Ultrafiltration as tertiary treatment for industrial reuse

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Abstract In this paper we present the results obtained from tests conducted on ultrafiltration treatment of a secondary effluent designated for possible industrial reuse. Tests were carried out at Empoli Wastewater Treatment Plant (WWTP) (Florence, Italy) with the use of a hollow fiber pilot plant (mod. ZW-10, Zenon Environmental). Testing lasted for about 4 months, during which the pilot plant was fed by effluent from the wastewater treatment plant. Results show that the permeate was of high quality. The membranes were very efficient in reducing turbidity (94.5%) and TSS (98.7%). The reduction of COD (around 35%) was also good. The permeate also showed low SDI values (usually < 3%). With respect to microbiological parameters, treatment was shown to be above all efficient in the removal of Escherichia coli. The permeate already respects required quality standards set forth by a new technical law decree on water reuse, soon to be approved within Italian Legislation. Based on these test results, a preliminary design of a wastewater treatment plant with the complex of structures necessary to the distribution of the treated wastewater in the industrial area located closed to the plant, has been created. Under this design, treated water could be produced at a cost of 0.38 €/m³, which includes investment, financial charges and maintenance costs.

Keywords Effluent reuse; industrial water reuse; membrane treatment; ultrafiltration

Introduction
In the past, the use of membranes for water treatment (wastewater treatment, refinement, drinking water treatment) was limited to particular situations due to their high costs (management and plant costs). The technological development and the increased water request, make nowadays membranes a competitive and convenient choice for wastewater treatment in particular in the field of refinement of purified water for reuse. The relative literature is wide and technical problems with respect to quality of treated water and managerial parameters seem to have been solved. This is especially true for microfiltration (MF) and ultrafiltration (UF) treatment processes. In 1995, Freeman and Morin described numerous applications for the use of MF/UF as pretreatment of reverse osmosis for wastewater reclamation and reuse (e.g. industrial, irrigation, groundwater recharge). This solution is still little spread in Italy, and experiences are necessary to evaluate, in the specified national context, the sustainability of the treatment system, with respect to law bounds and costs of treated water. The main advantages of MF/UF systems are:
(a) reduction of TSS and turbidity much improved with respect to traditional treatments,
(b) permeate with very low turbidity and TSS, regardless of the feed properties, and
(c) optimal pre-treatment for reverse osmosis.

As a matter of fact, according to Leslie et al. (1998), a combined membrane process using either MF or UF hollow fiber membrane followed by reverse osmosis (RO) spiral wound membranes is recognized as the low cost alternative for municipal wastewater reuse plants. Of substantial importance is also the capacity of MF and UF in the significant removal of microorganisms including viruses (Madaeni et al., 1995), reducing or eliminating expenses for disinfection. Within the field of water treatment, MF/UF processes can be installed in several ways:
(a) MF can be coupled with biological systems (especially MF) for the direct filtration of activated sludge in the case of membrane bioreactors,
(b) for direct treatment of wastewater designated for reuse (see for example Abdessembed et al., 1999; Ahn and Song, 1999; Tay and Jeyaseelan, 1995), and
(c) as a tertiary treatment for the refinement of effluent from wastewater treatment plants designated for reuse (see for example Tchobanoglous et al., 1998; Parameshwaran et al., 2001).

In this paper, the possibility of carrying out an UF treatment for the refinement for industrial reuse of the effluent of Empoli WWTP, has been investigated. The plant of Empoli is located close to a large industrial area, for which further expansion is planned in the near future. In the area, natural water sources are not present in sufficient quantity, therefore some companies are directly supplied from the local municipal waterworks. Tests were carried out with the following objectives:
(a) to test the quality of permeate and its variability with respect to feed,
(b) to verify that the permeate respects the water quality standards set forth for industrial reuse defined in the proposed law decree (still to be approved), and
(c) to assess whether this water is suitable for further refinement through reverse osmosis.

This last objective is highly relevant, since the process of reverse osmosis not only allows the integration of UF (in case reverse osmosis proves unsuccessful alone), but also increases potential uses, by including cases where reverse osmosis is provided for in the treatment cycle. In hypothesizing construction of a full scale UF plant and its water distribution system, an analytical cost evaluation of water per cubic metre has been carried out.

