Pilot plant study on the performance and optimization of submerged membranes for taste and odor removal

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Abstract: Hybrid sorption-membrane processes are an attractive alternative for meeting a range of water treatment goals in a single process that is compact and cost-effective. This study investigated the performance and optimization of a hybrid sorption-membrane process using powdered activated carbon (PAC) and submerged-style membranes for odor control. Specifically, this study focused on the removal of 2-methylisoborneol (2-MIB) from a Lake Michigan source water and investigated the effects of PAC dose, dosing method, backwash interval, and aeration. Adsorption performance was predicted using a mathematical model, and tested using a pilot-scale, submerged membrane system. Modeling continuously dosed PAC performance agreed well with pilot results, but pulse dosed PAC performance was overestimated by the model. Non-ideal mixing effects were identified as important factors in explaining the pulse dosing results. Pilot results with aeration and pulse dosing were also overestimated by the model. It is hypothesized that aeration disturbs the PAC cake layer that forms on the membrane, and thus eliminates the beneficial effects that this cake layer can have on adsorption. Extending backwash intervals from 30 to 180 minutes with continuous dosing increased the percent removal of 2-MIB by up to 30% in both the model predictions and pilot results. This study highlights some important considerations in the design of full-scale systems and future mathematical models.

Keywords: Activated carbon; adsorption; MIB; membrane; 2-methylisoborneol; taste and odor; ultrafiltration

Introduction
The use of low-pressure, microfiltration (MF) and ultrafiltration (UF) membranes in drinking water treatment has expanded quickly over the last decade, because of superior removal of pathogens and particles. Meanwhile, an important emerging technology is the combination of adsorption and low pressure MF/UF membranes for the efficient removal of dissolved contaminants that are not excluded by a low-pressure membrane alone (Adham et al., 1991, 1993; Jack and Clark, 1998; Campos et al., 1998, 2000a,b). This hybrid sorption-membrane process is a powerful treatment technology that can remove a wide range of contaminants in a single process that is very compact and cost-effective.

To date, most of the attention in hybrid sorption-membrane processes has focused on adding powdered activated carbon (PAC) to conventional MF or UF membrane reactors. There have been several bench and pilot-scale studies and a few full-scale plants built with this approach (Anselme et al., 1997; Baudin et al., 1997). While the references noted above provide important information on PAC-UF processes including performance data, models and optimization insights, they do not provide data based on submerged-style membrane units, which are getting increasing attention for many drinking water applications.

Best et al. (1999) presented data on the use of PAC with submerged-style membranes, but their research was primarily focused on demonstrating the compatibility of submerged membranes with large PAC doses and they did not attempt to optimize the PAC dose. Schideman et al. (2001) used the mathematical model developed by Campos et al. (2000a,b) to predict taste and odor removal in a submerged membrane system. This later study
demonstrated good agreement between the mathematical model results and a bench-scale, submerged-style membrane system for the case of 2-MIB removal from Lake Michigan water.

Building on this past work, the present study presents data gathered from a pilot-scale, submerged-style membrane system and compares it to mathematical model results for removal of the common taste and odor (T&O) causing compound, 2-MIB. Various operating procedures were modeled and pilot tested in an attempt to improve the performance of a hybrid sorption-membrane process for this application.

Materials and methods
A mathematical model was used to predict adsorption performance in a pilot-scale, submerged-style, hybrid sorption-membrane reactor under various operating conditions. The operational parameters investigated in this study included the following: carbon dose, carbon dosing method (pulse vs. continuous PAC dosing), aeration, and backwash interval. The following paragraphs describe the materials, equipment and experimental approach used in this study.

Adsorbent and adsorbent characterization
WPH powdered activated carbon (PAC) as manufactured by Calgon Carbon Corporation (Pittsburgh, PA) was the primary adsorbent used for modeling and pilot testing experiments in this study. It has been used by several Lake Michigan water utilities for T&O control. The adsorbent was oven dried at 105°C overnight and stored in a desiccator until used in pilot testing. The equilibrium and kinetic parameters of WPH carbon for 2-MIB adsorption in Lake Michigan water are inputs to the mathematical model described below. The raw data needed to generate the Freundlich equilibrium parameters ($K$ and $1/n$) and the surface diffusion kinetic parameter ($D_s$) for MIB adsorption on WPH in Lake Michigan were taken from isotherm and batch kinetic tests reported previously by Gillogly et al. (1998a,c). Table 1 presents the model inputs used in this study including the equilibrium and kinetic parameters for WPH PAC.

