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Redox Stratification Controlled Biofilm Reactors For Completely Autotrophic Nitrogen Removal

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1. Towards a novel reactor technology

Growing biofilms on oxygen permeable membranes, whereby oxygen supply to the bottom part of the biofilm can be easily controlled, can create redox stratication in the biofilm and, subsequently, micro niches for different bacterial communities which can perform simultaneous oxidation and reduction of pollutants from wastewater. These Redox-Stratified Controlled Biofilm Reactors (ReSCoBiR) are a promising technology for stable completely autotrophic nitrogen removal.

Aerobic Ammonium Oxidizing Bacteria (AOB) ***[1]***:

$$NH_4^+ + 1.38\,O_2 + 1.98\,HCO_3^- \rightarrow 0.018\,C_5H_7NO_2 + 0.98\,NO_2^- + 0.98\,NO_2 + 1.89\,CO_2 + 2.93\,H_2O$$

Anaerobic Ammonium Oxidizing Bacteria (AnAOB) ***[2]***:

$$NH_4^+ + 1.32\,NO_2^- + 0.13\,H^+ + 0.066\,HCO_3^- \rightarrow 0.066\,CH_2O_{0.5}N_{0.15} + 1.02\,N_2 + 0.26\,NO_3^- + 2.03\,H_2O$$

Modeling studies have confirmed that counter-diffusion biofilm (in which substrates are supplied from both sides of the biofilm) are more advantageous for completely autotrophic nitrogen removal than a conventional co-diffusion biofilms ***[3]***. Thus, this configuration will be the object of the present study.

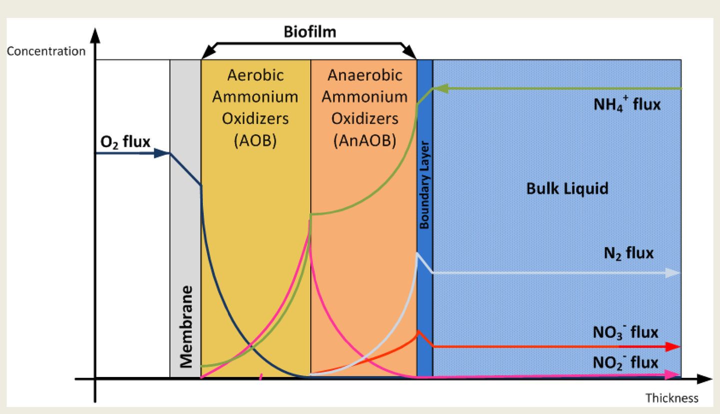


***FIGURE 1**: Substrate fluxes in counter-diffusion ReSCoBiR*

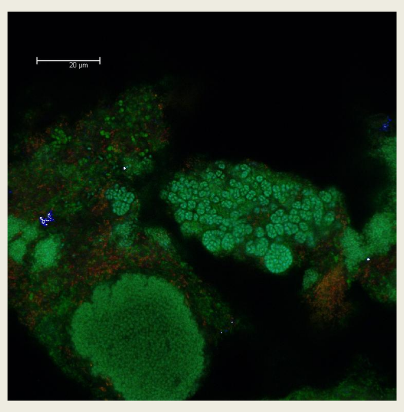


***FIGURE 2**:* FISH image of biofilm (Cyan: AOB, Yellow –orange-: NOB, Green: All bacteria).

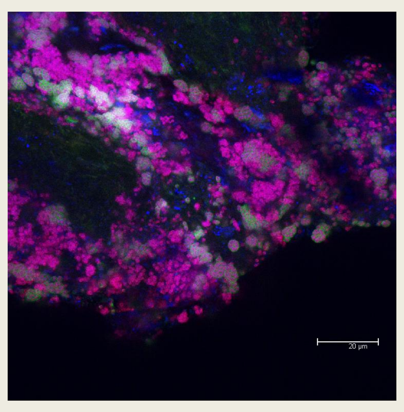


***FIGURE 3**:* FISH image of biofilm (Cyan: Chloroflexi, Magenta: Anammox, Blue: All bacteria).

2. Challenges in ReSCoBiR

Advantages

- High removal efficiencies are expected. AnAOB have reached removal rates up to 26 kg-N/m³/day *[4]*.
- Oxygen demand is 63% lower than in the nitrification-denitrification process ***[5]***.
- Oxygen transfer is very efficient in these systems, leading to lower aeration costs ***[6], [7], [8]***.
- Compact rector configuration with high biomass retention where control systems are easily applicable.
- No organic carbon is required. The carbon for bacterial growth is provided as HCO_3^- ***[2]***.
- Low CO_2 and N_2O emissions, causing a minor climate change impact ***[9]***.