Material and methods

Pilot plant

For experimentation a pilot plant ZenoGem® (Zenon Environmental S.r.L., Milan) equipped with a module of UF ZeeWeed® (Mod. ZW-10) was used. The pilot plant was positioned inside the wastewater treatment plant of Empoli (60,000 p.e., mean daily flow 13,500 m³/d). This is a typical activated sludge plant: pretreatments, primary sedimentation, oxidation and secondary sedimentation. Disinfection through chlorination is carried out only in emergency. Presently, pre-denitrification module is being constructed. The pilot plant was continuously fed with the effluent from the wastewater treatment plant. During about 4 months, from November 2001 to March 2002. For the preliminary definition of working parameters of the pilot plant, laboratory tests were carried out in the previous 3 months, by use of membranes.

Description. The pilot plant is made up of: a process tank (V = 220 L); a UF module, type ZeeWeed® (Table 1); a control panel; a process pump; a blower. The following instrumentation was also present: pressure/vacuum meter, located after UF modules, water temperature thermometer placed in the process tank and a flux meter for the measurement of air flux sent in the process tank. The layout of the pilot plant is presented in Figure 1. The UF ZeeWeed® 10 (ZW-10) module is formed of a series of hollow fibers, connected to superior and inferior heads, where outlets for the various instruments (process pump, pressure meter, blower) are present. The ZW-10 module is submerged in a process tank which works with an invariable hydraulic head; this is controlled by use of a buoy that controls a valve situated on the pipeline of the feed. A diffuser disperses air in the bottom of the module. Rising air bubbles induce an upward liquid flow and shaking of fibers, which controls particle deposition.

The system works through alternation of filtration and backwash (BW) phases, whose duration can be adjusted by the control panel. The system runs at constant flow and varying
pressure. The process pump connected to the head of the module creates a depression inside
the module, which gives rise to a filtration outside-in mode. The permeate that is produced
is partly stored in a 25 l reservoir (CIP) for backwash and the surplus is discharged. At the
end of the filtration, the process pump reverses the flow and sends the contents of the CIP
upstream and into the UF module for backwashing. To limit the concentration of sub-
stances in the membrane, a flux of concentrate is extracted from the process tank.

Start-up and operational parameters. Before activating the filtration system, a procedure
of conditioning was carried out, in order to remove the residual of the solution of mainte-
nance of membranes. The membrane was immersed in hot water (35–40°C), with 200 mg/L
sodium hypochlorite added. Conditioning lasted 12 hours. Secondly, the integrity of mem-
branes was verified using a bubble test, which blew air upstream into the module at 0.35 bar
for a duration of 5 minutes as to observe the dimension of the bubbles exiting from the
module. The next step was to start-up the plant. The operational parameters are shown in
Table 2.

The control of the pilot plant was made on the basis of specific flux ($J_s$), obtained from
the equation:

$$J_s = \frac{J_{(@20^\circ C)}}{\text{TMP}}$$

Flux $J$ was therefore previously normalized at 20°C through the following relation (Adham
and Gramith, 2000):

### Table 1 Specific parameters of ZW-10 module

<table>
<thead>
<tr>
<th>Model</th>
<th>ZW-10</th>
<th>Allowed range of pressure</th>
<th>–0.7/-0.6 psi</th>
</tr>
</thead>
<tbody>
<tr>
<td>Configuration</td>
<td>Hollow fibers</td>
<td>Max temperature allowed</td>
<td>40°C</td>
</tr>
<tr>
<td>Material</td>
<td>Organic, non-ionic,</td>
<td>Allowed pH range</td>
<td>5–9</td>
</tr>
<tr>
<td></td>
<td>hydrophilic</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Membrane surface</td>
<td>0.93 m²</td>
<td>Allowed pH range for cleaning</td>
<td>2–10.9</td>
</tr>
<tr>
<td>Nominal pore size</td>
<td>40 nm</td>
<td>Max concentration of ClO⁻ allowed</td>
<td>1,000 mg/L</td>
</tr>
<tr>
<td>Flux (average/max)</td>
<td>15/35 L/h·m²</td>
<td>Air flow</td>
<td>3.6 Nmc/h</td>
</tr>
</tbody>
</table>
The maximum allowable TMP value was fixed at 0.35 bar. When this value was reached, the chemical cleaning of membranes was carried out. The procedure of chemical cleaning was carried out by flushing a solution of 200 mg/L sodium hypochlorite downstream through the membranes for 30 min.