Mathematical modeling of adsorption performance
A computerized version of a mathematical adsorption model developed previously by Campos et al. (2000a) was used to investigate the effects of various operating parameters in a submerged-style, hybrid sorption-membrane system. The model was originally developed and verified for pressure-style membrane systems and single-solute adsorption situations (Campos et al., 2000a,b). Campos et al. (2000c) also used the model for predicting the removal of natural organic matter (NOM) from a natural water by lumping all the competing NOM species together as a single equivalent background compound (EBC). This study showed good correlation between the model and pilot-scale results for NOM removal in a pressure-style membrane system. Matsui et al. (2000) and Schideman et al. (2001) applied this modeling approach to predict trace organic removal in the presence of competing NOM by using the EBC approach and a pseudo single-solute approach. This later study showed good agreement between model predictions and a bench-scale, submerged-style membrane system. However, the model has not previously been compared to experimental results for trace organic removal in a pilot-scale, submerged membrane reactor, which is the subject of this study.

A general description of the modeling approach and the primary model assumptions are summarized here and Campos et al. (2000a) give additional details on the model development. Adsorption in the membrane reactor is modeled as a non-steady-state process with adsorption kinetics controlled by internal surface diffusion; adsorption equilibrium is
described by the Freundlich isotherm equation. Analytical solutions of the homogeneous surface diffusion model with no external mass transfer limitation are used to evaluate adsorbate concentrations in the solid phase as a function of time. In the model, the sorption-membrane reactor is assumed to be a continuously stirred tank reactor (CSTR) with ideal mixing. All of the PAC is assumed to leave the reactor during each backwash of the membrane system. Also, the model uses input parameters from batch equilibrium and kinetic tests to predict results in a flow-through situation where PAC is held in place by the membrane while water passes through. Other researchers have demonstrated that pore blockage and displacement by NOM components can result in lower adsorption capacities and slower kinetics for trace organic compounds in flow through systems than what is measured in batch isotherm and kinetic tests (Li et al., 1999, 2001). By using the batch test equilibrium and kinetic parameters for model inputs, the model neglects any effects of pore blockage and displacement.

Note that the equilibrium parameters in a natural water system change as a function of initial MIB concentration. Thus, the initial MIB concentration used in the computer model was set to match the previous isotherm data from Gillogly et al. (1998a). However, this reference also demonstrated that for typical concentrations of MIB in natural waters, the percent of MIB removed by adsorption on PAC is essentially independent of initial concentration. Thus, the results of this study are presented as percent of MIB removed, and the data can be applied to any reasonable initial concentration of MIB that is naturally occurring.

Pilot-scale hybrid sorption-membrane system

A pilot-scale membrane system was installed at the Racine Water Utility in Racine, Wisconsin and was fed with water from the full-scale water plant, which uses Lake Michigan as its water source. The pilot membrane unit selected for this project was the ZeeWeed®-1000 submerged ultrafiltration (UF) pilot unit as manufactured by ZENON Environmental Inc. (Oakville, Ontario). Compared to previous systems, this unit is a relatively new product that uses a smaller diameter hollow fiber membrane with a higher packing density, and thus has a larger capacity per unit surface area occupied by the process. Also, this unit uses a smaller trans-membrane pressure, and it does not require continuous aeration to scour the membrane.

