



Siloxane removal and sludge disintegration using thermo-alkaline treatments with air stripping prior to anaerobic sludge digestion



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ABSTRACT

A thermo-alkaline treatment with air stripping was applied before anaerobic sludge digestion for both siloxane removal and sludge disintegration. The treatment was expected to increase the amount of biogas produced and to reduce the amount of siloxane in the gas. Adding sodium hydroxide (NaOH) to the sludge improved the removal of siloxane from the sludge, with approximately 90% of the siloxane removed to the gas phase using a thermo-alkaline treatment. Over 80% of decamethylcyclopentasiloxane (D5) could be removed under the following conditions: 55 °C treatment temperature, 135 g-NaOH kg⁻¹ volatile total solids (VTS), and 0.5 L min⁻¹ air-stripping rate. The disintegration ratio of volatile suspended solids (VSS) in the sludge was correlated with the D5 removal ratio. Because most of the siloxane was adsorbed to, or was contained in the VSS, the siloxane removal ratio increased with VSS disintegration. Finally, the energy consumption and operational costs of this system were evaluated for several scenarios. Thermo-alkaline treatment at the indicated operational conditions had the lowest operating costs for a 400 m³ day⁻¹ anaerobic sludge digestion system.

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1. Introduction

Biogas produced from sewage sludge and landfilled waste generally contains siloxanes, which can potentially cause problems during energy recovery operations in e.g. gas engines, micro-gas turbines, and fuel cells [1,2]. Siloxanes are organic silicon compounds used in various industrial processes as a replacement for organic solvents, in organic Rankin cycle as working fluids [3,4] and in consumer products [5], and a part of them are emitted to wastewater and condensed in sewage sludge. Biogas derived from sewage sludge eventually contains volatile siloxane compounds [6]. Siloxanes are moved into biogas from sewage sludge during methane fermentation because the boiling points are relatively low: 100–210 °C, and vapor pressures are relatively high: 27–5613 Pa at 25 °C in siloxanes for the molecular weight: 162–370 g mol⁻¹ [7].

Many technologies have been investigated and developed to reduce the siloxane concentration in the biogas, including adsorption, absorption, cryogenics (+pressurization), biological

degradation, catalytic decomposition, membrane separation, chemical abatement, and gas stripping (Table 1). It is only adsorption, absorption and cryogenics to be currently implemented in the above-mentioned techniques [8]. Cryogenic condensation of the siloxanes is possible, but the low temperature necessary to achieve a high removal efficiency is energy-consuming [9]. The absorption process involves transferring siloxane from the gas to the liquid phase. Although water cannot remove siloxanes from biogas [10], silicone oil [11] are very effective. However, these absorption technologies require large amounts of energy for the regeneration of the absorbent oil (heating or distillation) [11]. The chemical abatement of siloxanes is partially effective if sulfuric acid (H₂SO₄), hydrogen peroxide (H₂O₂) or dimethyldioxirane (DMDO) is used [12]. However, this method not only has costs associated with the use of chemicals but also generates health and environmental risks. The biological degradation of siloxanes has recently been considered as an affordable technology [13,14]. The removal efficiency is currently however low and only octamethylcyclotetrasiloxane (D4) has been used in experiments. Biogas adsorption methods using activated carbon, silica gel, alumina, and zeolites are the most commonly applied and implemented technologies and they have high removal efficiencies for siloxanes in biogas.