Disadvantages

- Out competition of Nitrite oxidizers is not as easy as modelling studies reveal.
- Long start-up times until AnAOB show activity in the system ***[10]***.
- Fouling, durability and hydrodynamic behaviour of the membrane modules ***[8]***.
- Control strategy for both processes in the same reactor is difficult ***[11]***.

In this study:

- Construction of the reactor system.
- Study of the nitritation in order to achieve AnAOB stoichiometry.
- Modeling of the constructed reactor.

3. Materials and methods

Reactor monitoring

Most of the process variables are tracked on-line:

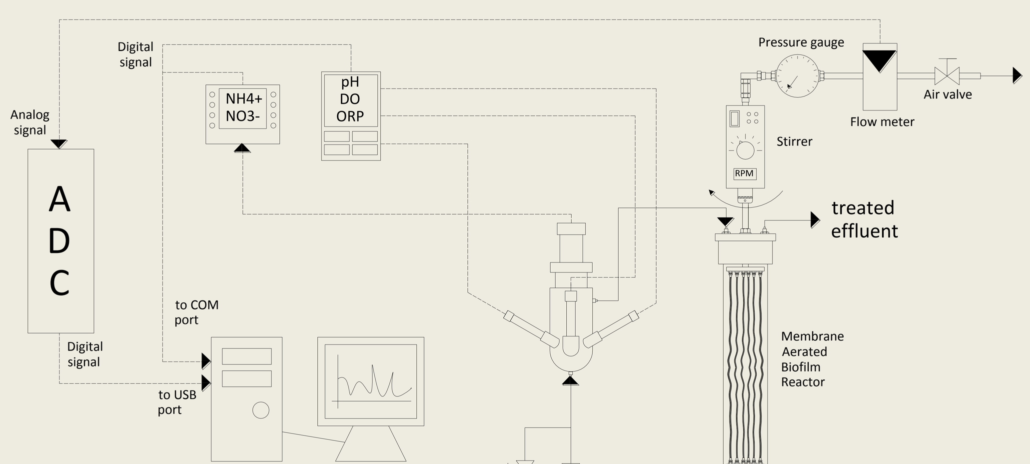
- pH.
- Dissolved Oxygen.
- Oxidation/Reduction Potential.
- NH_4^+ and NO_3^- concentrations.
- Air Flow.
- Reactor Temperature.

Variables tracked offline:

- Influent flow.
- Gas line pressure.
- Total Organic Carbon.
- NO_2^- Concentration.
- Total Suspended Solids

These readings are accessible from any computer with internet.

***FIGURE 4**:* Caption of the on-line monitoring system



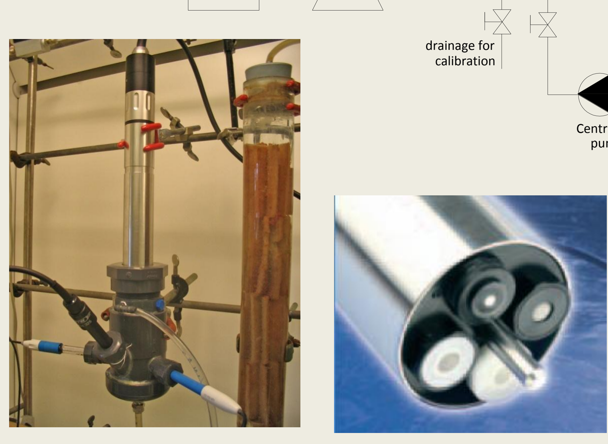


***FIGURE 5**:* Flow cell with electrodes and detail of the NH_4^+/NO_3^- electrode *[12]*

***FIGURE 6**:* Reactor setup

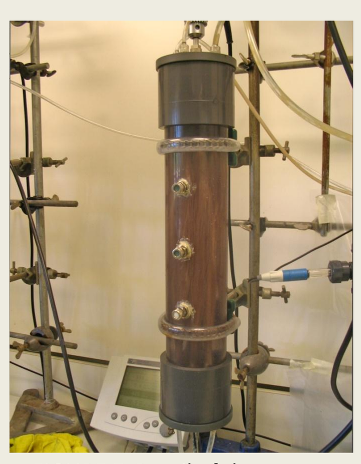


***FIGURE 7**:* Detail of the reactor

Substrate Composition

The substrate is synthetic wastewater adapted from ***[15]***, consisting of:

- $(NH_4)SO_4$ (300-600 mg-N/l)
- $KHCO_3$
- KH_2PO_4
- $MgSO_4{\cdot}7H_2O$
- $CaCl_2{\cdot}2H_2O$
- Trace elements

Oxygen supply

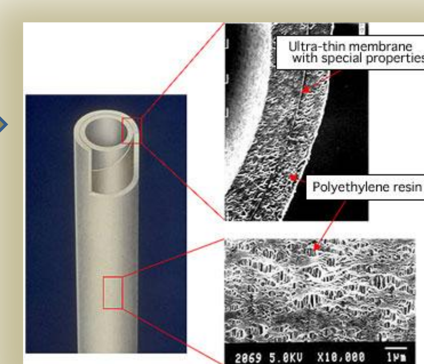


FIGURE 8**:* Hollow fibre membrane ***[13]

- Hollow fibre membranes in flow through configuration and O_2 supply by air.
- Composite membranes (Polyurethane + Polyethylene) for bubbleless operation.
- Specific surface area of 142 m²/m³.
- Oxygen transfer tests in clean water at 10, 30 and 50 KPa showed fluxes of O_2 of 2.12, 5 and 6.49 g O_2/m²/day.
- Higher global mass transfer coefficients when higher pressures are applied.

