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Can graphene oxide enhance methane production and pharmaceutical removal under anaerobic digestion?

Oriol Casabella-Font[1,2] *, Jelena Radjenovic [1,3], Maite Pijuan [1,2]

[1] Catalan Institute for Water Research (ICRA), C. Emili Grahit 101, 17003 Girona, Spain
[2] University of Girona, Girona, Spain
[3] Catalan Institution for Research and Advanced Studies (ICREA), Passeig Lluís Companys 23, 08010 Barcelona, Spain

*Corresponding author: ocasabella@icra.cat

Introduction

- The occurrence and destiny of pharmaceutical compounds in wastewater are a big concern for their environmental impact.
- Anaerobic processes exhibit slow reaction rates, prompting research efforts to enhance their efficiency and effectiveness in wastewater treatment.
- The anaerobic digestion process produces methane that can be recovered as renewable energy source.
- The phenomenon of direct interspecific electron transfer (DIET) within anaerobic environments is being investigated.
- The addition of graphene oxide is being explored to enhance the direct interspecific electron transfer, offering potential advancements in wastewater treatment strategies.

Methodology

- The methane production and the removal kinetics of four pharmaceutical compounds (i.e., sulfamethoxazole (SMX), trimethoprim (TMP), iohexol (IHX), and carbamazepine (CBZ) were monitored.
- The addition of 0.075 g of graphene oxide (GO), and hydrothermally reduced graphene oxide (rGO) per g of volatile solids was studied.
- Two sets of experiments were prepared:

Set 1:
- Batch methodology.
- On-line methane monitoring.
- Pharmaceutical removal kinetics

Set 2:
- Fed-batch methodology (three sequential batches).
- Online methane monitoring.
- Pharmaceuticals removal kinetics thereafter the third batch.

Results

Set 1

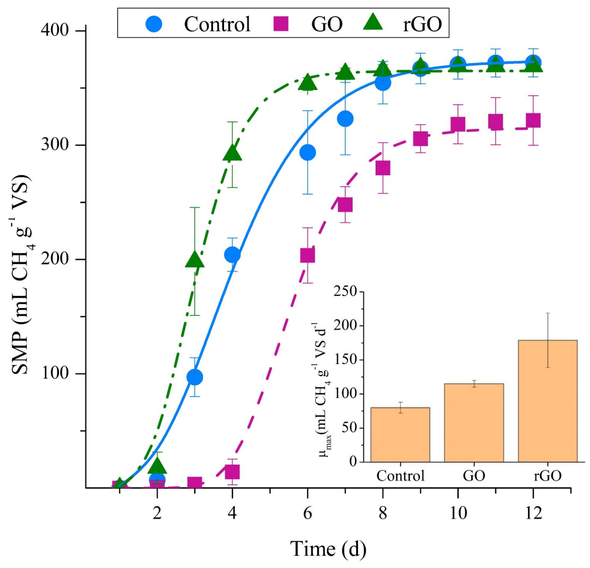

Figure 1: Experimental specific methane production (SMP) for each condition tested (symbols) and Gompertz model curve (lines) obtained during the **Set 1**. Representation of the specific methane production rates (μ_{max}) with standard deviation for each condition.

- Contrary to rGO, the addition of GO significantly reduced the maximum specific methane production (M_∞) by ~15% compared with the control.
- The maximum methane production rate (μ_{max}) increased by ~40% (GO) and ~120% (rGO), compared with the maximum production rate obtained in the Control.
- The lag phase (λ) increased by adding GO, doubling the estimated value compared with the Control and rGO conditions, probably caused by the biological reduction process of GO.

Set 2

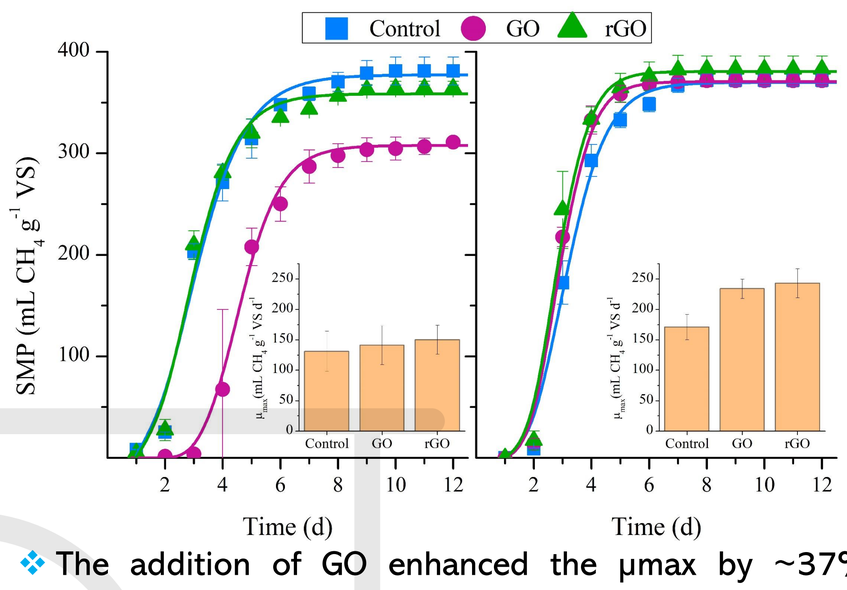

Figure 3: Experimental specific methane production (SMP) for each condition tested (symbols) and Gompertz model curve (lines) obtained during the first and the third batch of the **Set 2**. Representation of the specific methane production rates (μ_{max}) with standard deviation for each condition.