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Nomenclature

E	siloxane removal ratio, %	E_{ad}	electricity for anaerobic digestion, MJ day ⁻¹
R	siloxane recovery ratio, %	<i>Greek symbols</i>	
M_0	mass of siloxanes in the input sludge, mg	η	overall thermal efficiency of treatment tank
M_r	mass of siloxanes in the treated sludge, mg	ρ	specific gravity of the sludge, kg m ⁻³
M_a	mass of siloxanes collected on the adsorbent from the exhaust gas, mg	<i>Acronyms</i>	
VSE	VSS disintegration ratio, %	L2	hexamethyldisiloxane
VSS_0	VSS concentration of the input sludge, mg L ⁻¹	L3	octamethyltrisiloxane
VSS_r	VSS concentration of the treated sludge, mg L ⁻¹	L4	decamethyltetrasiloxane
Q	sludge input volume, m ³	L5	dodecamethylpentasiloxane
Q_v	heat value of biogas, MJ m ⁻³ -biogas	D3	hexamethylcyclotrisiloxane
C_w	evaporation coefficient of water, h ⁻¹	D4	octamethylcyclotetrasiloxane
C_p	specific heat of the sludge, MJ kg ⁻¹ K ⁻¹	D5	decamethylcyclopentasiloxane
T_1	treatment temperature, K	D6	dodecamethylcyclohexasiloxane
T_2	temperature outside the tank, K	ORP	oxidation reduction potential
L	amount of moisture evaporated from the sludge, kg day ⁻¹	TS	total solids
IT_1	evaporative latent heat of water at T_1 , MJ kg ⁻¹	SS	suspended solids
P_w	saturated vapor pressure at T_1 , kg m ⁻³	DS	dissolved solids
P_a	vapor pressure of air in the tank, kg m ⁻³	VTS	volatile total solids
A	surface area of sludge in the treatment tank, m ²	VSS	volatile suspended solids
V	biogas volume per sludge, m ³ -biogas m ⁻³ -sludge	VDS	volatile dissolved solids
H	heat requirement for pretreatment and mesophilic anaerobic digestion, MJ day ⁻¹	FTS	fixed total solids
H_r	recovered heat, MJ day ⁻¹	FSS	fixed suspended solids
H_v	total heat of biogas, MJ day ⁻¹	FDS	fixed dissolved solids
H_f	auxiliary fuel requirement, MJ day ⁻¹	DMDO	dimethyldioxirane
E_n	net production of electricity, MJ day ⁻¹	WWTP	wastewater treatment plant
E_a	additional electricity for pretreatment, MJ day ⁻¹	PE	population equivalent
E_g	gross production of electricity, MJ day ⁻¹	CHP	combined heat and power system
		LPG	Liquefied petroleum gas

The most favorable adsorbents and properties required to remove siloxanes have been suggested in previous studies [15–18]. However, most adsorbents are difficult to regenerate by heating because adsorbed siloxanes are polymerized to high-molecular-weight siloxanes, silicone, or decomposed into silica [15,18]. Moreover biogas contains not only methane, carbon dioxide, and siloxanes, but also high-molecular weight hydrocarbons (α -pinene, limonene, camphene, and decane) [19]. Some of the pore space of the adsorbents is consumed by compounds other than siloxanes. Because the cost of the replacement of the activated carbon is significant [9], reducing siloxanes prior to energy-application of the biogas is very important. This is also true for the use of catalytic decomposition [20] and membrane separation [21].

Ajhar et al. reviewed the use of gas-stripping technologies before digestion to reduce siloxanes in primary biogas, which has been investigated for volatile hexamethyldisiloxane (L2) on a lab scale, and concluded that gas stripping is technically feasible for siloxane removal, preferably in the stage between sludge heating and digestion [8]. Oshita et al. applied a thermal treatment (80 °C) with gas stripping before anaerobic digestion to remove siloxane from sludge and achieved 90% removal of decamethylcyclopentasiloxane (D5) from sewage sludge and a 1.6-fold increase in the biogas volume from the treated sludge as compared with the untreated sludge [7]. In this case, the siloxane concentration in biogas has been calculated to be lower than the limits of gas engine manufacturers reported by Wheless and Pierce [9]. Such a treatment could hence omit or minimize the adsorption process. However, the high energy consumption required to maintain heating at 80 °C is a disadvantage of this technique.

Various pretreatment methods have been developed that can disintegrate the organic content of sludge before anaerobic

digestion, such as sonication, ozonation, microwave or thermal treatments, and the addition of alkaline reagents [22–26]. Carrere et al. reviewed the pretreatment methods used to improve sludge anaerobic degradability and proposed combining a thermal treatment with alkaline addition to decrease the temperature of the thermal treatment [27]. Moreover siloxanes have low water solubility and can be adsorbed or contained in organic sludge solids [7], so more organic solids is dissolved by thermo-alkaline treatment, and siloxanes would become easy to be removed from sludge to a vapor phase with low energy.

In this study, an alkaline treatment was combined with a thermal treatment and air stripping before anaerobic digestion to reduce the heat energy required to remove siloxanes from sludge. Thermo-alkaline treatments and air-stripping experiments using sewage sludge were conducted to determine the optimal conditions for siloxane removal. The correlation between the removal of siloxanes and the disintegration of sludge was further estimated. Finally, an energy and operational cost analysis of the thermo-alkaline treatment and air stripping was conducted.

2. Materials and methods

2.1. Materials

The following eight types of siloxanes were used in the experiments reported here: L2 (99%), octamethyltrisiloxane (L3; 98%), decamethyltetrasiloxane (L4; 97%), dodecamethylpentasiloxane (L5; 97%), hexamethylcyclotrisiloxane (D3; 99%), D4 (99%), D5 (99%), and dodecamethylcyclohexasiloxane (D6; 99%). L2–L4 and D3–D6 were obtained from Shin-Etsu Chemical Co., Ltd. (Tokyo,

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