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Understanding the effects of the origin, occurrence, monitoring, control, fate and removal of siloxanes on the energetic valorization of sewage biogas—A review

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ABSTRACT

This paper reviews the effects of the origin, occurrence, monitoring, control, fate and removal of siloxanes on the energetic valorization of sewage biogas, which can be severely compromised by its volatile organic silicon compound (VOSiC) content. Almost 25 years after identifying silicon dioxide in the exhaust gases from engines powered using sewage and landfill gas, a wide range of studies have been conducted addressing the different stages of the siloxane life cycle. The cycle starts with the production and use of polydimethylsiloxane polymers in a wide range of industrial and domestic applications and its further dispersal into environmental compartments. Siloxanes are subsequently introduced into wastewater treatment plants, where as a result of their low biodegradability and high affinity to dissolved and particulate matter, they are first transferred from wastewater into sludge and later volatilized in biogas in anaerobic digesters. Biogas treatment technologies can reduce siloxane concentrations to less than 0.1 mg/m³; adsorbent materials with micro- and mesoporous structures appear to be the most relevant technology in technical and economic terms. The state-of-the-art on siloxanes is vast and extensive, but there are still some knowledge gaps to be addressed in the future, such as the standardization of the methodology for off-line analysis, the development of on-line monitoring equipment, better understanding the fates of siloxanes in wastewater treatment processes to operate at specific conditions to avoid siloxanes-related problems, the development of more selective and regenerative removal technologies from biogas to reduce operating costs and even to recover silicon, and better understand the detrimental effects on energy recovery technologies to determine the inlet concentration limits. This work compiles the most relevant results available in the literature for each stage of the siloxane life cycle.

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Contents

1. Introduction	367
2. Anthropogenic cycle of siloxanes: Production, uses and release into the environment	368
2.1. Classifications and definitions	368
2.2. Industrial and domestic uses of silicones and siloxanes	368
2.3. Releases of siloxanes into the environment	369
3. Environmental and health risk concerns of siloxanes	369
4. Quantification of siloxanes in environmental matrices: Air, water, soils, sediments, sludge and biogas	369
4.1. Quantification of siloxanes in gaseous matrices	369
4.2. Quantification of siloxanes in aqueous samples	370
4.3. Quantification of siloxanes in solid samples (soils, sediments, sludge)	371

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5. The fate of siloxanes in the urban/industrial wastewater cycle.....	371
5.1. The physico-chemical properties of siloxanes affecting their fates in WWTPs.....	372
5.2. Sewage treatment line.....	374
5.3. Sludge treatment lines and the final disposal sludge.....	375
6. Removal of siloxanes in the energetic valorization of biogas.....	375
6.1. Adsorption processes.....	375
6.2. Absorption processes.....	375
6.3. Refrigeration/condensation and deep chilling processes.....	375
6.4. Membrane separation processes.....	375
6.5. Biological degradation processes.....	376
7. Effect of the presence of siloxanes on biogas-powered energy conversion technologies: Siloxane limits and operating concerns for energetic valorization.....	376
7.1. Internal combustion engines (ICE).....	376
7.2. Micro-turbines.....	377
7.3. Fuel cells.....	377
7.4. Stirling engines.....	377
7.5. Domestic gas boiler.....	378
7.6. Siloxane limits in energy conversion systems.....	378
8. Conclusions, knowledge gaps and outlook.....	378
Symbols and acronyms.....	378
Acknowledgements.....	378
References.....	378

1. Introduction

Biogas produced in landfills and wastewater treatment plants (WWTPs) by microorganisms during the anaerobic degradation process of organic compounds is commonly used in energy production. Moreover, in several countries, methane-rich gas from landfills must be collected and burned or used for energy production to prevent the methane from being released into the atmosphere. For this reason and increasing interest in renewable fuels, biogas has become a notable alternative to conventional fuels in the production of electricity and heat. Indeed, biomethane, upgraded from biogas, has also become an interesting alternative for vehicle fuel.

Biogas contains mainly methane (from 40 to 60%) and carbon dioxide (40 to 55%); however, in the case of landfills, it may also contain nitrogen and oxygen [1]. Along with the main compounds, biogas may also contain trace compounds, such as hydrogen sulphide, halogenated compounds, and organic silicon compounds.

The energetic utilization of biogas is severely compromised by its volatile organic silicon compound (VOSiC) content. The scientific community has determined that VOSiCs in biogas are produced by the degradation and/or volatilization of organosilicon materials [1,2]. Organosilicons are a broad family of synthetic polymers with a wide variety of forms and uses. All of these polymers are manmade; the organosilicon linkage (the linkage between a silicon atom and a methyl group or any other organic group) is not found in nature. The organosilicon compounds present in biogas are oxidized during biogas combustion into microcrystalline silicon dioxide, a residue with chemical and physical properties similar to glass. Silicon dioxide deposits on valves, cylinder walls, and liners, cause abrasion and

blockage of pistons, cylinder heads, and valves. In gas turbines, siloxane deposits usually form on the nozzles and blades, causing erosion of the turbine blades and subsequently decreasing the operating efficiency [3]. Moreover, the glassy residues can deactivate the surface of the emission control system catalyst [1,3–5]. The use of spark ignition engines has increased the number of engine failures caused by siloxanes, as previous dual fuel engines were less vulnerable to silica deposits [6]. A correlation between increasing CO emissions and the build-up of silicates in engine oil after the combustion process was reported by Tower [3]. In this case, the oil in the engine needs to be changed more frequently, which resulted in the introduction of a 1 mg/L limit for silicon in the oil of gas engines by some gas engine manufacturers [7]. Furthermore, the deposited layers can break off and clog lines. Other undesired effects include the poisoning of the catalysts used in steam reforming [8] or fuel cells [9]. The catalysts used for both pre-combustion [10] and post-combustion gas purification (i.e., to reduce formaldehyde concentrations in exhaust gas) are also prone to deactivation by siloxanes. All of these negative effects are associated with increases in operating costs. Therefore, plant operators are facing a choice between either installing gas purification equipment or controlling the problems with more frequent maintenance (i.e., more frequent oil changes, engine inspections, downtime and associated loss of financial reimbursement [11]).

Reviews addressing the topic of “siloxanes” [4,6,12,13] are mainly focused on siloxane removal technologies, with minor references to quantification and occurrence issues. Within this context, no thorough study has been conducted compiling all of the information available in the literature at all stages of the siloxane life cycle. In this work,

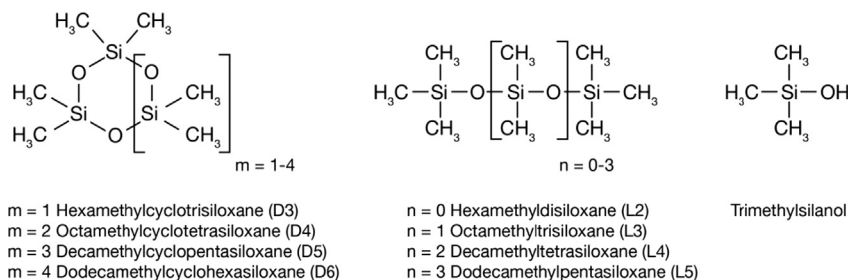


Fig. 1. Structures of common siloxane compounds, adapted from Schweigkofler and Niessner [1].

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