OPERATIONAL LEARNINGS FROM MEMBRANE BIOFOULING OF A GROUNDWATER REPLENISHMENT SCHEME

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ABSTRACT
One of the major operational challenges of operating membranes in wastewater effluent recycling applications is the control of biological fouling. Biofouling in the RO system has been a significant and ongoing issue in the operation of the Groundwater Replenishment Scheme (GWR 1.5 GL) in Perth, WA. This paper outlines the studies that were conducted to control biofouling in order to increase the operational life of the RO membranes.

INTRODUCTION
As part of its wide ranging response to a drying climate, the Water Corporation conducted the Groundwater Replenishment Trial (GWRT) from 2010-2012, in Perth, Western Australia to build knowledge of the local technical, health, environmental and social issues associated with groundwater replenishment as a future drinking water source. The Trial was successfully completed in December 2012 and all Trial Regulators – the Department of Environment Regulation (DER), Department of Health (DoH) and Department of Water (DoW) indicated that the Trial met all the project objectives¹. Since December 2012, the GWRT Advanced Water Recycling Plant (AWRP) has continued to operate as the GWR 1.5 GL/year scheme (GWR 1.5 GL). It was announced in August 2013 by the WA Water Minister that groundwater replenishment will be the next new climate independent water source for the Integrated Perth Water Supply Scheme². The full-scale groundwater replenishment plant will be built in the same site in 2014-2015, and be operational in late 2016.

The GWRT/GWR 1.5 GL scheme recharged treated recycled water to the Leederville Aquifer. The Beenyup Wastewater Treatment Plant (WWTP) provides the AWRP with a portion of its secondary treated effluent. This effluent was further treated to drinking water standards before being recharged to the Leederville aquifer. The AWRP treatment process utilised ultrafiltration (UF) and reverse osmosis (RO) followed by ultraviolet (UV) disinfection (Figure 1).

One of the major operational challenges of membrane filtration in wastewater effluent recycling applications is the control of biological fouling. Fouled membranes require chemical cleaning and ultimately result in water production loss, integrity loss, poorer water quality and a shorter membrane life which imposes a large economic weight on membrane plant operation³. Prevention and/or control of biofouling is essential for maintaining membrane operation to meet production volumes. In order to maintain long term operational stability, an intermittent cleaning process is required. The various cleaning processes used for fouling control fall into three categories: prevention, maintenance and recovery⁴. Operational costs also increase with the requirement for frequent chemical cleaning of membranes, so management of biofouling is crucial.

The level of biofouling that occurs on membranes is dependent on a variety of factors including the raw water quality entering the system (including the nutrient loading), and operational parameters at the AWRP. The Water Corporation currently operates two AWRPs with comparable treatment systems (the groundwater replenishment scheme and one scheme for industrial purposes). The industrial application AWRP has not experienced any significant biofouling issues compared to the groundwater replenishment AWRP. RO membranes at the industrial AWRP have only begun to be replaced after 10 years of service, well above the recommended lifespan expected from the membrane manufacturer.

As biofouling in the RO system has been a major ongoing issue in the operation of the groundwater replenishment AWRP, it has become the subject of a number of studies. This paper will describe the studies conducted to investigate the cause of the biofouling and how to control it by the use of biofouling management strategy in order to increase the operational life of the RO membranes.
**METHODOLOGY**

**AWRP Membranes**

The AWRP consisted of three UF trains operating in parallel (13 x 6) with Siemens Memcor L20 N membranes. The RO system consisted of two parallel trains. Both trains were two stage, single pass (15 x 8) with 7 elements in each vessel. The RO membranes were Hydranautics ESPA2-LD membranes.

**RO Membrane Autopsy and Biofilm Investigations**

The RO membrane autopsy was carried out by Advanced Water Management Centre from the University of Queensland. Characterisation of the membrane was done by several analytical techniques including:

- Loss of ignition (LOI)
- Metals
- Polysaccharides/Proteins
- Biomass quantification
- Scanning electron microscopy (SEM)
- Filtration trials

Debris was also collected by scaping a 1m² surface area on the wall of the UF and RO feed tanks within the AWRP. The debris was characterised by several analytical techniques by the Chemistry Centre of Western Australia including:

- Extraction of polysaccharides
- Hydrolysis of polysaccharides
- Sugar analysis (including HPLC analysis)
- ATP
- Metals
- LOI 550°C
- Protein

The full autopsy report and biofilm results can be presented on request.

