Biotrickling filtration as sustainable technology for biogas upgrading to renewable fuel

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Introduction
Anaerobic digestion has proved to be one of the most suitable processes to treat the high amounts of sludge generated in wastewater treatment plants. Its digestion in the absence of O2 leads to the generation of biogas, mainly composed by CO2 (15-50%) and CH4 (35-70%), which can be transformed into heat or electricity, it can be injected into the gas grid or can be used as a vehicle fuel. Depending on the final use of biogas, upgrading processes will be required prior to the energy conversion unit. Besides H2S, siloxane removal is mandatory in most biogas applications due to the formation of abrasive SiO2 during biogas combustion.

The most widely used technology to abate such compounds is adsorption onto activated carbon despite the high operational costs associated to the frequent carbon bed replacements. Alternatively, biological technologies present lower footprints, among other advantages such as lower investment and operation costs. Biotrickling filters (BTF), where the impurities of the gas stream are biodegraded by a biofilm grown attached to a packing bed and provided with nutrients by a trickling solution, have been successfully applied for odour abatement and waste gas treatment (Estrada et al., 2012).

The application of this sustainable technology for biogas upgrading is currently under research. The scarce existing publications have studied siloxane removal in air streams, i.e. using O2 as the electron acceptor, and reported 10-20% removal efficiencies (RE) of hexamethyldisiloxane (Accettola et al., 2008), and a 43-60% RE of octamethyldisiloxane (D4) (Popat and Deshusses, 2008), depending on the empty bed residence time (EBRT). However, O2 composition in biogases is limited if any [ref].

The present work aims at investigating the removal of siloxanes from a synthetic biogas in an anaerobic biotrickling filter and evaluate the influence of the operation conditions (i.e. EBRT) and the presence of VOCs usually found in real biogas.

Materials and methods
The lab-scale biotrickling filter (Figure 1) was packed with inert lava rock as biofilm support and inoculated with anaerobic digestor sludge from an urban wastewater treatment plant. Synthetic mineral medium, containing NO3-, other nutrients and vitamins, recirculated from an external reservoir at 13 cm h-1 sprayed from the top of the column. The feed gas was regulated with a mass flow controller, where the target compounds were injected by a syringe pump to a N2 stream.

The BTF was firstly operated with D4 at 62 mg m-3 as carbon source provided in the feed gas at an EBRT of 14.5 min.

On the second research stage, D4 in the feed gas was switched for a multicomponent mixture of D4, decamethylcyclopentasiloxane (D5) and VOCs (hexane, toluene and limonene) commonly found in biogases (Papadias et al., 2011), at the concentrations described in Table 1. After the operation of the BTF, samples of the lava rock were obtained for analysing the microbial communities.

Results and discussion
During the first period of the BTF operation, the concentration of D4 was monitored for over 30 days to evaluate the acclimation of the inoculum to siloxanes. The low water solubility of this compound (Table 1) limited its mass transfer to the biofilm. However, an average RE of 14% was achieved. At this point, lava rock within the

Table 1. Water solubility and concentrations in the BTF feed gas for each compound

<table>
<thead>
<tr>
<th>Compound</th>
<th>Water solubility [mg L-1]</th>
<th>Feed gas concentration [mg m-3]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hexane</td>
<td>9.5</td>
<td>375</td>
</tr>
<tr>
<td>Toluene</td>
<td>526</td>
<td>24</td>
</tr>
<tr>
<td>Limonene</td>
<td>13.8</td>
<td>220</td>
</tr>
<tr>
<td>D4</td>
<td>0.056</td>
<td>54</td>
</tr>
<tr>
<td>D5</td>
<td>0.017</td>
<td>102</td>
</tr>
</tbody>
</table>

Figure 1. Lab-scale anaerobic biotrickling filter setup
reactor was observed in scanning electron microscopy. SEM images (Figure 2) revealed the presence of a high number of microbial cells attached in the surface of the packing rock.

Within the second period, the performance of the bioreactor for each compound in the mixture was monitored and the influence of the EBRT was evaluated and is displayed in Figure 3, where removal efficiencies are plotted against the EBRT tested. Limonene and toluene REs above 99% were quickly obtained with 14.5 min of EBRT. Decreasing this parameter to 4.1 did not influence the capacity to remove these compounds, demonstrating fast biodegradation rates. In the contrary, hexane’s elimination did not improve with longer EBRT, remaining between 10 and 13.

Concerning siloxanes, a positive relationship between their RE and the EBRT was obtained, both for D4 and for D5. D4 was removed in an average 21% at 14.5 min. Dropping the contact time to 7.2 and 4.1, REs decreased to 13 and 6%, respectively. The results obtained suggest that cometabolism of VOCs and methylsiloxanes as carbon sources increased the elimination capacity towards siloxanes.

DNA extractions from the lava rock samples revealed different species belonging to the phylum Actinobacteria and Proteobacteria. Among the 18 isolates obtained, some of the more representative species identified were Pseudomonas aeruginosa, Arthrobacter sp. and Microbacterium hydrocarbonoxydans, which have been previously observed capable to degrade siloxanes in soils and/or aromatic hydrocarbons (Sabourin et al., 1996).

**Figure 2.** SEM pictures of the lava rock after operating the BTF with D4 as carbon source.

**Figure 3.** EBRT influence on the removal efficiency of the target compounds during the multicomound operation stage of the BTF

Conclusions

This work constitutes the first investigation on biotrickling filter in anaerobic conditions exploring the removal of siloxanes in the presence of other VOCs for biogas upgrading. This study proved the potential of BTFs to reduce the concentration of siloxanes while removing completely other VOCs without hindering siloxanes abatement.

When the final use of biogas requires complete siloxane removal, a secondary treatment with activated carbon would be required for polishing the BTF effluent. However, the complete VOC removal accomplished by the BTF together with the considerable siloxane abatement achieved would extend the lifetime of the activated carbon beds and thus the frequent deposition of wasted activated carbon and its associated operational costs.

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References


