Treatment of yellow water by membrane separations and advanced oxidation methods
Z. Lazarova and R. Spendlingwimmer

ABSTRACT

Comparative experimental study is performed on purification of yellow wastewaters separated and collected in solarCity, Linz, Austria. Three membrane methods (micro-, ultra-, and nano-filtration), and two advanced oxidations (gamma radiation and electrochemical oxidation) were applied. Best results concerning the removal of pharmaceuticals and hormones from urine by membrane separation were achieved using the membrane NF-200 (FilmTec™). Pharmaceuticals (ibuprofen and diclofenac), and hormones (oestrone, β-oestradiol, ethynylestradiol, oestriol) were removed completely from urine. NF-separation also has some disadvantages: losses of urea, and lowering the conductivity in the product (permeate). The retentates (concentrates) received have to be treated further by oxidation to destroy the “problem” compounds. The results showed that electrochemical oxidation is more suitable than gamma radiation. Gamma-radiation with intensities higher than 10 kGy has to be applied for efficiently destroying of ibuprofen, and especially diclofenac. A high quantity of intermediate “problem” substances with oestrone structure was formed during the gamma oxidation of hormone containing urine samples. The electrochemical oxidation can be successfully applied for elimination of pharmaceuticals such as diclofenac, and hormones (oestrone, β-oestradiol) from yellow wastewater without loss of urea (nitrogen fertiliser).

Key words | hormones, pharmaceuticals, urine treatment, wastewater purification, yellow water

INTRODUCTION

Urine must be considered not only as a problematic wastewater pollutant but also as a valuable natural resource of nutrients. Its volume is relatively small, less than 1% of the municipal wastewater volume. However, up to 80% of the total nitrogen, and around 45% of the total phosphorus load in the municipal wastewaters originate from urine (Larsen & Gujer 1996). It is disadvantageous to mix such a “high loaded” waste flow with the other wastewaters to be purified further in the sewer treatment plants. Recently, growing activities have been undertaken to collect and purify the urine separately. The separation of urine can contribute to a reduction of the nitrogen discharge into the wastewaters by approximately 60% (Johansson & Nykvist 2001). This will result in smaller dimensions of the purification plants and, respectively, lower investment and operating costs. In the future, separate urine collection will be more sustainable than tertiary wastewater treatment (Wilsenach & van Loosdrecht 2006).

An important advantage of urine separation is not only the production of high quality effluents, but also the saving of natural resources by using urine for fertilising and irrigating landscapes. The concentration of nitrogen and phosphorus in urine is 100 times higher than in wastewater, therefore, the separate collection of urine substantially improves the potential for nutrient recovery (Wilsenach & van Loosdrecht 2003). Moreover, because of decoupling of the different wastewaters flows, a fertiliser without heavy metal impurities can be produced.

The collected urine, however, contains many micro-impurities which are secreted from the bodies of living
organisms into the urine. Therefore, it has to be pre-treated before reaching the purification plant or before being used as nitrogen fertiliser. The most problematic substances contained in the urine can be summarised into two main groups: pharmaceuticals residues and endocrine disruptors (e.g. natural and synthetic hormones).

Researchers at Eawag, Switzerland studied a series of methods for urine treatment (complexation, precipitation, and ozonation, and membrane processes such as nanofiltration and electrodialysis) with the purpose of producing a fertiliser from urine (Pronk et al. 2006a,b; Pronk 2007). A combination of electrodialysis and ozonation is proposed as the most effective treatment of urine to produce fertiliser.

The application of membrane filtration, in particular of nano- and ultra-filtration, as well as reverse osmosis, is examined intensively for the removal of endocrine disrupting substances and pharmaceuticals from waters (Nghiem & Schafer 2005; Yoon et al. 2006). A combination of bioreactor and reverse osmosis (Oppenheimer et al. 2004) or of bioreactor and a conventional wastewater treatment plant (Clara et al. 2005) are also reported.

The literature review showed that besides ozonation other oxidation processes such ionising (gamma) radiation (Gehringer & Eschweiler 1996; Gehringer et al. 2004), and electrochemical oxidation (Debus 2004; Sires et al. 2004; Sizykh et al. 2004; Abhijit et al. 2005; Kim et al. 2005; Menini et al. 2005; Yoon et al. 2005) can be applied to also reduce or eliminate the concentration of pharmaceuticals, hormones or other toxic organic substances in urine.

