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A MATHEMATICAL MODEL DESCRIBING DENITRIFICATION COUPLED TO AEROBIC METHANE OXIDATION

Oskar Modin*, Soroush Saheb Alam, Britt-Marie Wilén

Chalmers University of Technology, Department of Civil and Environmental Engineering,
Division of Water Environment Technology, Bioresource Labs, Gothenburg, Sweden
*Contact: oskar.modin@chalmers.se

1. Introduction

Methane could potentially be used as external carbon source for denitrification of wastewaters or leachates. There are methanotrophic denitrifiers (e.g. Kampman et al. 2014); however, they are slow-growing and difficult to enrich. In this study, we instead focus on aerobic methane oxidation coupled to denitrification.

2. Aerobic methane oxidation coupled to denitrification

The process is carried by aerobic methanotrophs that oxidize methane and release soluble microbial products, which are organic substances that can be used by co-existing facultative heterotrophs for denitrification (see Fig. 1 below).

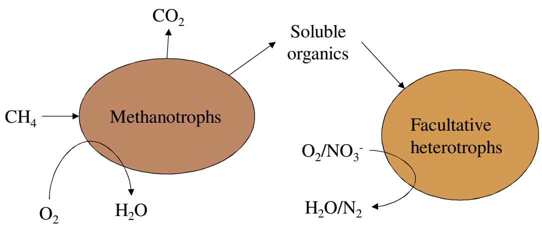

Fig. 1. The aerobic methane oxidation coupled to denitrification process.

3. Goal of the study

Aerobic methane oxidation coupled to denitrification has been investigated in several studies using different types of reactor systems. Membrane biofilm reactors have been shown to be especially effective (Modin et al. 2008). Methane (and optionally oxygen) are supplied from the interior of a membrane, to a biofilm growing on the membrane surface (Fig. 2).

The goal of this study is to develop a mathematical model describing aerobic methane oxidation coupled to denitrification, and use the model to investigate how factors such as intramembrane partial pressures of methane and oxygen affect nitrate removal in the process.

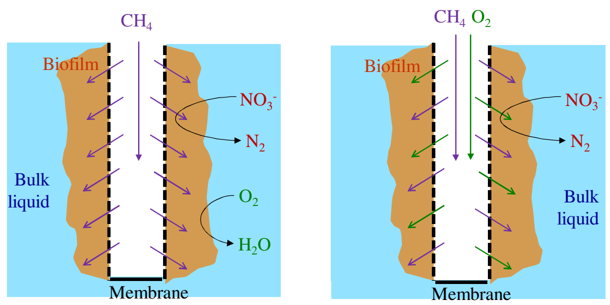

Fig. 2. Membrane biofilm reactor designs for aerobic methane oxidation coupled to denitrification.

3. Mathematical biofilm model

Components

The model considers three biomass components (X) and five soluble components (S):

- Methanotrops (X_M), Facultative heterotrophs (X_H), Inert biomass (X_I)
- Methane (S_M), Oxygen (S_O), Soluble organics (S_C), Nitrate (S_{NO}), Ammonium (S_{NH})

Processes

The turnover of biomass- and soluble components is modelled by six biochemical reactions:

- Growth and substrate utilization by (1) methanotrophs and (2) heterotrophs
- Endogenous respiration by (3) methanotrophs and (4) heterotrophs
- Hydrolysis of (5) methanotrophic biomass and (6) heterotrophic biomass

Algorithm

The model was solved for a 1-dimensional polar coordinate system simulating the development of a biofilm on a tubular membrane (Fig. 3). Transport of soluble components was described using Fick's law of diffusion. The total biomass density was set at a constant value and biomass was redistributed between neighbouring biofilm layers if this value was exceeded.

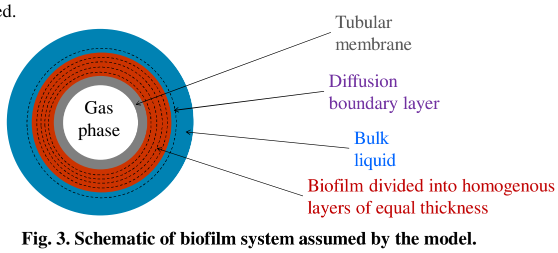

Fig. 3. Schematic of biofilm system assumed by the model.