The test program included measurement of the concentration of 2-MIB before and after the pilot PAC-UF process while varying operational parameters. On most days the pilot was operating without any 2-MIB or PAC being added. On selected days, 2-MIB and PAC were dosed to the pilot influent water from concentrated stock solutions using peristaltic pumps. The UF influent line was sampled continuously at a point downstream of the 2-MIB

<table>
<thead>
<tr>
<th>Mathematical model parameters used in this study</th>
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<tr>
<td><strong>Model input parameter</strong></td>
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<tr>
<td>Freundlich K</td>
</tr>
<tr>
<td>Freundlich 1/n</td>
</tr>
<tr>
<td>Surface Diffusion Coefficient $D_s$</td>
</tr>
<tr>
<td>Average PAC Particle Radius</td>
</tr>
<tr>
<td>MIB Influent Concentration</td>
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<tr>
<td>Initial Reactor Concentration of MIB</td>
</tr>
<tr>
<td>Backwash Interval (Max. Carbon Retention Time)</td>
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<tr>
<td>Carbon Dose</td>
</tr>
<tr>
<td>Carbon Dosing Method</td>
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<tr>
<td>Reactor Volume</td>
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<tr>
<td>Flow Rate</td>
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</table>
addition, but upstream of the PAC addition point. The UF permeate was also sampled continuously. Influent and effluent samples were composited over the testing period (usually one 3-hour backwash interval) and then sent to a commercial analytical laboratory for analysis (Montgomery Watson Labs, Pasadena CA). 2-MIB analysis was done according to the proposed standard method 6040-D, which is a gas chromatography-mass spectrometry (GC-MS) assay; samples were prepared using solid phase microextraction (SPME).

Results and discussion

Continuous PAC dosing experiments

In most drinking water applications, PAC is added in a continuous (or step) dose that is chosen to produce a desired amount of contaminant removal. The dose is changed as the influent concentration of the target contaminant changes. The pilot system was operated with different continuous PAC doses while 2-MIB was spiked into the membrane system influent. Composite samples of the influent and effluent streams were analyzed for 2-MIB corresponding to each continuous dose and compared to mathematical model predictions.

Table 2 presents the predicted and measured percent removal of 2-MIB for a range of continuous PAC doses. The mathematical model results correlate very well with the measured results and are considered to be within the inherent experimental variability.

Pulse PAC dosing experiments

With the PAC-UF process, it is also possible to dose PAC in a single large “pulse” at the beginning of the membrane filtration cycle as an alternative to conventional continuous dosing procedures. Campos et al. (2000b,c) demonstrated the benefit of pulse dosing in mathematical modeling predictions and verified those results with a pressure-style membrane system. The pulse dosing procedure increases the average carbon residence time (CRT). In turn, this results in more efficient PAC usage, because the average PAC particle has more time to approach its equilibrium capacity. To confirm these effects, the pilot system was operated with two different pulse PAC doses. Once again, composite samples of the influent and effluent were analyzed for 2-MIB for each pulse dose and compared to mathematical model predictions. Table 3 presents the predicted and measured percent removal of 2-MIB for the pulse PAC doses that were tested. The mathematical model results did not correlate well with the measured results, and in fact the observed
removal of 2-MIB with a 10 mg/L pulse dose was slightly lower than for the same continuous dose.

The likely explanation for these results is that there are dead-water zones in the reactor. Upon visual inspection of the reactor, PAC was lodged between the membrane housing and tank walls where it would not be in good contact with the water flowing through the membranes. Also, it is expected that PAC could accumulate at the bottom of the reactor, because the flow enters on one end of the reactor and no attempt is made to distribute the influent evenly across the bottom. Continuous dosing would be less susceptible to PAC accumulating in dead-water zones, because all the water enters the reactor being well mixed with PAC. With pulse dosing the water must become mixed with PAC after it enters the reactor. For pulse dosing, the assumption of CSTR conditions appears to neglect an important physical phenomenon in the pilot-scale, submerged PAC-UF reactors.

Effects of aeration with pulse dosing

In an attempt to mitigate the effects of reactor dead-water zones with pulse dosing, the membrane aeration was turned on throughout the entire filtration cycle for a couple of PAC experiments. The aeration was intended to keep the reactor contents well mixed and hopefully restore the advantage of pulse dosing. Table 4 presents the results of the aeration testing. Once again, the mathematical model significantly overestimated the removal of 2-MIB. In fact, the observed percent removals of 2-MIB with aeration and pulse dosing were lower than for the equivalent pulse dose without aeration. A possible explanation of these results is that the aeration breaks up the PAC cake that would otherwise form on the membrane surface. The PAC cake could act like a miniature carbon column that is more efficient at removing PAC than a CSTR arrangement, because the mini-column effluent concentration can be lower than the reactor bulk concentration. The formation of a PAC cake is visually apparent, especially for pulse dosing experiments, where the membrane reactor contents become progressively clearer as the PAC is deposited on the membrane surface.

