# Effect of Biofilm control on Nitrification in a Membrane Aerated Biofilm

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#### Abstract

The Membrane Aerated Biofilm reactor has shown in previous studies the potential for high-rate/high-efficiency treatment of various wastewater streams. This makes it an ideal candidate for treatment of streams with high ammonia concentrations. Landfill leachate is one such stream which despite large amounts of hard COD is possible to treat using biological processes. In this study a pilot scale MABR (Oxymem) was used for the treatment of leachate. Using both air and pure oxygen treatment rates of 1.3 and 6 gN-NH<sub>4</sub> m<sup>-2</sup> day<sup>-1</sup> were achieved respectively. High Oxygen Transfer Efficiencies of 50% and 90% were achieved for air and oxygen. Investigations into the influence of operational parameters on ammonia oxidation without subsequent nitrite oxidation were also examined and evidence highlighting the importance of biofilm control was found. The biofilm control while maintaining a thin active biofilm, caused the washout of biomass. This along with the reactor environment promoted the growth of AOB and stifled the growth of NOB. In fixed support biofilm reactor biofilm control is a critical parameter to ensure continued high performance.

## Keywords

Membrane Aerated Biofilm Reactor; Detachment, Partial Nitrification.

# **INTRODUCTION**

As we move towards the energy neutral wastewater treatment plant, novel energy efficient treatment technologies are gaining in importance. The Membrane Aerated Biofilm Reactor (MABR) is one such technology with its ability to supply oxygen to a pollutant degrading biofilm at high rate and high efficiency. The high rate of oxygen transfer and the potential of maintaining slow growing organisms in the biofilm are two of the advantages of the MABR which are particularly suited for the nitrification of ammonia in wastewater. The MABR has already shown its potential as for the high rate treatment of ammonia wastewater streams. Brindle et al. (1998), reported nitrification rates of 5.4gN-NH<sub>4</sub> m<sup>-2</sup> day<sup>-1</sup> while, Terada et al. (2003) achieved rates of 4.5gN-NH<sub>4</sub> m<sup>-2</sup> day<sup>-1</sup> (HRT 15 days) with a very high strength ammonia stream (N-NH<sub>4</sub>=3000mg/l). This high rate performance coupled with high the high Oxygen Transfer Efficiency (OTE) could, if deployed at full scale, significantly reduce the energy requirements for biological nitrification (Martin and Nerenberg 2012).

Landfill Leachate which is the water collected from the bottom of municipal landfills is a challenging wastewater stream. Although the composition of leachate changes with the age of the landfill it is typically characterised by high levels of ammonia, hard COD and dissolved solids. Treatment options include both biological and physical/chemical means. Currently in Ireland the treatment methods are either off-site by transportation to a municipal sewage treatment plant or onsite using an SBR for the nitrificiation of ammonia followed by a subsequent de-nitrification

process typically reed beds. Nitrification of the leachate using SBRs is very expensive and uses large amounts of caustic buffer and energy to provide oxygen and mixing, which in many instances results in equivalent treatment costs with off-site transportation.

Along with SBR's attached growth biofilm systems have proved successful with both RBCs and MBBRs being used for the treatment of landfill leachate. We lander et al. (1997) achieved nitrification rates of 11gN-NH<sub>4</sub> m<sup>-3</sup> hour<sup>-1</sup> (2.2gN-NH<sub>4</sub> m<sup>-2</sup> day<sup>-1</sup>) in a lab study, while complete nitrification was achieved at a rate of 1.92gN-NH<sub>4</sub> m<sup>-2</sup> day<sup>-1</sup> with an RBC (Kulikowska et al. 2010).

The potential use of an MABR for nitrification could lead to significant cost savings for the treatment of landfill leachate via both the reduced energy requirements and, if simultaneous denitrification is achieved, reduced chemical requirements. Another advantage of the MABR is the reduction in the emission of Volatile Organic Compounds (VOC) as was identified by Dollerer and Wilderer (1996), when using a silicone membrane to provide the oxygen for the biological treatment of a hazardous waste landfill leachate, a decrease in biologically degradable VOC in the flue gas was recorded although non-biodegradable VOCs were still emitted in the flue gas. With the advent of anaerobic ammonia oxidation and shortcut nitrogen removal technologies and their potential savings in terms of both oxygen and alkalinity requirement for nitrogen removal further potential saving could be achieved by conducting these processes in an MABR. An investigation into the treatment of municipal landfill leachate via combined shortcut nitrification. Anammox and achieved aerobic ammonia oxidation rates of hetrotrophic denitrification by Xu et al. (2010)  $2.83(kg NH_4-N kg_{dw}^{-1}day^{-1})$ . With a more efficient ammonia oxidation step by the MABR, potential energy savings of over 80 % could be achieved over the traditional treatment option of Nit-DeNit.