**Chemical in Place (CIP) Cleanings**

A variety of CIPs were attempted at the AWRP to control biofouling. A CIP was initiated once the differential pressure (dP) across each stage was > 20 % from the previous clean. The type of cleans attempted (with the recirculation time and soak times) is shown in Table 1.

**Chlorine: Ammonia Dosing Experiments**

Bench scale experiments were conducted to determine the optimum chlorine to ammonia dosing ratio, and to also avoid nitrosamine formation above the health guideline value of 100 ng/L. AWRP feedwater was dosed with chlorine to ammonia ratios of 4:1 – 8:1 in a nitrosamine formation potential experiment under both “plant conditions at the time of sampling” (4 hours and 20 minutes) and a “worst case scenario” (6 hours). Samples were quenched with sodium thiosulphate and analysed for 9 nitrosamine compounds. The 9 nitrosamines were:

- N-nitrosodi-n-butylamine (NDBA)
- N-nitrosodi-n-propylamine (NDPA)
- N-nitrosodiethylamine (NDEA)
- N-nitrosodimethylamine (NDMA)
- N-nitrosoethylmethylamine (NEMA)
- N-nitrosopiperidine (NPIP)
- N-nitroso-morpholine (NMOR)
- N-nitroso-pyrrolidine (NPYR)
- N-nitroso-diphenylamine (NDHPA)

The AWRP treatment process flow during the experiments is shown in Figure 2.
Table 1: CIPs Attempted at the AWRP

<table>
<thead>
<tr>
<th>CIP Type</th>
<th>Recirculation Time</th>
<th>Soak Time</th>
<th>Max dP recovered (kPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Permeate</td>
<td>2 hours</td>
<td>1 hour</td>
<td>1</td>
</tr>
<tr>
<td>Acid (usually after a caustic CIP)</td>
<td>45 mins</td>
<td>1.5 hours</td>
<td>15</td>
</tr>
<tr>
<td></td>
<td>1 hour</td>
<td>1.5 hours</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2 hours</td>
<td>1.5 hours</td>
<td></td>
</tr>
<tr>
<td>Biocide</td>
<td>2 hours</td>
<td>N/A</td>
<td>8</td>
</tr>
<tr>
<td></td>
<td>2 hours</td>
<td>2 hours</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2 hours</td>
<td>24 hours</td>
<td></td>
</tr>
<tr>
<td>Biocide + citric acid</td>
<td>15 mins</td>
<td>15 mins</td>
<td>10</td>
</tr>
<tr>
<td>Caustic</td>
<td>1 hours</td>
<td>30 mins</td>
<td>14.5</td>
</tr>
<tr>
<td></td>
<td>2 hours</td>
<td>1 hour</td>
<td></td>
</tr>
<tr>
<td>Hot Water</td>
<td>4 hours</td>
<td>N/A</td>
<td>10</td>
</tr>
<tr>
<td>Hydrex (normal CIP)</td>
<td>2 hours</td>
<td>1 hour</td>
<td>16</td>
</tr>
<tr>
<td>Hydrex (reverse lead and tail)</td>
<td>2 hours</td>
<td>1 hour</td>
<td>21</td>
</tr>
<tr>
<td>Hydrex (reverse pipework)</td>
<td>2 hours</td>
<td>1 hour</td>
<td>33</td>
</tr>
<tr>
<td>Salt</td>
<td>90 mins</td>
<td>N/A</td>
<td>12</td>
</tr>
</tbody>
</table>

Figure 2: Treatment Process of the AWRP at time of sampling for the chlorine: ammonia experiments.
RESULTS/OUTCOMES
The AWRP RO system encountered significant biofouling issues, which resulted in an increase in differential pressure and permeate conductivity due to membrane damage from frequent chemical cleaning. The original membranes in the AWRP were replaced after 18 months of operation due to severe biofouling and were replaced with Hydranautics ESPA2 LD membranes. After installation of the membranes in November 2011, biofouling was identified within two months of operation as the normalised differential pressure increased for both trains. Operation of the membranes has been recorded daily since installation. Figure 3 shows the differential pressure of RO Train 2 with the first stage in blue and the second stage in red for the lifetime of Train 2 with ESPA2 LD membranes.

An RO membrane element was taken out of the membrane system on 12th March 2013 for an autopsy. The fouling layer deposited on the membrane surface is presented in Figure 4. Scrapings from the RO and UF feed tanks are presented in Figure 5.

