane production during anaerobic digestion of 2.96 vol.-% over 44 days compared to mono-digestion of liquid dairy manure (DM). Despite, using the modified Gompertz model the predicted maximum methane production potential decreased for lignite addition (-5.64%). This indicates, that lignite was not microbiologically degraded during AD, acting as an inert material. The modified Gompertz model showed an increase of the maximum CH_4 production rate during the anaerobic digestion for lignite addition of +34.7%. In addition, the lag-phase decreased by 15.17% for DM + L compared to DM. These results are consistent with previous studies concerning

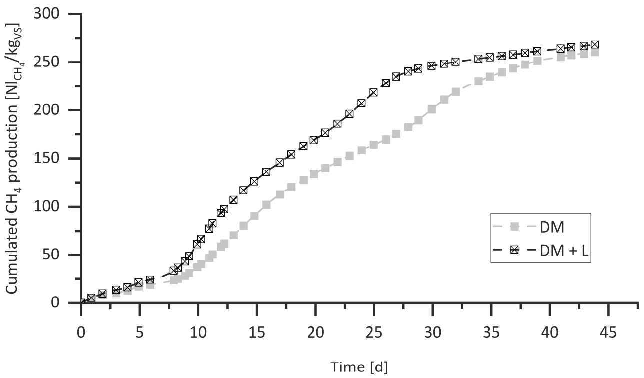

Figure 1: Experimental cumulated CH_4 production over time

the impact of biochar on the kinetics of anaerobic digestion [10,11], and show, that lignite can be used likewise in AD. Acceleration of anaerobic digestion caused by the addition of carbon-rich materials is favored by several reasons like their potential to act as catalysts in metabolism by increasing interspecies electron exchange [12] or stabilization of AD [13]. As shown in Figure 1 a diauxic degradation can be detected during anaerobic digestion, indicating either a two-step degradation of the liquid dairy manure or a light inhibition of the process [7]. Wijesinghe et al. [5] stated, that the addition of lignite at ≥70 g/L reduced ammonia inhibition of methanogenesis during anaerobic digestion of swine manure by lowering the free ammonia nitrogen (FAN) concentration. Hence, a light ammonia inhibition of the process might be avoided by lignite addition.

3.2 Fertilizer Properties

As an indicator for the production of slow-release organic fertilizers from digestate, the amount of total nitrogen in the dry solid phase was determined. The amount of TN in the dry solid phase depends on the loading of nitrogen in this phase as well as on the liquid-to-solid-phase ratio (LSR) of the digestate.

The addition of lignite to the AD of liquid dairy manure decreases the LSR of the digestate after centrifugation from (6.5/1) for DM to (4.5/1) for DM + L. This is mainly due to the high TS content of lignite. If TS of lignite is deducted from the solid phase, an LSR of (5.85/1) is reached. Hence, the change in LSR results not only from the TS of lignite, but an increased affiliation of digestate to the solid phase can be detected. Mumme et al. [14] stated the ability of biochar to support the formation of methanogenic microflora. Assuming similar effects of lignite, this could be a reason

Table 3: Load, phase distribution and total amount of TN

Sample	TN in solid phase (g/kg)	TN in liquid phase (g/l)	Solid phase in digestate (%)	Total amount of TN in dry solid phase (kg_N/t_D)
DM	35.54±0.04	1.962	13.30 ± 0.23	0.767 ± 0.002
DM + L	32.26±0.06	1.701	17.97 ± 3.41	1.499 ± 0.004

for the mass increase in the solid phase. The TN load in the liquid and solid phase decreases for lignite addition (Table 3). For the solid phase, this is mainly caused by the high carbon content in lignite (Table 1), increasing the C:N-ratio of the solid phase. Even though the TN load of the solid phase decreased by 9.36%, the absolute amount of nitrogen attached to the solid phase, which can be obtained from 1 t digestate after centrifugation, increased by 95.36% from 0.767 kg_N/t_D (DM) to 1.499 kg_N/t_D (DM + L) for lignite addition. Nitrogen concentration in the liquid phase decreased by 13.3% from 1.96 g/l (DM) to 1.70 g/l (DM + L), leading to an overall decrease of nitrogen in the liquid phase by -17.62% for DM + L. Hence, lignite addition during AD of liquid dairy manure leads to a nitrogen shift from liquid to solid phase, enabling a higher quantity of nitrogen recovery as solid organic fertilizer.

4. CONCLUSION

In this study, the addition of lignite during anaerobic digestion of liquid dairy manure was investigated. In a new approach, the impact on biogas production and the production of slow-release organic fertilizer was assessed simultaneously. It was found, that addition of lignite to AD of liquid dairy manure accelerates methane production during BMP tests and increases the amount of nitrogen in the solid phase after separation, which can be used for fertilizer production. Hence, by utilizing the benefits from both processes, biogas and fertilizer production, the economic viability of lignite addition can be increased. Nonetheless, further research is required to investigate optimal additive characteristics for both applications simultaneously.

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