Keeping these potential savings for the treatment of leachate in mind the application of an MABR, referred to here as Oxymem, for the nitrification of ammonia in landfill leachate was examined and the factors affecting the potential of shortcut nitrogen removal were investigated

# EXPERIMENTAL DESIGN

As part of the study into the application of the MABR called Oxymem for ammonia oxidation, the effect of membrane cleaning/ biofilm detachment on the nitrification process was examined

# METHODS

A 60 litre pilot plant (Figure 1) was constructed to treat the leachate from Arthurstown municipal landfill, Co Kildare, Ireland. The pilot consisted of hollow fibre PDMS membranes which have an internal diameter of 300 $\mu$ m and an external diameter of 500 $\mu$ m giving a total surface area of 4.1 m<sup>2</sup> and a specific surface area of  $68m^2m^{-3}$ . The temperature in the reactor was not controlled but remained between 21- 27°C throughout the experiment. The leachate feed was supplied to the reactor using a Watson Marlow 323D peristaltic pump and was between 0.4 to 0.6 L hr<sup>-1</sup>. Due to logistical constraints the raw leachate from the site was diluted by a factor of 3-5 during operation and then the ammonia concentration was controlled by the addition of ammonium chloride. The pH and DO were measured using a Mettler Toledo M300 meter and the pH was maintained between 7.5 and 8 with the addition of 1 molar Sodium Hydroxide to promote the growth of Ammonia Oxidising Bacteria (AOB) over Nitrite Oxidising Bacteria (NOB) (Kim et al. 2006). The Dissolved Oxygen was continuously monitored but after the start-up phase never rose above 0.2mg/l unless due to a process upset. Mixing is provided via a recirculation pump with a flow rate of 30 litres per minute.



Figure 1 Schematic of reactor layout.

Initial Oxygen Transfer Rates OTR experiments were carried out using air and oxygen and results of between 20-50  $gO_2 m^{-2} bar^{-2} day^{-1}$  were obtained. Raw leachate and treated leachate samples were analysed between once and twice a week, with samples being analysed for COD, N-NH<sub>4</sub>, N-NO<sub>3</sub> and N-NO<sub>2</sub>, using Hach Lang LCK kits and a CADAS 30 spectrophotometer. Periodically (once a month), the treated leachate was analysed for phosphate to ensure that it was not a limiting substrate. Gas analysis of the flu gas for oxygen concentration started on day 140, and samples were taken 3-4 times a week this data was used to calculate the Oxygen Transfer Efficiency (OTE).

#### **Start-Up and operation**

The reactor was inoculated with nitrifying sludge from the Arthurstown SBR and no feed was added for the first two weeks. After this time the ammonia loading rate was very gradually increased over the first two months by increasing the feed flowrate. All the while air was supplied to the PDMS membranes at a pressure of 100mBar. After 63 days the loading rate was further increased by the addition of ammonia chloride to the feed and the gas supply to the membranes was switched to pure oxygen. Further adjustments were made to the operational parameters of the reactor throughout the experiment, these are summarised in Table 1.

## RESULTS

## **Operational Results**

The Oxymem pilot plant has been running for over 400 days and despite some operational setbacks has achieved a number of periods of stable performance. The major process issues have been associated with the integrity of the membrane potting; this cumulated in a long period of unsteady operation, and almost complete removal of the biofilm from the membrane surface occurred on day 300 to allow for intensive repairs. Since then there have been 100 days of operation without any issues. Some of the major results from periods of stable operation are also given in Table 1 and figure 2.

Examining these results it can be seen that the reactor has achieved consistently good ammonia oxidation efficiencies between 70%-90%. Oxymem is able to achieve two modes of operation 1) extremely high reaction rates when supplied with sufficient oxygen and buffering capacity (Days 121 to Day235) and 2) very efficient ammonia oxidation when supplied with air (Day 35-66 and Day 320 to date). Oxygen transfer efficiencies of 50% in membranes of 40cm.

#### Shortcut nitrification results

After the initial start-up period of 66 days, Ammonia oxidation rates of over 90% at loading rates of 1.3gN-NH4/m<sup>2</sup> day were achieved (figure 2).At this point the reactor was switched from air to pure oxygen. During this early phase most of the ammonia was oxidised to nitrite only, but after 100

days when the intra-membrane oxygen pressure was increased to 200mbar complete oxidation of the nitrite also occurred.