In this study, a combination of membrane separation and two oxidation processes, gamma radiation and electrochemical oxidation, is investigated to remove (first step) and destroy (second step) pharmaceuticals and hormones contained in the urine, achieving a maximum yield of urea in the product solution. During the first (separation) step, the “problem” compounds will be separated from the yellow water, and “pure” urine (permeate) and concentrates (retentates) will be produced. At the second step, the retentates will be oxidised to reduce or destroy the organic compounds.

The study was performed in the framework of the EcoSan full scale pilot project solarCity, Linz, Austria. Besides new technologies, such as solar technology, and energy-saving technology, an ecological sanitation concept with urine separation toilets is applied. Urine separation toilets are installed, and the yellow water solutions used in our experiments are samples collected in this city (460 population equivalent).

**EXPERIMENTAL**

**Materials**

The following membranes were chosen for the membrane filtration of the yellow water:

- for micro-filtration: membrane Celgard 3400 (Celgard Co. LLC, Charlotte, USA) made of polypropylene, coated with a wetting agent; porosity 37%, pore size $0.117 \times 0.042 \mu m$; thickness $25 \mu m$;
- for ultra-filtration: membrane Nadir UF-CA-1 (Hoechst High Chem, Germany);
- for nano-filtration: three membranes NF90, NF200, NF270 (FilmTec™ Membranes, Dow, USA); according to the manufacturer, the NF-membranes are structured as follows: $0.2 \mu m$ polyamide, $40 \mu m$ polysulphone and $120 \mu m$ polyester as a support layer (Product Information, Film Tec, 2006).

**Analytical methods**

Preliminary analysis of the yellow water samples collected separately in the SolarCity (Linz AG, Austria) showed that they contain relatively high concentrations of ibuprofen, caffeine and diclofenac. Pharmaceuticals such as carbamazepin and clofibric acid were not identified. The concentration of hormones in the samples was very low, therefore, hormones (for example, oestrone and β-oestradiol) were additionally spiked to the samples to be treated further by membrane separations and/or oxidation methods.

The analysis of these compounds was performed by GC-MS (GC-MS-system 6890-GC/5973inert-MSD from Agilent (Palo Alto, CA, USA) with an HP-5 MS capillary column (30 m, 0.25 mm i.d., 0.25 μm film thickness). The detection limit of the analytical method applied was 0.2 μg/L.

The analysis of urea was accomplished using the Berthelot method. The measurements were performed on a UV/VIS spectrophotometer (Cary 3, Varian) at 700 nm wavelength.
Temperature, pH-values and conductivity of the samples were measured on-line using a pH-meter (HQ20, HACH-Lang), and a conductivity meter (LDO™).

**Experimental procedures**

**Membrane separation**

A cross-flow pressure-driven filtration mode was used, whereby the feed (yellow water sample) was flowed parallel to the membrane surface, and divided into two streams (permeate and retentate) due to the pressure difference on both sides of the membrane. The stream, which was pressed through the membrane pores, is a purified product (permeate), whereas the other stream (retentate) is a concentrated solution of the organic compounds which are not able to pass the membrane. The retentate solution has to be treated further or discharged.

The cross-flow membrane unit used (Amafilter BV, Nederland) is shown in Figure 1. Five different membranes were first tested with distilled water (with low salt content), and afterwards with a synthetic aqueous solution of urea. In this way, the permeate fluxes and the permeability of the urea molecules through the membranes were determined and compared.

In the next step, a yellow water sample (provided by Linz AG) was pre-treated by means of a blue band filter (BBF, Schleicher & Schuell) with a pore size of 2 µm, in order to separate the rough suspended particles from the original urine solution. Then, the BB-filtrates were further treated by means of three membrane separation methods (micro-, ultra- and nano-filtration), and both oxidation methods, electrochemical oxidation and gamma radiation.

