A pilot-plant study of a moving-bed biofilm reactor system using PVA gel as a biocarrier for removals of organic carbon and nitrogen

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Abstract A pilot-plant study was conducted to evaluate the performance of a moving-bed biofilm reactor process using PVA-gel beads as a biocarrier. Real primary-settled wastewater was fed to the pre-denitrification system and removals of nitrogenous and organic contaminants were evaluated over a 1-year period. The results demonstrated that at a total nitrogen (TN) loading of 18 mg/Lh, a TN removal efficiency in keeping with and even exceeding the theoretical maximum efficiency based on the level of internal recycle, was possible and a nitrification rate of 15 mg/Lh was sustained with a HRT of only 2.5 h at 15 °C. Furthermore, soluble COD and BOD5 in the effluent of the pilot plant were reduced to levels well below most regulatory discharge limits. In addition, the possibility of using this biocarrier in a system, including the elimination of waste organic sludge, was discussed.

Keywords Attached growth; denitrification; moving-bed biofilm reactor; nitrification

Introduction Biological wastewater treatment processes commonly rely on suspended biomass (activated sludge) for removals of organic carbon and nutrients. Attached-growth (biofilm) processes, however, have demonstrated greater efficiency and sustainability than suspended-growth processes, especially at low temperatures, in the presence of inhibitory compounds and at high or variable loadings (Levstek et al., 2003). In addition, higher treatment rates are achievable due to selective retention of large amounts of effective biomass with no need for sludge recycle (Rouse et al., 2004), thus allowing for compact treatment plants.

Fluidised-bed and moving-bed biofilm reactors have demonstrated further improvements in performance over fixed-bed and packed-bed reactors by eliminating dead-space resulting from clogging and channelling and eliminating the need for backwashing (Pastorelli et al., 1997; Hoa et al., 2006). In addition, these innovative processes can be applied to existing plants to enhance treatment power without major new construction. A potential drawback of biofilm processes, however, is diffusion limitation of substrates and oxygen across biofilm layers, which becomes more critical with increasing biofilm thickness (Vanhooren et al., 2002; Dulkadiroglu et al., 2005).

When nitrogen removal is required, a further advantage of biofilm processes is realised because attached biomass can easily be cultivated and retained in respective aerobic and anaerobic units to meet the metabolic requirements of nitrifying and denitrifying organisms, respectively. In single-sludge suspended-growth systems, the sludge passes through all the existing conditions in the plant, thus at any point in the system only a fraction of the biomass is active (Yuan et al., 2001). In addition, autotrophic nitrifying
organisms grow very slowly and thus require a long solids retention time, which biofilm processes provide; conversely, the solids retention time of suspended-growth processes is often inadequate, resulting in washout of nitrifying organisms.

Various biomass carriers are available on the market including polyvinyl alcohol (PVA) gel, which has been shown to be effective for cultivation and retention of slowly growing bacteria (Rouse et al., 2005). The PVA-gel beads consist of 4-mm diameter spheres and have a specific gravity of 1.025 (Kuraray Co., Osaka, Japan), thus being easily fluidised in solution. They are hydrophilic in nature and have a very porous structure with only 10% solids and a continuum of passages 10 to 20 μm in diameter tunnelling throughout each bead. Bacteria cultivated inside the core of the beads do not slough off and are protected from predation, thus being highly retained. The beads are typically used at volumetric packing ratios of only 5 to 15% versus much higher ratios of 50 to 70% common to other carriers. Loading rates are determined with respect to the settled-bed volume of the PVA-gel beads (or total reactor volume with consideration to filling ratio) as opposed to the surface area of the carrier because the biomass is cultivated and retained inside the beads and not on the surface.

Being slightly heavier than water, PVA-gel beads can be used in a packed-bed mode of operation (Rouse et al., 2005); furthermore, the beads can easily be kept in suspension by applying internal recycle in a column reactor for use in a fluidised-bed mode (Hoa et al., 2006). In the context of this study, however, the circulation of beads in the reactors is not due to internal recycle, but rather mechanical mixing; thus, the “moving-bed” terminology is adopted to describe the biofilm reactors used here.

**Experimental protocol**

The novel PVA-gel beads were used as biocarriers in a pilot-plant treating real wastewater from the primary (mechanical) treatment stage of the Domžale-Kamnik treatment plant which, though primarily municipal, receives considerable input from various industries. The objective of this study is to determine the capability of the pilot-plant with respect to removals of nitrogen and organic carbon under real treatment conditions experiencing daily and seasonal variations in influent contaminant concentrations and environmental conditions.