Results from the membrane autopsy and tank scrapings confirmed that the fouling layer is a mixture of biological and organic matter. Visual resemblance of the samples collected from RO feed tank (Figure 5b) suggest that the source of the fouling in the membrane system (Figure 4) is possibly the microorganisms from the biofilm that was deposited in the feed tank.

![Figure 4: RO Membrane Surface Visual Aspect and Fouling Layer Deposited on Membrane Surface.](image)

The current design of the AWRP (Figure 2) whereby the treatment process consists of UF, an RO feed tank and the RO units is strongly dependant on the disinfection efficacy of the
chlorine and ammonia dosing prior to the UF. As shown in the disinfection efficacy review (discussed in detail in the “Disinfection Efficacy Review and Chlorine: Ammonia Dosing Experiments” Section in this paper), disinfection in the AWRP was ineffective, allowing biofilms to grow in the system (as shown in Figure 5). This biofilm built up in the UF and RO feed tanks and was seeded onto the RO units. A cartridge filter was proposed prior to the RO feed tank to prevent this debris entering the RO system, but was never installed.

Chemical in Place (CIP) Cleanings
Performance of the RO membranes has been recorded daily since installation, and data shows a very high frequency of CIP cleaning. Cleaning should occur when the RO membranes show evidence of fouling. The work instruction created for CIPs detailed the actions to follow in order to determine which CIP was required, the strength of the chemical and the procedure for batching, recirculating and flushing the CIP solution. A CIP would usually occur at the AWRP if:

- Normalised permeate flow decrease < 15% since the last CIP
- Normalised salt passage increases > 15% since the last CIP
- Normalised pressure drop increase > 15 % since the last CIP
- Prior to and after an extended shutdown (> 1 week)
- Feed fouling index (FFI) < 0.45

Cleaning of the RO membranes should be carried out before any of these values are exceeded to maintain the elements in a clean condition. Effective cleaning is evidenced by the return of the normalised parameters to their initial value. From Table 1, a variety of CIPs were attempted at the AWRP, and the most effective cleans were:

- Hydrex (reverse pipework)
- Hydrex (reverse lead and tail)
- Hydrex (normal CIP)

Over the operational life of the RO membranes, because the biofouling had physically settled on the RO membrane surface, cleaning by standard acid and base CIPs became relatively ineffective and frequent (fortnightly).

After consultation with Hydranautics personnel, membrane specialists within Water Corporation and operational personnel, inadequate cross flow velocity during a CIP was identified as a potential problem with keeping the membranes clean. The RO trains in the AWRP were modified to allow reverse flow CIPs at a higher flow rate. Reversing the flow of the CIP theoretically allows shear force to destabilise the biofilm and remove it. The AWRP was not originally designed for reverse CIPs, and pipework modifications and membrane protection devices (e.g. anti-telescoping on lead end, shims) were installed to initiate these CIPs.

Prior to the design for the pipework modifications being completed, another reverse clean whereby the head and tail membranes in each element were switched in location was attempted. One issue with the reverse head and tail CIP is a potential risk of membrane integrity breaches. Conductivity mapping was undertaken to confirm seals were not rolled or telescoping occurred. However, every time an RO membrane element is opened and the membranes are removed there is a potential for an integrity breach, creating an inherent risk to membrane performance. Operational staff also advised that the reverse head and tail CIP involved significant manual handling compared to other CIPs. As a result of these concerns, the reverse head and tail CIP was discontinued once the reverse pipework modifications were complete.

Another CIP trialled for the RO membranes was a high salt dose for a short period of time. Dosing a high concentration of salt causes a shift from reverse osmosis to osmosis within the membrane, cleaning the biofilm at a molecular level on the surface of the membranes. This CIP was only attempted four times (after two years of operation of the membranes) with limited success. This limited success could be due to the entrenched biofouling on the membrane surface by this point of time.

An RO permeate flush was also introduced on a daily basis (3 x 5 minutes) to dislodge any biofilm from adhering to the membrane surface for an extended period of time. By including an RO permeate flush, followed by a hydrex reverse pipework CIP; this has reduced chemical cleaning of the membranes to every 4-6 weeks (rather than fortnightly). This is estimated to have increased the operational life of the membranes by an extra two years from the previous projected lifespan.