**Gamma-radiation**

The gamma-radiation is an advanced oxidation method, based on the production of the strongest oxidising agent in the aqueous systems—the active OH-radicals. Gamma-emitters are chemical elements, respectively materials, which send energy in the form of electromagnetic waves. As gamma-emitters, usually iridium 192 (¹⁹²Ir) and cobalt 60 (⁶⁰Co) are used. The gamma-rays resemble the visible light and the X-ray, however they are stronger, and can penetrate materials without to be reflected and/or broken. A part of the radiation is absorbed during the passage, depending on the density and the thickness of the medium.

The gamma radiation experiments included in this study were performed by means of the irradiator Gammacell 200 (Nuclear Engineering Seibersdorf GmbH) with a ⁶⁰Co-cell (Figure 2). All the yellow water samples (untreated, and treated by BBF, MF, UF, and NF) were illuminated with two radiation intensities: 5 and 10 kGy.

**Electrochemical oxidation**

The experiments were accomplished with the experimental plant shown in Figure 3. The electrochemical cell was
provided with graphite electrodes (or graphite electrodes with TiO₂ coating), and functioned as a flow-through reactor. Yellow waters sample treated with BBF were used as a feed.

Two experimental series were performed. The first experiment was carried out in a recycle mode with a constant flow rate of 20 L/h. The power of the direct current applied was changed to examine the influence of the electric field strength on the oxidation efficiency. At first, 1.5 V was applied for 120 min, then, the voltage was increased to 2.5 V for a further 2 hours. Process parameters such as pH, conductivity of the oxidised yellow water, electrical power (voltage and amperage), as well as the temperature, were measured on-line. Periodically, samples were taken from the inlet and outlet of the cell, and the concentration of pharmaceuticals, hormones and urea was analysed.

During the second experimental series, four subsequent cycles were performed with a sample of 35 L yellow water at higher flow rate (150 L/h) and voltage of 2.5 V.

RESULTS AND DISCUSSION

Membrane separation

In Figure 4, samples of untreated yellow water, and after membrane filtrations are shown: original yellow water (YW), BB-filtrate, MF (retentate, permeate), UF (retentate, permeate), and NF (retentate, permeate). Following pressure differences are applied: for MF—2.5 bar; for UF—10.5 bar; for NF—10.5 bar.

It was supposed that the urea molecules (the source of N) contained in the yellow water will pass the membrane pores because of their low molecular weight (60 g/M), whereas the larger molecules of the pharmaceuticals and hormones (>200 g/M) will be rejected from the membrane surface, and will be retained in the concentrated stream (retentate).

Permeation of urea through the membrane

Results concerning the permeation of urea through the micro-, ultra- and nano-filtration membranes are summarised in Table 1. The preliminary experiments showed that the membrane NF-200 is more appropriate than NF-270 and NF-90, if the purpose of the treatment is to receive a product (permeate) containing urea as an N source. Therefore, the results with the NF-membrane NF-200 only are given in Table 1. The pH-values of permeates (P) and retentates (R) as well as the conductivities χ before and after the membrane separation are compared.

Table 1 | Membrane separations: Urea in the yellow water (YW) samples

<table>
<thead>
<tr>
<th>YW sample</th>
<th>Urea (mg/dL)</th>
<th>pH</th>
<th>χ (mS/cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>YW original</td>
<td>150.0</td>
<td>8.36</td>
<td>9.77</td>
</tr>
<tr>
<td>YW after BBF</td>
<td>148.3</td>
<td>8.56</td>
<td>6.82</td>
</tr>
<tr>
<td>MF</td>
<td>151.0 (R)</td>
<td>8.68 (R)</td>
<td>8.33 (R)</td>
</tr>
<tr>
<td></td>
<td>144.8 (P)</td>
<td>8.63 (P)</td>
<td>8.18 (P)</td>
</tr>
<tr>
<td>UF-01-C</td>
<td>105.3 (R)</td>
<td>8.69 (R)</td>
<td>9.69 (R)</td>
</tr>
<tr>
<td></td>
<td>90.9 (P)</td>
<td>8.67 (P)</td>
<td>7.77 (P)</td>
</tr>
<tr>
<td>NF-200</td>
<td>101.7 (R)</td>
<td>8.58 (R)</td>
<td>7.95 (R)</td>
</tr>
<tr>
<td></td>
<td>57.9 (P)</td>
<td>8.79 (P)</td>
<td>3.51 (P)</td>
</tr>
</tbody>
</table>
The results show that all the membrane separation methods do not influence the pH-value of both retentates (R) and permeates (P). The conductivity of the treated samples, however, changes substantially, particularly in the case of nano-filtration. In all cases, the conductivity in the retentates is higher than the corresponding value in permeates. The conductivity of the permeates depends on the applied membrane separation methods decreasing from 8.18 mS/cm (MF), to 7.77 (UF), and 3.51 (NF). In the case of ultra-filtration, and especially of the nano-filtration,

