



# Impact of silica deposition on the performance of gas-fired domestic appliances caused by the combustion of siloxanes in the fuel

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## ARTICLE INFO

### Article history:

Received 27 September 2017

Received in revised form

19 July 2018

Accepted 30 July 2018

Available online 31 July 2018

### Keywords:

Biogas

Siloxane

Silica

Deposition

Appliances

## ABSTRACT

The effects of silica deposition resulting from the combustion of siloxane-containing biomethane on the performance of seven domestic appliances have been studied. The results show that silica deposition increases the flow resistance across the heat exchangers, which consequently reduces the air intake from the surroundings. For the flow-through hot water heater studied, the reduction in air intake in the appliance results in a substantial increase in the CO emission with time, ultimately resulting in a safety risk for the end user. For the fully premixed boilers examined, the increased flow resistance reduces the thermal input, and thus the thermal output, of the appliance. The extent of the reduction in thermal input is seen to depend strongly upon the concentration silicon in the fuel, and the configuration and material of the heat exchanger. For the boilers studied, the ion current measured by the flame safety device decreases with time because silica deposits on the probe used to measure the current in the flame; this ultimately results in boiler failure. Based on the results, extrapolation methods are proposed that can be used to assess the impact of low silicon concentrations in biomethane on appliance performance and safety with time.

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## 1. Introduction

The global drive towards sustainability has resulted in an increasing trend towards the use of biogas. An effective way to transport biogas to end users, both residential and industrial, is to inject the biogas into the existing natural gas grid after upgrading it to meet the natural gas specification, so-called biomethane. Depending on the point of injection of biomethane in the gas grid, end users who are located downstream of these points can receive up to 100% biomethane, especially in the summer when gas consumption is low.

Biogases from fermentation processes, for example from dairy waste, wastewater treatment or landfill, contain mainly CO<sub>2</sub> and CH<sub>4</sub>. However, depending on the source, biogas can contain varying quantities of trace contaminant species, such as halogenated compounds, sulfur compounds and volatile organic compounds,

including compounds like siloxanes, which are organosilicon compounds [1–3].

Siloxanes and other silicon-containing compounds that are found in many types of biogas deserve special attention. These compounds are present in many consumer products, such as detergents and shampoos, and ultimately can end up in the biogas produced. Volatile siloxanes present in the fermenting waste products can be present in the resulting biogas. The siloxane concentration found in biogases varies strongly depending on the biogas production/process conditions and the properties of the siloxanes present in the biomass. The largest concentrations of siloxanes, up to 400 mg/m<sup>3</sup>, are found in fermentation gases from landfill and wastewater treatment [2]. Siloxanes have either linear (L) or cyclic (D) structures. Different types of siloxanes have been reported from landfill gases [3]. The literature further discusses several techniques to remove siloxanes from biogas [2, 4–6].

Upon combustion, siloxanes are converted to silicon dioxide particles (silica, SiO<sub>2</sub>) [7–26], which deposit in combustion equipment such as engines, turbines and boilers [20–23]. In gas

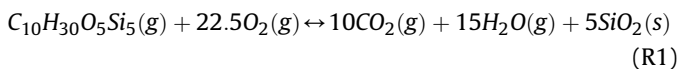
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appliances for heating and hot water production, silica particles may deposit on various surfaces like the heat exchanger and can cause deterioration in equipment performance [22,23]. Since domestic appliances tend to have compact heat exchangers, which can be relatively easily blocked, these appliances may be particularly susceptible to clogging by silica formation. Progressive clogging of heat exchangers leads to increased flow resistance in an appliance, which can result in a decreased thermal output and/or increased carbon monoxide (CO) emissions. For these reasons, domestic appliances can be considered among the most sensitive applications for silica clogging caused by siloxanes in biomethane. From the perspective of the end user, this deterioration is unwanted since no significant deterioration in performance should occur during the lifetime of the equipment when normally used with natural gas as a fuel. Moreover, any silica particles formed that are not deposited in combustion equipment are instead expelled into the air. Since combustion-generated silica particles smaller than 100 nm have been reported in the literature [see, for example, 7–11, 18], the particles exhausted could contribute to the environmental burden of ultrafine particles and potential health issues [22,23].

Given the growing trend towards the injection of biomethane into the natural gas grid, the development of a well-founded specification for the maximum amount of siloxanes in biomethane is necessary to guarantee appliance performance. Technical information needed to develop these specifications for domestic appliances is currently lacking. The goal of this study is to determine a hierarchy of impacts of the silicon content in biomethane on the performance of commonly used domestic appliances, varying from “nuisance” effects to the potential impairment of safe operation. Moreover, this study provides insight to the extent to which the impact of silica deposition on appliance performance can depend on appliance design, such as the configuration and material of heat exchanger present and on the performance of flame safety devices [20–23]. To maximize the generality of the results while avoiding large scale testing the results obtained are used to develop methods based on extrapolation that can be used to assess the impact of low silicon concentrations in biomethane on appliance performance over long periods of time, i.e., approaching the 15–20 year lifetime of domestic appliances. These results can in turn be used to estimate maximum siloxane concentrations in biomethane that maintain appliance performance.