Disinfection Efficiency Review and Chlorine: Ammonia Dosing Experiments

Chloramines are widely used as an alternative disinfectant to chlorination. Dosing chloramines has been proven to be very effective in minimising biofouling through membrane processes. Unlike free chlorine, chloramines do not cause significant damage to RO membranes. Chloramines are formed by mixing sodium hypochlorite with ammonia. Depending on the process conditions, chloramines can be found under three different species: monochloramine (NH2Cl), dichloramine (NCl2) and trichloramine (NCl3).
One part of the investigation into biofouling found that the variability in the current chloramination concentrations through the treatment stages of the AWRP indicated that the chloramine dosing system was not controlling biogrowth at the RO membrane surface and that the compounds formed could be of an organochloramine nature. Organochloramines are disinfection by-products which exhibit little or no biocidal activity and can react as chloramines under test conditions used in most methods for analysing monochloramine and dichloramines. Other treatment plants treating WWTP effluent found that a concentration of at least 3 mg/L of monochloramine is required to be maintained from UF feed to RO concentrate to effectively control biological growth, even when relatively high levels of nutrients are present.

An initial set of experiments determined the nitrosamine formation of the AWRP at plant conditions, and a worst case scenario. From Figure 6, only two nitrosamines were detected from the 9 compounds analysed at the various chlorine:ammonia dosing ratios – N-nitrosodimethylamine (NDMA) and N-nitrosomorpholine (NMOR). The remainder of the nitrosamines were not detected above their limit of reporting. From these initial experiments, the acceptable chlorine:ammonia ratios that could be used at the AWRP without exceeding the guideline value for NDMA was determined to be 4:1 – 6:1.

![Figure 6: Nitrosamine formation at the AWRP using various chlorine: ammonia dosing ratios for plant conditions (4 hours and 20 minutes).](image)

A number of experiments were then conducted to optimise the disinfection efficacy to see if it was possible to improve the monochloramine production. The chlorine to ammonia ratios varied from 4:1 – 8:1, with an ammonia residual from 1 – 3 mg/L. The experiments tested different injection scenarios as follows:
- Ammonia injection followed by chlorine injection (similar to the plant, Figure 3)
- Simultaneous ammonia and chlorine injection
- Preformation of chloramines then injection into the samples

Further experiments tested the difference in chloramine formation at various locations in the AWRP as follows:
- Before UF
- After UF

Analysis of the data from the various experiments produced the following conclusions:
1) Preforming monochloramines was the best option to maximise monochloramine formation. This eliminates all other competing reactions between ammonia, chlorine and other components in the water.
2) Two ratios were found to provide the optimal monochloramine concentration for RO disinfection:
   - 4:1 with 3 mg/L ammonia residual
   - 6:1 with 2 mg/L ammonia residual
3) Post UF injection appears to be the best option as it maximises the monochloramine ratio over total chloramines.

The AWRP increased the chlorine: ammonia ratio to 6:1 with a 2 mg/L ammonia residual after these experiments. From the time the ratio was increased (February 2014) the next clean of the RO membranes was in June 2014, allowing continual operation of the AWRP for over 4 months. This was an excellent achievement considering CIPs were being conducted on a fortnightly basis prior to adoption of any formal biofouling management plan. The AWRP was decommissioned in September 2014, so further research on performance at the higher dose rate with the RO membranes could not be attempted.

CONCLUSIONS
No single control measure was found to solve biofouling of RO membranes at the AWRP. One significant issue found during the investigations was due to the presence of the UF and RO feed tanks. These feed tanks allow the monochloramine dosed into the system to be degraded to organochloramine, which allowed biofouling to occur on the RO membranes.

A number of experiments found the optimal disinfection dose required in the AWRP to produce monochloramines. Increasing the chlorine:ammonia ratio to 6:1, with a 2 mg/L ammonia residual allowed continual operation of the AWRP without the need for a CIP, in excess of the
recommended cleaning frequency from membrane manufacturers.

A number of CIPs were also trialled in the AWRP to control biofouling. The preferred CIP method for the AWRP was found to be hydrex using the reverse pipework method. The variety of CIPs attempted in the AWRP has created a greater understanding of membrane cleaning procedures for operational staff.

All of the investigations undertaken at the AWP have created a “biofouling management strategy”. Implementation of the “biofouling management strategy” has shown that biofouling has been minimised at the AWRP and increased the lifetime of the RO membranes.

It is anticipated that the methodology undertaken at the AWRP with respect to biofouling, will be applied to the future full-scale groundwater replenishment scheme to be implemented in 2016, as well as other recycling applications undertaken by the Water Corporation.

REFERENCES

1. ‘Groundwater Replenishment Trial – Final Report Summary 2013’

2. ‘Advanced Recycling to Help Secure Water Supply’