<table>
<thead>
<tr>
<th>YW samples</th>
<th>Ibuprofen Initial concentration (µg/L)</th>
<th>Conc. after treatment (µg/L)</th>
<th>Retention (%)</th>
<th>Diclofenac Initial concentration (µg/L)</th>
<th>Conc. after treatment (µg/L)</th>
<th>Retention (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>YW (BBF)</td>
<td>185</td>
<td>150 (BBF)</td>
<td>18.9 (rel. to YWorg)</td>
<td>3.1</td>
<td>2.5</td>
<td>19.3 (rel. to Worg)</td>
</tr>
<tr>
<td>Micro-filtration</td>
<td>150</td>
<td>150 (P)</td>
<td>0 (rel. to BBF)</td>
<td>2.5</td>
<td>2.5</td>
<td>0 (rel. to BBF)</td>
</tr>
<tr>
<td>Ultra-filtration</td>
<td>150</td>
<td>115 (P)</td>
<td>23.3 (rel. to BBF)</td>
<td>2.5</td>
<td>1.5</td>
<td>40 (rel. to BBF)</td>
</tr>
<tr>
<td>Nano-filtration</td>
<td>150</td>
<td>3.5 (P)</td>
<td>97.7 (rel. to BBF)</td>
<td>2.5</td>
<td>0</td>
<td>100 (rel. to BBF)</td>
</tr>
</tbody>
</table>

The results show that all the membrane separation methods do not influence the pH-value of both retentates (R) and permeates (P). The conductivity of the treated samples, however, changes substantially, particularly in the case of nano-filtration. In all cases, the conductivity in the retentates is higher than the corresponding value in permeates. The conductivity of the permeates depends on the applied membrane separation methods decreasing from 8.18 mS/cm (MF), to 7.77 (UF), and 3.51 (NF). In the case of ultra-filtration, and especially of the nano-filtration,

Table 3 | Gamma irradiation with two intensities—5 and 10 kGy: Comparison of urea-concentrations, pH-values and conductivities in Retentates (R) and Permeates (P)

<table>
<thead>
<tr>
<th>YW sample</th>
<th>Urea (mg/dl) 5 kGy</th>
<th>Urea (mg/dl) 10 kGy</th>
<th>pH 5 kGy</th>
<th>pH 10 kGy</th>
<th>Conductivity 5 kGy (mS/cm)</th>
<th>Conductivity 10 kGy (mS/cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>YW original</td>
<td>99.15</td>
<td>61.03</td>
<td>8.61</td>
<td>8.6</td>
<td>8.76</td>
<td>8.68</td>
</tr>
<tr>
<td>YW after BBF</td>
<td>87.09</td>
<td>80.0</td>
<td>8.26</td>
<td>8.59</td>
<td>7.87</td>
<td>7.45</td>
</tr>
<tr>
<td>Ultra-filtration (UF-01-C)</td>
<td>72.82 (R)</td>
<td>61.48 (R)</td>
<td>8.55 (R)</td>
<td>8.83 (R)</td>
<td>9.86 (R)</td>
<td>9.55 (R)</td>
</tr>
<tr>
<td></td>
<td>69.54 (P)</td>
<td>64.93 (P)</td>
<td>8.79 (P)</td>
<td>8.78 (P)</td>
<td>7.73 (P)</td>
<td>7.71 (P)</td>
</tr>
<tr>
<td>Nano-filtration (NF-200)</td>
<td>110.95 (R)</td>
<td>89.73 (R)</td>
<td>8.53 (R)</td>
<td>8.73 (R)</td>
<td>7.95 (R)</td>
<td>7.83 (R)</td>
</tr>
<tr>
<td></td>
<td>57.52 (P)</td>
<td>49.03 (P)</td>
<td>8.96 (P)</td>
<td>8.87 (P)</td>
<td>3.48 (P)</td>
<td>3.53 (P)</td>
</tr>
</tbody>
</table>