Hydrodynamic behaviour

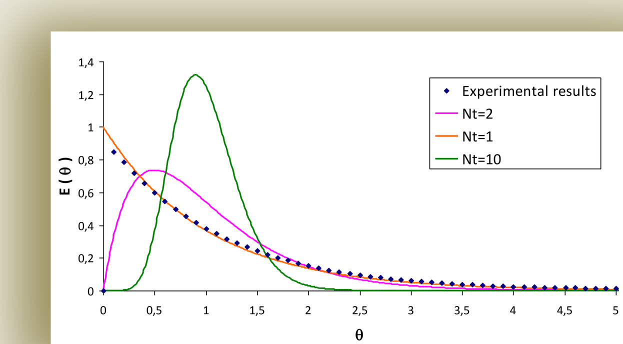


***FIGURE 9**:* Trace experiment results vs theoretical values from the N tank in series model for different N values

- High liquid recirculation ratio (80) and membrane module rotation (20 rpm) promote good dispersion characteristics and control of biofilm thickness.
- Packing density of 91.5 %, calculated according to ***[14]***.
- Residence Time Distribution tests and comparison to the N CSTR in series model show that the reactor behaves as a single CSTR.

4. Experimental results

Operation started in February, with nitrifying biomass from the Lundtofte wastewater treatment plant (Lundtofte, Denmark). The objective was to keep the J_{O2}/J_{NH4+} ratio at the optimal level ***[3]*** for attainment of the Anammox stoichiometry.

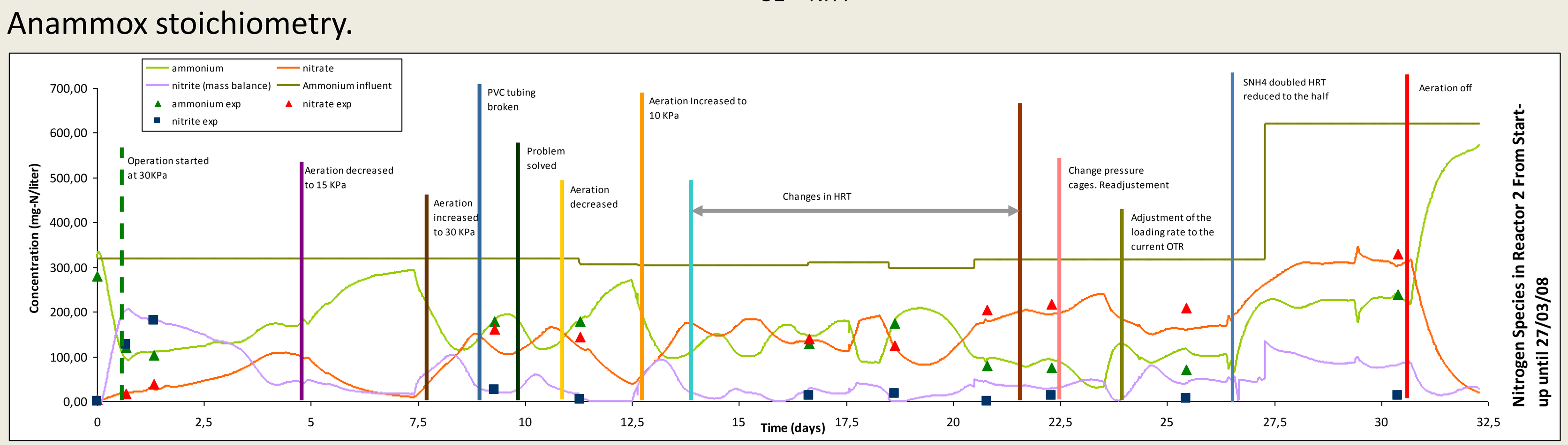


***FIGURE 10**:* Reactor performance during the first month of operation

- Difficulties to reach the proper NH_4^+/NO_2^- ratio, but fast response of the bacteria to disturbances.
- Calculations based on stoichiometric relations with NH_4^+, NO_2^- and NO_3^- steady state concentrations show an oxygen transfer rate about 10 times higher than the one predicted in the clean water tests.