- The addition of GO enhanced the μmax by ~37% compared to the Control.
- The μ_{max} was not significantly enhanced during the different batches.
- Non-significant differences ($p < 0.05$) were observed between GO and rGO maximum specific methane production rate.
- The μ_{max} values reached ~140 mL CH4 g^{-1} VS d^{-1}.

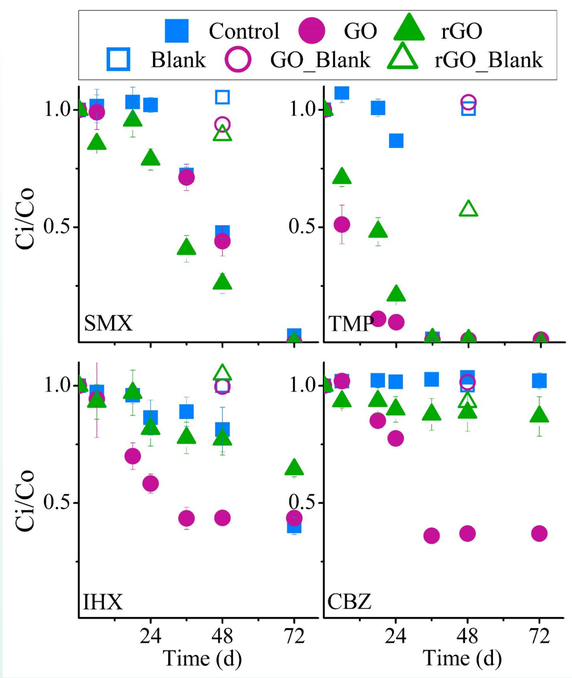

Figure 2: Kinetic removal profiles for sulfamethoxazole (SMX), trimethoprim (TMP), iohexol (IHX), and carbamazepine (CBZ).

- CBZ presented the highest removal enhancement by the addition of GO.
- TMP transformation was highly influenced by the addition of GO and rGO, with removals of 90±2% and 79±2% in just 24 hours.
- SMX was stable during the first 20 hours. After that point, SMX was removed with the same rate in all three conditions tested.
- The sludges that contained GO reduced IHX's initial concentrations by 60%.

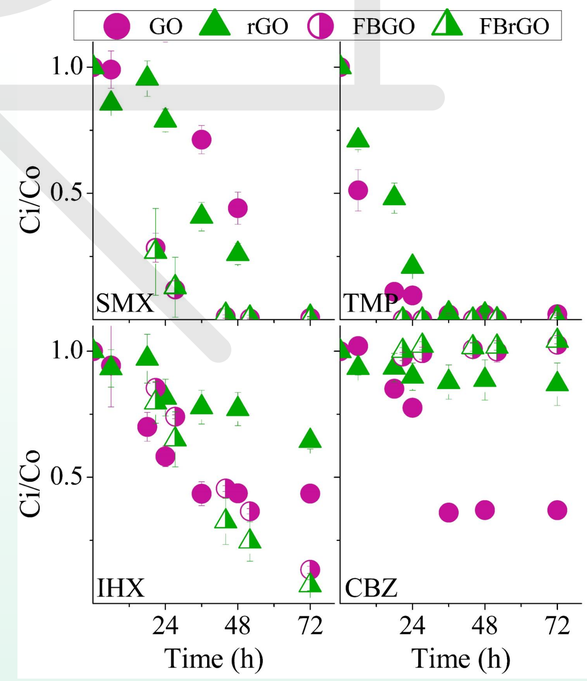

Figure 4: Kinetic removal profiles comparison between the first and the second set of experiments for sulfamethoxazole (SMX), trimethoprim (TMP), iohexol (IHX), and carbamazepine (CBZ).

- Although non-adapted sludge presented high removals in the presence of GO (Figure 2), adapted sludge (Figure 4) was not able to remove CBZ, as well as the other adapted sludge.
- TMP was removed in less than 18 hours, and SMX in 36 hours, much faster than in the first experimental set (Figure 4).
- Contrary to non-adapted sludges, IHX was ~90% removed from the liquid phase after 90h.

Conclusions

- Although the addition of graphene oxide negatively affects on the methane production in the first contact, after a short adaptation period the maximum methane production rate was enhanced by ~37%.
- The addition of graphene oxide enhances the removal of pharmaceutical under anaerobic conditions, and for three out of the four pharmaceuticals monitored, the adaptation to the conductive material positively affected to the removal kinetics.
- The highest kinetic constant in the presence of GO were associated to the DIET phenomena.

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