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Enhancement of polycyclic aromatic hydrocarbons (PAHs) removal during anaerobic digestion of sludge thanks to the addition of graphite felt

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Abstract
Carbon-based materials have been recently shown to improve interspecies electron transfer thereby increasing methane production in anaerobic digestors. Our study deals with the use of such conductive materials during anaerobic digestion of sewage sludge and the assessment of their impact on organic contaminant removal, PAHs being the model compounds. Our results not only demonstrate that graphite felt acts positively on dry matter (DM) hydrolysis but also on PAH removal. Indeed, DM and PAHs were removed 20% more with graphite felt than without. Abiotic control reactors revealed that volatilization and sorption are minor processes implied in such removal. The presence of graphite felt in anaerobic digestion could improve the bioavailability of PAHs (even those with high molecular weight) as indicated by an increase of PAHs concentration in the aqueous phase. Thus, graphite felt addition could activate PAH degradation.

Keywords
Conductive materials, bio-electrochemical systems, polycyclic aromatic hydrocarbons

INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are recognized as persistent and toxic organic pollutants that are hardly eliminated during wastewater treatment. They preferentially attach to hydrophobic organic matter with concentrations at µg-mg/kg DM in sewage sludge (Mailler et al., 2014).

Anaerobic digestion (AD) can partly degrade PAHs but performances have been limited by bioavailability of the compounds in the aqueous phase (Barret et al., 2010) and the absence of terminal electron acceptors (Meckenstock and Mouttaki, 2011).

Conductive materials have been found to stimulate the indigenous microorganisms by enhancing direct interspecies electron transfer (Zhao et al., 2016). In our study, the use of graphite felt or bioelectrochemical systems (BESs) with graphite felt electrodes to treat PAH-contaminated sewage sludge is reported for the first time.

MATERIALS AND METHODS

Three anaerobic batch digesters, fed with secondary sludge spiked with 13 PAHs (1-5 mg/kg DM) were run for a period of 42 days parallel to five reactors that either contained graphite felt as conductive material (3x) or the same amount of graphite felt to constitute two electrodes in one single chamber at closed circuit with an applied potential of 0.8V vs. Standard Hydrogen Electrode (SHE) (2x). Similar reactors were run on the same spiked sludge that was sterilized (2 cycles at 120°C, 20 min.).

Operating conditions were the same for all reactors (pH 7, 37°C, 650 rpm). Measurement of biogas quantity and composition was determined all along the experiment whereas PAHS and DM in the particulate and total phases were quantified at the beginning and at the end of the experiment. PAHs were extracted from the sludge matrix by accelerated solvent extraction (ASE 200, Dionex)

and analysed by HPLC with a fluorimetric detector (Waters 2475).

RESULTS AND CONCLUSIONS

Total solids (TS) and volatile solids (VS) removals were enhanced by 20% in the graphite felt reactors in comparison to the control digesters (Figure 1).