Water analysis
Along with the experimental period the quality of feed and permeate were monitored for physical–chemical and microbiological parameters. From November to February, 1 or 2 weekly samples were collected and analyzed for COD, nitrogen forms (ammonium, nitrite and nitrate), Total Suspended Solids (TSS), turbidity. The Silt Density Index (SDI) was also determined according to the procedure of ASTM Standards (1982). All samples were taken and immediately analyzed according to the IRSA-CNR methods (IRSA-CNR, 1994). Analytical measurement were carried out using the following instruments: Hach mod. 2100P for turbidity, Varian mod. Cary 50 for spectrophotometric measures. For microbiological analysis, all the samples (1 L) have been taken in asepsis conditions and analyzed for TC (MPN Technique) and \( E.\ coli \) (Membrane Filter Technique). For microbiological analysis, all the samples (1 liter) have been taken in asepsis conditions and analyzed for TC (MPN Technique) and \( E.\ coli \) (Membrane Filter Technique). Two hours before microbiological sampling the hydraulic circuit found downstream from the membrane was sterilized with a solution at 500 ppm of sodium hypochlorite for 30 minutes.

Results
The values over time of specific flux \( (J_s) \) and of Transmembrane Pressure (TMP) during testing are shown in Figure 2. Plots outline an initial phase of conditioning of the membrane

\[
J(@20^\circ C) = J \cdot \exp(-0.0239 \cdot (T - 20))
\]  

Figure 2 Specific flux \( (J_s) \) and TMP over the experimental period

<table>
<thead>
<tr>
<th>Table 2</th>
<th>Operational parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Permeate flow ( (Q_p) ) 22 l/h</td>
</tr>
<tr>
<td>Filtration</td>
<td>Flux ( (J) ) 23.7 L/h m²</td>
</tr>
<tr>
<td>Duration</td>
<td>420 s</td>
</tr>
<tr>
<td>Backwash</td>
<td>Flux ( (J) ) 35.5 L/h m²</td>
</tr>
<tr>
<td>Duration</td>
<td>40 s</td>
</tr>
<tr>
<td>Duration of 1 cycle</td>
<td>420 s</td>
</tr>
<tr>
<td>Concentrate withdrawal</td>
<td>2 L/h</td>
</tr>
<tr>
<td>Recovery</td>
<td>90%</td>
</tr>
<tr>
<td>Recovery (including BW)</td>
<td>73%</td>
</tr>
</tbody>
</table>

by guest
during a period of about 900 h. This was characterized by a rapid decrease of $J_s$ (from 1.5 to 0.35 m$^3$/m$^2$·d bar after first 240 h), following which $J_s$ began to oscillate around a mean value of 0.42 m$^3$/m$^2$·d bar. In the same phase, TMP was shown to be very low (average 0.07 bar). Next, with the start of the phenomenon of membrane fouling, $J_s$ decreased to 0.08 m$^3$/m$^2$·d bar after around 1550 h and TMP increased to 0.35 bar. At this point a chemical washing was carried out in accordance to the previously described procedure. Values of $J_s$ and TMP immediately following washing, show a full recover of filtration efficiency. Before the second chemical washing (1,300 h), the trends of $J_s$ and TMP were repeated in a similar manner. Also after the second washing, filtration efficiency was fully recovered.

Values of COD, turbidity and TSS, measured in the feed, in the permeate and in the process tank are listed in Table 3. UF treatment provided notably good results for what concerns removal of TSS and turbidity. These parameters were characterized by oscillations, even of notable significance, in the feed and in the process tank, also affected by the wastewater treatment plant’s bulking phenomenon. Despite this, in the permeate, values were always found to be < 1 mg/L for TSS and < 1 NTU for turbidity (Figure 3). Respective removal rates were on average 98.7% and 94.5%. Also remarkable, even though less evident, is removal of COD, clearly limited to particulate fraction.

The treatment proved to be efficient also for SDI parameter (Table 4). The SDI showed an average ($n = 11$) of 3.1 and only in 3 tests were values > 3.3 recorded. An on average slight decreasing trend in SDI was noted during testing.

The module ZW-10 also proved to be highly capable in the removal of microorganisms (Table 5). In the case of *E. coli*, in particular, membranes turned out to be an absolute barrier, as the permeate showed no trace of *E. coli*. The barriers also proved capable, although not absolute, in reducing TC as was expected.

It was noted that the maintenance of microbiological quality of permeate relies on the adding of disinfectant, to avoid bacterial re-growth. Sampling carried out without the preliminary disinfection of the circuit, resulted in a significatively poorer microbiological quality.