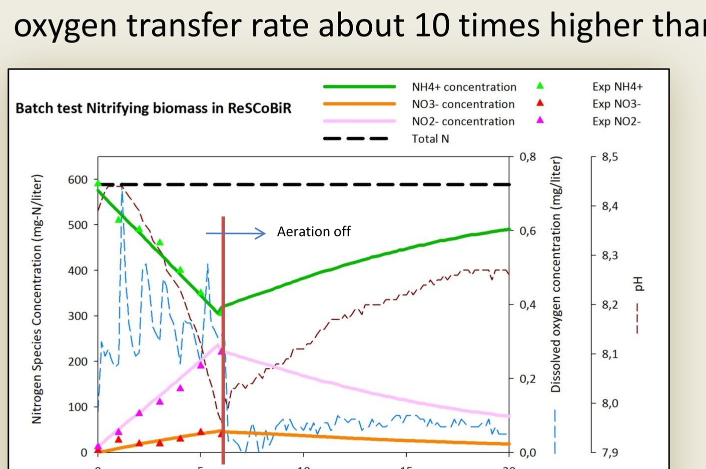


***FIGURE 11**:* Batch operation results. Evolution of parameters in the bulk liquid

Batch tests were carried to estimate bio-kinetic parameters and the current global mass transfer coefficient via parameter estimation with the implemented model. Another objective was to test the potential to reach the Anammox stoichiometry.

Current status and future work:

- Reactors inoculated with Anammox biomass cultivated in a plug flow reactor at 37 ºC in laboratory conditions.
- Continued operation to achieve complete nitrogen removal at room temperature.

5. Modeling

A mathematical model for the presented Counter-diffusion ReSCoBiR was built using AQUASIM ***[16]***, taking the model in ***[17]*** as guideline. A parameter estimation to fit the data presented in FIGURE 11 was performed.

Bio-kinetic parameters and global mass transfer coefficient obtained were used to build a simplified MATLAB model, which accounted for the following processes in the biofilm as they are presented in ***[18]*** and ***[19]***:

Particulates:

- Growth, decay and hydrolysis processes.
- Advection due to biofilm growth.
- Diffusion (implementation improved the convergence of the model).

Solutes:

- Diffusion.
- Reaction.

The model represent quite accurately the batch and the dynamics of the posterior continuous operation without aeration

- Empirical and modeling results confirmed that the mass transfer is 10 times higher than expected.
- AOB growth rate is 37% higher than the expected.
- NOB growth rate is 60% lower than the one in the literature.

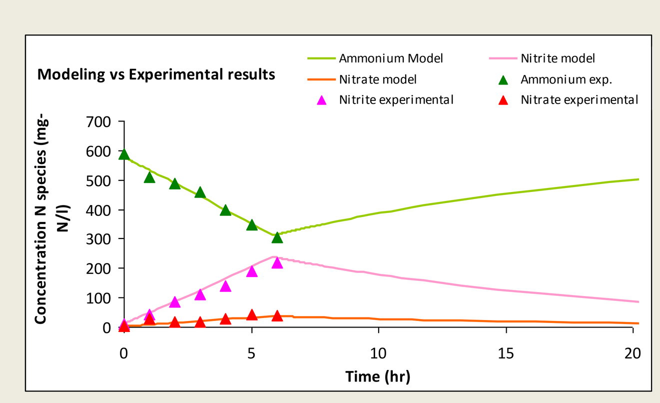


***FIGURE 12**:* Batch modeling results. Comparison to experimental values

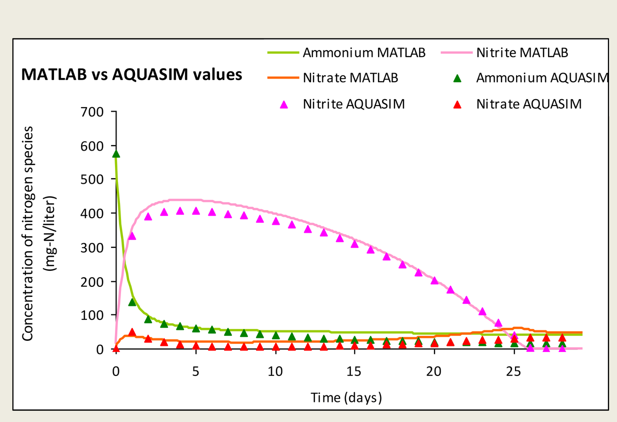


***FIGURE 13**:* Comparison of the results of the MATLAB and AQUASIM model

- The MATLAB model was extended and Anammox bacteria activity were incorporated. The model shows similar behaviour when compared to respective AQUASIM simulations.
- The model should be refined to represent better the biofilm growth and the stratification.
- A control strategy to shorten Anammox start-up time is being studied.
- MATLAB model will give the possibility to implement real time control in the system.

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