The pilot-plant system consisted of two biological parts: the first being a nitrogen-removal process based on pre-denitrification with recycle of nitrified liquor (Figure 1). This process included an anaerobic (anoxic) reactor followed by an aerobic (oxic) reactor, both containing the PVA-gel biocarrier. Biological treatment activity is attributed to attached growth because suspended activated sludge was not returned to (or retained in) this process. Subsequently, a total-oxidation (TOX) process was used for elimination of excess organic solids (biomass). While the removals of nitrogenous and organic contaminants across the complete system are covered in this report, details pertaining to the

![Figure 1](https://iwaponline.com/wst/article-pdf/55/8-9/135/439821/135.pdf)

**Figure 1** Schematic diagram of the pilot plant used in this study
balance of solids across the system (including the elimination of organic sludge in the TOX or “zero sludge” process) and methods of solid–liquid separation will be the subject of a subsequent report. With respect to the nitrogen-removal process, the experimental program included a series of seven runs conducted at various loading rates (dependent on hydraulic retention time (HRT) and influent composition), internal recycle levels and temperatures, as shown in Table 1.

Materials and methods
All reactors used in this study were constructed of Plexiglas and had operational volumes of 200 L (Figure 1). The anoxic and oxic reactors of the nitrogen-removal process contained a 15% volume of PVA-gel beads that were kept in suspension by mechanical mixing and retained in their respective zones by using slotted strainers. The subsequent TOX process consisted of two aerated reactors, each containing mixed-liquor suspended solids with no PVA-gel beads. All reactors functioned in a complete-mix mode. The system was concluded with a 150-L sedimentation tank providing return sludge only to the TOX process.

Wastewater was fed to the system and recycled between units by using peristaltic pumps. Inflow parameters measured on-line consisted of TOC and total nitrogen (TN) (Shimadzu, Japan), NH₄-N (WTW, Germany), and nitrification inhibition (LAR, Germany). Treatment performance was monitored by following total Kjeldahl nitrogen (TKN), NH₄-N, NO₂-N, NO₃-N, COD and BOD₅ as determined on spot samples. All analyses of spot samples were conducted in accordance with ISO methods. Samples for determination of soluble components were passed through Sartorius cellulose nitrate membrane filters prior to analyses. Filtration at 1.0 μm was used for Run I and 0.45 μm was used for Runs II through VII. Filtration at 0.45 μm was necessary for complete separation of the very fine solids yielded by the biofilm process; this filter variation, however, should not affect analyses for NH₄⁺, NO₂⁻ and NO₃⁻, but could have a small effect for organic components.

Results and discussion
At start-up, activated sludge from the full-scale treatment plant was added to the reactors containing PVA-gel beads and the process was operated with continuous recycle of the sludge (from the sedimentation basin) for approximately 1 month to achieve biomass growth in the PVA-gel carriers. Then, 1 L of PVA-gel beads that had been enriched using a high NH₄-N concentration was added to the oxic reactor and operation commenced with no further recycle of sludge to the reactors containing PVA gel.

Table 1

<table>
<thead>
<tr>
<th>Run</th>
<th>I</th>
<th>II</th>
<th>III</th>
<th>IV</th>
<th>V</th>
<th>VI</th>
<th>VII</th>
</tr>
</thead>
<tbody>
<tr>
<td>Influent flow (L/h)</td>
<td>50</td>
<td>67</td>
<td>50</td>
<td>50</td>
<td>67</td>
<td>67</td>
<td>80</td>
</tr>
<tr>
<td>HRT (h)*</td>
<td>4.0</td>
<td>3.0</td>
<td>4.0</td>
<td>4.0</td>
<td>3.0</td>
<td>3.0</td>
<td>2.5</td>
</tr>
<tr>
<td>Influent TN (mg/L)</td>
<td>48</td>
<td>37</td>
<td>34</td>
<td>50</td>
<td>55</td>
<td>47</td>
<td>44</td>
</tr>
<tr>
<td>TN loading (mg/L.h)**</td>
<td>12</td>
<td>12</td>
<td>8.5</td>
<td>12</td>
<td>18</td>
<td>16</td>
<td>18</td>
</tr>
<tr>
<td>Internal recycle (%)</td>
<td>200</td>
<td>200</td>
<td>200</td>
<td>200</td>
<td>200</td>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>HRTr (h)***</td>
<td>1.3</td>
<td>1.0</td>
<td>1.3</td>
<td>1.0</td>
<td>1.0</td>
<td>1.5</td>
<td>1.25</td>
</tr>
<tr>
<td>TOC:TN</td>
<td>3</td>
<td>3</td>
<td>2–7</td>
<td>1–3</td>
<td>3</td>
<td>3</td>
<td>3</td>
</tr>
<tr>
<td>DO, oxic reactor (mg/L)</td>
<td>6</td>
<td>5</td>
<td>7</td>
<td>6</td>
<td>5</td>
<td>7</td>
<td>5–9</td>
</tr>
<tr>
<td>Water temperature (°C)****</td>
<td>22</td>
<td>22</td>
<td>20</td>
<td>11</td>
<td>13</td>
<td>14</td>
<td>15</td>
</tr>
</tbody>
</table>