The results show that all the membrane separation methods do not influence the pH-value of both retentates (R) and permeates (P). The conductivity of the treated samples, however, changes substantially, particularly in the case of nano-filtration. In all cases, the conductivity in the retentates is higher than the corresponding value in permeates. The conductivity of the permeates depends on the applied membrane separation methods decreasing from 8.18 mS/cm (MF), to 7.77 (UF), and 3.51 (NF). In the case of ultra-filtration, and especially of the nano-filtration,

Table 4 | Oxidation of pharmaceuticals with gamma radiation (5 and 10 kGy)

<table>
<thead>
<tr>
<th>YW samples</th>
<th>Ibuprofen Concentration before radiation (µg/L)</th>
<th>Oxidation degree (%)</th>
<th>Diclofenac Concentration before radiation (µg/L)</th>
<th>Oxidation degree (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>YW original</td>
<td>185</td>
<td>87.05</td>
<td>96.22</td>
<td>2.5</td>
</tr>
<tr>
<td>YW (BBF)</td>
<td>150</td>
<td>94.00</td>
<td>97.00</td>
<td>2.0</td>
</tr>
<tr>
<td>MF (R)</td>
<td>210</td>
<td>85.24</td>
<td>97.38</td>
<td>2.5</td>
</tr>
<tr>
<td>UF (R)</td>
<td>160</td>
<td>87.50</td>
<td>98.13</td>
<td>2.5</td>
</tr>
<tr>
<td>NF (R)</td>
<td>235</td>
<td>78.7</td>
<td>95.32</td>
<td>2.5</td>
</tr>
<tr>
<td>MF (P)</td>
<td>185</td>
<td>84.86</td>
<td>97.57</td>
<td>1.5</td>
</tr>
<tr>
<td>UF (P)</td>
<td>115</td>
<td>95.22</td>
<td>99.57</td>
<td>2.8</td>
</tr>
<tr>
<td>NF (P)</td>
<td>3.5</td>
<td>100</td>
<td>100</td>
<td>n.d.</td>
</tr>
</tbody>
</table>

*no detection.
†under detection limit.
some urea can not pass the membrane, it is held back in the retentate.

**Removal of pharmaceuticals by cross flow membrane filtration**

The results on the removal of pharmaceuticals (ibuprofen and diclofenac) from yellow water by different membrane filtrations methods are shown in Table 2.

Very high rejection of ibuprofen (approximately 98%) was achieved with the nano-filtration membrane NF-200. The concentration of ibuprofen was reduced from 150 µg/L (in the YW-feed) to 3.5 µg/L (in the permeate). It can be supposed that the yellow water can be completely purified from such pharmaceuticals which have a similar structure to that of ibuprofen. Diclofenac was not found in the permeate flow, probably because of its rejection from the membrane surface. It must be considered, however, that the initial concentration of diclofenac in the yellow water was substantially lower (2.5 µg/L) than of ibuprofen (185 µg/L).

**Gamma radiation**

**Oxidation of urea by gamma irradiation**

The results summarised in Table 3 show that the irradiation of yellow water with gamma rays does not substantially change the pH-values and the conductivities of retentates and permeates. However, some urea is oxidised, whereby the oxidation degree depends on the intensity of the gamma radiation.

**Oxidation of pharmaceuticals by gamma radiation**

The analysis of ibuprofen in the treated retentate samples (Table 4) shows that higher intensity (at least 10 kGy) is necessary in order to oxidise this pharmaceutical more effectively. Only solutions with relatively low concentrations of ibuprofen (such the permeates with 3.5 µg/L) can be oxidised completely with 5 kGy. The oxidation degree at 5 kGy depends on the concentration of the substance in the solution, as illustrated in Figure 5.