**Discussion**

Trends of TMP and $J_s$ suffered from the use of a new membrane, for which the resistance to filtration is limited to the specific resistance of the material. After the initial phase,
filtration cycles lasted around 700 h before reaching the pre-designed lower threshold for \( J_s \). The efficiency of chemical washing proved to be very high in both cases. We can therefore state that, with respect to the testing period, the effluent of the water treatment plant is suitable for UF treatment, as no particular foulants (oils, silicones, etc) seem to be present. Otherwise, obstructions would much more rapidly appear, and the full recovery of filtration efficiency would require, during washing, the use of specific washing products, suggested by the producer.

In the light of these results, it can be stated that the quality of permeate in terms of turbidity and TSS content is independent from feed quality. Results agree with what reported in literature. Similar results were found also from Cotè et al. (2001), who used ZeeWeed\textsuperscript{®} modules to refine secondary effluent from different wastewater plants. Using a hollow fiber treatment, van Hoof et al. (2001) also noted turbidity values for a treated permeate of < 1 NTU with feed being between 5 and 20 NTU. With respect to the present SDI values, permeate can be considered suitable for further eventual reverse osmosis treatment.

Other authors report even better results: van Hoof et al. (2001) noted SDI values between 1 and 2 from UF treatments with hollow fibers; also in this case, nevertheless, some oscillations of the parameter were reported. These oscillations may be due to iron that can form insoluble oxide and/or hydroxide in an oxidative environment. In the secondary effluent concentrations higher than 0.8 mg/L were noted rather frequently. Luo and Wang (2001), for example, noted that iron concentration of 0.3 mg/L can cause fouling phenomenon in RO modules. The pilot plant showed the ability to generate an effluent with no \( E. \) coli. This is very important for reuse: the new Italian Law, presently being approved, defines a more restrictive threshold than previously, 10 \( E. \) coli in 100 ml. The tertiary effluent is already suitable without the need of further disinfection treatments.

**Water cost assessment**

To estimate treated water cost, the overall design of a system comprised of UF, a compensation/reservoir, and a water distribution system providing for potential users, has been made. In the calculation, investment, amortization, working and maintenance costs for the various components of the system have been taken in account. In Table 6 and Table 7 the main parameters used in calculation and the final results are shown.

The total cost of water is therefore very competitive when compared to the relative cost of sampling the waterworks, fixed as 1.23 €/m\(^3\) for industrial users.

**Table 4** Mean, max and min values of SDI and Plugging Factor (PF)

<table>
<thead>
<tr>
<th>SDI</th>
<th>PF (%)</th>
</tr>
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<tbody>
<tr>
<td>Mean</td>
<td>3.1</td>
</tr>
<tr>
<td>Max</td>
<td>6.2</td>
</tr>
<tr>
<td>Min</td>
<td>0.8</td>
</tr>
</tbody>
</table>

**Table 5** Results of microbiological analysis

<table>
<thead>
<tr>
<th>n.</th>
<th>TC (MPN/100 ml)</th>
<th>E. Coli (UFC/100 ml)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Concentrate</td>
<td>Permeate</td>
</tr>
<tr>
<td>1</td>
<td>380,000</td>
<td>1,950</td>
</tr>
<tr>
<td>2</td>
<td>93,000</td>
<td>11,000</td>
</tr>
<tr>
<td>3</td>
<td>93,000</td>
<td>11,000</td>
</tr>
</tbody>
</table>
Conclusions
In this paper, the possibility of using UF for tertiary refinement of the effluent of Empoli wastewater treatment plant designated for industrial reuse, was investigated. For this purpose a pilot plant was set up and water quality (feed and permeate) as well as main operational parameters were monitored. Operational parameters were maintained static. In particular, TMP and $J_s$ showed gradual and relatively gradual increases, which agree with those found in previous tests. Given the chosen operational conditions, it was necessary to carry out chemical cleansing of membranes once a month. Permeate showed very low TSS and turbidity values (respectively $<1$ mg/L and 1 NTU), regardless of the feed quality. The produced flux appears to be suitable for RO, with SDI values generally being less than 3. Furthermore, a high ability of removing indicator microorganisms was noted. In particular, E. coli was completely removed by the treatment. The permeate has therefore the microbiological characteristics requested by the Technical Decree, presently being approved, that will regulate reuse of treated water in Italy.

On the basis of these experimental results, an economical analysis for the evaluation of investment, amortization, exercise and maintenance costs of the UF treatment of a compensation reservoir and the relative distribution network was carried out. The total cost is estimated to be 0.38 €/m³, therefore highly competitive when compared to 1.23 €/m³, the cost of water for industrial reuse from municipal waterworks.

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