*Nominal HRT in each tank with respect to influent flow
**TN loading with respect to each individual reactor volume (i.e. 200 L)
***HRTr: Real HRT in each tank including impact of internal recycle flow
****Lab windows opened at beginning of Run IV to lower the temperature
Dissolved oxygen levels in the oxic reactor were 5–7 mg/L throughout the study (Table 1) and in the anoxic reactor they were usually below 0.1 mg/L (rarely exceeding 0.2 mg/L). The influent TN contained approximately 40% organic nitrogen (Org-N) and ammonification efficiency across the nitrogen removal process averaged 82% over the runs of this study (data not shown). With respect to the volume of the denitrification unit (200 L), the TOC loading ranged from 10 to 90 mg/L.h (data not shown) and TOC:TN ratios in the influent wastewater were usually about 3, with extremes of 1 and 7 (Table 1). Apart from the lowest TOC:TN extreme, organic carbon was not considered limiting (see following discussion).

In Figure 2, the time courses of TN in the influent, NO$_2$-N + NO$_3$-N (NOx-N) in the anoxic reactor and NH$_4$-N in the oxic reactor during the pilot study are shown. Nitrification and denitrification in the oxic and anoxic reactors, respectively, were well developed by the onset of Run I. During Runs 1, 2 and 3, influent TN gradually decreased due to seasonal shutdowns of local industries and onset of the rainy season. As shown in Figure 3, TN removals were near the theoretical maximum efficiency of 67% during this period when internal recycle was maintained at 200% and the wastewater temperature was approximately 20°C (Table 1).

For Run IV, when the influent TN level had recovered to a relatively high level (ca. 60 mg/L, Figure 2), the internal recycle was increased to 300% and the laboratory windows were opened to allow for a cold-weather treatment study. However, as shown in Figure 2, NOx-N concentrations in the anoxic reactor were higher during Run IV, which may have been impacted by the large quantity of aerated recycle liquor from the oxic reactor. Under these conditions, the HRTr (i.e. HRT including the impact of internal recycle) in the anoxic reactor was only 1.0 h, which may not have been sufficient to avoid oxygen inhibition. In Runs II and V the HRTr levels were also 1.0 h, however the internal recycle levels were only 200%, thus avoiding excessive oxygen intrusion and allowing for better denitrification performance. In addition, during Run IV the influent TOC:TN ratio fell to its lowest level of approximately 1 (Table 1), which may have further inhibited denitrification performance. The low temperature (11°C) during Run IV
was not thought to significantly impact denitrification because nitrification performance was strong in this run (Figure 2, Table 2). In subsequent runs, internal recycle levels were kept at 200% or lower to avoid inhibition of denitrification; however, an anoxic reactor could be sized to handle higher internal recycle flows if needed for greater TN removal efficiencies.

For Run V the influent flow rate was increased to lower the process HRT to 6 h (unit HRT, 3 h). During this run, when the TN loading rate was at its highest level of 18 mg/L.h, denitrification was complete; however, NH4-N concentrations in the oxic reactor were high (Figure 2) and TN removal efficiency fell 10 percentage points below the theoretical maximum (Figure 3). The nitrification rate, though, was approximately the same as in Run IV, thus the inferior efficiency during Run V may have been largely due to the higher NH4-N loading rate (Table 2). During Run V, though, two major nitrification inhibition responses (from on-line monitoring) occurred, indicating that nitrifying bacteria may have been stressed during much of this run.

Runs VI and VII continued at the same high TN loading (Table 1) and nitrification performances were improved (Table 2), reaching an average rate of approximately 15 mg/L.h in the last run with a HRT of 2.5 h at 15°C. On most sampling occasions in the last two runs, NH4-N concentrations in the oxic reactor were very low (1–3 mg/L); however, there were a few high responses that corresponded with the higher influent TN concentrations (Figure 2). For those events, removal efficiencies were diminished, but

![Figure 3](https://iwaponline.com/wst/article-pdf/55/8-9/135/439821/135.pdf)

**Table 2** Ammonium loading and nitrification rates and efficiencies during the pilot study

<table>
<thead>
<tr>
<th>Run</th>
<th>I</th>
<th>II</th>
<th>III</th>
<th>IV</th>
<th>V</th>
<th>VI</th>
<th>VII</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ammonium load (mg/L.h)*</td>
<td>10.0</td>
<td>11.3</td>
<td>8.3</td>
<td>11.5</td>
<td>17.0</td>
<td>14.8</td>
<td>17.3</td>
</tr>
<tr>
<td>Nitrification rate (mg/L.h)**</td>
<td>9.3</td>
<td>10.5</td>
<td>8.2</td>
<td>11.0</td>
<td>11.5</td>
<td>13.2</td>
<td>14.6</td>
</tr>
<tr>
<td>Nitrification efficiency (%)</td>
<td>93</td>
<td>93</td>
<td>99</td>
<td>96</td>
<td>68</td>
<td>89</td>
<td>84</td>
</tr>
</tbody>
</table>