The theory of a beneficial PAC cake could also explain why the continuous dose pilot results given earlier could achieve removals equal to or slightly greater than the predicted removals in spite of the negative influence of pore blockage and displacement effects that are not accounted for in the model. The negative effects of pore blockage and displacement mechanisms, which have been documented by other researchers (Li et al., 1999, 2001), could be counteracted by the positive effect of forming a PAC cake that acts like a mini-column of PAC on the membrane surface.

The data interpretation given above is plausible and it avoids the postulation that continuous aeration exacerbated reactor dead-water zones, which seems unlikely. Nonetheless, the data presented here are not sufficient to conclusively demonstrate that the proposed explanation of the data is accurate and complete. Further work will be required to confirm the hypothesis presented here. What is clear is that the assumptions and simplifications of currently available models for the PAC-UF process need to be carefully considered when

<table>
<thead>
<tr>
<th>Pulse PAC dose (mg/L)</th>
<th>Removal of 2-MIB in pilot plant (%)</th>
<th>Predicted removal of 2-MIB (%)</th>
<th>Ratio of measured to predicted removal</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>2%</td>
<td>0%</td>
<td>n/a</td>
</tr>
<tr>
<td>9</td>
<td>35%</td>
<td>70%</td>
<td>0.50</td>
</tr>
<tr>
<td>17</td>
<td>49%</td>
<td>86%</td>
<td>0.57</td>
</tr>
</tbody>
</table>

Notes: Results are for 180 minute backwash interval and a reactor volume of 220 L
Aeration was on throughout these PAC tests, and reactor flow rate was 55 L/min
considering process scale-up. Model refinements are likely to be needed to more fully
and accurately address non-ideal reactor mixing, pore blockage, displacement and other
phenomena that can dominate in certain reactor configurations and alternative operational
protocols.

Effects of backwash interval

In most membrane systems, the backwash interval determines the carbon retention time
(CRT), because the reactor contents are eliminated with each backwash to limit the buildup
of solids in the membrane reactor. CRT has a significant influence on adsorption perfor-
mance, because it determines the amount of time PAC has to approach its equilibrium
capacity. Table 5 presents a comparison of pilot and model results for various backwash
intervals and PAC dosing conditions. The first part of the table presents data for continuous
dosing, which has a reasonable correlation between the model and pilot results.

Unfortunately, the water plant was adding chlorine to the raw water during these con-
tinuous dose tests and carried a free chlorine residual in the water that was fed to the mem-
brane reactor. Chlorine has been shown to reduce the capacity of PAC for MIB in Lake
Michigan water (Gillogly et al., 1998b), and these effects are not quantified in the model.
Thus, these pilot plant results are biased towards lower percent removals. Even with this
complication, the trend of increasing percent removals by increasing the backwash interval
was clearly demonstrated for continuous dosing in both the model and pilot results.

The second part of Table 5 compared the effects of backwash interval for pulse dosing.
In this case, the pilot data and model predictions did not correlate well. The pilot data for
pulse dosing did not reflect a significant increase in the percent removal of 2-MIB for
extended backwash intervals as predicted by the model. In fact, the pilot results for a
180-minute backwash interval with pulse PAC dosing were actually slightly worse than for
30 and 90 minute backwash intervals. Once again, these results are likely explained by
reactor dead-water zones and short-circuiting. The effects of dead-water zones becomes
more pronounced with longer backwash intervals as the PAC is added in a bigger pulse and
it has more time to settle into areas where it is not effective. It is also noteworthy, that the
30 minute backwash interval, pulse dose pilot test result actually exceeded the predicted
removals. This supports the idea of a beneficial PAC cake formation, and indicates that for
short backwash intervals the effects of reactor dead-water zones is minimal.