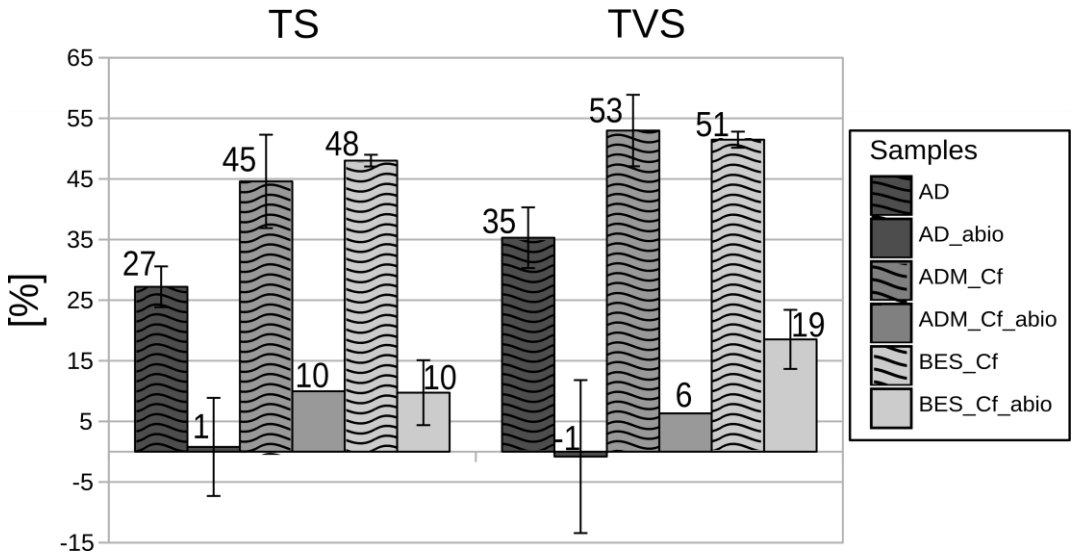

Figure 1. Removal of total solids (TS) and total volatile solids (TVS) during anaerobic sludge digestion (AD=anaerobic digester, ADM_Cf= graphite addition to AD, BES_Cf= graphite felt addition with current application, abio= abiotic controls).

The beneficial effect of carbonaceous material to interspecies electron transfer and bio-electricity generation was demonstrated recently (Chen et al., 2014; Li et al., 2016) and is probably based on enhanced direct interspecies electron transfer (DIET). Our results not only demonstrate that graphite felt acts positively on DM removal but also on PAHs with 20% more removal (Figure 2).

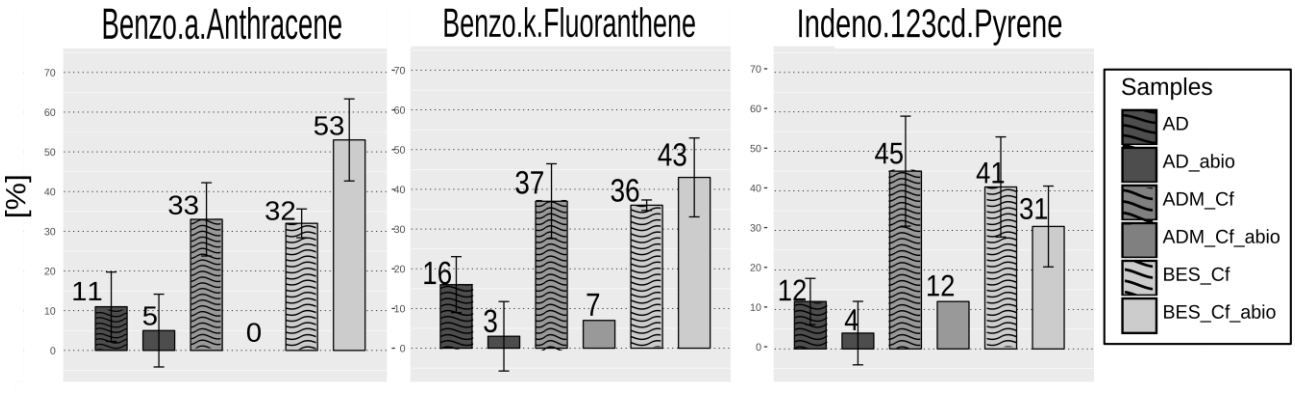

Figure 2. Total removal of indeno(123cd)pyrene, benzo(k)fluoranthene and benzo(a)anthracene in the aqueous phase after anaerobic digestion of municipal sludge. (AD=anaerobic digester, ADM_Cf= graphite felt addition to AD, BES_Cf= application of current to ADM_Cf, abio= abiotic controls).

Bioelectrochemical removal of DM and PAHs

The action of BES on sludge digestion is known to enhance organic matter degradation and methane yield (Zhao et al., 2014), yet, a beneficial effect of BES on sludge digestion and PAH removal could not be proven in this study. Polarized graphite felt electrode reactors did not perform

better than the reactors with graphite felt material alone BES_Cf compared to ADM_Cf (Figure 2). The reason could be that 0.8V of applied potential damages cytochrome c proteins on the outer anaerobes' membrane (Schröder et al., 2015) and disturbs further stimulation of electrode interacting microbial communities.