## 2. Silica particle formation in siloxane-containing flames

In this study, siloxane-containing biomethane is simulated by doping different quantities of siloxanes in natural gas. The siloxane decamethylcyclopentasiloxane ( $C_{10}H_{30}O_5Si_5$ , often simply referred to as D5) is used in the experiments. As an illustration, the overall oxidation reaction of D5 is shown below (R1).



It should be noted that reaction R1 is an overall representation of a chain of chemical reactions [12,17], similar to those occurring in hydrocarbon oxidation. Equilibrium calculations indicate that silicon-containing combustion products are mainly present as gaseous silicon oxide, SiO and silica, SiO<sub>2</sub>, both in gaseous and condensed phases depending on the temperature. As an example, the calculated equilibrium fraction of silicon oxides present in the gas phase for a methane/air mixture doped with D5 with an equivalence ratio of  $\phi = 0.7$  (typical for modern lean-premixed central heating boilers) is calculated as a function of temperature and D5 concentration and presented in Fig. 1. In these calculations,

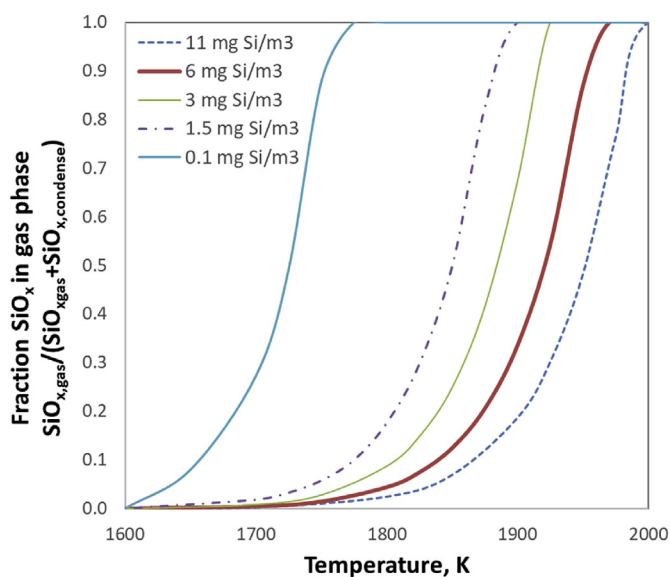


Fig. 1. Equilibrium fraction of gas-phase silicon oxides (SiO<sub>x</sub>) for different silicon content in the fuel, as a function of temperature at an equivalence ratio of  $\phi = 0.8$ .

the siloxane concentration is given in mg Si/m<sup>3</sup>n in the fuel. The fraction of silicon oxides in the gas phase is calculated as the sum of the mole fractions of SiO and SiO<sub>2</sub> (together denoted as SiO<sub>x</sub>) in the gas phase divided by the total fraction of SiO<sub>x</sub> in the gas and condensed phases. Since different siloxanes can have both different numbers of silicon atoms and different molar masses, two different siloxanes at equal mole fraction can yield different masses of silica when burned. Expressing siloxane concentration as mg Si/m<sup>3</sup> of fuel, as opposed to mole/volume fraction (in ppm) or mg siloxane/m<sup>3</sup> fuel, provides an unambiguous measure of the potential to form silica during combustion. Fig. 1 shows that the fraction of silicon oxides in the gas phase depends strongly upon the temperature and concentration of silicon present in the combustion products. Therefore, we expect these parameters to affect the deposition processes of silica in domestic appliances, and their subsequent impact on appliance performance, when using silicon-containing fuel.

While the thermodynamic calculations accurately describe the final state of a system, which includes chemically reacting components, it is possible to observe deviation from the equilibrium when the characteristic times of the chemical reactions driving the system to equilibrium are of the same order as, or larger than, the characteristic residence times in a combustion chamber. In the reaction zone of the flame, siloxanes are quickly converted to gas-phase SiO and SiO<sub>2</sub> creating a supersaturated vapor, which proceeds to condense, forming nanoparticles or nanoclusters [e.g. 7–11, 18]. At high temperatures, the nanoclusters will continue to grow into larger clusters by taking up free SiO<sub>2</sub>(g) molecules. These clusters, which are small particles with a diameter in the order of a few nanometers, become larger through collisions between SiO<sub>2</sub> particles. Depending on the temperature, the morphology of these new particles can be either spherical (clusters) or tightly attached fractal-like structures (“aggregates”) [8–11,26]. Further downstream the temperature decreases due to heat losses, particularly during heat exchange in the appliance, resulting in the condensation of more SiO<sub>2</sub> from the gas phase and continued particle and aggregate growth, as well as the formation of agglomerates of aggregates. All these forms of condensed silica will deposit, particularly via thermophoresis [22], in the appliance, for example the heat exchanger [22].

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