In spite of the very low concentration of diclofenac in the yellow samples, it was not possible to destroy more than 92% of diclofenac in the retentates. This means that...
pharmaceuticals with similar structures are more stable to the gamma rays, and higher intensities are needed for their successful elimination.

**Electrochemical oxidation**

The data received by means of electrochemical oxidation at two voltages (first experimental series) are summarised in Table 5. It can be seen that there is very small decrease in pH-values and conductivities in the oxidised solutions. The urea concentration remains unchanged after treatment with 1.5 V; at higher voltage (2.5 V) only a small amount of urea is oxidised. The efficiency of the process concerning destroying pharmaceuticals depends on the strength of the applied electric field, and the initial concentration of the pharmaceuticals. More than 90% of ibuprofen (initial concentration of 120 mg/L) was oxidised with 2.5 V (the current measured was 0.7 A). Diclofenac (initial concentration of 1.5 µg/L) was destroyed completely using the electrochemical oxidation.

As a next step (second experimental series), electrochemical oxidation of solutions containing hormones and pharmaceuticals was performed in four subsequent cycles at the following operating conditions: feed volume 35 L, flow rate 150 L/h; voltage 2.5 V. The YW was spiked with relatively high concentrations of some “problem” endocrine disruptors: the hormones oestrone, β-oestradiol, and the pharmaceutical diclofenac. The corresponding initial concentrations (relatively high values) are given in Figure 6. The results show that diclofenac was completely destroyed. Moreover, approximately 80% of the hormones were eliminated from the yellow water.

In Table 6, the results of the chemical analysis concerning hormones in some YW samples after membrane separation (NF-retentate and NF-permeate) and gamma irradiation are given.

It can be seen that very efficient separation of all hormones available in the yellow water samples is achieved by nano-filtration separation with the membrane NF-200 (sample NF-P).

Oxidation of the retentates with gamma radiation (5 and 10 kGy) is not efficient. The NF-retentates, which are illuminated with 5 kGy, contain higher oestrone and β-oestradiol concentrations after the radiation than the same sample before the radiation (see NF-R). It could be supposed that, in this case, intermediate products with oestrone structure are formed during the oxidative splitting by gamma radiation.

**CONCLUSIONS**

The following conclusions can be drawn on basis of the experimental results of this study.

- Nano-filtration (membrane NF-200) can be applied for efficient separation of pharmaceuticals such as ibuprofen and diclofenac from yellow waters. However, it has to be taken into consideration that NF leads to changes in the conductivity, and some urea will be lost.
- NF can be used for the removal of hormones such as oestrone, oestradiol, ethenyl-oestradiol and oestriol from yellow waters.
- Gamma radiation with an intensity higher than 10 kGy is necessary for completely destroying pharmaceuticals in yellow water; some urea will also be destroyed.
- Gamma radiation with 10 kGy leads to the formation of splitting intermediate products with an oestrone structure, therefore it is not appropriate for the treatment of yellow waters containing hormones.

<table>
<thead>
<tr>
<th>Yellow water sample</th>
<th>Oestrone (µg/L)</th>
<th>β-oestradiol (µg/L)</th>
<th>Ethenyl-oestradiol (µg/L)</th>
<th>Oestriol (µg/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>NF (R) membrane separation</td>
<td>112</td>
<td>0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
</tr>
<tr>
<td>NF (P) membrane separation</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
</tr>
<tr>
<td>NF (R) gamma radiation with 5 kGy</td>
<td>753</td>
<td>1.1</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
</tr>
<tr>
<td>NF (R) gamma radiation with 10 kGy</td>
<td>42</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
</tr>
</tbody>
</table>

In Table 6, the results of the chemical analysis concerning hormones in some YW samples after membrane separation (NF-retentate and NF-permeate) and gamma irradiation are given.
• Electrochemical oxidation can be successfully applied for destroying pharmaceuticals such as ibuprofen and diclofenac in yellow waters.
• The electrochemical oxidation does not affect the urea concentration, and destroyed effectively hormones.
• Pilot experiments are needed to optimise the electrochemical oxidation treatment of the yellow water.

**ACKNOWLEDGEMENTS**

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**REFERENCES**