Most PAC-UF systems in operation today operate with a backwash interval between 20
and 90 minutes, which typically leaves a significant amount of adsorption capacity unused.
However, there is a tradeoff between adsorption efficiency and membrane fouling or
flux decline, which increases as backwash intervals are extended. Discussions with the

<table>
<thead>
<tr>
<th>Backwash interval (min)</th>
<th>PAC dose and dosing method</th>
<th>Removal of 2-MIB (%)</th>
<th>Predicted removal of 2-MIB (%)</th>
<th>Ratio of measured to predicted removal</th>
</tr>
</thead>
<tbody>
<tr>
<td>22</td>
<td>40 mg/L continuous</td>
<td>48%</td>
<td>55%</td>
<td>0.87</td>
</tr>
<tr>
<td>180</td>
<td>40 mg/L continuous</td>
<td>78%</td>
<td>87%</td>
<td>0.90</td>
</tr>
<tr>
<td>30</td>
<td>10 mg/L pulse</td>
<td>46%</td>
<td>42%</td>
<td>1.09</td>
</tr>
<tr>
<td>90</td>
<td>10 mg/L pulse</td>
<td>48%</td>
<td>62%</td>
<td>0.77</td>
</tr>
<tr>
<td>180</td>
<td>10 mg/L pulse</td>
<td>44%</td>
<td>73%</td>
<td>0.60</td>
</tr>
</tbody>
</table>

Notes: Continuous dose pilot plant experiments were conducted during a time of prechlorination at the
water plant. Prechlorination will reduce PAC capacity, and this is not accounted for in the model
predictions. Results are for a reactor volume of 220 L. Flow rates were 92 L/min for continuous dose experiments and 50 L/min for pulse dose experiments.
membrane system supplier led to the conclusion that a backwash interval of 180 minutes would be reasonable for pilot testing with this source water. The membrane performance with these extended backwash intervals was not significantly impacted in the pilot testing. A previous study of the mathematical modeling of backwash interval effect on the efficiency of 2-MIB removal from Lake Michigan water indicated that extending backwash intervals to 180 minutes would allow the adsorbent to achieve approximately 90% of its maximum capacity (Schideman et al., 2001). This is a good example of the type of multi-objective optimization that should take place with hybrid sorption-membrane systems to achieve a reasonable balance between the membrane performance and adsorption performance.

Summary

One approach to improving the adsorption performance in hybrid sorption membrane processes is to assess the effects of operational parameters and attempt to control these parameters to optimize performance. A mathematical model facilitates optimization because it significantly reduces the cost and time associated with optimization testing. In this study, a mathematical model was used to predict advantageous operational approaches, and the predictions were tested with pilot plant experiments. This process verified the model for certain operating conditions and identified some situations where the model deviates from observed results. Explanations for the deviations are suggested in terms of model assumptions that may oversimplify the actual system.

This study investigated various ways to improve the performance of hybrid adsorption-membrane systems, with a focus on the removal of 2-MIB from a Lake Michigan source water. The effects of various operational parameters were evaluated with a mathematical adsorption model and a pilot-scale submerged membrane reactor. With continuous PAC dosing procedures, the model and pilot data agreed reasonably well. However, with pulse PAC dosing, the model consistently overpredicted adsorption performance. The authors suggest that differences with pulse dosing results are likely due to non-ideal mixing conditions not included in the current model. These mixing conditions can have a dominant effect with pulse dosing in a submerged membrane process. Aeration during pulse dosing runs was attempted to increase reactor mixing and hopefully eliminate inefficiencies due to non-ideal mixing. However, aeration actually decreased adsorption removals in comparison to pulse dosing without aeration. The authors hypothesize that the negative effects of aeration are due to disturbance of the PAC cake layer that forms on the surface of the membrane. The PAC cake can act as a miniature column of activated carbon that is more efficient for adsorption than a CSTR condition. This study highlights the need to consider reactor configuration and scale when applying mathematical models of adsorption in hybrid adsorption-membrane processes, and provides some general insights for optimizing these processes.

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