Along with the continually increasing loading rate which occurred trough out the first 150 days, biofilm control (Partial biofilm detachment) was initiated after day 120, using an air scour over the membrane surface for 5 minutes, this occurred at 8 time points between day 121 and 220, figure 3. Following from these events the nitrite concentration in the treated leachate began to rise. This was probably due to combination of the washout of the NOB from the biofilm and also the inhibition of the NOB caused by the increase in ammonia concentration in the reactor (Kim, Lee et al. 2006).

During the biofilm control period the nitrate concentration in the reactor slowly increased, possibly due to changes in the biofilm structure and/or population stratification. After 220 days the scouring events were stopped to observe the response of the reactor to uncontrolled biofilm growth. Along with complete oxidation of the ammonia to nitrate the percentage of ammonia nitrified decreased during this time. Because of the reduced ammonia oxidation rate the ammonia concentration in the reactor increased similarly as was seen at day 120 but in this case the measured nitrite concentration in the reactor remained less than 10mg/l indicating the importance of biofilm control in preventing complete nitrification

			N-NH <sub>4</sub>	Lumen	Lumen	$N-NH_4$	% N-NH <sub>4</sub>	
		Flowrate	Concentration	Gas	Pressure	Loading	Removal	OTE
Phase	Day	l/hr	mg/l		mbar	g/m² day		%
0	0 -14	0	-	Air	100	-	-	-
I	14 - 35	0.05	740 ± 76	Air	100	-	-	-
П	35 - 66	0.33	714 ± 103	Air	100	1.4	90	-
111	66 to 100	0.5	1134 ± 276	O <sub>2</sub>	100	3	90	-
IV	100 to 121	0.5	1248 ± 57	O <sub>2</sub>	207	3 to 4	80-95	80%
V	121 to 143	0.5	2172 ± 360	O <sub>2</sub>	190	5 to 7	40-80%	93%
VI	143 to 200	0.6 (0.26)	2364 ± 356	O <sub>2</sub>	210	4 to 8	80-90%	77%
VII	200 to 235	0.54	2200 ± 296	O <sub>2</sub>	235	6 to 7	80-90%	85%
VIII	235 to 280	0.54	2544 ± 201	O <sub>2</sub>	250	8	70-80%	40-50%
IX	280 to 337	0.4 to 0.6	2400 ± 263	O <sub>2</sub>	190	8 to 5	70%	70%
Х	337 to 365	0.4	1100 ± 429	Air	176	2	50%	80%
XI	370-400	0.4	563 ± 72	Air	200	1.38	76%	48%
XII	400-420	0.4	595 ± 61	Air	190	1.42	90%	55%

 TABLE 1: Operating conditions for Oxymem



Figure 2: Ammonia Loading and percentage removal over the course of the Oxymem operation to date.



Figure3: Concentrations of the Nitrogen species in the influent and in the reactor over the course of the first 250 days. The second x-axis indicates the periods of biofilm control events

After the almost complete biofilm removal on day 300 Oxymem regained stable operation around day 350 and biofilm scour recommenced on day 357. In this period the reactor was operated under a reduced load and was passed through the membrane. Biofilm scouring recommenced and occurred every two weeks, following each scour again there was a slight yet noticeable increase in the nitrite concentration in the effluent. (Figure 4)



Figure4: Concentrations of the Nitrogen species in the influent and in the reactor from day 350 day onwards. Along with an indication of the biofilm control events

Although the main focus of this study has been the investigation into the performance of Oxymem for the treatment of landfill leachate, from the results observed during both biofilm control periods it can be observed that when the biofilm control had a more larger impact on the performance of the NOB population than on the AOB.

## Conclusions

The conclusions from this study highlight the importance of detachment and regrowth of biofilms in wastewater treatment reactors. Unlike the MBBR and other biofilm reactors with mobile carriers, where large scale detachment is constantly occurring; in reactors with immobile biofilm supports, and indeed little or no gas sparging, the biofilm control mechanism and schedule play a huge role in preserving reactor performance. Maintaining the biofilm in a constant state of flux prevents a true "steady state" from occurring but allows for a measure of biological population control leading to improved reactor performance over long periods of time. Ultimately the entire effects of a biofilm

control strategy will need to be fully understood so that the appropriate implementation can lead to successful installation of a large scale Oxymem.

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