4. Results of model simulations

We used the model to simulate the growth of a biofilm on a tubular silicone membrane (ID=0.5mm, OD=1.0mm) for a time period of 35 days. The effect of changing the intramembrane partial pressures of CH_4 and O_2 between 21 and 42 kPa was investigated.
If both CH_4 and O_2 are fed from the interior of the membrane at 21 kPa, an anoxic layer is formed in the outer part of the biofilm and denitrification accounts for 82% of the total nitrate removal rate, which is 0.29 mol/m²·d. If the intramembrane oxygen pressure is increased to 42 kPa, the entire biofilm is aerobic and denitrification only accounts for 23% of the total nitrate removal rate, which is 0.07 mol/m²·d. (Note that the calculation of denitrification also includes NO_3^- assimilation by facultative heterotrophs)

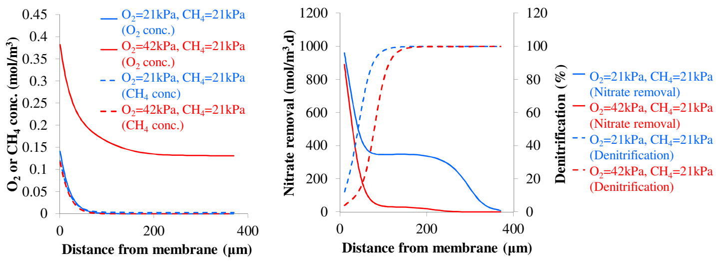

Fig. 4. (Left) CH_4 and O_2 concentrations, and (right) nitrate removal rate and fraction nitrate removed by denitrification at different biofilm depths. Two simulations with varying partial pressure of O_2 are shown.

If O_2 is fed from the bulk liquid instead of the membrane, the concentration profiles change significantly (Fig. 5). The interior of the biofilm becomes anoxic and allows denitrification to take place.

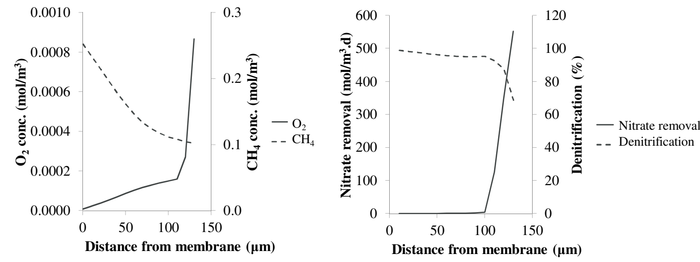

Fig. 5. (Left) CH_4 and O_2 concentrations, and (right) nitrate removal rate and fraction nitrate removed by denitrification at different biofilm depths. Simulations with O_2 in the bulk liquid.

5. Comparison to experimental data

We used data from Modin et al. (2010) who operated a membrane biofilm reactor for 37 days with both CH_4 and O_2 fed from the interior of the membrane. The first 23 days the CH_4/O_2 partial pressures were 24/46 kPa. From day 23 to 37 the partial pressures were 37/78 kPa. Although the biofilm thickness is similar to the experimental values, the predicted nitrate removal is initially significantly lower than the measured values in the initial period of the run.

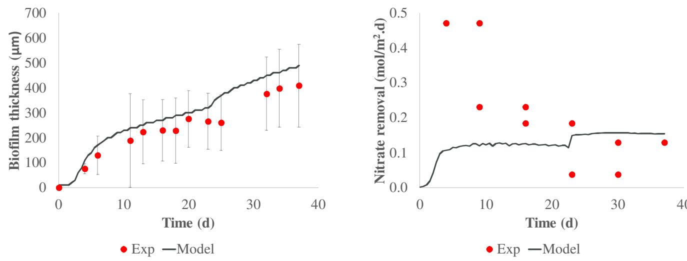

Fig. 6. Experimentally measured (Modin et al. 2010) and modelled biofilm thickness (left) and nitrate removal rate (right).

6. Discussion and conclusions

This is the first attempt to develop a mathematical model describing the aerobic methane oxidation coupled to denitrification process. Here, we have used the model to investigate the effect of different gas supply regimes on nitrate removal and denitrification in membrane biofilm reactors. The model could potentially also be used to examine different types of reactor setups and to investigate the microbial interactions in the process in greater detail (e.g. to quantify the carbon transfer between methanotrophs and heterotrophs). When the model was applied to experimental data it underpredicted nitrate removal rates despite realistic prediction of biofilm thickness. Further work is needed to analyze the sensitivity of various model outputs to the input parameters.

References

Kampman C., Temmink H., Hendrickx T. L., Zeeman G. and Buisman C. J. (2014). Enrichment of denitrifying methanotrophic bacteria from municipal wastewater sludge in a membrane bioreactor at 20 degrees C. *J Hazard Mater* **274**, 428-35.
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