Volatilization and sorption are minor processes

Abiotic control reactors AD_abio and ADM_Cf_abio showed little organic matter (VS, TS) and PAH reduction which indicates that volatilization and sorption on the graphite felt material are minor mechanisms by which PAH are removed from sludge.

Electrochemical removal of PAHs

The abiotic control with electrochemical treatment BES_Cf_abio showed high removal of PAHs that exceeded biotic removal of the graphite felt treatment for 4 to 5 rings PAHs. It suggests that the experimental conditions (dead biomass after autoclave treatment, the presence of graphite felt and current application) favoured the electrochemical removal of PAHs (Figure 2.)

Mechanism of PAH removal by graphite felt

At the end of the experiment, graphite felt reactors ADM_Cf exhibited 24, 31 and 37 % more PAHs Benzo(a)[…],Benzo(k) […], Indeno[…] in the aqueous phase than digesters without graphite felt addition. BES_Cf reactors showed the same trend (Figure 3).

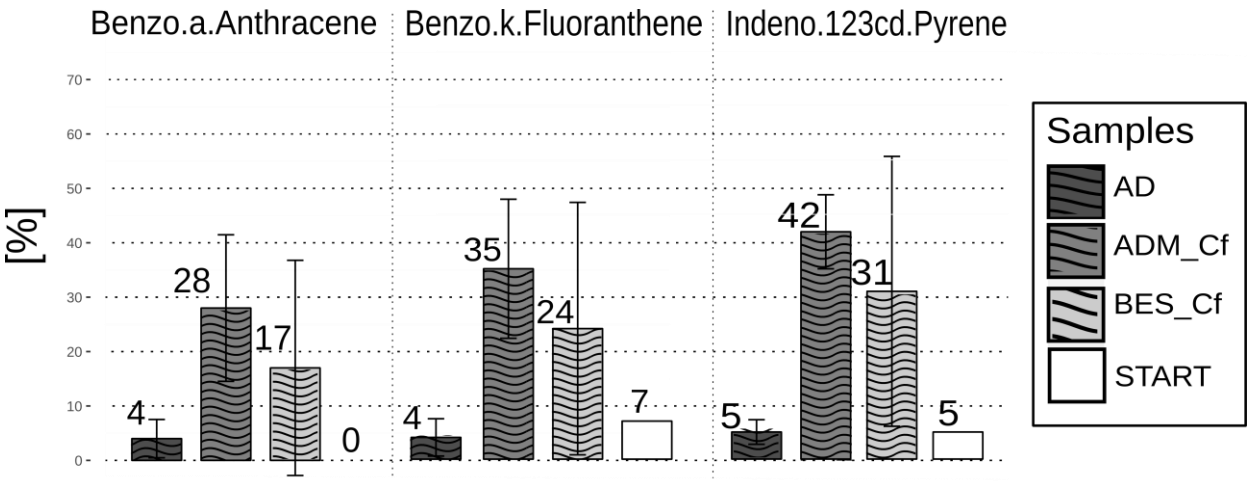

Figure 3. Presence of indeno(123cd)pyrene, benzo(k)fluoranthene and benzo(a)anthracene in the aqueous phase after anaerobic digestion of municipal sludge. (AD=anaerobic digester, ADM_Cf= graphite felt addition to AD, BES_Cf= application of current to ADM_Cf)

Apparently, the addition of graphite felt to anaerobic digesters relocated organic matter and PAHs from the particulate to the aqueous phase. DIET can help explain why graphite felt enhanced DM and PAH removal since mediatorless electron exchange between the conductive material and the anaerobic synthrophic community may facilitate hydrolysis. Carbonaceous conductors such as graphite felt seem to circumvent two limitations of bioremediation of PAHs, the lack of terminal electron acceptors as well as PAHs' limited bioavailability. Graphite felt could be considered a permanent solid electron exchange facility and may trigger the solubilisation of particulate matter in anaerobic sludge digestion, thereby rendering PAH more bioavailable.

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