*Ammonium load = (Inf TKN − Eff Org-N)/(HRT of oxic reactor)

**Nitrification rate = (Inf TKN − Eff TKN)/(HRT of oxic reactor)
nitrification rates were not, thus these results suggest that the process was approaching a maximum nitrification rate for the conditions of testing used here. For one grab sample near the end of Run VII when the influent TN peaked at 62 mg/L, the highest individual nitrification rate of 21 mg/L.h was recorded. Furthermore, with the internal recycle set at only 100% in Runs VI and VII, TN removal efficiencies were 10 to 14 percentage points higher than the theoretical maximum level of 50% (Figure 3). In these final runs of the study, when the biofilm in the PVA-gel beads would have been at its most mature level, the possibility that denitrification was also occurring in the protective core of the PVA-gel beads in the oxic reactor is considered as a explanation for this observation.

The elimination of soluble organic carbon across the complete system during Run III is described in Table 3. Soluble COD and BOD₅ were progressively cut in the successive stages of the system and the results following both the nitrogen-removal process and the TOX process were comfortably below typical discharge limits.

The fractions of nitrogenous compounds across the complete system during Run III are shown in Figure 4. These results show that TN was reduced to 8–9 mg/L in the reactors of the nitrogen-removal process, which – in addition to nitrification and denitrification efficiencies – is a function of the internal recycle (200%). As was the case in all runs (not shown), Org-N levels of 2–3 mg/L remained in all stages of the pilot plant. The increase in TN of 5 mg/L (as NO₃–N) in the final effluent is due to release of nitrogen from endogenous decay of biomass in the TOX process. If discharge standards require that effluent TN be reduced to less than 10 mg/L, additional internal recycle in the proceeding nitrogen-removal process or an additional post-treatment process would be required. The TOX process, though, is optional and the effluent of the nitrogen-removal process (i.e. of the oxic reactor) can be discharged following adequate solids–liquid separation.

While not detailed as a part of this report, no excess sludge was wasted from the system over the course of the TOX study, which was conducted during the first half year

<table>
<thead>
<tr>
<th>Stage (location)</th>
<th>Influent</th>
<th>Anoxic unit</th>
<th>Oxic unit</th>
<th>TOX effluent</th>
</tr>
</thead>
<tbody>
<tr>
<td>COD (mg/L)</td>
<td>367</td>
<td>61</td>
<td>50 [86% removal]</td>
<td>37 [90% removal]</td>
</tr>
<tr>
<td>BOD₅ (mg/L)</td>
<td>213</td>
<td>27</td>
<td>12 [94% removal]</td>
<td>6 [97% removal]</td>
</tr>
</tbody>
</table>

Table 3 Soluble COD and BOD₅ at the different stages of the pilot plant during Run III. Data of the oxic unit describes the effluent of the nitrogen removal process.

![Figure 4](https://iwaponline.com/wst/article-pdf/55/8-9/135/439821/135.pdf)
(through Run III). The solids yielded by the nitrogen-removal process were retained in the TOX process, which reached a steady-state level of sludge mass that was maintained over the final 2 months, thus demonstrating the feasibility of a balance between sludge production and decay in conjunction with a moving-bed biofilm reactor system.

Conclusions
Results of this moving-bed biofilm pilot-plant study demonstrated the effectiveness of using PVA-gel beads as a biocarrier for removal of nitrogen and organic carbon from a real municipal wastewater. At TN loadings of approximately 18 mg/L.h, TN removal efficiencies in keeping with and even exceeding the theoretical maximum based on the level of internal recycle were achieved. Furthermore, at 15 °C with a unit HRT of only 2.5 h, a nitrification rate of 15 mg/L.h was sustained and a peak rate of 21 mg/L.h was observed. In addition, levels of soluble COD and BOD₅ in the effluent of the pilot plant were well below most regulatory discharge limits. The process evaluated here would be suitable for a full-scale application where unit processes are sized to handle internal recycle levels as required to meet nitrogen discharge limits.

Acknowledgements
The authors wish to thank Mr. Boris Kalcic of Interchem, Austria, for his continual support with the logistics and coordination of the project as well as Mrs. Barbara Brajer Humar and Mr. Podbevsek Sebastjan for their kind cooperation with the analytical work and various technical aspects of the project.

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